Neutron scattering study of spin waves in one-dimensional antiferromagnet $KCuF_3$

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Inelastic neutron scattering has been used to obtain the spin-wave spectrum in $KCuF_3$. The nature of magnetic interactions in this material is quasi-one-dimensional (chainlike) as evidenced by a highly anisotropic spin-wave dispersion. The spin-wave dispersion along the chain direction fits very well with the exact solution for an $S=\frac{1}{2}$ linear-chain antiferromagnet given by des Cloizeaux and Pearson. Using linear spin-wave theory, the ratio of interchain to intrachain magnetic exchange parameters J_a/J_c is found to be -0.01 ± 0.001. A zone-center gap of 1.1 meV in the spin-wave mode with a spin component out of the easy (xy) plane shows that the spin system is predominantly of Heisenberg nature with a small (0.02%) xy-like component.

I. INTRODUCTION

The realization of many compounds which display nearly one-dimensional magnetic properties has given rise to a great deal of experimental and theoretical work in this area in the past few years.[1] In most such materials, the interchain exchange interaction is very weak, due to the presence of large nonmagnetic ions which separate the chains. Several years ago, Hirakawa *et al.*[2] pointed out that $KCuF_3$ displays properties characteristic of one-dimensional antiferromagnets. The one dimensionality in $KCuF_3$ is, however, attributed to the distortion of octahedral environment of the Cu^{2+} ions due to Jahn-Teller effect. This distortion leads to a spatial alignment of the $3d$ orbitals in Cu^{2+} ions, giving rise to a strong superexchange interaction J_c between nearest-neighbor ions along the c axis, which we refer to as the chain axis. In the direction perpendicular to the chain axis, the exchange interaction J_a is very small because of the poor overlap of the orbitals even though the distances between magnetic atoms is essentially the same along all principal axes. This spatial overlap of orbitals is depicted in Fig. 1 for both (a) and (d) polytype structures of $KCuF_3$.

In a previous neutron-diffraction work, Hutchings *et al.*[3] did not find any direct evidence for one-dimensional order above the Néel temperature $T_N=38$ K where three-dimensional ordering occurs. Ikeda and Hirakawa[4] have, however, confirmed the one-dimensional nature of $KCuF_3$ above T_N, from quasielastic neutron scattering experiments. They estimate that even well above the Néel temperature the spin-correlation length in the chain direction is of the order of 500 Å, with interchain correlation length being considerably smaller. The ratio of interchain to intrachain coupling was estimated to be about 0.027 by Ikeda and Hirakawa. The strong exchange interaction along chain axis was found to be of the order of -190 K from magnetic-susceptibility measurements[2] using the theory of Bonner and Fisher[5] for one-dimensional (1-D) antiferromagnets with spin $S=\frac{1}{2}$.

An intersting feature of the present system is the spin $S=\frac{1}{2}$ and hence the quantum nature of the system. The dispersion relation for the excited states of a 1-D Heisenberg antiferromagnet with $S=\frac{1}{2}$ was first calculated by des Cloizeaux and Pearson[6] (dCP) to be

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

where J_c is the nearest-neighbor (nn) exchange interaction and c is nn separation. This spectrum is

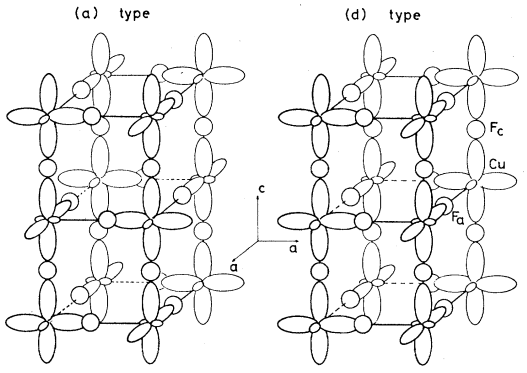

FIG. 1. Schematics of type (a) and (d) structures observed in $KCuF_3$.

qualitatively similar to the linear spin-wave-theory result for classical spins, except that the coefficient of J is equal to 2 for classical spins, as compared to π for dCP states. The spin-wave spectrum of a classical, $S=\frac{5}{2}$, linear-chain antiferromagnet, $(CD_3)_4NMnCl_3$ (TMMC), has been extensively studied by Birgeneau *et al.*[7] A striking confirmation of the dCP spectrum for an $S=\frac{1}{2}$ system was presented by Endoh *et al.*[8] in their inelastic neutron scattering experiments on $CuCl_2{\cdot}2N(C_5D_5)$ (CPC). It is clearly of interest to study if a similar dCP spin-wave spectrum is shown by $KCuF_3$ also. In this paper we report the results of inelastic neutron scattering experiments on $KCuF_3$. The experimental results show that spin-wave spectrum of this compound below the Néel temperature is highly anisotropic, thereby confirming the quasi-one-dimensional nature of the magnetic interactions. Using linear spin-wave theory, we have obtained quantitative values for intrachain and interchain coupling constants. The plan of the paper is as follows. In Sec. II we discuss the crystal structure and experimental techniques and establish the polytype structure of the present crystal. Section III gives the expected dispersion relations based on linear spin-wave theory. In Sec. IV we present and discuss the experimental results in the light of dCP and linear spin-wave-theory results. We summarize our results in Sec. V.

II. CRYSTAL STRUCTURE AND EXPERIMENTAL TECHNIQUE

The sample used in the present study was a single crystal of an approximate size $1.5\times1\times0.5$ cm^3 grown by Bridgman techniques. The crystal had a mosaic spread of less than 10 min. The presence of two polytype structures for $KCuF_3$ was shown by Okazaki using x-ray diffraction.[9] The two polytypes are characterized as (a) type and (d) type as shown in Fig. 1. In each structure the fluorines are displaced from the midpoint of the Cu^{2+}–Cu^{2+} bond in the *ab* plane. As we show below, the structure for the crystal used in the present study is predominantly (a) type.

An *A*-type ordering pattern of the spins in the 3-D magnetically ordered phase was established for both the (a)- and (d)-type crystals by Hutchings *et al.*[3] In *A*-type spin ordering, antiferromagnetic *c* chains are ferromagnetically ordered in the *ab* plane, giving a tetragonal magnetic unit cell with sides of length $(a,a,2c)$.

The neutron scattering experiments were conducted on a triple-axis spectrometer at the Brookhaven High Flux Beam Reactor. A portion of the experiments were also performed using the ISSP neutron diffractometer located at JRR-2, Japanese Atomic Energy Research Institute, Japan. At Brookhaven, pyrolytic graphite was used for both the monochromator and analyzer. Because of the steepness of the dispersion curve in the *c* direction, both constant-Q and constant-E scans were made with initial neutron energies varying between 5 and 70 meV. Various combinations of collimators were used in the experiments, depending on the scattered neutron intensity and resolution requirements. A tuned pyrolytic graphite filter was used to remove contamination from higher-order scattered neutrons. The crystal was wrapped in aluminum foil and mounted in a helium gas-filled container. The scattering plane for all the measurements was $(h0l)$.

The present sample showed weak nuclear Bragg reflections of the type 201, indicating a doubling of unit cell along the *c* direction and hence a predominantly (a)-type structure. Predominant (a)-type structure was also established from the measurement of $(001)_M$ magnetic Bragg-scattering intensity which is shown in Fig. 2. From the peak intensity of the $(001)_M$ Bragg point, we determine T_N to be 39.8 ± 0.1 K, which is in fair agreement with the value of 38 K for (a)-type structure by Hutchings *et al.* Hutchings *et al.* also estimated the Néel point of the crystal with (d)-type structure to be ~22 K. We observed a slight discontinuity in the $(001)_M$ intensity curve at ~23 K as shown in the figure. From the discontinuity we estimate that the present crystal contains $1(\pm0.5)\%$ of the (d)-type structure. The lattice con-

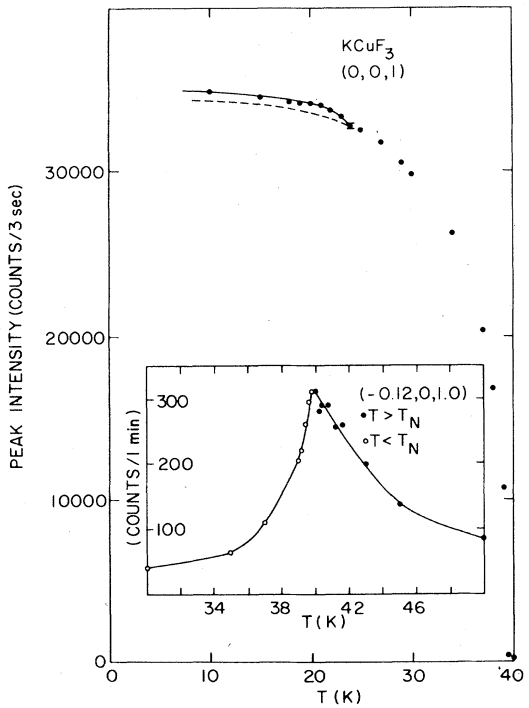

FIG. 2. Temperature variation of $(001)_M$ intensity of $KCuF_3$. The inset shows the variation of critical scattering intensity at $(-0.12,0,1)$ as a function of temperature.

stants for the crystal at 10 K determined by neutron scattering $a=4.126$ and $c=3.914$ Å are in very good agreement with those measured by Hutchings *et al.*[3] The reciprocal lattice used in this study is based on a unit cell which is doubled in the c direction, $c=2c_0$. Also shown in the inset in Fig. 2 is the temperature variation of magnetic critical scattering at the $(-0.12,0,1)$ position in reciprocal space. The Néel temperature determined from these measurements is also 39.8 ± 0.1 K.

III. DISPERSION RELATIONS

As already mentioned, $KCuF_3$ orders in a type-A pattern, which implies an xy-like anisotropy in the spin system. For an xy-like antiferromagnet, neutron inelastic scattering experiments should exhibit two distinct contributions. An in-plane component (IPC) due to fluctuations of S_k^x and S_k^y and an out-of-plane component (OPC) due to fluctuations of S_k^z. For scans near the antiferromagnet Bragg point $[001]_M$ only IPC modes would contribute to the inelastic scattering, whereas, in scans near $[201]_M$ or $[401]_M$, one should see both the IPC and OPC modes.

If, above the Néel temperature, $KCuF_3$ displays properties characteristic of 1-D magnetic materials, then, above T_N one expects spin-wave dispersion only along the chain axis, due to short-range correlations along the chain. For spin-$\frac{1}{2}$ Heisenberg antiferromagnetic linear chain, the dispersion relation is given by Eq. (1). If we include the effects of weak interchain coupling, we can write the Hamiltonian of the anisotropic Heisenberg exchange system to be of the form

$$H=2\sum_{l,m}(J_{lm}\vec{S}_l\cdot\vec{S}_m+D_{lm}S_l^zS_m^z) -2\sum_{ll'}J_{ll'}\vec{S}_l\cdot\vec{S}_{l'}-2\sum_{mm'}J_{mm'}\vec{S}_m\cdot\vec{S}_{m'}\ , \quad (2)$$

where the summation is over pairs, with indices l,l' running over a sublattice, m,m' over the b sublattice. $J_{lm}(=J_c)$ represents the antiferromagnetic intrachain interaction among nearest-neighbor (nn) ions on different sublattices and $J_{ll'}=J_{mm'}(=J_a)$ is the ferromagnetic interchain interaction among nn ions on the same sublattice. The term $D_{lm}(=D_c)$ produces the xy anisotropy.

According to linear-classical spin-wave theory,[10,11] the Hamiltonian (2) gives the following spin-wave dispersion:

$$\hbar\omega(q)=2S(\{Z_aJ_a[1-\gamma(q)]+Z_cJ_c(1\mp\gamma_c(q))\}\{Z_aJ_a[1-\gamma_a(q)]+Z_cJ_c[1\pm\gamma_c(q)\mp Z_cD_c\gamma_c(q)]\})^{1/2}\ , \quad (3)$$

where

$$\gamma_a(q)=\tfrac{1}{2}(\cos q_aa+\cos q_ba)\ ,$$

$$\gamma_c(q)=\cos q_cc\ ,$$

and

$$Z_a=4,\quad Z_c=2\ .$$

The lower sign in Eq. (3) corresponds to modes with a spin component out of the easy (xy) plane (OPC modes) and the upper sign corresponds to modes with spin fluctuations in the easy plane (IPC modes). The spin-wave theory thus predicts a zone-center mode of finite energy for the OPC mode. In the event that there is an Ising-like anisotropy in the xy plane, the Hamiltonian (2) will have to be modified in such a way as to produce an energy gap at the zone center for the IPC mode also. For a linear antiferromagnetic chain with isotropic Heisenberg interaction, $J_a=D_c=0$, and relation (3) reduces to the classical spin-wave dispersion formula

$$\hbar\omega(q)=2(J_c\sin qc)\ . \quad (4)$$

IV. EXPERIMENTAL RESULTS

The spin waves in $KCuF_3$ were investigated via their excitation, the neutrons losing energy to the system. Representative magnon groups along q_c and q_a at 10 K are shown in Figs. 3 and 4. The points in the figures are experimental counts, the solid lines are the best fit convolution of the cross section with the resolution function of the instrument assuming the dispersion relation given by Eq. (3). In the constant E scans the fitted spectra agree very well with the experimental ones except around $q=0$.

Temperature variation of the magnon modes was investigated only qualitatively. The results indicate that spin-wave scattering along q_c exists well above the T_N of 38 K. One such constant-E scan at 60 K is also shown in Fig. 3, which demonstrates the presence of short-range correlations along the chain direction above T_N. These persistent short-range correlations are characteristic of quasi-one-dimensional materials. There is no change in the peak position of the magnon modes along the chain direction up to 60 K, although the peaks are found to broaden considerably.

The dispersion relation for the excitations is shown in Fig. 5. We first discuss the dispersion along q_c. In this direction, the fitted dispersion follows a sine curve as predicted by Eq. (1) with $J_c=203\pm2$ K. This value of J_c is in very good agreement with the value of 190 K, obtained from susceptibility measurements fitted by using the Bonner and Fisher theory.[2] In Fig. 6, we have shown the dispersion relation

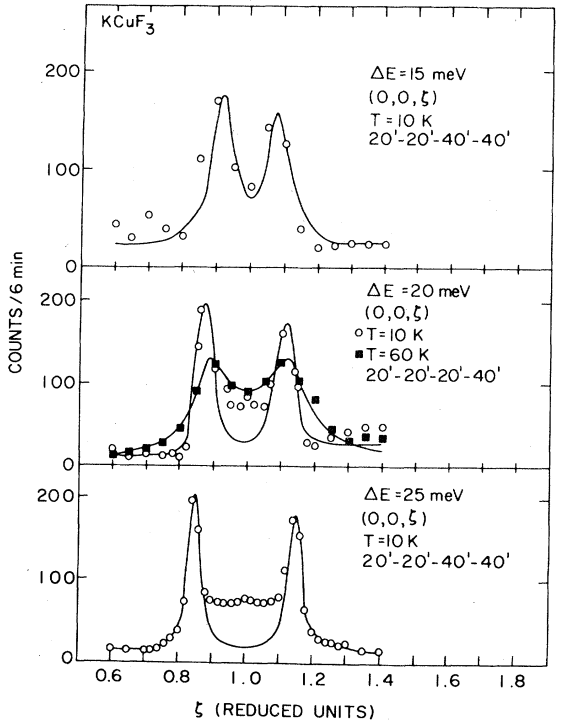

FIG. 3. Intensity observed in constant-E scans in the [001] direction at various energies. The lines drawn are fits to experimental spectra as described in the text, except that the line through 60-K scan is only drawn as a guide to the eye.

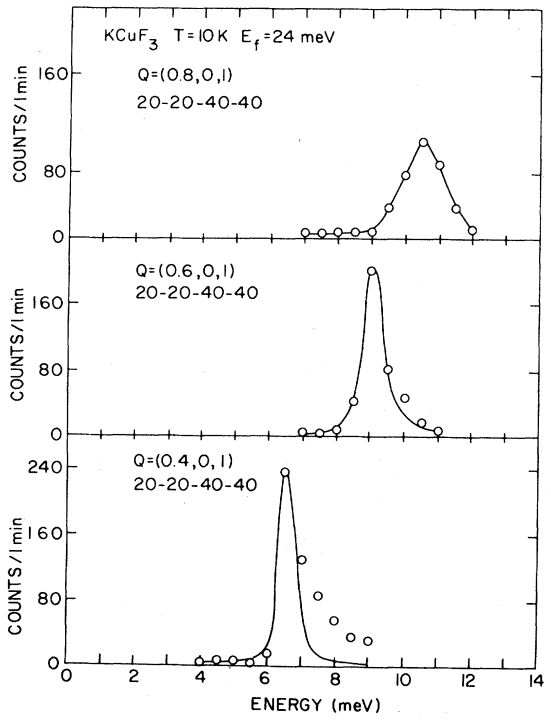

FIG. 4. Constant-Q scans at various points along the direction perpendicular to the chain axis. The solid lines are best fit to the data.

along q_c obtained from magnon modes measured at various different values of q_a, the momentum component perpendicular to the chain axis. One clearly sees that, at least above 15 meV, the spin-wave excitations along q_c are purely 1-D in character. Thus $KCuF_3$ is the second example, together with CPC, in which dCP result for a quantum system of spin $\frac{1}{2}$ is in excellent agreement with the experimental results.

We would, however, like to mention that the fit to dCP spectrum clearly depends on how the value of J_c was obtained from other experimental measurements, e.g., magnetic susceptibility. Magnetic-susceptibility data for $KCuF_3$ can be fitted by using two different theories. Hirakawa *et al.*[2] have used the numerical results of Bonner and Fisher for the spin $S=\frac{1}{2}$ system and obtain a value for $J_c=190$ K, as already mentioned. The susceptibility data can also be fitted to the Fisher results for linear chain of classical spins.[12] The fit, while not as good as with the Bonner and Fisher results, is qualitatively satisfactory. This fitting gives the value of $J_c \equiv 320$ K. If one fits the experimental dispersion along q_c in $KCuF_3$ using classical spin-wave formula, Eq. (2), the value of J_c comes out to be 318 ± 3 K, in excellent agree-

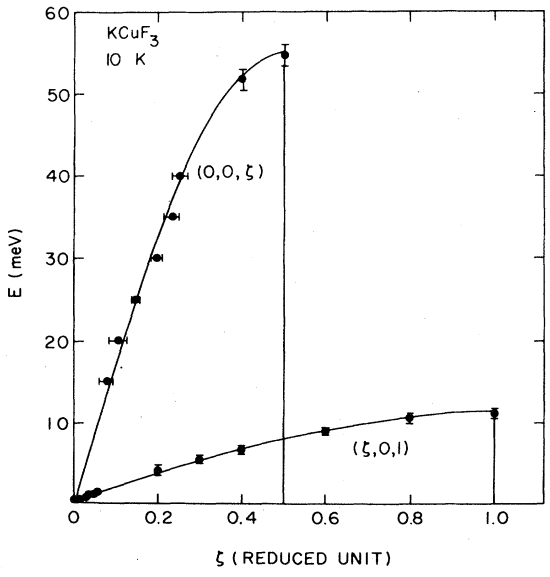

FIG. 5. Magnon dispersion in $KCuF_3$ along q_c and q_a at 10 K. The solid line is a fit to the data as described in the text. ζ is the reduced wave vector.

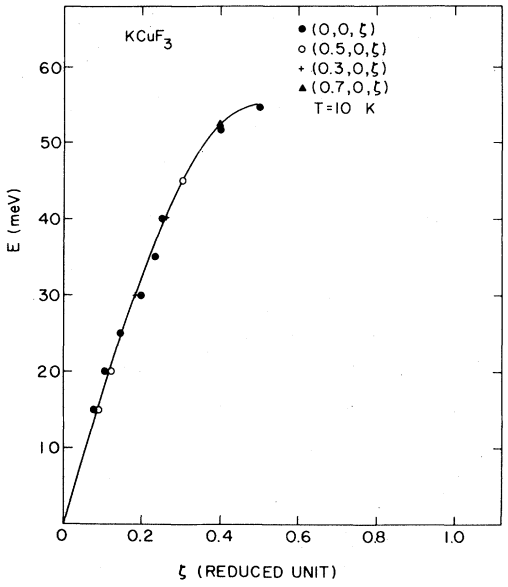

FIG. 6. Magnon dispersion in $KCuF_3$ along q_c at various values of q_a demonstrating the predominant one-dimensionality of the material.

ment with the above value of J_c! Thus, on the basis of neutron scattering alone, it is difficult to say unequivocally that one is observing dCP spectrum and hence the quantum nature of the system. An independent and accurate measurement of the exchange parameter is, therefore, necessary. It is of interest to mention that a similar situation occurs in the case of CPC.

Experimentally measured spin-wave dispersion along q_a at 10 K is also shown in Fig. 5. For a pure 1-D system with negligible interchain coupling, one should not observe any dispersion in this direction, whereas the data of Fig. 5 clearly show such dispersion. The solid line through the experimental points in the figure is a fit to the dispersion relation given by Eq. (3). The ratio of interchain to intrachain coupling constants given by such a fit is $J_a/J_c = 0.01 \pm 0.001$. Not shown in Fig. 5 is the magnon energy at the (110) Brillouin-zone corner in reciprocal space. The experimentally measured value for this mode is 14.5 ± 0.5 meV. This also gives a J_a/J_c ratio in very good agreement with the above value. If one uses the ratio J_a/J_c as the criterion, then it seems that $KCuF_3$ is a fairly good 1-D material even though the dispersion of the low-energy spin waves perpendicular to the chain axis is substantial.

We are not aware of existing calculations of the spin dynamics of 3-D spin-$\frac{1}{2}$ systems. In the absence of theoretical guidance, we have chosen to modify the classical results in the most obvious way to make them consistent with spin-$\frac{1}{2}$ 1-D results, by simply replacing 2 by π in the dispersion given by Eq. (3) and apply it to a 3-D spin-$\frac{1}{2}$ system. If one does this for $KCuF_3$, assuming J_c to be 203 K, the value of J_a comes out to be 2.0 ± 0.2, in good agreement with a value of 3 K predicted by theory of Oguchi.[13] It is not clear to us, however, that this procedure is legitimate, and J_a may thus be in error by a factor of order $2/\pi$.

We have studied the temperature dependence of the energy gap at the zone corner (1,0,1) as a function of temperature to determine if spin-wave scattering becomes predominantly one-dimensional (i.e., if the gap vanishes) as one approaches the Néel temperature. The spin-wave energy at this point decreases from 11.0 ± 0.5 meV at 10 K to 9.8 ± 0.8 meV at 40 K. Because of the increasing linewidth with temperature, it was not possible to observe this mode above 40 K. However, it is quite clear that the value of this energy gap does not go to zero at the Néel temperature, which indicates that three-dimensional effects are prevalent at least up to a few degrees above the 3-D ordering temperature in this material.

We now turn to spin-wave scattering at very small $\vec{q}$ vectors. It can be easily seen from Eq. (3) that depending upon the value of anisotropy D_c, the IPC and OPC modes of spin waves will differ in energy at small $\vec{q}$ vectors and will be nearly degenerate at higher $\vec{q}$ vectors. Figure 7 shows several magnon groups at small wave vectors. Scans around $(001)_M$

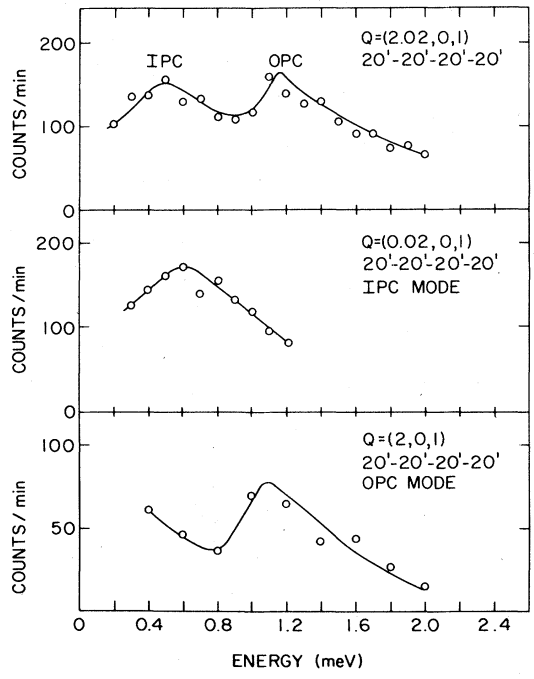

FIG. 7. Various constant-Q scans close to the antiferromagnetic Bragg point displaying the IPC and OPC spin-wave modes. The lines are drawn to guide the eye.

Bragg point display only contribution from IPC modes, whereas around $(201)_M$ the scattering from OPC modes also contributes to the spectrum. In particular, the magnon groups at (2.02,0,1) clearly show contribution from both the in-plane and out-of-plane components of the spin wave. In Fig. 8, we show in enlarged form, the spin-wave dispersion at small wave vectors along q_a for both the IPC and OPC modes. Due to very steep dispersion along q_c, it is not possible to measure the dispersion at small wave vectors along this direction. It is worth mentioning that all the scans in Fig. 8 correspond to a reduced wave-vector component along q_c^* equal to zero. The two branches of excitations probably merge at $q_a^* \equiv 0.2$ or higher; however, it was not possible to resolve the two branches at scans around $(201)_M$ for q_a greater than 0.08. The solid lines in Fig. 8 are calculated with $J_c = 203$ K, $J_a = 2$ K, and $D_c = 0.04$ K and by replacing 2 by π in Eq. (3). The experimental points have been corrected for small shifts due to instrumental resolution. Thus we estimate the anisotropic exchange interaction (xy-like) to be 0.02% of the major Heisenberg exchange interaction. This is much smaller than the theoretical estimates for anisotropy, which predict[14] that D_c should be of the order of $(g-2)^2 J$.

We have not been able to resolve clearly the IPC mode along q_a at wave vectors very close to zero. Thus, it is not possible to say, unequivocallly, whether there is a gap in this mode at $q = 0$. However, from the measurements of this mode at $q_a > 0.015$, we do not detect any flattening of the dispersion curve which might indicate a substantial gap at $q = 0$. As mentioned earlier, according to the estimates of Ikeda and Hirakawa from elastic neutron scattering experiments, the zone-center gap in the IPC mode should be of the order of 0.1 K, much too small to be observed by an inelastic neutron scattering experiment. From our measurements, one can put an upper limit on the value of the IPC mode gap to be 0.2 meV. It is also not possible to fit the dispersion curve of the IPC mode along q_a at both $q = 0$ and at zone boundary, using Eq. (3). As shown in Figs. 7 and 8, assuming the value of J_a to be equal to 2 K gives a satisfactory fit at higher q values, while overestimating the frequencies of the modes close to zone center. This indicates that simple linear spin-wave theory cannot describe the magnon spectrum in this case.

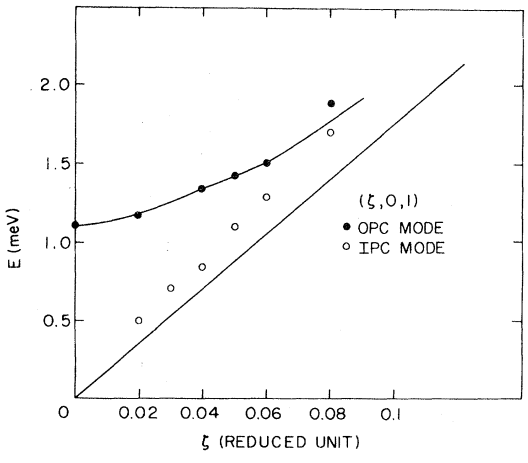

FIG. 8. Spin-wave dispersion close to zone center in the q_a direction, for OPC and IPC magnon modes in $KCuF_3$.

Finally, we discuss the possible source of extra intensity observed near $q = \pi$ shown in some of the scans in Fig. 3. It has been shown by several authors[15] that, for the case $S = \frac{1}{2}$ dCP excitations form a lower boundary to a continuum of higher-lying triplet states. Recent finite-chain calculations by Müller *et al.*[16] show that all the states of this continuum have nonzero spectral weight. Contribution from the continuum states becomes more pronounced around the antiferromagnetic superlattice peaks, i.e., for $q \simeq \pi$, but is not observable for small $q \simeq 0$. As shown in Fig. 3, the constant-E scans at 20 and 25 meV have considerably more intensity around $q = \pi$ than can be accounted for only from the dCP states. The scan at 15 meV, however, can be fitted to a pair of Lorentzians with appropriate width and shows no anomalous increase in the cross section at $q = \pi$. We would, however, like to point out that the peaks in 15-meV scan are not very well resolved and that even if extra intensity is present at $q = \pi$, the fitting program can apparently account for it by simply increasing the width of the Lorentzian peak. Thus it is possible that the enhanced cross section at $q = \pi$ in scans at 20 and 25 meV is due to quantum effects in this $S = \frac{1}{2}$ system. This effect has been observed by Heilmann *et al.*[18] in the case of CPC. Further experimental work, especially scans in the vicinity of nuclear Bragg points are needed to verify this conjecture.

V. SUMMARY AND CONCLUSIONS

In this study we have investigated magnetic inelastic scattering from spin waves in $KCuF_3$. Our measurements show this to be a quasi-one-dimensional antiferromagnet in which the spin-wave dispersion along the chain direction agrees very well with the des Cloizeaux and Pearson[6] result for the excited eigenstates of a linear antiferromagnetic chain with $S = \frac{1}{2}$. The ratio of interchain to intrachain coupling constants J_a/J_c is found to be 0.01 and there is no spin-wave dispersion perpendicular to the chain direction above the energies of 15 meV. It seems, therefore, that $KCuF_3$ would seem to be a very good 1-D magnet as far as the high-temperature-susceptibility

measurements are concerned. In fact, susceptibility measurements confirm $KCuF_3$ to be an excellent 1-D material. The neutron scattering experiments have shown that there is sizable 3-D character in the low-energy spin waves due to the presence of interchain coupling constant of order 2 K.

A zone-center gap of 1.1 meV in the OPC mode of the spin wave shows that the spin system is predominantly of Heisenberg nature with $\sim 0.02\%$ of the interaction being xy-like. The in-plane anisotropy in $KCuF_3$ is estimated to be less than 0.2 meV.

It is of interest to mention that $KCuF_3$ is a possible system for confirmation of some of the quantum effects recently predicted for $S=\frac{1}{2}$ system by various authors.[15,17,18] Thus, a thorough study of temperature and magnetic-field dependence of spin waves should reveal many interesting results about the quantum nature of magnetic systems in general.

After the preparation of this manuscript, we learned of a recent neutron scattering study by Hutchings *et al.*[19] Hutchings *et al.* have not been able to cover as large a range in wave vector $\vec{q}_c$ as was done in the present study. However, the agreement between the two measurements, wherever they overlap, is very good. By converting the Hutchings *et al.* values to proper units of the present study, the agreement between J_c and J_a is within 2%. The values of anisotropic constants D_c differ considerably: $D_c=0.04$ K compared with the value of 0.023 K obtained by Hutchings *et al.* The value of D_c in the present study should be more accurate since we were able to observe the OPC mode directly at $q_c=0$.

ACKNOWLEDGMENT

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