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A Quantum Theoretical Approach to Hard X-ray Time-Domain Interferometry

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In connection with the work on this thesis, the following article was published in a refereed journal

- P. Uhrich, S. Castrignano, H. Uys, and M. Kastner, "*Noninvasive measurement of dynamic correlation functions*", Phys. Rev. A (2017)

Moreover, also in connection with the work on this thesis, the following preprint was submitted for publication on a refereed journal

- S. Castrignano and J. Evers, "*Probing Quantum Dynamical Couple Cor- relations with Time-Domain Interferometry*", ArXiv e-prints (2018), 1805.01672.

Zusammenfassung

In dieser Arbeit präsentieren wir eine quantentheoretische Darstellung der Zeitbereich-Interferometrie mit harten Röntgenstrahlen und schlagen eine experimentelle Technik zur Untersuchung der zeitlichen Korrelationen zwischen Teilchen in einem kondensierten Materie-System über ihre Wechselwirkung mit harter Röntgenstrahlung vor.
Diese Technik wurde bereits erfolgreich auf klassische Systeme angewendet. Der jüngste Vorschlag, die gleiche Technik für Systemen zu verwenden, in denen Quanteneffekte eine große Rolle spielen, erfordert eine detaillierte Analyse aufgrund der dramatischen Auswirkungen, die der Messakt auf die Dynamik eines Quantensystems haben kann. Insbesondere der Versuch, über direkte Messungen auf die Korrelationen in einem Quantensystem zuzugreifen, würde nur unvollständige Informationen über sie liefern.
Indem wir sowohl das System der sondierten Materie als auch die sondierende Strahlung als Quantensysteme betrachten, die schwach miteinander wechselwirken, zeigen wir, dass die Strahlung in der Zeitbereich-Interferometrie das System im obigen Sinne nicht beeinflusst, sodass die zeitlichen Korrelationen der Teilchen vollständig erfasst werden können. Darüber hinaus wird im Hinblick auf aktuellen Fortschritt bei der Kontrolle von Röntgenpulsen vorgeschlagen, dass die Zeitbereich-Interferometrie für die Rekonstruktion von Teilchenkorrelationen und für die Erkennung des Vorhandenseins von Quanteneffekten im untersuchten System verwendet werden kann.

Abstract

In this work we present a quantum theoretical account of hard x-ray time-domain interferometry, which is an experimental technique to probe the correlations in time between particles in a condensed matter system via their interaction with hard x-radiation.
This technique has so far been successfully applied to classical systems. The recent proposal of using the same technique on systems for which quantum effects play a major role requires a detailed analysis due to the dramatic effect that the measurement act can have on the dynamics of a quantum system. In particular, trying to access the correlations in a quantum system via direct measurements would give only incomplete information about them.
Treating both the probed matter system and the probing radiation as quantum systems which interact weakly, we show that in time-domain interferometry the radiation does not affect the system in the above sense, such that it can access the particles correlations in time fully. Furthermore, in view of some recent advancements in x-ray control, it is proposed that time-domain interferometry can be used for the reconstruction of particles correlations and for detecting the presence of quantum effects in the probed system.

Contents

Chapter 1

Introduction

The discovery in 1895 of x-rays by Wilhelm C. Röntgen paved the way to a major breakthrough in our understanding of the structure of matter. Indeed the successive intuition and demonstration in 1912 by Max von Laue that the – at the time debated – wave nature of x-rays could be exploited to probe the crystalline structure of some solids, consecrated x-rays as a fundamental tool for the systematic experimental investigations of the atomic structure of matter by x-rays. An account of the different possibilities offered by x-ray scattering by matter for the exploration of static properties of the latter can be found in [1].

X-ray scattering methods allow as well to probe dynamical properties of condensed matter samples on several different length and time scales – ranging from $\sim$ 0.1 Å to $\sim$ 1000 Å and from $\sim fs$ to $100s$ – and a big effort is being done in the last years to push these limits further, since the understanding of complex phenomena at microscopic scales is key for future technological development[2, 3, 4].
One possibility to recover information about the dynamics in a sample is represented by inelastic x-ray scattering techniques (IXS) [5], in which the spectrum of excitation in the sample is retrieved by measuring the energy losses of an x-ray beam during its scattering from matter. This technique grants access to dynamics on timescales between $\sim fs$ and $\sim \mu s$ [1].

For probing phenomena that happen on slower time scales, like e.g. dif-

[1]This kind of experiments can be also run using neutrons as a probe, accessing complementary length and time scales [6]

fusion dynamics [7] or glass formation and other phenomena [8], it may be preferable to access them directly in the time domain. Two techniques are available that can achieve this result. X-ray photon correlation spectroscopy [9], is a technique in which the speckle patterns created at different times by scattering of x-rays from particles in the sample are correlated. This technique grants access to dynamics happening on timescales slower than the millisecond , and recently it has been demonstrated in the picoseconds to nanosecond range [10].
The second technique, and the one on which the work in this thesis is based, is Time-Domain Interferometry (TDI) [11]. This technique exploits quasi-elastic scattering of hard x-rays from a sample to access dynamics on timescales ranging from tens to hundreds of nanoseconds, and spatial scales from fractions to tens of Ångstrom .
In this technique, the target system is placed between two parallel filtering foils. A hard x-ray short pulse from a synchrotron source is split by the first foil in two pulses of different duration, which are later scattered by the target at different times. The two scattered pulses will then extract information on the spatial distribution of atoms in the target at different times, and are recombined by the second foil in such a way to make them interfere at the detector. The interference pattern generated in this way, will then give access to the correlations among the positions of atoms in the target at different times.
In particular via TDI, the spatial Fourier transform of the space-time correlations among particles is probed, a quantity known as intermediate scattering function (ISF).
TDI has been proposed and successfully demonstrated originally in [11] and since then has been further developed [12, 13], and used to study slow diffusion dynamics in glass-forming fluids [14, 15, 16] and crystalline solids [17].

In recent times TDI has been suggested for probing dynamics in strongly correlated systems [2]. Indeed the interplay among the different degrees of freedom of these systems can bring to the appearance of interesting dynamics on a variety of different space and time scales [18]. However, in order to understand the features of these systems a quantum description of matter is needed [19, 20, 18]. The perspective of applying TDI to quantum systems rises the problem of what will be actually measured in such experiments.
The measurement act on quantum systems is indeed one of the most controversial and debated issues in the foundation of quantum theory [21, 22].

In a classical framework, the back action of a measurement apparatus on a system plays no major role as it can, at least in principle, made arbitrarily small. In this case then a measurement on a system does not affect the outcome of subsequent measurements on the same system, and time correlations between physical observables can be obtained just by directly measuring the physical quantities of interest at successive times and correlating the results obtained.
If one tries to apply this scheme to obtain time correlations for a quantum system, the back action of the first measurement would provoke an abrupt change in the dynamics of the system [23], affecting thus all the outcomes of subsequent measurements. Therefore, the result obtained by correlating these outcomes is not a faithful representation of correlations that develop spontaneously in the system, because it accounts for an external intervention on the system itself that altered its dynamics.
The full correlation functions for a quantum system, that is the ones that do not account for measurement back action, are thus different from the ones obtained by direct measurements [24, 25].
Even in less extreme situations, in which the interaction between the measuring apparatus and the system is weak, the interpretation of measurement outcomes is a delicate issue [26]. For instance, in [27] it has been demonstrated that, even for minimally disturbing measurements on quantum systems in thermal equilibrium, the measured time correlations between two observables may not correspond to the full ones.
These issues are thus not only of foundational relevance, but pose also practical problems in devising experiments; recently a number of proposals for the measurement of the full two-time correlation functions on ensembles of quantum mechanical spin-1/2 systems has been put forward [28, 29, 25].

The main aim of the work on this thesis is thus to analyse the working of TDI when used for measurements on ensembles of quantum particles, in order to understand whether the full – in the sense that measurement back action is not accounted for – space-time correlations between the particles, or only a part of them, is accessed. In particular we present a theoretical account of this technique in which both the target and the radiation are considered as quantum systems in interaction. The main result presented in this thesis is that TDI is suitable to access the full particles correlation of the quantum system and not just parts of it.

The structure of this work

In chapter 2 definitions of dynamical correlation functions are given for generic pairs of physical observables, in both classical and quantum theoretical frameworks. It is shown that the full quantum dynamical correlation functions are complex valued functions given by the sum of two contributions: the first one, called the projective part, which is accessed when direct measurements of the correlated observables are done on the system, while the second one originates from the fact that these values are in general undefined if the system is in a quantum superposition of states and thus would be deleted by the back action of a direct measurement.
A first result of this work is then shown [25], i.e. that the projective part can equal the real part of quantum dynamical correlations only if the correlated observables can take only two possible values equal in magnitude and discord in sign.
In the end a model experimental protocol to avoid measurement back action by indirect coupling of the system to a measuring device is proposed and discussed.

In chapter 3, the differences between classical and quantum dynamical correlation functions are specialized to space-time correlations among particles in an ensemble, which are the correlations of interest for TDI experiments. In particular the dynamical couple correlation function (DCF) and its Fourier transforms are defined. These three functions enjoys some spatial symmetries in the classical case, that can be violated in the quantum mechanical case.
It is then argued that a control of these symmetries for an unknown system, can represent a method to experimentally assess whether a quantum model is in order for its correct decription. By analysing two quantum mechanical model systems, it is shown however that quantum systems can be in superposition of states that give space-time correlations with the same symmetries of the classical case.

In chapter 4, after a brief summary of the working principles of TDI, the second, major result of this work is given: via an analysis of this technique in which both the target sample and the probing hard x radiation are considered as quantum systems in weak interaction, it is shown that TDI can access the full quantum space-time correlations among the particles of the

sample – in particular the full ISF –, avoiding the measurement back action that would give only a partial access to them [30].
The method of our analysis is first shown on a model scheme analogous to TDI, in which the foils are substituted with split and delay units. After that the method is specialized to the case of the actual TDI.
By a perturbative analysis of the interaction between the hard x radiation and the electrons distribution in the sample, a picture of the photon-sample dynamics happening in the scheme emerges: a photon has two different scattering channels to reach the detector, each one extracting information from the sample at a different time. The recombination of this channels at the detector provokes a temporal interference in the signal generated by photon detection, from which the spatial Fourier transform of the quantum DCF can be extracted.
Moreover, some new usages of TDI are proposed.

Finally, in chapter 5, the work presented in this thesis is summarized and some directions for new research are proposed.

Three appendices come with the main text. The first one briefly resumes the time-dependent perturbation theory in quantum mechanics, whereas the second resumes the quantization of the free electromagnetic field. These two appendices contain no new results, their only function is to make the text self-consistent. The third appendix contains the details of calculation of some integrals that appear in chapter 4, which would have made the main text more difficult to follow.

Chapter 2

The dynamical correlation functions in quantum theory

In this chapter we define dynamical correlation functions in both classical and quantum theory. The differences between classical and quantum dynamical correlation function is addressed starting from the different structures of the two theories, in particular the origin of the difference between the quantum dynamical correlations and their classical counterparts is traced back to the fact that the configuration of a quantum system can be a superposition of different configurations.
It is then shown that the quantum dynamical correlation function can be split in the sum of a contribution related to the probabilities that the physical quantities take definite values in a direct measurement, and a second contribution that accounts for the fact that for quantum superposition of states the value of these quantities can be not defined.
Finally a model of experimental protocol, that we will call two-times ancilla measurement, is illustrated that allows to retrieve the full quantum dynamical autocorrelation function of an observable of a quantum system. This model is actually an abstraction of the main features of Time Domain Interferometry, so that the analysis here proposed is nothing else than a translation on a more general level of the detailed calculations given in chapter 4 for the specific case of TDI.

2.1 The Classical case dynamical correlations

Let us consider a classical system of which we want to determine the correlations between the value of the quantity A at time t_1 and the value of quantity B at time t_2. In a classical statistical theory A and B are random variables defined over the sets of all their possible values. We will call these values $\{a_i\}_i$ and $\{b_j\}_j$ respectively, with the labels i and j that can assume continuous or discrete values.
At each instant of time, we can define a probability that A assumes a given value a_i and the same we can do for B and b_j. Moreover we can assign a probability for the joint event that A evaluates to a_i at time t_1 and B to b_j at time t_2, that we will call $P(A = a_i, t_1; B = b_j, t_2)$.
The classical dynamical correlation function is then calculated as the following mean value

$$C_{AB}^{(cl)}(t_1, t_2) \equiv \sum_{i,j} a_i \, b_j \, P(A = a_i, t_1; B = b_j, t_2) \tag{2.1}$$

The classical dynamical correlation function is directly accessible experimentally. Indeed we can make in principle direct repeated measurement of A at t_1 and then of B at t_2, obtaining for each measurement a couple of values. Through the sequence of pairs of values $\{(a_{i(1)}, b_{j(1)}),\ (a_{i(2)}, b_{j(2)}),\ \ldots,(a_{i(n)}, b_{j(n)})\}$ obtained in this way, the function (2.1) can be reconstructed as

$$C_{AB}^{(cl)}(t_1, t_2) = \frac{1}{n} \sum_{\alpha=1}^{n} a_{i(\alpha)} \, b_{j(\alpha)} \tag{2.2}$$

This identification of the arithmetic average on the right hand side of (2.2) and the definition given above of the classical dynamical correlation function, is totally legit for classical systems because, in each run of the combined measurements, the influence of the measuring apparatus at t_1 on the outcome of the measurement at t_2 can be considered negligible in principle.

2.2 The Quantum dynamical correlations

2.2.1 The structure of quantum theory

What happen if we apply the same experimental protocol to a quantum system? In order to understand that, it is useful to recall briefly the basic

postulates of quantum theory. In doing this we rely on the most common interpretation of the theory, that is the so-called Copenhagen interpretation.

Postulate I The state of a system at a given time t is represented by a normalized vector $|\psi(t)\rangle$ belonging to a suitably defined Hilbert space $\mathcal{H}$.

Postulate II The physical observables of the system are represented by hermitian linear operators defined over the Hilbert space of states. The set of possible outcomes for a measurement of a physical quantity O is the set of the eigenvalues of the corresponding operator $\hat{O}$. We will refer with the word "observable" interchangeably to both the operator and the physical quantity it represents.

Postulate III Given that the system is in the state $|\psi(t)\rangle$, the probability that a measurement of O gives the outcome o_i is given by the Born rule

$$P(O = o_1, t) = \langle\psi(t)|\hat{\Pi}_{o_i}|\psi(t)\rangle \tag{2.3}$$

being $\hat{\Pi}_{o_i}$ the projector on the eigenspace of $\hat{O}$ in $\mathcal{H}$ corresponding to the eigenvalue o_i.

Postulate IV The evolution in time of the state of the system is given by the Schrödinger equation

$$i\hbar\frac{d}{dt}|\psi(t)\rangle = \hat{H}|\psi(t)\rangle \tag{2.4}$$

Here $\hat{H}$ is the hermitian operator corresponding to the Hamiltonian of the system [1].

Postulate V During a measurement of an observable, the dynamics of the state of the system is no more described by the Schrödinger equation. In particular if a measurement of the observable O gives the value o_i, the state of the system after the measurement is

$$|\psi(t)\rangle \longrightarrow \frac{\Pi_{o_i}|\psi(t)\rangle}{\sqrt{\langle\psi(t)|\Pi_{o_i}|\psi(t)\rangle}} \tag{2.5}$$

[1] In what follows we assume that the systems are closed ones, so that thei hamitlonians are time-independent.

Some remarks on these postulates are in order.
The first two postulates represent a shift in the mathematical description of physics. Whereas in a classical theory the state of a system corresponds with the collection of values of relevant observables, in quantum mechanics the concept of state is "detached" from that of observable. The state becomes an abstract object in a linear space, this meaning, at variance with classical theories, that a sum of states is as well a fair description of the system.

On the other hand postulate II and III help us to give a meaning to these superposition of states. In fact, by virtue of postulate II, the set of eigenvectors of an observable $\hat{O}$ – or of a system of observables mutually commuting – is an orthonormal basis for the Hilbert space of states making it possible to re-express at any time the quantum state $|\psi(t)\rangle$ as a sum of eigenvectors of $\hat{O}$ with complex valued coefficients

$$|\psi(t)\rangle = \sum_i c_i(t)|o_i\rangle \tag{2.6}$$

Postulate III tells us how to interpret the coefficients $c_i(t)$ of such expansion: substituting the right hand side of (2.6) in the expression (2.3) for the Born rule, we obtain

$$P(O = o_i, t) = |c_i(t)|^2 \tag{2.7}$$

Formulae (2.6) and (2.7) inform us that the best knowledge we can ever have about a quantum system is probabilistic in nature.
So in quantum theory the state of the system represents in general a multiplicity of possible values for its physical properties.
From the probability distribution defined by the coefficients $c_i(t)$ one can calculate the average value of an observable expected in many measurements as $\langle O\rangle(t) = \langle\psi(t)|\hat{O}|\psi(t)\rangle$.

The only way we can be sure of the value of an observable is by measuring it, but this action singles out one of the manifold possibilities expressed by the quantum state, as stated by postulate V. A measurement spoils the linear superposition of eigenstates that constitutes the state and this spoiling action is what we referred to before as measurement back action on the system.
Actually one may be interested in measuring more than one observable. It may happen that the operators corresponding to different observables do not commute, this meaning that there is no common system of eigenvectors.

To fix the ideas let us consider two non-commuting observables $\hat{A}$ and $\hat{B}$: the state $|\psi(t)\rangle$ can be expanded in one or the other of the two sets of eigenvectors $\{|a_i\rangle\}_i$ or $\{|b_j\rangle\}_j$ obtaining either the probability distribution for the values of $\hat{A}$ or for those of $\hat{B}$, but determining one spoils the definition of the other. This fact is more evident if we refer to what happens to $\hat{B}$ in a measurement of $\hat{A}$. A measurement of the latter provokes the collapse of the state in one of the eigenvectors $|a_i\rangle$, which in turn can be expanded as a linear combination of eigenvectors of $\hat{B}$ with coefficients $c_j^{(a_i)}$

$$|a_i\rangle = \sum_j c_j^{(a_i)} |b_j\rangle \tag{2.8}$$

A probability distribution for the values of $\hat{B}$ remains then defined as $P(B = b_j) = |c_j^{(a_i)}|^2$, whereas the value of A is determined with certainty. It is then said that two non-commuting observables are incompatible, as the determination of one of them by measurement forbids the knowledge of the values of others non-commuting with the first, as stated in the famous indeterminacy principle.
This fact is of capital importance for the properties of dynamical correlation functions in quantum theory as will be shown below.

For what concerns the dynamics of a quantum system instead, postulate IV tells us that the hamiltonian is the infinitesimal generator of the dynamics of the system, which in turn can be represented by a unitary transformation $\hat{U}(t)$ of its initial state

$$|\psi(t)\rangle = \hat{U}(t, t_0)|\psi(t_0)\rangle \tag{2.9}$$

The dynamical problem expressed by the Schrödinger equation can be translated in an initial value problem for $\hat{U}(t)$

$$\begin{cases} i\hbar \frac{d}{dt}\hat{U}(t) = \hat{H}\hat{U}(t) \\ \hat{U}_0(t_0, t_0) = \mathbb{1} \end{cases} \tag{2.10}$$

The prediction of quantum theory are given in terms of expectation values which can be re-expressed with the aid of the evolution operator as

$$\langle\psi(t)|\hat{O}|\psi(t)\rangle = \langle\psi|\hat{U}(t, t_0)^\dagger \hat{O}\hat{U}(t, t_0)|\psi\rangle \tag{2.11}$$

one can think of the initial state of the system as a static object and attribute a dynamical nature to the observables defining the corresponding time-dependent operators as

$$\hat{O}(t) \equiv \hat{U}(t, t_0)^\dagger \hat{O} \hat{U}(t, t_0) \tag{2.12}$$

In this way we can restate the problem of dynamics on the observables, writing down the differential equations they obey

$$i\hbar \frac{d}{dt} \hat{O}(t) = [\hat{O}(t), \hat{H}] \tag{2.13}$$

and considering the initial state as fixed.
These two different but equivalent perspectives on the dynamics, the one in which the state evolves with time and the one in which observables evolve instead, are known respectively as the Schrödinger and the Heisenberg picture of quantum theory.

2.2.2 The incomplete dynamical correlation function retrieved by projective measurements

Now we are able to understand the consequences of the experimental protocol proposed in the beginning on a quantum system.

Let us assume that the system at time $t_0 = 0$ is represented by the state

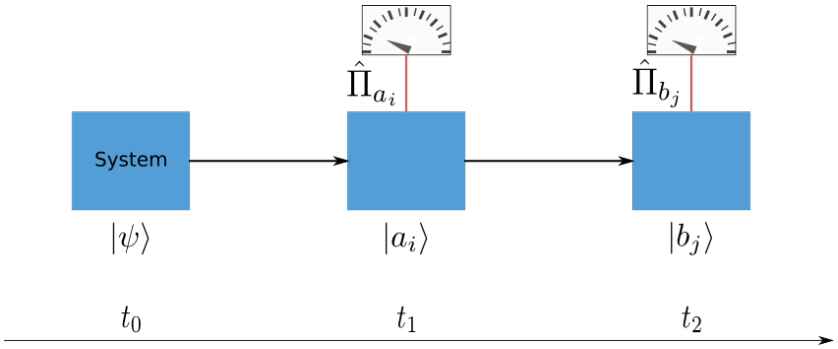

Figure 2.1: Schematic representation of the direct measurements of the two observables at different times. The first measurement at t_1 meakes the state of the system collapse into an eigenstate of $\hat{A}$, whereas the second provokes a second collapse of the state at t_2 into an eigenstate of $\hat{B}$.

$|\psi\rangle$ and that the measurement of A is done at time t_1 giving the value a_i. By

postulate V we know that the state of the system after the measurement is

$$\frac{\hat{\Pi}_{a_i}\hat{U}(t_1)|\psi\rangle}{\sqrt{\langle\psi|\hat{U}(t_1)^\dagger\hat{\Pi}_{a_i}\hat{U}(t_1)|\psi\rangle}} \tag{2.14}$$

Then the probability of measuring at t_2 the value b_j for the observable B having obtained a_i at t_1, that is the conditional probability $P(B = b_j, t_2|A = a_i, t_1)$, is obtained by the Born rule expressed in Postulate III and amounts to [24, 25]

$$P(B = b_j, t_2|A = a_i, t_1) = \frac{\langle\psi|\hat{U}(t_1)^\dagger\hat{\Pi}_{a_i}\hat{U}(t_2,t_1)^\dagger\hat{\Pi}_{b_j}\hat{U}(t_2,t_1)\hat{\Pi}_{a_i}\hat{U}(t_1)|\psi\rangle}{\langle\psi|\hat{U}(t_1)^\dagger\hat{\Pi}_{a_i}\hat{U}(t_1)|\psi\rangle} \tag{2.15}$$

Again according to the Born rule, the denominator in the right hand side of (2.15) is the probability $P(A = a_i, t_1)$, so that bringing it to the left hand side we obtain the chain rule for the joint probability for measuring a_i at time t_1 and b_j at t_2

$$\begin{aligned} P(B = b_j, t_2; A = a_i, t_1) &= P(B = b_j, t_2|A = a_i, t_1)P(A = a_i, t_1) = \\ &= \langle\psi|\hat{U}(t_1)^\dagger\hat{\Pi}_{a_i}\hat{U}(t_2,t_1)^\dagger\hat{\Pi}_{b_j}\hat{U}(t_2,t_1)\hat{\Pi}_{a_i}\hat{U}(t_1)|\psi\rangle \end{aligned} \tag{2.16}$$

Actually (2.16) cannot be considered a proper joint probability distribution as it does not provide the correct marginal distribution for the outcomes of measurements of $\hat{B}$ at t_2, that is

$$\sum_i P(B = b_j, t_2; A = a_i, t_1) \neq P(B = b_j, t_2) \tag{2.17}$$

This inequality is a consequence of the difference in the processes that define the two sides. We have seen in fact that the terms in the sum of the left hand side of (2.17) accounts for the outcomes of measurements of $\hat{B}$ only after a measurement of $\hat{A}$ is performed, that is after a collapse of the state of the system has taken place, while the right hand side of the inequality does not consider any other action on the system before $\hat{B}$ is measured.
Actually, no consistent way of defining a proper joint probability distribution exists for non-commuting observables, as is discussed and shown in [31, 32, 33].

The dynamical correlation function that we would obtain by making direct subsequent measurements of $\hat{A}$ and $\hat{B}$ on a quantum system, is defined analogously to the classical dynamical distribution function (2.1) using the joint probability (2.16)

$$\begin{aligned}\mathscr{C}_{AB}(t_1,t_2) &= \sum_{i,j} a_i\, b_j \langle\psi|\hat{U}(t_1)^\dagger \hat{\Pi}_{a_i} \hat{U}(t_2,t_1)^\dagger \hat{\Pi}_{b_j} \hat{U}(t_2,t_1) \hat{\Pi}_{a_i} \hat{U}(t_1)|\psi\rangle = \\ &= \sum_i a_i \,\langle\psi|\hat{U}(t_1)^\dagger \hat{\Pi}_{a_i} \hat{U}(t_2,t_1)^\dagger \hat{B} \hat{U}(t_2,t_1) \hat{\Pi}_{a_i} \hat{U}(t_1)|\psi\rangle\end{aligned} \tag{2.18}$$

The quantity (2.18) will be called in the following projective correlation function. It accounts automatically for the measurement back action, and as such it does not contain the full information stored in the initial linear superposition representing the initial quantum state.

2.2.3 The full quantum dynamical correlations and their relationship with the pojective correlation function

The results cited in the introduction of this chapter, that is the ones about coherences of the electromagnetic fields and the linear response theory for thermodynamic systems, are actually not formulated in terms of correlations of the kind of (2.18). Results like the fluctuation dissipation theorem and the Kubo formula involve a different kind of correlation function which is defined as

$$C_{AB}(t_1,t_2) \equiv \langle\psi|\hat{U}(t_1)^\dagger \hat{A} \hat{U}(t_1) \hat{U}(t_2)^\dagger \hat{B} \hat{U}(t_2)|\psi\rangle \tag{2.19}$$

and to which we will refer as the quantum dynamical correlation function.

The argument that brought to the definition (2.18) of projective correlation function shows that one has to analyse carefully how dynamical correlations are probed, as the outcome depends dramatically on the experimental protocol.
The problem of a correct analysis of the measuring process is not only of interest when considering correlation measurements. Actually the systematization of the measuring process in quantum theory is a major issue in the debate on the foundations of the subject itself, and it is the main point on

which different interpretations of the theory differentiate [22, 21].
Except the foundational issues, a detailed modelling of the measurement back action on the system has also practical importance for the correct interpretation and design of experiments [26, 34].
In the context of dynamical correlation measurements, this problem has been addressed in [27], where the authors show that for a quantum system the measured dynamical correlation function does not correspond with (2.19).

The question then arise whether a kind of experiment able of measuring the quantum dynamical correlation (2.19) is conceivable.
In order to address this problem, we first have to investigate the relationship and differences between $\mathscr{C}_{AB}(t_1, t_2)$ and $C_{AB}(t_1, t_2)$, in such a way that we can understand why the direct measurement protocol described at the end of the previous subsection forbids the access to the quantum dynamical correlation function.
The most direct difference between these two quantities is that the projective correlation function is real valued, as can be immediately verified taking the complex conjugate of (2.18), whereas the quantum dynamical one is a complex valued quantity. Explicitly, the real and imaginary parts of the quantum dynamical correlation function are given in terms of anticommutator and commutator of the operators involved

$$\begin{aligned} \mathrm{Re}\, C_{AB}(t_1, t_2) &= \frac{1}{2}\langle\psi|\{\hat{U}(t_1)^\dagger \hat{A}\hat{U}(t_1), \hat{U}(t_2)^\dagger \hat{B}\hat{U}(t_2)\}|\psi\rangle \\ \mathrm{Im}\, C_{AB}(t_1, t_2) &= \frac{1}{2i}\langle\psi|[\hat{U}(t_1)^\dagger \hat{A}\hat{U}(t_1), \hat{U}(t_2)^\dagger \hat{B}\hat{U}(t_2)]|\psi\rangle \end{aligned} \tag{2.20}$$

The issue of the relationship between the projective and the quantum dynamical correlation functions has been addressed in [25] for the case in which the correlated observables $\hat{A}$ and $\hat{B}$ are two Cartesian components of the spin of two spin-1/2 particles. In that work, it has been found that for correlations among spin-1/2 components, the real part of the quantum dynamical correlation function equals the projective correlation function independently of the state of the system.
Then the real part of quantum dynamical correlation function between components of spin-1/2 observables is accessible via the direct measurement protocol, and it can be concluded that such real part is not affected from measurement back action.
In the same work it has been further asked if interpreting $\mathrm{Re}\, C_{AB}(t_1, t_2)$ as

the back action-immune part of quantum dynamical correlations is correct for generic systems and generic pairs of observables. The attempt to give an answer produced a mathematical condition that the correlated observables must satisfy in order to have $\mathrm{Re}\, C_{AB}(t_1,t_2) = \mathscr{C}_{AB}(t_1,t_2)$. In the following we give a derivation of this condition and, extending the result in [25], we show that it can only be satisfied by operators whose spectrum is made of just two eigenvalues with the same magnitude and opposite signs.

We start from the definition of quantum dynamical correlation function (2.19) and substitute in it the decomposition of $\hat{A}$ in the sum of its projectors

$$\hat{A} = \sum_i a_i \hat{\Pi}_{a_i} \tag{2.21}$$

and introduce the following resolution of the identity operator

$$1 = \hat{U}(t_1)^\dagger \Big(\sum_l \hat{\Pi}_{a_l} \Big) \hat{U}(t_1) \tag{2.22}$$

$C_{AB}(t_1,t_2)$ can then be re-expressed in the following way

$$\begin{aligned} C_{AB}(t_1,t_2) =& \langle\psi|\hat{U}(t_1)^\dagger \Big(\sum_i a_i \hat{\Pi}_{a_i} \Big) \hat{U}(t_2,t_1)^\dagger \hat{B} \hat{U}(t_2,t_1) \Big(\sum_l \hat{\Pi}_{a_l} \Big) \hat{U}(t_1)|\psi\rangle = \\ &= \sum_i a_i \langle\psi|U(t_1)^\dagger \hat{\Pi}_{a_i} \hat{U}(t_2,t_1)^\dagger \hat{B} \hat{U}(t_2,t_1) \hat{\Pi}_{a_i} \hat{U}(t_1)|\psi\rangle + \\ &+ \sum_{\substack{i,l \\ i\neq l}} a_i \langle\psi|\hat{U}(t_1)^\dagger \hat{\Pi}_{a_i} \hat{U}(t_2,t_1)^\dagger \hat{B} \hat{U}(t_2,t_1) \hat{\Pi}_{a_l} \hat{U}(t_1)|\psi\rangle \end{aligned} \tag{2.23}$$

The first term on the right hand side of the last identity is nothing else than the definition of $\mathscr{C}_{AB}(t_1,t_2)$, formula (2.18), whereas the second term

$$K_{AB}(t_1,t_2) \equiv \sum_{\substack{i,l \\ i\neq l}} a_i \langle\psi|\hat{U}(t_1)^\dagger \hat{\Pi}_{a_i} \hat{U}(t_2,t_1)^\dagger \hat{B} \hat{U}(t_2,t_1) \hat{\Pi}_{a_l} \hat{U}(t_1)|\psi\rangle \tag{2.24}$$

is a complex quantity.
The real and imaginary parts of $C_{AB}(t_1,t_2)$ can then be re-expressed in terms

of these two quantities

$$\begin{aligned} \mathrm{Re}\, C_{AB}(t_1,t_2) &= \mathscr{C}_{AB}(t_1,t_2) + \mathrm{Re}\, K_{AB}(t_1,t_2) \\ \mathrm{Im}\, C_{AB}(t_1,t_2) &= \mathrm{Im}\, K_{AB}(t_1,t_2) \end{aligned} \tag{2.25}$$

The identity $\mathrm{Re}\, C_{AB}(t_1,t_2) = \mathscr{C}_{AB}(t_1,t_2)$ that we are interested into, is satisfied when $\mathrm{Re}\, K_{AB}(t_1,t_2)$ vanishes. Using the explicit form of $K_{AB}(t_1,t_2)$ this condition amounts to

$$\begin{aligned} &\mathrm{Re}\, K_{AB}(t_1,t_2) = \\ &\quad = \sum_{\substack{i,l \\ i\neq l}} (a_i + a_l) \langle\psi|\hat{U}(t_1)^\dagger \hat{\Pi}_{a_i} \hat{U}(t_2,t_1)^\dagger \hat{B} \hat{U}(t_2,t_1) \hat{\Pi}_{a_l} \hat{U}(t_1)|\psi\rangle = 0 \end{aligned} \tag{2.26}$$

irrespective of the initial state $|\psi\rangle$ of the system.
It is convenient to expand the evolved state $\hat{U}(t_1)|\psi\rangle$ in formula (2.26) on the basis of eigenstates of $\hat{A}$

$$\hat{U}(t_1)|\psi\rangle = \sum_m c_m(t_1)|a_m\rangle \tag{2.27}$$

so that it is immediate to evaluate the action of the projectors $\hat{\Pi}_{a_l}$, $\hat{\Pi}_{a_i}$ on it

$$\begin{aligned} \hat{\Pi}_{a_l} \hat{U}(t_1)|\psi\rangle &= c_l(t_1)|a_l\rangle \\ \langle\psi|\hat{U}(t_1)^\dagger \hat{\Pi}_{a_i} &= c_i^*(t_1)\langle a_i| \end{aligned} \tag{2.28}$$

We obtain thus a new form of equation (2.26) by substitution of formulae (2.28) into it, that is

$$\sum_{\substack{i,l \\ i\neq l}} c_i^*(t_1) c_l(t_1)(a_i + a_l) \langle a_i|\hat{\Pi}_{a_i} \hat{U}(t_2,t_1)^\dagger \hat{B} \hat{U}(t_2,t_1) \hat{\Pi}_{a_l}|a_l\rangle = 0 \tag{2.29}$$

We are momentarily interested in solving equation (2.29) with respect to the observables alone, without any reference to the specific quantum state $|\psi\rangle$ the system is in. In turn the quantum state figures in the equation via the products of the expansion coefficients $c_i^*(t_1)c_l(t_1)$, that we will call

coherences, and then equation (2.29) must be valid whatever the value of these coefficients is, and this implies that

$$(1-\delta_{i,l})(a_i+a_l)\langle a_i|\hat{U}(t_1)^\dagger\hat{U}(t_2)\hat{B}\hat{U}(t_2)\hat{U}(t_1)^\dagger|a_l\rangle = 0 \qquad \forall\, i,l \tag{2.30}$$

The set of equations (2.30) is the condition for the observables that we were looking for. The set of equations has two kinds of solutions.
The first and most trivial is

$$\langle a_i|\hat{U}(t_1)\hat{U}(t_2)^\dagger\hat{B}\hat{U}(t_2)\hat{U}(t_1)^\dagger|a_l\rangle = 0 \qquad \text{for } i\neq l \tag{2.31}$$

At a closer look this equality means that $\hat{U}(t_2)^\dagger\hat{B}\hat{U}(t_2)$ is diagonal in the representation of eigenvectors of $\hat{U}(t_1)^\dagger\hat{A}\hat{U}(t_1)$, which equivalently means that these two operators commute.
In this case the full quantum dynamical correlation function is exactly equal to the projective one. This happens because the commutativity of $\hat{U}(t_1)^\dagger\hat{A}\hat{U}(t_1)$ with $\hat{U}(t_2)^\dagger\hat{B}\hat{U}(t_2)$ means that the measurement of $\hat{A}$ does not influence the later outcome for $\hat{B}$, and then the probability (2.16) is well defined and can be taken as the joint probability of measurement outcomes for the two observables.

The second and more interesting kind of solution is

$$a_i = -a_l \qquad \text{for } i\neq l \tag{2.32}$$

It is easy to verify that the above condition can be satisfied only if the whole spectrum of $\hat{A}$ is composed of two eigenvalues, which are equal in magnitude and have opposite signs.
The Cartesian components of spin-1/2 operators evidently satisfy this condition.

Next we investigate further the relationship between the projective and the quantum dynamical correlation function, asking under what conditions these two types of correlation functions are equal.
Looking back at formulae (2.25), this stricter condition of equality amounts to

$$K_{AB}(t_1,t_2) = 0 \tag{2.33}$$

We have found previously that already the vanishing of $\mathrm{Re}\, K_{AB}(t_1,t_2)$ independently of the state is possible for a very small set of observables. Insisting

on state-independency and imposing on this observables the additional condition $\mathrm{Im}\, K_{AB}(t_1, t_2) = 0$, we would find at best that an even smaller set of observables could satisfy equation (2.33).

For this reason we find more interesting to investigate condition (2.33) in light of the features of the quantum state of the system.
From formulae (2.24) and (2.28) we find that

$$K_{AB}(t_1, t_2) = \sum_{\substack{i,l \\ i \neq l}} a_i c_i^*(t_1) c_l(t_1) \langle a_i | \hat{U}(t_2, t_1)^\dagger \hat{B} \hat{U}(t_2, t_1) | a_l \rangle \tag{2.34}$$

The dependency of the last formula on the coherences $c_i^*(t_1)c_l(t_1)$, makes it explicit that the difference between $C_{AB}(t_1, t_2)$ and $\mathscr{C}_{AB}(t_1, t_2)$ is that the first one accounts for the fact that the system at t_1 can be in a linear superposition of eigenstates of $\hat{A}$, whereas the second cannot because it is defined considering the collapse of the state into only one of this eigenvalues provoked by the measurement at t_1.
In this sense $C_{AB}(t_1, t_2)$ is richer in information about the system than its projective counterpart, and the only way they can be equal is that the state of the system has evolved spontaneously at t_1 in an eigenstate of $\hat{A}$, as in this case the direct measurement of this observable would not affect the subsequent evolution of the state.
Mathematically this means that the expansion coefficients reduce to $c_l(t_1) = \delta_{il}$, and this indeed satisfies equation (2.33). If the state is described more generally by a density matrix, this last condition amounts to say that the system is in a statistical mixture of eigenstates of $\hat{A}$.

2.2.4 Two-time ancilla measurement to access the quantum dynamical correlation function

From the section 2.2.3 it is clear that if we wish to measure the full quantum dynamical correlation function $C_{AB}(t_1, t_2)$, we have to find a way of probing the system twice without spoiling the coherences of the state of the system in the basis of eigenstates of $\hat{A}$. Thus a direct coupling of the system with a measuring apparatus must be avoided.
Hereafter a measurement protocol based on an indirect coupling of the measuring devices to the system is discussed. This protocol is an abstraction of

the features of TDI technique and we will refer to this as two-time ancilla measurement.

The basic idea of two-time ancilla measurement is to use an auxiliary quantum system, the ancilla, which can interact weakly with the system for a finite time.
The ancilla is initially prepared in a linear superposition of states, in such a way to generate two parallel "histories" of the dynamics of composite system made up of the ancilla and our system. In each of these two histories the two subsystems begin to interact at a different instant for a finite time. Via the interaction, in each of the parallel history the ancilla extract information on the properties of the system around the time of interaction characteristic of that history.
Later on a direct measurement of an observable of the ancilla is made. The result of such measurement will depend on both the parallel histories of the composite system, thus containing information on the properties of the system at the two different interaction times.

In particular the result of the direct measurement on the ancilla depends on the quantum dynamical correlation function of the observables of the system involved in the interaction with the ancilla.
In the following we show the validity of this statement.
We call the states of the system and of the ancilla at the initial time $t = 0$ respectively $|\psi\rangle$ and $|\gamma\rangle$. Assuming that the two subsystems are initially uncorrelated the state of the composite system is just the product

$$|\Psi\rangle = |\psi\rangle|\gamma\rangle \tag{2.35}$$

The initial state of the ancilla is actually a linear superposition of two states

$$|\gamma\rangle = |\gamma_\alpha\rangle + |\gamma_\beta\rangle \tag{2.36}$$

and consequently (2.35) is also a superposition

$$|\Psi\rangle = |\psi\rangle|\gamma_\alpha\rangle + |\psi\rangle|\gamma_\beta\rangle \tag{2.37}$$

Now if the ancilla is in the state $|\gamma_\alpha\rangle$ it interacts with the system from time t_α to time t_1, whereas if it is in $|\gamma_\beta\rangle$ the interaction takes place from $t_\beta > t_1$ to t_2.

The evolution operator that rules the dynamic of the composite system during these two interaction intervals, that we indicate by $\hat{U}_I$ is generated by the interaction hamiltonian

$$\hat{H}_I = g\hat{A} \otimes \hat{R} \tag{2.38}$$

where $\hat{R}$ is an observable of the ancilla and $\hat{A}$ is the observable of the system whose dynamical correlation will be tested via this scheme[2]. Before, in between and after the interaction intervals the hamiltonian of the composite system is instead the sum of the free hamiltonians of the system and the ancilla. This implies that, in this interaction-free intervals, the two subsystems evolve independently from each other according to the free evolution operator $\hat{U}_0$.
The evolution of the state of composite system can thus be split in four steps:

1. Free evolution for $0 \leq t \leq t_\alpha$

$$|\psi\rangle|\gamma_\alpha\rangle + |\psi\rangle|\gamma_\beta\rangle \to \hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\beta\rangle \tag{2.39}$$

2. Interaction in the first branch for $t_\alpha \leq t \leq t_1$

$$\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\beta\rangle \to \hat{U}_I(t_1, t_\alpha)\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \hat{U}_0(t_1)|\psi\rangle|\gamma_\beta\rangle \tag{2.40}$$

3. Free evolution for $t_1 \leq t \leq t_\beta$

$$\begin{aligned} &\hat{U}_I(t_1, t_\alpha)\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \hat{U}_0(t_1)|\psi\rangle|\gamma_\beta\rangle \to \\ &\quad \to \hat{U}_0(t_\beta, t_1)\hat{U}_I(t_1, t_\alpha)\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \hat{U}_0(t_\beta)|\psi\rangle|\gamma_\beta\rangle \end{aligned} \tag{2.41}$$

4. Interaction in the second branch $t_\beta \leq t \leq t_2$

$$\begin{aligned} &\hat{U}_0(t_\beta, t_1)\hat{U}_I(t_1, t_\alpha)\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \hat{U}_0(t_\beta)|\psi\rangle|\gamma_\beta\rangle \to \\ &\quad \to \hat{U}_0(t_2, t_1)\hat{U}_I(t_1, t_\alpha)\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \hat{U}_I(t_2, t_\beta)\hat{U}_0(t_\beta)|\psi\rangle|\gamma_\beta\rangle \end{aligned} \tag{2.42}$$

[2]For the sake of simplicity the form of the interaction hamiltonian has been taken as a simple product of observables. The more general case is $H_I = g\sum_k \hat{A}_k \otimes \hat{R}_k$ and we will see that this is the case in TDI, and the Fourier transform of dynamical correlation function is obtained rather than the function itself.

For $t \geq t_2$ the interaction does not contribute anymore to the dynamics and the evolution of the composite system is free again.

The assumption of weak coupling of the ancilla means that the coupling parameter g in the interaction hamiltonian (2.38) is small enough to allow a perturbative evaluation of $\hat{U}_I$ (see appendix A).
Thus the state at a time $t > t_4$ is

$$\begin{aligned}|\Psi(t)\rangle =& \hat{U}_0(t)|\Psi\rangle + \\ &- \frac{ig}{\hbar}\hat{U}_0(t,t_\alpha)\left[\int_{t_\alpha}^{t_1} dt'\, \hat{U}_0(t',t_\alpha)^\dagger \left(\hat{A}\otimes\hat{R}\right)\hat{U}_0(t',t_\alpha)\right]\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle + \\ &- \frac{ig}{\hbar}\hat{U}_0(t,t_\beta)\left[\int_{t_\beta}^{t_2} dt'\, \hat{U}_0(t',t_\beta)^\dagger \left(\hat{A}\otimes\hat{R}\right)\hat{U}_0(t',t_\beta)\right]\hat{U}_0(t_\beta,0)|\psi\rangle|\gamma_\beta\rangle\end{aligned} \tag{2.43}$$

The last two terms in (2.43) can be usefully rewritten exploiting the unitarity of the free evolution operator

$$\begin{aligned}|\delta\Psi_\alpha(t)\rangle \equiv -\frac{ig}{\hbar}\hat{U}_0(t,t_\alpha)\int_{t_\alpha}^{t_1} dt' \left[\hat{U}_0(t',t_\alpha)\hat{U}_0(t_\alpha)\hat{U}_0(t_\alpha)^\dagger\right]^\dagger \left(\hat{A}\otimes\hat{R}\right) \\ \left[\hat{U}_0(t',t_\alpha)\hat{U}_0(t_\alpha)\hat{U}_0(t_\alpha)^\dagger\right]\hat{U}_0(t_\alpha)|\psi\rangle|\gamma_\alpha\rangle = \\ = -\frac{ig}{\hbar}\hat{U}_0(t,0)\left[\int_{t_\alpha}^{t_1} dt'\, \hat{U}_0(t')^\dagger \left(\hat{A}\otimes\hat{R}\right)\hat{U}_0(t')\right]|\psi\rangle|\gamma_\alpha\rangle\end{aligned} \tag{2.44}$$

$$\begin{aligned}|\delta\Psi_\beta(t)\rangle \equiv -\frac{ig}{\hbar}\hat{U}_0(t,t_\beta)\int_{t_\beta}^{t_2} dt' \left[\hat{U}_0(t',t_\beta)\hat{U}_0(t_\beta)\hat{U}_0(t_\beta)^\dagger\right]^\dagger \left(\hat{A}\otimes\hat{R}\right) \\ \left[\hat{U}_0(t',t_\beta)\hat{U}_0(t_\beta)\hat{U}_0(t_\beta)^\dagger\right]\hat{U}_0(t_\beta)|\psi\rangle|\gamma_\alpha\rangle = \\ = -\frac{ig}{\hbar}\hat{U}_0(t)\left[\int_{t_\beta}^{t_2} dt'\, \hat{U}_0(t')^\dagger \left(\hat{A}\otimes\hat{R}\right)\hat{U}_0(t')\right]|\psi\rangle|\gamma_\beta\rangle\end{aligned} \tag{2.45}$$

If $\hat{W}$ is the observable on the ancilla that we measure after the interaction with the system, the average value for such an observable at $t > t_2$ will be

determined by the scalar product $\langle\Psi(t)|(1\otimes\hat{W})|\Psi(t)\rangle$. Looking at expression (2.43) for the the evolved state $|\Psi(t)\rangle$, we realize that the expectation value of $\hat{W}$ is the sum of different contributions at different orders of the coupling constant g:

- 0th order
$$\langle\Psi|\hat{U}_0(t,0)^\dagger(1\otimes\hat{W})\hat{U}_0(t,0)|\Psi\rangle \tag{2.46}$$

- 1st order
$$\langle\Psi|\hat{U}_0(t,0)^\dagger(1\otimes\hat{W})|\delta\Psi_\alpha(t)\rangle+\langle\Psi|\hat{U}_0(t,0)^\dagger(1\otimes\hat{W})|\delta\Psi_\beta(t)\rangle+c.c. \tag{2.47}$$

- 2nd order
$$\begin{aligned}&\langle\delta\Psi_\alpha(t)|(1\otimes\hat{W})|\delta\Psi_\alpha(t)\rangle+\langle\delta\Psi_\beta(t)|(1\otimes\hat{W})|\delta\Psi_\beta(t)\rangle+\\&+\langle\delta\Psi_\alpha(t)|(1\otimes\hat{W})|\delta\Psi_\beta(t)\rangle+\langle\delta\Psi_\beta(t)|(1\otimes\hat{W})|\delta\Psi_\alpha(t)\rangle\end{aligned} \tag{2.48}$$

It will be shown that the operators $\hat{W}$ and $\hat{R}$, and the choice of the initial state of the ancilla specific to the TDI technique make the 0th and 1st order terms negligible, so that only the 2nd order terms are necessary for the determination of the average value of W. Substitution of (2.44) into (2.48) gives

$$\begin{aligned}\langle W\rangle(t)\simeq\frac{g^2}{\hbar^2}\Bigg\{&\int_{t_\alpha}^{t_1}\int_{t_\alpha}^{t_1}dt'dt''\,\langle\psi|\hat{A}(t')\hat{A}(t'')|\psi\rangle\langle\gamma_\alpha|\hat{R}(t')\hat{W}(t)\hat{R}(t'')|\gamma_\alpha\rangle+\\&+\int_{t_\beta}^{t_2}\int_{t_\beta}^{t_\alpha}dt'dt''\,\langle\psi|\hat{A}(t')\hat{A}(t'')|\psi\rangle\langle\gamma_\beta|\hat{R}(t')\hat{W}(t)\hat{R}(t'')|\gamma_\beta\rangle+\\&+2\mathrm{Re}\Bigg[\int_{t_\alpha}^{t_1}\int_{t_\beta}^{t_2}dt'dt''\,\langle\psi|\hat{A}(t')\hat{A}(t'')|\psi\rangle\langle\gamma_\alpha|\hat{R}(t')\hat{W}(t)\hat{R}(t'')|\gamma_\beta\rangle\Bigg]\Bigg\}\end{aligned} \tag{2.49}$$

In the last formula we adopted for simplicity the notation $\hat{O}(t)=\hat{U}_0^\dagger(t)\hat{O}\hat{U}_0(t)$ for the operators.
If the target free dynamics is slower than the ancilla free dynamics, the correlation function $\langle\psi|\hat{A}(t')\hat{A}(t'')|\psi\rangle$ can be evaluated at one of the two

edges of the time integration intervals and brought out of it, in such a way that (2.49) becomes

$$\begin{aligned}\langle W\rangle(t) \simeq \frac{g^2}{\hbar^2}\Bigg\{ & C_{AA}(t_1,t_1)\int_{t_\alpha}^{t_1}\int_{t_\alpha}^{t_1} dt'dt''\,\langle\gamma_\alpha|\hat{R}(t')\hat{W}(t)\hat{R}(t'')|\gamma_\alpha\rangle + \\ & + C_{AA}(t_2,t_2)\int_{t_\beta}^{t_2}\int_{t_\beta}^{t_2} dt'dt''\,\langle\gamma_\beta|\hat{R}(t')\hat{W}(t)\hat{R}(t'')|\gamma_\beta\rangle + \\ & + 2\,\mathrm{Re}\big[C_{AA}(t_1,t_2)\big]\,\mathrm{Re}\Bigg[\int_{t_\alpha}^{t_1}\int_{t_\beta}^{t_2} dt'dt''\,\langle\gamma_\alpha|\hat{R}(t')\hat{W}(t)\hat{R}(t'')|\gamma_\beta\rangle\Bigg] + \\ & - 2\,\mathrm{Im}\big[C_{AA}(t_1,t_2)\big]\,\mathrm{Im}\Bigg[\int_{t_\alpha}^{t_1}\int_{t_\beta}^{t_2} dt'dt''\,\langle\gamma_\alpha|\hat{R}(t')\hat{W}(t)\hat{R}(t'')|\gamma_\beta\rangle\Bigg]\Bigg\}\end{aligned} \tag{2.50}$$

This last formula tells us that from repeated measurement on the ancilla system we can retrieve the dynamical quantum correlation function of the observable $\hat{A}$ through which the system couples to the ancilla.
We will see in chapter 4 how the result (2.50) can be firmly established for the TDI technique. Moreover, in that case the assumption of a slow dynamical variation of the internal dynamics of the target is not strictly necessary.

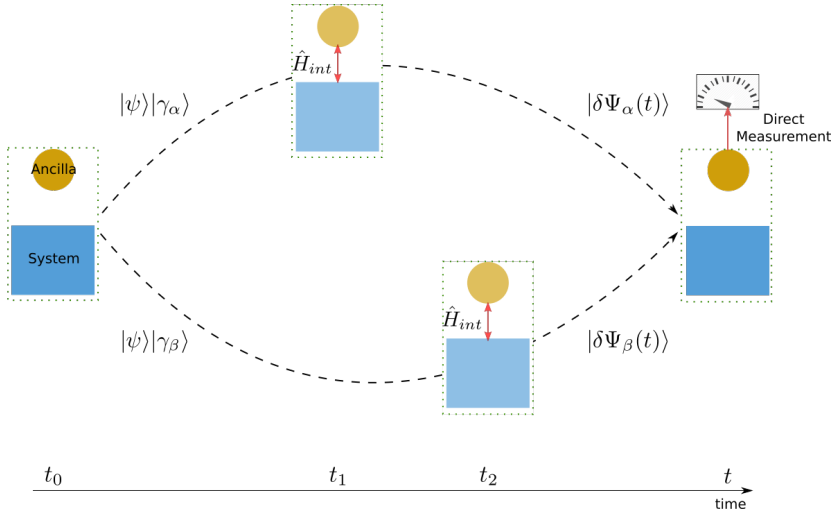

Figure 2.2: Schematic representation of the two-time ancilla measurement. The dashed black lines represent the two possible histories of the ancilla and the system under their mutual interaction. In the upper history the ancilla interacts with the system at time t_1 generating the contribution $|\delta\Psi_\alpha(t)\rangle$ to the final state of the composite system, which depend on the $\hat{A}(t_1)$. Similarly in the second history the interaction takes place at t_2 bringing in the final state $\hat{A}(t_2)$-dependent contribution $|\delta\Psi_\beta(t)\rangle$. The direct measurement on the ancilla mixes the two contributions, so that the outcome will depend on the quantum dynamical correlation function for $\hat{A}$ evaluated at the two interaction times.

Differently from the the measurement of correlation described in section 2.2.2, where one truly has to act on the system twice, in the two-time ancilla measurement there is only one intervention on the system by the ancilla but it cannot be stated when this intervention takes place.
Indeed the form of the final state (2.43), neglecting the zero-th order contribution, can be interpreted stating that either the ancilla has probed the system at t_1 or it has probed the system at t_2 but this two possible events do not mutually influence.
As suggested by figure 2.2, the two perturbative contributions $|\delta\Psi_\alpha(t)\rangle$ and $|\delta\Psi_\beta(t)\rangle$ to the final state of the composite system can be thought as two parallel histories, in each of which the ancilla has probed the system at a different time.
The final measurement on the ancilla does not distinguish the two histories, therefore the the outcome of it will depend on both the histories, that is on the properties of the system at the two interaction times.

Chapter 3

The Dynamical couple correlation function and its Fourier transforms

This chapter is dedicated to the dynamical couple correlation function of an ensemble of particles (DCF), and its Fourier transforms.
In this chapter we give the generic definitions for the DCF, the ISF and the dynamic structure factor which are valid irrespective of whether the system is a classical or a quantum one. Then we show that if a system is classical, these functions enjoy some symmetry properties, and that this same symmetries can be violated if the system is quantum mechanical as a consequence of a non-vanishing imaginary part of the DCF.

This fact is of big relevance for TDI or inealstic x-ray or neutron scattering experiments. Indeed if an experimenter finds that the above mentioned symmetries of the ISF or of the dynamic structure factor are violated, he must conclude that the dynamics of the particles in the system is ruled by quantum mechanical laws. It is then legit to ask if, on the contrary, it can concluded that a classical model for the particles of a system is sufficient when these functions are experimentally found to be symmetric.
We answer this question in the second part of the chapter by analysing two model systems of quantum particles hopping between the minima of some external potential. There we find that the particles can be in a state that is a quantum superposition of different position configurations, but, despite this, give a DCF and an ISF which are symmetric like the ones of a classical

system.
These results for the first of the two model systems have been presented in [30].

3.1 The definition and properties of the DCF and its Fourier transforms

The DCF for a system of particles enclosed in a volume V is the correlation function between the densities of particles at different points in the system at different times, averaged over its whole volume.
Calling the density of particles at a given point and time $\rho(\mathbf{r}, t)$, the definition of the DCF is[1]

$$G(\mathbf{r}, t_1, t_2) \equiv \int_V d^3r' \, \langle \rho(\mathbf{r}', t_1)\rho(\mathbf{r}' + \mathbf{r}, t_2)\rangle \tag{3.1}$$

where the angular brackets have the meaning of a statistical average over different possible configurations of particles in the system, and therefore they have to be intended as an ensemble average for a classical system and as an expectation value for a quantum system.

The DCF tells us how likely is to have a particle somewhere in the target at time t_1 and to have a second particle at a distance $\mathbf{r}$ from the first at a later time t_2.

From the DCF we can define the ISF and the dynamic structure factor. The ISF is the spatial Fourier transform of $G(\mathbf{r}, t_1, t_2)$

$$S(\mathbf{Q}, t_1, t_2) \equiv \int_V d^3r \, G(\mathbf{r}, t_1, t_2)e^{-i\mathbf{Q}\cdot\mathbf{r}} \tag{3.2}$$

The ISF represents the degree of correlations between two particles on a length-scale $\lambda = 2\pi/|\mathbf{Q}|$ along the direction $\mathbf{Q}/|\mathbf{Q}|$.

The dynamic structure factor is usually defined for systems in states whose statistical properties do not depend on time, like e.g. systems in

[1] Many different normalizations of the DCF are adopted in the scientific literature. Here no special normalization is used in order not to make the notation cumbersome

thermal equilibrium [35].
The stationarity condition implies that the DCF, and as a consequence the ISF, do not depend on the two instants t_1 and t_2 separately but only on their difference $\tau = t_2 - t_1$. When such a condition holds, it is possible to define the space-time Fourier transform of the DCF as

$$S(\mathbf{Q}, \omega) \equiv \int_{-\infty}^{+\infty} d\tau \, S(\mathbf{Q}, \tau) e^{i\omega\tau} \tag{3.3}$$

3.1.1 The properties of the DCF, the ISF and the dynamic structure factor for classical systems

We know that dynamical correlation functions for classical systems are real valued quantities (see chapter 2), so that the same can be said of the DCF. From definition (3.1) of the DCF, it is easy to verify by the change of variables $\mathbf{r}' \longrightarrow \mathbf{r}' - \mathbf{r}$ that the following relation holds

$$G^{(cl)}(\mathbf{r}, t_1, t_2) = \int_V d^3r' \langle \rho(\mathbf{r}', t_2) \rho(\mathbf{r}' - \mathbf{r}, t_1) \rangle = G^{(cl)}(-\mathbf{r}, t_2, t_1) \tag{3.4}$$

The fact that $G^{(cl)}(\mathbf{r}, t_1, t_2)$ is real valued and that it enjoys the inversion symmetry (3.4), implies that the classical ISF satisfies two identities.
From the reality condition and the definition (3.2) it follows that the classical ISF is an hermitian function with respect to the variable $\mathbf{Q}$, that is its complex conjugate its equal to the original function evaluated at the inverse value $-\mathbf{Q}$, indeed we have

$$S^{(cl)*}(\mathbf{Q}, t_1, t_2) = \int_V d^3r \, G(\mathbf{r}, t_1, t_2) e^{i\mathbf{Q}\cdot\mathbf{r}} \equiv S^{(cl)}(-\mathbf{Q}, t_1, t_2) \tag{3.5}$$

which translates on its real and imaginary parts as

$$\mathrm{Re}\, S^{(cl)}(\mathbf{Q}, t_1, t_2) = \mathrm{Re}\, S^{(cl)}(-\mathbf{Q}, t_1, t_2) \tag{3.6}$$

$$\mathrm{Im}\, S^{(cl)}(\mathbf{Q}, t_1, t_2) = -\mathrm{Im}\, S^{(cl)}(-\mathbf{Q}, t_1, t_2) \tag{3.7}$$

From identity (3.4) instead it follows that

$$\begin{aligned} S^{(cl)}(\mathbf{Q}, t_1, t_2) &= \int_V d^3r \, G(-\mathbf{r}, t_2, t_1) e^{-i\mathbf{Q}\cdot\mathbf{r}} = \\ &= \int_V d^3r' \, G(\mathbf{r}', t_2, t_1) e^{i\mathbf{Q}\cdot\mathbf{r}'} \equiv S^{(cl)}(-\mathbf{Q}, t_2, t_1) \end{aligned} \tag{3.8}$$

that is the classical ISF is symmetric with respect to the inversion of $\mathbf{Q}$ and the exchange of the time arguments.
Moreover combining identities (3.5) and (3.8) we obtain that the ISF for classical systems is also hermitian with respect to the exchange of time arguments, that is

$$S^{(cl)*}(\mathbf{Q}, t_1, t_2) = S^{(cl)}(\mathbf{Q}, t_2, t_1) \tag{3.9}$$

The symmetry conditions (3.4) and (3.8) for stationary systems become

$$G^{(cl)}(\mathbf{r}, \tau) = G^{(cl)}(-\mathbf{r}, -\tau) \tag{3.10}$$

$$S^{(cl)}(\mathbf{Q}, \tau) = S^{(cl)}(-\mathbf{Q}, -\tau) \tag{3.11}$$

which imply that the dynamic structure factor must satisfy the following conditions of reality and symmetry

$$S^{(cl)*}(\mathbf{Q}, \omega) = S^{(cl)}(\mathbf{Q}, \omega) \tag{3.12}$$

$$S^{(cl)}(\mathbf{Q}, \omega) = S^{(cl)}(-\mathbf{Q}, -\omega) \tag{3.13}$$

The dynamic structure factor represent the excitation spectrum of the particles density in the system, e.g. the spectrum of phonon excitations in a crystalline solid in thermal equilibrium at a given temperature [6, 36].

3.1.2 The properties of the DCF, the ISF and the dynamic structure factor for quantum systems

The DCF of a quantum system takes complex values. As a consequence not all the symmetries for the ISF and the dynamic structure factor shown for classical systems hold in this case.
The DCF of a quantum system in the initial state $|\psi\rangle$ is [37]

$$G^{(qu)}(\mathbf{r}, t_1, t_2) \equiv \int_V d^3r' \, \langle\psi|\hat{U}(t_1)^\dagger \hat{\rho}(\mathbf{r}')\hat{U}(t_1)\hat{U}(t_2)^\dagger \hat{\rho}(\mathbf{r}' + \mathbf{r})\hat{U}(t_2)|\psi\rangle \tag{3.14}$$

with the particle-density operator being a function of the position operators of all the N particles $\hat{\mathbf{r}}_1, \ldots \hat{\mathbf{r}}_N$

$$\hat{\rho}(\mathbf{r}) = \sum_{j=1}^{N} \delta(\mathbf{r} - \hat{\mathbf{r}}_j) \tag{3.15}$$

The $G^{(qu)}(\mathbf{r}, t_1, t_2)$ enjoys the hermiticity property with respect to distance and times, that is its complex conjugate is equal to the function itself evaluated at $-\mathbf{r}$ and at times exchanged. In fact taking the complex conjugate of (3.14) we get

$$G^{(qu)*}(\mathbf{r}, t_1, t_2) = \int_V d^3r' \, \langle\psi|\hat{U}(t_2)^\dagger \hat{\rho}(\mathbf{r}' + \mathbf{r})\hat{U}(t_2)\hat{U}(t_1)^\dagger \hat{\rho}(\mathbf{r}')\hat{U}(t_1)|\psi\rangle =$$
$$= \int_V d^3r'' \, \langle\psi|\hat{U}(t_2)^\dagger \hat{\rho}(\mathbf{r}'')\hat{U}(t_2)\hat{U}(t_1)^\dagger \hat{\rho}(\mathbf{r}'' - \mathbf{r})\hat{U}(t_1)|\psi\rangle \equiv G^{(qu)}(-\mathbf{r}, t_2, t_1) \tag{3.16}$$

This hermiticity condition endows the quantum ISF with the same hermitian symmetry (3.9) with respect to times exchange that holds for classical systems

$$S^{(qu)*}(\mathbf{Q}, t_1, t_2) = S^{(qu)}(\mathbf{Q}, t_2, t_1) \tag{3.17}$$

but conversely to the classical case, the presence of an imaginary part in the DCF invalidates the identity (3.5), so that in general for quantum systems we have

$$S^{(qu)*}(\mathbf{Q}, t_1, t_2) \neq S^{(qu)}(-\mathbf{Q}, t_1, t_2) \tag{3.18}$$

In the stationary case, the hermiticity condition on the quantum ISF (3.17) still allows for the definition of a real valued dynamic structure factor, but the inequality (3.18) spoils the inversion symmetry (3.13) that the dynamic structure factor enjoys in the classical case, so that for quantum systems one in general has that

$$S^{(qu)}(\mathbf{Q}, \omega) \neq S^{(qu)}(-\mathbf{Q}, -\omega) \tag{3.19}$$

3.1.3 The experimental relevance of the symmetry properties of the DCF and its transforms

The different properties and symmetries stated for the DCF of classical and quantum systems and its Fourier transforms are summed up in the following

table

	Classical	Quantum
DCF	$G^{cl}(\mathbf{r}, t_1, t_2) \in \mathbb{R}$ $G^{cl}(\mathbf{r}, t_1, t_2) = G^{cl}(-\mathbf{r}, t_2, t_1)$	$G^{qu}(\mathbf{r}, t_1, t_2) \in \mathbb{C}$ $G^{qu*}(\mathbf{r}, t_1, t_2) = G^{qu}(-\mathbf{r}, t_2, t_1)$
ISF	$S^{cl*}(\mathbf{Q}, t_1, t_2) = S^{cl}(-\mathbf{Q}, t_1, t_2)$ $S^{cl}(\mathbf{Q}, t_1, t_2) = S^{cl}(-\mathbf{Q}, t_2, t_1)$ $S^{cl*}(\mathbf{Q}, t_1, t_2) = S^{cl}(\mathbf{Q}, t_2, t_1)$	$S^{qu*}(\mathbf{Q}, t_1, t_2) \neq S^{qu}(-\mathbf{Q}, t_1, t_2)$ $S^{qu*}(\mathbf{Q}, t_1, t_2) = S^{qu}(\mathbf{Q}, t_2, t_1)$
Dynamical structure factor	$S^{cl}(\mathbf{Q}, \omega) \in \mathbb{R}$ $S^{cl}(\mathbf{Q}, \omega) = S^{cl}(-\mathbf{Q}, -\omega)$	$S^{qu}(\mathbf{Q}, \omega) \in \mathbb{R}$ $S^{qu}(\mathbf{Q}, \omega) \neq S^{qu}(-\mathbf{Q}, -\omega)$

In particular we have seen that the fact that $G^{(cl)}$ is real valued implies that

$S^{(cl)}$ is hermitian with respect to the momentum and this in turn implies that the dynamic structure factor is symmetric with respect to inversion of momentum and frequency.
Actually, also the opposite statements are valid. Indeed if the ISF is hermitian with respect to the momentum, it follows that

$$\begin{aligned} G^{(cl)}(\mathbf{r}, t_1, t_2) = \frac{1}{(2\pi)^3} \int_V d^3Q \, S^{(cl)*}(-\mathbf{Q}, t_1, t_2) e^{i\mathbf{Q}\cdot\mathbf{r}} = \\ = \frac{1}{(2\pi)^3} \left(\int_V d^3Q' \, S^{(cl)}(\mathbf{Q}', t_1, t_2) e^{i\mathbf{Q}'\cdot\mathbf{r}} \right)^* = \\ = G^{(cl)*}(\mathbf{r}, t_1, t_2) \end{aligned} \tag{3.20}$$

Whereas, in the case of stationary system, a symmetric dynamic structure factor implies an hermitian ISF

$$S^{(cl)}(\mathbf{Q},\tau) = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega\, S^{(cl)}(-\mathbf{Q},-\omega)e^{-i\omega\tau} = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega'\, S^{(cl)}(-\mathbf{Q},\omega')e^{i\omega'\tau} =$$

$$= \frac{1}{2\pi}\left(\int_{-\infty}^{\infty} d\omega\, S^{(cl)}(-\mathbf{Q},\omega)e^{i\omega\tau}\right)^{*} = S^{(cl)*}(-\mathbf{Q},\tau) \tag{3.21}$$

Thus if in an experiment on a system it is found that the ISF is non-hermitian, or that the dynamic structure factor is not symmetric, then it can be stated that the corresponding DCF is complex valued, that is that a classical model for the particles is not sufficient to describe their correlations and that a quantum mechanical model instead is necessary.
In particular the need of taking into account quantum corrections to the dynamic structure factor in order to correctly fit the data from inelastic x-ray and neutron scattering experiments, has been recognized in [38, 39, 40], and measurements of the asymmetry of such function have been performed for different liquid systems in thermal equilibrium at low temperatures, e.g. for liquid Neon [41], liquid Hydrogen and Deuterium [42].
In particular, for a generic quantum system in thermal equilibrium it can be shown that the dynamic structure factor satisfies the identity [43]

$$S^{(qu)}(\mathbf{Q},\omega) = e^{\frac{\hbar\omega}{k_B T}} S^{(qu)}(-\mathbf{Q},-\omega) \tag{3.22}$$

which is evidently non-symmetrical with respect to inversion of the sign of $\mathbf{Q}$ and ω.

Relation (3.22) provides us with an estimate of the temperature scales at which we shall expect quantum effects to show up in the dynamic structure factor.
If the temperature of a system is of order $T \sim o(\hbar\omega/k_B)$, the asymmetric character of the dynamic structure factor is not negligible. If we consider the temperature of the system as fixed, the last condition means that the asymmetry of the dynamic structure factor is more pronounced for frequencies $\omega \geq k_B T/\hbar$. This also means that in the time domain we should expect quantum effects to be non negligible on a time-scale $\tau \simeq \hbar/(k_B T)$, that is

on such time-scales one should expect the ISF to be non-hermitian and the DCF to be complex-valued.

Such an easy estimate of the time-scales of quantum features of the DCF and ISF is not possible for systems out of thermal equilibrium. An experimental assessment of this time-scale could then help in the understanding of quantum effects in non-equilibrium physics.

In the next chapter we discuss the cases of two quantum model systems which admit states with no classical features, being given by superpositions of other states, that lead to real valued DCF. This means that if in an experiment on a system it is found that its ISF is hermitian, or that its dynamic structure factor is symmetric, it cannot be stated that the particles in the system are classical only on the basis of such experiment.

3.2 Model systems

3.2.1 One particle in a 1D Double Well potential

The first model we consider is the minimal constituent block of more general quantum systems, that is a single particle in a double well potential.

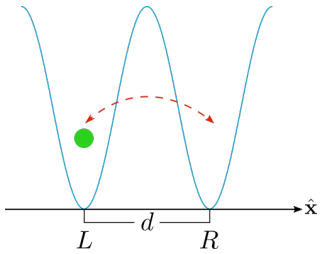

Figure 3.1: Particle trapped in a double well potential

In a simplified description of this system, we assume that the particle subject to the double-well potential can only be found at two positions that is, with reference to figure 3.1, either the left or the right minimum of the potential which lie at a mutual distance d.
These two possible configurations of the particle, corresponding to two mutually orthogonal quantum states $|L\rangle$ and $|R\rangle$ with L and R standing for

"left" and "right", represent all the possibilities for the system. These two quantum states then constitute a basis for the Hilbert space of the particle.

Assuming that the particle can hop from one position to the other, the hamiltonian of the system can be represented in the $\{|L\rangle, |R\rangle\}$ basis as

$$\hat{H} = -\frac{\hbar\tilde{\omega}}{2}(|L\rangle\langle R| + |R\rangle\langle L|) \tag{3.23}$$

Adapting the definition (3.14) for the case of a one-dimensional system in which positions of particles can only take discrete values, the DCF for the particle in the double well is defined as

$$G_{DW}(x, t_1, t_2) =$$

$$= \begin{cases} \langle\psi|\hat{U}(t_1)^\dagger\hat{\rho}(L)\hat{U}(t_1)\hat{U}(t_2)^\dagger\hat{\rho}(R)\hat{U}(t_2)|\psi\rangle & \text{if} \quad x = d \\ \\ \langle\psi|\hat{U}(t_1)^\dagger\hat{\rho}(R)\hat{U}(t_1)\hat{U}(t_2)^\dagger\hat{\rho}(L)\hat{U}(t_2)|\psi\rangle & \text{if} \quad x = -d \\ \\ \langle\psi|\hat{U}(t_1)^\dagger\hat{\rho}(L)\hat{U}(t_1)\hat{U}(t_2)^\dagger\hat{\rho}(L)\hat{U}(t_2)|\psi\rangle + \\ + \langle\psi|\hat{U}(t_1)^\dagger\hat{\rho}(R)\hat{U}(t_1)\hat{U}(t_2)^\dagger\hat{\rho}(R)\hat{U}(t_2)|\psi\rangle & \text{if} \quad x = 0 \end{cases} \tag{3.24}$$

From the definition (3.15) for the particle density operator, we can find the representations of $\hat{\rho}(L)$ and $\hat{\rho}(R)$ in the left/right basis

$$\begin{aligned} \hat{\rho}(L) &= \delta_{L,L}|L\rangle\langle L| + \delta_{L,R}|R\rangle\langle R| = |L\rangle\langle L| \\ \hat{\rho}(R) &= \delta_{R,L}|L\rangle\langle L| + \delta_{R,R}|R\rangle\langle R| = |R\rangle\langle R| \end{aligned} \tag{3.25}$$

whereas the evolution operator corresponding to the hamiltonian (3.23) is

$$\hat{U}(t) = e^{-i\frac{\hat{H}}{\hbar}t} = 1\,\cos\left(\frac{\tilde{\omega}}{2}t\right) + i\big(|L\rangle\langle R| + |R\rangle\langle L|\big)\sin\left(\frac{\tilde{\omega}}{2}t\right) \tag{3.26}$$

Having at hand the explicit forms of all the factors appearing in (3.24), the DCF can be calculated for any initial state of the particle.
The generic initial state of the particle at time $t = 0$ is

$$|\psi\rangle = c_L|L\rangle + c_R|R\rangle \tag{3.27}$$

where the complex coefficients c_L and c_R satisfy the normalization condition $|c_L|^2 + |c_R|^2 = 1$, but are otherwise arbitrary.
The explicit form of the DCF of the particle is

$$G_{DW}(x, t_1, t_2) =$$

$$= \begin{cases} \sin^2\left[\dfrac{\tilde{\omega}(t_2 - t_1)}{2}\right] P(L, t_1) + \dfrac{1}{2}\sin\left[\tilde{\omega}(t_2 - t_1)\right] \operatorname{Im}\Gamma(t_1) + \\ \qquad + \dfrac{i}{2}\sin\left[\tilde{\omega}(t_2 - t_1)\right] \operatorname{Re}\Gamma(t_1) \quad \text{if} \quad x = d \\ \sin^2\left[\dfrac{\tilde{\omega}(t_2 - t_1)}{2}\right] P(R, t_1) - \dfrac{1}{2}\sin\left[\tilde{\omega}(t_2 - t_1)\right] \operatorname{Im}\Gamma(t_1) + \\ \qquad + \dfrac{i}{2}\sin\left[\tilde{\omega}(t_2 - t_1)\right] \operatorname{Re}\Gamma(t_1) \quad \text{if} \quad x = -d \\ \cos\left[\dfrac{\tilde{\omega}(t_2 - t_1)}{2}\right] - i\sin\left[\tilde{\omega}(t_2 - t_1)\right] \operatorname{Re}\Gamma(t_1) \quad \text{if} \quad x = 0 \end{cases} \tag{3.28}$$

Here $P(L, t)$ and $P(R, t)$ are the probabilities at time t for the particle to be at the left or right minimum, and amount to

$$\begin{aligned} P(L, t) &\equiv |\langle L|\hat{U}(t_1)|\psi\rangle|^2 = \\ &= \frac{1}{2}\left[1 + (|c_L|^2 - |c_R|^2)\cos(\tilde{\omega}t)\right] + \operatorname{Im}(c_L^* c_R)\,\sin(\tilde{\omega}t) \end{aligned} \tag{3.29}$$

$$\begin{aligned} P(R, t) &\equiv |\langle R|\hat{U}(t)|\psi\rangle|^2 = \\ &= \frac{1}{2}\left[1 - (|c_L|^2 - |c_R|^2)\cos(\tilde{\omega}t)\right] - \operatorname{Im}(c_L^* c_R)\,\sin(\tilde{\omega}at) \end{aligned} \tag{3.30}$$

while the quantity $\Gamma(t)$ is the coherence of the state of the particle in the $\{|L\rangle, |R\rangle\}$ basis at t_1, that is

$$\begin{aligned} \Gamma(t) &\equiv \langle\psi|\hat{U}(t)|L\rangle\langle R|\hat{U}(t)|\psi\rangle = \\ &= \operatorname{Re}(c_L^* c_R) + i\left[\operatorname{Im}(c_L^* c_R)\,\cos(\tilde{\omega}t) + \frac{|c_L|^2 - |c_R|^2}{2}\sin(\tilde{\omega}t)\right] \end{aligned} \tag{3.31}$$

Expression (3.28) provides us with the explicit expression for the real and imaginary parts of the DCF, and in particular we immediately see that the

imaginary part is directly proportional to the real part of the coherence $\Gamma(t_1)$ of the quantum state of the particle.
Therefore, consistently with the general discussion on quantum correlation functions in section 2.2.3, we find that a non-vanishing $\mathrm{Im}\, G_{DW}(x, t_1, t_2)$ is a direct consequence of the fact that the particle can be in a superposition of position eigenstates, which is possible only if the dynamics is ruled by quantum mechanical law.

Moreover, the explicit for of $\mathrm{Im}\, G_{DW}(x, t_1, t_2)$ allows us to inquiry if there exist any quantum states which are still superposition of eigenstates of position, that is $\Gamma(t) \neq 0$ for some t, but that give a real-valued DCF so that it would not be possible to spot the quantum nature of the particle by measurements of this function or of the asymmetries in the corresponding ISF.
As stated above we have

$$\mathrm{Im}\, G_{DW}(x, t_1, t_2) \propto \mathrm{Re}\, \Gamma(t_1) \tag{3.32}$$

but from the expression for the coherence (3.31) it turns out that

$$\mathrm{Re}\, \Gamma(t_1) = \mathrm{Re}\, (c_L^* c_R) \tag{3.33}$$

that is, the real part of the coherence is constant during the dynamics of the particle. Combining (3.32) and (3.33), we find that the condition to have a real valued DCF is that the expansion coefficients of the initial state of the particle $|\psi\rangle$ satisfy the equation

$$\mathrm{Re}\, (c_L^* c_R) = 0 \tag{3.34}$$

The coefficients satisfying this equation can be given in the form

$$c_L = \cos\left(\frac{\theta}{2}\right), \quad c_R = \pm i \sin\left(\frac{\theta}{2}\right) \tag{3.35}$$

with $\theta \in [0, \pi]$.
That the coherence corresponding to these coefficients does not identically vanish, that is that the initial state corresponding to the coefficients (3.35) is a legit superposition of quantum states, can be seen by a direct calculation. Substitution of (3.35) into (3.31) leads to

$$\Gamma_{\theta,\pm}(t) = \frac{i}{2} \sin\big(\theta \pm \omega t\big) \tag{3.36}$$

The ISF for the particle can also be explicitly calculated.
Because $G_{DW}(x, t_1, t_2)$ can only take three possible discrete values, then the ISF is defined over this discrete values as the discrete sum

$$S_{DW}(\mathbf{Q}, t_1, t_2) = \sum_{x=-d,0,d} G_{DW}(x, t_1, t_2) e^{-ix\mathbf{Q}\cdot\hat{\mathbf{x}}} \tag{3.37}$$

Substitution of (3.28) into (3.37) gives

$$\begin{aligned} S_{DW}(\mathbf{Q}, t_1, t_2) = \sin\left[\frac{\tilde{\omega}(t_2 - t_1)}{2}\right]^2 \cos\left(d\mathbf{Q}\cdot\hat{\mathbf{x}}\right) + \cos\left[\frac{\tilde{\omega}(t_2 - t_1)}{2}\right]^2 + \\ + i\left\{\left[P(L, t_1) - P(R, t_1)\right] \sin\left[\frac{\tilde{\omega}(t_2 - t_1)}{2}\right]^2 \sin\left(d\mathbf{Q}\cdot\hat{\mathbf{x}}\right) + \right. \\ \left. + \left[\operatorname{Im}\Gamma(t_1) \sin\left(d\mathbf{Q}\cdot\hat{\mathbf{x}}\right) + \operatorname{Re}\Gamma(t_1)\left(\cos\left(d\mathbf{Q}\cdot\hat{\mathbf{x}}\right) - 2\right)\right] \frac{\sin\left[\tilde{\omega}(t_2 - t_1)\right]}{2}\right\} \end{aligned} \tag{3.38}$$

We notice that if the condition (3.34) for the state of the particle holds, the ISF is not hermitian with respect to $\mathbf{Q}$ as expected when the DCF is real valued.

This simple model shows explicitly that a complex valued DCF, or equivalently an hermitian ISF, is a consequence of the particle being in a coherent superposition of position eigenstates. But we have also found that among these coherent superpositions there are some for which such conditions on the correlation functions can be violated.
This means that in principle it is not correct to claim that an hermitian ISF is an exclusive property of classical systems.
This is anyway a very minimal system on which to base such a conclusion. For this reason in the next section we analyse a generalization of this model.

3.2.2 Many particles in one dimensional periodic potential in tight binding approximation

A direct generalization of the previous system is given by particles in a periodic potential in the tight binding approximation [44]. In this model a system of non-interacting particles, that we assume to be spin-0 bosons, is trapped in a periodic potential. For simplicity we consider a one-dimensional

potential with M minima in positions $\{x_i\}_{i=1}^{M}$, with two adjacent minima at distance $x_{i+1} - x_i = d$.
Additionally we consider the potential to satisfy periodic boundary conditions, that is $x_{M+1} = x_1$.

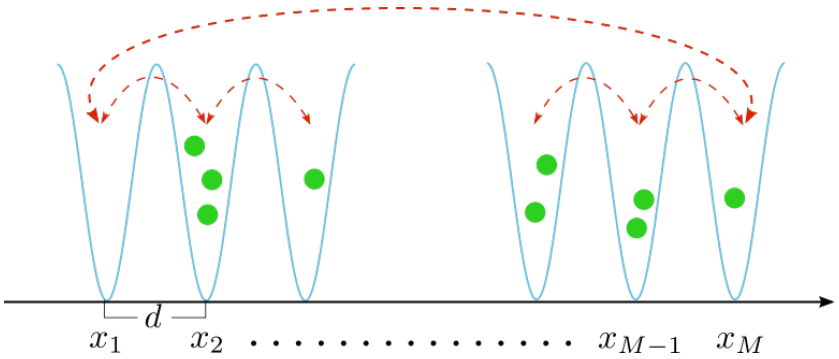

Figure 3.2: One-dimensional bosonic tight binding approximation.

Analogously to the case of a particle in a double well potential, we assume that the particles can only be found at the minima of the potential, and that they can hop between neighbouring minima.

As we are interested in treating the case of a generic number of particles in the system it is useful to adopt a second quantization formalism [43].
We introduce then a set of M operators $\{\hat{c}_i\}_{i=1}^{M}$ and their hermitian conjugates $\{\hat{c}_i^\dagger\}_{i=1}^{M}$. The operator $\hat{c}_i^\dagger$ creates a particle at position x_i, whereas $\hat{c}_i$ destroys a particle at the same site.
These $2M$ operators obey bosonic commutation rules, that is

$$\begin{aligned} \left[\hat{c}_i, \hat{c}_j\right] = \left[\hat{c}_i^\dagger, \hat{c}_j^\dagger\right] = 0 \\ \left[\hat{c}_i, \hat{c}_j^\dagger\right] = \delta_{i,j} \end{aligned} \tag{3.39}$$

The hamiltonian of the N non-interacting bosons hopping between the position of the minima of the potential can be expressed in terms of the creation and destruction operators just introduced

$$\hat{H} = -\hbar\frac{\tilde{\omega}}{2}\sum_{i=1}^{M} \hat{c}_i^\dagger \hat{c}_{i+1} + \hat{c}_{i+1}^\dagger \hat{c}_i \tag{3.40}$$

As for the case of a single particle in the double well potential, in order to analyse the DCF for this model we need the explicit form of the number of particle operators $\hat{\rho}(i)$ at each of the x_i positions. Moreover we have to find the time evolution of these operators.
The first task is straightforward, as we have that

$$\hat{\rho}(i) = \hat{c}_i^\dagger \hat{c}_i \tag{3.41}$$

For the second task it is necessary to introduce a new set of creation and destruction operators to recast the hamiltonian in diagonal form. This new operators are given in terms of the old ones as

$$\begin{aligned} \hat{c}_k^\dagger &= \frac{1}{\sqrt{Md}} \sum_{i=1}^{M} e^{ikx_i} \hat{c}_i^\dagger \\ \hat{c}_k &= \frac{1}{\sqrt{Md}} \sum_{i=1}^{M} e^{-ikx_i} \hat{c}_i \end{aligned} \tag{3.42}$$

where the parameter k takes the discrete values

$$k = \frac{2\pi}{Md} m\,, \qquad m = 0, \pm 1, \ldots, \pm \frac{M}{2} \tag{3.43}$$

The relations (3.42) can then be inverted to give

$$\begin{aligned} \hat{c}_i^\dagger &= \frac{1}{\sqrt{Md}} \sum_{k=-\pi/d}^{\pi/d} e^{-ikx_i} \hat{c}_k^\dagger \\ \hat{c}_i &= \frac{1}{\sqrt{Md}} \sum_{k=-\pi/d}^{\pi/d} e^{ikx_i} \hat{c}_k \end{aligned} \tag{3.44}$$

Substitution of relations (3.42) into (3.40) we arrive at the diagonal form of the hamiltonian

$$\hat{H} = -\hbar \frac{\tilde{\omega}}{2} \sum_{k=-\pi/d}^{\pi/d} \cos(kd) c_k^\dagger c_k \tag{3.45}$$

From this expression it is clear that the operators $\hat{c}_k / \hat{c}_k^\dagger$ destroy/create a particle in the eigenstate of the hamiltonian corresponding to the energy

$$\varepsilon_k = -\hbar \frac{\tilde{\omega}}{2} \cos(kd) \tag{3.46}$$

Moreover we can calculate the evolution of the $\hat{c}_k$ operators via the Heisenberg equation

$$\begin{cases} \frac{d}{dt}\hat{c}_k(t) = \frac{i}{\hbar}[\hat{H}, \hat{c}_k(t)] = -\frac{i}{\hbar}\hat{c}_k(t) \\ \hat{c}_k(0) = \hat{c}_k \end{cases} \tag{3.47}$$

from which we obtain

$$\hat{c}_k(t) = \hat{c}_k e^{-i\frac{\varepsilon_k}{\hbar}t} \qquad\qquad \hat{c}_k^\dagger(t) = \hat{c}_k^\dagger e^{i\frac{\varepsilon_k}{\hbar}t} \tag{3.48}$$

The time dependency of $\hat{\rho}(i)$ can now be explicitly calculated by noticing that

$$\begin{aligned} \hat{\rho}(i,t) &= \frac{1}{Md}\sum_{k,k'=\pi/d}^{\pi/d} e^{-i(k-k')x_i}\, \hat{c}_k^\dagger(t)\hat{c}_{k'}(t) = \\ &= \frac{1}{Md}\sum_{k,k'=\pi/d}^{\pi/d} e^{-i(k-k')x_i}\, e^{\frac{i}{\hbar}(\varepsilon_k-\varepsilon_{k'})t}\, \hat{c}_k^\dagger\hat{c}_{k'} \end{aligned} \tag{3.49}$$

With the explicit form (3.49) we can calculate the imaginary part of the DCF and find the conditions under which it vanishes. This quantity is given by

$$\begin{aligned} &\operatorname{Im} G_{TB}(l,t_1,t_2) = \frac{1}{2i}\sum_i \langle\psi|\big[\hat{\rho}(i,t_1), \hat{\rho}(i+l,t_2)\big]|\psi\rangle = \\ &= \frac{1}{Md}\operatorname{Im}\left(\sum_{k=-\pi/d}^{\pi/d} e^{-ikld}e^{i\varepsilon_k(t_2-t_1)}\sum_{k'=-\pi/d}^{\pi/d} e^{ik'ld}e^{-i\varepsilon_{k'}(t_2-t_1)}\langle\psi|c_{k'}^\dagger c_{k'}|\psi\rangle\right) \end{aligned} \tag{3.50}$$

Asking that (3.50) vanishes, means that the initial state $|\psi\rangle$ of the particles must be such to make real valued the quantity

$$\sum_{k=-\pi/d}^{\pi/d} e^{-ikld}e^{i\varepsilon_k(t_2-t_1)}\sum_{k'=-\pi/d}^{\pi/d} e^{ik'ld}e^{-i\varepsilon_{k'}(t_2-t_1)}\langle\psi|c_{k'}^\dagger c_{k'}|\psi\rangle \in \mathbb{R} \tag{3.51}$$

This last condition of reality can be satisfied if the state of the system is such that the average population of all the energy eigenstates are equally populated, namely if the expectation value $\langle\psi|c_k^\dagger c_k|\psi\rangle$ has the same value for

all the modes k.
We have at least two states satisfying the condition above, namely

$$|\psi_n\rangle = \prod_{k=-\pi/d}^{\pi/d} \frac{\left(c_k^\dagger\right)^n}{\sqrt{n!}}|0\rangle \qquad |\psi_N\rangle = \frac{1}{\sqrt{M}} \sum_{k=-\pi/d}^{\pi/d} \frac{\left(c_k^\dagger\right)^N}{\sqrt{N!}}|0\rangle \tag{3.52}$$

where $|0\rangle$ is the state with zero particles, and if the number of particles is set to N, the number n is an integer such that $nM = N$. For the two states (3.52) we have that the average population of the modes is

$$\langle\psi_n|c_k^\dagger c_k|\psi_n\rangle = \langle\psi_N|c_k^\dagger c_k|\psi_N\rangle = \frac{N}{M} \qquad \forall k \in \left[-\frac{\pi}{d}, \frac{\pi}{d}\right] \tag{3.53}$$

However the two states (3.52) are not necessarily the only ones that can give a constant population for all the energy eigenstates, neither satisfying such condition on populations is necessarily the only way to satisfy condition (3.51).
This means that there could be many more quantum states for this system that give a real valued DCF, hiding in this way the quantum nature of the particles in scattering experiments.

Chapter 4

The quantum theory of Time-Domain interferometry with filtering foils

We open this chapter with a brief account of the working principles of TDI with filtering foils and after that, we present the results obtained in [30].
We provide our quantum theoretical analysis of this experimental technique. As the method relies on a perturbative treatment of the interaction between the target and the radiation, we identify the parts of the matter-radiation hamiltonian that can account for the scattering of photons off of the target.
Before facing the quantum theoretical analysis of TDI with Mössbauer filtering foils, we propose the analysis of a model scheme in which the foils are substituted by split and delay lines, which are arrengements of beamsplitters and mirrors. The action of these devices on the photons of the x-ray pulses can be considered, for our ends, similar to the action of Mössbauer foils.
This alternative scheme does not only serve to illustrate the methods of our analysis, but, given the progeress in the development of split and delay lines for x-ray and gamma radiation [45], is a new concrete experimental proposal for particles correlation measurements.
Then, it is shown how our analysis applies to the actual TDI with filtering foils and the results obtained for the two schemes are explained in a unified way as a "which path information" erasure experiment.
Exploiting the insights obtained via this interpretation, and considering some recent developement in the control of Mössbauer emission by the filtering foils, in the last part of the chapter we propose new usages of TDI.

4.1 Working principle of TDI with Mössbauer filtering foils

In TDI experiments with Mössbauer filtering foils, the target system whose ISF we want to retrieve, is placed between two parallel foils enriched with Mössbauer isotopes and one of the two foils is mounted on a drive.
A short hard x-ray synchrotron pulse, of duration of order $\sim 100ps$, propagates orthogonally toward the first foil, which is moved at constant velocity. The Mössbauer isotopes present in the foil, are excited by the incoming pulse from their ground state to a longlived excited state. Thus, the spectral component of the incident pulse is partially absorbed, and then re-emitted without recoil by the first foil on a timescale τ given by the lifetime of the Mössbauer isotope.
As a consequence, downstream of the first foil, the radiation along the incidence direction is split in two wave packets: a short wave packet, representing the part of the incoming pulse that went through undisturbed through the first foil, and a long wave packet resulting from excitation and de-excitation of the nuclei, whose carrier frequency is given by the frequency ω_0 of the nuclear transition plus the Doppler shift Ω provoked by the motion of the foil.

These two radiation wave packets interact then with the target, whose excitation spectrum is assumed to be non-resonant with the spectra of the wave packets. The radiation is then scattered quasi-elastically in different directions by the electrons distribution in the target.
Each radiation wave packet is scattered at a different time, so that their scattering amplitudes are proportional to the electron density of the target $\rho(\mathbf{r})$ at two distinct times.
In particular the short wave packet recovers information about the target at its arrival time, whereas the long wave packet is scattered during a timelapse τ which is supposed on the same timescale of the dynamics in the target, so that the scattered amplitude in this second case is modulated in time.

The two scattered wave packets go then through the second Mössbauer foil, which is at rest with respect to the first foil so that its resonance frequency is ω_0. Thus the second foil lets the scattered long wave packet propagate undisturbed, while it absorbs and re-emit the ω_0 component of the

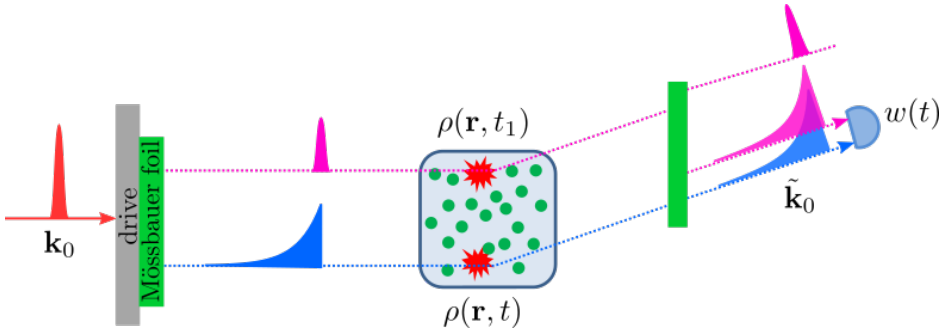

Figure 4.1: Schematics of TDI with Mössbauer filtering foils. The short incoming pulse (red) is split by the first foil mounted on a moving drive. A part of the original pulse goes through the foil (purple) and is scattered by the target at time t_1. Another part excites the nuclei in the foil and is then reemitted in a long pulse (blue) trailing the first one. This long pulse is scattered by the target throughout its duration $\sim \tau$. The short scattered pulse is split by the second foil, so that in the end two overlapping long pulses interfere in the detector.

scattered short wave packet.
By suitably timing the detector, the part of the original incoming pulse that does not interact with any of the two Mössbauer foils can be excluded. Therefore, if we look at the radiation scattered along the direction connecting the target and the detector, we will find two long wave packets: the one that interacted with the first foil, with carrier frequency $\omega_0 + \Omega$ and amplitude depending on instantaneous electron density $\rho(\mathbf{r}, t_1)$, and the one that interacted with the second foil, with carrier frequency ω_0 and a time varying amplitude depending on $\rho(\mathbf{r}, t)$.
Because of the frequency difference, the overlap of these wavepackets in the detector will generate an interference pattern in the recorded signal, and the amplitude of the pattern will depend on the electron density at different times.

To fix the scales of frequencies and times involved in this kind of experiments, we can take as an example the experiment in [11], where the foils were enriched with the Mössbauer isotope ^{57}Fe. For this nucleus we have that $\omega_0 \simeq 2,2 \times 10^7\, THz$, $\tau = 141\, ns$, and the Doppler shift Ω of the resonance frequency of the first foil is of order $\sim 10^2 MHz$.
The general considerations above are the basis for the classical analysis of TDI that can be found in [11, 46, 47]. In the next section we will work on this considerations to build our quantum theoretical analysis of TDI

4.2 The quantum theoretical analysis of TDI

For a consistent analysis of TDI experiments in a quantum theoretical framework, we need to treat both the ensemble of the electrons in the target and the radiation wave packets as quantum systems in interaction.
When the radiation is considered as a quantum system, it can be shown that the signal produced by a photon detector is proportional to the probability per unit time that a photon is absorbed at a given time at the position of the detector [48].
Then, fixing the initial time t_0 at the instant at which the prompt wave packet crosses the first foil, and calling $|\Psi\rangle$ the state of the joint quantum system made up of the electrons in the target and a photon from the incoming x-ray pulse, the above mentioned probability at a time t for an ideal photon detector placed at position $\mathbf{R}$ is

$$w(t) = \langle\Psi|\hat{U}(t,t_0)^\dagger\ \hat{\mathbf{E}}^{(-)}(\mathbf{R})\cdot\hat{\mathbf{E}}^{(+)}(\mathbf{R})\ \hat{U}(t,t_0)|\Psi\rangle \tag{4.1}$$

Here $\hat{\mathbf{E}}^{(\pm)}$ are the positive/negative frequency part of the electric field operator (see appendix B) and $\hat{U}(t,t_0)$ is the evolution operator of the joint system.
The aim of the analysis that we propose here is to calculate $w(t)$ for an initial state $|\Psi\rangle$ that represents a situation in which the photon was acted upon by the first foil, and for an evolution operator $\hat{U}(t,t_0)$ that can account for the quasi-elastic scattering of the photon off of the electron distribution in the target. The expression thus obtained for $w(t)$, will be corrected for the action of the second foil on the pulses.

4.2.1 The electron-photon interaction hamiltonian

In order to find the form of $\hat{U}(t,t_0)$ it is necessary to consider the hamiltonian of the electrons of the target in the presence of an electromagnetic field, and to isolate from it the terms that can contribute to photon scattering at the lowest perturbative order. Considering the electrons as non-relativistic, spinless particles, and assuming that the vector potential of the field satisfies the Coulomb gauge, the hamiltonian is [49]

$$\hat{H} = \hat{H}_{em} + \sum_{j=1}^{N}\frac{\hat{\mathbf{p}}_j^2}{2m} + \hat{V}_j + \frac{e}{m}\hat{\mathbf{p}}_j\cdot\hat{\mathbf{A}}(\mathbf{r}_j) + \frac{e^2}{2m}\hat{\mathbf{A}}(\mathbf{r}_j)^2 \tag{4.2}$$

where $\hat{\mathbf{p}}_j$ is the momentum of the j-th electron, m and e its mass and charge respectively and $\hat{V}_j$ stands for all the interactions that the electron has with the other electrons and constituent elements of the target. $\hat{H}_{em}$ is the hamiltonian of the free electromagnetic field and $\hat{\mathbf{A}}$ is its vector potential operator.

The $\hat{\mathbf{p}}_j \cdot \hat{\mathbf{A}}(\mathbf{r}_j)$ interaction term in (4.2) contributes to photon scattering only at second order in perturbation theory, via virtual absorption and emission of one photon by the electrons. As we suppose that the spectrum of the incoming radiation pulse is far away from any resonances in the target, this probability of photon scattering via the abovementioned virtual processes is heavily suppressed [50].
Therefore we will keep only the $\hat{\mathbf{A}}(\mathbf{r}_j)^2$ interaction term. This term can be recast using the normal mode expansion of $\hat{\mathbf{A}}$ in the following form (see appendix B)

$$\frac{e^2}{2m}\hat{\mathbf{A}}(\mathbf{r}_j)^2 =$$

$$\begin{aligned} = r_e \frac{\hbar c^2}{8\pi} \sum_{j=1}^{N} \sum_{\sigma,\sigma'} \int d^3k \int d^3k' \frac{1}{\sqrt{\omega_k \omega_{k'}}} \big(& \mathbf{e}_{\mathbf{k},\sigma} \cdot \mathbf{e}_{\mathbf{k}',\sigma'} \hat{a}_{\mathbf{k},\sigma} a_{\mathbf{k}',\sigma'} e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{r}_j} + \\ + \mathbf{e}^*_{\mathbf{k},\sigma} \cdot \mathbf{e}^*_{\mathbf{k}',\sigma'} \hat{a}^\dagger_{\mathbf{k},\sigma} \hat{a}^\dagger_{\mathbf{k}',\sigma'} e^{-i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{r}_j} & + \mathbf{e}_{\mathbf{k},\sigma} \cdot \mathbf{e}^*_{\mathbf{k}',\sigma'} \hat{a}_{\mathbf{k},\sigma} \hat{a}^\dagger_{\mathbf{k}',\sigma'} e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_j} + \\ & + \mathbf{e}^*_{\mathbf{k},\sigma} \cdot \mathbf{e}_{\mathbf{k}',\sigma'} \hat{a}^\dagger_{\mathbf{k},\sigma} \hat{a}_{\mathbf{k}',\sigma'} e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_j} \big) \end{aligned} \tag{4.3}$$

where $r_e = e^2/(4\pi\epsilon_0 mc^2)$ is the classical radius of the electron.
The operators $\hat{a}_{\mathbf{k},\sigma}, \hat{a}^\dagger_{\mathbf{k},\sigma}$ destroy and create a photon with momentum $\mathbf{k}$ polarized in the direction $\mathbf{e}_{\mathbf{k},\sigma}$, thus the first two terms in (4.3) do not preserve the number of photons and they cannot contribute to photon scattering, so we drop them.
The last two terms are the only one we will keep from (4.3), because they destroy and create one photoninstead contributing to photon scattering.
Rearranging the order of creation and destruction operators we obtain an effective interaction hamiltonian

$$\hat{H}_{eff} = r_e \frac{\hbar c^2}{4\pi} \sum_{\sigma,\sigma'} \int d^3k \int d^3k' \int_V d^3r \frac{1}{\sqrt{\omega_k \omega_{k'}}} \mathbf{e}^*_{\mathbf{k},\sigma} \cdot \mathbf{e}_{\mathbf{k}',\sigma'} \hat{a}^\dagger_{\mathbf{k},\sigma} \hat{a}_{\mathbf{k}',\sigma'} \hat{\rho}(\mathbf{r}) e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} \tag{4.4}$$

where the density of electrons operator $\hat{\rho}(\mathbf{r}) \equiv \sum_{j=1}^{N} \delta(\mathbf{r} - \mathbf{r}_j)$ has been in-

troduced. Through this operator we can make the substitution

$$\sum_{j=1}^{N} e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_l} = \int_V d^3r\hat{\rho}(\mathbf{r})e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} \tag{4.5}$$

with V the volume of the target.

The evolution operator $\hat{U}(t,t_0)$ appearing in (4.1), is then given at first order in perturbation theory by (see appendix A)

$$\hat{U}(t,t_0) \simeq \hat{U}_0(t,t_0)\left[\mathbb{1} - \frac{i}{\hbar}\int_{t_0}^{t} dt'\,\hat{U}_0(t,t_0)^\dagger \hat{H}_{eff}\hat{U}_0(t,t_0)\right] \tag{4.6}$$

4.2.2 A model of TDI with split and delay lines

For what concerns the initial state of the joint system, we assume in first place that no correlations between the target and the radiation exist at time t_0, so that the $|\Psi\rangle$ is a product of a state for the electrons and a state for the photon

$$|\Psi\rangle = |\psi\rangle|\gamma\rangle \tag{4.7}$$

where $|\psi\rangle$ refers to the electrons and $|\gamma\rangle$ to the photon.

The state $|\gamma\rangle$ must represent the result of the action of a Mössbauer foil on a photon from the incoming radiation pulse
To understand what this action amounts to, and also to show how the detection probability $w(t)$ is calculated, we analyze in the following a model scheme in which the foils are substituted with two identical split and delay lines (SDL).

A SDL is a device that splits an incoming radiation pulse in two, and leads these two pulses on paths of different length before they are collimated back on the same propagation direction.
As a result, at the end of the SDL the second pulse is delayed with respect to the first (see figure 4.2)

Therefore when a photon enters a SDL, it can either propagate along the shorter path or it gets delayed by going the longer path (see figure 4.2). These two possibilities are represented by two quantum states for the photon,

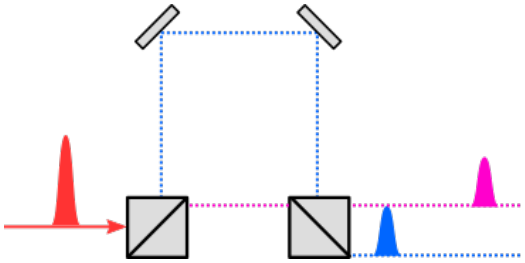

Figure 4.2: A split and delay is an arrangement of beam splitters and mirrors that splits an incoming pulse (in red) in two pulses (in purple and blue). The blue pulse accumulates a delay during its propagation as it follows a longer path with respect to the blue pulse. The two pulses are then collimated along the same direction (the vertical distance between the pulses in the picture is introduced only for the sake of clarity and has not to be meant as a distance between the directions of prpagation.

that we call $|\gamma_1\rangle$ and $|\gamma_2\rangle$, so that the state of the photon after the SDL is the superposition of these two, that is

$$|\gamma\rangle = |\gamma_1\rangle + |\gamma_2\rangle \tag{4.8}$$

The two states $|\gamma_l\rangle$ $(l = 1, 2)$ are photon wave packets with the same shape of the incoming pulse, propagating along the same direction z. Assuming that the polarization of the photon is fixed in the direction $\mathbf{e}_{\sigma_0}$, we can represent these wave packets as follows

$$|\gamma_l\rangle = \frac{1}{\sqrt{\mathcal{A}}} \int_0^\infty dk g(\omega_k - \omega_0) e^{-ikz_l} |1_{\mathbf{k},\sigma_0}\rangle \tag{4.9}$$

where $|1_{\mathbf{k},\sigma_0}\rangle$ are single photon states with momentum parallel to z, z_l is the distance the wave packet must travel to reach the target, $\mathcal{A}$ is the transverse area of the wave packet and $g(\omega_k - \omega_0)$ is a function reproducing the features of the spectrum of the incoming pulse. For the sake of definiteness we can consider the function g to be a gaussian centered at ω_0 with a variance $\Delta\omega \ll \omega_0$

Taking then the superposition (4.8) as the initial state of the photon, also the initial state of the joint system will be a superposition of quantum states

$$|\Psi\rangle = |\psi\rangle|\gamma_1\rangle + |\psi\rangle|\gamma_2\rangle \tag{4.10}$$

Applying the evolution operator (4.6) to this state, we obtain that the evolved state of the joint system is the sum of three contributions

$$\hat{U}(t, t_0)|\Psi\rangle \simeq \hat{U}_0(t, t_0)\Big[|\Psi\rangle + |\delta\Psi_1(t)\rangle + |\delta\Psi_2(t)\rangle\Big] \tag{4.11}$$

where

$$|\delta\Psi_l(t)\rangle = -\frac{i}{\hbar}\int_0^t dt' \hat{U}_0^\dagger(t')\hat{H}_{eff}\hat{U}_0(t')\,dt'|\gamma_l\rangle|\psi\rangle. \tag{4.12}$$

The first term in (4.11) represents a situation in which there is no interaction between the photon and the electrons so that the former cannot be deflected towards the detector, and for this reason does not contribute to the probability of detection. We will neglect this term in the following.
Therefore $w(t)$ is

$$w(t) \simeq \sum_{l,l'=1,2} \langle\delta\Psi_l(t)|\hat{\mathbf{E}}^{(-)}(\mathbf{R},t)\cdot\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_{l'}(t)\rangle \tag{4.13}$$

with $\hat{\mathbf{E}}^{(\pm)}(\mathbf{R},t) = \hat{U}_0(t,t_0)^\dagger\,\hat{\mathbf{E}}^{(\pm)}(\mathbf{R})\hat{U}_0(t,t_0)$.

Since $|\delta\Psi_l(t)\rangle$ are single photon states and $\hat{\mathbf{E}}^{(+)}$ destroys one photon, the quantity $\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_{l'}(t)\rangle$ is proportional to the vacuum state of the electromagnetic field $|0\rangle$. Thus we can rewrite the expression (4.13) as [51]

$$w(t) \simeq \sum_{l,l'=1,2} \langle\delta\Psi_l(t)|\hat{\mathbf{E}}^{(-)}(\mathbf{R},t)|0\rangle\cdot\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_{l'}(t)\rangle \tag{4.14}$$

The quantities $\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_l(t)\rangle$ represent the quantum probability amplitude of detecting the photon in the l-th scattering channel.

With the explicit form (4.4) for the effective interaction hamiltonian and the form (4.9) for the single photon states we find that

$$\begin{aligned}|\delta\Psi_l(t)\rangle = -i\frac{r_e c^2}{4\pi\sqrt{\mathcal{A}}}\sum_\sigma \int_{t_0}^t dt' \int_V d^3r \int d^3k' \int_0^\infty dk \frac{e^{-i(\mathbf{k}'\cdot\mathbf{r}-\omega_{k'}t')}}{\sqrt{\omega_{k'}}} \\ (\mathbf{e}_{\mathbf{k}',\sigma}\cdot\mathbf{e}_{\sigma_0})\frac{e^{i(kz-\omega_k t')}}{\sqrt{\omega_k}} e^{-ikz_l} g(\omega_k-\omega_0)\hat{\rho}(\mathbf{r},t')|\psi\rangle|1_{\mathbf{k}',\sigma_0}\rangle\end{aligned} \tag{4.15}$$

Using the explicit form of $\hat{\mathbf{E}}^{(+)}$ (see appendix B) and of $|\delta\Psi_l(t)\rangle$ we obtain

that the l-th detection amplitude is

$$\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_l(t)\rangle = -\frac{r_e c^2}{4\pi}\sqrt{\frac{\hbar}{2(2\pi)^3\mathcal{A}\epsilon_0}}\sum_\sigma \int_{t_0}^{t} dt' \int_V d^3r\hat{\rho}(\mathbf{r},t')|\psi\rangle\times$$
$$\times \int d^3q\,(\mathbf{e}_{\mathbf{q},\sigma}\cdot\mathbf{e}_{\sigma_0})\mathbf{e}_{\mathbf{q},\sigma}\,e^{i[\mathbf{q}\cdot(\mathbf{R}-\mathbf{r})-\omega_q(t-t')]}\int dk e^{-i[\omega_k t'-k(z+z_l)]}\frac{g(\omega_k-\omega_0)}{\sqrt{\omega_k}} \tag{4.16}$$

and being $g(\omega_k-\omega_0)$ peaked around the value ω_0 with a finite support, we approximate the integral over dk with the Fourier transform of g

$$\int dk e^{-i[\omega_k t'-k(z+z_l)]}\frac{g(\omega_k-\omega_0)}{\sqrt{\omega_k}} \simeq \frac{1}{\sqrt{\omega_0}}e^{-i\omega_0\left(t'-\frac{z+z_l}{c}\right)}f\left(t'-\frac{z+z_l}{c}\right) \tag{4.17}$$

Here the function f is the moving envelope of the photon wavepacket, which again can be taken as a guassian whose maximum is at $t'=(z+z_l)/c$ and has a variance of order $\sim 1/\Delta\omega$. The physical dimension of f is that of an inverse square root of a length.
Beacause of these features of the envelope, the integrand in the time integral appearing in (4.16) is non-null while the envelope f is contained in the volume occupied by the target, that is during the time that the envelope takes to cross the transverse size of the target. Assuming that such crossing time is shorter than the typical timescale of the dynamics in the target, the electron density $\hat{\rho}(\mathbf{r},t')$ can then be considered "frozen" at the instant of arrival of the photon wave packet t_l and brought out of the time integral.
Expression (4.16) then becomes

$$\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_l(t)\rangle = -\frac{r_e c^2}{4\pi}\sqrt{\frac{\hbar}{2(2\pi)^3\omega_0\mathcal{A}\epsilon_0}}\sum_\sigma \int_V d^3r\hat{\rho}(\mathbf{r},t_l)|\psi\rangle$$

$$\int d^3q\,(\mathbf{e}_{\mathbf{q},\sigma}\cdot\mathbf{e}_{\sigma_0})\mathbf{e}_{\mathbf{q},\sigma}\,e^{i[\mathbf{q}\cdot(\mathbf{R}-\mathbf{r})-\omega_q t]}\int_{t_0}^{t} dt' f\left(t'-t_l-\frac{z}{c}\right)e^{i\omega_q t'}e^{-i\omega_0\left(t'-t_l-\frac{z}{c}\right)} \tag{4.18}$$

Integrating over the directions of the wavevector $\mathbf{q}$ and the time t', and taking into account that the detector is placed far away from the target, that

is $|\mathbf{R}| \gg \sqrt[3]{V}$ the detection amplitude becomes

$$\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_l(t)\rangle = -\frac{r_e}{2}\sqrt{\frac{\hbar\omega_0}{2(2\pi)^3\mathcal{A}\epsilon_0}}\frac{e^{i\omega_0(R/c-t)} - e^{-i\omega_0(R/c+t)}}{R}e^{i\omega_0 t_l}$$
$$\mathbf{e}_\perp f\left(t - t_l - \frac{R}{c}\right)\int_V d^3r e^{-i(\tilde{\mathbf{k}}_0-\mathbf{k}_0)\cdot\mathbf{r}}\hat{\rho}(\mathbf{r},t_l)|\psi\rangle \tag{4.19}$$

where $\mathbf{e}_\perp$ is the projection of the photon polarization $\mathbf{e}_{\sigma_0}$ on the direction of $\mathbf{R}$, $\tilde{\mathbf{k}}_0 \equiv \mathbf{R}\omega_0/(cR)$ and $\mathbf{k}_0 \equiv \mathbf{z}\omega_0/(cz)$, by which we define the momentum exchanged between the target and the photon as $\mathbf{Q} \equiv \tilde{\mathbf{k}}_0 - \mathbf{k}_0$.

Expression (4.19) is a superposition of an outgoing and an ingoing spherical wave propagating from/to the target. The ingoing spherical wave represents a path in which the photon propagates from infinity to the target, which is a situation not compatible with our scattering problem, and therefore is dropped.
The final form of the detection amplitude is then

$$\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_l(t)\rangle =$$

$$= -\frac{r_e}{2}\sqrt{\frac{\hbar\omega_0}{2(2\pi)^3\mathcal{A}\epsilon_0}}\mathbf{e}_\perp\frac{e^{i\omega_0(R/c-t)}}{R}e^{i\omega_0 t_l}f\left(t - t_l - \frac{R}{c}\right)\int_V d^3r e^{-i\mathbf{Q}\cdot\mathbf{r}}\hat{\rho}(\mathbf{r},t_l)|\psi\rangle \tag{4.20}$$

Substitution of (4.20) into (4.13) brings to the explicit expression of the

photon detection probability per time unit

$$w(t) \simeq \frac{r_e^2}{8}\frac{\hbar\omega_0}{(2\pi)^3\mathcal{A}\epsilon_0}\frac{|\mathbf{e}_\perp|^2}{R^2}$$

$$\left\{\left|f\left(t-t_1-\frac{R}{c}\right)\right|^2 S^{(qu)}(\mathbf{Q},t_1,t_1)+\left|f\left(t-t_2-\frac{R}{c}\right)\right|^2 S^{(qu)}(\mathbf{Q},t_2,t_2)+\right.$$

$$\left.+2\mathrm{Re}\left[e^{i\omega_0(t_1-t_2)}f\left(t-t_1-\frac{R}{c}\right)f^*\left(t-t_2-\frac{R}{c}\right)S^{(qu)}(\mathbf{Q},t_1,t_2)\right]\right\} \tag{4.21}$$

where $S^{(qu)}$ stands for the quantum ISF of the electrons as defined in section 3.1.2, that is the space Fourier transform of the expectation value over the quantum state of the product of the electron density operators at two different times

$$S^{(qu)}(\mathbf{Q},t_1,t_2)=\int_V d^3r\int_V d^3r'\,e^{-i\mathbf{Q}\cdot|\mathbf{r}-\mathbf{r}'|}\langle\psi|\hat{\rho}(\mathbf{r},t_1)\hat{\rho}(\mathbf{Q},t_2)|\psi\rangle \tag{4.22}$$

The first two terms in (4.21) give respectively the probability that the photon is detected after it has been scattered in the channel 1 or 2. The third term is instead an interference term between the two scattering channels, and its magnitude depends on the product of the wave packets scattered in the different channels and of the ISF of our interest. Thus the more these wave packets overlap, the bigger the interference term and the more significant is the contribution of the ISF to the interferogram. Hence, If we want to recover the ISF, the overlap between the envelopes of the scattered wave packets has to be enhanced.
This can be done if a second SDL, with same characteristics of the first, is introduced just before of the detector. This second SDL splits each scattering pathway into two other pathways

Thus the photon can take four possible pathways from the first SDL to the detector (see figure 4.3) :

- it is scattered by the target at time t_1 and goes straight through the second SDL

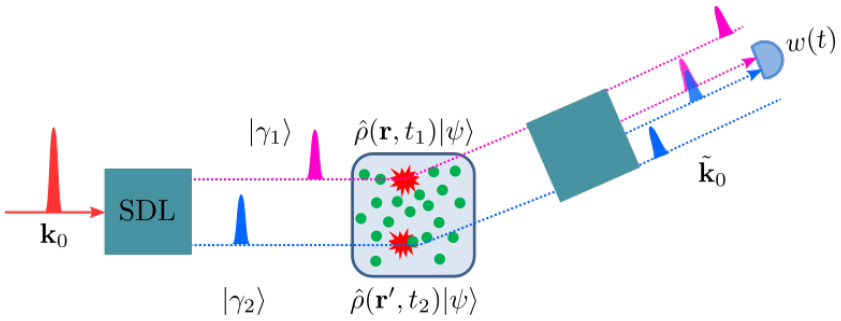

Figure 4.3: Time domain interferometry realized with split and delay lines. In blue and purple are represented the envelopes of the single photon wave packets.

- it is scattered at time t_1 and is delayed by the second SDL
- it is scattered at time t_2 and goes straight through the second SDL
- it is scattered at time t_2 and is delayed by the second SDL

Because the two SDLs are equal, the envelopes of the wave packets scattered in the second and third pathways overlap completely and arrive at the same time at the detector. The envelopes in the first and fourth path arrive instead at the detector either before or after, and by a proper timing of the detector their contribution to the detection probability can be neglected.
The final for of detection probability is [30]

$$w(t) \simeq \frac{r_e^2}{8} \frac{\hbar\omega_0}{(2\pi)^3 \mathcal{A}\epsilon_0} \frac{|\mathbf{e}_\perp|^2}{R^2} \left| f\left(t - t_2 - \frac{R}{c}\right) \right|^2 \tag{4.23}$$
$$\left\{ S^{(qu)}(\mathbf{Q}, t_1, t_1) + S^{(qu)}(\mathbf{Q}, t_2, t_2) + 2\mathrm{Re}\left[e^{i\omega_0(t_1 - t_2)} S^{(qu)}(\mathbf{Q}, t_1, t_2) \right] \right\}$$

4.2.3 TDI realization with Mössbauer filtering foils

The action of a Mössbauer foil on a single photon is analogous to the action of a SDL: when a single photon wave packet containing the resonance frequency of the foil goes through the foil, either it goes through undisturbed or the foil absorbs the photon and starts immediately to re emit it during a time of order of the lifetime of the excited state of ^{57}Fe. Therefore, as in the case of

the SDL, the state of the photon after downstram of the foil is a quantum superposition of two single photon wavepackets

$$|\gamma\rangle = |\gamma_1\rangle + |\tilde{\gamma}_2\rangle \tag{4.24}$$

$|\gamma_1\rangle$ is a short single photon wave packet of the same kind as (4.9), while $|\tilde{\gamma}_2\rangle$ is the state of the photon re-emitted by the Mössbauer foil

$$|\tilde{\gamma}_2\rangle = \frac{1}{\mathcal{A}} \int dk\, \mathcal{L}(\omega_k - \omega_0 - \Omega) e^{-ikz_1} |1_{\mathbf{k},\sigma_0}\rangle \tag{4.25}$$

The function $\mathcal{L}(\omega_k - \omega_0 - \Omega)$ is a Lorentzian spectral shape centered at the resonance frequency $\omega_0 + \Omega$ of the moving foil. The envelope of $|\tilde{\gamma}_2\rangle$ is then an exponential function decaying with time, with decay constant τ.
Moreover, because the re-emission of the photon starts immediately after the photon hits the foil, the envelope of $|\tilde{\gamma}_2\rangle$ and that of $|\gamma_1\rangle$ are at the same initial distance z_1 from the target.

As in the previous case with SDLs, we can explicitly compute the detection amplitudes for the two scattering channels $|\delta\Psi_1(t)\rangle$ and $|\delta\tilde{\Psi}_2(t)\rangle$ corresponding to $|\gamma_1\rangle$ and $|\tilde{\gamma}_2\rangle$.
The calculation of the amplitude for $|\delta\tilde{\Psi}_2(t)\rangle$ needs a slightly different treatment than the one for $|\delta\Psi_1(t)\rangle$. This is due to the fact that the decay constant τ of the exponential envelope, is comparable with the timescale of the internal dynamics of the target.
This fact does not allow us to assume that the electron density operator is constant during the crossing time of the wave packet.

In order to take into account the time dependency of $\hat{\rho}$ in the expression for the scattering amplitude we introduce the energy eigenbasis of the target $\{|\varepsilon_n\rangle\}_n$ and decompose on it the electron density operator

$$\hat{\rho}(\mathbf{r}, t') = \sum_{m,n} e^{i\omega_{mn}t'} |\varepsilon_m\rangle\langle\varepsilon_m|\hat{\rho}(\mathbf{r})|\varepsilon_n\rangle\langle\varepsilon_n| \tag{4.26}$$

where $\omega_{mn} \equiv (\varepsilon_m - \varepsilon_n)/\hbar$ are the characteristic frequencies of the target's internal dynamics. The assumption that the target is non resonant with the radiation means that the ω_{mn} do not fall within the support of the Lorentzian spectral shape $\mathcal{L}(\omega - \omega_0 - \Omega)$.

As a consequence, $\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\tilde{\Psi}_2(t)\rangle$ is

$$\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\tilde{\Psi}_2(t)\rangle = -\frac{r_e c^2}{4\pi}\sqrt{\frac{\hbar}{2(2\pi)^3\mathcal{A}\epsilon_0}}$$
$$\sum_{m,n}\int_V d^3r|\varepsilon_m\rangle\langle\varepsilon_m|\hat{\rho}(\mathbf{r})|\varepsilon_n\rangle\langle\varepsilon_n|\psi\rangle\int d^3q\sum_\sigma(\mathbf{e}_{\mathbf{q},\sigma}\cdot\mathbf{e}_{\sigma_0})\mathbf{e}_{\mathbf{q},\sigma}$$
$$e^{i[\mathbf{q}\cdot(\mathbf{R}-\mathbf{r})-\omega_q t]}\int_0^t dt' e^{i(\omega_q+\omega_{mn})t'}\int dk e^{-i[\omega_k t'-k(z+z_1)]}\frac{\mathcal{L}\big(\omega_k-\omega_0-\Omega\big)}{\sqrt{\omega_k}} =$$

$$= -\frac{r_e c^2}{4\pi}\sqrt{\frac{\hbar}{2(2\pi)^3\omega_0\mathcal{A}(\omega_0+\Omega)\epsilon_0}}\sum_{m,n}\int_V d^3r|\varepsilon_m\rangle\langle\varepsilon_m|\hat{\rho}(\mathbf{r})|\varepsilon_n\rangle\langle\varepsilon_n|\psi\rangle e^{i\omega_{mn}\frac{z+z_1}{c}}$$

$$\int d^3q\sum_\sigma(\mathbf{e}_{\mathbf{q},\sigma}\cdot\mathbf{e}_{\sigma_0})\mathbf{e}_{\mathbf{q},\sigma}e^{i\left[\mathbf{q}\cdot(\mathbf{R}-\mathbf{r})-\omega_q\left(t-\frac{z+z_1}{c}\right)\right]}\mathcal{L}\big(\omega_q+\omega_{mn}-\omega_0-\Omega\big) \tag{4.27}$$

Integration over the momentum $\mathbf{q}$ leads to (see appendix C)

$$\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\tilde{\Psi}_2(t)\rangle = -\frac{r_e}{2}\sqrt{\frac{\hbar}{2(2\pi)^3\mathcal{A}(\omega_0+\Omega)\epsilon_0}}\frac{\mathbf{e}_\perp}{R}$$

$$\sum_{m,n}(\omega_0+\Omega-\omega_{mn})^2\int_V d^3r|\varepsilon_m\rangle\langle\varepsilon_m|\hat{\rho}(\mathbf{r})|\varepsilon_n\rangle\langle\varepsilon_n|\psi\rangle e^{i\omega_{mn}\left(t-\frac{|\mathbf{R}-\mathbf{r}|}{c}\right)} \tag{4.28}$$

$$\frac{e^{i(\omega_0+\Omega)\left(\frac{|\mathbf{R}-\mathbf{r}|+z+z_1}{c}-t\right)}}{|\mathbf{R}-\mathbf{r}|}\Theta\left(t-\frac{|\mathbf{R}-\mathbf{r}|+z+z_1}{c}\right)e^{-\frac{t-(|\mathbf{R}-\mathbf{r}|+z+z_1)/c}{\tau}}$$

with Θ the Heaviside step function.

To obtain the final form of this detection amplitude, several considerations are in order. In first place the Doppler shift Ω is big enough to set the two Mössbauer foils off resonance, but it is anyways smaller than the resonance

frequency at rest ω_0. Indeed taking as a reference the experiments in [11], Ω is of order $\sim 10^8\, Hz$.
Also the characteristic frequencies of the target, ω_{mn}, have to be compared to ω_0. The condition of non-resonance between the target and the pulse implies that $\omega_{mn} \ll \omega_0$, and as a consequence we have that the sum over the energy eigenstates of the target can be approximated as

$$\begin{aligned}\sum_{m,n}(\omega_0+\Omega-\omega_{mn})|\varepsilon_m\rangle\langle\varepsilon_m|\hat{\rho}(\mathbf{r})|\varepsilon_n\rangle\langle\varepsilon_n|\psi\rangle e^{i\omega_{mn}\left(t-\frac{|\mathbf{R}-\mathbf{r}|}{c}\right)} \simeq \\ \simeq \omega_0\,\hat{\rho}\left(t-\frac{|\mathbf{R}-\mathbf{r}|}{c}\right)|\psi\rangle\end{aligned} \tag{4.29}$$

Moreover, invoking again the hypothesis that $R \gg V^{1/3}$ and assuming that the dynamics in the target has a typical timescale larger than $V^{1/3}/c$, we simplify expression (4.28) as follows

$$\begin{aligned}\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\tilde{\Psi}_2(t)\rangle = -\frac{r_e}{2}\sqrt{\frac{\hbar\omega_0}{2(2\pi)^3\mathcal{A}\epsilon_0}}\mathbf{e}_\perp\frac{e^{i(\omega_0+\Omega)((R+z_1)/c-t)}}{R} \\ \Theta\left(t-\frac{R+z_1}{c}\right)e^{-\frac{t-(R+z_1)/c}{\tau}}\int_V d^3r\,\hat{\rho}\left(\mathbf{r},t-\frac{R}{c}\right)|\psi\rangle e^{-i(\tilde{\mathbf{k}}_0-\mathbf{k})\cdot\mathbf{r}}\end{aligned} \tag{4.30}$$

where again we defined the wave vectors $\tilde{\mathbf{k}}_0 \equiv (\omega_0+\Omega)\mathbf{R}/(cR) \simeq \omega_0\mathbf{R}/(cR)$ and $\mathbf{k}_0 \equiv (\omega_0+\Omega)\mathbf{x}/(cx) \simeq \omega_0\mathbf{z}/(cz)$

Expression (4.30) is the final form of the detection amplitude in the second scattering channel. We find that it has the form of a spherical wave of frequency $\omega_0+\Omega$, being then non resonant with the second Mössbauer foil.

On the other hand, if there were not the second foil, the detection amplitude $\langle 0|\hat{\mathbf{E}}^{(+)}(\mathbf{R},t)|\delta\Psi_1(t)\rangle$ would be similar to expression (4.16). However, the second foil modifies it by scattering its component at frequency ω_0, so that the envelope f that appears in (4.16) must be substituted in this case with the same exponentially decaying function appearing in (4.30), that is

$$f\left(t-\frac{R+z_1}{c}\right) \longrightarrow \Theta\left(t-\frac{R+z_1}{c}\right)e^{-\frac{t-(R+z_1)/c}{\tau}} \tag{4.31}$$

Again the part of the photon wave packet $|\gamma_1\rangle$ that is not affected by none of the two foils can be excluded from detection by timing the detector, so that the detection probability is

$$w(t) \simeq -\frac{r_e^2}{8}\frac{\hbar\omega_0}{(2\pi)^3\mathcal{A}\epsilon_0}|\mathbf{e}_\perp|^2\,\Theta\left(t-t_1+\frac{R}{c}\right)\frac{e^{-2\frac{t-t_1+R/c}{\tau}}}{R^2}\Bigg\{S^{(qu)}(\mathbf{Q},t_1,t_1)+$$

$$+S^{(qu)}\left(\mathbf{Q},t_1,t-\frac{R}{c}\right)+2\mathrm{Re}\left[e^{i\Omega\left(t-t_1+\frac{R}{c}\right)}S^{(qu)}\left(\mathbf{Q},t_1,t-\frac{R}{c}\right)\right]\Bigg\} \tag{4.32}$$

with $t_1 = z_1/c$.
Also in this case we obtain an interference term in the photon detection probabilty which is proportional to the ISF of the electrons. This shows that in principle TDI with Mössbauer filtering foil is fit to study the dynamical correlations of particles even if the target is a quantum system.

The results so far obtained for the detection probability $w(t)$ in the two cases of TDI realized with SDLs and Mössbauer filtering foils, expressions (4.23) and (4.32), can be understood in a unifying picture of the experimental schemes that follows the line of reasoning proposed by Feynman for calculating probabilities of events in quantum theory [52].
Following Feynman, given an event that can occur via different indistinguishable paths (for instance an electron can get from a source to a detector placed behind a screen, through two holes pinched in the screen), each of this paths has an amplitude probability associated with it, and the total probability that the event occurs is given by the modulus square of the sum of these amplitudes.
Therefore the different amplitudes will give rise to interference terms in the expression for the probability of occurrence of the event. However if the different paths through which the event can occur can be somehow experimentally discriminated, the probability for the event is just the sum of the squares of their amplitudes.

In our case, for both the realization of TDI technique, the photon can be absorbed by the detector following two different paths: either the photon is scattered by the target at time t_1 and then reaches the detector, or it scatters

at time t_2 and then get to the detector[1].
We have seen that these amplitude amounts to

$$\mathrm{Amp}_1 \propto f(t_1) \int_V d^3r\, e^{-i\mathbf{Q}\cdot\mathbf{r}} \hat{\rho}(\mathbf{r}, t_1)|\psi\rangle \tag{4.33}$$

$$\mathrm{Amp}_2 \propto e^{i\phi(t_1,t_2)} f(t_2) \int_V d^3r\, e^{-i\mathbf{Q}\cdot\mathbf{r}} \hat{\rho}(\mathbf{r}, t_2)|\psi\rangle \tag{4.34}$$

where $\phi(t_1, t_2)$ is the relative phase between the two paths, and amounts to $\omega_0(t_1 - t_2)$ for the SDL realization and to $\Omega(t - t_1 - R/c)$ for the realization with the Mössbauer foils .
The only information that could allow us to understand what path the photon took to the detector, and thus to destroy the interference between the probability amplitudes forbidding us to retrieve the ISF, are the different scattering instants and therefore the times of arrival of the photon at the detector.

From this point of view, the overlap of the scattered wavepackets caused by the SDL, or by the Mössbauer foil placed before the detector, make the paths experimentally indistinguishable by erasing the information about the scattering times.
Thus, the photon detection probability is

$$\begin{aligned} w(t) = |\mathrm{Amp}_1 + \mathrm{Amp}_2|^2 \propto S^{(qu)}(\mathbf{Q}, t_1, t_1) + S^{(qu)}(\mathbf{Q}, t_2, t_2) + \\ + \cos\,\phi(t_1, t_2)\ \mathrm{Re}\, S^{(qu)}(\mathbf{Q}, t_1, t_2) - \sin\,\phi(t_1, t_2)\ \mathrm{Im}\, S^{(qu)}(\mathbf{Q}, t_1, t_2) \end{aligned} \tag{4.35}$$

4.2.4 The reconstruction of the ISF via relative phase control

In both the schemes we have found that the relative phase $\phi(t_1, t_2)$ strictly depends from the scattering times.
However it has been demonstrated recently that it is possible to control the phase of the pulse emitted by Mössbauer foils by controlling their motion with sub-Ångstrom precision [53]. This means that it is in principle possible to introduce in TDI with Mössbauer filtering foils an additional relative

[1]We will use t_2 also for the Mössbauer case, with the understanding that for this case $t_2 = t - R/c$ as follows from (4.32)

phase ϕ_0 between the paths that can be controlled and is independent of the scattering times.

The introduction of the controllable relative phase ϕ_0 opens up the possibility of assessing experimentally the quantum nature of correlations among particles in a target whose nature is not known *a priori*.
Let us consider a TDI experiment in which two detectors are placed behind the second foil, in correspondence of two opposite values of the exchanged momentum $\mathbf{Q}$.
The sum and the difference of the signals recorded by the two detectors will be

$$\begin{aligned} I^{(\pm)}(\phi_0, t_1, t_2) \propto & \sum_{l=1,2} S(\mathbf{Q}, t_l, t_l) \pm S(-\mathbf{Q}, t_l, t_l) + \\ & + 2\Big\{ \cos\big(\phi(t_1, t_2) + \phi_0\big) \big[\mathrm{Re}\, S(\mathbf{Q}, t_1, t_2) \pm \mathrm{Re}\, S(-\mathbf{Q}, t_1, t_2)\big] + \\ & - \sin\big(\phi(t_1, t_2) + \phi_0\big) \big[\mathrm{Im}\, S(\mathbf{Q}, t_1, t_2) \pm \mathrm{Im}\, S(-\mathbf{Q}, t_1, t_2)\big] \Big\} \end{aligned} \tag{4.36}$$

In section 3.1 we have seen that when the DCF is real-valued, the corresponding ISF satisfies the following symmetries

$$\begin{aligned} \mathrm{Re}\, S(\mathbf{Q}, t_1, t_2) &= \mathrm{Re}\, S(-\mathbf{Q}, t_1, t_2) \\ \mathrm{Im}\, S(\mathbf{Q}, t_1, t_2) &= -\mathrm{Im}\, S(-\mathbf{Q}, t_1, t_2) \end{aligned} \tag{4.37}$$

this meaning that in this case the signals $I^{(\pm)}$ become

$$I^{(+)}(\phi_0, t_1, t_2) \propto 2 \sum_{l=1,2} S(\mathbf{Q}, t_l, t_l) + 4 \cos\big(\phi(t_1, t_2) + \phi_0\big) \mathrm{Re}\, S(\mathbf{Q}, t_1, t_2) \tag{4.38}$$

$$I^{(-)}(\phi_0, t_1, t_2) \propto -4 \sin\big(\phi(t_1, t_2) + \phi_0\big) \mathrm{Im}\, S(\mathbf{Q}, t_1, t_2) \tag{4.39}$$

As ϕ_0 can be controlled, one could record the signals $I^{(\pm)}(\phi_0, t_1, t_2)$ for different values of the phase ϕ_0.
Then, if for some pairs of scattering instants t_1 and t_2, the dependency of $I^{(\pm)}$ on ϕ_0 is not compatible with the sinusoidal behaviours (4.38)- (4.39), the experimenter must conclude that the ISF of the target does not satisfy the symmetries (4.37), this meaning that a quantum mechanical model is needed

to correctly describe correlations among particles. For instance such a check can be done even just by looking at the values of ϕ_0 for which $I^{(-)}$ is null: if these values are not $\phi_0^{(m)} = -\phi(t_1, t_2) + m\pi$ it can be claimed straight away that quantum phenomena determine the behaviour of the target.

On the other hand, finding out that the behaviour of $I^{(\pm)}$ is compatible with (4.38), (4.39), that is finding that the ISF is symmetrical according to (4.37), does not automatically imply that a classical model for particles correlation is sufficient.

Another possibility offered by the control of the relative phase ϕ_0 is the reconstruction of the ISF from the data, as will be shown below.
At the knowledge of the authors, there is no other proposals in the literature for operating such reconstruction. Indeed usually, in TDI experiments, a classical model for the ISF of the target is presumed and the data are used to extract the value of the parameters appearing in the model. For instance, in the application of TDI for the study of diffusive dynamics already cited [14, 15, 16, 17], an exponentially decaying behaviour of the ISF with time is assumed, and the decaying constant, as well as other parameters, are extracted from the data.
The reconstruction of the ISF can be useful if no clues are available to suggest a model for the features of the target, or to obtain more accurate validations of existing models when these exist.

Let us imagine that a TDI experiment is done on a generic target, whose features are unknown. Its ISF $S(\mathbf{Q}, t_1, t_2)$ is also unknown and in principle it can be a complex valued function, so that the signal at the detector will be

$$\begin{aligned} I(\phi_0, t_1, t_2) \propto & \sum_{l=1,2} S(\mathbf{Q}, t_l, t_l) + \\ & + \cos\left(\phi(t_1, t_2) + \phi_0\right) \operatorname{Re} S(\mathbf{Q}, t_1, t_2) - \sin\left(\phi(t_1, t_2) + \phi_0\right) \operatorname{Im} S(\mathbf{Q}, t_1, t_2) \end{aligned} \tag{4.40}$$

By measuring the signal (4.40) for different values of ϕ_0, an interferogram is obtained whose visibility is given by

$$\mathcal{V} = \frac{I_{max} - I_{min}}{I_{max} + I_{min}} = \frac{|S(\mathbf{Q}, t_1, t_2)|}{2 \sum_{l=1,2} S(\mathbf{Q}, t_l, t_l)} \tag{4.41}$$

The denominator in (4.41) can be estimated from the average value of the interferogram, being it

$$\bar{I}(t_1, t_2) = \frac{1}{2\pi} \int_0^{2\pi} d\phi_0 \, I(\phi_0, t_1, t_2) \propto \sum_{l=1,2} S(\mathbf{Q}, t_l, t_l) \tag{4.42}$$

and thus the modulus of the ISF can be extracted from the visibility of the interferogram
Moreover the argument of the ISF, that is the arcotangent of the ratio between its imaginary and real part, can also be obtained from (4.40). Indeed by subtracting to I its average value (4.42), the following signal is obtained

$$\begin{aligned} I(\phi_0, t_1, t_2) - \bar{I}(t_1, t_2) \propto \cos\left(\phi(t_1, t_2) + \phi_0\right) \operatorname{Re} S(\mathbf{Q}, t_1, t_2) + \\ - \sin\left(\phi(t_1, t_2) + \phi_0\right) \operatorname{Im} S(\mathbf{Q}, t_1, t_2) \end{aligned} \tag{4.43}$$

which vanishes for values of ϕ_0 given by

$$\phi_0^{(m)} = \arctan\left[\frac{\operatorname{Im} S(\mathbf{Q}, t_1, t_2)}{\operatorname{Re} S(\mathbf{Q}, t_1, t_2)}\right] - \phi(t_1, t_2) + m\pi \tag{4.44}$$

Therefore extrapolating the values of $\mathcal{V}$ and $\phi_0^{(m)}$ from data, an experimenter can extract both the real and the imaginary part of the ISF at a given pair of scattering times, obtaining such function entirely from the experimental data.
This reconstruction strategy works of course also when the target is known to be a classical system, and can be particularly useful to face the study of target systems out of equilibrium. Indeed in this context, the modelling of particles correlations can be a very though task and an experimental study of them could be of great help giving inputs for the theoretical understanding of non-equilibrium.

Chapter 5

Conclusions and Outlooks

Time-Domain interferometry with Mössbauer filtering foils (TDI) can access the dynamical pair correlation function (DCF) of condensed matter systems by probing the space Fourier transform of this function, known as the intermediate scattering function (ISF). This experimental technique has been demonstrated so far only for classical systems. Therefore, in the theoretical accounts of this experimental technique treat the probing radiation and the probed systems as classical systems.
The suggestion [2] that TDI could be suited to measure the ISF of systems that exhibit quantum features calls for an anlysis of this technique in a quantum theoretical framework, which still lacks in the current literature.

The special role played by the act of measurement in quantum theory requires special attention in the analysis of experimental schemes in which a quantum system is probed. This situation is even more delicate when one considers successive measurement on the same system, such as for measurements of dynamical correlation functions like the DCF.
In chapter 2 it has been illustrated that dynamical correlation functions between two non-commuting observables of a quantum system are in general complex valued, and that these functions can be given as a sum of two parts: a part that we called projective, which depends only on the probabilities that the observables have definite values, and a part that originates from the possibility that these values are in general undefined if the system is in a quantum superposition of states, and therefore will be called the coherent part.
Measuring dynamical correlations by direct action of the measurements ap-

paratus on the system can only give access to the projective part [24, **?**]. Therefore the projective part is immune to measurement backaction and the possibility has been investigated in [**?**] that the real part of dynamical correlations is always equal the projective part. In that work it has been concluded that this claim is true only if the correlated observables have two eigenvalues, equal in magnitude but with opposite signs, like spin-1/2 components.
Pushing this analysis further, it has been asked if a generic quantum system can be in a state for which the dynamical correlation between two non-commuting observables exactly equals the projective part. The result obtained is that only eigenstates of one of the observables have this property, as in that case the coherence part of the correlation function must be zero, as one would expect. From this we conclude that in general terms the quantum dynamical correlation functions are richer in information with respect to their projective part, and that direct measurements reduce the accessible information.
Therefore, in order to access to the complete dynamical correlation of quantum systems, the direct coupling of the measurement apparatus to the system has to be avoided. The two-time ancilla protocol proposed in the final part of chapter 1, is a model of experimental protocol that can accomplish this direct coupling. This protocol can be taken as a paradigm to understand the TDI technique when operated on quantum systems; moreover, it could serve as a guideline for devising new experiments for the measurement of quantum dynamical correlation functions.

In chapter 3 we considered the dynamical pair correlation function for an ensemble of particles, and its Fourier transforms. When these functions are defined for ensembles of classical particles, they enjoy some spatial and temporal symmetries. In particular the ISF enjoys hermitian symmetry with respect to reflection of the exchanged momentum (see chapter 2 for a definition of hermiticity).
This symmetries are no more granted when a quantum mechanical description of the particles is adopted. In particular, the appearence of an imaginary part of the DCF breaks the hermiticity of the ISF. In section 3.1.3 it is argued that the hermiticity of the ISF can be exploited in experiments to assess the quantum or classical nature of the particles composing a system. In particular it is concluded that if an experimenter finds a non-hermitian ISF, any classical model for the target can be ruled out.
On the contrary however, it is not correct to claim that the behaviour of a

system is classical only on the basis of the experimental finding of an hermitian ISF. In order to explicitly show this, two model systems of quantum particles hopping between the minima of potential wells are considered in section 3.2. One of the result of this work is the finding, for these two models, of superpositions of quantum states that give a vanishing imaginary part of the DCF, i.e. a hermitian ISF.
In [54] an interpretation of the imaginary part of the DCF was proposed: this quantity was intepreted as the local variation in the density of particles consequent to a weak external stimulus of the system. This would mean that the density of a system would not respond to small external perturbations, if its quantum state is such to make the imaginary part of the DCF vanish. From the theoretical side it would be then interesting to characterize these special states, to understand if and how they can be prepared in laboratories to investigate the interpretation above. Such theoretical studies could be supported experimentally, as nowadays it is possible to actually implement model hamiltonians – like the ones analysed in this work – which can simulate the behaviour of natural systems [55, 44].

Chapter 4 contains the second, major result of this work, that is the quantum theoretical analysis of TDI. In this chapter the general working principles of this experimental technique have been summarized, on the basis of which the quantum analysis has been built.
The main result of the analysis is that if TDI is applied to a quantum target, the whole quantum ISF of the target placed between the foils is accessed, and not just some parts of it.
This has been shown by calculating the probability in the unit time that a photon is absorbed by the detector – to which the recorded signal is proportional – after it is scattered by the target.
A photon going through the apparatus, can get to the detector by being scattered off of the target at two different times, and a quantum probability amplitude is associated to each of these scattering channels. Therefore the two amplitudes have been calculated explictly showing that they depend on the electrons density operator in the target at different times. However the two scattering channels can be distinguished in principle by the different times of arrival of the photon at the detector, and the final detection probability for the photon would then be the sum of the squares of the probability amplitudes, and would not depend on the ISF.
In this respect, we concluded that the second foil is necessary to erase the

information about the times of arrival, and make the two scattering channels indistinguishable. As a consequence, the detection probability is given by the modulus squared of the sum of the probability amplitudes, so that the signal at the detector contains an interference term which is given by a combination of both the real and the imaginary part of the quantum ISF of the target.
The possibility of of controlling the phase of one of the two scattering channels, demonstrated in [53], paves the way to new applications of TDI in condensed matter studies. In section 4.2.4 it has been proposed to use the phase control to verify the hermiticity of the ISF of a system (see chapter 2), and therefore to exclude classical models for it, and also to reconstruct the ISF from the experimental data.
These two proposals are different with respect to the applications of TDI realized so far, in which a model ISF is presumed for the target and the experimental data are used to extract the values of the carachteristic parameters of the model. On the contrary, the two proposed applications do not require any previous knowledge about the behaviour of the target, and in this respect they can help to explore correlations in systems whose modeling is difficult or at the moment non existent.
Few comments are in order about the model scheme used in section 4.2.2 to introduce our method for the analysis of TDI. In this scheme the two foils are replaced by two split and delay lines (SDL). In view of the development of SDLs in the hard-x part of the electromagnetic spectrum [45], a realization of this model scheme using hard-x radiation represents a real prospect to access correlations in condensed matter systems on shorter timescales, going from $\sim ps$ to $\sim ns$, than the one available with TDI that range from $\sim 10\,ns$ to $\sim 100ns$

Appendix A

Time-dependent perturbation theory

Let us assume that the hamiltonian of a quantum system can be given as a sum

$$\hat{H} = \hat{H}_0 + \hat{H}_I \tag{A.1}$$

where $\hat{H}_0$ is hereafter called the free hamiltonian and $\hat{H}_I$ is called the interaction.
We assume that the evolution operator associated with the free hamiltonian is known, that is that we know the solution $\hat{U}_0(t)$ to the following initial value problem

$$\begin{cases} i\hbar\frac{d}{dt}\hat{U}_0(t,t_0) = \hat{H}_0\hat{U}_0(t,t_0) \\ \\ \hat{U}_0(t_0,t_0) = \mathbb{1} \end{cases} \tag{A.2}$$

We then search the evolution operator corresponding to the full hamiltonian $\hat{H}$ as a product of the known $\hat{U}_0$ and a second unknown unitary operator $\hat{U}_I$. This product must therefore satisfy the following initial value problem

$$\begin{cases} i\hbar\frac{d}{dt}\Big(\hat{U}_0(t,t_0)\hat{U}_I(t,t_0)\Big) = \Big(\hat{H}_0 + \hat{H}_I\Big)\hat{U}_0(t,t_0)\hat{U}_I(t,t_0) \\ \\ \hat{U}_I(t_0,t_0) = \mathbb{1} \end{cases} \tag{A.3}$$

Substituting (A.2) into (A.3), and exploiting the unitarity of $\hat{U}_0$, we find an equation for $\hat{U}_I$

$$i\hbar\frac{d}{dt}\hat{U}_I(t,t_0) = \hat{U}_0(t,t_0)^\dagger \hat{H}_I \hat{U}_0(t,t_0)\hat{U}_I(t,t_0) \tag{A.4}$$

The formal solution of this equation, satisfying the initial condition in (A.3), is

$$\hat{U}_I(t,t_0) = \mathbb{1} - \frac{i}{\hbar}\int_{t_0}^{t} dt' \hat{U}_0(t',t_0)^\dagger \hat{H}_I \hat{U}_0(t',t_0)\hat{U}_I(t',t_0) \tag{A.5}$$

which can be iterated to obtain $\hat{U}_I$ as a formal infinite series of terms at increasing order in the interaction hamiltonian

$$\hat{U}_I(t,t_0) = \mathbb{1} + \sum_{n=1}^{\infty}\left(\frac{1}{i\hbar}\right)^n \int_{t_0}^{t} dt' \int_{t_0}^{t'} dt'' \cdots \int_{t_0}^{t^{(n-1)}} dt^{(n)} \hat{U}_0(t',t_0)^\dagger \hat{H}_I \hat{U}_0(t',t_0)$$
$$\hat{U}_0(t'',t_0)^\dagger \hat{H}_I \hat{U}_0(t'',t_0)\cdots \hat{U}_0(t^{(n)},t_0)^\dagger \hat{H}_I \hat{U}_0(t^{(n)},t_0) \tag{A.6}$$

Therefore the time evolution operator is

$$\hat{U}(t,t_0) = \hat{U}_0(t,t_0) + \hat{U}_0(t,t_0)\sum_{n=1}^{\infty}\left(\frac{1}{i\hbar}\right)^n \int_{t_0}^{t} dt' \int_{t_0}^{t'} dt'' \cdots \int_{t_0}^{t^{(n-1)}} dt^{(n)}$$
$$\hat{U}_0(t',t_0)^\dagger \hat{H}_I \hat{U}_0(t',t_0)\hat{U}_0(t'',t_0)^\dagger \hat{H}_I \hat{U}_0(t'',t_0)\cdots \hat{U}_0(t^{(n)},t_0)^\dagger \hat{H}_I \hat{U}_0(t^{(n)},t_0) \tag{A.7}$$

Appendix B

The quantized electromagnetic field

The set of Maxwell equations describing the dynamics of electromagnetic fields given a distribution of charges ρ_{ch} and currents $\mathbf{j}$ is

$$\begin{cases} \nabla \cdot \mathbf{E}(\mathbf{r},t) = \frac{\rho_{ch}(\mathbf{r},t)}{\epsilon_0} \\ \\ \nabla \cdot \mathbf{B}(\mathbf{r},t) = 0 \\ \\ \nabla \times \mathbf{E}(\mathbf{r},t) = -\frac{\partial \mathbf{B}}{\partial t}(\mathbf{r},t) \\ \\ \nabla \times \mathbf{B}(\mathbf{r},t) = \mu_0 \mathbf{j}(\mathbf{r},t) + \frac{1}{c^2}\frac{\partial \mathbf{E}}{\partial t}(\mathbf{r},t) \end{cases} \tag{B.1}$$

The second and third equations allow to define the vector and scalar potentials of the fields, respectively $\mathbf{A}$ and Φ, as these equations are automatically satisfied if one makes the substitution

$$\begin{cases} \mathbf{B}(\mathbf{r},t) = \nabla \times \mathbf{A}(\mathbf{r},t) \\ \\ \mathbf{E}(\mathbf{r},t) = -\nabla\Phi(\mathbf{r},t) - \frac{\partial \mathbf{A}}{\partial t}(\mathbf{r},t) \end{cases} \tag{B.2}$$

For ease of notation we will omit from now on the space-time dependecy of the fields and the potentials. With the introduction of the potentials the first

and fourth maxwell equations become

$$\begin{cases} \nabla^2\Phi + \frac{\partial}{\partial t}\nabla\cdot\mathbf{A} = -\frac{\rho_{ch}}{\epsilon_0} \\ \\ \nabla^2\mathbf{A} - \frac{1}{c^2}\frac{\partial^2\mathbf{A}}{\partial t^2} - \nabla\left[\nabla\cdot\mathbf{A} + \frac{1}{c^2}\frac{\partial\Phi}{\partial t}\right] = \mu_0\mathbf{j} \end{cases} \tag{B.3}$$

The equations (B.9) and (B.6) are invariant by the gauge transformation of potentials

$$\begin{cases} \mathbf{A}' = \mathbf{A} + \nabla\Lambda \\ \\ \Phi' = \Phi - \frac{\partial\Lambda}{\partial t} \end{cases} \tag{B.4}$$

this meaning that we can choose the gauge function Λ in such a way to simplify the equations (B.6). Many different choices of Λ are valid and often used in physics. The one used in this thesis is the Coulomb gauge, that is the gauge function is such that the vector potential is divergenceless

$$\nabla\cdot\mathbf{A}' = 0 \tag{B.5}$$

Omitting the apex, the dynamical equations in such a gauge are

$$\begin{cases} \nabla^2\Phi = -\frac{\rho_{ch}}{\epsilon_0} \\ \\ \nabla^2\mathbf{A} - \frac{1}{c^2}\frac{\partial^2\mathbf{A}}{\partial t^2} - \nabla\frac{1}{c^2}\frac{\partial\Phi}{\partial t} = \mu_0\mathbf{j} \end{cases} \tag{B.6}$$

As we are interested to the quantization of the free fields, we put $\rho_{ch} = 0$, $\mathbf{j} = 0$. This gives a constant scalar potential being

$$\Phi = \frac{1}{4\pi\epsilon_0}\int d^3r' \frac{\rho_{ch}}{|\mathbf{r}-\mathbf{r}'|} + const \tag{B.7}$$

and the only left equation is a wave equation for the vector potential

$$\nabla^2\mathbf{A} - \frac{1}{c^2}\frac{\partial^2\mathbf{A}}{\partial t^2} = 0 \tag{B.8}$$

with the electric and magnetic fields depending now only on $\mathbf{A}$

$$\begin{cases} \mathbf{B} = \nabla\times\mathbf{A} \\ \\ \mathbf{E} = -\frac{\partial\mathbf{A}}{\partial t} \end{cases} \tag{B.9}$$

The solution of equation (B.8) is easily obtained expressing $\mathbf{A}$ in terms of its Fourier transform

$$\mathbf{A} = \int d^3k \, \tilde{\mathbf{A}}_{\mathbf{k}} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} \tag{B.10}$$

with $\omega_k^2 = c^2|\mathbf{k}|^2$ Imposing that $\mathbf{A}$ must be a real vector and that it must satisfy the Coulomb gauge, one obtains that its Fourier transform satisfies the two conditions

$$\begin{aligned} \tilde{\mathbf{A}}_{\mathbf{k}} &= \tilde{\mathbf{A}}^*_{-\mathbf{k}} \\ \mathbf{k}\cdot\tilde{\mathbf{A}}_{\mathbf{k}} &= 0 \end{aligned} \tag{B.11}$$

in particular the second condition tells us that the vector $\tilde{\mathbf{A}}$ can be belongs to the plane perpendicular to $\mathbf{k}$. Choosing in this plane the basis of circular polarization $\{\mathbf{e}_{\mathbf{k},\sigma}\}_{\sigma=1,2}$ such that

$$\mathbf{e}_{\mathbf{k},\sigma}\cdot\mathbf{e}^*_{\mathbf{k},\sigma'} = \delta_{\sigma,\sigma'}, \ \mathbf{e}_{\mathbf{k},\sigma} = \mathbf{e}^*_{-\mathbf{k},\sigma} \tag{B.12}$$

we can re-express the Fourier transform $\tilde{\mathbf{A}}_{\mathbf{k}}$

$$\tilde{\mathbf{A}}_{\mathbf{k}} = \sum_\sigma \alpha_{\mathbf{k},\sigma}\mathbf{e}_{\mathbf{k},\sigma} \tag{B.13}$$

with $\alpha_{\mathbf{k},\sigma} = \alpha^*_{-\mathbf{k},\sigma}$, in order to satisfy the reality condition for $\mathbf{A}$

The quantization procedure for is based on the similarity of the hamiltoniana for the free electromagnetic field with the hamiltonian of a sum of independent harmonic oscillators. In order to make this similarity evident, we need to express the hamiltonian in terms of the amplitudes $\alpha_{\mathbf{k},\sigma}$. The hamiltonian for the free field is

$$H_{em} = \frac{1}{2}\int d^3r \ \epsilon_0|\mathbf{E}|^2 + \frac{|\mathbf{B}|^2}{\mu_0} \tag{B.14}$$

On the other hand the electric and magnetic field are expressed in terms of the $\tilde{\mathbf{A}}_{\mathbf{k}}$ as

$$\begin{aligned} \mathbf{E} &= -i\sum_\sigma \int d^3k \ \omega_k \left[\alpha_{\mathbf{k},\sigma}\mathbf{e}_{\mathbf{k},\sigma}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} - \alpha^*_{\mathbf{k},\sigma}\mathbf{e}^*_{\mathbf{k},\sigma}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}\right] \\ \mathbf{B} &= i\sum_\sigma \int d^3k \ \mathbf{k}\times\left[\alpha_{\mathbf{k},\sigma}\mathbf{e}_{\mathbf{k},\sigma}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} - \alpha^*_{\mathbf{k},\sigma}\mathbf{e}^*_{\mathbf{k},\sigma}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}\right] \end{aligned} \tag{B.15}$$

Substituting these forms for the fields into (B.14) and using the properties of the polarization vectors (B.12) and the identities $|\mathbf{k}|^2/\mu_0 = \epsilon_0\omega_k$, after the integration over the positions and one of the momenta we end up with the desired form for the hamiltonian

$$H_{em} = (2\pi)^3\epsilon_0 \sum_\sigma \int d^3k\ \omega_k^2\big(\alpha_{\mathbf{k},\sigma}\alpha^*_{\mathbf{k},\sigma} + \alpha^*_{\mathbf{k},\sigma}\alpha_{\mathbf{k},\sigma}\big) \tag{B.16}$$

the $(2\pi)^3$ coming from the integration over $\mathbf{r}$ of the exponentials, which gives delta functions that are subsequently integrated, and the order of multiplication of the amplitudes is kept in view of their substitution with non-commuting operators.
Evidently (B.16) is the hamiltonian of a continuum of harmonic oscillators associated with a given wavevector and polarization oscillating with frequency ω_k. The quantization is thus accomplished substituting the amplitudes and their complex conjugates of such oscillators with pairs of operators

$$\begin{aligned} \alpha_{\mathbf{k},\sigma} &\longrightarrow \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0\omega_k}}\hat{a}_{\mathbf{k},\sigma} \\ \alpha^*_{\mathbf{k},\sigma} &\longrightarrow \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0\omega_k}}\hat{a}^\dagger_{\mathbf{k},\sigma} \end{aligned} \tag{B.17}$$

whose commutators are assigned in analogy with the ones for the destruction and creation operators of independent quantum harmonic oscillators

$$[\hat{a}_{\mathbf{k},\sigma}, \hat{a}^\dagger_{\mathbf{k}',\sigma'}] = \delta(\mathbf{k}-\mathbf{k}')\delta_{\sigma,\sigma'} \tag{B.18}$$

With the introduction of these operators we also need to introduce a space of quantum states for the free electromagnetic field. This can be done by constructing a suitable basis for such a space. From the theory of quantum harmonic oscillators, we know that the number operator $\hat{n}_{\mathbf{k},\sigma} \equiv \hat{a}^\dagger_{\mathbf{k},\sigma}\hat{a}_{\mathbf{k},\sigma}$ has a discrete spectrum corresponding to the set of natural numbers. The eigenvector corresponding to the null eigenvalue is called vacuum and is defined by the condition

$$\hat{a}_{\mathbf{k},\sigma}|0\rangle = 0 \quad \forall\ \mathbf{k},\sigma \tag{B.19}$$

and the eigenvector of $\hat{n}_{\mathbf{k},\sigma}$ with eigenvalue m is obtained through the action of the creation operator on the vacuum

$$|m_{\mathbf{k},\sigma}\rangle = \frac{\left(\hat{a}^{\dagger}_{\mathbf{k},\sigma}\right)^m}{\sqrt{m!}}|0\rangle \tag{B.20}$$

Varying m in the whole set of natural numbers one obtains the basis for the space of quantum states $\mathcal{H}_{\mathbf{k},\sigma}$ of the $\mathbf{k},\sigma$-th oscillator. The whole state space is then given by the union

$$\mathcal{H}_{em} = \bigcup_{\mathbf{k},\sigma} \mathcal{H}_{\mathbf{k},\sigma} \tag{B.21}$$

which is referred to as the Fock space.

The hamiltonian and linear momentum of the field in terms of the creation and destruction operators are

$$\hat{H}_{em} = \sum_{\sigma} \int d^3k \; \hbar\omega_k \left(\hat{n}_{\mathbf{k},\sigma} + \frac{1}{2}\right) \tag{B.22}$$

$$\hat{\mathbf{P}}_{em} = \sum_{\sigma} \int d^3k \; \hbar\mathbf{k}\, \hat{n}_{\mathbf{k},\sigma} \tag{B.23}$$

Evidently the eigenstates of both these operators are the number eigenstates: $|m_{\mathbf{k},\sigma}\rangle$ is then to be interpreted as a configuration of the free electromagnetic field made up of m excitations of its $(\mathbf{k},\sigma)$ normal mode each carrying an energy $\hbar\omega_k$ and a momentum $\hbar\mathbf{k}$, that is of m photons.

The quantized vector potential reads

$$\hat{\mathbf{A}} = \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0}} \sum_{\sigma} \int d^3k \; \frac{1}{\sqrt{\omega_k}} \left[\hat{a}_{\mathbf{k},\sigma}\mathbf{e}_{\mathbf{k},\sigma} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} + \hat{a}^{\dagger}_{\mathbf{k},\sigma}\mathbf{e}_{\mathbf{k},\sigma} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} \right] \tag{B.24}$$

The electric field operator in terms of the creation and destruction operator is

$$\hat{\mathbf{E}} = \hat{\mathbf{E}}^{(+)} + \hat{\mathbf{E}}^{(-)} \tag{B.25}$$

where

$$\hat{\mathbf{E}}^{(+)} = -i\sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0}}\sum_{\sigma}\int d^3k\,\sqrt{\omega_k}\,\hat{a}_{\mathbf{k},\sigma}\mathbf{e}_{\mathbf{k},\sigma}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}$$
$$\hat{\mathbf{E}}^{(-)} = i\sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0}}\sum_{\sigma}\int d^3k\,\sqrt{\omega_k}\,\hat{a}^{\dagger}_{\mathbf{k},\sigma}\mathbf{e}_{\mathbf{k},\sigma}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} \tag{B.26}$$

Appendix C

Integration of geometric factors

In chapter 3 we had to calculate the following integral

$$\sum_{\sigma} \int_V d^3r \hat{\rho}(\mathbf{r}, t_l)|\psi\rangle \int d^3q \, (\mathbf{e}_{\mathbf{q},\sigma} \cdot \mathbf{e}_{\sigma_0}) \mathbf{e}_{\mathbf{q},\sigma} \, e^{i[\mathbf{q}\cdot(\mathbf{R}-\mathbf{r})-\omega_q t]} \int_{t_0}^{t} dt' f\left(t' - t_l - \frac{z}{c}\right) e^{i\omega_q t'} e^{-i\omega_0 \left(t' - t_l - \frac{z}{c}\right)} \tag{C.1}$$

and we started with the integration over the different directions of the $\mathbf{q}$ wavevector. There only the final result was shown with the understanding that only radiative terms are kept, that is terms whose square is proportional to the inverse square of the distance between the source (in our case the target) and the detection point.
Here we show that indeed the integration over $\mathbf{q}$'s direction produces non-radiative terms. The integral of our interest is

$$\int d^3q \sum_{\sigma} (\mathbf{e}_{\mathbf{q},\sigma} \cdot \mathbf{e}_{\sigma_0}) \mathbf{e}_{\mathbf{q},\sigma} e^{i\mathbf{q}\cdot(\mathbf{R}-\mathbf{r})} \tag{C.2}$$

The polarization terms in the integral (C.2) can be rewritten expliting the fact that the vectors $\mathbf{e}_{\mathbf{q},\sigma}$ and the direction of momentum $\tilde{\mathbf{q}}$ form a basis of the ordinary space, and thus $\sum_\sigma (\mathbf{e}_{\mathbf{q},\sigma} \cdot \mathbf{e}_{\sigma_0}) \mathbf{e}_{\mathbf{q},\sigma}) = \mathbf{e}_{\sigma_0} - \tilde{\mathbf{q}}(\tilde{\mathbf{q}} \cdot \mathbf{e}_{\sigma_0})$.
For the calculation we introduce an auxiliary reference frame X, Y, Z, whose Z axis is parallel to the $|\mathbf{R} - \mathbf{r}| = Z$ vector and adopt spherical coordinates in this frame.
The expression of polarization and of the versor $\tilde{\mathbf{q}}$ in these new reference

frame (see figure C.1) is then

$$\begin{aligned} \mathbf{e}_{\sigma_0} &= \sin\theta_0 \cos\phi_0 \hat{\mathbf{X}} + \sin\theta_0 \sin\phi_0 \hat{\mathbf{Y}} + \cos\theta_0 \hat{\mathbf{Z}} \\ \tilde{\mathbf{q}} &= \sin\theta_q \cos\phi_q \hat{\mathbf{X}} + \sin\theta_q \sin\phi_q \hat{\mathbf{Y}} + \cos\theta_q \hat{\mathbf{Z}} \end{aligned} \tag{C.3}$$

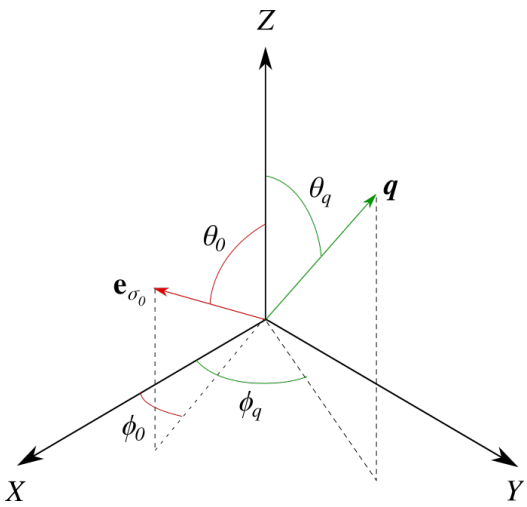

Figure C.1: Auxiliary reference frame used in the calculation of angular integral.

The angular part of the integral (C.2) is then

$$
\begin{aligned}
&2\pi \mathbf{e}_{\sigma_0} \int_0^\pi d\theta_q \sin\theta_q e^{iqZ\cos\theta_q} + \\
&- \hat{\mathbf{X}} \Bigg(\sin\theta_0 \int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \sin^3\theta_q \int_0^{2\pi} d\phi_q \cos\phi_q \cos(\phi_q - \phi_0) + \\
&+ \cos\theta_0 \int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \sin^2\theta_q \cos\theta_q \int_0^{2\pi} d\phi_q \cos\phi_q \Bigg) + \\
&- \hat{\mathbf{Y}} \Bigg(\sin\theta_0 \int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \sin^3\theta_q \int_0^{2\pi} d\phi_q \sin\phi_q \cos(\phi_q - \phi_0) + \\
&+ \cos\theta_0 \int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \sin^2\theta_q \cos\theta_q \int_0^{2\pi} d\phi_q \sin\phi_q \Bigg) + \\
&- \hat{\mathbf{Z}} \Bigg(\sin\theta_0 \int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \sin^2\theta_q \cos\theta_q \int_0^{2\pi} d\phi_q \cos(\phi_q - \phi_0) + \\
&+ 2\pi \cos\theta_0 \int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \cos^2\theta_q \sin\theta_q \Bigg)
\end{aligned}
\tag{C.4}
$$

all the terms containing an integral of a single sine or cosine function of ϕ_q vanish, whereas the only non-vanishing integrals in the last expressions are

$$
\int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \sin\theta_q = 2\frac{\sin(qZ)}{qZ} \tag{C.5}
$$

$$
\int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \sin^3\theta_q = 4\left[\frac{\sin(qZ)}{(qZ)^3} - \frac{\cos(qZ)}{(qZ)^2}\right] \tag{C.6}
$$

$$
\int_0^\pi d\theta_q e^{iqZ\cos\theta_q} \cos^2\theta_q \sin\theta_q = 2\frac{\sin(qZ)}{qZ} - 4\left[\frac{\sin(qZ)}{(qZ)^3} - \frac{\cos(qZ)}{(qZ)^2}\right] \tag{C.7}
$$

$$
\int_0^{2\pi} d\phi_q \cos\phi_q \cos(\phi_q - \phi_0) = \pi\cos\phi_0 \tag{C.8}
$$

$$
\int_0^{2\pi} d\phi_q \sin\phi_q \cos(\phi_q - \phi_0) = \pi\sin\phi_0 \tag{C.9}
$$

Grouping the terms in the angular integral by descending power of Z, we obtain that (C.4) is

$$\begin{aligned} &4\pi(\mathbf{e}_{\sigma_0} - \cos\theta_0\hat{\mathbf{Z}})\frac{\sin(qZ)}{qZ} + \\ &+ 4\pi\big[\sin\theta_0(\cos\phi_0\hat{\mathbf{X}} + \sin\phi_0\hat{\mathbf{Y}}) - 2\cos\theta_0\hat{\mathbf{Z}}\big]\left[\frac{\cos(qZ)}{(qZ)^2} - \frac{\sin(qZ)}{(qZ)^3}\right] \end{aligned} \tag{C.10}$$

of which only the radiative term of order Z^{-1} is kept in the results of chapter 4.

C.1 The polarization factor

It is useful checking to what amounts to the vector factor $\mathbf{e}_{\sigma_0} - \cos\theta_0\hat{\mathbf{Z}}$ of the radiative contribution to the angular integral. In order to do this we analyze two cases for the incoming polarization.
In the first case the polarization, see figure C.2a, $\mathbf{e}_{\sigma_0}$ is orthogonal to the plane containing the axes z and Z. This means that $\theta_0 = \pi/2$, that the vector factor reduces to $\mathbf{e}_{\sigma_0}$.
The second case, see figure C.2b, is the one of $\mathbf{e}_{\sigma_0}$ contained in the z, Z plane. If we call the observation angle α, that is the angle between z and Z, in this special case it holds that $\theta_0 = \pi/2 - \alpha$ and the vector factor reduces to

$$\cos\alpha(\cos\phi\hat{\mathbf{X}} + \sin\phi\hat{\mathbf{Y}}) \tag{C.11}$$

whose modulus square is $\cos^2\alpha$
The case of generic incoming polarization will be a sum of these two possibilities, that is if s and $1-s$ are the perecentages of polarization respectively orthogonal and parallel to the z, Z plane, the polarization factor appearing in the detection rate is

$$s\sin^2\alpha + \cos^2\alpha \tag{C.12}$$

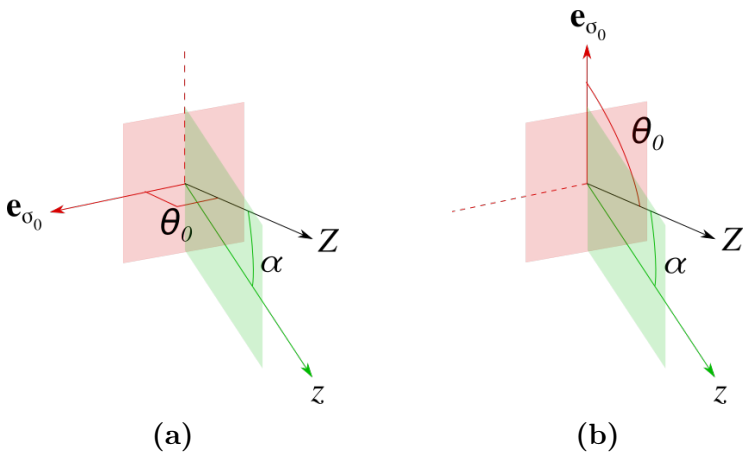

Figure C.2: The two possible incoming polarization cases: (a) polarization orthogonal to the z, Z plane (in green), (b) polarization parallel to the z, Z plane. The red plane represents the polarization plane

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