Electron spin resonance in high-field critical phase of gapped spin chains

A. K. Kolezhuk*

Institute of Magnetism, National Academy of Sciences and Ministry of Education of Ukraine 36(B) Vernadskii avenue, 03142 Kiev, Ukraine
and Institut für Theoretische Physik, Universität Hannover, Appelstraße 2, D-30167 Hannover, Germany

H.-J. Mikeska

Institut für Theoretische Physik, Universität Hannover, Appelstraße 2, D-30167 Hannover, Germany

Motivated by recent experiments on $Ni(C_2H_8N_2)_2Ni(CN)_4$ (commonly known as NENC), we study the electron spin resonance in the critical high-field phase of the antiferromagnetic $S=1$ chain with strong planar anisotropy and show that the ESR spectra exhibit several peculiarities in the critical phase. Possible relevance of those results for other gapped spin systems is discussed.

I. INTRODUCTION

Recently, there has been a growing interest in the properties of low-dimensional spin systems subject to strong external magnetic field.[1–9,10,11,12] In gapped spin systems, when the external magnetic field H exceeds a critical value H_c, it closes the gap and drives the system into a new critical phase with finite magnetization and gapless excitations. When the field is further increased, the system may stay in this critical phase up to the saturation field H_s, above which the system is in a saturated ferromagnetic state. Under certain conditions, however, the excitations in this high-field phase may again acquire a gap, making the magnetization "locked" in some field range; this phenomenon is known as magnetization plateaus and has been receiving much attention as well.[13–16]

Recently, electron spin resonance (ESR) experiments on $Ni(C_2H_8N_2)_2Ni(CN)_4$ (commonly abbreviated as NENC) in strong magnetic fields were conducted.[7] This compound is believed to be a realization of the $S=1$ chain with strong planar and weak in-plane anisotropy.[17] The theory describing ESR response for this system outside the critical phase (i.e., $H<H_c$ or $H>H_s$) was developed by Papanicolaou *et al.*[6] However, much of the ESR data of Ref. 7 belongs presumably to the field range $H_c<H<H_s$, i.e., exactly to the region where the theory is lacking, which makes the interpretation of the data rather difficult.

Motivated by those experiments, in the present paper we study theoretically the zero-temperature ESR response in the critical phase of a planar $S=1$ chain. It is shown that the typical feature of ESR in the critical phase is the appearance of continua with resonances being determined by power-law singularities instead of well-defined quasiparticle peaks. We predict that a characteristic change of the slope in the field dependency of the ESR resonance frequency takes place at the critical field $H=H_c$.

The paper is organized in the following way. In Sec. II we introduce the effective Hamiltonian for the planar chain, Sec. III is devoted to the calculation of resonance frequencies and exponents characterizing the corresponding singularities, and, finally, in Sec. IV we discuss possible existence of similar features in other gapped one-dimensional spin systems.

II. PLANAR $S=1$ CHAIN: EFFECTIVE MODEL

We start from the model of strongly anisotropic antiferromagnetic $S=1$ chain described by the Hamiltonian

$$\hat{H}=J\sum_n \mathbf{S}_n\cdot\mathbf{S}_{n+1}+D\sum_n (S_n^z)^2+E\sum_n \{(S_n^x)^2-(S_n^y)^2\}$$
$$-h\sum_n S_n^z, \tag{1}$$

where the planar anisotropy D is assumed to be much stronger than the exchange constant J, $E\ll D$ is a weak in-plane anisotropy, and $h=g\mu_B H$, where g is the Landé factor, μ_B is the Bohr magneton, and H is the external magnetic field. The in-plane anisotropy should be included since it spoils S^z as good quantum number and thus allows certain transitions which are otherwise forbidden; generally, the in-plane anisotropy constant E may be comparable to J.

Let us consider first the noninteracting case $J=0$, then Eq. (1) amounts to a single-ion problem. The spectrum of a single ion consists of three states, whose wave functions and energies read as follows:

$$|v\rangle=|0\rangle, \quad e_v=0,$$

$$|a\rangle=\cos\alpha|+\rangle-\sin\alpha|-\rangle, \quad e_a=D-\tilde{h},$$

$$|b\rangle=\sin\alpha|+\rangle+\cos\alpha|-\rangle, \quad e_b=D+\tilde{h},$$

$$\tilde{h}\equiv(E^2+h^2)^{1/2}, \quad \sin\alpha\equiv\frac{1}{\sqrt{2}}(1-h/\tilde{h})^{1/2}, \tag{2}$$

where $|0\rangle,|\pm\rangle$ denote the spin-1 states. For $J\ll D$ and weak field $h<h_c$, the ground state of the model (1) can be described, to a good approximation, as a direct product of $|v\rangle$ states of separate magnetic ions. The elementary excitations are propagating $|a\rangle$ and $|b\rangle$ states, the spectrum has a gap, and the lowest excitation is a degenerate doublet. This doublet gets split off by the external field, $|a\rangle$ state coming down

with the field, and $|b\rangle$ states going up. The low-temperature ESR response for $h<h_c$ is determined by the transitions from the ground state of the type $|v\rangle\rightarrow|a\rangle$ and $|v\rangle\rightarrow|b\rangle$.[6]

When the field h exceeds the critical value

$$h_c=\{(D-2J)^2-E^2\}^{1/2},$$

the system enters critical phase with a finite density M of $|a\rangle$ states, and a new type of ground state transitions, namely, of the $|a\rangle\rightarrow|b\rangle$ type, becomes possible (note that this transition is allowed only in presence of the finite in-plane anisotropy E). The "old" types $|v\rangle\leftrightarrow|a\rangle$ and $|v\rangle\rightarrow|b\rangle$ still remain possible. As the density M increases from 0 to 1, the $|v\rangle\rightarrow|b\rangle$ signal should become weaker, while the intensity of the $|a\rangle\rightarrow|b\rangle$ transition should increase. Above the saturation field h_s, determined by the equation

$$\tilde{h}=D+2J\{1+(h/\tilde{h})^2\},$$

the density of a particles is equal to 1.

In order to describe this picture quantitatively, we first introduce the hardcore boson operators $a_n^\dagger$, $b_n^\dagger$ creating, respectively, the $|a\rangle$ and $|b\rangle$ states from the vacuum state $|v\rangle$. Not more than one boson is allowed to be present on any site, which defines the set of physical states. The effective Hamiltonian in terms of hardcore bosons can be written as $\hat{H}_{\text{eff}}=\mathcal{P}(\mathcal{H}_0+\mathcal{H}_{\text{int}})\mathcal{P}$, where $\mathcal{P}$ is the projector onto the set of physical states. The "unperturbed" Hamiltonian $\mathcal{H}_0$ has the following form:

$$\mathcal{H}_0=\sum_n (\Delta_a a_n^\dagger a_n+\Delta_b b_n^\dagger b_n)+\sum_n (t_a a_n^\dagger a_{n+1}+t_b b_n^\dagger b_{n+1}$$
$$+\text{H.c.}),\qquad (3)$$

where the self-energies are $\Delta_{a,b}=e_{a,b}$ and the hopping amplitudes $t_a=t_b\equiv t=J$. The interaction part $\mathcal{H}_{\text{int}}$ looks as follows:

$$\mathcal{H}_{\text{int}}=U_{aa}\sum_n (a_n^\dagger a_n)(a_{n+1}^\dagger a_{n+1})+U_{ab}\sum_n \{(a_n^\dagger a_n)$$
$$\times(b_{n+1}^\dagger b_{n+1})+(b_n^\dagger b_n)(a_{n+1}^\dagger a_{n+1})\},\qquad (4)$$

where the interaction constants are given by $U_{aa}=-U_{ab}=J(h/\tilde{h})^2$.

Our goal is to take into account the effects arising in the first order in the coupling J, and therefore we have neglected the pair creation term

$$\Phi_{ab}\sum_n (a_n b_{n+1}+b_n a_{n+1}+\text{H.c.}),$$

with $\Phi_{ab}=J(h/\tilde{h})$, whose contribution to any energy level arises only in the second order in J. This simplifies the problem considerably, making the total numbers N_a and N_b of a and b particles good quantum numbers. At the same level of approximation, one can neglect the processes of exchanging the positions of neighboring a and b particles, described by the term

$$t_{ab}\sum_n \{b_n^\dagger a_{n+1}^\dagger a_n b_{n+1}+\text{H.c.}\},$$

since the "interparticle" hopping amplitude $t_{ab}=J(E/\tilde{h})^2$ is very small in the critical region, $E/\tilde{h}\sim E/D\ll 1$. Further, we will be interested in configurations with at most one b boson, so that the interaction between b particles can be also safely neglected.

Let us first consider the "unperturbed" Hamiltonian $\mathcal{P}\mathcal{H}_0\mathcal{P}$ (note that the physics described by this Hamiltonian is nevertheless nontrivial due to the single occupancy constraint). In absence of b particles $(N_b=0)$ the spectrum of the problem can be in principle obtained by mapping to noninteracting fermions with the help of the well-known Jordan-Wigner transformation, but it is more convenient to stick to the hardcore boson language. From the Bethe-ansatz solution[18] one knows that the energy spectrum is given by

$$E(\{k_l\})=\sum_l (\Delta_a+\cos k_l),\quad k_l=\pi+\frac{2\pi}{L}I_l,\qquad (5)$$

where the numbers I_l are all different and respectively half-integer (integer) if the total number of particles $N=N_a$ is even (odd), and L is the number of sites which will be assumed even for convenience. The total momentum P of a state defined by a certain choice of N different numbers I_l is $P=\Sigma_l k_l$. The ground state $|\text{g.s.}\rangle$ is given by a symmetrical dense distribution of I_l around zero:

$$I_l=-\frac{N-1}{2},-\frac{N-3}{2},\ldots,\frac{N-1}{2},\qquad (6)$$

describing the Fermi sea of particles with momenta in the interval $[k_F,2\pi-k_F]$, where we shall define

$$k_F=\pi(1-N/L).\qquad (7)$$

The total momentum of the ground state configuration is $P=0(\text{mod }2\pi)$ if N is even, and $P=\pi$ if N is odd. Since the hopping amplitudes for a- and b-particles are equal, it is easy to realize that the above picture of the distribution of wave vectors remains true when one has the total number of particles $N=N_a+N_b$, N_a of them being of the a type and N_b of the b type: they form a single "large" Fermi sea.

III. ESR TRANSITIONS

We would like to study the ESR transitions from the ground state, which survive in the low-temperature limit. The ESR intensity $I(\omega)$ for the ground state transitions is determined by the formula[19]

$$I(\omega)\propto\omega\sum_f |\sum_\alpha h_r^\alpha\langle f|S_{\text{tot}}^\alpha|\text{g.s.}\rangle|^2\delta(E_f-E_{\text{g.s.}}-\omega),\quad (8)$$

where h_r^α are the components of the radio frequency field, and f labels all possible final states.

A. $|v\rangle \mapsto |b\rangle$ transitions

Let us first consider the $|v\rangle \mapsto |b\rangle$ process with $\Delta N_a = 0$ and $\Delta N_b = 1$, as the most interesting one (note that it is allowed even in absence of the in-plane anisotropy E). This process is determined by the operator S_{tot}^-, which is in this case equivalent to $\sqrt{1 - h/\tilde{h}} \Sigma_n b_n^\dagger$. Let us assume for convenience that the initial number of particles $N = N_a$ is even, so that $N = 2N_0$. Changing the total number of particles by one causes, according to Eq. (6), the rearrangement of the allowed wave vectors. This leads to the vanishingly small transition matrix elements in Eq. (8) (orthogonality catastrophe[20]), however, it is compensated by a diverging number of states $|f\rangle$ having nearly the same energy. The problem of calculating the ESR response in this case amounts to that of calculating the spectral properties of a mobile hardcore impurity suddenly created in a hardcore boson system, the impurity having the same mass as the other particles, and the dispersion law being of the type $\Delta_{a,b} + 2t \cos k$. A similar problem was studied in Refs. 21–24 and is closely related to the so-called Fermi edge singularities in the photoemission/adsorption spectra (see, e.g., Ref. 25, and references therein).

The excited wave function $|f\rangle$ can be generally represented in the following form:

$$|f\rangle = \sum_{x_0'} e^{iP'x_0'} \sum_{y_1' \ldots y_N'} \Phi(y_1', \ldots, y_N') |x_0'; y_1', \ldots, y_N'\rangle, \tag{9}$$

where x_0' denotes the position of the b particle and $x_i' = x_0' + y_i'$, $i = 1, \ldots, N$ denote the positions of the a particles (so that y_i' are their coordinates relative to the b particle), and P' is the total momentum of the excited state.

Following Castella and Zotos,[22] one can write the reduced wave function Φ in the determinantal form

$$\Phi(y_1', \ldots, y_N') = (-)^{\Sigma'} \frac{1}{\sqrt{LN!}} \det\{\varphi_i(y_j')\},$$

$$\varphi_l(y) = A_l \left\{ e^{i(k_l'y + \delta_l)} - \frac{1}{N} \sum_{n=1}^N e^{i(k_n'y + \delta_n)} \right\}, \tag{10}$$

where A_l is the normalization factor, and the phase shift $\delta_l \equiv \delta = -\pi/2$ is independent of the state label l in our case of noninteracting hardcore particles. The factor $(-)^{\Sigma'} = \pm 1$ is the sign of the permutation

$$\begin{pmatrix} y_1', y_2', \ldots, y_N' \\ y_{i_1}', y_{i_2}', \ldots, y_{i_N}' \end{pmatrix},$$

where $y_{i_1}' < y_{i_2}' < \cdots < y_{i_N}'$, which ensures that the total wave function is symmetric under the permutation of any two particles. The allowed values for the wave vectors k_l' of the excited state are given by Eq. (5) with integer I_l. The energy of this state is given by

$$E_f = \sum_l (\Delta_a + 2t \cos k_l') + \Delta_b + 2t \cos \lambda, \tag{11}$$

where λ is defined by

$$\lambda = P' - \sum_l k_l' \tag{12}$$

and plays the role of the momentum of the b particle.

It is easy to see that the wave function $b_{x_0}^\dagger |\text{g.s.}\rangle$, obtained from the ground state by creating a b particle at site x_0, can be written in a similar determinantal form

$$b_{x_0}^\dagger |\text{g.s.}\rangle = e^{iPx_0} \sum_{y_1, \ldots, y_N} f(y_1, \ldots, y_N) |x_0; y_1, \ldots, y_N\rangle, \tag{13}$$

where $y_i = x_i - x_0$ are again relative coordinates of a-particles, $P = \Sigma_l k_l$ is the total momentum, and the reduced wave function

$$f(y_1, \ldots, y_N) = (-)^\Sigma \frac{1}{\sqrt{N!}} \det\{\psi_i(y_j)\}, \tag{14}$$

is built from simple plane waves $\psi_l(y) = (1/\sqrt{L}) e^{ik_l y}$, $(-)^\Sigma$ is the sign of the permutation

$$\begin{pmatrix} y_1, y_2, \ldots, y_N \\ y_{i_1}, y_{i_2}, \ldots, y_{i_N} \end{pmatrix},$$

where $y_{i_1} < y_{i_2} < \cdots < y_{i_N}$.

Now, one can see that the matrix element entering Eq. (8) takes the form

$$\langle f| \sum_n b_n^\dagger |\text{g.s.}\rangle = \sqrt{L} \delta(P, P') M_{fi},$$

$$M_{fi} = \sum_{y_1, y_2, \ldots, y_N} \Phi^*(y_1, y_2, \ldots, y_N) f(y_1, y_2, \ldots, y_N), \tag{15}$$

where $\delta(P, P')$ is the Kronecker symbol telling us the well-known fact that the total momentum should not change during the ESR transition, $P = P' \pmod{2\pi}$. The reduced matrix element M_{fi} can be represented as a determinant of the overlap matrix between "old" and "new" wave functions

$$M_{fi} = \det\{\langle \varphi_{l'} | \psi_l \rangle\}. \tag{16}$$

As shown in Ref. 22, the wave functions $\varphi_l(y)$ defined in Eq. (10) behave asymptotically as plane waves, $\varphi_l(y) \mapsto \tilde{\varphi}_l(y) = (1/\sqrt{L}) e^{i(k_l'y + \delta_l)}$, and the overlaps Eq. (16) calculated with φ_l can be with the accuracy of $O(\ln L/L)$ replaced by those calculated with $\tilde{\varphi}_l$. Then the matrix element M_{fi} is asymptotically equal to the overlap between two Slater-type wave functions, one describing the system with the total momentum $P = \Sigma_l k_l$ and the other corresponding to the system with the momentum $Q = \Sigma_l k_l' \equiv P' - \lambda$. It is thus clear that M_{fi} can be nonzero only if $P = Q$. This gives us the complete set of selection rules as

$$P = P', \quad \lambda = 0. \tag{17}$$

The ground state configuration is given by the following distribution of the wave vectors:

$$k_l = \pi \pm \frac{2\pi}{L}\left(l + \frac{1}{2}\right), \quad l = 0, \ldots, (N_0 - 1)$$

and has the total momentum $P = 2\pi N_0$, while in the excited state the numbers I_l are integer, and the lowest energy configuration built according to Eq. (6) would have the total momentum $\pi(2N_0 + 1)$. In order to get the momentum change ΔP back to zero, one has to introduce some excitation. In the simplest way this can be achieved by creating a hole at $k = \pi$, i.e., one gets the symmetric excited state configuration $|f_s\rangle$ given by

$$k'_n = \pi \pm \frac{2\pi}{L}(n + 1), \quad n = 0, \ldots, N_0 - 1. \tag{18}$$

The overlap M^s_{fi} can be easily calculated by means of passing to the even/odd states [i.e., to sines and cosines of $k_l y$ and $(k'_l y + \delta)$, respectively]. The determinant then factorizes into a product of two (in our case equal) Cauchy-type determinants $M^{(\pm)}$,

$$M^{(\pm)} = \det\left\{\frac{1}{\pi(n - n') - \delta}\right\}.$$

Determinants of this type were calculated by Anderson[20] and shown to be algebraically vanishing in the thermodynamic limit $M^{(\pm)} \propto L^{-\beta_\pm/2}$, with the exponents $\beta_\pm = (\delta/\pi)^2 = 1/4$, so that

$$|M^s_{fi}|^2 \propto L^{-\beta_s} \tag{19}$$

with the orthogonality catastrophe (OC) exponent $\beta_s = \beta_+ + \beta_- = 1/2$. This exponent can be also calculated in a different way, using the results of boundary conformal field theory (BCFT).[26] For this purpose it is necessary to calculate the energy difference ΔE_f between the ground state and the excited state $|f\rangle$, including the $1/L$ corrections. Then in case of *open boundary conditions* the OC exponent β, according to BCFT, can be obtained as

$$\beta = \frac{2L\widetilde{\Delta E_f}}{\pi v_F} \equiv \frac{2\widetilde{\Delta E_f}}{\Delta E_{min}}. \tag{20}$$

Here $v_F = 2t \sin k_F$ is the Fermi velocity, so that $\Delta E_{min} = \pi v_F/L$ is the lowest possible excitation energy, and $\widetilde{\Delta E_f}$ is the $O(1/L)$ part of ΔE_f (i.e., with the bulk contribution subtracted). In this last form this formula should be also valid for the *periodic boundary conditions*, then ΔE_{min} should be replaced by $2\pi v_F/L$. It is easy to calculate ΔE_f for the symmetric configuration $|f_s\rangle$: one gets $\Delta E_f^s = \Delta_b + 2t + \Delta E'_s$, where

$$\Delta E'_s = 4t \sum_{l=0}^{N_0-1}\left\{\cos\frac{2\pi}{L}\left(l + \frac{1}{2}\right) - \cos\frac{2\pi}{L}(l+1)\right\}$$

$$= 2t(1 + \cos k_F) + t\frac{\pi}{L}\sin k_F + O(1/L^2). \tag{21}$$

Applying Eq. (20), one gets the correct value $\beta = 1/2$ which coincides with that obtained through a traditional way by calculating determinants.

The symmetric excited state configuration with $Q = P$ is obviously *not* the configuration with the lowest energy. The symmetric configuration *is*, however, the configuration with the highest overlap with the ground state. Indeed, there are configurations with asymmetric distribution of momenta around $k = \pi$, which nevertheless satisfy the selection rules, e.g., the following one which we will denote as $|f_a(n)\rangle$:

$$k'_l = \pi + \frac{2\pi}{L}l, \quad l = -(N_0 - n), \ldots, (N_0 + n - 1). \tag{22}$$

If we require that $Q = P + 2\pi\Lambda$, where Λ is an integer, the following condition on k_F is obtained:

$$k_F = \pi\frac{2m - 1}{2n - 1}, \quad m \equiv n - \Lambda, \tag{23}$$

which gives the dense set of allowed values of k_F, at which the configuration of the type (22) will satisfy the selection rules. The energy difference from the ground state energy is $\Delta E_f^a(n) = \Delta_b + 2t + \Delta E'_a(n)$, with

$$\Delta E'_a(n) = t(2n - 1)^2\frac{\pi}{L}\sin k_f + O(1/L^2). \tag{24}$$

The corresponding OC exponent $\beta_a(n)$, according to Eq. (20), is given by

$$\beta_a(n) = \frac{1}{2}(2n - 1)^2 \tag{25}$$

and is always larger than the OC exponent for the symmetric configuration $\beta_s = 1/2$ (except for $n = 1$, which would mean $k_F = \pi$, i.e., the vanishing Fermi sea).

The main contribution into the ESR response is thus coming from the states whose energy is close to that of the symmetric configuration. The summation over "shake-up" (i.e., particle-hole) excitations around the symmetric configuration leads to the singularity in the ESR intensity $I(\omega)$ of the form[25]

$$I(\omega) \propto 1/(\omega - \omega_0)^\alpha, \quad \omega > \omega_0 \tag{26}$$

with $\alpha = \alpha_s = 1 - \beta_s = 1/2$, and the threshold frequency $\omega_0 = \Delta E_f^s = \Delta_b + 4t + 2t\cos k_F$. It is remarkable that the contribution from the asymmetric configuration displays no singularity, since the corresponding OC exponent $\beta_a > 1$, so that $\alpha_a < 0$; this contribution yields just some scattered intensity at frequencies $\omega \gtrsim \Delta_b + 2t$, see Fig. 1. The integrated intensity of $|v\rangle \to |a\rangle$ contribution is proportional to the total num-
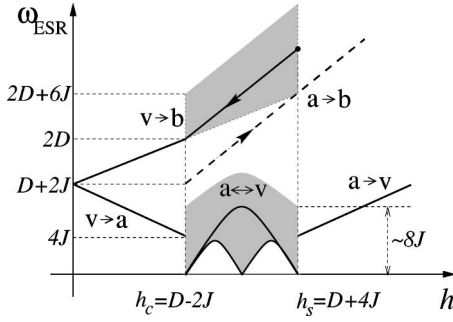
FIG. 1. Schematic picture of ground state transitions in the ESR spectrum of the planar $S=1$ chain as functions of the field in the critical region. For simplicity, the case of zero in-plane anisotropy E is shown. The dashed line corresponds to the quasiparticle peak determined by $a \to b$ processes which are forbidden in absence of the in-plane anisotropy E. Filled areas show the continua, and solid lines within the continua indicate the position of the singularities. Arrows on the lines indicate the direction of the intensity increase when varying the magnetic field.

ber $L(1-M)$ of empty sites, and thus decreases with the magnetic field; this contribution does not exist above the saturation field H_s.

Taking into account the definition of k_F, namely $\Delta_a + 2t \cos k_F = 0$, the formula for threshold ω_0 can be rewritten as

$$\omega_0(v \to b) = \Delta_b - \Delta_a + 4t. \tag{27}$$

One may notice the following remarkable feature: for weak magnetic fields below h_c the $|v\rangle \mapsto |b\rangle$ process leads to a quasiparticle peak in the ESR response at the frequency $\omega_0' = \Delta_b + 2t$, so that *below the critical field* the slope $d\omega_0'/dh$ of the ESR line as a function of the field h is approximately 1 (for $E \ll D$). For $h > h_c$ the same process yields the Fermi-edge-type singularity (26) in the ESR response, with the edge frequency ω_0 being given by Eq. (27). If one makes a reasonable assumption, that experimentally observed ESR line for $h > h_c$ follows the behavior of the edge singularity, we get for the slope of the ESR line *above the critical field* the value $d\omega_0/dh \approx 2$. Thus, the slope of the experimentally observed line should change abruptly at the critical field $h = h_c$, as shown schematically in Fig. 1. It is worthwhile to note that such a behavior is reminiscent of the picture experimentally seen in NENC (Ref. 7) in the intermediate field regime.

Though taking into account the effects of temperature is beyond the scope of the present paper, we would like to remark that the singular contribution of $|v\rangle \to |b\rangle$ processes to the ESR response should be enhanced at finite temperature, due to the presence of a finite number of holes in the ground state. The density of states has a singularity at the bottom of the Fermi sea, so that the main contribution will come at the same frequency (27). This could be important for interpreting the results of experiments on NENC,[7] where the temperature was of the order of J.

If the interaction terms (4) are taken into account, the corresponding effective model cannot be solved exactly anymore, and we cannot calculate the OC exponents in this

case. However, one can treat the effect of interactions in a sort of the ''mean-field'' approximation, i.e., replacing simply

$$\Delta_a \mapsto \Delta_a(M) = \Delta_a + 2U_{aa}M,$$

$$\Delta_b \mapsto \Delta_b(M) = \Delta_b + 2U_{ab}M, \tag{28}$$

where the particle density $M = 1 - k_F/\pi$ has to be calculated self-consistently from the equation

$$\Delta_a(M) + 2t \cos k_F = 0. \tag{29}$$

One can check that such an approximation, though being crude, delivers correct values for the critical fields H_c and H_s. Actually, in absence of b particles one can show that the effect of U_{aa} is not only to change the self-energy Δ_a, but also to renormalize the hopping amplitude t_a [in the first order in U_{aa} one gets $t_a \mapsto t_a + (2U_{aa}/\pi)\sin k_F$], and both t_a and U_{aa} are of the order of J, so this correction is not negligible. According to the numerical studies,[27] the OC exponent β is rather sensitive to the ratio of hopping amplitudes $r = t_b/t_a$ [the behavior of $\beta(r)$ is approximately linear around $r=1$], so that the corresponding OC exponents will certainly change compared to the noninteracting (hardcore) case, and they will become k_F dependent. One may nevertheless expect that at the *qualitative* level our picture of transitions remains true, particularly, the effect of changing the slope of ESR lines at $H = H_c$ should survive.

B. $|a\rangle \mapsto |b\rangle$ transitions

The $|a\rangle \mapsto |b\rangle$ processes are possible only in presence of the finite in-plane anisotropy E, which mixes $|-\rangle$ and $|+\rangle$ states into $|a\rangle$ and $|b\rangle$, respectively. Those transitions are determined by the operator S_{tot}^z, which is in this case equivalent to $(E/\tilde{h})\Sigma_n b_n^\dagger a_n$. One can easily see that now the wave functions $\Sigma_n b_n^\dagger a_n |\text{g.s.}\rangle$ and $|f\rangle$ can be both represented in the form (9), (10) with $N \to (N-1)$. The total momentum of the ground state can be represented as $P = \Sigma_{i=1}^{N-1} \tilde{k}_i + \tilde{\lambda}$, and the momentum of the excited state has a similar form, $P' = \Sigma_{i=1}^{N-1} k_i' + \lambda$. Through the same line of arguments as before one gets the selection rules

$$P = P', \quad \lambda = \tilde{\lambda}.$$

Since the number of particles was not changed during the transition, the allowed values of wave vectors k_i' are now the same as $\tilde{k}_i$ and are given by $\pi + (2\pi/L)I_i$ with half-integer I_i. Thus, there is *no orthogonality catastrophe* in this case, and one gets a well-defined *quasiparticle peak* at the frequency

$$\omega_{a \to b} = \Delta_b - \Delta_a. \tag{30}$$

One can visualize this process as simply replacing one of the a particles in the Fermi sea of the ground state by a b particle, without changing the wave vector distribution. The corresponding ESR line goes parallel to the $|v\rangle \to |b\rangle$ line, slightly below it, as shown schematically in Fig. 1. The in-

tensity of this line is proportional to the total number LM of a particles and increases with the field; this line continues to exist at $H > H_s$.

C. $|v\rangle \leftrightarrow |a\rangle$ transitions

Those transitions are allowed in absence of the in-plane anisotropy. The number of particles changes by one, which leads to a change in the distribution of momenta and indicates absence of the quasiparticle peak. Since only two states per site are involved (N_b remains zero), those processes can be treated within the effective spin-$\frac{1}{2}$ model, similarly to the way it was done for spin ladders.[8,14] If one neglects the small corrections of the order of $(E/h)^2$, this effective model is the easy-plane XXZ chain in an effective longitudinal magnetic field with the Hamiltonian[28]

$$H_{\mathrm{eff}} = \sum_n \tilde{J}_{xy}(\tilde{S}_n^x \tilde{S}_n^x + \tilde{S}_n^y \tilde{S}_n^y) + \tilde{J}_z \tilde{S}_n^z \tilde{S}_n^z - \tilde{B}\tilde{S}_n^z,$$

where $\tilde{S}^\mu$ are the spin operators of the effective spin-$\frac{1}{2}$ chain, $J_{xy} = 2J$, the easy-plane anisotropy $J_z/J_{xy} = 1/2$, and the field $\tilde{B} = h - J - D$.

Calculating the low-frequency ESR response amounts then to the knowledge of the dynamical structure factor $S^{+-}(q, \omega)$ of the effective XXZ chain $I(\omega) \propto S^{+-}(q=0, \omega)$. The asymptotic behavior of the correlation functions for the XXZ chain is known from the bosonization results[11,29]

$$\langle \tilde{S}^+(x,t)\tilde{S}^-(0,0)\rangle = \frac{C_1}{(x^2 - v_F^2 t^2)^{1/(4K)}}\cos(\pi x)$$

$$+ \frac{C_2}{(x^2 - v_F^2 t^2)^{K+1/(4K)-1}}\left\{ \frac{e^{i(2k_F-\pi)x}}{(x-v_F t)^2} \right.$$

$$\left. + \frac{e^{-i(2k_F-\pi)x}}{(x+v_F t)^2} \right\}. \tag{31}$$

Here $C_{1,2}$ are some constants, v_F is the Fermi velocity, and K is the so-called Luttinger liquid parameter. For $\tilde{B} = 0$ [which corresponds to $k_F = \pi/2$, or $M = 1/2$, or equivalently to the physical field H in the middle of the critical region, $H = \frac{1}{2}(H_c + H_s)$] and $J_z/J_{xy} = 1/2$ one has[12]

$$v_F = 3J\sqrt{3}/2, \quad K = 3/4. \tag{32}$$

At nonzero $\tilde{B}$ (i.e., for $M \neq 1/2$) the values of v_F and K can be obtained numerically by solving the Bethe ansatz equations;[30] the corresponding solutions are presented in Fig. 2 of Ref. 12. For $k_F \to \pi$ (0), which corresponds to $H \to H_c$ (H_s), respectively, i.e., when the number of particles (holes) becomes small, K tends to 1 and v_F to zero.

Fourier-transforming Eq. (31), one gets the $q=0$ dynamic structure factor $S^{+-}(q=0, \omega)$ having edge-type singularities at the frequencies

$$\omega_0(Q) = v_f Q, \tag{33}$$

where $Q = \pi$ and $Q = 2k_F - \pi$ for the contributions of the first and second term in Eq. (31), respectively. The singularities are of power-law type, with the exponent being determined by the Luttinger liquid parameter K:

$$I(\omega) \propto \frac{1}{[\omega - \omega_0(Q)]^{\eta(Q)}},$$

$$\eta(\pi) = 1 - \frac{1}{4K}, \quad \eta(2k_F - \pi) = 2 - K - \frac{1}{4K}. \tag{34}$$

Thus, two lines corresponding to the singularities in the $|v\rangle \leftrightarrow |a\rangle$ processes at $\omega = \omega_0(\pi)$ and $\omega = \omega_0(2k_F - \pi)$ should be visible in the ESR spectrum, as shown schematically in Fig. 1. Inclusion of the finite temperature would lead to damping of the singularities.[9]

IV. DISCUSSION

In the model of a planar $S = 1$ chain considered in the previous section we have seen several features of the ESR spectrum in the high-field critical phase: appearance of low-lying and high-lying continua with power-law singularities, counterintuitive change of the slope of ESR lines at the critical field $H = H_c$, etc. One may ask oneself whether similar features can be present also in other one-dimensional spin systems, e.g., strongly coupled $S = \frac{1}{2}$ ladders such as $Cu_2(C_5H_{12}N_2)_2Cl_4$ [usually abbreviated CuHpCl (Ref. 3)] and $(C_5H_{12}N_2)_2CuBr_4$ [abbreviated BPCB (Ref. 31)] or $S = 1$ Haldane chain $Ni(C_5H_{14}N_2)_2N_3(PF_6)$ [known as NDMAP (Refs. 4,5)].

As far as the model of $S = \frac{1}{2}$ ladder with strong rung interaction J_R and weak leg coupling $J_L \ll J_R$ is concerned, one can construct the effective model exactly in the same way as for the large-D $S = 1$ chain. The role of vacuum state $|v\rangle$ will be played by singlet state $|s\rangle$ on a single rung, and one will have three types of "particles" corresponding to the three triplet states $|t_\pm\rangle$, $|t_0\rangle$. Applying a strong magnetic field will lead to closing the gap and to a finite density of $|t_+\rangle$ states in the ground state in the critical phase. However, in absence of any additional couplings (e.g., Dzyaloshinskii-Moriya interaction or nonuniaxial anisotropies) the only possible direct transitions from the ground state are determined by the processes of the $|t_+\rangle \to |t_0\rangle$ type, which, similarly to the $|a\rangle \to |b\rangle$ processes considered in the previous section, yield a quasiparticle peak with the resonance frequency $\omega_{+0} = h$. Only in presence of such additional interactions providing finite admixture of triplets in $|v\rangle$ one could see $|v\rangle \to |t_0\rangle$ and $|v\rangle \to |t_-\rangle$ lines, which should then exhibit the change of slope at $H = H_c$ with the simultaneous appearance of the continuum above them. Similar arguments apply also to the $S = 1$ Haldane chains: generally, in order to observe the "interesting" lines, one needs to have some perturbations allowing the direct transitions from the ground state at $H = 0$.

In this sense, the planar $S = 1$ chain is a remarkable model, where features such as the slope change or the low-lying continuum can be observed "generically," without appealing to the existence of any additional interactions. However, one may in principle hope that similar effects could be

observed in other one-dimensional spin systems as well. There are, for example, experimental indications[32] that such additional terms are really present in CuHpCl.

Some of the features predicted by the present study, namely, the change of the slope of the ESR line at $H = H_c$, resemble the picture experimentally seen in NENC (Ref. 7) at $H \simeq 4.5$ T. One might speculate that such an identification suggests that the actual value of D is somewhat larger than one adopted in Ref. 7, and is about 7.2 K, and that J is respectively smaller, about 0.25 K. However, a quantitative comparison of our predictions with the experimental data is hindered by several factors: first of all, we have studied only $T = 0$ case, while the experimental data[7] are given for the temperature $T = 4.2$ K, which is large compared to the exchange constant J. Second, since the local anisotropy axes in adjacent nickel chains in NENC are slightly tilted with respect to the crystallographic c axis, one actually has to include a finite transversal field component into the Hamiltonian (1), which would lead to considerable difficulties in the further analysis (e.g., the necessity to handle unequal hopping amplitudes t_a and t_b along with a finite exchange amplitude t_{ab}). Our analysis includes also certain oversimplifying approximations (such as neglecting interaction between the hardcore bosons). Finally, as one can conclude from the disagreement between the results of fitting the susceptibility data for different field directions,[7] at present there is no reliable data on the Hamiltonian parameters for NENC, which makes a conclusive quantitative comparison rather difficult.

In recent experiment[5] on the $S = 1$ Haldane chain compound NDMAP it was observed, that one of the ESR branches was just continuing into the critical region, without any noticeable features at $H = H_c$. It is worth noting that this feature closely resembles our conclusions for the $|a\rangle \rightarrow |b\rangle$ processes in the planar chain: if one denotes the (field-dependent) magnon gaps for NDMAP as $\Delta_\pm$ and Δ_0, then at $H < H_c$ the abovementioned ESR branch corresponds to the thermally excited transition within the magnon triplet, with the frequency $\omega = \Delta_0 - \Delta_+$. At $H > H_c$ there is a finite density of Δ_+ magnons in the ground state, and, if we exploit the analogy with our picture of $|a\rangle \rightarrow |b\rangle$ processes for the planar chain (identifying the $S^z = +1$ magnon with a and the $S^z = 0$ magnon with b), one can expect the presence of a quasiparticle peak at exactly the same frequency, in agreement with the experiment.

Finally, some words of caution are here in order. In the present paper, we have studied only the *ground state transitions*, in other words the ESR response at zero temperature, and only in purely one-dimensional (1D) model. When interpreting the experimental data, one should have in mind that, because of the gapless nature of the ground state in the critical phase of the 1D system, the effects of temperature become important, as well as those of weak 3D coupling; particularly, the system should be 3D ordered under certain critical temperature. Thus, the results presented here should only be taken as a guide displaying features of the purely 1D behavior. Further work is required to analyze the possibility of observation of those phenomena in realistic systems.

*Permanent address: Institute of Magnetism, National Academy of Sciences and Ministry of Education of Ukraine, 36 B Vernadskii avenue, 03142 Kiev, Ukraine.

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