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Interchain coupling effects and solitons in $CuGeO_3$

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The effects of interchain coupling on solitons and soliton lattice structures in $CuGeO_3$ are explored. It is shown that interchain coupling substantially increases the soliton width and changes the soliton lattice structures in the incommensurate phase. It is proposed that the experimentally observed large soliton width in $CuGeO_3$ is mainly due to interchain coupling effects. [S0163-1829(97)51022-8]

The inorganic spin-Peierls (SP) system, $CuGeO_3$, has attracted much attention[1] recently. Pure $CuGeO_3$ has a SP transition at $T_{sp} \simeq 14.3$ K. Below T_{sp}, the system is in a dimerized spin singlet state, and the gap to spin triplet excitations is $\Delta \sim 24.5$ K.[2,3] It has been argued[4,5] that the spin susceptibility measurements in this system can be understood only if the nearest-neighbor Heisenberg Hamiltonian is augmented by a substantial next-nearest-neighbor (NNN) coupling, $J_2 \sim 0.2J$, where J is the nearest-neighbor coupling. The NNN coupling has been shown to be important in other experiments[6] as well. Recently, the soliton lattice structure in $CuGeO_3$ was studied using neutron and NMR measurements.[7,8] An important observation is that the soliton width $\xi \sim 13.6a$, where a is the lattice spacing, is much larger than the theoretical prediction[9] for the unfrustrated SP chain, $\xi_0 = Ja\pi/(2\Delta) \sim 8a$.

The soliton width in $CuGeO_3$ can be changed by interchain coupling and frustration J_2. Without interchain coupling, the soliton width is generally $\xi \propto \overline{v}/\Delta$, where $\overline{v}$ is the spin-wave velocity and Δ is the *triplet* gap in the SP chain. From the nonlinear sigma model (Ref. 10), $\overline{v} = \overline{v}_0\sqrt{1-4J_2/J_1}$, and the dimensionless coupling constant (Ref. 11) $g_s(J_2) = g_s(0)/\sqrt{1-4J_2/J_1}$. So, the frustration will decrease the soliton width. One can show that the soliton width $\xi = Ja\overline{v}/\Delta$. The triplet gap Δ will depend on both SP coupling and frustration. However, to compare with experiments, the triplet gap $\Delta \sim 24.5$ K, while $\overline{v}$ decreases with increase of frustration, so the frustration will make the soliton width in a $CuGeO_3$ chain smaller than $\sim 8a$. Thus, the observed soliton width $\xi/\xi_0 \sim 1.7$ is not an effect due to the frustration J_2 in $CuGeO_3$.

It is known that the interchain coupling in $CuGeO_3$ is not small (Ref. 2): $J_c \simeq 120$ K, $J_b \simeq 0.1 J_c$, and $J_a \simeq -0.01 J_c$. The phase diagram of the unfrustrated SP system in the presence of interchain coupling was studied by Inagaki and Fukuyama.[12] In the present paper we calculate the effect of interchain coupling on soliton width. Although the singlet SP ground state is destroyed by large interchain coupling, in the strongly dimerized regime weak interchain coupling cannot affect the spin singlet ground state. Similarly, weak interchain coupling has very little effect on the susceptibility above T_{sp}. From mean-field theory, it can be shown that the effects are typically of the order of 3%.

The low-energy excitations involving broken spin singlets are more sensitive to interchain coupling. Once a singlet pair is broken in one chain, it is energetically less costly to break another pair in a neighboring chain due to the interchain coupling. Thus, the interchain coupling will affect the low-energy excitations in the dimerized phase. For the same reason, the interchain coupling can lead to an increased soliton width: the local magnetization of a soliton in SP chain is $M(x) \sim \cosh^{-1}(x/\xi)$ (cf. below), and the gain in the interchain coupling energy increases with the increased soliton width ξ, hence an increased soliton width.

In this paper, the effects due to the interchain coupling are studied using bosonization. We consider the Hamiltonian

$$\mathcal{H} = \sum_{i,j} [J(1+\beta u_{i,j})\vec{S}_{i,j}\cdot\vec{S}_{i+1,j} + \alpha_0 J \vec{S}_{i,j}\cdot\vec{S}_{i+2,j}] + \gamma J \sum_{i,j,\mu} \vec{S}_{i,j}\cdot\vec{S}_{i,j+\mu} + \frac{K}{2}\sum_{i,j} u_{i,j}^2, \tag{1}$$

where $\vec{S}_{i,j}$ ($u_{i,j}$) is the spin operator (lattice distortion) of site i in chain j, and K is the spring constant. β is the spin-lattice coupling constant. $\alpha_0 = J_2/J$ is the strength of frustration and $\gamma = J_\perp/J$ is the interchain coupling constant. Note that we have used the adiabatic approximation for the phonons.

The interchain coupling is treated in the mean-field approximation (Refs. 12 and 13): $\vec{S}_{i,j}\cdot\vec{S}_{i,j+\mu} = S^z_{i,j}\langle S^z_{i,j+\mu}\rangle$. Thus, the Hamiltonian is transformed into a single chain model and the interchain coupling term transforms into $-\Sigma_i \gamma J \widetilde{Z} h_i S^z_i + \frac{1}{2}\gamma J Z \Sigma_i h_i^2$, where $h_i = \langle S^z_{i,j+\mu}\rangle = -(\widetilde{Z}/Z)\langle S^z_{i,j}\rangle$ is the effective field. We have introduced the notion of an effective coordination number $\widetilde{Z}$ to account for $\langle S^z_{i,j+\mu}\rangle \neq -\langle S^z_{i,j}\rangle$. In the soliton lattice phase, $\widetilde{Z}$ is site dependent. The effective Hamiltonian is then transformed to a fermionic Hamiltonian using the Jordan-Wigner transformation, and the fermionic model is bosonized. The resulting continuum Hamiltonian is[14]

$$\frac{\mathcal{H}}{Ja}=\frac{\bar{v}}{2\pi K_\rho}\int dx[K_\rho^2(\partial_0\phi)^2+(\partial_x\phi)^2]+\int dx\frac{Ku^2(x)}{2Ja^2}-\frac{g_3}{2a^2}\int dx\cos(4\phi)-\frac{\beta}{a^2}\int dx u(x)\sin(2\phi)-\frac{\gamma\tilde{Z}}{a^2}\int dx h(x)\cos(2\phi)+\int dx\frac{\gamma Z}{2a^2}h^2(x), \tag{2}$$

where a is the short distance cutoff, $u(x)=(-1)^i u_i/a$ and $h(x)=(-1)^i h_i/a$ are slowly varying variables. The bare value of the Umklapp term is $g_3=1-3\alpha_0$ and we shall assume that the renormalized g_3 has approximately the same α_0 dependence.

Here $\bar{v}$ is the spin-wave velocity and K_ρ is a critical exponent. For the unfrustrated antiferromagnetic (AFM) Heisenberg spin chain, the following renormalized values should be used (Ref. 15): $\bar{v}=\pi/2$ and $K_\rho=1/2$. However, the values of $\bar{v}$ and K_ρ in the frustrated Heisenberg model is dependent on α_0. With increase of frustration, the spin-wave velocity decreases. In the nonlinear sigma model (Ref. 10), $\bar{v}=v_0\sqrt{1-4\alpha_0}$ for small α_0. In a recent numerical calculation, it is shown that the variation of *renormalized* $\bar{v}$ with α_0 is approximately (Ref. 16) $\rho_v=2\bar{v}/\pi\simeq(1-1.12\alpha_0)$. Although we do not know K_ρ at finite α_0 precisely, it is known[17] that $K_\rho\geqslant 1/2$ at small frustration $\alpha_0<\alpha_0^c\sim 0.3$. Define $\rho_K=2K_\rho-1$, then $\rho_K\ll 1$ and $\rho_v\lesssim 1$.

For the ground state, $u(x)=u_0$, $h(x)=h_0$, and $\tilde{Z}=Z$. If there is no Umklapp interaction g_3, the model (2) is exactly soluble by Bethe ansatz.[18] Due to the g_3 term, an approximation has to be employed. We use the self-consistent Gaussian approximation, which was previously shown to be reliable[9] for the SP chains. To this end, we write $\phi(x)=\phi_s(x)+\tilde{\phi}(x)$, where ϕ_s is the semiclassical solution and $\tilde{\phi}$ is the fluctuation around ϕ_s. We retain $\tilde{\phi}$ to quadratic order in the action. The first-order term vanishes at the saddle point. Then,

$$\partial_x^2(4\phi_s)+\frac{1}{\xi^2}\sin(4\phi_s)=0, \tag{3}$$

where

$$\frac{1}{\xi^2}=\frac{1}{\xi_0^2}-\frac{4\gamma\tilde{Z}^2}{a^2 Z}\sigma^2, \tag{4}$$

and

$$\frac{1}{\xi_0^2}=\frac{4\beta^2 J}{a^2 K}\sigma^2-\frac{8g_3}{a^2}\sigma^4. \tag{5}$$

The quantity σ is given by

$$\sigma=e^{-2\langle\tilde{\phi}^2\rangle}. \tag{6}$$

σ describes the renormalization of g_1 and g_3 due to fluctuations of $\tilde{\phi}$ around ϕ_s. Note the derivation is appropriate for systems with a finite gap, otherwise, σ has *infrared* divergence. In deriving these equations, we have used the self-consistent equations

$$u(x)=(\beta J/K)\sigma\sin(2\phi_s), \tag{7}$$

$$h(x)=(\tilde{Z}/Z)\sigma\cos(2\phi_s). \tag{8}$$

The ‘‘uniform’’ ground state [i.e. $u_i=(-1)^i u_0 a$ and $h_i=(-1)^i h_0 a$] calculated from Eq. (3) corresponds to $\sin(4\phi_s)=0$. Therefore, either $\phi_s=0$ or $\pi/4$. When $\phi_s=0$, $u(x)=0$, $h_0\neq 0$, the system has long-range AFM order; when $\phi_s=\pi/4$, $h(x)=0$, $u_0\neq 0$, the system is dimerized. The spin-lattice coupling and interchain coupling strengths dictate the solution that has the lowest energy. It is clear that on symmetry grounds these two homogeneous solutions are mutually exclusive.

The calculation of ground-state energy and excitation spectrum in the self-consistent Gaussian approximation is straightforward and can be found in Ref. 9, so we will just state the results. The excitation spectrum corresponding to uniform ϕ_s is

$$\omega_q=Ja\bar{v}\sqrt{q^2+q_0^2}\,. \tag{9}$$

In the dimerized phase $\phi_s=\pi/4$ and $\phi_0=0$, and it can be easily shown that $q_0=1/\xi_0$. From Eq. (9), the spin triplet excitation gap is $\Delta=Ja\bar{v}q_0$. The scaling of the spin triplet excitation gap is

$$\left(\frac{\Delta}{2\pi J\bar{v}}\right)^{1-\rho_K}\propto\delta^{(2-2\rho_K)/(3-\rho_K)}, \tag{10}$$

where $\delta=\beta u$ is the bond alternation induced by lattice dimerization. For the unfrustrated SP chain, $K_\rho=1/2$, $\rho_K=0$, and we obtain the correct scaling relation[19] $\Delta\propto\delta^{2/3}$.

Comparing the ground-state energies between dimerized phase ($\phi_s=\pi/4$) and AFM phase ($\phi_s=0$), we get, for $\rho_K\ll 1$, that the crossover from $\phi_s=0$ to $\phi_s=\pi/4$ is determined by

$$C_K\equiv\frac{J\gamma Z}{\Delta\rho_v}\left(\frac{\Delta}{\pi^2 J\rho_v}\right)^{\rho_K}=1. \tag{11}$$

If $C_K<1$ then $\phi_s=\pi/4$, $\delta=\beta u$, $h=0$, and the system is dimerized; otherwise, $\delta=\gamma Zh$, $u=0$, and the ground state has long-range AFM order. A similar critical value of the interchain coupling for the unfrustrated SP model was derived by Inagaki and Fukuyama.[12] Using $\Delta\simeq 24.5$ K and $J\simeq 120$ K, we find that $C_K\sim 0.98\rho_v(0.021/\rho_v)^{\rho_K}<1$, which is consistent with the fact that $CuGeO_3$ is dimerized at zero temperature. Since ρ_v (ρ_K), which is less (greater) than 1(0), decreases (increases) with increase of frustration, the frustration pushes the SP-Néel transition to stronger interchain coupling. If $\rho_v\lesssim 1$ and $\rho_K\ll 1$, the interchain coupling is close to the critical value for the long-range AFM order to be realized. This is probably significant, as materials with only 3% Zn exhibit long-range AFM order.

Consider the nonuniform solutions of Eq. (3), which correspond to the solitonic excitations of the dimerized phase.

In zero magnetic field, the soliton solution has lower energy[9] than the spin-triplet excitation with the gap Δ. In this case, we can continue to assume that $\tilde{Z}/Z$ is a constant, and the soliton solution from Eq. (3) is

$$u(x) \propto \sin(2\phi_s) = \pm \tanh\frac{x}{\xi}, \tag{12}$$

with the half-width

$$\xi = \frac{Ja\overline{v}}{\Delta} \Big/ \sqrt{1-(\tilde{Z}/Z)^2 C_K}. \tag{13}$$

The corresponding magnetization, $M(x)$, is

$$M(x) = \cosh^{-1}(x/\xi)/(2\pi\xi) \tag{14}$$

and the staggered magnetization, $N(x)$, is

$$N(x) = (-)^x \sqrt{\frac{q_0 a}{2\pi}} \cosh^{-1}(x/\xi). \tag{15}$$

From Eq. (13), we can see that the soliton width is increased by interchain coupling, while the effects of frustration $\alpha_0 = J_2/J$ appear only in Δ and $\overline{v}$.

Without interchain coupling, the soliton width is $\xi_0 = Ja\pi\rho_v/(2\Delta) \lesssim 8a$ for $CuGeO_3$. With $\tilde{Z}=Z$, $\langle S^z_{i,j+\mu}\rangle = -\langle S^z_{i,j}\rangle$, the soliton excitation described by Eq. (12) is an array of antiferromagnetically coupled solitons in a direction perpendicular to the SP chains. As discussed in the introduction, the spins are antiferromagnetically ordered inside the soliton, therefore increased soliton size will lead to a gain in the interchain coupling energy. This is the physical origin of the increased soliton width in the presence of interchain coupling. When the interchain coupling is so strong that the soliton width diverges, i.e., $C_K = 1$, the system crosses over to the long-range antiferromagnetically ordered phase. This criterion $C_K = 1$, derived from consideration of the soliton size, is the same as that derived from Eq. (11) for small ρ_K.

If the solitons are excited locally without forming a regular array, $\langle S^z_{i,j+\mu}\rangle \neq -\langle S^z_{i,j}\rangle$, and $\tilde{Z}/Z$ is smaller than unity. The soliton size will still be increased with the increased interchain coupling γ, and AFM domains will form and increase in size. We propose that for the solitonic excitations due to magnetic or nonmagnetic impurities similar AFM domains will form. The percolation of these AFM domains must be relevant to impurity induced AFM ordering in the doped $CuGeO_3$ systems.[20,21]

In a high magnetic field, there will be a transition from the zero-field commensurate dimerized phase to the incommensurate soliton lattice phase.[22] The ground state of this soliton lattice phase corresponds to the self-consistent periodic solutions of the coupled nonlinear equations similar to Eq. (3). Here, we consider only the qualitative aspects of the effect of the interchain coupling in the soliton lattice phase. To this end, we assume a constant ξ in Eq. (3) for each chain. The effect of the interchain coupling is to renormalize ξ, as in Eq. (13). The periodic solution of Eq. (3) is

$$u(x) \propto \mathrm{sn}\left(\frac{x}{k\xi}, k\right), \tag{16}$$

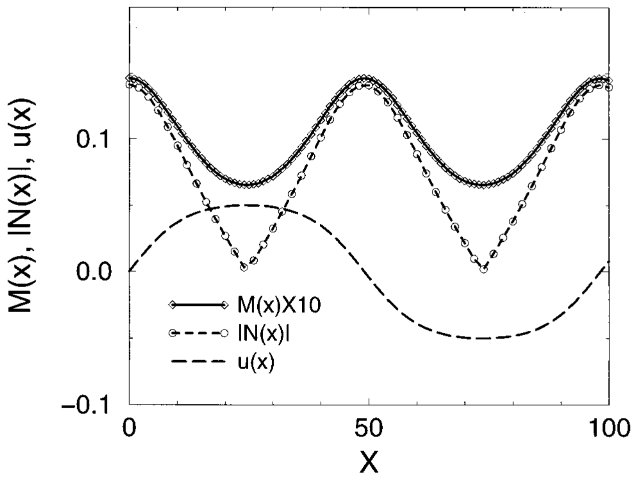

FIG. 1. The lattice distortion and magnetizations in the soliton lattice phase with $\xi = 13.6$, $k = 0.8$. The scale of $u(x)$ is arbitrary.

$$M(x) = \frac{1}{2\pi k\xi} \mathrm{dn}\left(\frac{x}{k\xi}, k\right), \tag{17}$$

$$N(x) = (-)^x \sqrt{\frac{q_0 a}{2\pi}} \mathrm{cn}\left(\frac{x}{k\xi}, k\right), \tag{18}$$

where ξ is the soliton width defined in Eq. (13), and sn, dn, and cn are the Jacobi elliptic functions.[23] The intersoliton distance is $2K(k)k\xi$ [see Fig. 1], where the modulus k of the elliptic integral K is related to the total magnetization induced by the external magnetic field that can be derived from the minimization of the energy.[22] These equations are the same as those of the soliton lattice without interchain coupling.[24] Close to the commensurate-incommensurate transition, $M(x) \sim 0 \ll |N(x)|$, we can assume that these equations still approximately describe the soliton lattice in the presence of interchain coupling: each chain has a magnetization $M(x)$, and the neighboring chains have staggered magnetizations $N(x)$ and $-N(x)$. The effective coordination number $\tilde{Z}/Z$ is smaller than unity. Thus, $\tilde{Z}/Z = \text{constant}$ is a reasonable approximation close to the commensurate-incommensurate phase transition, where most of the experiments are carried out.

In the experiment of Kiryukhin *et al.*,[7] the value of ξ is measured in the soliton lattice phase. The measured value of ξ is $\xi = (13.6 \pm 0.3)a$, which is substantially larger than the value $\xi_0 \sim 8a$ without interchain coupling. If we use an average $\tilde{Z}/Z$, from $\xi = (13.6 \pm 0.3)a$ we estimate $\tilde{Z}/Z \sim 0.8$ at $\alpha_0 = 0$. With finite α_0, the estimated $\tilde{Z}/Z$ will be larger. In these experiments the intersoliton distance is 40−70a, so $k \sim 0.7-0.9$. The magnetization $M(x)$ and staggered magnetization $N(x)$ are shown in Fig. 1 for $k = 0.8$. The ratio $\langle M(x)\rangle/\sqrt{\langle N^2(x)\rangle} \sim 0.1$, which is the right order of magnitude to give effective (Ref. 25) $\tilde{Z}/Z \sim 0.8-1.0$. In the NMR experiments,[8] the maximal value of the effective local spin is measured to be $S_{max} \sim 0.065$, which is smaller than the value corresponding to a single chain given by $\sqrt{\Delta/(J\pi^2\rho_v)} \gtrsim 0.14$. However, because the solitons in the neighboring chains are antiferromagnetically coupled, the effective local magnetization at each site is reduced by the interchain effects; a reduction factor of the order of 2–3 of $S_{\max}$ is quite reasonable.

In conclusion, we have studied the combined effects of the frustration and interchain coupling on solitons and soliton-lattice excitations in $CuGeO_3$. We find that the interchain coupling can substantially increase the soliton size, while the NNN frustration will decrease it. When the interchain coupling strength is close to the SP-Néel transition, the size of the soliton diverges. The analysis of the soliton structure presented here is consistent with experimental observations.[7,8]

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[24] Note that our expressions of soliton-lattice solutions for $M(x)$ and $N(x)$ are slightly different from those of Fujita and Machida (Ref. 22), but the nature of these solutions are the same, as shown in Fig. 1.

[25] The relation between $\langle M(x)\rangle/\sqrt{\langle N^2(x)\rangle}$ and $\widetilde{Z}/Z$ is subtle, since $\langle S^z_{i,j+\mu}\rangle/\langle S^z_{i,j}\rangle$ oscillates between $1-\epsilon \sim 1+\epsilon$.