

On the theory of microwave absorption by the spin-1/2 Heisenberg-Ising magnet

Michael Brockmann, Frank Göhmann, Michael Karbach, and Andreas Klümper
Fachbereich C – Physik, Bergische Universität Wuppertal, 42097 Wuppertal, Germany

Alexander Weiße

Max-Planck-Institut für Mathematik, P.O. Box 7280, 53072 Bonn, Germany

We analyze the problem of microwave absorption by the Heisenberg-Ising magnet in terms of shifted moments of the imaginary part of the dynamical susceptibility. When both, the Zeeman field and the wave vector of the incident microwave, are parallel to the anisotropy axis, the first four moments determine the shift of the resonance frequency and the line width in a situation where the frequency is varied for fixed Zeeman field. For the one-dimensional model we can calculate the moments exactly. This provides exact data for the resonance shift and the line width at arbitrary temperatures and magnetic fields. In current ESR experiments the Zeeman field is varied for fixed frequency. We show how in this situation the moments give perturbative results for the resonance shift and for the integrated intensity at small anisotropy as well as an explicit formula connecting the line width with the anisotropy parameter in the high-temperature limit.

The total magnetization in an isotropic system of interacting spins rotates as a whole about the axis of a homogeneous external field (see e.g. [1]). We consider L spins- $\frac{1}{2}$, combining to a total spin $\mathbf{S} = S^x \mathbf{e}_x + S^y \mathbf{e}_y + S^z \mathbf{e}_z$, $S^\alpha = \sum_{j=1}^L s_j^\alpha$, in a magnetic field of strength h in z -direction. The Heisenberg equation of motion for \mathbf{S} with a Zeeman term $-hS^z$ is solved by $\mathbf{S}(t) = (\cos(ht)S^x + \sin(ht)S^y)\mathbf{e}_x - (\sin(ht)S^x - \cos(ht)S^y)\mathbf{e}_y + S^z \mathbf{e}_z$, a rotation counterclockwise about the z -axis. In ESR experiments this can be probed by circularly polarized microwave radiation propagating along the z -direction. Since its wavelength is large compared to typical distances in spin systems, we may assume a magnetic field component of the form $\mathbf{h}(t) = A(\cos(\omega t)\mathbf{e}_x - \sin(\omega t)\mathbf{e}_y)$, $A > 0$. It couples to the total spin as $V(t) = -h^\alpha(t)S^\alpha$ and produces a sharp resonance at $\omega = h$ (see (4) below). If the spin system is perturbed by anisotropic interactions, this resonance is broadened, shifted or even split in a way that is characteristic of the microscopic interactions between the spins.

For any spin system with Hamiltonian H linear response theory relates the observed absorbed intensity to the (imaginary part of the) dynamical susceptibility [2]

$$\chi''_{+-}(\omega, h) = \frac{1}{2L} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle [S^+(t), S^-] \rangle_T. \quad (1)$$

Here $S^\pm = S^x \pm iS^y$ and $\langle \cdot \rangle_T$ stands for the canonical average at temperature T calculated by means of the statistical operator $\rho = e^{-(H-hS^z)/T} / \text{tr} e^{-(H-hS^z)/T}$. Through this average the dynamical susceptibility depends on h and T . The absorbed intensity per spin, normalized by the intensity A^2 of the incident wave and averaged over a half-period π/ω of the microwave field, is

$$I(\omega, h) = \frac{\omega}{2} \chi''_{+-}(\omega, h). \quad (2)$$

In current ESR experiments in solids $I(\omega, h)$ is measured as a function of h for fixed ω . Of particular interest are experiments on quasi one-dimensional compounds (reviewed e.g. in [3, 4]) which provide prototypical realizations of interacting

many-body systems with strong quantum fluctuations. Still, the data are not always easy to interpret, because of a lack of reliable theoretical predictions.

Most of the existing theories are based on a priori assumptions about the line shape and typically apply for limited ranges of temperature and magnetic field. Field theoretical approaches [1] are restricted to small temperatures and small (but not too small) magnetic fields. The more traditional approaches [2, 5] rely on the high temperature approximation. Purely numerical approaches [6] are unbiased, but the extrapolation of the data to the thermodynamic limit of large chains may be difficult.

A remarkable result for the resonance shift in one-dimensional antiferromagnetic chains, being valid at arbitrary temperatures, was obtained in [7]. It utilizes the exact nearest-neighbor correlation functions of the isotropic spin- $\frac{1}{2}$ Heisenberg chain. Here we present an alternative framework for the derivation of the resonance shift which, in the limit of small anisotropy, reproduces [7]. In our approach the anisotropy is treated non-perturbatively, and it allows us to derive an exact formula for the line width ‘in frequency direction’ at fixed magnetic field, as well as a new explicit expression for the ESR-line width in the high temperature regime.

We consider an important example of anisotropic interactions described by the Heisenberg-Ising Hamiltonian

$$H = J \sum_{\langle ij \rangle} (s_i^x s_j^x + s_i^y s_j^y + (1 + \delta) s_i^z s_j^z). \quad (3)$$

Here the sum is over nearest neighbors, and δ is the anisotropy parameter. If $\delta = 0$, then (1), (2) imply that the normalized absorbed intensity is

$$I(\omega, h) = \pi \delta(\omega - h) h m(T, h). \quad (4)$$

It is proportional to the magnetic energy $hm(T, h)$ per lattice site. This case includes the familiar paramagnetic resonance (Zeeman effect) for $J = 0$, for which the magnetization is $m(T, h) = \frac{1}{2} \text{th}(\frac{h}{2T})$.

For non-zero δ the function χ''_{+-} is unknown and hard to calculate. Still, some more elementary spectral characteristics,

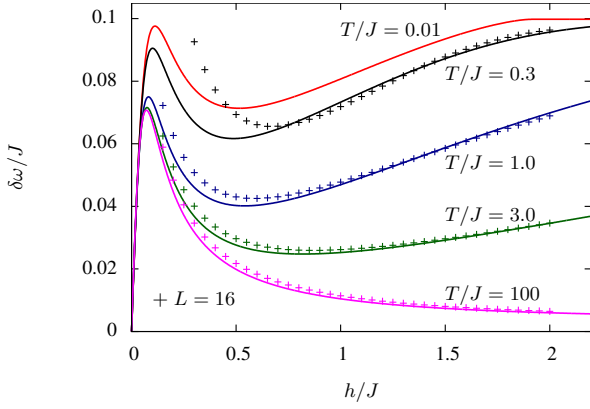


FIG. 1. Resonance shift $\delta\omega/J$ for the 1D model in the critical regime at $\delta = -0.1$ as function of the magnetic field. Crosses from fully numerical calculation for a finite chain Hamiltonian of 16 sites.

such as the position of the resonance or the line width, may be expressed in terms of certain static correlation functions that determine the moments of the normalized intensity function.

Let us assume for a while that our chain is large but finite. Then the spectrum is bounded and the integrals

$$I_n = \int_{-\infty}^{\infty} d\omega \omega^n I(\omega, h) \quad (5)$$

exist for all non-negative integers n . Since $I(\omega, h)$ is non-negative everywhere and since $I_0 > 0$, we may interpret $I(\omega, h)/I_0$ as a probability distribution and the I_n as its moments. As we shall see, it is convenient to express the I_n in terms of another closely related sequence of integrals

$$m_n(T, h) = J^{-n} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (\omega - h)^n \chi''_{+-}(\omega, h) \quad (6)$$

which, by slight abuse of language, will be called (shifted) moments. Again they exist for every finite chain.

By definition the shift of the resonance for fixed h is

$$\delta\omega = \frac{I_1}{I_0} - h = J \frac{Jm_2 + hm_1}{Jm_1 + hm_0}. \quad (7)$$

A measure for the line width is the mean square deviation

$$\Delta\omega^2 = \frac{I_2}{I_0} - \frac{I_1^2}{I_0^2} = J^2 \frac{Jm_3 + hm_2}{Jm_1 + hm_0} - \delta\omega^2. \quad (8)$$

Hence, in order to calculate the resonance shift and the line width, we need to know the first four shifted moments m_0, m_1, m_2, m_3 of the dynamic susceptibility χ''_{+-} .

In the following we shall employ the notation $\text{ad}_X \cdot = [X, \cdot]$ for the adjoint action of an operator X . Then $S^+(t) = e^{-iht} e^{it \text{ad}_H} S^+$, since $[H, S^z] = 0$ and $[S^z, S^+] = S^+$, and it follows with (1) and (6) that

$$m_n = \frac{1}{2L} \langle [S^+, \text{ad}_{h/J}^n S^-] \rangle_T. \quad (9)$$

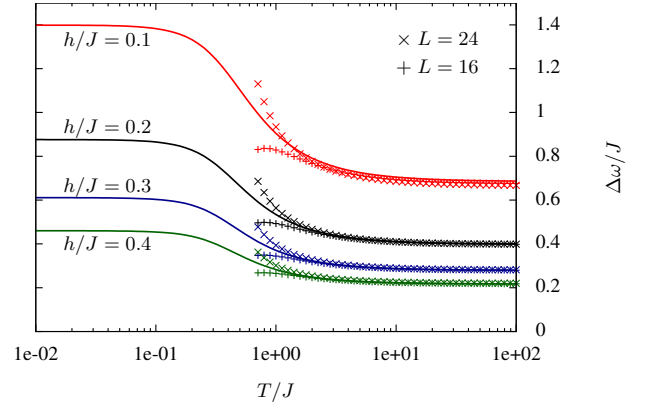


FIG. 2. Line width $\Delta\omega/J$ for the 1D model in the critical regime at $\delta = -0.1$ as function of temperature. Crosses from fully numerical calculation for finite chain Hamiltonians of 16 and 24 sites.

The latter formula shows that the moments m_n are static correlation functions whose complexity grows with growing n . The first few of them can be easily calculated. We shall show the results for the one-dimensional model, for which

$$m_0 = \frac{1}{2L} \langle [S^+, S^-] \rangle_T = \frac{1}{L} \langle S^z \rangle_T, \quad (10)$$

which is the magnetization per lattice site. The subsequent moments are less intuitive,

$$m_1 = \delta \langle s_1^+ s_2^- - 2s_1^z s_2^z \rangle_T, \quad (11a)$$

$$m_2 = \frac{1}{2} \delta^2 \langle s_1^z + 4s_1^z s_2^z s_3^z - 4s_1^z s_2^+ s_3^- \rangle_T, \quad (11b)$$

$$m_3 = \frac{1}{4} \delta^2 \langle 2s_1^+ s_2^+ s_3^- s_4^- + 4s_1^+ s_2^- s_3^+ s_4^- - 2s_1^+ s_2^- s_3^- s_4^+ - 8s_1^z s_2^z s_3^+ s_4^- - 4s_1^z s_2^+ s_3^z s_4^- + 8s_1^z s_2^+ s_3^- s_4^z - 4s_1^+ s_2^- s_3^- s_4^z - s_1^+ s_3^- + 8s_1^z s_2^z s_3^z s_4^z + 2s_1^z s_3^z - 4s_1^z s_2^z + \delta(8s_1^z s_2^+ s_3^- s_4^z + 2s_1^+ s_2^- - 8s_1^z s_2^z) \rangle_T. \quad (11c)$$

They are certain combinations of static short-range correlation functions. This implies, in particular, that they all exist in the thermodynamic limit $L \rightarrow \infty$. It follows that the line shape for fixed h cannot be strictly Lorentzian, as is often assumed in the literature.

All static correlation functions of the one-dimensional Heisenberg-Ising model are polynomials in the derivatives of three functions ω, ω' and φ [8] which, as is common in integrable models, can be expressed in terms of the solutions of certain well behaved linear and non-linear integral equations [9]. This is the reason why in this case the moments m_0, m_1, m_2, m_3 can be calculated exactly by means of the techniques developed in [10].

We parameterize the anisotropy as $\delta = (q - 1)^2/2q$. Then, with the shorthand notations $\varphi_{(n)} = \partial_x^n \varphi(x)|_{x=0}$, $f_{(m,n)} = \partial_x^m \partial_y^n f(x, y)|_{x=y=0}$, for $f = \omega, \omega', \varphi$, we obtain

$$\begin{aligned}
m_0 &= -\frac{1}{2}\varphi_{(0)}, \quad m_1 = \frac{(q-1)^2(q^2+4q+1)\omega'_{(0,1)}}{16q^2} - \frac{(q^3-1)\omega_{(0,0)}}{4q(q+1)}, \\
m_2 &= \frac{(q-1)^2}{256q^4} \left[4q(q+1)(q^3-1)(\omega_{(0,2)}\varphi_{(0)} - 2\omega_{(1,1)}\varphi_{(0)} - \omega_{(0,0)}\varphi_{(2)}) \right. \\
&\quad \left. + (q^2-1)^2(q^2+4q+1)(\omega'_{(1,2)}\varphi_{(0)} + \omega'_{(0,1)}\varphi_{(2)}) - 16q^2(q-1)^2\varphi_{(0)} \right], \\
m_3 &= \frac{(q-1)^4}{98304q^8(q^4-1)(q^6-1)} \\
&\quad \left[16q^2(q^2-1)^3(q^4-1)(q^6-1)(q^2+4q+1)(\omega_{(0,2)}\omega'_{(0,1)} + \omega_{(0,0)}\omega'_{(1,2)}) \right. \\
&\quad + 64q^2(q^2-1)^4(2q^{10}-q^9+4q^8-4q^7-12q^6-14q^5-12q^4-4q^3+4q^2-q+2)\omega_{(0,0)}\omega_{(1,1)} \\
&\quad - 16q^2(q^2-1)^2(q^4-1)(q^6-1)(3q^4+14q^2+3)\omega'_{(0,3)} \\
&\quad + 8q^2(q^2-1)(q^4-1)^2(q^6-1)(q+1)^2(8\omega'_{(1,2)} - \omega'_{(2,3)}) \\
&\quad + 192q^4(q^2-1)^2(q^4-1)(q^6+18q^4+8q^3+18q^2+1)\omega_{(0,2)} \\
&\quad + 64q^2(q^2-1)^2(q^4-1)(q+1)^2(2q^8-5q^7+26q^6-49q^5+28q^4-49q^3+26q^2-5q+2)\omega_{(1,1)} \\
&\quad - 16q^2(q^2-1)^2(q^4-1)(q+1)^2(q^8-q^7+q^6+q^5+2q^4+q^3+q^2-q+1)(2\omega_{(1,3)}-3\omega_{(2,2)}) \\
&\quad + 64q^2(q^4-1)(q^6-1)(3q^8+2q^6+24q^5-130q^4+24q^3+2q^2+3)\omega'_{(0,1)} \\
&\quad + (q^4-1)(q^6-1)(q+1)^2(q^{10}-2q^9+25q^8+16q^7+118q^6+164q^5 \\
&\quad \quad + 118q^4+16q^3+25q^2-2q+1)(\omega'_{(0,3)}\omega'_{(1,2)} + \omega'_{(0,1)}\omega'_{(2,3)}) \\
&\quad - 1536q^5(q^4-1)(4q^8-9q^7-2q^6-6q^5+8q^4-6q^3-2q^2-9q+4)\omega_{(0,0)} \\
&\quad + 4q^2(q^6-1)(q+1)^2(q^2+1)(5q^8-2q^7+32q^6+50q^5+70q^4+50q^3+32q^2-2q+5) \\
&\quad \quad (2\omega_{(1,3)}\omega'_{(0,1)} - 3\omega_{(2,2)}\omega'_{(0,1)} + \omega_{(0,2)}\omega'_{(0,3)} - 2\omega_{(1,1)}\omega'_{(0,3)} - 3\omega_{(0,2)}\omega'_{(1,2)} - \omega_{(0,0)}\omega'_{(2,3)}) \\
&\quad - 16q^2(q+1)^2(q^{16}-q^{15}+8q^{14}+9q^{13}+47q^{12}+45q^{11}+96q^{10}+91q^9+128q^8+91q^7+96q^6 \\
&\quad \quad + 45q^5+47q^4+9q^3+8q^2-q+1)(3\omega_{(0,2)}^2 - 6\omega_{(1,1)}\omega_{(0,2)} + 2\omega_{(0,0)}\omega_{(1,3)} - 3\omega_{(0,0)}\omega_{(2,2)}) \left. \right]. \quad (12)
\end{aligned}$$

These functions represent the moments in the thermodynamic limit. Since they can be calculated to arbitrary precision, we obtain numerically accurate results for the resonance shift and for the line width as functions of temperature or magnetic field over the whole range of the phase diagram. In particular, our approach is not restricted to small anisotropies. Examples for $\delta = -0.1$ are shown in figures 1 and 2. We find a broadening of the line width as defined by (8) for small temperatures in the critical ($\delta < 0$) as well as in the massive ($\delta > 0$) regime (latter case not shown here).

At first sight this seems to contradict experimental results [4] which claim a narrowing. Still, one has to take into account that usually in the analysis of experimental data rather different definitions of the line width, as e.g. the distance between the turning points right and left to the maximum of the intensity, are used. In particular, if the intensity distribution has long shallow tails the definition (8) will give considerably larger values than the distance between the turning points. We have performed a detailed numerical study of the dynamical susceptibility for chains of 16, 20 and 24 lattice sites (to be published elsewhere) and we see indeed such long tails at low temperature (compare also [6]). In experiments they may be

misinterpreted as background stemming from couplings of the spin chain to other degrees of freedom, but in fact they are due to the spin-spin interactions and are part of the true absorption line. For the determination of the resonance shift tails are expected to have less influence. As long as they are not too much asymmetric the shift calculated by means of (7) should agree with the shift of the maximum of the absorbed intensity.

In current ESR experiments the microwave frequency ω is kept fixed and the Zeeman field h is modulated. This means that, as opposed to most of the theoretical treatments, including our considerations above, the absorbed intensity $I(\omega, h) = \omega\chi''_{+-}(\omega, h)/2$ is determined as a function of h for fixed ω , and the resonance shift and line width are measured in ‘ h -direction’. Away from the isotropic point ($\delta = 0$), where $\chi''_{+-}(\omega, h)$ is symmetric and the absorption line is extremely narrow, this may clearly lead to rather different values. To be closer to present-day ESR experiments one should calculate resonance shift and line width in terms of the moments of the dynamical susceptibility in ‘ h -direction’.

We define

$$M_n(T, \omega) = J^{-n} \int_{-\infty}^{\infty} \frac{dh}{2\pi} (h - \omega)^n \chi''_{+-}(\omega, h). \quad (13)$$

For these functions we obtain the representation

$$M_n(T, \omega) = (-1)^n \sum_{k=0}^{\infty} \frac{(-J)^k}{k!} m_{k+n}^{(k)}(T, \omega), \quad (14)$$

where the superscript (k) denotes the k th derivative with respect to the second argument. We see that the M_n are determined by infinitely many of the m_n and their derivatives, i.e., they depend on static correlation functions for arbitrarily large distances. For this reason they cannot be calculated by our exact method above. Yet, in certain cases only finitely many terms of the series are needed for a good approximation.

We first of all express the resonance shift $\delta h = \langle h \rangle - \omega$ and the mean square deviation from the center of the absorption peak $\Delta h^2 = \langle h^2 \rangle - \langle h \rangle^2$ in terms of the M_n ,

$$\frac{\delta h}{J} = \frac{M_1}{M_0}, \quad \frac{\Delta h^2}{J^2} = \frac{M_2}{M_0} - \frac{M_1^2}{M_0^2}. \quad (15)$$

We have identified two cases, where these formulae simplify and finitely many of the m_n are enough to determine δh and Δh approximately.

The equation for the resonance shift simplifies for small anisotropy $|\delta| \ll 1$. Since $M_0 = m_0 + \mathcal{O}(\delta)$, $M_1 = m_1 + \mathcal{O}(\delta^2)$ and, generically, m_1 itself is of order δ (see (11), (14)) we obtain to linear order in δ

$$\frac{\delta h}{J} = -\frac{m_1}{m_0}. \quad (16)$$

In [5, 7] the same equation was obtained by a more intuitive reasoning. It leads to results which compare rather well to experiments [7]. However, some care is necessary with the interpretation of (16). m_1/δ vanishes at $\delta = h = 0$. It follows that $m_1 = \delta(ah + b\delta + \dots)$ with some coefficients a, b , whence h must be large compared to δ for (16) to be applicable. Note that all higher moments m_n are of order δ^2 . Hence, there is no simplification for small anisotropy, like in (16), for the line width. But the integrated intensity M_0 has again a finite approximation to first order in δ , $M_0 = m_0 - Jm'_1$.

The representation (14) is a series in ascending powers of J/T (with still temperature dependent coefficients). This can be used to evaluate (15) asymptotically for high temperatures. It turns out that the leading terms in the J/T expansion of m_1 and Jm'_2 cancel each other: $\delta h \sim \frac{h}{2T}\delta \rightarrow 0$ in the high-temperature limit $T \gg J$, and

$$\frac{\Delta h}{J} = \frac{|\delta|}{\sqrt{2}} \quad (17)$$

for arbitrary microwave frequency ω . This formula provides a simple means to directly measure the anisotropy parameter δ .

It may be instructive to illustrate our formula with one of the few explicit results, namely with the formula for the intensity in the free fermion case $\delta = -1$ at $T \rightarrow \infty$ [11]. In this case $I(\omega, h) \sim (\omega^2/J^2) \exp(-(\omega - h)^2/J^2)$ and, in agreement

with (17), we obtain the line width $\frac{\Delta h}{J} = \frac{1}{\sqrt{2}}$, whereas the width in omega direction depends on h .

Our work is the first exact result for the resonance shift and the line width in microwave absorption experiments on the Heisenberg-Ising chain. The reduction to moments is not restricted to the integrable case and may be interesting for the two- and three-dimensional models as well. Our approach is unbiased. It makes no a priori assumptions about the shape of the spectral line. As opposed to all other approaches it is valid for all temperatures and magnetic fields and in addition for arbitrary values of δ . For small δ close to the isotropic point we recover the result of [7] for the line shift. The resonance shift $\delta\omega/J$ or $\delta h/J$ and the line width $\Delta\omega/J$ or $\Delta h/J$ defined in terms of moments show a simple scaling behavior. They depend on the exchange interaction only through the ratios T/J and h/J . In this sense the curves in figures 1 and 2 are universal.

The intensity $I(\omega, h)$ is a function of ω and h . With our definitions of $\delta\omega$ and $\Delta\omega$ we determine the resonance shift and the line width in ω -direction as functions of h , while in standard ESR experiments the resonance shift δh and line width Δh in h -direction are measured as functions of ω , which should be clearly distinguished. For the resonance shift it follows from (7), (16) that $\delta h(T, \omega) = -\delta\omega(T, h)|_{h=\omega}$ to linear order in δ . For the line width there is no such simple relation between $\Delta\omega$ and Δh . However, for Δh we obtained the simple high-temperature formula (17) which we suggest to be useful to measure the anisotropy directly. We are further convinced that it may be worth trying to measure $\Delta\omega$, which is now known exactly, directly in multi-frequency ESR experiments.

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