Comment on "Spin Correlations of 2D Quantum Antiferromagnet at Low Temperatures and a Direct Comparison with Neutron-Scattering Experiments"

Ding and Makvić[1] (DM) found with their Monte Carlo treatments that spin-$\frac{1}{2}$ antiferromagnets (AFM) exhibit the same exponentially divergent correlation length $\xi(T)=A\exp(2\pi\rho_s/k_BT)$ as is known for classical Heisenberg AFM with the nearest-neighbor interaction $\mathcal{H}=J\sum \mathbf{S}_i\cdot\mathbf{S}_j$ in two dimensions (2D). However, the quantum-mechanical (QM) spin stiffness ρ_s^{QM} is $0.199J$, demonstrating that QM fluctuations change the behavior by a factor of 5 when compared to the classical value $\rho_s/J=1$.

However, the only comparison which is adequate is to a model of classical spins of magnitude S yielding $\rho_s=S^2J=0.25J$. Expressing the DM result by a QM renormalizing factor $f_{\mathrm{QM}}^{(1/2)}=\rho_s^{\mathrm{QM}}/\rho_s^{\mathrm{class}}=0.80$ would indicate that quantum fluctuations cause only a rather small effect of 20%.

Chakravarty, Halperin, and Nelson[2] (CHN) and DM extracted from the same experimental data (see Fig. 8, sample NTT-2, of Endosh *et al.*[3]) the very different experimental fit values $b=1175$ K and $b=1813$ K, respectively, thus differing by 50%, much larger than the 20% to be tested. Both authors display figures with an excellent fit.

This Comment would like to clarify this discrepancy: A close look at the highest simulation point in Fig. 4 of DM reveals that, in contrast to CHN, DM did not use the two experimental points above $T=500$ K, which is certainly justified due to the structural phase transition around this temperature.

Since the important quantum factor to be tested is rather small, possible systematic experimental deviations should be tested.

A possible influence of interlayer coupling can be tested by analyzing experimental data on layer compounds where the quantum effects are expected to be much smaller or negligible, with $S=\frac{5}{2}$ as an example. Using the analytic expression of CHN, the factor $f_{\mathrm{QM}}^{(5/2)}\cong 0.95$ would be close to the classical $f_{\mathrm{class}}=1$.

It is also the aim of this Comment to bring attention to recent neutron-scattering data of $S=\frac{5}{2}$ spins (Mn^{+2}). A similar fit by $\xi\sim\exp(b/T)$ has been reported by Higgins and Cowley[4] for the layered AFM $Rb_2Mn_xCr_{1-x}Cl_4$ ($x=0.95$, far beyond the percolation limit) with $b=132\pm 10$ K, yielding $f_{\mathrm{expt}}^{(5/2)}=bk_B/2\pi S^2J=0.90\pm 0.08$ in agreement with the above QM value $f_{\mathrm{QM}}^{(5/2)}=0.95$, but not far from the classical value $f_{\mathrm{class}}=1$.

A more general remark on investigations of 2D Heisenberg magnets might be added. Spatial effects are explained quantitatively with excited spin waves alone also at elevated temperatures, with the results of DM as an excellent example. This is in contrast to 2D XY magnets, where additional localized excitations are well established for elevated temperatures.

It is finally the aim of this Comment to point out that the excellent fit to spin waves alone does not fully exclude that, in addition to spin waves, localized excitations could be present, because they would cause also $\xi\sim\exp(b/T)$ with b comparable to $b_{\text{spin waves}}$. Such a behavior was proposed long ago by Belavin and Polyakov[5] with 2D localized excitations of density $n(T)\sim\exp(-E/k_BT)$ in the shape of Skyrmions[6] with classical excitation energy $E=4\pi S^2J$. Correcting their[5] obvious error for the average distance $r_{\mathrm{av}}\sim n^{-1}$ to $r_{\mathrm{av}}^2\sim n^{-1}$ for localized excitations in 2D, their assumption[5] $\xi\sim r_{\mathrm{av}}$ would be identical to the classical spin-wave form $\xi\sim\exp(E/2k_BT)=\exp(2\pi\rho_s/k_BT)$ with $\rho_s=S^2J$. Only their assumption[5] that quantum spin renormalization is not essential for Skyrmions would have to be reconsidered, keeping in mind that this reduction is only 20% for spin waves. Therefore, the spatial behavior is not a very discriminating test between the spin-wave or Skyrmion model, in contrast to temporal relaxation effects, where also the form of the temperature dependence would be different for the two models.

Indeed, the line broadening of electron-spin-resonance (ESR) lines shows an Arrhenius behavior in accord with the Skyrmion model. Unfortunately, the only experiments that are known[7] are for $S=\frac{5}{2}$, yielding $f_{\mathrm{expt}}=E_{\mathrm{expt}}/4\pi S^2J$ values of 0.95 ± 0.09, 1.1 ± 0.1, 0.97 ± 0.08, 1.1 ± 0.1 for K_2MnF_4, Rb_2MnF_4, $(CH_3NH_3)_2MnCl_4$, $(C_2H_5NH_3)_2MnCl_4$, respectively, not discriminating between the classical $f_{\mathrm{class}}=1$ and $f_{\mathrm{QM}}^{(5/2)}\cong 0.95$. ESR measurements and other relaxation-type investigations of AFM with $S=\frac{1}{2}$ should be performed.

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Received 13 April 1990
PACS numbers: 75.10.Jm, 74.70.Hk, 75.40.Mg

[1] H.-Q. Ding and M. S. Makivić, Phys. Rev. Lett. **64**, 1449 (1990).

[2] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. Lett. **60**, 1057 (1988).

[3] Y. Endosh *et al.*, Phys. Rev. B **37**, 7443 (1988).

[4] S. Higgins and R. A. Cowley, J. Phys. C **21**, 2215 (1988). Note that their Table 1 is based on the exchange energy $-2J\mathbf{S}_i\cdot\mathbf{S}_j$ between neighboring spins $\mathbf{S}_i$ and $\mathbf{S}_j$, instead of the usual $-J\mathbf{S}_i\cdot\mathbf{S}_j$.

[5] A. A. Belavin and A. M. Polyakov, Pis'ma Zh. Eksp. Teor. Fiz. **22**, 503 (1975) [JETP Lett. **22**, 245 (1975)].

[6] T. H. R. Skyrme, Proc. Roy. Soc. London **262**, 237 (1961).

[7] F. Waldner, J. Magn. Magn. Mater. **54-57**, 873 (1986); **31-34**, 1203 (1983), and references therein.