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Dynamics of an $S=\frac{1}{2}$, One-Dimensional Heisenberg Antiferromagnet

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We report a detailed neutron-scattering study of the spin dynamics in $CuCl_2 \cdot 2N(C_5D_5)$, a physical realization of the one-dimensional $S=\frac{1}{2}$ Heisenberg antiferromagnet. At $T=1.3$ K well-defined excitations are observed over the whole zone with energies given by $E(q)=\pi J_{nn}\sin(qc)$, the celebrated des Cloizeaux–Pearson exact solution for the spectrum of first excited states, but with intensities approximately those expected from classical spin-wave theory. At $T=8$ K the excitations are broad and ill defined.

The one-dimensional (1D), nearest-neighbor (nn), $S=\frac{1}{2}$ Heisenberg antiferromagnet is one of the few nontrivial many-body problems with interesting dynamics for which exact solutions exist. In 1931, Bethe[1] found the ground-state eigenfunction, and he showed that no long-range order exists even at 0 K. Somewhat later, Hulthén[2] derived the ground-state energy $E_0=-|J|N(2\ln 2-\frac{1}{2})$. In 1962, des Cloizeaux and Pearson[3] (dC-P) found that the first excited states obey the simple dispersion relation

$$E(q)=\pi J|\sin(qc)|, \tag{1}$$

where c is the nn separation, and they identified these excitations as "spin waves." They noted that, quite remarkably, the dispersion relation, Eq. (1), has a double periodicity of π just as in standard spin-wave theory[4] starting from an assumed Néel ground state. However, the coefficient of J in Eq. (1) is equal to π, compared with 2 for classical spins.[4] It has not proven possible, however, to go from the exact dC-P calculation to a response function $\chi''(\vec{q},\omega)$. In order to elucidate this problem, we have carried out a detailed inelastic neutron scattering study of the spin dynamics in $CuCl_2 \cdot 2N(C_5D_5)$, dichlorobis pyridine copper II (CPC). As discussed below, CPC is a good physical approximation to the idealized 1D nn $S=\frac{1}{2}$ Heisenberg antiferromagnet. We find that at $T=1.3$ K there are sharp excitations with a dispersion relation given precisely by Eq. (1); this work thence constitutes the first experimental observation of the dC-P states.

We first discuss the crystal structure[5] and the consequent magnetic properties[6] of CPC. As shown in Fig. 1, CPC is body-centered monoclinic, with space group $P2_{1/n}$. Each Cu^{++} ion is located at the center of a rhombus of chlorine and nitrogen atoms which are stacked along the c axis, thence giving rise to a linear chain structure. The chains are kept well separated by the large intervening pyridine molecules; the ratio of in-

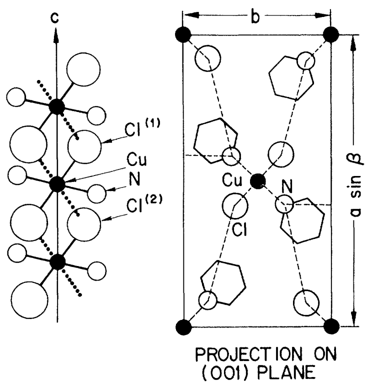

FIG. 1. Crystal structure of CPC; left-hand side, $CuCl_2 \cdot N$ chains; right-hand side, view of the unit cell normal to the chain axis (c axis). Room-temperature lattice constants: $a=17.00$ Å, $b=8.59$ Å, $c=3.87$ Å; $\beta=91°52'$.

terchain to intrachain exchange is about 3×10^{-3} so that for the purposes of this report the magnetic properties may be taken as purely one-dimensional. The low-symmetry crystal field completely lifts the Cu^{++} $3d^9$ orbital degeneracy, thence rendering lowest an $S=\frac{1}{2}$ doublet.

Electron-spin-resonance measurements[6] of CPC at room temperature show that the Cu^{++} ions have an extremely narrow resonance line; this necessitates that the effective exchange Hamiltonian be isotropic to within ~2%. Both the bulk susceptibility and magnetic specific heat[6] of CPC are accurately described by the Bonner and Fisher[7] numerical calculations for the $S=\frac{1}{2}$ isotropic Hamiltonian with nn exchange $J=13.4\pm0.2$ K. Thus, CPC corresponds rather well to the idealized 1D model. The only possible caveat is next-nearest-neighbor (nnn) exchange along the chain. However, by comparison with other materials, we estimate that $J_{\text{nnn}}/J_{\text{nn}}<0.03$ which introduces an uncertainty in Eq. (1) of ~6%, much less than the predicted $100(\pi/2-1)=57\%$ renormalization due to quantum effects. Within an overall accuracy of about 10% then, above the 3D ordering temperature of 1.14 K, CPC should be well described by the simple 1D Hamiltonian

$$\mathcal{H}=2J\sum_i \vec{S}_i\cdot\vec{S}_{i+1}, \quad S=\tfrac{1}{2}, \tag{2}$$

with $J=13.4$ K.

The experiments were performed on a triple-axis spectrometer at the Brookhaven National Laboratory high-flux beam reactor using neutrons of 13.7 meV incident energy. Needles of deuterated CPC typically 20 mm in length and 0.5 mm in diameter were grown in the manner described by Takeda, Matsukawa, and Haseda.[6] A sample volume of 1.5 cm^3 was obtained by aligning several hundred needles on grooved aluminum plates, thence producing an effective 1D single crystal with a mosaicity of several degrees. The experimental results may be briefly stated.

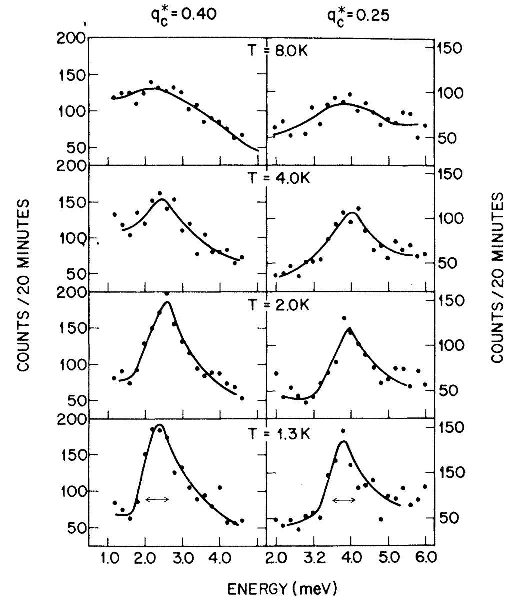

FIG. 2. Variation with temperature of scans at $q_c^*=0.40$ and 0.25. Solid lines, guides for the eye. The arrows at $T=1.3$ K give the instrumental width.

Constant-Q scans were made relative to the $l=1$ plane with the reduced momentum transfers varying from $q_c^*=0.07$ to $q_c^*=0.46$, where $q_c^*=qc/2\pi$; the zone boundary is at $q_c^*=0.5$. Values of q_c^* closer to 0 were ruled out by a vanishing structure factor, whereas those closer to 0.5 were prevented by the large incoherent scattering around $E=0$. Typical experimental scans at $q_c^*=0.25$ and $q_c^*=0.40$ for temperatures between 1.3 and 8 K are shown in Fig. 2. We consider first the 1.3-K results. At this temperature well-defined peaks are observed at all $\vec{q}$'s examined. As a note of caution, we should emphasize that the asymmetric line shapes may be purely due to an instrumental artifact. High-resolution scans on a single crystal are required to determine the intrinsic cross section. The peak energies are plotted in Fig. 3 in units of $J=13.4$ K. As is evident in the figure, the agreement of the measured dispersion relation with the dC-P result, Eq. (1), is excellent. Indeed, the agreement is better than might have been expected considering the uncertainties discussed above. Measurements at fixed q_c^* with varying $q_\perp^*$, the momentum component perpendicular to the chains,

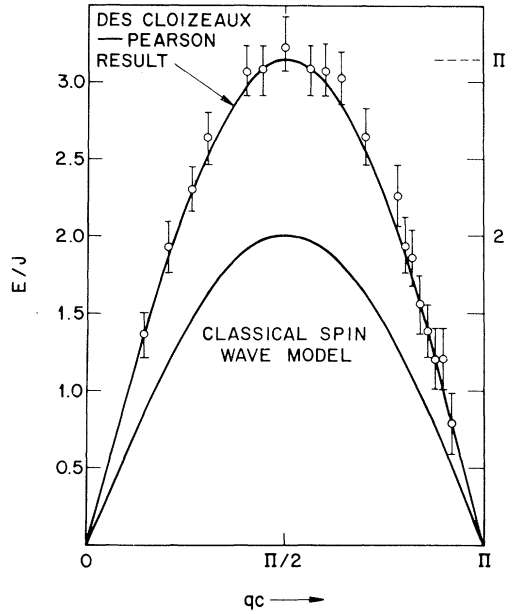

FIG. 3. Dispersion of the excitations in CPC at $T=1.3$ K with energy in units of $J=13.4$ K.

show that the peak energy is independent of $q_\perp{}^*$; thus, these are pure 1D excitations propagating along individual CuCl$_2$N chains. These results, therefore, both illustrate experimentally the dC-P exact solution for the lowest-excited-state energies and, in addition, they show that these dC-P states comprise a major part of the spectral weight.

The neutron scattering cross section, proportional to $\chi''(\vec{q},\omega)$, contains information not only about the elementary excitation eigenvalues but also their eigenfunctions through the measured intensities. As discussed by Hutchings *et al.*,[8] spin-wave theory for a classical nn linear chain predicts that the intensity should vary like $(1-\cos qc)/\sin qc$ for $0 \leqslant qc \leqslant \pi$. As is evident in Fig. 2, it is only possible to obtain a coarse measure of the integrated intensity of the scattering in these experiments because of large uncertainties in the background and the high-energy cutoff. Nevertheless, with these difficulties in mind, we see from Fig. 4 that the experimental intensity follows rather well the classical spin-wave result. Thus, following dC-P's original identification, these excitations may be validly described as *spin waves*.

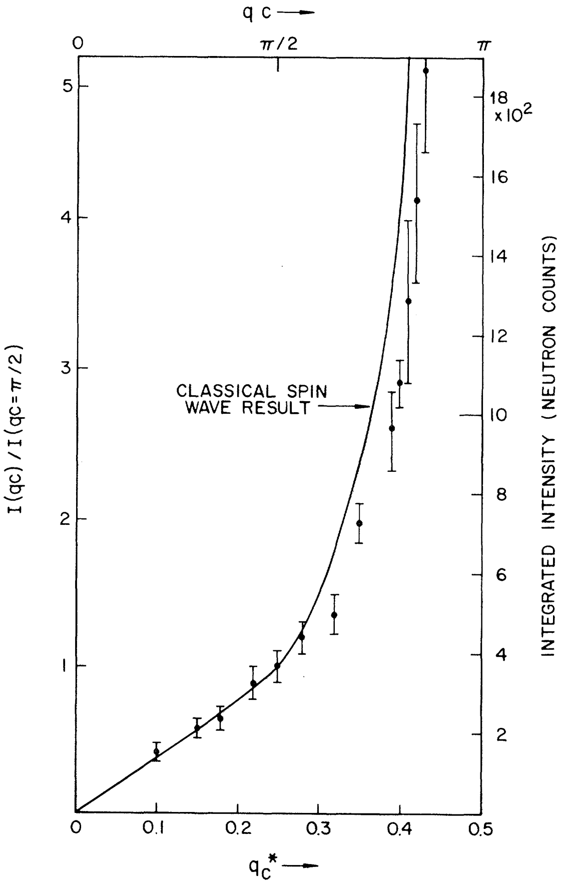

FIG. 4. Integrated intensity of the $T=1.3$ K peaks as a function of wave vector. Solid line, theoretical spin-wave intensity curve $(1-\cos qc)/\sin qc$ multiplied by an instrumental and Cu^{++}-form-factor correction factor. The theoretical curve is normalized at $qc=\pi/2$.

We now consider the temperature dependence of the scattering. There is at the present time no theory for the temperature dependence of $\chi''(q,\omega)$ for the $S=\frac{1}{2}$ 1D system except for some exact calculations on finite chains.[9] However, extensive information does exist both for classical spins from computer experiments and theory[10] and for $S=\frac{5}{2}$ from the experiments in $(CD_3)_4NMnCl_3$ (TMMC).[8] The experimental results for CPC are shown in Fig. 2. As the temperature is raised, the excitations broaden rapidly; however, there is no appreciable change either in the peak position or in the integrated intensity. For $T=8$ K and $q_c{}^*=0.40$ so that $k_BT/\hbar\omega(\text{peak})\simeq 0.3$, the excitation is already highly damped. Thus the thermodynamic properties may not be calculated in this temperature range in terms of sharp dC-P excitations with definite statistics, either Fermi or Bose. It is of interest to compare these results directly with the experiments in TMMC.[9] In order to make a proper comparison it is necessary to define relative temperature scales. These would be given most directly via the correlation lengths $\xi(t)$, but in the absence of such information for CPC, we instead use $JS(S+1)/k_{\rm B}$ as the appropriate scaling factor. The temperature of 1.3 K here then corresponds to 8 K in TMMC. With this taken into account, comparison of Fig. 2 here with Figs. 11 and 12 of Hutchings *et al.*[8] shows that the general qualitative features are quite similar in CPC and TMMC in spite of the difference in spin values. Thus the extreme nonclassical nature of the $S=\frac{1}{2}$ system does not seem to play a central role in the temperature evolution in the dynamics, at least at the wavelengths we have probed.

Finally, we have also carried out a series of quasielastic scans around $q_c^*=0.5$ for various values of $q_\perp^*$ at 1.3 K. No appreciable 3D critical scattering is observed, indicating that at this temperature the correlations are still largely 1D. Planar 1D scattering is clearly observable; however, as a result of the anomalously large incoherent scattering and small signal (signal:background ~1:10) no determination of $\xi(T)$, the correlation length, could be made.

To conclude, we emphasize that continued experiments together with a renewed theoretical effort on the $S=\frac{1}{2}$ system in one dimension are clearly called for.

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Study of Deuteron-Cluster Deformation Using the Reaction ^{6}Li(d, tp)^{4}He

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The reaction ^{6}Li(d,tp)^{4}He has been studied for two values of residual ^{4}He momentum ($\vec{k}_\alpha=0$ and -30 MeV/c). The shift of the first minimum in the angular distribution of the reaction ^{6}Li(d,tp)^{4}He, for fixed $\vec{k}_\alpha$, with respect to the first minimum in the angular distribution of the reaction $d(d,t)p$, has been used to investigate the structure of the d cluster in the ^{6}Li nucleus.

There have been several attempts to explain the ground-state characteristics of ^{6}Li using an α-d cluster model.[1–4] In the analysis of electron scattering on ^{6}Li,[5] the low-energy d-α scattering and ^{6}Li bound-state calculations,[6,7] and the study of medium-energy cluster knock-out reactions[8] on ^{6}Li, it was noticed that the d cluster in ^{6}Li has a smaller size than a free deuteron. However, it may be interesting to note that in optical-model analysis of deuteron elastic scattering on medium and heavy nuclei[9] there are indications of deuteron stretching. A more direct test is therefore necessary for the shrinkage of the d cluster in the ^{6}Li nucleus. In the case of electron scatter-