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Predictions for neutron scattering and photoemission experiments on $CuGeO_3$

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Applying numerical techniques to a model recently proposed for the one-dimensional spin-Peierls compound $CuGeO_3$, we calculate dynamical properties that can be directly compared with inelastic-neutron-scattering data and angle-resolved photoemission (ARPES) experiments. The momentum and energy dependence of the dynamical structure factor $S(q,\omega)$ are discussed, as well as the static structure factor $S(q)$. The latter is shown to be specially useful to estimate the strength of the spin interaction beyond nearest neighbors. The spectral function $A(q,\omega)$ calculated from the one-particle Green's function at half-filling is shown at several values of the hole-hopping amplitude t. The results have some unique features characteristic of one-dimensional systems including small weight near the Fermi energy. The presence of "shadow bands" induced by short distance antiferromagnetic correlations is predicted to appear in ARPES experiments for $CuGeO_3$ and also for Sr_2CuO_3.

The behavior of low-dimensional magnetic materials continues to attract much experimental and theoretical interest. A variety of exotic ground states can be observed in different compounds depending on anisotropies, the spin of the magnetic ions, and the coupling to the lattice degrees of freedom. Recently, $CuGeO_3$ was found to be an inorganic system that shows a spin-Peierls (SP) phase.[1] With decreasing temperature a regime described by uniform $S=1/2$ Heisenberg chains undergoes a phase transition at $T_{SP}=14$ K into a dimerized system due to the coupling of the spin-1/2 Cu^{2+} ions to the three-dimensional lattice phonons. At T_{SP} a structural transition takes place where alternate atoms are displaced in opposite directions, and an energy gap for magnetic spin-triplet excitations appears. The results of Hase, Terasaki, and Uchinokura[1] have generated much excitement, and the presence of a spin gap in $CuGeO_3$ has been confirmed by several groups.[2,3]

$CuGeO_3$ has a c-direction lattice constant of 2.941 Å, much smaller than the a and b lattice parameters. Although the c-axis lattice constant corresponds to the Cu^{2+}-Cu^{2+} distance, the actual links between coppers are provided by the edge sharing of CuO_6 octahedra. The angle defined by each Cu-O-Cu bond is close to 90°.[4] This induces an exchange coupling J_1 in the effective nearest neighbor (NN) $S=1/2$ Heisenberg model for the Cu-O chains which is much smaller than those found in the two-dimensional (2D) cuprate superconductors. The measured susceptibility, χ, above 14 K, while showing characteristics of 1D antiferromagnets, is not quantitatively reproduced by calculations corresponding to a NN $S=1/2$ Heisenberg model. Actually, Lorenzo *et al.*, based on a neutron-scattering study, suggested that a next-nearest-neighbor (NNN) coupling J_2 may arise from the Cu-O-O-Cu path.[5] Riera and Dobry[6] arrived to the same conclusion studying numerically the temperature dependence of χ. They reported a substantial ratio $J_2/J_1=0.36$. In this regime both phonons *and* frustration contribute to the opening of the spin gap. Castilla, Chakravarty, and Emery[7] also remarked the relevance of NNN terms reporting a smaller ratio $J_2/J_1=0.24$, with $J_1=150$ K, which is right below the ratio where the effects of frustration open a spin gap in the model in the absence of phonons. Actually, exact diagonalization (ED) techniques locate the critical ratio at $J_2/J_1|_c \approx 0.2412 \pm 0.0001$.[8,7] While the microscopic origin of J_2 is not quite clear, we include it in our studies simply on phenomenological grounds to model accurately $CuGeO_3$. Since some experimental evidence shows that the SP transition is exclusively driven by phonons,[3] in our study we use Castilla, Chakravarty, and Emery's estimation of J_2. However, note that working so close to $J_2/J_1|_c$ likely induces strong dimerizing fluctuations of nonphononic origin in the ground state.

Based on this discussion,[5–7] the model studied here corresponds to a Heisenberg model with NN and NNN interactions and a term that dimerizes the lattice to mimic the effects of phonons. The Hamiltonian is

$$H=J_1\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j+J_2\sum_{\langle\langle im\rangle\rangle}\mathbf{S}_i\cdot\mathbf{S}_m+\delta\sum_{\langle ij\rangle}(-1)^i\mathbf{S}_i\cdot\mathbf{S}_j, \quad (1)$$

where i,j,m denote sites of a 1D chain with N sites, $\mathbf{S}_i$ are S=1/2 spins, and $\langle ij\rangle$ ($\langle\langle im\rangle\rangle$) corresponds to NN (NNN) spins along the chain. The static properties of the special case $J_2=0$ have been widely studied in the literature,[9] while the frustrated J_1-J_2 model has also received much attention.[8] However, dynamical studies with controlled approximations have not been previously reported to the best of our knowledge, and here we provide this information using ED techniques.[10]

Inelastic-neutron-scattering experiments have been recently performed on $CuGeO_3$ single crystals.[11,12] The opening of a gap at $q=0$ was observed in the dispersion of the magnetic excitations. An ED study by Castilla, Chakravarty, and Emery showed that the inelastic-neutron-scattering (INS) triplet dispersion is reproduced using Eq. (1) if $J_2/J_1=0.24$ and $\delta=0.03J_1$. We adopt these parameters in the present paper. Thus far the *dynamical* information contained in the energy dependence of $S(q,\omega)$ has not been analyzed in the INS literature for $CuGeO_3$. The reason is that the experimental data are usually contaminated by spurious effects, and a complicated convolution between the actual

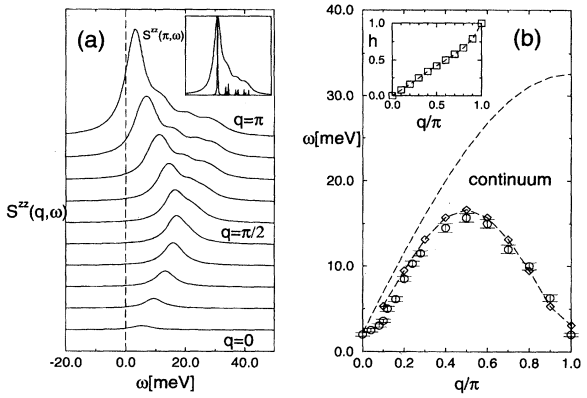

FIG. 1. (a) $S^{zz}(q,\omega)$ obtained on a chain of $N=20$ sites with a broadening $\epsilon=0.3J_1$ at several momenta. In the inset, we show $S^{zz}(\pi,\omega)$ with more resolution using $\epsilon=0.02J_1$; (b) position of the first pole in (a) for each q, defining the spin-triplet dispersion (diamonds). The circles denote results from Refs. 11 and 12. The agreement is good as remarked before in Ref. 7. The dashed line near these points guides the eye, while the dashed line at higher energies signals the position of the pole with the highest energy for each q. In the inset, the intensity h of the highest peak in (a), normalized with respect to $q=\pi$, is shown as a function of momentum, using $J_2/J_1=0.24$, $\delta/J_1=0.03$, and $J_1=150$ K.

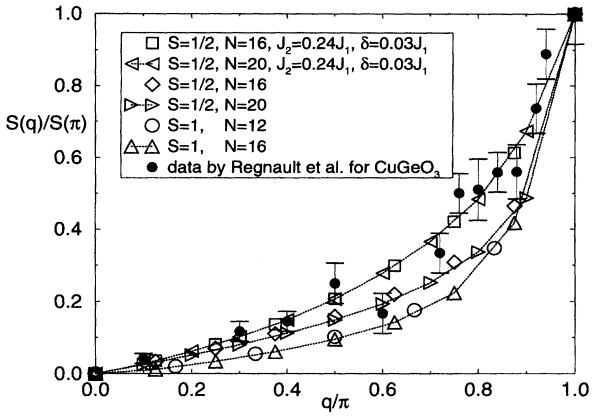

FIG. 2. $S(q)$, normalized with respect to its value at $q=\pi$, as a function of q. The convention for the symbols is shown in the inset. We present results for the model Eq. (1), for the (gapless) NN spin-1/2 Heisenberg chain, and for the spin-1 Heisenberg chain which has a disordered ground state and a finite spin gap. We also show recent experimental data by Regnault *et al.* (Ref. 17).

data and the instrumental resolution is needed to extract $S(q,\omega)$. To guide this experimental effort it is useful to have a *theoretical* prediction for $S(q,\omega)$. The purpose of this paper is to provide spin-dynamical information obtained from model Eq. (1) to compare theory with experiments in $CuGeO_3$. Here we also calculate the spectral function $A(q,\omega)$ which can be directly compared with angle-resolved photoemission spectroscopy (ARPES) experiments. In the context of high-temperature superconductors, recent ARPES data for $Sr_2CuO_2Cl_2$ at half-filling have provided important constraints on theoretical models for the cuprates.[13] A similar study in $CuGeO_3$ would likely prove as useful.

In Fig. 1(a), $S^{zz}(q,\omega)$ is shown at several momenta. The position of the dominant peak defines the energy of the first triplet excitation for each q.[14] The intensity is the highest at $q=\pi$ as expected, due to the power-law decaying antiferromagnetic (AF) spin-spin correlations in the ground state. The intensity of the dominant peak for each momentum, normalized with respect to $q=\pi$, is shown in the inset of Fig. 1(b). This ratio decays away from $q=\pi$ more slowly than in the case of an $S=1$ Heisenberg chain,[15] which has a spin gap caused by a disordered but translationally invariant ground state. In Fig. 1(b), the first peak in the spectrum is shown as a function of momentum and compared with experiments.[11,12] As previously remarked by Castilla, Chakravarty, and Emery,[7] the agreement between theory and experiment is excellent. From our numerical study we can also analyze the position of the peak with the highest energy to put bounds on the multiparticle continuum.[16] The result is shown in Fig. 1(b), which in principle could be compared directly with INS results. However, note that the rapidly decaying intensity of the spectral weight makes such comparison difficult. If the resolution of the INS experiment only allows for the observation of intensities within, e.g., 10% or 20% of the maximum at $q=\pi$, then the apparent width of the continuum would appear considerably reduced.

In Fig. 2, the static structure factor $S(q)$ $[=\int d\omega\, S(q,\omega)]$ is presented. As shown in the figure, finite-size effects are small and we consider our results representative of the bulk limit. As expected, the large AF correlations in the model cause $S(q)$ to peak at $q=\pi$, slowly decreasing away from it as the momentum is varied. The normalized results for Eq. (1), with the parameters introduced in Ref. 7, can be clearly distinguished from those obtained at $J_2=0$. Recent INS measurements of $S(q)$ by Regnault *et al.*,[17] also shown in Fig. 2, confirm the presence of important NNN couplings in $CuGeO_3$. The agreement between theoretical expectations and experimental data is excellent. Note that δ here is so small that it does not influence $S(q)$ as much as J_2, contrary to what would occur in transport calculations at zero temperature. For completeness, results for the $S=1$ chain are also shown.

We have also calculated the spectral function $A(q,\omega)$ which corresponds to the creation of holes by the removal of spins on the chains in the sudden approximation. As a Hamiltonian for this calculation we use the t-J model generalization of Eq. (1), i.e., we add a hopping term

$$H_t=-t\sum_{\langle ij\rangle\sigma}(\tilde{c}^{\dagger}_{i,\sigma}\tilde{c}_{j,\sigma}+\text{H.c.}), \tag{2}$$

where $\tilde{c}$ are hole operators and the rest of the notation is standard.[10] In the absence of experiments testing the properties of carriers in $CuGeO_3$, it is difficult to predict the value of the hopping amplitude t. Thus, in the results quoted below, three different amplitudes are used such that more information is available to compare with experiments once ARPES data become available. However, if the couplings J_1 and J_2 are caused by an exchange mechanism, it is likely that the amplitude t would be larger than both. For simplicity, we have neglected the influence of the dimerization on t, which in principle should also be modulated as the exchange

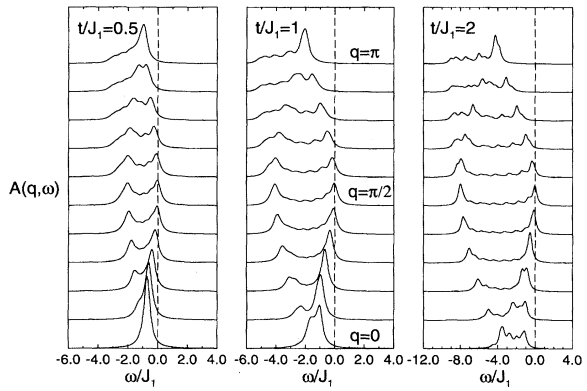

FIG. 3. Spectral function $A(q,\omega)$ on a chain of 20 sites, at several values of t/J_1. The broadening is $\epsilon=0.25J_1$, and the Fermi energy is arbitrarily located at $\omega=0$. The other couplings are as in Fig. 1.

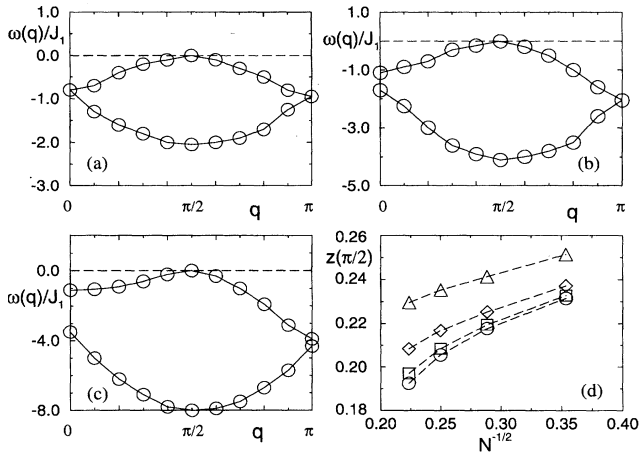

FIG. 4. Dispersion of the poles with the lowest and highest binding energy found in $A(q,\omega)$ using a 20-site chain at different q and for coupling t/J_1 equal to (a) 0.5, (b) 1.0, and (c) 2.0. The rest of the couplings are as in Fig. 1. (d) Spectral weight $Z(\pi/2)$ of the pole with the lowest binding energy appearing in our calculation at $q=\pi/2$ vs $N^{-1/2}$, where N is the number of sites. The triangles, diamonds, squares, and circles correspond to t/J_1 equal to 2.0, 8.0, 32.0, and 128.0, respectively. $Z(\pi/2)$ is normalized such that it is bounded between 0 and 1.

J_1. Since the dispersive features observed in our calculations have bandwidths of order J_1 ($\gg\delta$), the influence of δ on the ARPES data is expected to be small. Actually J_2 is more relevant than δ for ARPES, where states deep in energy are tested. This is true even for a $T=0$ calculation such as ours.

In Fig. 3, $A(q,\omega)$ is shown for different momenta and three couplings t/J_1, with J_1, J_2, and δ defined as before. A substantial width is given to the δ functions to mimic the effect of the large backgrounds appearing in ARPES data. Note the presence of a dominant peak at low binding energy which moves towards the Fermi energy (at $\omega=0$ in the figure) as the momentum is changed from $q=0$ to $q=\pi/2$ for all couplings. The lowest binding energy is precisely reached at $q=\pi/2$. Based on previous experience with the 2D copper oxide compounds,[10] this large peak may correspond to a hole immersed in a *locally* ordered AF spin background which increases its effective mass. An interesting feature of our results is the presence of substantial weight at large ω for all q and couplings. At $q=\pi/2$, the shape of $A(q,\omega)$ at the top and bottom of the band seems similar, i.e., a symmetry line seems to run through the middle of the total bandwidth. This feature also appears in the 2D t-J model but only at very small J/t.[18] This is another effect predicted by theoretical calculations that an ARPES experiment could analyze, although we are aware that in such experiments it is difficult to investigate structure at large binding energies due to the presence of core backgrounds in the data that grow with binding energy, as well as the influence of other bands not taken into account in simple one-band models like the one used in the present paper.

A curious effect obvious to the eye in Fig. 3 is the presence of a robust peak in A(q,ω) for momenta larger than $q=\pi/2$. This behavior is quite different from what is observed in normal metals, and here we argue that it is caused by the strong AF correlations in the ground state. Recently, this effect, which is usually referred to as "shadow bands," has received much attention in the context of the 2D cuprates after ARPES experiments addressed their existence in Bi2212 at optimal doping.[19] The origin of the shadow bands is simple: in the presence of AF order the unit cell doubles its size due to the nonzero staggered order parameter, reducing the Brillouin zone in half. This induces an extra symmetry in the spectrum since now, e.g., on a square lattice momenta along the diagonal from (0,0) to $(\pi/2,\pi/2)$ become equivalent to those from $(\pi/2,\pi/2)$ to (π,π).[20] ARPES techniques[13] have shown that antiferromagnetically induced bands can be clearly seen in the insulator $Sr_2CuO_2Cl_2$. If AF order is replaced by a finite but robust AF correlation, the effects of shadow bands are expected to be still visible in ARPES data. The present calculations have shown that shadow bands can be robust *even* in 1D systems, where we are used to the concept that there are no ordered phases at zero temperature. Our prediction can be extended to materials like Sr_2CuO_3, where the NN Heisenberg model accurately describes its properties.[21] The existence of shadow bands in 1D systems should not be surprising since the hole dispersion tested in ARPES experiments is dominated by *short* distance correlations.

In Figs. 4(a)–4(c), the dispersion of the large features (peaks) observed in Fig. 3 are plotted, both at low and high binding energy. The approximate symmetry of the spectrum with respect to the middle of the total bandwidth is clear. In Fig. 4(d) the weight Z of the lowest energy pole appearing in $A(\pi/2,\omega)$ is shown as a function of $N^{-1/2}$ as presented in previous studies for 1D systems.[22] The finite size of the clusters does not allow us to extrapolate Z to the bulk. However, the shape of the curve in Fig. 4(d) leads to the speculation that Z may remain finite as the system size increases, contrary to what occurs for the pure t-J model in 1D.[22] It has recently been argued[23] that in the present calculation at *half-filling* with a spin gap in the system, the charge degrees of freedom tested in $A(q,\omega)$ indeed should have a one-particle Green's function with a finite quasiparticle peak (see also Ref. 22). The strong correlations may reduce Z to just a fraction of its maximum value 1 [as shown in Fig. 4(d)], but

ARPES experiments should still observe a finite weight peak in their data. However, as the density of carriers becomes finite, the holes will tend to form a Luttinger liquid with $Z=0$. The crossover between these two regimes is interesting both theoretically and experimentally if $CuGeO_3$ is doped away from half-filling. These speculations deserve further study.

Summarizing, here we presented dynamical properties of a model for $CuGeO_3$. Theoretical predictions are made that can be directly compared with experiments. In the context of INS, we have calculated the intensity and momentum dependence of $S(q,\omega)$, as well as the integrated static structure factor $S(q)$. This quantity is particularly useful to estimate the strength of J_2, and here we confirmed that J_2 is nonzero as predicted in Refs. 5–7. For ARPES experiments at half-filling, the dispersion of quasiparticlelike features was provided. The shape of the spectrum is somewhat unusual with substantial weight concentrated at high energies. We also predict the presence of "shadow bands" in ARPES data induced by AF correlations in the ground state of the 1D compounds $CuGeO_3$ and Sr_2CuO_3.

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