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Angaben zur Veröffentlichung / Publication details:

Büchner, B., H. Fehske, Arno P. Kampf, and G. Wellein. 1999. "Lattice dimerization in the Spin-Peierls compound CuGeO3." *Physica B: Condensed Matter* 259-261: 956–57. https://doi.org/10.1016/S0921-4526(98)00720-0.

Lattice dimerization in the spin–Peierls compound $CuGeO_3$

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Abstract

The uniaxial pressure dependence of the exchange coupling and the structural distortion in the dimerized phase of $CuGeO_3$ are analyzed. A minimum magnetic dimerization of 3% is obtained, incompatible with an adiabatic approach to the spin–Peierls transition. Exploring the properties of an Heisenberg spin chain with *dynamical* spin–phonon coupling, the dimerization dependence of the spin excitation gap is found to be in qualitative agreement with experiment.

Keywords: Spin–Peierls transition; Dynamical spin–phonon coupling; Exact diagonalization

There has been a renewed interest in the spin–Peierls (SP) phenomenon since it was recognized that the displacive SP transition in the first inorganic SP compound $CuGeO_3$ shows no phonon softening. On the contrary, the Peierls-active optical phonon modes with frequencies $\omega \approx J$ and $\omega \approx 2J$ (J is the exchange coupling between nearest neighbour (nn) Cu spins) harden by about 5% with decreasing temperature, requiring a very strong spin–phonon coupling for the SP instability to occur [1,8].

In spite of this fact, up to now most theoretical studies rely on an adiabatic approach to the phonons. The Hamiltonian commonly used to model the dimerized (D) SP phase is

$$\mathscr{H} = \sum_j J[(1 + (-1)^j\delta)\boldsymbol{S}_j \cdot \boldsymbol{S}_{j+1}] + J'\boldsymbol{S}_j \cdot \boldsymbol{S}_{j+2}, \qquad (1)$$

where δ describes the alternating pattern of weak and strong bonds due to the structural distortion and J' is the next nn coupling. From the magnetic properties of the uniform (U) phase $J \simeq 160\,\mathrm{K}$ and $\alpha = J'/J \simeq 0.35$ have been estimated for $CuGeO_3$ [2]. However, if one attempts to reproduce the observed spin gap $\Delta^{\mathrm{ST}} \simeq 2.1\,\mathrm{meV}$ from Eq. (1), a very small value of $\delta \simeq 0.012$ results.

An alternative and model-independent way to determine δ is possible by combining the amount of the structural distortion in the D-phase with the magnetoelastic coupling in the U-phase using the uniaxial pressure derivatives of the magnetic susceptibility at high temperatures as measured by magnetostriction [3]. Rather large values $\partial \ln J/\partial p_i$ of 4, -9, and -2%/GPa are obtained for pressures p_i along the a-, b-, and c-axis, respectively, whereas $\partial J'/\partial p$ is essentially zero [2]. We stress that these results do not depend crucially on the values of J and J'. The decrease of J for pressure applied parallel to the spin chains (c-axis) signals the correlation between J and the Cu–O–Cu bond angle γ [1,3,4,8].

The c-axis lattice constant depends on γ only and the Cu–O bond length d_{CO} ($c = 2d_{\mathrm{CO}}\sin(\gamma/2)$). Obviously $\partial d_{\mathrm{CO}}/\partial p_c \leqslant 0$, $\partial J/\partial d_{\mathrm{CO}} \leqslant 0$, and from Ref. [5,9] we have $\partial \ln c/\partial p_c \simeq -0.3\%$/GPa. The observed negative $\partial J/\partial p_c$ is due to a positive $\partial J/\partial\gamma$ which is the most important source of spin–phonon coupling in $CuGeO_3$ [1,8]. By accounting for the bond angle and length dependence a clear-cut lower limit $\partial \ln J/\partial\gamma \leqslant 5\%/^\circ$ is obtained for the angular dependence of J. The true value is, however, much larger (of order 10%) for two reasons: uniaxial pressure also reduces d_{CO} causing an additional positive

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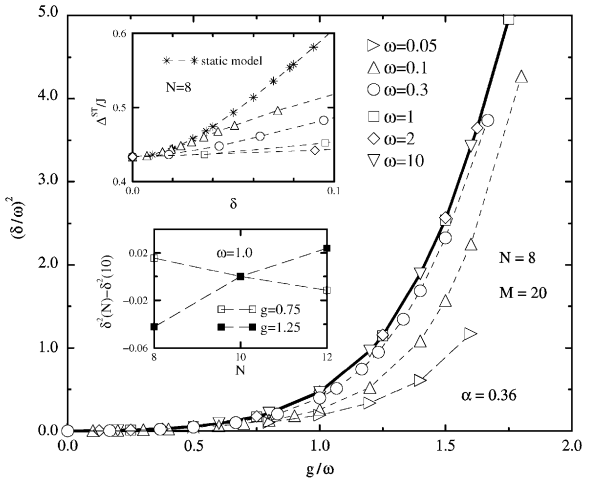

Fig. 1. Dimerization in the frustrated Heisenberg model with dynamic spin–phonon coupling. Results are presented for an eight-site chain with $M = 20$ phonons at different phonon frequencies.

pressure effect on J and the elastic reactions of the two other lattice constants also increase $\partial \ln J/\partial\gamma$.

Qualitatively the positive $\partial J/\partial p_a$ follows from $\partial \ln d_{CO}/\partial p_a \leqslant 0$ and $\partial\gamma/\partial p_a \geqslant 0$. In order to explain the very large negative pressure derivative with respect to p_b we have to take into account the hybridization between Ge and O orbitals [4]. This side group effect depends on the angle $\phi \simeq 159^\circ$ between the Ge–O bonds and the CuO_4 plaquettes [4]. The large compressibility of the b axis is related to a decrease of ϕ [1,5,9] and explains the strong reduction of J for $p \| b$; we estimate $\partial \ln J/\partial\phi \simeq 1\%/^\circ$.

In the D-phase of $CuGeO_3$ the structural dimerization is related to two independent phonon modes [1,8]. Both, γ and ϕ, alternate in the dimerized phase ($\gamma_1 - \gamma_2 = 0.83^\circ$ and $\phi_1 - \phi_2 = 1.8^\circ$), whereas d_{CO} hardly differs for neighboring exchange paths [1,8]. From the lower bounds of the coupling constants a minimum magnetic dimerization of $(J_1 - J_2)/2J \equiv \delta \geqslant 3\%$ is obtained. Thus the small value $\delta = 1.2\%$, which is necessary to reproduce the correct spin gap within the static model Eq. (1), is clearly excluded from our data analysis.

The simplest model that includes lattice dynamical effects may be obtained from Eq. (1) by replacing [6,7]

$$(-1)^i\delta \to gu_i = g(b_i^\dagger + b_i), \tag{2}$$

where g is the magneto-elastic coupling constant and the $b_i^{(\dagger)}$ are independent phonon destruction (creation) operators on each bond $(i, i+1)$. For modeling the optical phonons in $CuGeO_3$ we choose dispersionless Einstein modes with frequency ω, i.e. $\mathscr{H}_{ph} = \omega\sum_i b_i^\dagger b_i$. Recently it was shown that such a dynamical spin–phonon model describes the general features of the magnetic excitation spectrum of $CuGeO_3$ [6].

Here we focus on the behavior of the lattice dimerization which can be found from the displacement structure factor at $q = \pi$:

$$\delta^2 = \frac{g^2}{N^2}\sum_{i,j} \langle u_i u_j\rangle e^{i\pi(R_i - R_j)}. \tag{3}$$

Since in the physically most relevant *non-adiabatic* ($\omega \gtrsim J$) and *intermediate coupling* ($g \sim J$) regime the spin and phonon dynamics are intimately related, we performed a complete numerical diagonalization of the quantum phonon model (2) using a controlled phonon Hilbert space truncation [6].

Fig. 1 summarizes our main results: (i) in the non-adiabatic region the SP transition takes place provided that $(g/\omega) > (g/\omega)_c \simeq 1$, *irrespective* of the ratio ω/J. For $CuGeO_3$ with $\omega \sim J$ this implies $g > g_c \simeq J$, i.e. a strong-coupling situation. (ii) Below (above) $(g/\omega)_c$ the dimerization decreases (increases) with increasing lattice size (see lower inset), indicating that the *infinite* system (2) exhibits a true phase transition. (iii) The dynamic model (2) partially resolves the $\Delta^{ST} - \delta$ conflict we are faced within the static approach (1), because the dimerization δ *grows* with the phonon frequency at fixed Δ^{ST} (cf. upper inset). Thus matching Δ^{ST} to the neutron scattering data, a larger $\delta \simeq 5\%$ may result.

In conclusion, our findings are in qualitative agreement with experimental data on $CuGeO_3$, suggesting the necessity for a non-adiabatic spin–phonon approach to this material.

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