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Renormalization of Large-Wave-Vector Magnons in Ferromagnetic $CrBr_3$ Studied by Inelastic Neutron Scattering: Spin-Wave Correlation Effects*

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The renormalization of large-wave-vector magnons in $CrBr_3$ has been carefully studied using inelastic neutron scattering. The results are in disagreement with the first-order approximation to Dyson's spin-wave theory, which predicts that the spin-wave energy shift $\Delta E/E$ is independent of wave vector. Over part of the Brillouin zone our data can be fitted to a constant $\Delta E/E$. However, in the region $q=0.4$ to 0.6 Å^{-1} in the Δ direction, there is strong evidence for a wave-vector dependence of $\Delta E/E$, such as would arise from the higher-order spin-wave interactions, i.e., spin-wave correlation effects. Qualitative comparison is made to the predictions of t-matrix calculations, which treat the interactions to all orders (and are thus capable of reproducing Dyson's results). The observed q dependence agrees with these (qualitative) predictions, giving evidence for the direct observation of the effect of correlations between spin waves. Unfortunately, quantitative comparisons between theory and experiment are not possible at present, because of the difficulty of calculating the two-spin-wave t matrix for this system.

I. INTRODUCTION

$CrBr_3$ is an insulating ferromagnet[1] with $T_c = 32.5$ °K. It has been widely studied as a prototype of the Heisenberg model, both by thermodynamic and resonance techniques,[2–7] and recently by inelastic scattering of neutrons.[8] In the neutron experiment the magnon spectrum was measured both along the (weakly coupled) c axis and in two different symmetry directions within the (strongly coupled) c plane. The renormalization of the thermodynamically accessible (low-energy) spin waves was also measured, and a direct verification of Dyson's theory[9] for the energy shift was made. However, in a neutron experiment, in contrast to thermodynamic measurements, the higher-energy (nonthermal) magnons are also accessible, and it has been shown theoretically[10] that the nonthermal (large-wave-vector) spin waves show strong interaction effects.[11] Thus a careful study of the renormalization of the high-energy spin waves in $CrBr_3$ may yield additional interesting information about the nature of the spin-wave interaction. In particular, since these magnons interact strongly, their renormalization may be sensitive to the detailed correlations between spin waves.

The Heisenberg ferromagnet is a system in which the correlation effects can be calculated rigorously. At low temperatures, where one may utilize the low-density expansion derived by Dyson,[9] the correlations between spin waves are straightforwardly treated by calculating the two-spin-wave t matrix[10] and from it the effective dressed interaction. The result of such a calculation is the prediction that the spin-wave energy shift is not merely proportional to the excitation energy, but has a wave-vector dependence which reflects the density of two-spin-wave states.

Generally, the energy shift is small, and in cubic systems it is difficult to distinguish experimentally between Dyson's theory and the random-phase approximation (RPA). However, it was shown in Ref. 8 that the spin-wave renormalization in $CrBr_3$ cannot be described by RPA. The renormalization of the magnons in the c plane and along the c axis differs considerably from the RPA theory. In addition, the renormalization seems to show the directional dependence predicted by first-order spin-wave calculations.[4,12] Unfortunately, the experimental uncertainties were sufficiently large as not to permit a totally unambiguous statement about the detailed agreement with the theory. Nevertheless, it seems that $CrBr_3$ is one system in which it may be possible not only to test Dyson's theory in the lowest order (Hartree-Fock approximation), but also to observe effects due to the higher-order interactions (repeated scattering), i.e., those resulting from the correlated motions of the particles.[9,10] We have made a careful study of the magnon dispersion in $CrBr_3$ in two different directions at two temperatures and have some evidence that these correlation effects have been seen.

II. EXPERIMENTAL CONSIDERATIONS

The $CrBr_3$ sample used in these measurements is the same as that used in the previous neutron scattering study.[8] It was grown from chromium metal powder, in a Br_2 atmosphere, in a closed quartz tube furnace at 850 °C. It consists of three intergrown single crystals with a common [111] c axis. Fortunately, the orientation of these crystals is such that two of the three portions have virtually identical orientations. These were aligned with [11$\bar{2}$] axis vertical and the third portion then had its [1$\bar{1}$0] zone axis vertical. The Brillouin zones for these crystals are shown superimposed in Fig. 1. For small wave numbers the magnons in these two zones are truly degenerate (Fig. 2); at large wave numbers they are well separated in energy. Only in the intermediate region does the multidomain structure present any measurement problems.

These measurements were made at the Brookhaven High Flux Beam Reactor on a triple-axis spectrometer using pyrolytic-graphite monochromators and analyzers. Incoming neutron energies of 14 and 38 meV were chosen to provide the best focusing of the spin waves being studied. For 14-meV incoming neutron energy, the beam was passed through a tuned pyrolytic-graphite filter to remove higher-order contamination. The $\lambda/2$ component of the incoming neutron beam was reduced

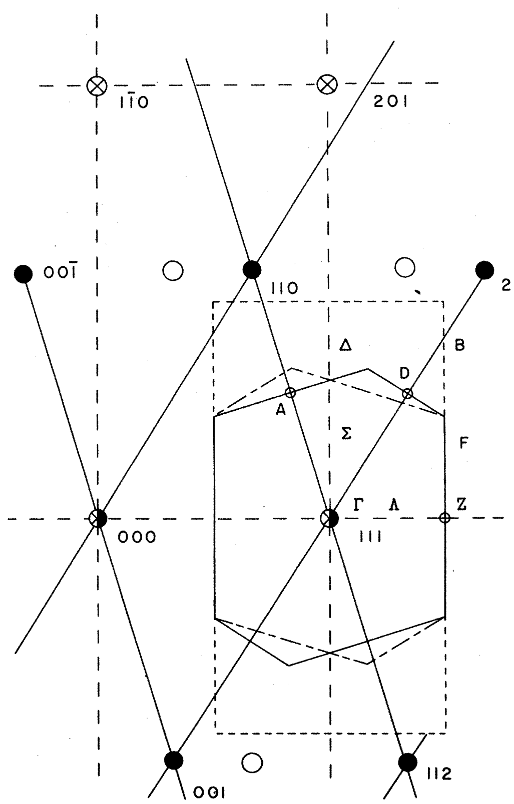

FIG. 1. The superposition of the (1$\bar{1}$0) and the (11$\bar{2}$) planes of reciprocal space, corresponding to the two crystal orientations present in the sample. The Σ direction of one crystal coincides with the Δ direction of the other. The directions studied are Λ ([111] or c axis), Δ [in the (11$\bar{2}$) plane through Γ] and Σ [in the (1$\bar{1}$0) plane through Γ]. For a complete description of the structure and behavior of $CrBr_3$ see Ref. 8.

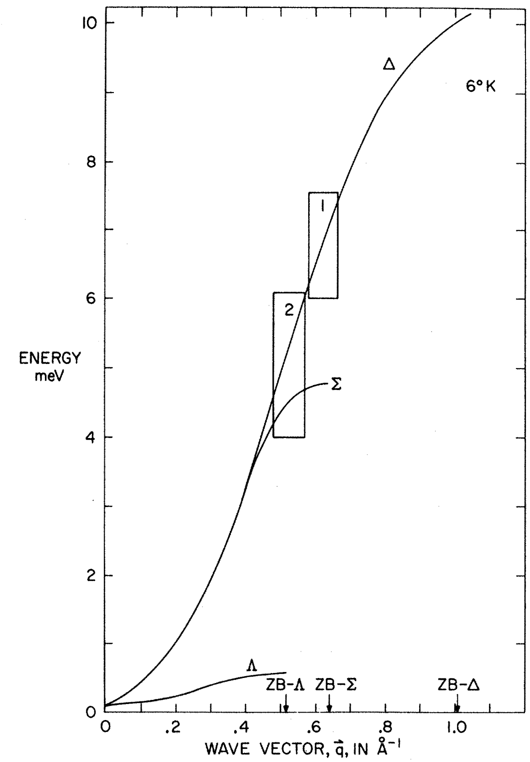

FIG. 2. The dispersion curves for the Λ, Δ, and Σ directions (from Ref. 8) at 6 °K. The region designated (2) is the portion of the fitted dispersion curve (Fig. 6) studied with the 14-meV incoming neutron energy, while that designated (1) was studied with 38 meV.

in this way to 1/1000 of its incident intensity, while the principal component was reduced by only about 30%. Measurements were made at (6 ± 0.1) and (20 ± 0.1)°K. The Curie temperature of the sample was (32.5 ± 0.5)°K, and because the system is nearly two dimensional, the spin waves were still quite well defined at 20 °K,[8] although there is measurable broadening. At 6 °K the magnon linewidth is less than the instrumental resolution.

The temperature control was not a critical parameter, since the rate of change of the magnon energy with temperature is quite small.[8] We were able to make some scans at one temperature, change temperature (for example, for refilling the Dewar), and return to the measuring temperatures without any evidence of a resulting increase in the scatter of the data.

Since the sample is "soft," particularly along the c axis, showing a considerable temperature dependence of the lattice constant, it was quite important to check the alignment whenever the temperature was changed. In particular, the lattice expanded (or contracted) and tended to twist somewhat, and it was necessary to give the sample adequate time to reach equilibrium and to adjust for the changes, in order to assure that the correct wave-vector magnons were being studied at each temperature.

It was necessary, also, to check the Bragg reflections from the crystal often in order to be certain that the spectrometer angles were remaining correct, to within their minimum uncertainty, 0.01°. Several runs were made: a preliminary set of measurements which gave some indication of a q-dependent renormalization, and two runs studying carefully a small portion of the magnon dispersion curve. The first of these, at the 38-meV incoming energy, was used to study the region of the Δ direction for which $q \geq 0.579$ Å^{-1} (Fig. 2). In this region, the Δ magnons are well focused, i.e., the slope of the resolution ellipse nearly coincides with the slope of the magnon dispersion curve, resulting in a small instrumental linewidth,[13] and this branch is reasonably well separated from that corresponding to the Σ direction. For wave vectors < 0.57 Å^{-1} the Δ and Σ branches overlap to a large extent, making it impossible to determine the energies accurately with the 38-meV incident energy. This run was itself divided into cycles at 6 and 20 °K since initially a few points were measured and then, later, other points were added, making necessary temperature cycling. There is no evidence, from the scatter in the data points, that any systematic errors were introduced in this manner.

Similar measurements were made with the 14-meV incoming energy in the region $q < 0.579$ Å^{-1}. Although the Δ branch was not as well focused at this energy the Σ branch was better focused and the two branches were clearly distinct, due to the narrower energy resolution, down to about 0.5 Å^{-1}. Unfortunately, it was not realized until all the data had been analyzed that this particular region of the dispersion curves was of maximum interest, or a few more data points might have been taken with this incident energy.

The intrinsic line shape of a magnon is Lorentzian,[9,10] and the neutron resolution function is Gaussian.[13] When the intrinsic width of the magnon is small compared to the resolution width ($T \ll T_c$), the observed line will thus be a Gaussian. For intrinsic width large compared to the resolution width a Lorentzian line should be observed. Consequently, in order to determine the magnon energies accurately, the neutron scattering peaks at 6 °K, where the broadening is negligible, were fitted to Gaussians. At 20 °K, the data were fitted to both forms. The peak position was the same for each form, but the Lorentzian clearly defined the shape better and put more realistic standard deviations on the parameters, and thus was accepted.

At 6 °K, because of the sharp peaks and the wide energy steps used in the measurements, the peaks were defined by relatively few points, as illustrated in Fig. 3. Therefore, the calculated uncertainty in the peak position was increased to include a contribution equal to one-half the error which would result from the analyzer being misset by the minimum error, 0.01°.

The peaks measured at 20 °K (Fig. 3) were broader than those measured at 6 °K and this analyzer uncertainty was not significant, since there were more measured points defining the peak, so that random misset errors tended to be averaged out. Initially, all of the peaks were fitted without any constraints on either the height or the width of the Gaussian or Lorentzian. This procedure may be susceptible to error if the peaks are not defined by enough points. Therefore, the peaks were refitted with the width constrained (for the 6 °K data to approximately the resolution width), and the height constrained to a smoothly varying function of energy. It was found that with no exceptions for the 6 °K data and only one exception for the 20 °K data, the peak position calculated for both methods was the same to within the standard deviation of the peak, and in most cases the same to within 0.003 meV. Furthermore, the standard deviations for each fit were approximately the same.

All of the fits included a constant background, and for part of the data taken with 38 meV incoming, it was necessary to fit to two Gaussians (at 6 °K) or one Lorentzian and one Gaussian (at 20°K) with the extra Gaussian peak corresponding to the broadly defocused Σ branch, which tended to overlap the Δ branch for some points. However, it was observed that the peak position of the Δ branch was virtually insensitive to the fitting of this broad peak.

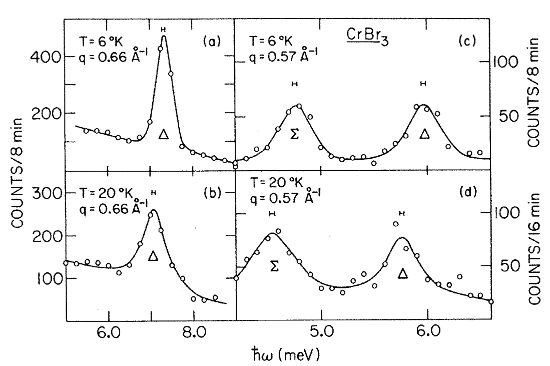

FIG. 3. Representative neutron scattering peaks observed at 6 and 20 °K. (a) and (b) are magnons in the Δ direction observed with 38 meV incoming at $q = 0.66$ Å^{-1}. The lower energy peaks in (c) and (d) are from the Σ branch; the higher energy peaks are from the Δ branch. These scans [(c) and (d)] were with 14 meV incoming at $q = 0.57$ Å^{-1}. The error bars were assigned on the basis of nonlinear least-squares fits to the appropriate functional forms.

III. THEORETICAL CONSIDERATIONS

In a neutron scattering experiment, spin-wave renormalization results from the interaction between the spin wave excited by the incoming neutron and a thermally excited one. In the first Born (or Hartree-Fock) approximation[12] to Dyson's theory[9] the strength of this interaction is proportional to the energies of the magnons in question, and thus $\Delta E/E$ is independent of q with magnitude given by the average energy of the thermally excited spin waves, $\int d_p^3 n_p \epsilon_p$. This approximation will be a good one as long as the bare interaction potential is weak, which is the case for small wave vector, as shown originally by Dyson. However, the potential increases with wave vector, and can become strong enough to produce bound spin-wave pairs at large q.[14,15] Thus as q increases, the first Born approximation loses its validity, and one must sum the entire Born series to reproduce correctly the results of Dyson's low-density expansion for the thermodynamics.[10] Physically, this summation corresponds to dressing the spin-wave interaction, to account for the correlated motions of the magnons, by using the correct (interacting) density of pair states for the magnon being excited and a thermal magnon. Since the bare interaction is attractive and increases with wave vector and the thermal magnon is one of low energy and momentum,[16] the largest effect may be expected to occur when the nonthermal magnon has an energy near the bottom of the two-spin-wave band and a relatively large wave vector. This expectation has been verified theoretically for both the simple-cubic lattice[10] and the Λ direction in $CrBr_3$,[7] where the summation of the full Born series may be performed exactly. Due to the complicated structure of $CrBr_3$, the full Born series is difficult to evaluate for the Σ and Δ directions (the interaction is not separable, so that the t-matrix integral equation is formidable). However, one may predict the qualitative behavior of $\Delta E/E$ as a function of q in the following manner.

Consider a spin wave propagating in the Δ direction. Its coupling to Λ direction magnons can be shown[17] to be of order $(J_Z/J_\perp)$ and is negligible (this coupling is in fact identically zero in the first Born approximation). Thus the energy shift results from interaction with the thermally excited spin waves *propagating within the c plane*. Although these thermal magnons propagate in all directions, they have very long wavelengths, so that their dispersion is isotropic.[8] Consequently, the density of states for two spin waves, one of which is nonthermal and propagating in the Δ direction, is *independent of the direction of the second (thermal) magnon*. For convenience, then, we can consider the thermal magnon also to be propagating in the Δ direction. The band of energies available to two noninteracting Δ direction spin waves is shown in Fig. 4, along with the two single-magnon branches. Interactions between the spin waves will alter the density of states within this band and may introduce bound states below, but will not change the boundaries of the band. Note that the bottom of the two-magnon band has an initially steep dispersion, but then levels off at about $q = 0.75$ Å^{-1} and decreases slightly at larger wave vector. In cubic systems, the band continues to rise so that it is narrowest near the zone boundary (this results in the splitting of bound states for $q \approx q_{ZB}$). Thus analogy is made between the band in Fig. 4 and that for the cubic $[11X]$ direction by comparing the value $q_\Delta(\sim\frac{3}{4} q_{ZB})$ where the zone becomes narrowest to the cubic zone boundary. Although the spin-wave interaction increases monotonically with increasing wave vector, in the cubic $[11X]$ case[10] the scattering is re-

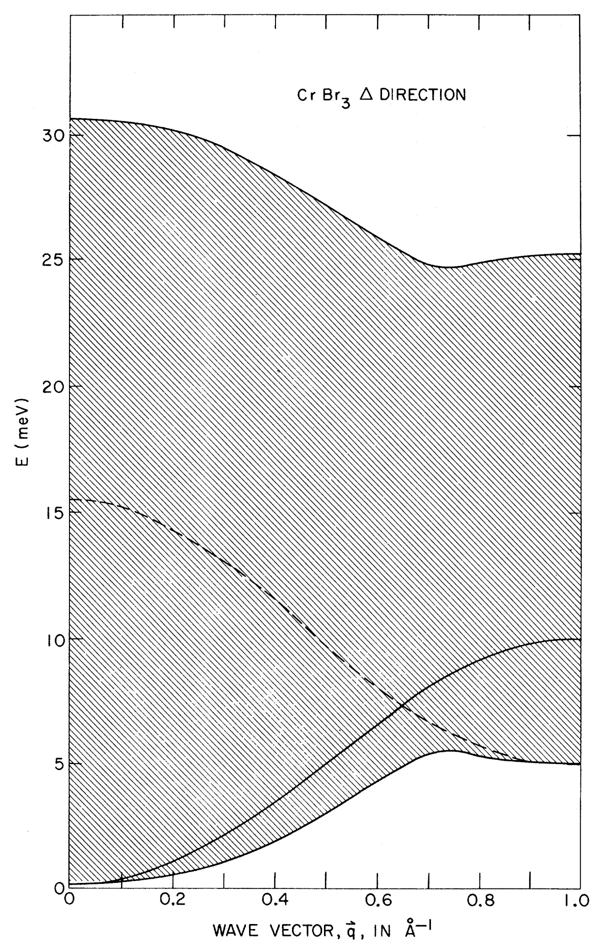

FIG. 4. The energy band for two noninteracting Δ direction magnons in $CrBr_3$ (shaded area) and the optic and acoustic magnon branches in that direction. The optic branch (dashed) was not observed due to a vanishing structure factor.

duced for q near q_{ZB} because of the rise of the single-magnon spectrum away from the lower part of the band (where the strong attractive interaction is most effective). Thus the maximum enhancement of the scattering occurs somewhere intermediate between zone center and zone boundary. By analogy, one may expect for the $CrBr_3$ Δ magnon a scattering which is most strongly enhanced for q between, say, 0.3 and 0.7 $Å^{-1}$. We note that if $q < 0.3$ $Å^{-1}$ one may actually sum the full Born series[7] by using a rectangular model of the lattice, and the effect of the higher-order terms is still small. In addition, at large wave vector the observed branch (solid curve) is relatively far from the lower edge of the band, so that the effect of the repeated scatterings will be quite small (for the reason described above), and the first Born approximation (fortuitously) quite good in this region ($q \gtrsim 0.75$ $Å^{-1}$). In summary, the qualitative effect of the spin-wave correlations on a Δ direction magnon, which can be predcited from Fig. 4, is that the first Born (Hartree-Fock) approximation of a constant $\Delta E/E$ will be valid for both small and large wave vector, with a region of enhanced scattering in between (0.3–0.7 $Å^{-1}$), where $\Delta E/E$ should rise to a maximum. Moreover, in all calculable cases,[7,9,10] the maximum enhancement is of the order of 40–60% of the first Born value, and one might expect this magnitude of effect for the $CrBr_3$ Δ magnon as well. As we shall see in Sec. IV, the observed $\Delta E/E$ seems to display precisely this structure.

One may also make an estimate of the correlation effects for a Σ direction spin wave. Since the Σ optical and acoustic branches are widely separated,[8] the band does not pinch, and there is no region of large wave vector where the Born approximation is valid. Thus $\Delta E/E$ has less structure and one can predict only a maximum $\Delta E/E$ for $q < q_{\Sigma ZB} = 0.6$ $Å^{-1}$ of order 40–60% the first-order result. Again this is consistent with our observation, as explained below.

IV. RESULTS AND DISCUSSION

In order to examine the portion of the energy shift resulting from spin-wave interactions, we treat the anisotropy as an effective field and subtract the experimentally determined[3] energy gap, $E_g(T)$, obtaining

$$\frac{\Delta E}{E} = \frac{[E(6\,°\mathrm{K}) - E_g(6\,°\mathrm{K})] - [E(20\,°\mathrm{K}) - E_g(20\,°\mathrm{K})]}{E(6\,°\mathrm{K}) - E_g(6\,°\mathrm{K})}.$$

We note that since the gap is small compared to $E(q)$ in the region studied, the q dependence of the gap will not be observable. This quantity is shown in Fig. 5 for the Δ direction (including a few points measured in the previous study). There appears to be a definite q dependence of the renormalization in the region 0.4–0.6 $Å^{-1}$ but the size of the experimental uncertainties demands careful statistical analysis to determine the degree of confidence one can place in the result. From this analysis several conclusions can be drawn.

(1) There is no overlap between the data measured in the Δ direction and those previously measured in the Λ direction[8] for any wave vector. Since the energies of the Λ branch are so small, there appears to be little chance of significantly reducing those experimental uncertainties and those measurements were not repeated. The new results for the Δ direction (with improved accuracy), however, reinforce the conclusion that the renormalization is different for the two directions.

(2) In the region 0.5–0.65 $Å^{-1}$, where most of the measurements were made, there is a statistically significant ($F > 0.9$) difference between the best average renormalization, 3.41%, and the first Born approximation prediction,[4] 2.9%. This could be a manifestation of either the q dependence of the higher-order terms or a gross failure in the lowest-order theory. This latter possibility is rather unlikely, in that the first-order theory describes reasonably well all of the measured thermodynamic properties of $CrBr_3$.[4,7,18] Furthermore, the exchange constants can be varied over the error limits of the neutron scattering measurements without affecting the predicted renormalization. The same result is obtained from both self-consistent and non-self-consistent calculations,[19] using either the correct structure (DO_5) or the simplified model used in the earlier work.[18]

(3) It can easily be seen that a constant is not the best fit to the data in the region 0.5–0.65 $Å^{-1}$. Since the predictions for $\Delta E/E$ vs q of the t-matrix summation of the full Born series cannot be ex-

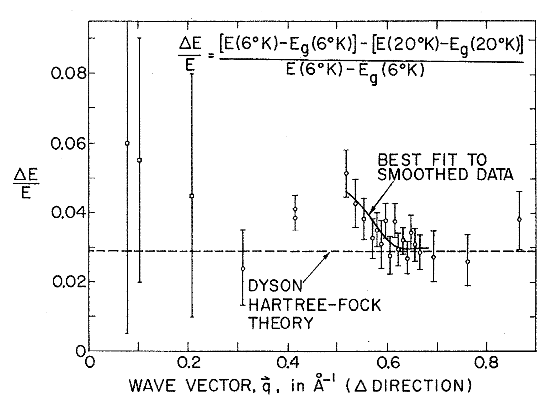

FIG. 5. Summary of experimental results for $\Delta E/E$ vs q. The circles are the new data in the Δ direction, while rectangles are from Ref. 8. The smoothed-fitted curve is derived from the dispersion curves in Fig. 6. Dashed line is the prediction of first-order spin-wave theory.

pressed in analytic form even in simple systems, it is not directly possible to fit our experimental results to an exact model. Thus we simply fit a quadratic polynomial in this region to describe the detailed shape better. A new fit is then produced which is significantly better ($F \approx 0.95$) than the best average (3.41%). The probability that the data, as described by this fit, differ from the first-order spin-wave theory is greater than 0.99. The maximum-fitted renormalization is 4.8%, agreeing reasonably well with the estimate of the full t-matrix calculation given in Sec. III.

In an effort to reduce the scatter, and better describe the data in this q-dependent region, we smoothed the data at each temperature prior to calculating $\Delta E/E$. The data between 0.5 and 0.6 $Å^{-1}$ were fitted to a polynomial in q, the order determined only by the quality of the fit. In the Δ direction only a quadratic polynomial was necessary, but for a similar analysis of the Σ direction data, cubic terms were needed as well. The results are shown in Fig. 6. The solid curves are the best quadratic fits to the data without any constraints. If we fix the 6 °K fit, and constrain the fit to the 20 °K data to a constant renormalization the result

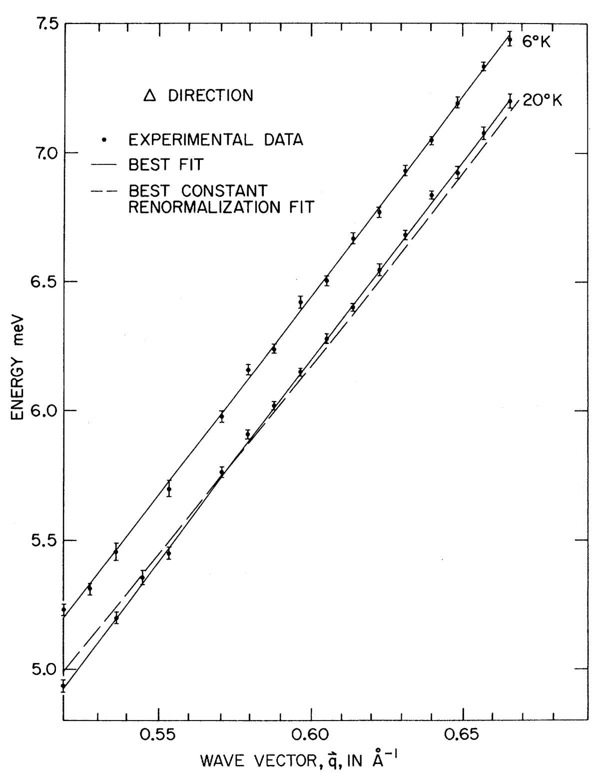

FIG. 6. A portion of the Δ direction dispersion curve measured at 6 and 20 °K. Error bars are 1 standard deviation. The solid curves are the best polynomial fits to the data. The dashed curve at 20 °K is the best polynomial fit constrained to a constant $\Delta E/E$.

is the dashed curve. Although few points fall far outside this curve, the over-all fit is clearly worse than the solid curve. Even when one takes into account the uncertainty in the 6 °K data, with 97% confidence, the data cannot be described by a constant renormalization. The solid curve in Fig. 5 is the renormalization derived from these fits rather than the quadratic fit to the nonsmoothed data.

Since the observed effect is relatively small, and the uncertainties are large, certain remarks should be made about the possibility of systematic errors affecting these observations. The Δ direction data consist of measurements made with two different incoming energies, with the maximum renormalization effects occurring in the region measured with 14 meV. It might be felt that some systematic error merely resulted in large apparent renormalization in this region, and that fitting the combined data yielded the shape observed. However, we have found that the individual dispersion curves show no sign of any such effects, and that fitting only the 38-meV data for $q > 0.57$ $Å^{-1}$ yields the same shape as fitting the combined data. It seems unlikely, then, that what we have seen represents systematic error effects.

Although detailed calculations do not exist for the theoretical renormalization of the Δ branch, the observed $\Delta E/E$ vs q is similar to that predicted in other, more easily calculated, cases including the Λ branch in $CrBr_3$, and is in qualitative agreement with the considerations of Sec. III. Specifically, in the region $q > 0.6$ $Å^{-1}$ the first-order theory result agrees completely with the measurement; below this there is a distinct increase in the renormalization down to $q \approx 0.5$ $Å^{-1}$. For smaller wave vectors, the details are quite fuzzy; the renormalization appears to first fall off toward 2.9% and then perhaps rise. However, the region from 0.4 to 0.5 $Å^{-1}$ could not be studied due to the overlap of the Δ and Σ branches, and below this the uncertainties are large because of the smaller energies (and consequently smaller shifts) involved, and therefore it would be misleading to make any definitive statement about the q dependence in the small-wave-vector region.

The Σ direction (which is truly degenerate with the Δ direction for small q) was accessible to study only over a limited range of q. The results (not shown) are also in disagreement with first-order spin-wave theory, with an average $\Delta E/E$ in the region 0.5–0.6 $Å^{-1}$ equal to 3.8%. It is not possible, however, to make any statements about the detailed q dependence of $\Delta E/E$ in this direction.

It is possible that the observed deviations from first-order spin-wave theory are not due to correlation effects, but rather to a more subtle interaction (q dependent) between the magnons and other excitations. While this cannot be definitely ruled out until detailed calculations of the correlation effects are done for the Δ branch, it is a rather unlikely possibility.

As a final point, we note that it would be convenient to find a system in which these effects might be more easily seen. However, in a truly three-dimensional magnetic system where the first-order spin-wave effects are larger (for fixed T/T_c), the magnons are too broad to be studied at a temperature at which the correlations are expected to be important, i.e., where there is a large population of thermal spin waves. In a purely two-dimensional system, the renormalization, as a function of temperature, is simply too small to be observed. Only in an intermediate system in which the renormalization is reasonably large, the population large, and the magnons relatively sharp can one hope to see these effects at all. In this respect, CrI_3 might have proven to be even better for this study than $CrBr_3$, being slightly more three dimensional.[20] In any case, we have been able to show some evidence for the existence of spin-wave correlation effects in the present system.

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