Comment on "Low-Temperature Behavior of Two-Dimensional Quantum Antiferromagnets"

In a recent Letter[1] Chakravarty, Halperin, and Nelson have shown that the long correlation length recently observed in the paramagnetic phase of the quasi-two-dimensional antiferromagnet La_2CuO_4 by Shirane *et al.*[2] can be quantitatively explained by a quasi-two-dimensional quantum Heisenberg model with an in-plane coupling chosen within the range of values for which the ground state of an isolated layer is ordered antiferromagnetically. In this Comment I want to point out that while the model is able to account accurately for the spatial extent of the instantaneous magnetic correlations, its predictions for the spin dynamics seem to be in disagreement with experiment. In the low-frequency limit ($\omega \ll kT$) the neutron-scattering cross section is proportional to the one-sided Fourier transform of the relaxation function $R(\mathbf{q},t)$ which, in the mode-mode coupling approximation, is the solution of[3]

$$\frac{\partial}{\partial t}R(\mathbf{q},t) = -\frac{kT}{\chi(\mathbf{q})}\frac{1}{N}\sum_k \frac{[\chi(\mathbf{k})-\chi(\mathbf{k}-\mathbf{q})]^2}{\chi(\mathbf{k})\chi(\mathbf{k}-\mathbf{q})}\int_0^t d\tau R(\mathbf{k},\tau)R(\mathbf{k}-\mathbf{q},\tau)R(\mathbf{q},t-\tau), \tag{1}$$

where $\chi(\mathbf{k})$ is the wave-vector-dependent static susceptibility and N is the number of spins. The mode-coupling vertex is determined by a Ward identity that enforces spin conservation. For an antiferromagnet (1) contains coupled equations for two sets of slow modes, one near $q=0$ and another near $\mathbf{q}=\mathbf{Q}$.

For the present purposes it is enough to discuss the linewidths defined by the inverse of the zero-frequency Fourier components of $R(\mathbf{q},t)$ and $R(\mathbf{Q}+\mathbf{q},t)$.

In the hydrodynamic region ($q\xi \ll 1$), I find the relaxation rates

$$\hbar\Gamma_0(q) = A\left(\frac{\xi}{a}\right)\left(\frac{\sqrt{2}}{\pi}\frac{\hbar c a^3 kT}{Z_\chi Z_c}\right)^{1/2} q^2,$$

$$\hbar\Gamma_Q(q) = B\left(\frac{a}{\xi}\right)\left(\frac{\sqrt{2}}{\pi}\frac{\hbar c kT}{a Z_\chi Z_c}\right)^{1/2} [1+C(q\xi)^2],$$

for momentum transfer near zero and near $\mathbf{Q}$, respectively. Here, a is the Cu-Cu distance and $A=0.4004$, $B=1.3573$, and $C=0.8811$ are universal constants (independent of the cutoff). The renormalization factors Z_χ and Z_c are defined as in Ref. 1. In the derivation I have assumed that the uniform field susceptibility is temperature independent at low temperatures. For large ξ the relaxation rates are very small. As an illustration consider the quasielastic linewidth at $\mathbf{Q}$. Using the experimentally determined values of ξ and setting (for example) $T=300$ K, one obtains $\hbar\Gamma_Q(0)=1.18$–1.56 meV, for c in the range 0.4–0.7 eV Å. These small rates correspond to the intuition that relaxation over long distances must proceed very slowly. At $T \ll J$ the total intensity in the quasielastic peak is roughly equal to the intensity of the 2D Bragg peak at $T=0$.

In contrast with this, the experiment[2] shows little or no 2D quasielastic intensity even very near T_N. This behavior is inconsistent with both the critical slowing down predicted by the model (and expected on physical grounds), and with the idea that 2D fluctuations drive the transition.

Since at $d=2$ the Heisenberg model is at its lower critical dimension the results of the mode-mode coupling approximation should be taken with caution because it ignores vertex renormalizations that could eventually be quantitatively important.[4] However, it would be extremely surprising if a more accurate treatment did not predict critical slowing down for large ξ. Theory and experiment could perhaps be reconciled if besides being coupled among themselves the spins were strongly coupled to another set of excitations subject to a fast dynamics. Whether this is true and, if it is, what is the nature of these excitations remains to be elucidated.

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[2] G. Shirane, Y. Endoh, R. J. Birgeneau, M. A. Kastner, Y. Hidaka, M. Oda, M. Suzuki, and T. Murakami, Phys. Rev. Lett. **59**, 1613 (1988).

[3] See, for example, K. Kawasaki, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, London, 1976), Vol. 5a, p. 165.

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