Effect of spin-orbit coupling on the high harmonics from the topological Dirac semimetal Na_3Bi: Supplementary Information

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I. EQUATION OF MOTION OF THE CHARGE CURRENT

As explained in the main text, we start from the single-particle Pauli Hamiltonian describing a particle of mass m and charge e in an external electromagnetic field,

$$\hat{h}(t) = \frac{1}{2m}\hat{\mathbf{\Pi}}^2(t) - eV(t) + \frac{e\hbar}{2mc}\mathbf{B}(t).\hat{\boldsymbol{\sigma}} + \frac{e\hbar}{8m^2c^2}\left[\hat{\mathbf{\Pi}}(t).(\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)) + (\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)).\hat{\mathbf{\Pi}}(t)\right], \quad (1)$$

While the previous Hamiltonian has been considered in many theoretical work, it is possible to obtain a local $U(1)\times SU(2)$ symmetry by including a term of the higher order $O(1/m^3)$ as explained in Ref. 1. The motivation for this choice lies in the fact that this will allow us later to define a $U(1)\times SU(2)$ gauge-invariant spin-current, which we defined in this work as the physical spin current. For this, we add the term $\frac{e^2\hbar^2}{32m^3c^4}(\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)).(\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)) = \frac{e^2\hbar^2}{16m^3c^4}|\mathbf{E}(t)|^2$, which is half of the corresponding term in the Foldy-Wouthuysen expansion [1]. Doing so, one arrives to a locally $U(1)\times SU(2)$ gauge-invariant single-particle Hamiltonian

$$\hat{h}(t) = \frac{1}{2m}\left[\hat{\mathbf{\Pi}}(t) + \frac{e\hbar}{4mc^2}(\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t))\right]^2 - eV(t) + \frac{e\hbar}{2mc}\mathbf{B}(t).\hat{\boldsymbol{\sigma}}, \quad (2)$$

which is the one we consider in the following.

In second quantization, all operators are expressed in terms of the field operators $\hat{\psi}(\mathbf{x})$ and $\hat{\psi}^\dagger(\mathbf{x})$. The many-body Hamiltonian $\hat{H}(t)$ consists of the time-dependent one-body part $\hat{h}(t)$ and the particle-particle interaction term $w(\mathbf{x},\mathbf{x'})$, and reads as

$$\hat{H}(t) = \sum_{\sigma\sigma'}\int d\mathbf{r}\hat{\psi}^\dagger(\mathbf{r},\sigma)h_{\sigma\sigma'}(\mathbf{r},t)\hat{\psi}(\mathbf{r},\sigma') + \frac{1}{2}\int d\mathbf{x}\int d\mathbf{x'}w(\mathbf{x},\mathbf{x'})\hat{\psi}^\dagger(\mathbf{x})\hat{\psi}^\dagger(\mathbf{x'})\hat{\psi}(\mathbf{x'})\hat{\psi}(\mathbf{x}). \quad (3)$$

We now define the operators

$$\begin{aligned}
\hat{t}(\mathbf{x}) &= \frac{\hbar^2}{2m}(\boldsymbol{\nabla}\hat{\psi}(\mathbf{x}))^\dagger(\boldsymbol{\nabla}\hat{\psi}(\mathbf{x})),\\
\hat{n}(\mathbf{x}) &= \hat{\psi}^\dagger(\mathbf{x})\hat{\psi}(\mathbf{x}),\\
\hat{\mathbf{m}}(\mathbf{x}) &= \hat{\psi}^\dagger(\mathbf{x})\hat{\boldsymbol{\sigma}}\hat{\psi}(\mathbf{x}),\\
\hat{\mathbf{j}}(\mathbf{x}) &= -\frac{i\hbar}{2m}\left[\hat{\psi}^\dagger(\mathbf{x})(\boldsymbol{\nabla}\hat{\psi}(\mathbf{x})) - (\boldsymbol{\nabla}\hat{\psi}(\mathbf{x}))^\dagger\hat{\psi}(\mathbf{x})\right],\\
\hat{\mathbf{J}}(\mathbf{x}) &= -\frac{i\hbar}{2m}\left[\hat{\psi}^\dagger(\mathbf{x})\hat{\boldsymbol{\sigma}}(\boldsymbol{\nabla}\hat{\psi}(\mathbf{x})) - (\boldsymbol{\nabla}\hat{\psi}(\mathbf{x}))^\dagger\hat{\boldsymbol{\sigma}}\hat{\psi}(\mathbf{x})\right],
\end{aligned}$$

which are respectively the kinetic energy, density, magnetization, current and spin-current operators.
In order to derive the equation of motion of the different observable, we start by writing the equation of motion of the creation and annihilation operators in the Heisenberg picture[2]

$$i\hbar\frac{d}{dt}\hat{\psi}(\mathbf{x},t) = \sum_{\sigma'}h_{\sigma\sigma'}(\mathbf{x},-i\overrightarrow{\boldsymbol{\nabla}},t)\hat{\psi}(\mathbf{x},\sigma',t) + \int d\mathbf{x'}w(\mathbf{x},\mathbf{x'})\hat{n}(\mathbf{x'})\hat{\psi}(\mathbf{x},t), \quad (4)$$

and

$$i\hbar\frac{d}{dt}\hat{\psi}^\dagger(\mathbf{x},t) = -\sum_{\sigma'}\hat{\psi}^\dagger(\mathbf{x},\sigma',t)h_{\sigma'\sigma}(\mathbf{x},i\overleftarrow{\boldsymbol{\nabla}},t) - \int d\mathbf{x'}w(\mathbf{x},\mathbf{x'})\hat{\psi}^\dagger(\mathbf{x},t)\hat{n}(\mathbf{x'}). \quad (5)$$

Following Ref. 2, we split the equation of motion of the field operators in terms of a contribution without any external potential nor field, and the part containing them.

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We obtain that

$$i\hbar\frac{d}{dt}\hat{\psi}(\mathbf{x},t) = i\hbar\frac{d}{dt}\hat{\psi}(\mathbf{x},t)\Big|_{V=\mathbf{A}=0} + \frac{ie}{2mc}\sum_p\Big[2A_p(t)\partial_p + (\partial_p A_p(t))\Big]\hat{\psi}(\mathbf{x},t) + u\hat{\psi}(\mathbf{x},t)$$
$$+\frac{e\hbar}{8m^2c^2}\left[\hat{\mathbf{\Pi}}(t).\,(\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)) + (\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)).\,\hat{\mathbf{\Pi}}(t)\right]\hat{\psi}(\mathbf{x},t) + \frac{e\hbar}{2mc}\mathbf{B}(t).\,\hat{\boldsymbol{\sigma}}\hat{\psi}(\mathbf{x},t)\,, \tag{6}$$

and

$$i\hbar\frac{d}{dt}\hat{\psi}^\dagger(\mathbf{x},t) = i\hbar\frac{d}{dt}\hat{\psi}^\dagger(\mathbf{x},t)\Big|_{V=\mathbf{A}=0} + \frac{ie}{2mc}\sum_p\Big[2A_p(t)\partial_p + (\partial_p A_p(t))\Big]\hat{\psi}^\dagger(\mathbf{x},t) - u\hat{\psi}(\mathbf{x},t)$$
$$-\frac{e\hbar}{8m^2c^2}\left[\hat{\mathbf{\Pi}}^*(t).\,(\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)) + (\hat{\boldsymbol{\sigma}}\times\mathbf{E}(t)).\,\hat{\mathbf{\Pi}}^*(t)\right]\hat{\psi}^\dagger(\mathbf{x},t) - \frac{e\hbar}{2mc}\mathbf{B}(t).\,\hat{\boldsymbol{\sigma}}\hat{\psi}^\dagger(\mathbf{x},t)\,, \tag{7}$$

where we defined

$$u = -eV(\mathbf{x},t) + \frac{e^2}{2mc^2}|\mathbf{A}(\mathbf{x},t)|^2 - \frac{e^2\hbar^2}{16m^3c^4}|\mathbf{E}(\mathbf{x},t)|^2\,. \tag{8}$$

Before obtaining the equation of motion for the physical charge current, we first need to define it. Such definition can be obtained from the continuity equation, which defines the conserved charge current for a given Hamiltonian. After some algebra, we arrive to the equation of motion of the charge density, the continuity equation,

$$\frac{\partial}{\partial t}\hat{n}(\mathbf{x},t) = -\boldsymbol{\nabla}.\,(\hat{\mathbf{j}}(\mathbf{x},t) + \hat{\mathbf{j}}_d(\mathbf{x},t) + \hat{\mathbf{j}}_{\mathrm{SO}}(\mathbf{x},t))\,. \tag{9}$$

where the physical, conserved, charge current in presence of spin-orbit coupling, contains the paramagnetic current ($\hat{\mathbf{j}}$), the diamagnetic current ($\hat{\mathbf{j}}_d$), and a spin-orbit current, $\hat{\mathbf{j}}_{\mathrm{SO}}(\mathbf{x},t) = \frac{e\hbar}{4m^2c^2}\hat{\mathbf{m}}\times\mathbf{E}(t)$.
This equation determines the physical conserved current up to a rotational part. However, as we are interested in the time-derivative of the macroscopic current (source term of the Maxwell equations), this rotational part of the current is irrelevant, and in the following, we use Eq. 9 to define the physical current associated with the Hamiltonian of Eq. 2.
In order to obtain the equation of the conserved total physical current $\hat{\mathbf{j}}_{\mathrm{phys}} = \hat{\mathbf{j}} + \hat{\mathbf{j}}_d + \hat{\mathbf{j}}_{\mathrm{SO}}$, we need the equation of motion of the magnetization density and of the paramagnetic current. Following the same approach as for the charge density, we obtain the equation of motion of the k component of the magnetization density

$$\frac{\partial}{\partial t}\hat{m}_k(\mathbf{x},t) = -\sum_p\partial_p\left[\hat{J}_{\mathrm{phys},kp}\right] + \frac{e}{mc}\sum_{qr}\epsilon_{kqr}B_q\hat{m}_r + \frac{e}{2mc^2}\sum_p\left[E_p\hat{J}_{\mathrm{phys},pk} - E_k\hat{J}_{\mathrm{phys},pp}\right], \tag{10}$$

where, by analogy to the continuity equation for the charge, we can defined a physical spin current as

$$\hat{J}_{\mathrm{phys},kp} = \left[\hat{J}_{kp} + \frac{e}{mc}\hat{m}_kA_p - \frac{e\hbar}{4m^2c^2}\sum_r\epsilon_{kpr}E_r\hat{n}_H\right], \tag{11}$$

where we recognize a paramagnetic, a diamagnetic, and a spin-orbit spin current. The motivation for this definition lies in the fact that the spin current defined in Eq. 11 is a mixed space and spin quantity, which is for our definition locally $U(1)\times SU(2)$ gauge invariant. This symmetry is the same as the one of the Hamiltonian of Eq. 2 and motivates us to use this expression to define the physical spin current, in the same way that the physical charge current, which is a spatial quantity only, is invariant under $U(1)$ gauge transformation. Here ϵ_{ijk} denotes the Levi-Civita symbol.
The equation of motion of the paramagnetic current is found to be

$$\begin{aligned}\frac{\partial}{\partial t}\hat{j}_k(\mathbf{x},t) &= -\sum_p\partial_p\hat{T}_{pk} - \hat{W}_k \\ &-\frac{1}{m}(\partial_k w(\mathbf{x},t))\hat{n}(\mathbf{x},t) - \frac{e\hbar}{2m^2c}\sum_p\partial_k(B_p)\hat{m}_p \\ &-\frac{e}{mc}\sum_p\Bigg[\partial_p\Big(A_p\hat{j}_k + \frac{\hbar}{4mc}\sum_{q,r}\epsilon_{pqr}E_r\hat{J}_{qk}\Big) \\ &+(\partial_kA_p)\Big[\hat{j}_p + \frac{e\hbar}{4m^2c^2}\sum_{q,r}\epsilon_{pqr}\hat{m}_qE_r\Big] \\ &+\frac{\hbar}{4mc}\sum_{q,r}\epsilon_{pqr}(\partial_kE_r)\Big[\hat{J}_{qp} + \frac{e}{mc}A_p\hat{m}_q\Big]\Bigg],\end{aligned} \tag{12}$$

where $\hat{T}_{pk}$ is the so-called momentum-stress tensor [2]

$$\hat{T}_{pk} = \frac{1}{2m^2}\left[(\partial_k\hat{\psi}^\dagger)(\partial_p\hat{\psi}) + (\partial_p\hat{\psi}^\dagger)(\partial_k\hat{\psi}) - \frac{1}{2}\partial_k\partial_p\hat{n}\right], \tag{13}$$

while the operator $\hat{W}_k$ is defined by [2]

$$\hat{W}_k(\mathbf{x},t) = \frac{1}{m}\int d\mathbf{x'}\hat{\psi}^\dagger(\mathbf{x})\hat{\psi}^\dagger(\mathbf{x'})(\partial_k v(\mathbf{x}-\mathbf{x'})\hat{\psi}(\mathbf{x'})\hat{\psi}(\mathbf{x})\,. \tag{14}$$

Putting everything together, we get the following expression for the equation of motion of the physical charge current

$$\frac{\partial}{\partial t}\hat{j}_{\mathrm{phys},k}(\mathbf{x},t) = -\sum_p \partial_p\Big[\hat{T}_{pk} + \frac{e}{mc}(A_p\hat{j}_k + \hat{j}_{\mathrm{phys},p}A_k) + \frac{e\hbar}{4m^2c^2}\sum_{qr}(\epsilon_{kqr}\hat{J}_{\mathrm{phys},pq}E_r + \epsilon_{pqr}\hat{J}_{qk}E_r)\Big]$$
$$-\hat{W}_k - \frac{e^2\hbar}{4m^3c^3}\left[(\mathbf{m}_H\times\mathbf{B})\times\mathbf{E}\right]_k$$
$$-\frac{e}{m}\Big[\hat{n}\mathbf{E} + \frac{1}{c}\hat{\mathbf{j}}_{\mathrm{phys}}\times\mathbf{B}\Big]_k + \frac{e\hbar}{4m^2c^2}\Big[\hat{\mathbf{m}}\times\partial_t\mathbf{E}\Big]_k - \frac{e\hbar}{2m^2c}\sum_p \hat{m}_p\Big(\partial_k(B_p)\Big)$$
$$+\frac{e\hbar}{4m^2c^2}\sum_{pqr}\hat{J}_{\mathrm{phys},pq}\left[\epsilon_{kpr}\partial_q - \epsilon_{qpr}\partial_k + \epsilon_{kqr}\frac{e}{2mc^2}E_p\right](E_r)\,. \tag{15}$$

We note that the conserved current is only defined up to a rotational part in the above discussion, and we need to add to it the magnetization current $\hat{\mathbf{j}}_m = \frac{\hbar}{2m}\boldsymbol{\nabla}\times\hat{\mathbf{m}}$. This current only contributes to the microscopic current, and not to the macroscopic current. Moreover, adding the contribution from the magnetization current, only adds a term to the stress tensor, as

$$\frac{d}{dt}\hat{j}_{m,k}(\mathbf{x},t) = \Bigg[-\sum_p \partial_p\frac{\hbar}{2m}\sum_{st}\partial_s\epsilon_{kst}\Big[\hat{J}_{\mathrm{phys},tp}\Big]$$
$$+\frac{e\hbar}{2m^2c}\sum_p \partial_p(B_k\hat{m}_p - B_p\hat{m}_k) + \frac{e\hbar}{4m^2c^2}\sum_{pqr}\partial_p\epsilon_{kpq}\Big[E_r\hat{J}_{\mathrm{phys},rq} - E_q\hat{J}_{\mathrm{phys},rr}\Big]\Bigg]$$
$$= -\sum_p \partial_p\hat{M}_{pk}\,. \tag{16}$$

which implies that the magnetization current does not contribute to the radiated light.

Supplementary References

[1] J. Fröhlich and U. M. Studer, Rev. Mod. Phys. **65**, 733 (1993).

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