Antiferromagnetic resonance in $KNiF_3$

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Far-infrared spectroscopy and electron spin resonance measurements were performed on single crystals of the cubic perovskite $KNiF_3$. We found the absorption at $48.3\,cm^{-1}$, which was thought to be a magnetic signal (P. L. Richards: J. Appl. Phys. **34**, 1237 (1963)) is not magnetic in origin. Instead, a new absorption line was observed and was well fitted by the theory of antiferromagnetic resonance with uniaxial anisotropy. The analysis yields an anisotropy energy of $8.7 \times 10^{-3}\,cm^{-1}$ and the ratio of the anisotropy field to the exchange field only 2.4×10^{-5}. Thus, $KNiF_3$ is an excellent example of a Heisenberg antiferromagnet.

In spite of the enormous studies since 1950's, magnetism of $3d$ transition metal compounds with the perovskite-type structure is still attracting considerable attention. $KNiF_3$ has a cubic perovskite structure and is regarded as an ideal example of Heisenberg antiferromagnets. The cubic symmetry is retained down to $78\,K$[1] which is significantly lower than the Néel temperature $T_N = 253\,K$ determined by measurements of specific heat.[2] Isotropy in crystal structure implies the isotropy of magnetic exchange interaction between neighboring Ni spins. Indeed, measurements of neutron diffraction[3] revealed that Ni spins on the neighboring sites are aligned antiferromagnetically in three directions (G-type antiferromagnet) below T_N. The magnitude of the exchange interaction (J) between the neighboring spins was derived to be $J = 89 \pm 4\,K$ from an analysis of magnetic susceptibility.[4] For this isotropic antiferromagnet $KNiF_3$, the magnetic excitation spectra below T_N were studied by far-infrared (FIR) spectroscopy at zero field.[5] In that work, an absorption line at $48.7 \pm 0.3\,cm^{-1}$ was observed and ascribed to an antiferromagnetic resonance (AFMR). If this absorption indeed corresponds to the AFMR, the value of the single ion anisotropy would be $3.2\,cm^{-1}$. This value is, however, incompatible with other experiments which suggested isotropic nature of $KNiF_3$.

To clarify this contradiction, we have performed measurements of FIR spectroscopy and electron spin resonance (ESR).[6] Our results indicate that the absorption observed by Richards[5] is not magnetic in origin. In addition to this absorption, we found resonance peaks whose position shifted by magnetic fields. An analysis of our results by a standard theory for AFMR[7] revealed that $KNiF_3$ is an excellent example of Heisenberg antiferromagnets.

Single crystals of $KNiF_3$ were grown by a flux method and cut into pellets with typical dimensions of $5 \times 2.5 \times 0.5\,mm^3$. FIR absorption at zero magnetic field has been measured in RIKEN by using a Fourier-transform spectrometer (Bruker IFS-120HR). FIR spectroscopy in magnetic fields up to $30\,T$ was done at the National High Magnetic Field Laboratory in Tallahassee, USA. Measurements of ESR have been performed using a high-frequency high-field spectrometer in RIKEN.

Figure 1 shows the FIR absorption spectra of $KNiF_3$ in zero field at various temperatures between 4.2 and $70\,K$. A strong peak at $49\,cm^{-1}$ is in good agreement with that reported by Richards.[5] In addition to the resonance at $49\,cm^{-1}$, weak peaks are found at around 86 and $94\,cm^{-1}$ (see arrows in Fig. 1. The intensity of each resonance is diminished drastically as temperature increases.

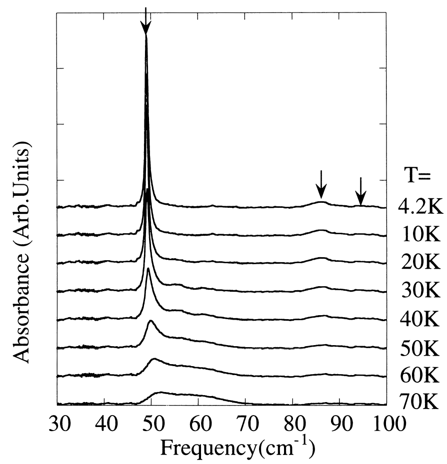

Fig. 1. Temperature dependence of the far-infrared absorption spectra of $KNiF_3$ in zero field. The curves have been offset in ordinate for clarity.

As for the absorption of FIR light, two origins can be considered, i.e., magnetic and phononic ones. For the former origin, external magnetic fields shift the resonance frequencies, whereas, do not for the latter.

To clarify the origin of the absorption in Fig. 1, we have performed an FIR spectroscopy in magnetic fields up to $30\,T$. As shown in the inset of Fig. 2, these three peaks did not show any field-dependence. Therefore, these absorption lines are not magnetic in origin. In addition to the three peaks shown in Fig. 1, new peaks were observed at lower frequencies of these measurements. Figure 3 shows the FIR transmission spectra of $KNiF_3$ at $4.2\,K$ in magnetic fields. To extract field-dependent component, each spectrum in a magnetic field was normalized by that at zero field. The resonance point (see arrows in Fig. 3) is plotted in the main panel of Fig. 2 as a function of magnetic field for different temperatures.

Because the low-frequency limit of the FIR spectrometer is about $10\,cm^{-1}$, we have carried out ESR measurements below $340\,GHz$ ($11.3\,cm^{-1}$). The inset of Fig. 4 shows a typical trace of the transmission of a millimeter wave with a frequency of $215.97\,GHz$. A strong absorption line is seen at $6.35\,T$, a weaker one at $6.07\,T$, and a very weak one at $3.18\,T$. All the resonance points obtained from this study for $KNiF_3$ are plotted in a frequency versus field plane in Fig. 4.

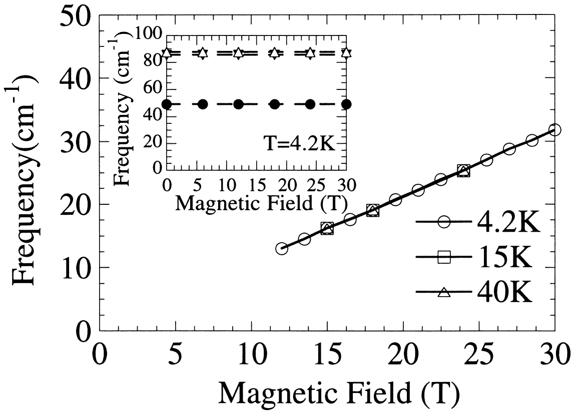

Fig. 2. An absorption mode in $KNiF_3$ measured at various fields and temperatures. The inset shows the magnetic-field dependence of the three low-frequency absorption lines in $KNiF_3$ at 4.2 K. The external magnetic fields are applied parallel to the [100] direction.

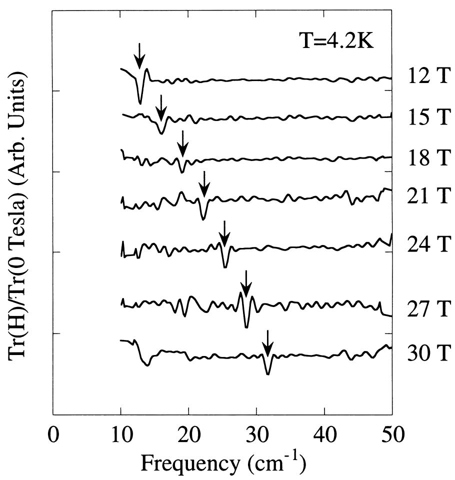

Fig. 3. Far-infrared transmission ratio spectra of $KNiF_3$ at 4.2 K in applied magnetic fields up to 30 T. The spectra taken at finite fields are normalized by the one at zero field. The curves have been offset in ordinate for clarity.

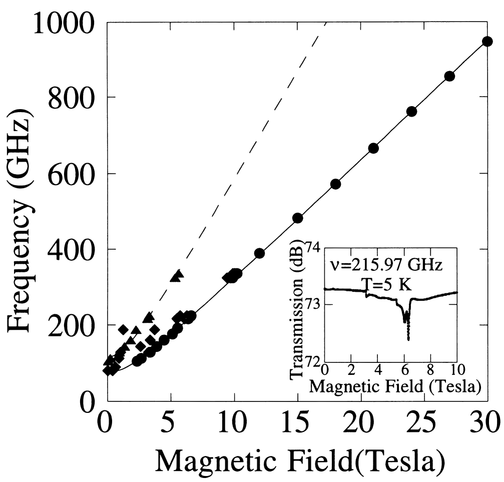

Fig. 4. The frequencies versus magnetic field (symbols) relation for the magnetic excitations for $KNiF_3$ from both far-infrared (T = 4.2 K) and ESR (T = 5 K) measurements. Solid and dashed lines are theoretical fits described in the text. The inset shows the ESR spectrum of $KNiF_3$ at 5 K taken at a frequency of 215.97 GHz.

Next, we analyze the magnetic excitation branches using the theory of AFMR[7] with uniaxial anisotropy. When the external magnetic field H is applied perpendicular to the easy axis, the AFMR frequency ν at low temperatures ($T \ll T_{\mathrm{N}}$) is given by

$$2\pi\nu/\gamma = (H^2 + 2H_{\mathrm{E}}H_{\mathrm{A}})^{1/2}. \tag{1}$$

where γ is the magnetomechanical ratio, H_{E} and H_{A} are the exchange and anisotropy fields, respectively. From the fit of Eq. (1) to the main peak of our data (see the solid line in Fig. 4), we determined the two parameters, $(\gamma/2\pi)\,(2H_{\mathrm{E}}H_{\mathrm{A}})^{1/2} = 76.1\,\mathrm{GHz}$ ($2.54\,\mathrm{cm}^{-1}$) and $g = 2.26$. Using the value of $J = 89\,\mathrm{K}$ ($= 62\,\mathrm{cm}^{-1}$) derived by Lines,[4] we obtain $g\mu_{\mathrm{B}}H_{\mathrm{E}} = 370\,\mathrm{cm}^{-1}$ at low temperatures ($T \ll T_{\mathrm{N}}$). Then, using the experimental value of $(\gamma/2\pi)\,(2H_{\mathrm{E}}H_{\mathrm{A}})^{1/2} = 76.1\,\mathrm{GHz}$, we estimate $g\mu_{\mathrm{B}}H_{\mathrm{A}} = 8.7 \times 10^{-3}\,\mathrm{cm}^{-1}$. This anisotropy energy is too small to be accounted for either by the magnetic dipole-dipole interaction (typically of the order of $0.1\,\mathrm{cm}^{-1}$) or by a single ion anisotropy (typically of the order of $1\,\mathrm{cm}^{-1}$). Thus, we conclude that the cubic symmetry is retained below T_{N}. The ratio between H_{A} and H_{E} in $KNiF_3$ is 2.4×10^{-5}. This result indicates that $KNiF_3$ is an excellent example of Heisenberg antiferromagnets.

Finally, we briefly discuss the weak absorption lines observed in the ESR measurements. The weak absorption near the main signal in the inset of Fig. 4 may be an interference fringe. We try to fit the weak absorption, e.g. at 3.18 T for $\nu = 215.97\,\mathrm{GHz}$, to the AFMR theory. The dashed line in Fig. 4 represents the fit by Eq. (1) with $g = 4.09$ and $(\gamma/2\pi)\,(2H_{\mathrm{E}}H_{\mathrm{A}})^{1/2} = 115.8\,\mathrm{GHz}$. One possible explanation for this excitation would be that it originates from a nonlinear excitation. Further studies are necessary to clarify this point.

In conclusion, we have carried out measurements of far-infrared spectroscopy and electron spin resonance for single crystals of $KNiF_3$. We confirmed that the absorption at $49\,\mathrm{cm}^{-1}$ reported before[5] is not magnetic in origin. Instead, we observed new branches of magnetic resonance. An analysis of the antiferromagnetic resonance mode gives a very small anisotropy energy ($8.7 \times 10^{-3}\,\mathrm{cm}^{-1}$) and confirms that $KNiF_3$ is an excellent example of Heisenberg antiferromagnets.

This work has been done in collaboration with H. L. Liu, D. B. Tanner, H. Yamaguchi, Y. J. Wang, and A. Zibold.

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