Light scattering from magnetic-energy fluctuations in the one-dimensional Heisenberg antiferromagnet $KCuF_3$

I. Yamada and H. Onda
Department of Physics, Faculty of Science, Chiba University, Yayoi-cho, Inage-ku, Chiba 263, Japan
(Received 6 July 1993)

Inelastic light scattering by magnetic-energy fluctuations has been observed in the one-dimensional Heisenberg antiferromagnet $KCuF_3$ using a conventional Raman spectrometer. The scattering, having a peak in the cross section at zero frequency and completely distinguished from Rayleigh scattering, is observed over an extremely wide range of temperature above $T_N = 39$ K. The scattered light is strongly polarized and its line profile around zero frequency is well fitted on a Lorentzian curve. The cross section increases with increasing temperature above T_N. Our experimental data are well explained by the theory which Halley developed introducing the hydrodynamic form for the correlation function of magnetic-energy density given by Halperin and Hohenberg. The absence of such inelastic scattering in K_2CoF_4 (a two-dimensional Ising antiferromagnet) and K_2CuF_4 (a two-dimensional Heisenberg ferromagnet) in a Raman spectrum is also discussed.

I. INTRODUCTION

Analysis of inelastic light scattering in magnetic compounds can be used to investigate the behavior of spin wave and spin-wave pairs. In addition, the same mechanisms may produce scattering with a peak in the differential cross section centered at zero frequency. Moriya[1] and L'vov[2] discussed independently such a quasielastic scattering based on the one-magnon process. On the other hand, Reiter[3] showed that, in the mechanism of two-magnon scattering, the light is coupled to the magnetic energy of the system. He further showed that the integrated intensity of scattering due to magnetic-energy fluctuations is proportional to the magnetic specific heat. A theory developed on the basis of a rigorous microscopic treatment was also proposed by Reiter. Consistent with Reiter's view, Halley[4] introduced a theory which facilitates analysis of the experimental results.

However, to our knowledge, only two experimental investigations have been conducted to prove these theories. One is on the three-dimensional antiferromagnet $KNiF_3$ by Lyons and Fleury.[5] The quasielastic line observed by them using a tandem pressure-scanned Fabry-Pérot interferrometer can be represented as the sum of two peaks both of which have peaks at zero frequency; the broader one with the half width at half maximum of approximately ~ 10 GHz at T_N was interpreted to result from scattering caused by the magnetic-energy fluctuations. The other report is on the layered antiferromagnetic semiconductor $FePS_3$ by Sekine *et al.*[6] They observed quasielastic scattering below and above $T_N = 118$ K.

Encouraged by these theories and experiments, we investigated light scattering around zero frequency in the one-dimensional Heisenberg antiferromagnet $KCuF_3$. Our motivation in choosing this compound is because the current theories[3,4] indicate that quasielastic scattering of $KCuF_3$ should be governed by the magnetic specific heat. If so, we expect scattering to be observed over a wide range of temperatures above T_N in one-dimensional Heisenberg magnetic systems like $KCuF_3$ because, contrary to three-dimensional systems, a great portion of the magnetic entropy remains above T_N in such systems. Furthermore, this compound is good for experiments of light scattering because it has no absorption band for visible light.

Employing a Raman spectrometer, we obtained a result which, we believe, is representative of quasielastic light scattering caused by magnetic-energy fluctuations. In Sec. II we shall first review the magnetic and crystallographic properties of $KCuF_3$ so far reported. After introducing the theoretical background in Sec. III, we shall show our experimental results and discuss them in Sec. V. A preliminary report[7] of the present study has already been published.

II. MAGNETIC AND CRYSTALLOGRAPHIC PROPERTIES OF $KCuF_3$

This compound is known to form magnetic linear chains along the c axis, in spite of its pseudocubic crystal structure.[8] The origin of such a peculiar magnetic coupling is solely due to the cooperative Jahn-Teller distortion of CuF_6 octahedra. That is, each F atom is slightly displaced from the center of adjacent Cu sites in the c plane as a result of the cooperative Jahn-Teller effect. As a result, the hole orbital of Cu^{2+}, i.e., $d_{x^2-z^2}$ or $d_{y^2-z^2}$, shows alternate ordering in the c plane as shown in Fig. 1. The overlap of orbitals along the c axis results in a strong superexchange interaction, whereas that perpendicular to the c axis is very weak because of poor overlap.

On the other hand, studies[9,10] of electron paramagnetic resonance (EPR) have revealed that this magnetic system should have the Dzyaloshinsky-Moriya (DM) exchange interaction $\sum_{i>j} \mathbf{d}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$ between nearest-neighbor (NN) spins on the c axis with the DM vector $\mathbf{d}_{ij}$ perpendicular to the c axis. Furthermore, a recent antiferro-

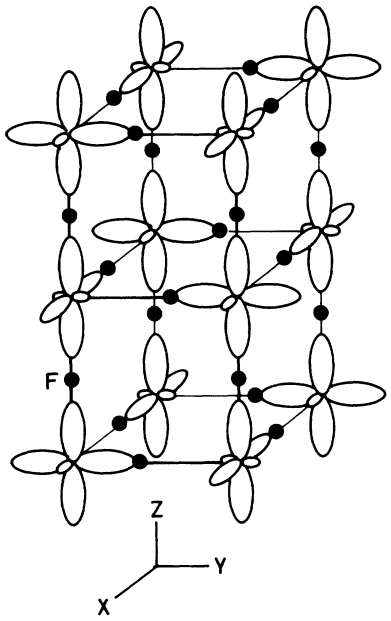

FIG. 1. Crystal structure of $KCuF_3$ so far accepted and the orbital ordering. The laboratory axes (X, Y, Z) are indicated; X, Y, and Z are parallel to the $[100]_p$, $[010]_p$, and $[001]_p$ axes, respectively.

magnetic resonance (AFMR) study[11] showed that $\mathbf{d}_{ij} \| [100]_p$ and its equivalents, where $[\ ]_p$ indicates an axis in the unit cell of a perovskite structure. Therefore, the adjacent DM vectors in the c plane are orthogonal to each other. The Hamiltonian in zero field is then given by

$$\mathcal{H} = -2 \sum_{i>j} (J_c \mathbf{S}_i \cdot \mathbf{S}_j + D_c S_i^z S_j^z) - 2J_a \sum_{l>m} \mathbf{S}_l \cdot \mathbf{S}_m + \sum_{i>j} \mathbf{d}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \quad (1)$$

where J_c and J_a are the exchange interactions between NN spins along the c axis and in the c plane, respectively. The term D_c produces an XY anisotropy by which the spins are laid in the c plane. The last term represents the DM interaction between NN spins along the c axis. The value of each parameter so far reported is $J_c = -187$ K (Ref. 12) (magnetic specific heat), -190 K (Ref. 8) (susceptibility), -197 K (Ref. 13) and -203 K (Ref. 14) (neutron diffraction), $J_a \simeq 0.01 |J_c|$ (Ref. 14) (neutron diffraction), $D_c \simeq 0.04$ K (Ref. 14) (neutron diffraction), and $|\mathbf{d}_{ij}| \equiv d \simeq 0.027 |J_c|$ (Ref. 11) (AFMR).

Since the value of the main term $|J_c|$ is extremely large compared with the other terms, it is possible to neglect the other terms where magnetic energy is concerned. Owing to excellent one-dimensionality and extremely large value of $|J_c|$ of $KCuF_3$, the magnetic specific heat shows a broad peak around 180 K, as observed by the measurement of birefringence[12] and therefore the bulk of the magnetic entropy calculated from the specific heat was found to remain above $T_N = 39$ K.

The crystal structure as well as the orbital ordering mentioned above is shown in Fig. 1. A space group D_{4h}^{18} was assigned to it.[15] The discovery of the DM interaction in this compound, however, raises questions about its crystal structure because the symmetry consideration based on the D_{4h} point group results in $\mathbf{d}_{ij} = 0$.[9] Recent studies of phonon-Raman scattering[16] and x-ray diffraction[17] strongly suggest a crystal symmetry lower than D_{4h}; we believe that the distorted CuF_6 octahedra make a superstructure which probably involves minute tilting of the octahedra around one of the principal axes of the pseudocubic structure. When the crystal symmetry is lower than current estimates, the configurations of the polarization of incident and scattered light that allow inelastic scattering around zero frequency should be different from those of the D_{4h} symmetry, which we shall explain more precisely in the following section.

III. THEORETICAL BACKGROUND

As established theory of light scattering shows, the scattered radiation is described as an electric dipole radiation from the induced dipole. Therefore, the differential cross section for the scattering of light from a field $\mathbf{E}_1$, wave vector $\mathbf{k}_1$, frequency ω_1, to a field $\mathbf{E}_2$, wave vector $\mathbf{k}_2$, frequency ω_2 by a magnetic system is given by

$$\frac{d^2h}{d\Omega\, d\omega_2} = \frac{\omega_1 \omega_2^3 n V}{16\pi^2 c^4} \sum_{\alpha\beta\mu\nu} \epsilon_1^\alpha \epsilon_2^\beta \epsilon_1^\mu \epsilon_2^\nu \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \langle M^{\alpha\beta}(\mathbf{k},t) [M^{\mu\nu}(-\mathbf{k},0)]^* \rangle \ , \quad (2)$$

where $\omega = \omega_2 - \omega_1$, $\mathbf{k} = \mathbf{k}_2 - \mathbf{k}_1$, ϵ_1, ϵ_2 are the polarization vectors of the incident and scattered light, c is the velocity of light, V is the scattering volume, n is the refractive index, while $\langle\ \rangle$ indicates a thermal average. The superscripts α, β, μ, and ν denote Cartesian components. The polarization factor $\epsilon_1^\alpha \epsilon_2^\beta \epsilon_1^\mu \epsilon_2^\nu$ is fixed to a nonzero value when the combination of the polarization of incident and scattered light allowed by its symmetry is determined. The tensor $M^{\alpha\beta}(\mathbf{k})$ which is given by

$$M^{\alpha\beta}(\mathbf{k}) = \sum_i e^{i\mathbf{k}\cdot\mathbf{r}_i} M_i^{\alpha\beta} \quad (3)$$

indicates a coupling between the light and a quantity coupled to the magnetic-energy density and (below the transition temperature) to other slowly varying hydrodynamic variables. Following the general approach of Moriya,[1] we may express the coupling in question as

$$M_i^{\alpha\beta} = \sum_\mu G^{\alpha\beta\mu} S_i^\mu + \sum_j \sum_{\mu\nu} H^{\alpha\beta\mu\nu} S_i^\mu S_j^\nu \ . \quad (4)$$

The first term in Eq. (4) involves a spin operator at a single ionic site i with a complex tensor $\mathbf{G}$ describing the strength of the coupling between the light and the mag-

netic system. This term leads to scattering from single-spin fluctuations. The second term in Eq. (4) involves a pair of spin operators at different sites and the coupling tensor $\mathbf{H}$, and gives rise to scattering by pairs of spin fluctuations. Nonzero components of $\mathbf{G}$ and $\mathbf{H}$ are determined by symmetry operations of the crystal.

Moriya[1] and L'vov[2] treated a one-spin process. They derived that the factor given by $\langle\ \rangle$ in Eq. (2) is expressed as $\langle m_\zeta(\mathbf{k},t)m_\zeta^*(-\mathbf{k},0)\rangle$ for both ferromagnets and antiferromagnets above and below the transition temperature,[1] where $m_\zeta=g\mu_B S^\zeta$ and ζ is one of the Cartesian components.

On the other hand, Reiter[3] and Halley[4] treated a two-spin process. According to them, the two-spin process results in $\langle E(\mathbf{k},t)E^*(-\mathbf{k},0)\rangle$ for $T>T_N$ for symmetry-allowed polarization, where $E(\mathbf{k},t)$ is the magnetic-energy density given by the Fourier transform of $E(\mathbf{r})=-\langle\sum_{i>j}(J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j)\delta(\mathbf{r}-\mathbf{r}_i)\rangle$ where J_{ij} is an exchange interaction and $\mathbf{r}_i$ is the position of the ith spin. As a result, a line profile of the symmetry-allowed ($\epsilon_1^\alpha\epsilon_2^\beta\epsilon_1^\mu\epsilon_2^\nu=1$) scattering is then given by

$$I(\omega)\propto\int_{-\infty}^{\infty}e^{-i\omega t}dt\,\langle m_\zeta(\mathbf{k},t)m_\zeta^*(-\mathbf{k},0)\rangle \tag{5}$$

for the one-spin process, while

$$I(\omega)\propto\int_{-\infty}^{\infty}e^{-i\omega t}dt\,\langle E(\mathbf{k},t)E^*(-\mathbf{k},0)\rangle \tag{6}$$

for the two-spin process.

The two-spin process is treated below. Introducing the hydrodynamic form given by Halperin and Hohenberg[18] for the correlation function $\langle E(\mathbf{k},t)E^*(-\mathbf{k},0)\rangle$ and based on the fluctuation-dissipation theorem, one obtains an expression, following the notations given by Halley, as

$$I(\omega)\propto\frac{\omega}{1-e^{-\beta\hbar\omega}}\frac{C_mTDk^2}{\omega^2+(Dk^2)^2}\ , \tag{7}$$

where $\beta=1/k_BT$, C_m is the magnetic specific heat at zero magnetic field, and D is the thermal diffusion constant which is given by

$$D=\frac{K}{C_m} \tag{8}$$

when K is the magnetic contribution to the thermal conductivity. When $\beta\hbar\omega\ll1$, one obtains the expression

$$I(\omega)\propto C_mT^2\frac{Dk^2}{\omega^2+(Dk^2)^2}\ . \tag{9}$$

Equation (9) indicates that the peak height of $I(\omega)$ is proportional to C_mT^2/Dk^2 and has a Lorentzian shape with the half width given by Dk^2.

For $T<T_N$, on the other hand, Halley[4] showed that the magnetization also contributes to $\langle\ \rangle$ in Eq. (2) and an essentially identical analysis gives

$$I(\omega)\propto\int_{-\infty}^{\infty}dt\,e^{-i\omega t}[\langle E(\mathbf{k},t)E^*(-\mathbf{k},0)\rangle + J^2\langle m_z(\mathbf{k},t)m_z^*(-\mathbf{k},0)\rangle]\ , \tag{10}$$

as long as the symmetry-allowed polarization of incident and scattered light for the two-spin process exists. Using the hydrodynamic results, Eq. (10) is reduced to

$$I(\omega)\propto\frac{\omega}{1-e^{-\beta\hbar\omega}}\left[\frac{C_mTDk^2}{\omega^2+(Dk^2)^2}+J^2\frac{\chi_\parallel D_\parallel k^2}{\omega^2+(D_\parallel k^2)^2}\right]\ , \tag{11}$$

where $D_\parallel$ is the diffusion constant for the component at the magnetization parallel to the sublattice magnetization, $\chi_\parallel$ is the parallel susceptibility in unit of $(g\mu_B)^2$, and J is the exchange interaction.

Citing Van Hove's argument[19] for critical slowing down, Halperin and Hohenberg[20] suggest that K depends primarily on the short-range behavior of the system, and should therefore remain finite at the critical point. This suggestion is also supported by more microscopic treatments by Kawasaki and his co-workers.[21,22] The magnetic specific heat C_m diverges at the critical point, so that D is predicted to vanish at T_N. When the mean-field temperature dependence is used for C_m, the thermal diffusion constant D is predicted to be linear to $T-T_N$.

IV. EXPERIMENTAL PROCEDURE

The sample used was cut out from a large single crystal grown by the Bridgman method.[9] It had neither twins nor stacking disorder, which we confirmed by x-ray diffraction. We shaped it into a rectangular parallelepiped of approximately $3\times3\times3$ mm^3 size; its surfaces were parallel to the $(100)_p$ plane and its equivalents. To reduce stray light, each surface was polished to an unevenness of about 1 μm. Raman-scattering measurements were performed using a laboratory-assembled instrument such as a double monochromator (CT-1000 D, Jasco), a cooled photomultiplier (R-943-02, Hamamatsu), a photon counter (C-767, Hamamatsu) and several optical elements such as mirrors, prisms, and lenses. Since this compound has no absorption band over 350–650 nm, a 514.5 nm polarized argon laser was employed as the light source and all spectra were collected in a microcomputer. The polarization of the scattered light was analyzed with Polaroid HN-38 film. A quartz wedge, placed just in front of the entrance slit of the monochromator, was used to depolarize the scattered light. Using a closed-cycle refrigerator, we obtained the spectra over the temperature range 14–300 K.

In the following discussion, the laboratory axes, X, Y, and Z, are taken as shown in Fig. 1; Z is parallel to the $[001]_p$ axis, X is parallel to one of the $[100]_p$ axes, and Y is perpendicular to X in the c plane. The orientation of the sample with respect to the directions of incident and scattered light under the 90° scattering geometry is chosen for the desired polarization analysis. Following the method given by Moriya[23] for one-spin and two-spin processes, we determine the polarization of the incident and scattered light which is symmetry allowed and produces inelastic scattering around $\omega=0$. As a result, cross configurations ZX and XY were found to be symmetry allowed for the one-spin process, while the ZZ and XX configurations yield the scattering from the two-spin process only as long as D_{4h} crystal symmetry is adopted.

Introducing tilting in CuF_6 octahedra relaxes the restriction for the polarization of the incident and scattered light. For instance, the XX and ZZ become allowed symmetries for the one-spin process as well as the cross configurations, ZX and XY, while the ZX, XX, and ZZ configurations are symmetry allowed for the two-spin process. The experimental spectrum in which quasielastic scattering appears is only the ZZ spectrum as it is not against the symmetry consideration for the two-spin process mentioned above.

As long as a conventional Raman spectrometer is used as the present case, it is sometimes difficult to detect quasielastic scattering because Rayleigh scattering (direct scattering), the intensity of which is usually stronger than the expected quasielastic line, covers an area of several cm^{-1} around $\omega=0$. As will be shown later, Rayleigh scattering appears only over the area of $|\omega|<2$ cm^{-1} in the spectra of the present experiments for $KCuF_3$. Then, if the full width of the quasielastic line is less than ~ 4 cm^{-1}, it becomes impossible to separate the quasielastic line from the Rayleigh scattering line.

V. EXPERIMENTAL RESULTS AND DISCUSSION

In our earlier paper,[7] it was shown that the shape of the Rayleigh scattering line is narrow enough not to mask a quasielastic line in the present experiments (see Fig. 1 in Ref. 7); Rayleigh scattering was confirmed to appear below $|\omega|\sim 2$ cm^{-1} as an abrupt increase in the scattering intensity and it was confirmed to be independent of temperature below room temperature. As a result, quasielastic scattering, which appears for $|\omega|>2$ cm^{-1}, is easily distinguished from Rayleigh scattering.

To see how the quasielastic line changes with temperature, we show in Fig. 2 several traces obtained at different temperatures. More accurate spectra near $\omega\sim 0$ are shown in Fig. 2 of Ref. 7. As can be seen in Fig. 2, the quasielastic scattering line is clearly seen except for $T=24$ K, and the scattering intensity increases with temperature above $T_N=39$ K. The scattering intensity observed below T_N is very weak, as can be seen in the trace obtained at $T=24$ K. Quasielastic scattering below T_N, if it occurs, probably has a narrow linewidth and is masked by the Rayleigh scattering line.

As the theory of Halley introduced in Sec. III suggests, the spectrum obtained at a fixed temperature above T_N can be fitted on a Lorentzian curve. We have tried to fit the theoretical curve on the experimental results using the least-squares method. The result is shown in Fig. 2 by solid lines. As a consequence, one finds that each experimental line is Lorentzian. Equation (9) indicates that the half width and the peak height of the fitted line are proportional to Dk^2 and C_mT^2/Dk^2, respectively. Therefore, D and C_m are obtained from the fitted lines taking into account the value $|\mathbf{k}|\sim k_1\sin(\theta/2)$ for the scattering angle $\theta=90°$. The C_m and D thus obtained are shown in Fig. 3(a) and Fig. 4, respectively, as a function of temperature. The magnetic specific heat[12] obtained by birefringence measurements is reproduced in Fig. 3(b). One finds that the dependence of C_m on temperature bears a remarkable resemblance with the one given in

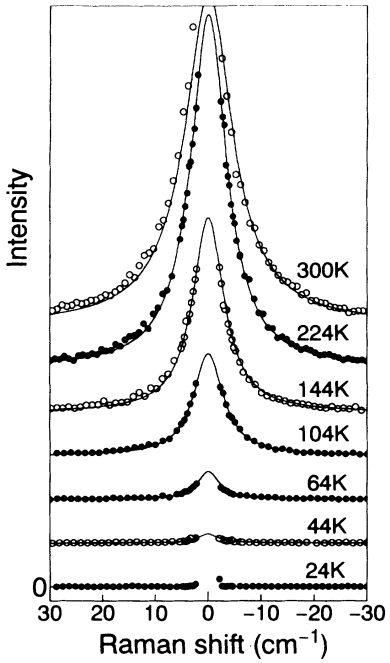

FIG. 2. Traces of quasielastic scattering obtained at several temperatures. Each solid line indicates a fitting of Lorentzian line shape. Intensity is shown on a scale of arbitrary units. More accurate spectra near $\omega\sim 0$ are shown in Fig. 2 in Ref. 7. The polarization phenomena of the scattered light are shown in Fig. 1 in Ref. 7.

Fig. 3(b). On the other hand, D decreases with temperature, as can be seen in Fig. 4, which is not against the theoretical prediction explained at the end of Sec. III.

Our present Raman experiments could not detect quasielastic scattering below T_N as referred to above. We cannot, however, rule out the possibility that quasielastic scattering may have a width far narrower than that of the Rayleigh scattering. A Brillouin scattering spectrometer is necessary to observe such a narrow inelastic line expected below T_N, because the line having the full width less than several cm^{-1} is masked by Rayleigh scattering as long as a conventional Raman spectrometer is used.

In addition to $KCuF_3$, we performed experiments on K_2CoF_4 and K_2CuF_4 using the same Raman spectrometer. A 633 nm line of a He-Ne laser was employed for K_2CoF_4 as the light source because the color of the sample was dark red, while a 514.5 nm line of an argon laser was used for K_2CuF_4. Both of these compounds are well-known two-dimensional magnetic systems; the former is an Ising antiferromagnet with $T_N=107$ K and the latter a Heisenberg ferromagnet having $T_C=6.25$ K. Our experimental conclusion is that both systems do not yield quasielastic scattering except over an area of $|\omega|<6$ cm^{-1} covered by Rayleigh scattering.

We believe that the absence of such a scattering in Raman spectra of these two compounds is caused by different phenomena. The magnetic specific heat of K_2CoF_4 shows a sharp peak[24] which diverges logarithmically at T_N owing to its excellent two-dimensional Is-

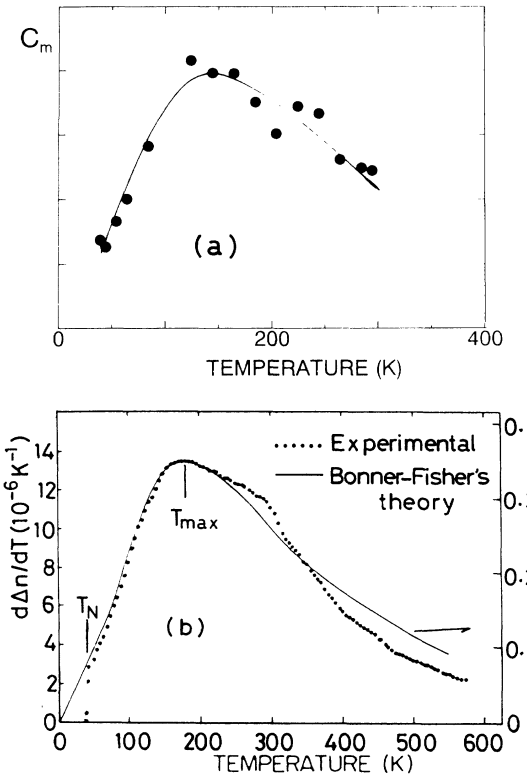

FIG. 3. (a) Temperature dependence of magnetic specific heat C_m derived from the peak height of the fitted Lorentzian line. The solid line is a guide to the eyes. (b) The magnetic specific heat obtained by birefringence measurements which is reported in Ref. 12 and is reproduced here by permission of The Physical Society of Japan.

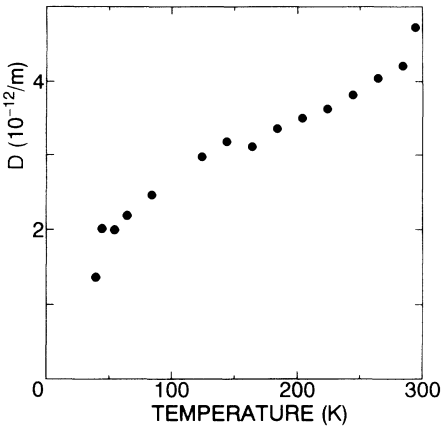

FIG. 4. Thermal diffusion constant D derived from the linewidth of quasielastic line.

ing character. If the thermal conductivity K diverges less strongly than C_m, the thermal diffusion constant D given by Eq. (8) should be small. As a result, the width of the quasielastic line, which is proportional to D, will be too narrow to be detected by a conventional Raman spectrometer because the Rayleigh line covers such a narrow quasielastic line. We believe that a Brillouin scattering method should be used in such a case because, by this method, quasielastic scattering can be observed over the frequency range above several GHz. The results for $KNiF_3$ (Ref. 5) correspond to this situation; a Brillouin scattering spectrometer instead of a Raman spectrometer was used for this compound because the linewidth is only ~ 10 GHz.

As theoretically shown, on the other hand, the exchange scattering mechanism, which is the origin of two-magnon scattering, is not effective in a ferromagnet. Thus the quasielastic scattering cannot be observed in a ferromagnet as long as the scattering mechanism is the same as the two-magnon process. This is why K_2CuF_4 does not yield inelastic scattering around $\omega=0$.

Before we conclude our discussion, we refer to quasielastic scattering from the one-spin process. Since the symmetry-allowed configurations of the polarization in the one-magnon process is generally different from that in the two-magnon process,[25] the possibility exists that the quasielastic scattering suggested by Moriya or L'vov in the configurations other than those allowed for two-magnon process may appear. The contribution of $\langle m_z(\mathbf{k},t)m_z^*(-\mathbf{k},0)\rangle$ below T_N deduced from the two-magnon process given by Halley should be strictly distinguished from that originated from the one-magnon process.

In K_2CoF_4, K_2CuF_4, and $KCuF_3$, however, no inelastic line around $\omega=0$ was observed in any of the configurations of the polarization of our Raman spectrometer except the ZZ case for $KCuF_3$. To make clear whether the one-magnon process produces quasielastic scattering in these compounds, further experimental investigation by a Brillouin spectrometer is desired over the are of low frequency.

In conclusion, quasielastic light scattering has been observed in $KCuF_3$. The scattering is exactly distinguished from Rayleigh scattering. The temperature behavior of the scattering intensity and the width of the line is well reproduced by Halley's theory which is developed on the basis of the hydrodynamic form of the correlation function of the magnetic-energy density.

ACKNOWLEDGMENTS

We are grateful to Dr. M. Hidaka for his critical examination of single crystals of $KCuF_3$. Thanks are also due to M. Suzuki for his help in experiments. This study was partly supported by a grant from Fujitsu Ltd.

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