Neutron study of the line-shape and field dependence of magnetic excitations in $CuCl_2 \cdot 2N(C_5D_5)$

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We have carried out inelastic neutron scattering on $CuCl_2 \cdot 2N(C_5D_5)$, at $T = 1.2$ K and at magnetic fields up to 70 kOe. The spin dynamics of this typical $s = \frac{1}{2}$ one-dimensional Heisenberg antiferromagnet have previously been investigated at zero magnetic field by Endoh *et al.*, using neutron scattering. They observed a spectrum of magnetic excitations in close agreement with the spectrum of lowest excited states as calculated exactly by des Cloizeaux and Pearson (dCP). The marked asymmetry in the line shape of the neutron response previously observed is carefully reexamined and is shown to be a true effect, in agreement with several theoretical predictions. At high magnetic field, a broadening of the neutron response is observed, especially pronounced at the antiferromagnetic zone boundary, where the peak smears out at 70 kOe. For wave vectors near an antiferromagnetic Bragg point a decrease in the peak energy is observed for increasing field, lending qualitative support to the calculations of Ishimura and Shiba of the field dependence of the dCP states.

I. INTRODUCTION

In recent years, a variety of magnetic materials has been found for which the magnetic interaction predominantly is between ions within well-separated chains. Consequently, such compounds constitute good physical approximations to ideal one-dimensional magnetic models, which in general are much more tractable than corresponding three-dimensional models. For this reason, these materials have attracted a great deal of interest from the experimentalists and both static and dynamical properties have been measured by various techniques.[1,2] As discussed by Endoh *et al.*,[3] the metalorganic compound $CuCl_2 \cdot 2N(C_5D_5)$, dichloro *bis* pyridine copper II (CPC) is a good realization of the ideal $S = \frac{1}{2}$ nearest-neighbor antiferromagnetic Heisenberg chain. The structure of this compound is body-centered monoclinic of space group $P2_{1/n}$,[4] and is shown in Fig. 1. The chains of Cu^{2+} ions are along the c axis and are well separated from each other by the large intervening pyridine groups, thus giving rise to the one-dimensional magnetic properties. It was previously argued[3] that above the three-dimensional ordering temperature of $T = 1.4$ K, the magnetic properties of CPC should be adequately described by the one-dimensional Heisenberg nearest-neighbor Hamiltonian

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

with the antiferromagnetic exchange constant equal to $J = 13.4$ K, as determined from bulk measurements.[5]

The determination of the lowest excited states of the Hamiltonian (1) is complicated by the fact that the exact ground state is not simply the antiferromagnetically ordered Néel state. Hence, standard linear spin-wave theory cannot be expected to be immediately applicable, especially not in the quantum limit $S = \frac{1}{2}$. Considerable interest has been devoted to the eigenvalue problem of the Hamiltonian (1),[6–8] and des Cloizeaux and Pearson[6] (dCP) were the first to calculate exactly the spectrum of the lowest excited eigenstates of (1). These authors found that they obey the simple dispersion relation

$$E(q) = \pi J |\sin(qc)| , \tag{2}$$

c being the nearest-neighbor distance. Apart from the fact that the coefficient of J is equal to π, the spectrum (2) is, quite surprisingly, similar to the standard spin-wave result which yields a coefficient of J equal to two. The dCP spectrum (2) is shown by

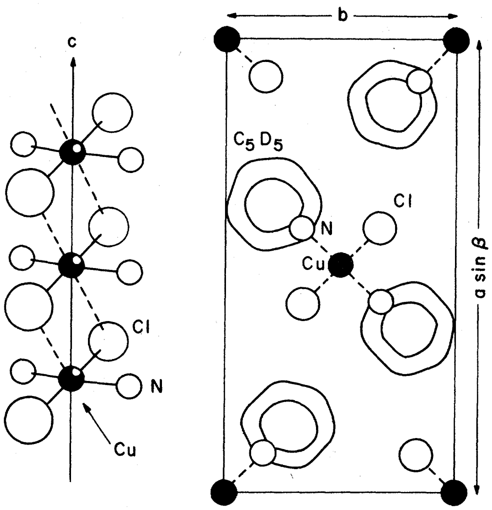

FIG. 1. Crystal structure of CPC. The right-hand side shows a projection of the unit cell normal to the c axis (chain direction). Left-hand side shows the Cu^{2+} chain. Room-temperature lattice constants are: $a = 17.00$ Å, $b = 8.59$ Å, $c = 3.87$ Å, and $\beta = 91°52'$.

the full curve in the upper part of Fig. 2. The calculation of dCP does not allow a determination of a neutron scattering function $S(Q,\omega)$, however. Endoh *et al.* found by means of inelastic neutron scattering a spectrum of well-defined magnetic excitations in CPC in close agreement with the dCP spectrum (2), and the dispersion was found to be in-

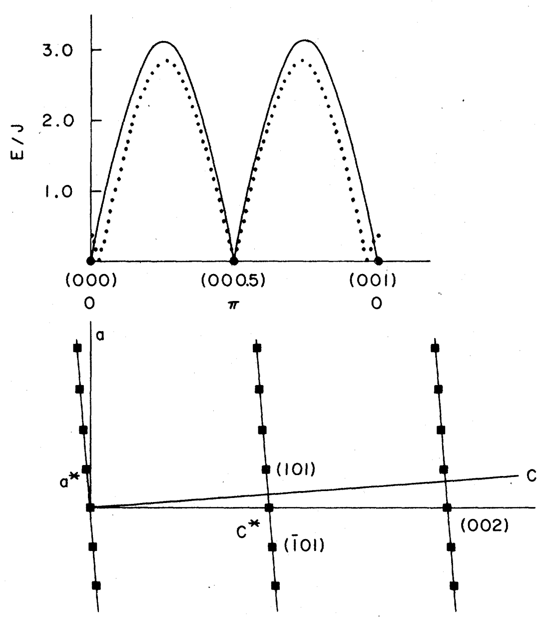

FIG. 2. dCP spectrum at zero field (solid curve) and at 70 kOe (dotted curve), upper part. Lower part shows the reciprocal-lattice plane of CPC used as the scattering plane in the experiments.

dependent of the wave-vector component perpendicular to the c direction, thus displaying the one-dimensional character. Moreover, they found, quite remarkably, that the variation over the Brillouin zone of the integrated intensity of these excitations followed quite well the predictions of classical spin-wave theory,[9] i.e.,

$$I \propto (1-\cos qc)/\sin qc, \quad 0 < qc < \pi \ .$$

In addition, Endoh *et al.* observed for values of qc close to the antiferromagnetic Bragg point (π) an asymmetrical line shape of the constant-Q scans, having relatively more weight at the high-energy flank.

It has been shown by Yamada[10] and by Bonner and Sutherland (for a finite chain)[11] that there exist low-lying $S=1$ states above the dCP states. In fact, these states form a continuum between the lower boundary of the dCP states (2) and an upper boundary,

$$E_u(q) = 2\pi J|\sin\tfrac{1}{2}(qc)| \ .$$

In the past few years, several attempts[11–14] have been made to calculate the spectral weight of these states (see Sec. IV), yielding the results that the dCP states indeed have the major spectral weight, but some weight is found at higher energies. This is in qualitative agreement with the observed asymmetrical line shapes and will be further discussed in Sec. IV in relation to the present measurements.

The dependence of an applied magnetic field on the spectrum of lowest excited states of (1) has been calculated approximately by Pytte[8] and later by Ishimura and Shiba,[15] who extended the exact treatment of des Cloizeaux and Pearson[6] to the case of an applied magnetic field. These authors find a field dependence of the dCP spectrum which is qualitatively different from the result obtained in classical spin-wave theory.[16] This is illustrated by Fig. 3, where the parameter h is defined through $h \equiv g\mu_B H/2J$, H being the applied magnetic field. The calculations are based on zero anisotropy field, which means that for small values of H the spins are virtually perpendicular to H and then cant along the direction of the field when this is increased. For $h \geq 2$ the spins are aligned along the applied field (at 0 K). The right-hand side of Fig. 3 shows the results of Ishimura and Shiba, while the left-hand side shows the H dependence of the classical spin-wave spectrum.[16] In the latter case, only one of the two spin-wave branches is shown for each value of h less than two; the other branches are obtained by folding the shown spectra about $qc = \frac{1}{2}\pi$. It is seen that in the presence of a magnetic field the dCP spectrum goes to zero at some point between 0 and π, in contrast to the classical case. Furthermore, the dCP energy at $qc = \frac{1}{2}\pi$ decreases with increasing field, whereas the classical spin-wave energy at $qc = \frac{1}{2}\pi$ is independent of the field (for $0 \leq h \leq 2$). The dCP spectrum at $H = 70$

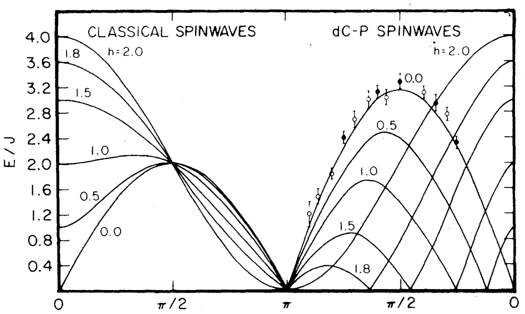

FIG. 3. Magnetic field dependence of spin-wave energies in a one-dimensional antiferromagnetic nearest-neighbor Heisenberg chain [Eq. (1)], where $h = g\mu_B H/2J$. The left-hand side shows the results obtained in classical linear spin-wave theory for zero anisotropy field. For $h < 2$, only one branch is shown for each value of h. The right-hand side shows the field dependence of the dCP states. Experimental points for $h = 0$ are shown with open circles (present investigation) and black circles (previous investigation).

kOe (experimental limit), corresponding to $h = 0.21$, is represented by the dotted curve on the upper part of Fig. 2. This curve was found approximately using the results shown in Figs. 1 and 2 of the paper by Ishimura and Shiba.[15] It is seen that the decrease in excitation energy in going from 0 to 70 kOe is rather small, especially in the vicinity of an antiferromagnetic Bragg point.

In this paper we present the results of new improved inelastic neutron scattering experiments on CPC at $T = 1.2$ K and zero magnetic field, with special attention to the asymmetrical line shapes. Moreover, we have carried out measurements at magnetic fields up to 70 kOe, the results of which will be discussed in relation to Fig. 3.

II. EXPERIMENTAL TECHNIQUE

A. Sample preparation and instrumentation

The sample was prepared in a manner similar to that described previously.[3] By aligning approximately 50 small elongated CPC crystals, a total sample volume of 2 cm^3 was obtained. The resulting mosaic spread was consequently large, about 2.5° (see Sec. III). For the measurements at zero magnetic field, the sample was placed in a pumped He cryostat reaching a lowest temperature of 1.2 K. The measurements at high field were performed by means of a pumped He cryostat in connection with a He-cooled superconducting magnet thus capable of reaching 1.6 K and 80 kOe.

The experiments were carried out with a neutron triple-axis spectrometer at the Brookhaven High Flux Beam Reactor. Pyrolytic graphite of mosaic spread equal to 24′, reflecting from (002) planes was used for monochromator as well as the bent analyzer. A fixed incoming energy of $E_i = 13.7$ meV and horizontal collimation equal to 20′–40′–40′–40′ was used. A tuned pyrolytic graphite filter was applied to remove higher-order contamination.

The sample was oriented in such a way as to make a (010) plane of reciprocal space coalesce with the scattering plane. This reciprocal-lattice plane is shown in Fig. 2 (lower part). The a and c (chain) directions of the direct lattice are also shown. At $T = 1.2$ K the values of a^* and c^* were found to be 0.3785 and 1.6675 Å^{-1}, respectively. All inelastic measurements were performed in the constant-Q mode and the scans were recorded for a fixed monitor count.

B. Intensity corrections and line-shape calculations

The measured intensity $I(Q,\omega)$ results from a convolution of the cross section $d^2\sigma/d\omega d\Omega$ of the sample with the resolution function $R(Q+\Delta Q, \omega+\Delta\omega)$ of the spectrometer

$$I(Q,\omega) = \int \frac{d^2\sigma}{d\omega d\Omega}(Q+\Delta Q, \omega+\Delta\omega) \times R(Q+\Delta Q, \omega+\Delta\omega)\, d(\Delta Q)\, d(\Delta\omega) \quad . \quad (3)$$

For a constant-Q scan with fixed incoming energy the measured intensity should be corrected for the variation in $R(Q,\omega)$ with ω (the size of the resolution function decreases with increasing ω). Moreover, correction should be made for a possible energy dependence of the reflectivity of the analyzer. The resolution effect can be corrected for by applying the factor $(k_F^3/\tan\theta_A)^{-1}$ to the measured points, as shown by Chesser and Axe.[17] The energy dependence of the pyrolytic graphite (002) reflectivity was measured by Shapiro and Chesser,[18] and for energies between 10 and 15 meV, corresponding to the present case, virtually no change was observed. Accordingly, no corrections for analyzer reflectivity have been carried out in the present analysis.

The integration (3) of the experimental line shape, based on four-dimensional Gaussian resolution functions[19] and Lorentzian cross sections is a standard computational task. The effect of a mosaic spread of the sample on the line shape (3) is in general an additional broadening, depending on the shape of the dispersion surface, and can effectively be taken into account through the elements of the 4 × 4 resolution matrix, as shown by Werner and Pynn.[20] Using the formulas herein we have extended the current program to allow for a finite sample mosaic spread. Thus, by applying Lorentzian cross sections and a value ~2.5° of the sample mosaic spread, we have adequately reproduced the observed phonon line shapes. However, similar calculations of line shapes of the magnetic excitations failed to explain observed asymmetry. This is discussed in the Sec. III.

III. EXPERIMENTAL RESULTS AND ANALYSIS

A. Experiments at zero magnetic field

The constant-Q scans at three different values of wave-vector transfer are shown in Fig. 4, displaying the dCP excitations. The peak positions are in close agreement with the results obtained previously.[3] The peak at ~4.5 meV observed for $\vec{Q}=(0,0,0.825)$ is due to a longitudinal optical phonon. Some of the observed excitation energies are plotted in Fig. 3 (open circles) together with some points from the earlier investigation (closed circles). The agreement with the dCP spectrum ($h=0.0$) is seen to be good.

While the neutron peaks for $\vec{Q}=(0,0,0.75)$ and $\vec{Q}=(0,0,0.825)$ seem fairly symmetric, a pronounced asymmetry is displayed for $\vec{Q}=(0,0,0.60)$. This feature is moreover established by other scans close to the antiferromagnetic Bragg point (0,0,0.5) as shown in Fig. 5, where the upper trace is from the present investigation while the middle and lower trace are data from the previous measurements.

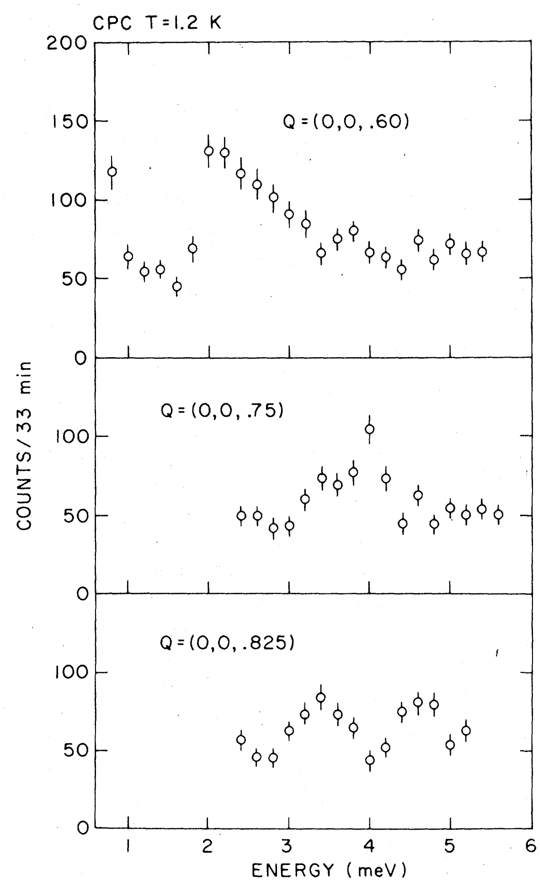

FIG. 4. Constant-Q scans at $T=1.2$ K and zero magnetic field showing magnetic excitations. The peak at ~4.5 meV for $Q=(0,0,0.825)$ arises from an optical phonon.

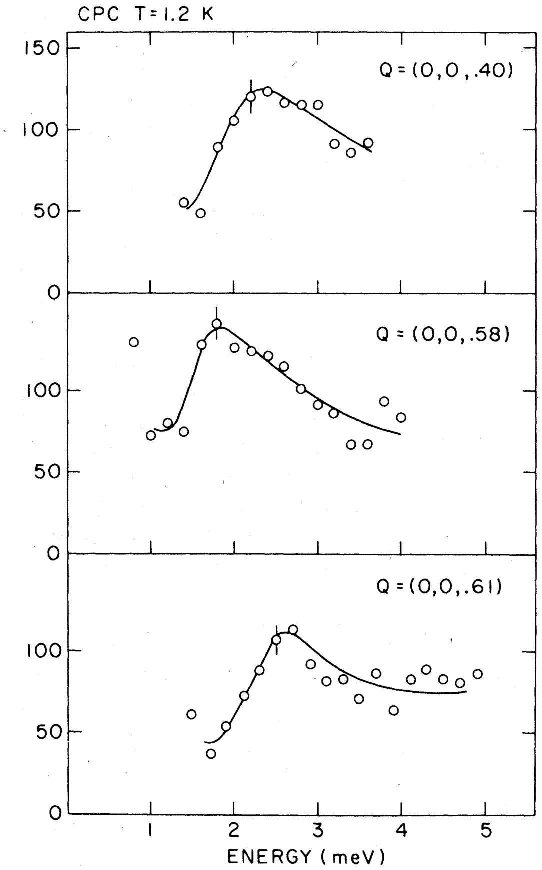

FIG. 5. Inelastic scans at $T=1.2$ K and zero magnetic field at three points in reciprocal space close to an antiferromagnetic Bragg point (0,0,0.50), showing magnetic excitations. The upper scan shows data from the present work, while the two others are from the previous work.

The asymmetry might *a priori* be due to either a pure resolution effect or be a true physical effect. In order to investigate the former possibility we have carried out various calculations of line shapes for one-dimensional magnetic excitations as well as phonons, allowing for a finite sample mosaic spread as described in Sec. II B.

In Fig. 6(a) we have plotted the neutron scans for a longitudinal acoustical phonon of wave vector $q=0.075c^*$ measured at $\vec{Q}=(0,0,2.075)$ and an optical zone-center mode measured at $\vec{Q}=(0,0,1.0)$. A horizontal background (= 30 counts/12 min) and a sloped background (indicated by the dashed line), respectively, were subtracted from the two sets of data points and the resulting points were then subjected to the resolution correction described in Sec. II. The points thus obtained are shown in Fig. 6(b). The solid curves are the results of the line-shape calculations based on (3). In the upper case, an acoustic dispersion relation $E=v|\vec{q}|$, with $v=18.2$ meV Å was used. For the optical zone-center mode (lower case) we used a flat dispersion $E = 2.6$ meV. In both cases, a Lorentzian cross section with a line width of 0.05 meV was applied. In order to reproduce the observed phonon line shapes calculations were performed for a variety of values of the sample mosaic spread η_s. The curves shown in Fig. 6(b) correspond to $\eta_s = 2.5°$, which value is seen to give an adequate fit to both observed phonon groups. Moreover, this value of η_s is in reasonable agreement with the irregular rocking scan profiles of the nuclear Bragg points. Using $\eta_s = 2.5°$, similar calculations were performed applying a one-dimensional dispersion relation $E(q) = 3.55|\sin q_{c^*} c|$ meV, corresponding to the dCP spectrum (q_{c^*} denotes the component of the wave vector along c^*). A Lorentzian cross section with a line width of 0.05 meV was applied. The calculated line shapes for the three values of q_{c^*} corresponding to the measurements of Fig. 4 are represented in Fig. 7 by the full lines. The intensity scale was adjusted separately in the three cases. Also shown are the experimental points from Fig. 4 after having been subjected to background and resolution corrections similar to those of Fig. 6. One sees that the calculated line shapes are approximately symmetric, and while the fit is reasonably good for $\vec{Q} = (0,0,0.825)$ (ignoring the optical-phonon peak), it is somewhat poorer at $\vec{Q} = (0,0,0.75)$ and totally fails at $\vec{Q} = (0,0,0.60)$. We may therefore conclude that the observed asymmetry in the neutron response of the magnetic excitations cannot be explained by a pure resolution effect but must be caused by a true asymmetry in the cross section. This will be further discussed in Sec. IV.

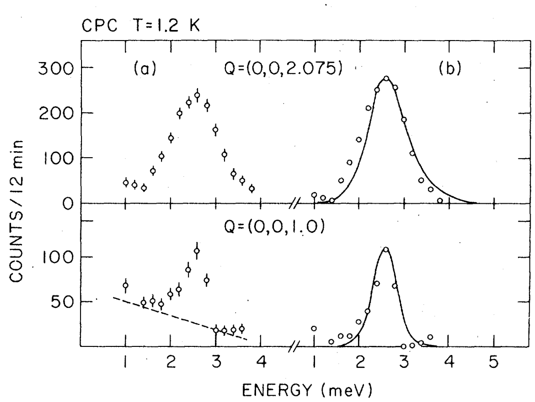

FIG. 6. Phonon groups showing longitudinal acoustical phonon (upper row) and optical zone-center phonon (lower row). (a) shows the raw experimental data while (b) shows the experimental points after background subtraction and resolution correction. The solid curves represent calculated line shapes using Eq. (3) with a sample mosaic spread equal to 2.5°.

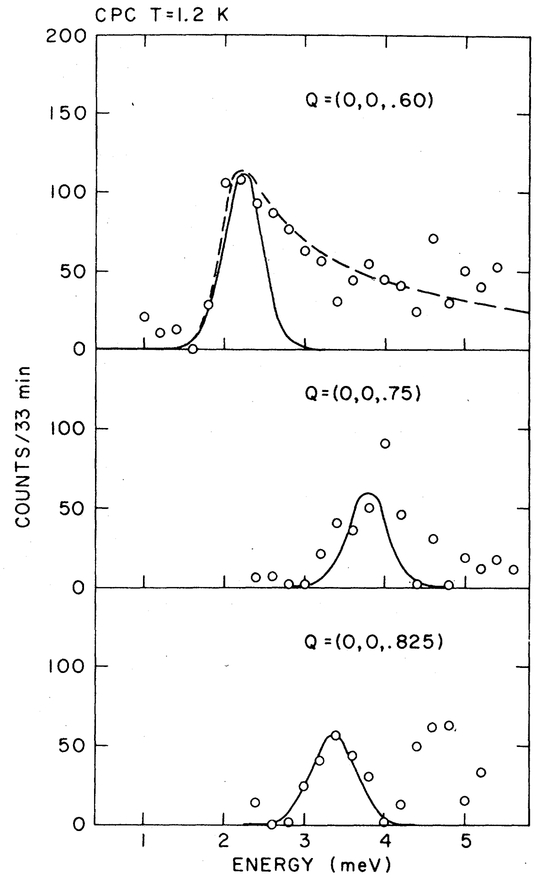

FIG. 7. Experimental points of Fig. 4 after background— and resolution correction. The full curves show the calculated line shapes based on Lorentzian cross sections and a sample mosaic spread of 2.5°. The dashed curve represents the calculated line shape corresponding to the scattering function Eq. (4).

B. Experiments at high magnetic field

In Fig. 8 we have plotted the results of the inelastic scans performed at $H = 0$ and at $H = 70$ kOe, using the superconducting magnet. The temperature is $T = 1.6$ K. Due to the presence of the superconducting magnet it is seen that all these scans suffer from a poor signal-to-background ratio, smearing out the details of the line shapes. The peaks for $\vec{Q} = (0,0,0.625)$ and $(-1,0,0.625)$ at a given field are seen to be at approximately the same positions as expected due to the one dimensionality of the magnetism. When the field is increased from 0 to 70 kOe the peaks exhibit a broadening, most pronounced for $\vec{Q} = (0,0,0.75)$, where the signal is smeared out at high field. This effect stands out clearly in the scans at $\vec{Q} = (0,0,0.75)$ at $H = 0$, 35 kOe, and 70 kOe, plotted in Fig. 9. The two phonons shown in Fig. 6 were remeasured at both zero field and at 70 kOe, showing that the phonon groups were unaffected by the field.

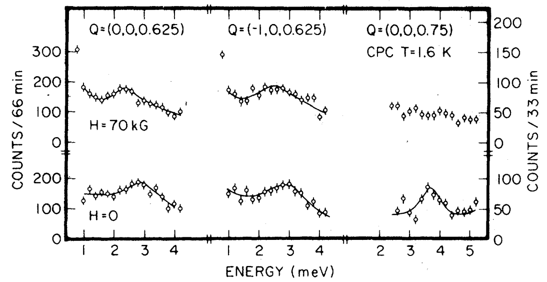

FIG. 8. Constant-Q scans at $T=1.6$ K showing magnetic excitations at zero magnetic field (lower row) and at $H=70$ kOe (upper row). The scans at $\vec{Q}=(0,0,0.625)$ plus the 70 kOe scan at $\vec{Q}=(-1,0,0.625)$ were recorded in 66 min at each point, while 33 min were used for the rest of the scans. Solid curves are guides to the eye.

Thus, the observed broadening of the magnetic neutron responses cannot be due to some field-induced change in the mosaic spread of the sample.

From Fig. 8 it is noted that both scans at $q_{c^*}=0.625$ seem to show a decrease of about 15% in the peak energy in going from 0 to 70 kOe.

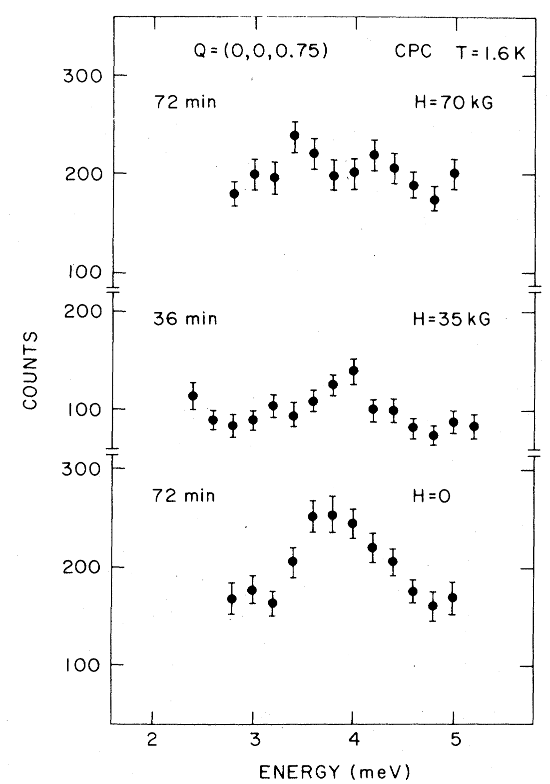

FIG. 9. Inelastic scans at $\vec{Q}=(0,0,0.75)$ and $T=1.6$ K at three values of applied magnetic field.

IV. DISCUSSION

A. Line shape at zero magnetic field

As mentioned in Sec. I the approach of dCP did not allow for a calculation of time-dependent spin correlation functions and thereby inelastic neutron scattering cross sections. It is by no means evident what is the contribution of the dCP states to the scattering function compared to that of the continuum of higher excited states above the dCP spectrum. Several authors have dealt with this problem. Bonner and Sutherland[11] have carried out numerical calculations on a chain containing eight spins ($N=8$). They find that for the wave vector k equal to $\frac{1}{4}\pi$ and $\frac{1}{2}\pi$ the lowest excited states (corresponding to the dCP states in the limit $N\rightarrow\infty$) contain all the spectral weight. However, for $k=\frac{3}{4}\pi$ and $k=\pi$ they find that the higher $S=1$ states do contribute a measurable amount (~10%) to the spectral weight function. Hohenberg and Brinkman[12] have calculated sum rules for the antiferromagnetic Heisenberg chain and have obtained an exact result for the peak value of the spectral weight function in the limit of small k's. This is found to be close to the dCP value.

Another investigation was carried out by Mikeska,[13] who found analytic expressions for spin correlation functions for small values of T/JS^2 and $1/S$. For q values close to an antiferromagnetic Bragg point Mikeska thus obtained in the zero temperature limit the scattering function

$$S(Q,\omega)\propto\frac{1}{1+e^{-\beta\omega}}\frac{1}{(\omega^2-\omega_q^2)^{\alpha}}\Theta(|\omega|-\omega_q)\ , \tag{4}$$

where $\alpha=1-1/\pi\hat{S}$, $\hat{S}=\sqrt{S(S+1)}$ and $\Theta(x)$ is the step function defined as $\Theta(x)=1$ for $x\geq 0$ and $\Theta(x)=0$ for $x<0$. Because of the presence of the step function the scattering function [Eq. (4)] exhibits a strong asymmetry in ω, being zero for $|\omega|<\omega_q$, and decreasing smoothly towards zero above the singular point at $\omega=\omega_q$. This result is in qualitative agreement with those obtained in Refs. 11, 12, and 14. Since, however, the expression [Eq. (4)] is derived assuming a small value of $1/S$, we might expect at most a qualitative application to the present system.[21] We have done this for $\vec{Q}=(0,0,0.60)$ by inserting in Eq. (4) for ω_q the corresponding dCP frequency and subsequently calculated the experimental line shape [Eq. (3)] using a resolution ellipsoid appropriate for the experimental configuration. In doing this we introduced an additional term $\Gamma^{-2\alpha}$ in the denominator of Eq. (4) in order to remove the divergence at ω_q. Putting Γ equal to 0.5 meV, the half-width at half-maximum becomes $\Delta\omega\simeq\Gamma^2/2\omega_q\simeq 0.05$ meV, comparable to the steps of the numerical integration and still much smaller than the resolution width. The resulting line shape is represented by the dashed curve in the upper part of Fig. 7. The agreement with the experimental points is surprisingly good, and demonstrates the step-function behavior of the cross section as predicted by Mikeska. The slope of the line profile at higher energies agrees well with the measurements, but since the data could allow for a range of values of α, this should probably not be taken too literally. Thus, in the light of the obtained experimental results, further theoretical work on the neutron cross section of the present system will be valuable.

B. Magnetic excitations at applied field

As can be seen from Fig. 3, the most striking difference between the field dependence of the dCP energies to the classical case are found for values of k between 0 and $\frac{1}{2}\pi$, that means in the vicinity of the nuclear Bragg points. Unfortunately, this is the range in reciprocal space where the scattering intensity is low. Due to the experimental conditions and the broadening of the peaks, reasonable neutron signals at high fields were obtainable only in the vicinity of the antiferromagnetic Bragg point, using very long counting times. From the calculation of the dCP spectrum at $H = 70$ kOe shown in Fig. 2, it is seen that the expected decrease in the excitation energy from zero field for $Q = 0.625$ is about 10%. In the corresponding classical case, the energy of one spin-wave branch decreases a negligible amount while the other branch increases about 10%, as can be seen from Fig. 3 by tracing out the curve for $h = 0.21$. The observed decrease of $\sim$15% in the peak at $Q_{c^*} = 0.625$ in going from 0 to 70 kOe thus constitutes some support to the calculations of Ishimura and Shiba. Furthermore, this is in explicit contradiction to expectations from classical spin-wave theory. The observed broadening of the neutron response at high field indicates a field dependence of the spectral weight function, tending to enhance the contribution of higher excited states in the presence of applied magnetic field.

V. SUMMARY AND CONCLUSION

The inelastic neutron scattering measurements on CPC carried out in the present work have shown that the strong asymmetry in the experimental line shape of magnetic excitations in the paramagnetic phase at low temperatures and small wave vectors is a demonstration of a true asymmetry in $S(Q, \omega)$ as predicted by Mikeska[13] and others.[11,12,14] At high magnetic field we have observed a broadening of the neutron peaks causing increased errors in the determination of peak positions. At the antiferromagnetic zone boundary, the peak is virtually smeared out at 70 kOe, whereas closer to the antiferromagnetic Bragg point, the experiments seem to produce a 15% decrease in the peak energy. The latter observation is in qualitative agreement with the calculations of Ishimura and Shiba[15] of the H dependence of the spectrum of spin-wave-like states found by dCP,[6] rather than with the results obtained in classical linear spin-wave theory. A more thorough investigation of the effect of applied field on the magnetic excitations of CPC seems to call for a better sample quality, and new attempts to grow one large crystal have been initiated.

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