# MAGNETIC FIELD-INDUCED GAP IN QUANTUM SPIN CHAINS: EPR CHARACTERISTICS*

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Recently several compounds revealed properties of a one-dimensional (1D) spin 1/2 antiferromagnetic Heisenberg chain (AHC) with gapless low-lying spin excitations in the absence of an external magnetic field $H$. A weak external magnetic field leads an appearance of a spin gap. The gap onset is related to the staggered Dzyaloshinskii–Moriya (DM) interaction or effective $g$-factors of magnetic ions. Experimentally measured thermodynamic properties cannot distinguish between those two origins for the spin–gap formation. We propose the experiment, by which one can distinguish between those two possibilities: an electronic paramagnetic resonance (EPR) in the parallel pumping geometry.

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1. The interest to quantum low-dimensional spin systems has grown considerably. The spin 1/2 AHC is probably one of the best studied models. This system is disordered even in the ground state in the absence of $H$ [1]. Low-lying excitations (LLE) are spinons, which are gapless in the absence of external fields and relativistic interactions. This gapless behavior of LLE is the origin of the finite zero-$T$ magnetic susceptibility and linear in $T$ behavior of the specific heat for this model. Due to the absence of the spin gap it has been difficult to find real materials with properties similar to low-$T$ features of the spin 1/2 AHC: Any inter-chain spin–spin interaction in a 3D system produces a magnetic ordering and the low-$T$ behavior of magnetic characteristics are determined by it. The behavior of 1D quantum spin systems with gapped LLE (which possess spin gaps) are very different,

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*e.g.*, for HAC with integer site spins [2], for half-integer spin systems with the "easy-axis" magnetic anisotropy [1] and systems with the spin-Peierls period doubling, where the spin gap persists at zero values of $H$, too. Recently new materials were discovered, which behavior is reminiscent of the spin 1/2 AHC in the absence of $H$ (*i.e.*, their LLE are gapless), while in the presence of a weak $H$ their properties manifest that the magnetic field can induce a spin gap for LLE: the copper benzoate [3], $Yb_4As_3$ [4], in which effective chains along special directions are formed and copper pyrimidine [5]. In those compounds $H$ causes the spin gap, either due to staggered (alternating) DM interaction [6] or because of staggered effective $g$-factors of magnetic ions [7]. Both, a staggered DM interaction and staggered external magnetic field can produce a spin gap for LLE. This gap defines exponentially small low-$T$ magnetic susceptibility and magnetic specific heat there. It is difficult to determine, which one of these two staggered interactions are present in real compounds.

2. Here we propose the set-up for an experiment, which can distinguish between those two mechanisms of the formation of the spin gap: the EPR. We start with the effective Hamiltonian

$$\mathcal{H}_0 = J\sum_j \vec{S}_j\vec{S}_{j+1} + (J_z - J)\sum_j S_j^z S_{j+1}^z - \sum_j [(\vec{H}+\vec{h}_0\cos\omega t)\times(\hat{g}_1\vec{S}_{2j}+\hat{g}_2\vec{S}_{2j+1})] + \sum_j (D_1[\vec{S}_{2j}\times\vec{S}_{2j+1}]^z + D_2[\vec{S}_{2j+1}\times\vec{S}_{2j+2}]^z)\,, \quad (1)$$

where $S_j^{x,y,z}$ are the operators of the projections of $S = 1/2$ spin at site $j$, $J \geq 0$ is the exchange coupling, $J^z$ $(-J \leq J^z \leq J)$ is the parameter of the magnetic anisotropy, $D_{1,2}$ are the constants of the DM interaction (DM vectors are supposed to be collinear to the direction of the magnetic anisotropy and $(D_1+D_2)/2$ describes a homogeneous DM coupling, whereas $(D_1 - D_2)/2$ is connected with a staggered DM interaction), the matrices $\hat{g}_{1,2}$ determine effective (staggered) $g$-factors of magnetic ions, $\omega$ and $h_0$ are the frequency and the magnitude of the ac magnetic field, respectively.

3. In the absence of any interactions the standard EPR geometry (*i.e.*, the ac magnetic field polarized perpendicular to the dc field) the EPR manifests two maxima in the absorbed power of the ac field at $\hbar\omega_{\rm res} = g_{1,2}^z H$, and $g_{1,2}^z$ are the components of the $g$-tensor. Here we propose to study the different EPR geometry, with the ac magnetic field collinear to the direction of the dc field: the parallel pumping. This geometry permits to find solutions for characteristics of the system under parallel pumping in the closed form, as we show below. On the other hand, it permits to detect the origin of the spin gap formation. For simplicity let us consider the case $H^x = H^y = 0$, $H^z = H$ and in the following we drop the indices $z$ for the components of the $g$-tensor. In this case we know [6, 8] that there are two branches of LLE

with the energies $\varepsilon_{k,1,2}$. It determines the onset of two critical magnetic fields in the ground state: $H_{c1}$, at which the spin gap is closed, and $H_{c2}$, of the transition to the spin-polarized state [8]. Then, using the small parameter $|g_1 - g_2|h_0/\hbar\omega \ll 1$, we perform the resonance approximation [8], *i.e.*, we single out the terms which make nonzero contribution to the linear response of the system, while other terms, explicitly dependent of time, we discard. The main resonance processes for a parallel pumping result from the merger of an elementary spin rotation of spin at even site of the chain (LLE of one branch of the spectrum) with a photon of the pumping field which gives rise to a spin rotation of the spin at an odd site of the chain in the opposite direction (LLE from the other branch) and vice versa. In the steady-state regime the absorption is essentially nonzero. It is finite (limited) for any magnitudes of the ac field. There is no any threshold value for the magnitude of the ac field (usually characteristic to the parallel pumping in magnetically ordered systems), *i.e.*, the parallel EPR pumping does not lead to the parametric instability. The reason is the reduced dimensionality (*i.e.*, LLE are spinons, which satisfy the Fermi statistics rather than spin waves, characteristic for ordered magnets, which obey the Bose statistics). The homogeneous and staggered total magnetic moments oscillate with the frequency of the ac field in the steady-state regime. The time-independent parts of the total staggered and homogeneous magnetic moments, caused by the pumping, are proportional to $h_0^2$ at small $h_0$, as expected. In the linear response regime the ac absorption has the form

$$Q = N\omega \frac{(g_1 - g_2)^2 h_0^2}{8} \int dk (n_{k,2}^0 - n_{k,1}^0)(1 - \alpha^2)\delta(\varepsilon_{k,1} - \varepsilon_{k,2} - \hbar\omega)\,, \quad (2)$$

where $n_{k,1,2}^0$ are the Fermi distribution functions with energies $\varepsilon_{k,1,2}$ and $\alpha = (g_1 - g_2)H/(\varepsilon_{k,1} - \varepsilon_{k,2})$.

4. In the spin-polarized state $H \geq H_{c2}$ the absorption is equal to zero (it is small for $T \neq 0$ for $H \geq H_{c2}$) for any $D_1$ and $D_2$. In the spin-gapped state $H \leq H_{c1}$, $Q$ is maximal for $H = 0$, and smoothly becomes smaller with the growth of the value of the dc magnetic field. $Q$ is small at high $T$. The ac field can affect the AHC only for $E_{\min} \leq \hbar\omega \leq E_{\max}$, *i.e.*, one has the frequency threshold effect. For $H \geq H_{c2}$, $E_{\min} = E_{\max} = (g_1 + g_2)H_{c2}$. For $H \leq H_{c2}$ we have $E_{\max} = \sqrt{(g_1 - g_2)^2 H^2 + 4g_1 g_2 H_{c2}^2}$. $E_{\min}$ is equal to $\sqrt{(g_1 - g_2)^2 H^2 + 2g_1 g_2 H_{c1}^2}$ for $H \leq H_{c1}$, and $(g_1 + g_2)H$ for $H_{c1} \leq H \leq H_{c2}$. In the resonance the response of the AHC is affected by the van Hove singularities $\sim ([(\hbar\omega)^2 - E_{\min}^2][E_{\max}^2 - (\hbar\omega)^2])^{-1/2}$, characteristic for 1D systems. $Q$ is maximal in the ground state for the resonance frequency, $\hbar\omega_{\rm res}$. It is (formally) equal to $\hbar\omega_{\rm res} = (g_1 + g_2)H_{c2}$ for $H \geq H_{c2}$ ($Q = 0$ in this region in the ground state), and for $H \leq H_{c2}$ we have $(\hbar\omega)_{\rm res}^2 =$

$(g_1 - g_2)^2 H^2 + 8 g_1 g_2 H_{c2}^2 A$, where $A = H_{c1}^2/(H_{c1}^2 + H_{c2}^2)$ for $H \le H_{c1}$, and $A = H^2/(H^2 + H_{c2}^2)$ for $H_{c1} \le H \le H_{c2}$. In the critical region, $H_{c1} \le H \le H_{c2}$, with gapless excitations, we expect the resonance characteristics of our system to be renormalized due to the inter-chain coupling (*i.e.*, due to the possible magnetic ordering). However, magnetic sub-lattices in the ordered phase must also have different nominal values of magnetizations because of different values of effective $g$-factors. Hence the process of the merger of an elementary rotation of a magnetic moment of one magnetic sub-lattice with a photon of the parallel pumping, which gives rise to a rotation of a magnetic moment of the other sub-lattice in the opposite direction, has to persist in the ordered phase, too.

5. The main conclusion is following: The effects of the staggered DM interaction and staggered $g$-factors are different under the condition of the EPR parallel pumping. The main difference comes from the fact that the real and imaginary parts of the dynamical magnetic susceptibility become zero for $g_1 = g_2$, while they are finite for $D_1 = D_2$, but for $g_1 \neq g_2$. In the case of $D_1 = D_2$ the spin-gapped phase does not exist ($H_{c1} = 0$) and the maximum for the absorption is manifested only as a smooth function of the applied dc field. On the other hand, for $D_1 \neq D_2$ one can see two different regimes for the resonance absorption as a function of $H$. In the spin-gapped phase the resonance frequency has an activation character. For small values of $|g_1 - g_2|$ the slope of the dependence of the resonance frequency as a function of $H$ is small in this phase. For $H_{c1} \le H \le H_{c2}$ the dependence of the resonance frequency has a negative curvature (with the effective $g$-factor renormalized by spin–spin interactions). This effect can be experimentally observed in the effectively 1D AHC, which manifest a magnetic field-induced spin gap of LLE, to check which mechanism, the staggered DM interaction, or staggered $g$-factors (or both) form the field-induced spin–gap there.

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