Spin Correlations in a One-Dimensional Heisenberg Antiferromagnet

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Quasielastic magnetic neutron scattering from the linear-chain antiferromagnet $(CD_3)_4NMnCl_3$ is reported. The system is found to exhibit *planes* of critical scattering perpendicular to the $MnCl_3$ chains from >40°K down to 1.1°K. Both the spatial and thermal variation of the scattering can be quantitatively accounted for at all temperatures using Fisher's theory for the classical Heisenberg linear chain.

Over the past several decades considerable attention has been devoted by theoretical physicists to the problem of the one-dimensional ([1]) antiferromagnet with interaction Hamiltonians varying between the Ising, XY, and Heisenberg limits.[1] For the physically important Heisenberg case this has led to a few exact results such as the determination of the ground-state[2] and first-excited-state[3] energies at $T=0$°K for $S=\frac{1}{2}$ and to the instantaneous correlations at all temperatures[4] for $S=\infty$, but there are still many unanswered questions, particularly with regard to the dynamics. It has also been evident that there are a variety of materials in nature which for structural reasons could approximate well over a certain range of reduced temperature to the Heisenberg linear antiferromagnet. However, in all cases which have been reported to date, the [3] aspects of the system seem to manifest themselves at a relatively early stage so that it has not proven possible to study the static and dynamic behavior of a [1] system with very long-range [1] correlations in the absence of [3] effects.[5] In this Letter we present measurements of the instantaneous correlations in the linear-chain antiferromagnet, $(CD_3)_4NMnCl_3$, which is found to be purely [1] at all temperatures. Direct comparison is then possible with Fisher's exact solution of the classical [1] Heisenberg antiferromagnet. As we shall see, the agreement is excellent at all temperatures. This work then represents the first experimental illustration of an exactly soluble model in the phase-transition problem.

The structure and magnetic properties of $(CD_3)_4NMnCl_3$ (hereafter denoted as TMMC) have been discussed extemsively elsewhere.[6,7] For our purposes here it is sufficient to note that TMMC consists of a hexagonal array of antiferromagnetic $MnCl_3$ chains (Mn^{++}, $S=\frac{5}{2}$) which are magnetically insulated from each other by intervening $(CD_3)_4N^+$ ions. The quasielastic neutron scattering cross section[8,9] for such a chain may be derived simply from Fisher's exact solution for the correlations $\langle \vec{S}_i \cdot \vec{S}_{i+n} \rangle = u^{|n|} S(S+1)$, where u is defined below. This assumes classical spins interacting with a nearest-neighbor Heisenberg exchange $-2J_{nn}\vec{S}_i \cdot \vec{S}_{i+1}$. The result is

$$\frac{d\sigma}{d\Omega_f} \propto S(\vec{Q}) = \frac{B}{\kappa^2 + (2/\pi^2)(\cos\pi Q^z + 1)}, \qquad (1)$$

with

$$B = (u^2 - 1/\pi^2 u)S(S+1), \quad \kappa = (1+u)/\pi\sqrt{-u},$$

where $u = \coth K - K^{-1}$, $K = 2J_{nn}S(S+1)/kT$. In the above we have written Q^z in units of $2\pi/2a$ with a the nearest-neighbor separation along the chain. Inspection of Eq. (1) shows two important features. Firstly, for an antiferromagnet where $-1 \leq u < 0$, the cross section has minima at even integer Q^z and maxima at odd integer Q^z. Secondly, as is obviously required for a [1] system, $S(\vec{Q})$ is independent of the two momentum coordinates Q^x and Q^y perpendicular to the chains so that these maxima at odd Q^z will have the form of *planes* of scattering. This is just the [1] analog of the ridges observed in the [2] antiferromagnet K_2NiF_4.[10] It should also be noted that for $Q^z \sim n$, where n is odd, Eq. (1) reduces to a simple Lorentzian in $q^z = Q^z - n$.

The experiments were performed on a double-axis spectrometer at the Brookhaven high-flux beam reactor using neutrons of wavelength 1.029 Å; 10-min collimation before and after the scattering was employed. We used different crystals, both with nominally 99% deuteration to minimize the large hydrogen incoherent scattering. The crystals had volumes of 0.7 and 0.4 cm^3, respectively, with mosaic spreads of less than 10 min each; they were oriented such that $(h,0,l)$ and (h,h,l) wave vectors, respectively, could be surveyed.[11] Scans were carried out both along and across the anticipated planes of diffuse scattering. Typical scans across the planes for various temperatures and positions are shown in Fig. 1. From the figure it may be seen that the scattering does indeed peak at integer values of l as anticipated. Scans with l fixed at 1, 2, 3, etc. in the $(h,0,l)$ and (h,h,l) directions show that the scattering is independent of h thus confirming that it is *planar*, or, in Fourier transform, that the fluctuations giving rise to the scattering are purely one-dimensional.

Somewhat surprisingly, however, the spectra exhibit maxima at *both* even- and odd-integer Q^z (denoted by l in Fig. 1) contrary to our expectations based on Eq. (1). At 22°K at the even-l position a sharp, resolution-limited peak with essentially a flat background is observed. At the odd-l position at 22°K the scattering clearly has two distinct components: a sharp peak at the center similar to that observed at the even position together with a much broader Lorentzian peak with an integrated intensity considerably larger than that of the central component. Studies of the temperature dependence between 1.1 and 30°K show that both the even-l peak and the central peak at odd l are temperature independent, whereas the residual scattering at odd l varies rapidly in peak intensity and width with temperature. It is evident that the temperature-independent scattering observed at both even and odd l has a unique origin; it must be nonmagnetic since it does not follow the Mn^{++} form factor. The probable explanation is that it arises from pseudo-one-dimensional correlated motion of the $(CD_3)_4N^+$ ions. The remaining scattering at the odd-l positions then may be identified as the anticipated [1] magnetic scattering.

The scans across the $l=1$ planes in the $(h,0,l)$, (h,h,l) crystals were analyzed as follows. Data taken above 8°K were fitted by Eq. (1) with the central points omitted so that the nuclear and magnetic contributions could be separated out.

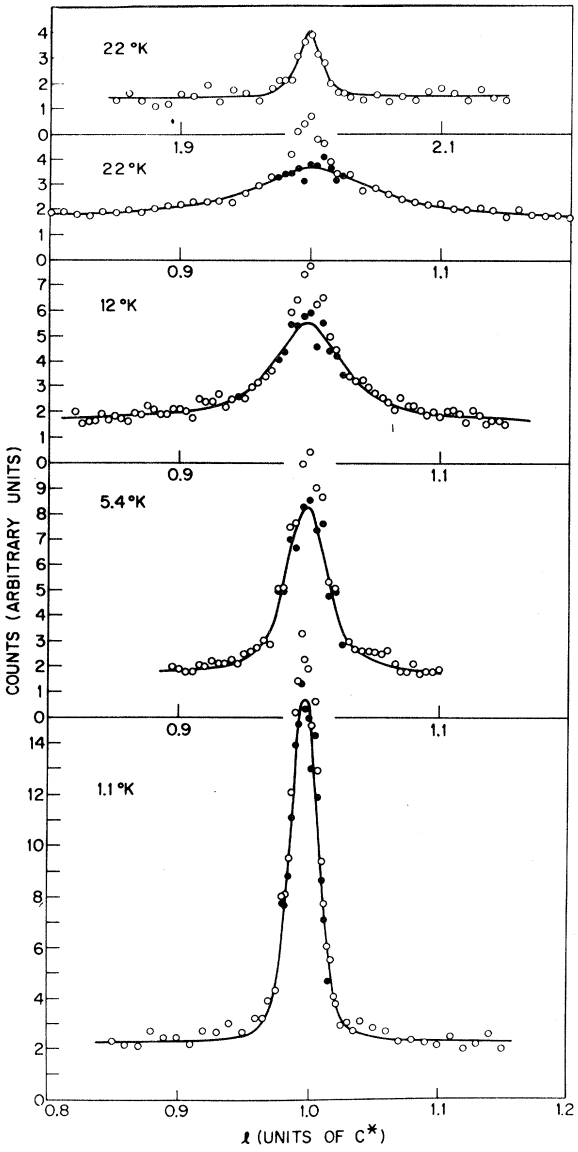

FIG. 1. Quasielastic scans across the planes in TMMC. The upper four scans were made in the $(h,0,l)$ crystal with h fixed at 0.28 reciprocal-lattice units. The lowest scan was made in the (h,h,l) crystal with $h=0.1$. The open circles are the actual experimental data; the closed circles give the residual magnetic scattering after the nuclear contribution has been subtracted off. The solid lines in the lowest four scans are theoretical fits as described in the text.

As noted above, the nonmagnetic scattering was found to be temperature independent up to 30°K so that it could be uniformly subtracted off from all of the data. This is necessary at the lower temperatures since the two components are not resolved. The magnetic scattering at all temperatures was then fitted by Eq. (1) convoluted with

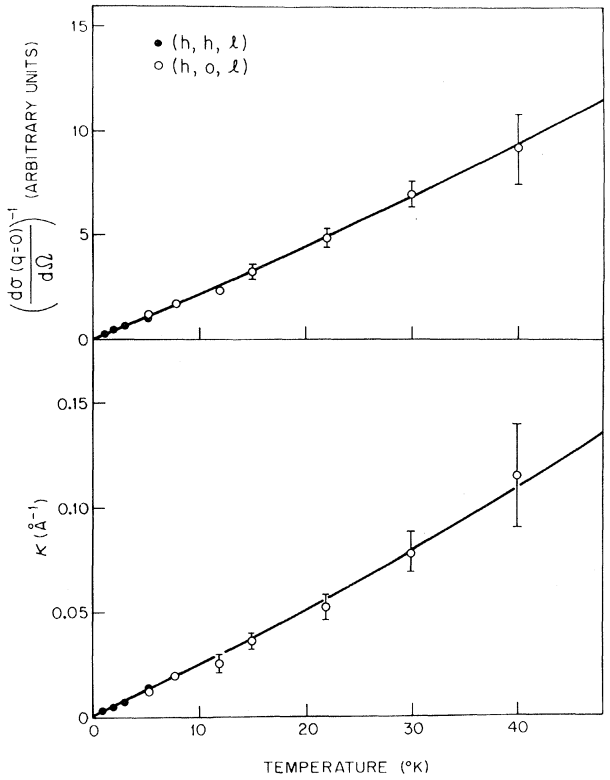

FIG. 2. Results obtained from the fits of the cross section, Eq. (3), to the magnetic scattering. The solid lines are fits by the classical theory as described in the text.

the instrumental resolution function. The solid lines in Fig. 1 are the curves generated by these fits. From the figure it may be seen that the fits are excellent ($\chi^2 \sim 1$) at all temperatures.

Figure 2 gives the deconvoluted inverse peak intensity and the inverse correlation length as a function of temperature. Both are observed to extrapolate to zero at 0°K as expected for the [1] system. This behavior is quite analogous to a normal second-order phase transition in a [3] antiferromagnet with the notable difference that in this [1] system $T_N^{(1)} = 0$°K. In three dimensions $d\sigma(q=0)/d\Omega$ is proportional to the staggered susceptibility.

It is of interest to compare our results quantitatively with the predictions of Fisher's classical model. From Eq. (1) it may be seen that the theoretical expression for κ involves only the one adjustable constant J_{nn}. The solid line in Fig. 2 for κ represents the best fit of the classical expression for κ, as given in Eq. (1), to our experimental results with $J_{nn} = -7.7 \pm 0.3$°K. This value for J_{nn} is somewhat higher than that determined from the bulk susceptibility, $J_{nn} \sim -6.5$°K, but probably lies within the uncertainty associated with the neglect of further neighbor interactions. The corresponding prediction of the classical model for $d\sigma(q^z = 0)/d\Omega$ is shown as the solid line in the upper part of the figure. Here one overall scaling factor is adjusted to give the best fit; again the agreement is excellent. These results may be stated as follows: *Fisher's classical nearest-neighbor Heisenberg model properly predicts both the spatial and thermal variation of the instantaneous correlations in TMMC at all temperatures between 40 and 1.1°K.* This is a remarkable result especially when one considers the tremendous labor involved in the corresponding theory for [2] and [3] systems.

There are several additional points of note. Firstly, we have searched for evidence of [3] magnetic correlations at temperatures down to 1.1°K without success. Our search was by no means exhaustive, but it does suggest that the [3] ordering temperature may be somewhat lower than the 0.84°K inferred by Dingle, Lines, and Holt.[6] Secondly, it is clear that the dynamics in a [1] Heisenberg antiferromagnet are of considerable interest. Inelastic measurements show that at low temperatures there are long-lived spin waves over almost the entire [1] Brillouin zone in spite of the absence of long-range order. This will be the subject of a detailed report to be published elsewhere.[12] Finally, our experimental results pose a number of interesting theoretical problems. Although the Mn^{++} spin is large ($S = \frac{5}{2}$), we (at least) find it surprising that quantum mechanical effects do not seem important at 1.1°K. To our knowledge no theoretical calculations have as yet been reported for the linear chain for finite S other than $S = \frac{1}{2}$. In addition, TMMC would seem to offer an ideal test case for the theories of spin dynamics in paramagnetic systems. So far the only published work on [1] Heisenberg systems at finite temperatures is that of McLean and Blume.[13] Clearly, further theoretical work is required.

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New Experimental Test of Coulomb's Law: A Laboratory Upper Limit on the Photon Rest Mass

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A high-frequency test of Coulomb's law is described. The sensitivity of the experiment is given in terms of a finite photon rest mass using the Proca equations. The null result of our measurement expressed in the form of the photon rest mass squared is $\mu^2 = (1.04 \pm 1.2) \times 10^{-19}$ cm^{-2}. Expressed as a deviation from Coulomb's law of the form $1/r^{2+q}$, our experiment gives $q = (2.7 \pm 3.1) \times 10^{-16}$. This result extends the validity of Coulomb's law by two orders of magnitude.

The testing of Coulomb's law (Gauss's law) by means of a null experiment dates back to Cavendish (1773).[1] The now classical test of Plimpton and Lawton[2] was performed in 1936, and showed that any difference in the exponent from 2 was smaller than 1×10^{-9}. Recently two other groups have extended the accuracy of that result by two[3,4] and four[5] orders of magnitude. The result reported here represents an extension in accuracy over that obtained by Plimpton and Lawton by six orders of magnitude.

The experiment described here is a "high-frequency" null test of Coulomb's law. We make use of the fact that a $1/r^2$ force law does not give rise to any electric field on the inside of a closed conductor. A conducting shell that is about $1\frac{1}{2}$ m in diameter is charged to 10 kV peak to peak with a 4-MHz sinusoidal voltage. Centered inside of this charged conducting shell is a smaller conducting shell. Any deviation from the $1/r^2$ force law is detected by measuring the line integral of the electric field between these two shells with a detection sensitivity of about 10^{-12} V peak to peak.

The results of the experiment can be expressed in terms of the Proca equations,[6,7] a relativistically invariant linear generalization of Maxwell's equations, which are appropriate to describe the experimental system when a finite rest mass is assumed. Proca's equations for a particle of spin 1 and mass m_0 are[8]

$$(\Box + \mu^2) A_\nu = (4\pi/c) J_\nu, \tag{1}$$

where $\mu = m_0 c/\hbar$. In three-dimensional notation, Gauss's law becomes

$$\nabla \cdot \vec{E} = 4\pi\rho - \mu^2 \varphi. \tag{2}$$

To calculate the sensitivity of the system, consider an idealized geometry consisting of two concentric, conducting, spherical shells of radii R_1 and R_2 ($R_2 > R_1$) with an inductor across (in parallel with) this spherical capacitor. To the outer shell is applied a potential $V_0 e^{i\omega t}$. An iterative solution for the field between the spheres may easily be found. Forming a spherical Gaussian surface at radius r between the two shells and then using the approximation $\varphi(r) = V_0 e^{i\omega t}$ for this interior region, the integral of Eq. (2) over the volume interior to the Gaussian surface becomes

$$\int [\nabla \cdot \vec{E} - 4\pi\rho + \mu^2 V_0 e^{i\omega t}] d^3x = 0. \tag{3}$$