Dilute quasi-1D antiferromagnets

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Even a small number of defects which break magnetic bonds along chains have a substantial effect on the properties of spin waves and the transverse susceptibility in quasi-1D antiferromagnets. The behavior of the susceptibility and of the spin-wave velocity on the defect concentration x has been found in the region $1 \gg x \gg (J_\perp / J_\parallel)^{1/2}$, where a perturbation theory in x is not applicable.

There is a well-developed theory for 3D Heisenberg antiferromagnets with a few exchange defects.[1] Impurities cause a damping of spin waves and slight renormalizations of the velocity of these waves and also of the Néel temperature T_N.

The situation should be different in quasi-1D systems, in which the exchange interaction along the chains, $J_\parallel$, is considerably stronger than the interaction across

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the chains, $J_\perp$. By breaking the chains, nonmagnetic defects reduce T_N sharply (this effect has been observed[2] in the alloys $K_2Fe_{1-x}Ga_xF_5$) and severely obstruct the propagation of spin waves, even if the defect concentration is low. We will show below that such defects also have a very strong effect on the transverse susceptibility. Let us assume for simplicity that the defects are ruptured bonds along a chain.[1)] The Hamiltonian of a two-sublattice antiferromagnet, in which the bond between spins $\mathbf{S}_m$ and $\mathbf{S}_n$ is broken, is

$$\mathcal{H} = \sum_{i,j} J_{ij}(\mathbf{S}_{1i}\mathbf{S}_{2j}) - \Delta \sum_{ip} (S_{ip}^z)^2 - \mathbf{H} \sum_{ip} \mathbf{S}_{ip} - J_\parallel(\mathbf{S}_m\mathbf{S}_n), \tag{1}$$

where $p = 1,2$ specifies the sublattice, $\Delta > 0$, H is the external magnetic field, and $J_\parallel$ is the exchange energy of nearest neighbors along a chain. At low temperatures, $T \ll T_N \propto (J_\parallel J_\perp)^{1/2}$, in lowest order in the concentration, we find the following expression for the transverse magnetic susceptibility:

$$\chi(\omega, \mathbf{q}) = 2S^2[J(0) + 2\Delta - J(\mathbf{q}) - xJ_\parallel(1 - \cos q_\parallel a)\Lambda^{-1}(\omega)]$$

$$\times [\omega^2 - \omega_{\mathbf{q}}^2 + x\Sigma(\omega, \mathbf{q})]^{-1}. \tag{2}$$

Here the eigenenergy part is

$$\Sigma(\omega, \mathbf{q}) = 2\Lambda^{-1}(\omega)J_\parallel S^2(J(0) + 2\Delta - J(\mathbf{q})\cos q_\parallel a), \tag{3}$$

the function $\Lambda(\omega)$ is

$$\Lambda(\omega) = 1 + 2J_\parallel S^2 \sum_{\mathbf{q}} [J(0) + 2\Delta - J(\mathbf{q})\cos q_\parallel a](\omega^2 - \omega_{\mathbf{q}}^2)^{-1}, \tag{4}$$

$q_\parallel$ is the momentum component along the chain, $J(\mathbf{q})$ is the Fourier transform of the exchange energy $J(\mathbf{r})$, and the spin-wave spectrum of an ideal crystal is

$$\omega_{\mathbf{q}}^2 = S^2[(J(0) + \Delta)^2 - J^2(\mathbf{q})]. \tag{5}$$

Under the condition $\omega \ll S(J_\perp(0)J_\parallel(0))$; the frequency on the right side of (4) can be set equal to zero. If $\Delta = 0$, we find

$$\Lambda(0) = \alpha(J_\perp(0)/8J_\parallel(0))^{1/2} \ll 1, \qquad \alpha = \sum_{\mathbf{q}_\perp} [1 - \frac{J_\perp(\mathbf{q})}{J_\perp(0)}]^{1/2} \approx 1. \tag{6}$$

As a result, we find the following expression for the correction to $c(\varphi)$, the velocity of spin waves which are propagating at an angle φ from the chain axis:

$$c_0(\varphi) - c(\varphi) = x\alpha^{-1}(\frac{8J_\parallel(0)}{J_\perp(0)})^{1/2}(c_{0\parallel}\cos^2\varphi + \frac{1}{2}c_{0\perp}\sin^2\varphi), \tag{7}$$

where $c_0(\varphi)$ is the velocity in an ideal crystal, $c_{0\parallel} = c_0(0) \sim J_\parallel$, and $c_{0\perp} = c_0(\pi/2) \sim (J_\parallel J_\perp)^{1/2}$. It can be seen that the parameter of the perturbation theory is $x(J_\parallel/J_\perp)^{1/2}$; i.e., the spectrum is greatly modified even at small values of x if $x \gtrsim x_0 = (J_\parallel/J_\perp)^{1/2}$.

The damping of the spin waves is

$$\gamma(\omega, \mathbf{q}) = \frac{x}{2\omega_{\mathbf{q}}} \operatorname{Im} \Sigma(\omega, \mathbf{q}) \approx \frac{\omega^3}{J_\perp^2 J_\parallel} \frac{c_{0\parallel}^2 q_\parallel^2 + c_{0\perp}^2 q_\perp^2}{\omega(\mathbf{q})} . \tag{8}$$

Under the conditions $\omega \ll S(J_\parallel/J_\perp)^{1/2}$ and $x < (J_\parallel/J_\perp)^{1/2}$, such that this expression holds, the damping satisfies $\gamma(\omega,\mathbf{q}) \ll \omega_{\mathbf{q}}$. It follows from (2)–(6) that the uniform susceptibility is

$$\chi = \frac{1}{J_\parallel(0)} (1 + \frac{2^{1/2} x}{x_0}). \tag{9}$$

If $\omega \gg S(J_\parallel/J_\perp)^{1/2} \approx \omega_{max}(\mathbf{q}_\perp)$, then $\Lambda(\omega) = i\omega/4J_\parallel(0)$, and the frequency of the spin waves is

$$\omega = \omega(q_\parallel) + i2xJ_\parallel(0) . \tag{10}$$

When the perturbation wavelength $\lambda \approx q_\parallel^{-1}$ is shorter than the average segment length x^{-1}, the damping is slight. In other words, well-defined spin excitations exist within a segment.

If $\Delta \neq 0$ but $\Delta \ll J_{\perp S}$, the defects shrink the gap ω_0 in the spectrum by an amount $\approx \omega_0(x/x_0)$. The most important effect of the anisotropy is that the damping becomes substantial at small values of q:

$$\frac{\operatorname{Im}\omega}{\omega - \omega_0} \approx x (\frac{\omega_0}{\omega - \omega_0})^{1/2} \frac{\Delta^{3/2}}{J_\perp J_\parallel^{1/2}} . \tag{11}$$

In other words, the hydrodynamic description of the spin waves becomes inapplicable at frequencies ω close to ω_0. This result does not depend on the relation between $J_\parallel/J_\perp$, because at $\Delta \neq 0$ the equations of motion of the transverse spin components do not have solutions which correspond to a uniform flip of the sublattice spins.

We turn now to the concentration region $1 \gg x \gg x_0$, in which the perturbation theory is not valid. The ruptured bonds break the chains up into segments which contain either an even number or an odd number of spins. We assume that only nearest neighbors interact along a chain, and we treat the interaction between chains in the molecular field approximation. We ignore the magnetic anisotropy. The energy of a segment containing n spins can then be written.

$$E = -J_\parallel S^2 \sum_{\substack{i, \\ j=i\pm 1}} \cos(\theta_i + \theta_j) - S^2 J_\perp(0) \sum_i \cos(\theta_i + \theta) - HS \sum_i \sin\theta_i , \tag{12}$$

where θ_i determines the direction of spin i, and θ determines the average direction of the spins in a sublattice (Fig. 1).

Minimizing E with respect to the angles θ_i, we find the following system of equations for determining θ_i under the condition $H/J_\parallel$, when all the angles are small:

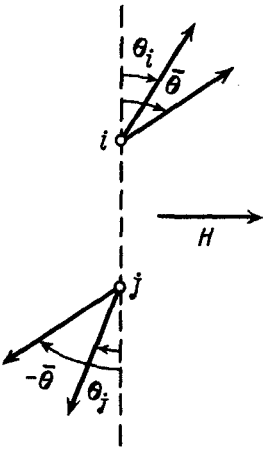

FIG. 1. The orientation of spins $\mathbf{S}_{1i}$ and $\mathbf{S}_{2j}$ belonging to the same segment. Here $\theta_i > 0$, $\theta_j < 0$, H is the external magnetic field, and θ determines the orientation of the magnetic moment of the sublattice.

$$\theta_1 + \theta_2 + h\theta_1 = \tilde{H}, \quad \theta_n + \theta_{n-1} + h\theta_n = \tilde{H} \quad 2\theta_i + \theta_{i+1} + h\theta_i = H, \quad i = 2, 3, \ldots n-1, \tag{13}$$

where $h = J_\perp(0)/J_\parallel$, and $\tilde{H} = H/J_\parallel - h\theta$. Solving these equations, we find

$$\theta_1 = \theta_n = \tilde{H}/hn \text{ for } n = 2k+1, \text{ where } k \text{ is an integer, and}$$
$$\theta_i = 0 \text{ for } n = 2k. \tag{14}$$

Taking an average of θ_i with the nearest-neighbor distribution function $P(x) = x\exp(-nx)$, we find $\theta = xH/J_\perp(0)\mathrm{S})\ln x^{-1}$, so the susceptibility per spin is

$$\chi = \frac{xS}{H} \sum_{i=1}^{n} \theta_i = J_\perp^{-1}(0)x^2 \ln x^{-1}. \tag{15}$$

The velocities of the spin waves can be found with the help of the Harris–Kirkpatrick relation,[4]

$$c_{\parallel,\perp} \propto (\sigma_{\parallel,\perp}/\chi)^{1/2}, \tag{16}$$

where $\sigma_{\parallel,\perp}$ are the conductivities of the resistance network with exchange energies J_{ij} between nodes i and j. According to Refs. 5 and 6, we have $\sigma_\parallel \propto J_\perp/x^2$, $\sigma_\perp \propto J_1$ in quasi-1D conductor. We thus have

$$c_\parallel \sim \frac{J_\perp(0)}{x^2}\left(\ln \frac{1}{x}\right)^{-1/2}, \quad c_\perp \sim \frac{J_\perp}{x}\left(\ln \frac{1}{x}\right)^{-1/2}. \tag{17}$$

As x increases, the spectrum becomes more nearly isotropic.

We have ignored the interactions between neighbors other than the nearest neighbors along a chain. It can be shown that this simplification is legitimate if the corresponding interaction satisfies $J' \ll (J_\parallel/J_\perp)^{1/2}$.

[1)] The properties of a 2D antiferromagnet with ruptured bonds were studied in Ref. 3.

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