

# Theory of the electron spin resonance in quantum spin chains with nearest, next-nearest neighbor, and alternating interactions

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(Received 21 November 2008; revised manuscript received 12 January 2009; published 27 February 2009)

The dynamical mean-field-like theory of the electron spin resonance (ESR) in spin-1/2 chains with nearest and next-nearest neighbor interactions (or zigzag spin ladders) and alternation is developed. It is explained that either the magnetic anisotropy of spin-spin couplings or the Dzyaloshinskii-Moriya interaction causes shifts of the resonance position in spin chains, comparing to only magnetically isotropic interactions. The power of the ac ESR field, absorbed by the spin chain, is calculated. It is shown that the temperature dependence of the shift of the resonance position is related to the temperature dependence of the ESR absorption. Depending on the strength of next-nearest neighbor interactions, the shift of the ESR position can become either exponentially small or can be increased near quantum critical points. The absorbed by the alternating spin chain power of the ac magnetic field is calculated, manifesting two resonances. The influence of mentioned spin-spin interactions on the linewidth of the ESR is analyzed.

DOI: 10.1103/PhysRevB.79.064422

PACS number(s): 75.10.Pq, 76.30.-v

## I. INTRODUCTION

The electron spin resonance (ESR) serves as a very good method to investigate the spin dynamics of many systems. In ESR experiments a dc magnetic field is applied to the investigated system, and one measures the absorption of an ac magnetic field, usually polarized perpendicular to the dc field direction. The theory of the ESR is well developed for the situation in which spins of electrons are weakly coupled to each other: the electron paramagnetic resonance (EPR).<sup>1</sup> On the other hand, the situation becomes also relatively simple, if spin-spin interactions are strong enough to form a magnetic long-range order. In that case the magnetic order parameter is the same for all magnetic sites. The low-temperature behavior of ordered magnets can be described in the quasiclassical approximation, i.e., ordered spins are replaced with classical vectors of the order parameter with small inhomogeneous deviations from their steady-state ordered configurations, called magnons (or spin waves). Theories explore this fact, calculating the magnon response to the dc field in the ferro- or antiferromagnetic ESR, called ferro- or antiferromagnetic resonance (FMR or AFMR).<sup>2</sup> The ESR response in magnetic insulators is usually related to the homogeneous situation; hence, as a rule, only magnons with zero quasimomentum form the FMR or AFMR.

The situation becomes much more complicated in low-dimensional spin systems. Here, on one hand, spins are strongly interacting with each other. On the other hand, quantum and thermal fluctuations are enhanced in low-dimensional systems. Those fluctuations usually destroy the long-range magnetic ordering in two- and one-dimensional (1D) systems (except of the ground state, where the ordering is possible).<sup>3</sup> Thus, when describing low-dimensional spin systems, one has to take into account strong enough short-range correlations, while the absence of the long-range order does not permit theorists to use advantages of the quasiclassical approach. The theory of low-dimensional spin systems

becomes even more complicated, if one takes into account possible quantum phase transitions there. Those quantum critical points strongly affect the behavior of thermodynamic and other characteristics of spin chains, including ESR. However, during the last decade the number of ESR experiments on low-dimensional magnetic systems increased considerably.<sup>4</sup>

The goal of the present work is to present a relatively simple theory of the ESR in quantum spin-1/2 chains with quantum critical points caused by intrachain nearest neighbor (NN) and next-nearest neighbor (NNN) interactions and alternation in spin chains.

## II. ESR IN SPIN SYSTEMS WITH MAGNETICALLY ISOTROPIC INTERACTIONS

Let us consider a spin system, in which interactions between spins do not form a magnetically ordered state at non-zero temperatures, in a dc magnetic field, and a circular polarized perpendicular to the dc one ac magnetic field. The Hamiltonian of the system can be written as

$$\mathcal{H} = \mathcal{H}_{\text{is}} + \mathcal{H}_{\text{an}} - H \sum_n S_n^z - h \sum_n [\cos(\omega t) S_n^x - \sin(\omega t) S_n^y], \quad (1)$$

where  $S_n^{x,y,z}$  are the operators of the projections of a spin-1/2, situated at the site  $n$ ,  $H$  is the value of the dc magnetic field (we use the units, in which the Bohr magneton and the  $g$  factor are equal to 1),  $h$  and  $\omega$  are the magnitude and the frequency of the ac magnetic field,  $\mathcal{H}_{\text{is}}$  is the part of the Hamiltonian, which describes isotropic exchange interactions, and  $\mathcal{H}_{\text{an}}$  describes magnetically anisotropic interactions. The power of the ac field, absorbed by the system per spin, can be written as

$$Q = \frac{\partial \langle \mathcal{H} \rangle_t}{N \partial t} = \frac{h\omega}{N} \sum_n [\sin(\omega t) \langle S_n^x \rangle + \cos(\omega t) \langle S_n^y \rangle], \quad (2)$$

where the subscript  $t$  implies time averaging,  $N$  is the total number of spins, and angular brackets denote averaging with the density matrix of the system (or with the ground-state wave function for  $T=0$ ). Using the unitary transformation we can remove the explicit time dependence from the Hamiltonian (transferring it into the time dependence of the density matrix, or the ground-state wave function). Similar situation appears if the magnetic field  $H$  is directed exactly along the axis of the uniaxial magnetic anisotropy: one can remove the explicit time dependence from the Hamiltonian with the help of the unitary transformation. Observe that if the polarization of the ac field is not circular, one can drop the other (nonresonance) proportional to  $h$  terms, present in the Hamiltonian, after the unitary transformation, because  $h \ll H$ ,  $\hbar\omega$  in standard ESR experiments. After such a transformation the effective Hamiltonian gets the form

$$\tilde{\mathcal{H}} = \mathcal{H}_{\text{is}} + \mathcal{H}_{\text{an}} - (H - \hbar\omega) \sum_n S_n^z - h \sum_n S_n^x, \quad (3)$$

and the absorbed power can be written as

$$Q = \frac{h\omega}{N} \sum_n \langle S_n^y \rangle. \quad (4)$$

Consider first the situation, in which all magnetically anisotropic terms are absent in the system,  $\mathcal{H}_{\text{an}}=0$ . Then, it is easy to see that all projections of the total spin of the system,  $S^{x,y,z} = (1/N) \sum_n S_n^{x,y,z}$  commute with  $\mathcal{H}_{\text{is}}$ . Now it is a simple task to calculate the absorbed power  $Q$ . The ‘‘quantum Boltzmann’’ equations,<sup>5</sup> which describe the dynamics of the system, are

$$i\hbar \left[ \frac{\partial \langle S^z \rangle}{\partial t} + \gamma (\langle S^z \rangle - S_0^z) \right] = -\frac{h}{2} (\langle S^+ \rangle - \langle S^- \rangle),$$

$$i\hbar \left[ \frac{\partial \langle S^\pm \rangle}{\partial t} + \gamma (\langle S^\pm \rangle - S_0^\pm) \right] = \pm (H - \hbar\omega) \langle S^\pm \rangle \mp h \langle S^z \rangle, \quad (5)$$

where  $S^\pm = S^x \pm iS^y$ ,  $\gamma$  is the relaxation rate (the damping can be connected with, e.g., the spin-lattice relaxation), and  $S_0^{\pm,z}$  are the average values of the spin projections to which relaxation tends. These quantum Boltzmann equations are obtained in the standard way: we write Heisenberg equations of motion with the Hamiltonian  $\tilde{\mathcal{H}}$  for the operators  $S^{\pm,z}$  and average them with the density matrix of the system (or with the ground-state wave function); then the relaxation is included phenomenologically in a simplest form.<sup>6</sup>

In the stationary limit  $t \gg \gamma^{-1}$ , where one can neglect the terms in Eq. (5) with time derivatives, we obtain

$$Q = \frac{h\omega \hbar \gamma [hS_0^z - (H - \hbar\omega)S_0^x + \hbar\gamma S_0^y]}{(H - \hbar\omega)^2 + h^2 + (\hbar\gamma)^2}. \quad (6)$$

Usually  $h \ll \hbar\omega$ ,  $H$  in the ESR experiments; hence, one can neglect the term  $h^2$  in the denominator, i.e., to use the linear-

response theory. Then, in the resonance, with  $\hbar\gamma$  being small, in the stationary limit the main contribution comes, naturally, from the first term in square brackets. Therefore usually only this term is taken into account (i.e., one can neglect terms with  $S_0^y$  and  $S_0^x$ ). One can see from Eq. (6) that the absorbed power of the ac field in the limit of  $\hbar\gamma \rightarrow 0$  is proportional to a  $\delta$  function  $\delta(H - \hbar\omega)$ , i.e., the linewidth of the ESR response is zero in this limit and the position of the resonance line is determined by the bare value of the dc magnetic field. In the case of, e.g., the linear polarization of the ac field, i.e.,  $h \cos(\omega t) \sum_n S_n^x$ , a resonance-antiresonance situation is possible with  $\hbar\omega = \pm H$ , usual for the ESR case. However, we shall consider only the resonance with  $\hbar\omega = H$  in detail in what follows.

The strength of the absorption is determined by the value of  $S_0^z$ . If, as usual for ESR,  $h \ll H$ ,  $\hbar\omega$ , it is natural to suppose that  $S_0^z = \text{Sp}(S^z \rho(H, T))$ , where  $\rho(H, T)$  is the density matrix with Hamiltonian (1) taken at  $h=0$ . In many cases (at least for spin systems with antiferromagnetic interactions, i.e., in the absence of the spontaneous magnetization, and for weak enough dc field values for  $H \ll H_s$ , where  $H_s$  is the value of the dc field at the quantum phase transition to the spin-polarized phase in the ground state, at which  $S_0^z = 1/2$ )  $S_0^z$  can be approximated by  $S_0^z \approx \chi(H, T)H$ , where  $\chi(H, T)$  is the static magnetic susceptibility of the system for  $h=0$  (in the magnetically isotropic case the susceptibility is, naturally, the same for any direction of the dc field).

In the framework of the linear response<sup>7</sup> theorists usually present the expression for the absorbed power as  $Q = \hbar^2 \omega^2 \chi''_{aa}(\omega, \mathbf{q}=0)/2$ , where  $\chi''_{aa}(\omega, \mathbf{q})$  is the imaginary part of the dynamical magnetic susceptibility taken at the wave vector  $\mathbf{q}=0$  (because usually the wavelength of the ac field is larger than the size of a sample in the ESR experiments) and  $a \perp z$  is the direction of the polarization of the ac field. The dynamical magnetic susceptibility is calculated using spin-spin correlation functions of the problem.

### III. ESR IN SPIN CHAINS WITH AN UNIAXIAL MAGNETIC ANISOTROPY

In what follows we will study the general case, where  $\mathcal{H}_{\text{an}} \neq 0$ , i.e., one has magnetically anisotropic terms in the Hamiltonian. Notice that if the magnetic anisotropy is along the  $x$  direction, then  $\hbar^2 \omega^2 \chi''_{xx}(0, \omega) = H^2 \chi''_{yy}(0, \omega)$  [or, if there is an angle  $\phi$  between the axis  $x$  and  $a$ , one has  $\hbar^2 \omega^2 \chi''_{aa}(0, \omega) = (H^2 \cos^2 \phi + \hbar^2 \omega^2 \sin^2 \phi) \chi''_{yy}(0, \omega)$ ]. However, from now on we study the case, in which the distinguished by the magnetic anisotropy direction coincides with the direction of the dc magnetic field, i.e., the magnetic anisotropy is along the  $z$  direction. For the system with such a magnetic anisotropy, presented in the simplest form, the Hamiltonian can be presented as

$$\mathcal{H}_{\text{an}} = - \sum_{n,\delta} A_\delta S_n^z S_{n+\delta}^z, \quad (7)$$

where  $A_\delta$  are the magnetic anisotropy constants (for spin-1/2 systems we, naturally, have only interior magnetic anisotropy; the single-ion one is absent). The anisotropy can be caused, e.g., by magnetic dipole-dipole interactions.  $\delta$  deter-

mines the range of interion interactions. The situation differs drastically from the above. Writing Heisenberg equations of motion for the operators of spin projections, we immediately see that other operators such as  $\pm A_\delta(S_{n-\delta}^z S_n^\pm + S_n^\pm S_{n+\delta}^z)$  appear in the equations of motion. The simplest possibility to take into account those terms is to use the dynamical mean-field-like approximation (we limit ourselves with the only one value of  $A_\delta=A$  for some fixed  $\delta$ , e.g., for NN interactions  $\delta=\pm 1$ )

$$i\hbar \left[ \frac{\partial \langle S^\pm \rangle}{\partial t} + \gamma \langle S^\pm \rangle \right] = \pm (H - \hbar\omega + ZA \langle S^z \rangle) \langle S^\pm \rangle \mp h \langle S^z \rangle, \\ i\hbar \left[ \frac{\partial \langle S^z \rangle}{\partial t} + \gamma (\langle S^z \rangle - S_0^z) \right] = -\frac{h}{2} (\langle S^+ \rangle - \langle S^- \rangle), \quad (8)$$

where  $Z$  is the coordination number ( $Z=2$  for the spin chain with NN interactions, where  $\delta=\pm 1$ ). In the linear-response limit,  $h \ll H$ ,  $\hbar\omega$ , one can write the answer for the power of the ac field absorbed by the system in this case as

$$Q = \frac{h^2 \omega \hbar \gamma S_0^z}{(H + ZA S_0^z - \hbar\omega)^2 + (\hbar\gamma)^2}. \quad (9)$$

Notice that  $\langle S^z \rangle$  is determined from the self-consistency-like equation

$$\langle S^z \rangle - S_0^z = \frac{h^2 \langle S^z \rangle}{(H + ZA \langle S^z \rangle - \hbar\omega)^2 + (\hbar\gamma)^2}. \quad (10)$$

It is clear that the difference between  $\langle S^z \rangle$  and  $S_0^z$  is proportional to the square of the (small) magnitude of the ac field. Therefore, in the linear-response theory we can use the approximation  $\langle S^z \rangle \approx S_0^z$  [cf. Eq. (9)]. It turns out that the dynamical mean-field-like approximation does not take into account, naturally, possible bound states, which contribute to the characteristics of quantum spin chains.<sup>3</sup> The result for  $Q$  implies that in this approximation the position of the ESR is shifted, with respect to the one of the isotropic case, by the value of the effective anisotropy field  $\hbar\omega = H + ZA S_0^z$ . Sometimes this shift of the resonance position is written as the effective  $g$  factor, i.e., in our case  $g_{\text{eff}} = g_0 [1 + (ZA S_0^z / \mu_B H)]$ , where  $g_0$  is the  $g$  factor of noninteracting spins and  $\mu_B$  is the Bohr magneton. It turns out that the temperature dependence of the shift of the resonance position approximately coincides with the temperature dependence of the absorbed ac power. On the other hand, the linewidth of the signal is not affected by the magnetic anisotropy in this approximation explicitly. Notice that we have not used yet the one dimensionality of the considered problem, and the above result is valid (in the framework of the used approximations, naturally) for a uniaxial spin system of any dimension. For many cases (at least for antiferromagnetic exchange interactions at weak dc fields  $H \ll H_s$ ) the last formula can be rewritten as  $\hbar\omega = H [1 + ZA \chi_{zz}(H, T)]$ . One can see that the shift of the resonance position in this case is determined by the constant of the magnetic anisotropy and the static magnetic susceptibility of the system in the direction of the dc field. We stress again that our theory implies similar temperature dependence

of the shift of the ESR position and the absorbed ac field power [cf. Eq. (9)].

In the case of the spin-1/2 Heisenberg antiferromagnetic chain with the easy-plane magnetic anisotropy of NN interactions,  $A > 1$ , the magnetic susceptibility is known exactly for small values of  $H$  (Refs. 3 and 8) and, therefore, this shift of the resonance position of the ESR can be approximated at low temperatures and small values of the dc field as

$$\hbar\omega = H \left[ 1 + \frac{A}{\pi v(J, A, H)} \left\{ K(J, A, H) + f_1(J, A) \left[ \frac{\pi T}{v(J, A, H)} \right]^2 + f_2(J, A) \left[ \frac{\pi T}{v(J, A, H)} \right]^{4K(J, A, H)-4} + \dots \right\} \right], \quad (11)$$

where  $J$  is the constant of the antiferromagnetic NN intrachain exchange interaction,  $v(J, A, H)$  is the velocity of low-energy excitations [at  $H=0$  it can be written as  $v = J\pi\sqrt{A(2J-A)}/2 \cos^{-1}(J-A)$ ],  $K(J, A, H)$  is the dressed charge<sup>3</sup> (for  $H=0$  it can be written as  $K = \pi/[\pi - \cos^{-1}(J-A)]$ ), and  $f_{1,2}(J, A)$  are functions of the anisotropy parameter.<sup>8</sup> For the easy-axis anisotropy, on the other hand, low-lying excitations of an antiferromagnetic chain have a spin gap and, therefore, the magnetic susceptibility is exponentially small for values of the dc field smaller than the critical one  $H_c$  (determined by the gap value). For the case of very small anisotropy  $|A| \ll J$ , as usual for transition-metal compounds, we can write the formula for the shift of the resonance position as

$$\hbar\omega = H \left[ 1 + \frac{|A|}{\pi v} \left( 1 + \frac{1}{2\mathcal{L}} - \frac{\ln 2\mathcal{L}}{4(\mathcal{L})^2} + \frac{(\ln \mathcal{L})^2 - \ln \mathcal{L} + 3/4}{8(\mathcal{L})^3} + \frac{\sqrt{3}T^2}{\pi J^2} + \dots \right) \right], \quad (12)$$

where  $v = \pi J/2$  and  $\mathcal{L} = \ln[\sqrt{\pi} \exp(\gamma+1/4)J/\sqrt{2}T]$  with the Euler constant  $\gamma$ . This result, which can be used at any sign of  $A$ , is very similar in the main approximation in  $|A|$  (however, it differs in the next logarithmic corrections) to the one of the bosonization<sup>9</sup> and conformal field theory<sup>10</sup> approximations (where the shift is proportional to  $|A|/v$ ) and to the Nagata-Tazuki<sup>11</sup> based theory,<sup>12</sup> which uses the Bethe ansatz results [where the shift is proportional to  $(|A|/J) \int S_0^z dJ$ ]. Notice different temperature dependencies of the shift of the resonance position of the present theory compared with the previous ones.<sup>9,10,12</sup> For larger values of  $H$  the magnetization of the antiferromagnetic Heisenberg chain is almost linear in  $H$ , and for  $H > H_s$ , where  $H_s = J+A$ , the magnetization is almost  $H$  independent at low enough temperatures.

#### IV. FEATURES OF THE ESR IN SPIN CHAINS WITH NEAREST AND NEXT-NEAREST NEIGHBOR INTERACTIONS

Now, let us study the ESR response of a spin-1/2 chain with NN and NNN interactions (or, in other words, in a zigzag spin ladder). According to the above, isotropic NN and NNN interactions do not change equations of motion, but they change the dependence of the average value of mag-

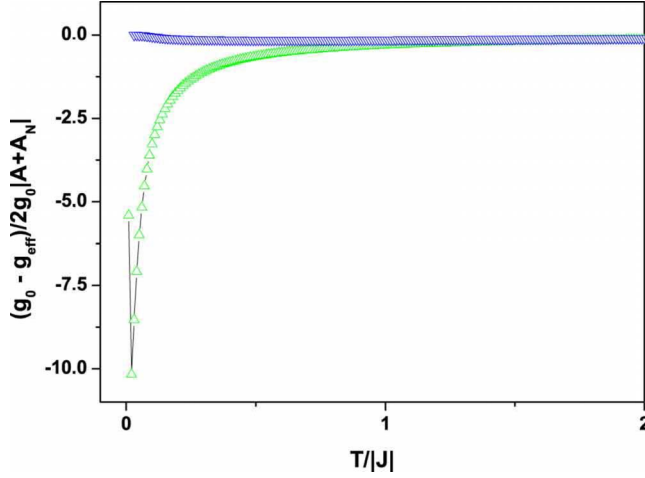


FIG. 1. (Color online) Temperature dependencies of normalized effective  $g$  factors for weakly anisotropic spin-1/2 chains for  $H \ll H_s$ . Upper curve (down-oriented blue triangles): the case with only antiferromagnetic NN interactions,  $J=1$ . Lower curve (up-oriented green triangles): the case with ferromagnetic NN interactions  $J=-1$  and antiferromagnetic NNN interactions  $J_N=0.29$ . Lines are guides for the eyes.

netization,  $S_0^z$ , on the temperature, dc magnetic field, and interaction constants. Hence, in the equations of motion one needs to take into account the magnetically anisotropic NNN couplings as  $\mathcal{H}_{\text{NNNan}} = -A_N \sum_n S_n^z S_{n+2}^z$ , i.e., we take into account the first and the second terms with different constants of the anisotropy in Eq. (7). In the mean-field-like approximation such a consideration yields the previous answer for the ESR absorbed power [Eq. (9)] with the replacement  $A \rightarrow A+A_N$ . It formally seems that the result for ESR is not very much affected by the NNN interactions. It is not true: the ESR response can be drastically changed due to NNN interactions. For example, it is known that for  $J_N > 0.24 \dots J$  ( $J_N$  is the constant of the isotropic exchange NNN interaction) for both antiferromagnetic NN and NNN couplings, low-lying excitations of such a spin-1/2 chain have a gap.<sup>13</sup> Hence, according to our theory,  $S_0^z$  and, therefore, the ESR shift and the absorbed power have to be exponentially small for the values of the dc field  $H$  smaller than  $H_c$ , at which the gap is closed. On the other hand, for several spin-1/2 chains with NN and NNN interactions the solution is known exactly.<sup>14</sup> In some of those models there exists a quantum phase transition to the incommensurate phase with gapless excitations, and such a situation seems to be generic for chains with competing NNN and NN spin-spin couplings. The quantum phase transition exists for NNN antiferromagnetic interactions with both ferro- and antiferromagnetic NN ones and often for both easy-axis and easy-plane magnetic anisotropies.<sup>14,15</sup> It was shown<sup>14,16</sup> that in this case the velocity of low-lying excitations  $v$  becomes smaller,  $v \rightarrow v[1 - |\alpha/\alpha_c|]$ , where  $\alpha$  is determined by the ratio of  $J_N$  and  $J$  and  $\alpha_c$  is the value at the quantum critical point. It follows from our theory that for  $\alpha$  close to the critical value the shift of the ESR position has to be much stronger than for the situation for  $\alpha$  being far from the  $\alpha_c$  value. In Fig. 1 we present the temperature dependencies for normalized effective  $g$  factors

of the spin-1/2 chain with antiferromagnetic NN interactions and the chain with ferromagnetic NN interactions and antiferromagnetic NNN interactions near a quantum critical point. Calculations were performed using exact diagonalization for a weakly anisotropic spin-1/2 chain with 14 spins. One can see the drastic difference in the behaviors of effective  $g$  factors. ESR experiments on frustrated spin-1/2 quasi-one-dimensional systems<sup>17,18</sup>  $\text{In}_2\text{VO}_5$  and  $\text{Li}_2\text{ZrCuO}_4$  reveal the qualitative features of their behavior similar to our theoretical results. Namely, due to the possible presence of quantum critical points, caused by NNN spin-frustrating interactions (see, e.g., Ref. 15), the special temperature behavior of effective  $g$  factors (shifts of the ESR positions) were observed in those compounds. Observed temperature behavior of the ESR shifts was too strong to be explained using the standard theory of the ESR in spin chains with only NN interactions (cf. Fig. 1). Notice, that ESR experiments<sup>17,18</sup> were made on oriented polycrystals, where one has to average the response over directions of the magnetic anisotropy of each domain of the polycrystal. In  $\text{Li}_2\text{ZrCuO}_4$  additional doubling of effective  $g$  factors takes place, probably via Jahn-Teller-type effect due to shifts of surrounding ligands (cf. Ref. 19).

## V. ESR IN SPIN CHAINS WITH THE DIMERIZATION

Now let us consider the ESR response of the spin-1/2 chain (or zigzag spin ladder), in which the alternation of the NN coupling constants takes place, i.e., each spin is coupled to the NN ones with different exchange constants,  $J_1$  and  $J_2$  (isotropic exchange), with the anisotropy parameters  $A_1$  and  $A_2$ , respectively. The Hamiltonian can be written as

$$\begin{aligned} \mathcal{H}_{\text{is}} &= J_1 \sum_n \mathbf{S}_{1,n} \mathbf{S}_{2,n} + J_2 \sum_n \mathbf{S}_{2,n} \mathbf{S}_{1,n+1} + J_N \sum_{i=1,2} \sum_n \mathbf{S}_{i,n} \mathbf{S}_{i,n+1}, \\ \mathcal{H}_{\text{an}} &= -A_1 \sum_n S_{1,n}^z S_{2,n}^z - A_2 \sum_n S_{2,n}^z S_{1,n+1}^z - A_N \sum_{i=1,2} \sum_n S_{i,n}^z S_{i,n+1}^z, \end{aligned} \quad (13)$$

where  $n$  enumerates elementary cells in the dimerized chain. Each cell contains two magnetic centers (spins), labeled by the index  $i=1,2$ . In this case the quantum Boltzmann equations, which describe the ESR dynamics, in the dynamical mean-field-like approximation can be written as

$$\begin{aligned} i\hbar \left[ \frac{\partial \langle S_{1,2}^\pm \rangle}{\partial t} + \gamma \langle S_{1,2}^\pm \rangle \right] &= \pm (H - \hbar\omega) \langle S_{1,2}^\pm \rangle \mp \hbar \langle S_{1,2}^z \rangle \\ &\pm (J_1 + J_2) \langle S_{1,2}^z \rangle \langle S_{2,1}^\pm \rangle \\ &\mp 2A_N \langle S_{1,2}^z \rangle \langle S_{1,2}^\pm \rangle \\ &\mp (J_1 + J_2 + A_1 + A_2) \langle S_{2,1}^z \rangle \langle S_{1,2}^\pm \rangle, \end{aligned}$$

$$\begin{aligned}
& i\hbar \left[ \frac{\partial \langle S_{1,2}^z \rangle}{\partial t} + \gamma (\langle S_{1,2}^z \rangle - S_{0,1,2}^z) \right] \\
& = -\frac{\hbar}{2} (\langle S_{1,2}^+ \rangle - \langle S_{1,2}^- \rangle) \mp \frac{J_1 - J_2}{2} (\langle S_1^- \rangle \langle S_2^+ \rangle - \langle S_2^- \rangle \langle S_1^+ \rangle),
\end{aligned} \tag{14}$$

where  $\langle S_{1,2}^{\pm} \rangle$  are average values of the projections of the spin in each elementary cell, with the subscript 1 or 2 noting even and odd spins. In these quantum Boltzmann equations we took into account that the ESR usually considers the homogeneous response to the ac field. As above, we introduced the relaxation  $\gamma$  and values  $S_{0,1,2}^z$ . The latter are the average values of operators of spin projections for even and odd spins, to which the relaxation tends. They are determined by the averaging with the density matrix determined by the Hamiltonian of the system for  $h=0$ .

We solve Eq. (14) in the linear-response regime, where  $h \ll H$ ,  $\hbar\omega$ , as above, and the result for the absorbed ESR power is

$$\begin{aligned}
Q = & \frac{\hbar^2 \omega \hbar \gamma}{2[A^2 + (\hbar\gamma)^2 B^2]} \left\{ [F^2 + (\hbar\gamma)^2] R_+ \right. \\
& \left. + \left[ 2(H - \hbar\omega) - R_+ \frac{A_1 + A_2 + 2A_N}{2} \right] f \right\}, \tag{15}
\end{aligned}$$

where

$$R_{\pm} = S_{0,1}^z \pm S_{0,2}^z,$$

$$f = \frac{1}{2} (A_1 + A_2 - 2A_N) R_-^2,$$

$$F = H - \hbar\omega - \frac{2J_1 + 2J_2 + A_1 + A_2 - 2A_N}{2} R_+,$$

$$B = 2(H - \hbar\omega) - (J_1 + J_2 + A_1 + A_2) R_+,$$

$$\begin{aligned}
A = & F[F + (J_1 + J_2 - 2A_N) R_+] - f \frac{2J_1 + 2J_2 + A_1 + A_2 - 2A_N}{2} \\
& - (\hbar\gamma)^2. \tag{16}
\end{aligned}$$

It is interesting to point out that the dimerized antiferromagnetic spin-1/2 chain in the ground state is in the spin-polarized state for  $H > H_s$ , where  $H_s$  can be written in our approximation as  $H_s = (2J_1 + 2J_2 + A_1 + A_2 - 2A_N)/2$ , i.e.,  $F = H - H_s R_+ - \hbar\omega$ . It is also worthwhile to mention that  $R_{\pm}/2H$  for a weak dc magnetic field can be related to the  $z$  components of the homogeneous and staggered susceptibilities of the alternating chain, respectively. It can be seen that the denominator of Eq. (15) can be rewritten as  $[(H - x_1 - \hbar\omega)^2 + (\hbar\gamma)^2][(H - x_2 - \hbar\omega)^2 + (\hbar\gamma)^2]$ . It means that Eq. (15) describes *two* resonances. Two resonances reflect the fact that the dimerization of the spin chain introduces two effective magnetic centers in the problem and the ESR studies the responses from the movement of both of them (two magnetic moments oscillate as a total and one relative to other). Similar situation was observed, e.g., in CuGeO<sub>3</sub> (see Refs. 4 and

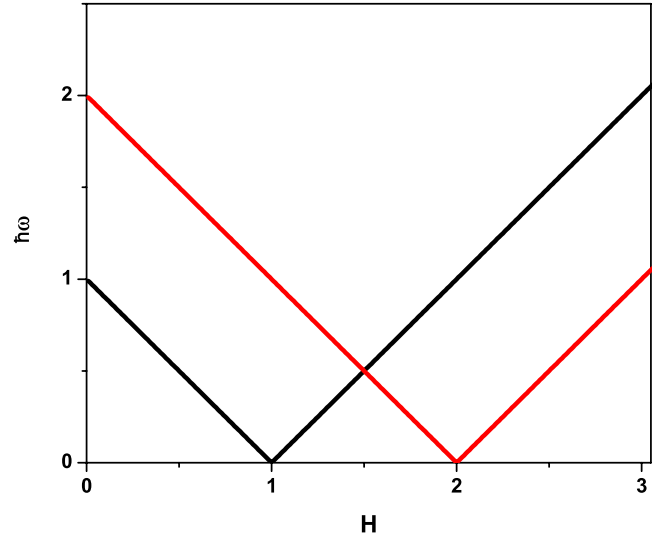


FIG. 2. (Color online) The resonance frequency-field ESR phase diagram for the resonance response of the spin-1/2 chain with alternating antiferromagnetic NN interactions and small NNN interactions;  $x_1=2$  and  $x_2=1$ .

20). The resonance conditions can be written as  $\hbar\omega_{1,2} = |H - x_{1,2}|$ , where the shifts of the resonances  $x_{1,2}$  are

$$\begin{aligned}
x_{1,2} = & \frac{1}{2} \{ (J_1 + J_2 + A_1 + A_2) R_+ \pm [(J_1 + J_2 - 2A_N)^2 R_+^2 \\
& + (A_1 + A_2 - 2A_N)(2J_1 + 2J_2 - 2A_N + A_1 + A_2) R_-^2]^{1/2} \}. \tag{17}
\end{aligned}$$

Obviously, the shifts for resonance fields exist only for non-zero magnetic anisotropy, according to the above. For  $R_- = 0$ , we have  $H - x_1 - \hbar\omega = F$ , and  $H - x_2 - \hbar\omega = H - \hbar\omega - (A_1 + A_2 + 2A_N)/2$ , i.e., Eq. (15) coincides for  $A_1 = A_2$  with the previous result for the homogeneous chain, as it must be. Also, one can see that in the absence of the anisotropy  $A_1 = A_2 = A_N = 0$ , Eq. (15) coincides with Eq. (6), as it must be too.

If  $x_{1,2} > 0$ , the resonance frequency first decreases with the growth of the value of the dc field until  $H = x_{1,2}$  and then increases with  $H$ . On the other hand, for  $x_{1,2} < 0$ , the resonance frequency increases with the growth of the dc field value. This is why, depending on the values of the NN and NNN anisotropic interactions, the behavior of the resonance shifts is different. For example, if  $(J_1 + J_2 + A_1 + A_2)(S_{0,1}^z + S_{0,2}^z) > 0$ , i.e., for antiferromagnetic NN interactions, and if  $(A_1 + A_2)S_{0,1}^z S_{0,2}^z > A_N[(S_{0,1}^z)^2 + (S_{0,2}^z)^2]$ , i.e., for small NNN interactions,  $x_{1,2} > 0$  with  $x_1 > x_2$  (see Fig. 2). Notice that the difference between  $x_1$  and  $x_2$  is taken large in Figs. 2–5 for illustration purpose. It is too large compared to known real experimental situations. For  $(J_1 + J_2 + A_1 + A_2)(S_{0,1}^z + S_{0,2}^z) < 0$ , i.e., for the ferromagnetic NN interactions, and  $(A_1 + A_2)S_{0,1}^z S_{0,2}^z > A_N[(S_{0,1}^z)^2 + (S_{0,2}^z)^2]$  (also small NNN couplings), one has  $x_{1,2} < 0$  with  $|x_1| < |x_2|$  (see Fig. 3).

On the other hand, if  $(J_1 + J_2 + A_1 + A_2)(S_{0,1}^z + S_{0,2}^z) > 0$  and  $(A_1 + A_2)S_{0,1}^z S_{0,2}^z < A_N[(S_{0,1}^z)^2 + (S_{0,2}^z)^2]$ , or for  $(J_1 + J_2 + A_1 + A_2)(S_{0,1}^z + S_{0,2}^z) < 0$ , and  $(A_1 + A_2)S_{0,1}^z S_{0,2}^z < A_N[(S_{0,1}^z)^2$

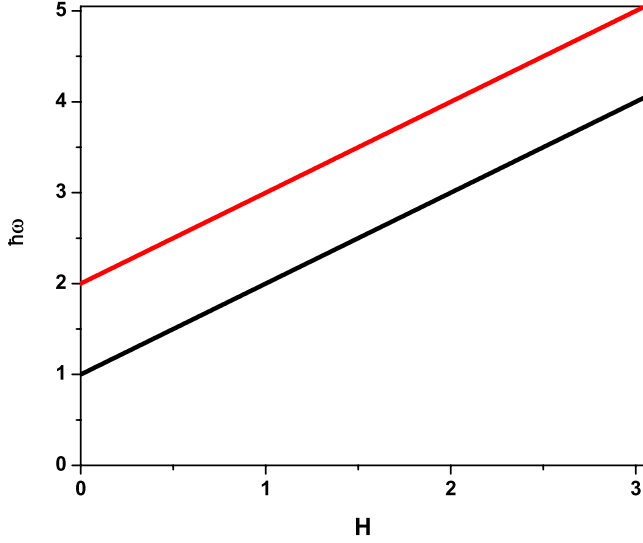


FIG. 3. (Color online) The same as in Fig. 2, but for the spin-1/2 chain with alternating ferromagnetic NN interactions and small NNN interactions;  $x_1=-1$  and  $x_2=-2$ .

$+(S_{0,2}^z)^2]$ , i.e., for large NNN interactions, one has  $x_1 > 0$  and  $x_2 < 0$  with  $x_1 > |x_2|$  for the antiferromagnetic NN couplings and  $x_1 < |x_2|$  for the ferromagnetic NN couplings, respectively (see Figs. 4 and 5). In the case of  $J_N=A_N=0$  we can write the approximate values for  $R_{\pm}$  for a quasi-one-dimensional spin system with alternation of exchange constants along chains, neglecting the renormalization of dressed charges and velocities of low-energy excitations<sup>3</sup> as

$$R_+ = \frac{2}{\pi} \int_{H_c}^{H_s} \frac{xdx}{\sqrt{(H_s^2 - x^2)(x^2 - H_c^2)}} \times \frac{\sinh(H/T)}{\cosh(H/T) + \cosh(x/T)},$$

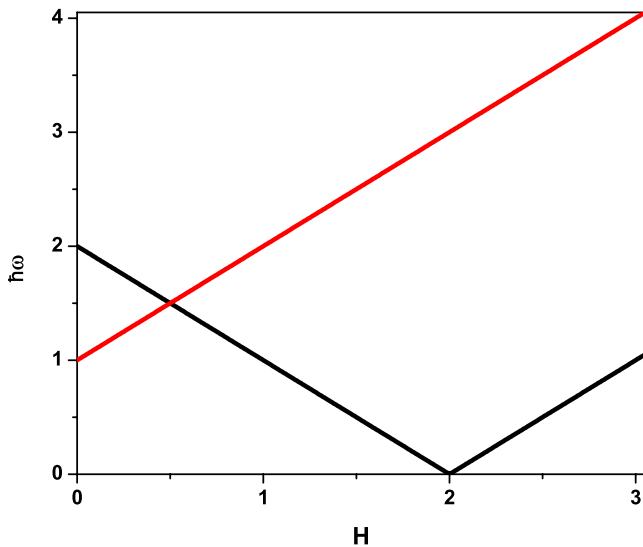


FIG. 4. (Color online) The same as in Fig. 2, but for the spin-1/2 chain with alternating antiferromagnetic NN interactions and strong NNN interactions;  $x_1=2$  and  $x_2=-1$ .

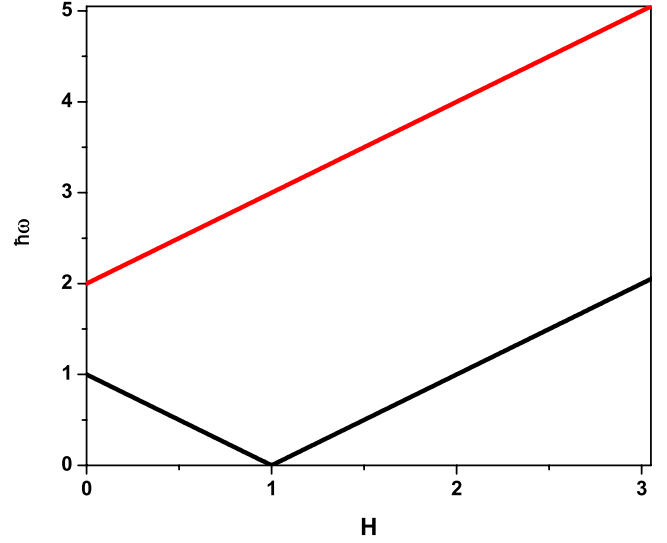


FIG. 5. (Color online) The same as in Fig. 2, but for the spin-1/2 chain with alternating ferromagnetic NN interactions and strong NNN interactions;  $x_1=1$  and  $x_2=-2$ .

$$R_- = \frac{2}{\pi} \int_{H_c}^{H_s} \frac{xdx}{\sqrt{(H_s^2 - x^2)(x^2 - H_c^2)}} \times \frac{\sinh(x/T)}{\cosh(H/T) + \cosh(x/T)}, \quad (18)$$

where  $H_c$  is the value of the field, at which the gap for the lowest-energy excitation of the dimerized spin chain is closed. The result was obtained for the case of a strong interchain antiferromagnetic coupling. Results of calculations are presented in Figs. 6–8 for several values of the magnetic field. Substituting Eq. (18) into Eq. (17) one obtains the magnetic field and temperature dependencies of the shifts of the ESR positions for the quasi-one-dimensional spin-1/2 antiferromagnetic system. One can see that at small values of the

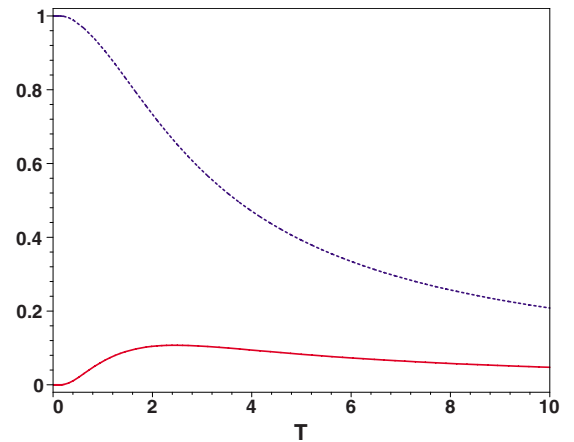


FIG. 6. (Color online) The temperature dependence of the doubled magnetization and staggered magnetization,  $R_+$  and  $R_-$  (lower red solid, and upper blue dashed-dotted lines, respectively), which determine the absorption and the shifts of the resonance positions in the quasi-one-dimensional antiferromagnetic spin-1/2 system with alternating intrachain NN interactions and strong interchain coupling for  $H_c=2$  and  $H_s=6$ ;  $H=1$ .

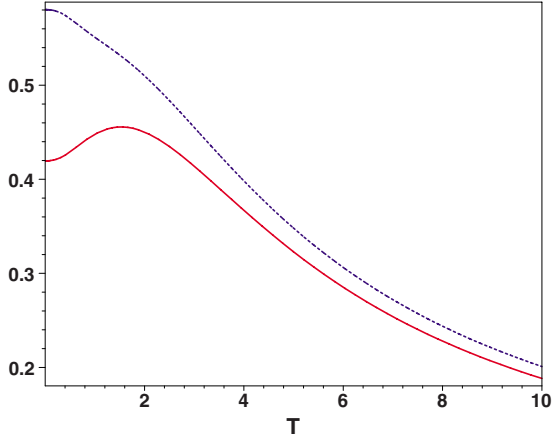


FIG. 7. (Color online) The same as in Fig. 6, but for  $H=4$ . The upper blue dashed-dotted line describes  $R_-(T)$  and the lower red solid one is for  $R_+(T)$ .

dc field the main contribution to the temperature dependence of the shift of the ESR position and absorbed power in the quasi-one-dimensional system with alternation comes from  $R_-(T)$ , while in the case of large dc field values the situation is opposite. It implies different behavior with relative phase shifts. For example, for small values of the dc field,  $H \ll H_c$ , according to Eq. (17) the shifts of the positions of two resonances have to be almost equal in absolute values,  $|x_1| \sim |x_2|$ , but different in signs,  $x_{1,2} \approx \pm (R_-/2)[(A_1+A_2)(2J_1+2J_2+A_1+A_2)]^{1/2}$  at low temperatures, similar to what happens in AFMR in standard three-dimensional magnets.<sup>2</sup> Notice the principal difference between ESR in the dimerized spin chains and AFMR. In three-dimensional ordered antiferromagnets, for temperatures lower than the Néel temperature, the order parameter (the vector of antiferromagnetism) persists, while in the one-dimensional case any nonzero temperature destroys the long-range magnetic order, and only short-range correlations determine the ESR response there. The resonance conditions for the case  $H \ll H_c$  can be approximated via the function of the staggered susceptibility as  $\hbar\omega = H [1 \pm \chi_{zz}(\mathbf{q} = \pi, H, T)[(A_1+A_2)(2J_1+2J_2+A_1+A_2)]^{1/2}$ .

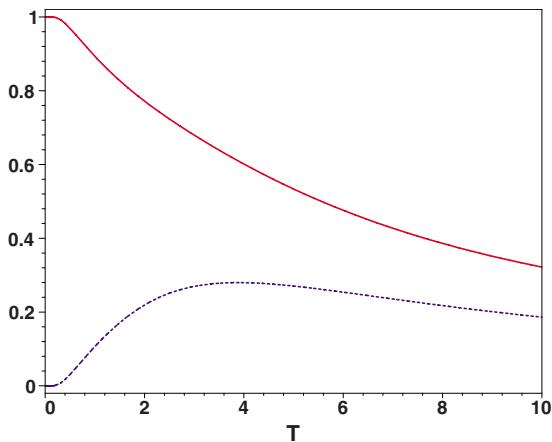


FIG. 8. (Color online) The same as in Fig. 6, but for  $H=7$ . The upper red solid line describes  $R_+(T)$  and the lower blue dashed-dotted one is for  $R_-(T)$ .

However, at higher temperatures, both  $R_+$  and  $R_-$  (or, in other words, both staggered and homogeneous susceptibilities) yield approximately similar contributions (cf. Fig. 6) and the difference in the positions of the zero-field resonances becomes more pronounced. The difference between  $|x_1|$  and  $|x_2|$  becomes more significant, if  $H_c$  is relatively small. In the general case of nonzero NNN interactions in the spin chain with alternation it is impossible yet to get the expressions for  $R_{\pm}$  in the closed form. However, we expect similar to Figs. 6–8 dependencies of  $R_{\pm}(T)$  and, therefore, all above conclusions to be qualitatively similar for weak NNN interactions too. For example, for isotropic antiferromagnetic intrachain alternating NN and weak antiferromagnetic intrachain NNN interactions, Eq. (18) is valid, with  $H_c = (1/2)|J_1 - J_2|$  and  $H_s = (1/2)[J_1 + J_2 - (16J_N/\pi)]$  (cf. Ref. 21). Temperature and magnetic field behaviors, similar to the ones presented in Figs. 6–8, take place for  $R_+$  and  $R_-$  in a single spin-1/2 chain with alternation of NN exchange couplings and alternation of effective  $g$  factors.<sup>3</sup>

Summarizing, for the alternating spin-1/2 chain the shifts of the ESR positions are determined by the magnetic anisotropy, exchange constants, and the components of the homogeneous and staggered magnetization (homogeneous and staggered susceptibilities for small dc fields), parallel to the direction of the dc field, unlike the situation with the homogeneous chain, where only the magnetic anisotropy and the homogeneous magnetization (susceptibility for small dc fields) determine the ESR shift. It is worthwhile to point out that our analysis for the dimerized spin-1/2 chain with small modifications (renotations) can be applied to the situation with the two-leg spin-1/2 ladders, which should reveal similar ESR behavior.

## VI. ESR IN SPIN CHAINS WITH DZYALOSHINSKII-MORIYA INTERACTIONS

We can include in our analysis the Dzyaloshinskii-Moriya (DM) interaction, with Dzyaloshinskii vectors directed along the direction of the dc magnetic field. It is known that DM interactions in spin chains can be of two kinds: homogeneous and alternating (staggered). For the homogeneous case one has to add to the Hamiltonian of the uniaxial spin chain the term

$$\mathcal{H}_{DMh} = D \sum_n (S_n^x S_{n+1}^y - S_n^y S_{n+1}^x), \quad (19)$$

where  $D$  is the constant of the DM coupling. The DM interaction produces effective easy-plane anisotropy,<sup>22</sup> i.e., in this case one has to replace  $A \rightarrow A + (\sqrt{D^2 + J^2} - |J|)$  in the kinetic equations and the ESR absorption. On the other hand, in the alternating case the additional part of the Hamiltonian of the spin chain has the form

$$\begin{aligned} \mathcal{H}_{DMa} = & D_1 \sum_n (S_{1,n}^x S_{2,n}^y - S_{1,n}^y S_{2,n}^x) \\ & - D_2 \sum_n (S_{2,n}^x S_{1,n+1}^y - S_{2,n}^y S_{1,n+1}^x), \end{aligned} \quad (20)$$

and one has to introduce the replacements  $A_{1,2} \rightarrow A_{1,2}$

$+\sqrt{D_{1,2}^2+J_{1,2}^2}-|J_{1,2}|$ , respectively,<sup>23</sup> in the formulas for the absorbed ESR power. Obviously, the alternation of the DM interaction produces the dimerization of the spin chain and, hence, two resonances in the ESR absorbed power, even in the case of homogeneous NN exchange couplings  $J_1=J_2$ . Clearly, shifts of the resonance positions can be caused by only DM interactions, even in the absence of the uniaxial magnetic anisotropy.

### VII. DIFFERENT GEOMETRY OF THE ESR IN SPIN CHAINS

If the direction of the dc field does not coincide with the direction of the anisotropy (including the anisotropy in the direction of the Dzyaloshinskii vector), in the framework of the linear response, neglecting nonresonance terms, the calculated shift of the resonance position has to be multiplied by, e.g., the standard direction-dependent factor  $1-3\cos^2\theta$  (cf. Ref. 24), where  $\theta$  is the angle between the directions of the dc field and the anisotropy axis. Nonresonance terms are small, because  $h\ll H$ ,  $\hbar\omega$  and give smaller contributions to the ESR response of the chain (notice that they can produce additional resonance lines, though).

### VIII. FEATURES OF THE ESR LINEWIDTH IN SPIN CHAINS

Now, let us briefly consider how spin-spin interactions affect the linewidth of the ESR in spin chains. Usually the relaxation in spin systems contain a contribution from spin-lattice (or spin-nuclear) interactions and from spin-spin interactions. To estimate the value of the spin-spin relaxation conventional theory of spin damping<sup>25,26</sup> implies  $\hbar\gamma_{ss}(\mathbf{q})\sim[T\chi(\mathbf{q})]^{-1}$ , where  $\mathbf{q}$  is the quasimomentum (in our case we need only  $\mathbf{q}=0$  for the homogeneous case and/or  $\mathbf{q}=\pi$  for the staggered magnetization appeared in the chain with alternation). This implies  $\hbar\gamma_{ss}\sim 2\pi v/T+\dots$  in a homogeneous spin-1/2 chain with antiferromagnetic interactions and exponentially large contributions at low temperatures for spin systems with gapped low-energy excitations, such as in the antiferromagnetic chain with NN and NNN interactions for  $J_N>0.24\dots J$ . However, experimental ESR studies of quasi-1D spin systems often show different behavior of the linewidth.

In the framework of the linear-response theory the shift of the resonance position of the ESR can be considered as the real part of the perturbation-caused correction to the real part of the dynamical magnetic susceptibility.<sup>9,10</sup> Kramers-Kronig relations

$$\chi''(\mathbf{q},\omega)=-\frac{1}{\pi}\mathcal{P}\int d\omega'\frac{\chi'(\mathbf{q},\omega')-\chi(\mathbf{q},\infty)}{\omega'-\omega} \quad (21)$$

establish the connection between the real part and the imaginary part of the susceptibility. The latter, in turn, can be

considered as the contribution from spin-spin interactions to the linewidth of the ESR. Hence, the shift of the resonance position and the linewidth are inter-related via the Kramers-Kronig relations in the framework of the linear-response theory. It implies the enhancement of the ESR linewidth caused by the magnetically anisotropic spin-spin interactions to be  $\hbar\gamma_{ss}\sim AT^2/v^2$ . Previous theories<sup>9,10</sup> imply a little different behavior,  $\hbar\gamma_{ss}\sim A^2T/v^2$ . Notice that similar relations with the replacement of  $A$  with necessary constants can be used for the case of the homogeneous DM interactions and the magnetic anisotropy of the NNN spin-spin interactions (see above). We point out that the frustrating NNN interactions produce the change  $v\rightarrow v[1-|\alpha/\alpha_c|]$  (see above), hence yielding near the quantum critical point larger ESR linewidth, compared to the case of the absent NNN competing couplings. On the other hand, weak alternating interactions produce  $\hbar\gamma_{ss}\sim v(A_1-A_2)^2/T^2$  (see Ref. 10), i.e., the growth of the ESR linewidth with the decrease in the temperature.<sup>27</sup>

### IX. CONCLUDING REMARKS

In summary, a simple dynamical mean-field-like theory of the electron spin resonance in spin-1/2 chains has been developed. It has been explained that either the magnetic anisotropy of spin-spin couplings or the Dzyaloshinskii-Moriya interaction can be the reason for the shift of the resonance position in spin chains, comparing to the case with only magnetically isotropic interactions (or, in the absence of interactions). The power of the ac ESR field, absorbed by the spin chain, has been calculated. It has been shown that the temperature dependence of the shift of the resonance position is related to the temperature dependence of the ESR absorption. The role of competing (spin-frustrating) next-nearest neighbor interactions in the spin chain (or zigzag ladder) has been studied. It has been shown that depending on the strength of NNN interactions, the shift of the ESR position can become either exponentially small (due to the NNN-coupling-induced spin gap) or can be increased (near NNN-governed quantum critical point). The absorbed by the alternating spin chain power of the ac magnetic field has been calculated. It reveals two resonance lines caused by the oscillations of two effective magnetic centers in the dimerized spin chain. The influence of all mentioned spin-spin interactions on the linewidth of the ESR is briefly analyzed.

### ACKNOWLEDGMENTS

I thank V. Kataev for interesting and helpful discussions and V. O. Cheranovskii for his help with the program for exact diagonalization. The financial support from the DFG (Grant No. BU 887/7-1) and the Ukrainian Fundamental Research State Fund (Grant No. F25.4/13) is acknowledged.



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