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Phonon Effects in the Two-Magnon Raman Scattering in Spin-Peierls Systems

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Abstract

The phonon assisted Fleury-Loudon two-magnon scattering in spin-Peierls systems is derived and its effect on the scattering processes is studied. We find that the phonon effects are important for two-magnon Raman scattering in both dimerized and uniform phases.

Keywords: Lattice dynamics, Many-body and quasiparticle theories, Infrared and Raman spectroscopy

Recently discovered spin-Peierls (SP) transition in the compound CuGeO_3 [1] revived the interest in theoretical studies of low dimensional spin systems and lattice spin coupling effects [2,3]. Special attention has been paid to the frustration effects and their consequence on the dynamical properties [2]. In fact, it was pointed out earlier by some of the present authors that there are frustrated SP fluctuations in the MX materials near the crossover region [4]. In the study of the excitations in the CuGeO_3 compound, several groups have conducted two-magnon Raman scattering experiments [5]. In addition, several theoretical studies were carried out which, however, neglected the phonon effects totally [6].

FIG. 1. Lattice distortions related to magnetic excitations in the dimerized phase. (a) ground state; (b) magnetic excited state with one spin flip. (c) Since the above state is unstable, the lattice near the flipped spin relaxes.

In the present study, we show that the phonon effects are important in the two-magnon Raman scattering. The phonon effects enter the Raman scattering process in two ways. First, there is a strong spin phonon coupling; naturally, the photon-magnon coupling has a phonon term, which has strength $\partial J/\partial u$, where u is lattice distortion. Second, without frustrated spin-spin coupling J_2 (see below), there is no two-magnon Raman scattering in the high temperature uniform phase, as the scattering term commutes with the Hamiltonian. However, since the lattice fluctua-

tions (quantum or thermal) will introduce disorder in the spin-spin coupling, there is phonon induced two-magnon Raman scattering even without J_2 .

Let us first derive the phonon-assisted two-magnon scattering coupling using Peierls-Hubbard model:

$$H = - \sum_{ij} [t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + H.c.] + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{1}{2} \sum_i K u_i^2, \quad (1)$$

where $t_{i,i+1} = t_1[1 + \frac{\lambda u_i}{2a_0}]$, $t_{i,i+2} = t_2$, and otherwise, $t_{i,j} = 0$. We have the lattice distortion $u_i = (-)^i u_0 + \hat{u}_i$ with $\hat{u}_i = [\frac{1}{2M\omega_0 N}]^{1/2} (b_i + b_i^\dagger)$. In the limit $t_i \ll U$ and half-filling, the system is in the insulating phase, the Hamiltonian (1) is equivalent to the frustrated SP Hamiltonian [7]:

$$H = \sum_i \left[J_{1,i} (\vec{S}_i \cdot \vec{S}_{i+1} - \frac{1}{4}) + J_2 (\vec{S}_i \cdot \vec{S}_{i+2} - \frac{1}{4}) + \frac{1}{2} K u_i^2 \right], \quad (2)$$

where $J_{1,i} = \frac{4t_1^2}{U} [1 + \frac{\lambda u_i}{a_0}] \equiv J_1 [1 + \frac{\lambda u_i}{a_0}]$ and $J_2 = \frac{4t_2^2}{U}$.

With the presence of a photon field $\vec{A}(q) = \vec{e}(hc^2/V\omega_q)^{1/2} (a_q + a_{-q}^\dagger)$, one gets a Peierls phase in the electron hopping term: $t_{ij} \rightarrow t_{ij} \exp(i \frac{e}{\hbar c} \int_i^j \vec{A} \cdot d\vec{l})$. After the expansion, we obtain electron-photon coupling

$$H_j = - \frac{e}{\hbar c} \sum_i \vec{j}_i^\nu(i) \cdot \vec{A}_\nu(i), \quad (3)$$

where $\vec{j}_i(i)$ is the current operator. Note that we have dropped the second order term in $\vec{A}$, which is unimportant for two-magnon Raman scattering. The Ra-

man scattering cross section is determined by the time-dependent second order perturbation, writing in the effective spin operators, the two-magnon scattering can be calculated [8] using:

$$\hat{R} = - \sum_n \frac{a_0^2}{U - \omega_n} [t_{1,n}^2 (\vec{S}_n \cdot \vec{S}_{n+1} - 1/4) + 4t_2^2 (\vec{S}_n \cdot \vec{S}_{n+2} - 1/4)]. \quad (4)$$

There are phonon assisted processes (up to 2-phonon) included here in the $t_{1,n}^2$ -terms. The derivation of Eq.(4) is following Shastry and Shraiman for a Mott-Hubbard system [8]. Here we have included the phonon-spin coupling and next-nearest-neighbor (NNN) spin-spin couplings. Usually, one would not use NNN spin scatterings but use NNN spin-spin coupling in the spin Hamiltonian. Then, we will have a natural two-magnon scattering due to NNN spin couplings. However, this is not true if the NNN two-magnon term derived here is also included. Without phonon-spin coupling, there is no two-magnon scattering even if J_2 effects are included. Here, we have neglected the high order spin scattering effects, which can contribute higher order (t/U) scatterings [8]. The most important two-magnon scattering terms come from the interchain coupling and phonon effects. The interchain coupling is neglected here and will be discussed elsewhere. The phonons can induce finite two-magnon scattering is because the phonons are three dimensional; otherwise, H_R commutes with Hamiltonian (2).

In the spin-Peierls systems, the ground state is dimerized. The magnetic excitations are dressed with finite lattice distortions, the phonon assisted scattering has important contributions. If the photon is scattered by a soliton, soliton-antisoliton pairs, etc., there is no doubt that the phonon assisted two-magnon Raman scattering is more important. Even if magnetic excitations in the *static* dimerized lattice distortion background are responsible for the two-magnon Raman scattering, we can see from Fig.(1) that a relaxation of lattice distortions is involved. Because of the relaxation of lattice involved near the flipped spin, phonon assistant two-magnon scattering is important.

At temperature above the spin-Peierls transition T_{sp} , the ground state has uniform lattice distortion. However, notice that if phonon effects are neglected, the scattering term commutes with the Hamiltonian (2); thus, there is no inelastic scattering. However, the (quantum and thermal) lattice fluctuations will induce randomness in the spin exchange couplings, which will induce two-magnon scatterings. In Fig.(2), we calculate the two-magnon Raman scattering using adiabatic approximations by assuming that the lattice fluctuations induce random spin-spin coupling in J_1 ($J_1 \rightarrow J_1 + \delta J$). This approximation has been used successfully for Peierls systems [9].

FIG. 2. Raman scattering for $N=10$ systems using Gaussian randomness of J_1 due to phonon fluctuations. (a) $J_2 = 0$; (b) $J_2/J_1 = 0.1$.

In summary, we study the phonon assisted two-magnon Raman scattering in the SP systems. We find that the phonon effects are important in both dimerized phase and high temperature uniform phase.

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