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Direct Numerical Analysis of ESR Line Shape in Low Dimensional Spin Systems

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We have developed a numerical method to estimate the behavior of electron spin resonance (ESR) absorption. By this method we investigated the dependence of the ESR line shape in various configurations of the external parameters in quantum spin systems with short range correlation. We found the dependence of the line shape on the temperature and also on the directions of the field. We propose to use ESR as the method to investigate the details of the geometrical and the magnetic structure of material.

Keywords electron spin resonance, resonant shift, dynamical shift, zigzag chain, metamagnetic transition

INTRODUCTION

Electron spin resonance (ESR) is a useful technique that enables us to search for energy structure of material. We have developed a numerical method to calculate the Kubo formula that gives the resonant absorption as a function of the frequency of the oscillating field[1]. The present method possesses a flexibility for the variety of the geometrical and the magnetic structure of the system, although this method has a limitation on number of spins because of computational capacity. We have investigated the behavior of the ESR line shape intensively in low-dimensional quantum spin systems. In particular, we have studied the dynamical shift, *i.e.*, dependence on the configuration of the static field and the oscillating field. The results are analyzed from a view point of the energy structure and the matrix elements between the microscopic states. We expect that our method is valid to analyze the experimental data in low-dimensional

systems, especially those with so-called spin gap where the spin correlation function is of short range. We also study the dependence of ESR line shape on the oscillating frequency, we obtained the overlook picture in the whole range of the frequency. We have also applied the method to the antiferromagnetic ordered state due to the uniaxial anisotropy. The field- and frequency-dependence of the resonant peak for this state has been investigated extensively as 'the antiferromagnetic resonance'. We found that the present method qualitatively reproduces the dependence, and we also found properties due to the short range fluctuation, *e.g.*, the coexistence of peaks due to the paramagnetic resonance and the antiferromagnetic resonance, which is not included in the conventional treatments[2]. In the present paper, however, we concentrate ourselves in the spin system without long range order.

FIELD-ANGLE DEPENDENCE OF RESONANT SHIFT

Extensive studies have been made on the ESR in interacting spin systems

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\mathrm{D}} - \left(\mathbf{H}_0 + \mathbf{H}_1 \mathrm{e}^{i\omega t}\right) \cdot \sum_i \mathbf{S}_i \tag{1}$$

where $\mathcal{H}_0$ is the spin exchange interactions and $\mathcal{H}_{\mathrm{D}}$ is for the dipole interaction. Kanamori and Tachiki deduced a method to treat the shift of the resonant field, solving the motion equation of an effective uniform magnetic moment of the system[3]. Nagata and Tazuke (NT) used this formalism to obtain the temperature dependence of the resonance in one-dimensional antiferromagnetic Heisenberg model with $\mathcal{H}_{\mathrm{D}}$. They explained successfully experimental data of the shift of the resonant field in $CsMnCl_3{\cdot}2H_2O$[4]. If the static field is applied perpendicular to the one-dimensional spin chain, the shift of the resonance from the paramagnetic resonant field is positive, while it is negative for the case of the field parallel to the chain. We introduced a direct numerical method calculating the Kubo formula for the imaginary part of the dynamical susceptibility

$$\chi''_{xx}(\omega) = \frac{1}{2}\left(1 - \mathrm{e}^{-\beta\omega}\right) \int_{-\infty}^{\infty} \langle M^x(0)\, M^x(t)\rangle\, \mathrm{e}^{-i\omega t}\, dt. \tag{2}$$

In this method we diagonalize the Hamiltonian of quantum spin systems which have various geometrical and magnetic structures. With eigenenergies and eigenfunctions of the system, we directly estimate

the imaginary part of the susceptibility $\chi''_{xx}(\omega)$. We succeeded in reproducing the result of NT by our method. We also studied another configuration. For example, we fix the static field H_0 perpendicular to the spin chain and rotate the direction of the oscillating field H_1 around it. We found that the line shape changes according to the angle between the spin chain and H_1. Furthermore we found that the shift is sensitive to the oscillating frequency.

FINE DETERMINATION OF MAGNETIC PARAMETERS

In this section we consider the possibility to use the ESR measurement to determine the fine structure of magnetic interactions of material. The lattice structure of the material can be determined very fine by the X-ray diffraction. The strength of the magnetic interactions are estimated from the temperature dependence of the specific heat and the magnetic susceptibility, and so on. When the spin system has a gap, it is difficult to determine them in general because of the weak response to the field. For example, if the system consists of ferromagnetic-antiferromagnetic bond-alternating zigzag chain, it is difficult to determine which bond is the antiferromagnetic. In such systems, we can expect that the line shape reflects the geometrical relation among H_0, H_1 and the directions of ferromagnetic bond and the antiferromagnetic bond. First let us study the difference of the line shapes between two configurations where H_0 directs along the ferromagnetic bond and along the antiferromagnetic bond, with H_1 fixed perpendicular to the zigzag chain (Fig. 1). As shown in Fig. 1 where we study a lattice in which the angle between the bonds is 90° and the strength of the both interactions, J_{F} and J_{AF}, respectively, are the same, we find a clear difference between the two configurations. When H_0 is perpendicular to the antiferromagnetic bond, the shift is positive which is the same sign as the case of the one dimensional case (Fig. 2). On the other hand, when H_0 is perpendicular to the ferromagnetic bond, the shift is negative. Thus we can determine which bond is the antiferromagnetic.

Even if we change the ratio of the interactions, the present tendency remains except for the case $|J_{\mathrm{F}}/J_{\mathrm{AF}}| \gg 1$. If we change the direction of the static field from that along the ferromagnetic bond to that along the antiferromagnetic bond, the resonant shift changes and shows the maximum value at one angle. If we change the angle between the bonds from 90°, the angle for the maximum value also changes. The details will be published elsewhere[5]. We also find dif-

ferent behavior in the dynamical shift from that in the linear chain, because $\mathcal{H}_{\rm D}$ consists of the dipole-dipole interactions of different directions in the zigzag lattice. For example, $(CH_3)_2CHNH_3CuCl_3$ and $Cu_{1-x}Zn_xNb_2O_6$ have zigzag structures. Although there are many other interactions which prevents to observe the present pure behavior, we hope that we can detect some signals which originates in the present mechanism and we can use the ESR measurements as a fine tool to investigate the magnetic interactions of the material.

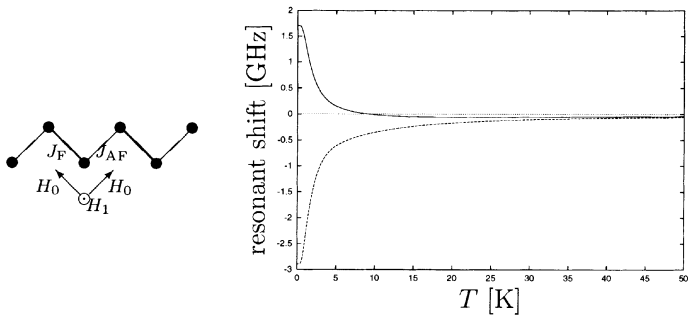

FIGURE 1: Geometrical configuration and temperature dependence of resonant shift in the ferromagnetic-antiferromagnetic bond-alternating zigzag chain. Solid line is corresponding to the case of the static field H_0 parallel to the ferromagnetic bond, and dashed line to the case of H_0 parallel to the antiferromagnetic bond.

ESR IN DIMERIZED SPIN SYSTEMS

Teki *et al.* have observed the temperature dependence of the line shape in a one-dimensional bond-alternating antiferromagnet N-[(2,4-dichlorophenyl)thio]-2,4,6-triphenylphenylaminyl[6], and found the resonant shift shows a qualitatively different behavior from that obtained by the method of NT[4]. We found that the difference is due to the frequency dependence of the shift. In Fig. 2, the dependence of the shift on the frequency is shown. In low-frequency region we find the same tendency as that in the experiment. When we decrease the frequency, the shift for H_0 perpendicular to the chain goes down to negative in low-temperature region. This frequency dependence can be understood from the energy structure and the matrix elements between them[7].

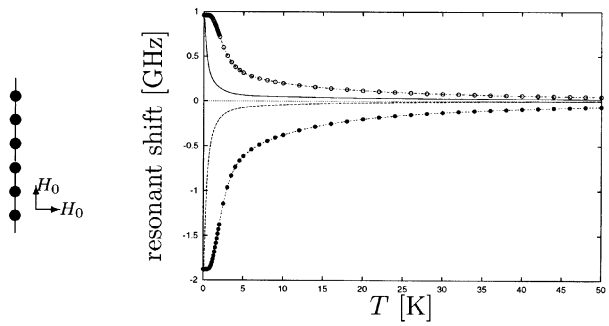

FIGURE 2: Geometrical configuration and temperature dependence of resonant shift in the bond-alternating antiferromagnetic chain. Solid and dashed lines correspond to the case of the static field H_0 perpendicular and parallel to the chain, respectively for $\omega = 9.5$ GHz. Open and closed circles are to the case of H_0 perpendicular and parallel to the chain, respectively for $\omega = 95$ GHz, and lines are guides for eye.

ESR AT METAMAGNETIC TRANSITION POINT

Ohta *et al.* observed the frequency-dependence of the resonant shift in the antiferromagnetic ladder system with a diagonal exchange integral that causes frustration Cu_2(1,4-diazacycloheptane)$_2Cl_4$ (abbreviated as CHpC)[8]. There the resonant shift shows a staircase-like increase with the resonant field which roughly corresponds to the oscillating frequency. This dependence means that the g-factor of the material varies in the staircase-like way. Magnetization process shows a rather smooth change even at lower temperature than the resonant shift. We study a small system which has a staircase-like magnetization process in the ground state. At finite temperatures, the staircase-like structure is smeared out and we see a smooth curve. While even in such temperatures, we observe the plateau structure in the frequency dependence of the shift, and we found that the ESR absorption is much sensitive to the energy structure than the magnetization (Fig. 3). Thus we propose that the magnetization process of CHpC also have a staircase-like structure and the observed resonant-field dependence of the shift represents the structure. We found a peculiar density of states for the excited levels in the present model. The effect of this distribution on the line shape will be reported elsewhere[9].

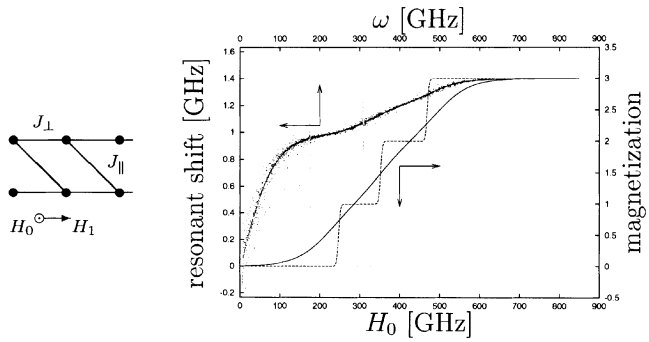

FIGURE 3: Frequency dependence of resonant shift and magnetization process in the ladder with $J_\parallel = 12.8$ K and $J_\perp = 6.4$ K. Resonant shift is plotted by dots for $T = 2.0$ K. Solid and dashed lines show magnetization process for $T = 2.0$ K and 0.1 K, respectively.

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