X-band antiferromagnetic resonance measurements in $KNiF_3$

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The antiferromagnetic resonance spectrum of $KNiF_3$ has been studied at temperatures close to $T_N=247$ K. Each spectrum consists of a very broad line and the line shape has been analyzed in terms of the Landau-Lifshitz phenomenological equations. The spectrum disappears at $T\sim 230$ K, a fact which, we suggest, is caused by the reorientation of the antiferromagnetic domains.

INTRODUCTION

The G-type antiferromagnet $KNiF_3$ has very simple crystallographic and magnetic structures.[1] Due to its strong nearest-neighbor exchange interaction and low anisotropy this cubic perovskite is one of the best examples of the Heisenberg three-dimensional (3D) system.[2] For this reason $KNiF_3$ has been extensively studied both experimentally and theoretically. However, the antiferromagnetic resonance properties of this compound seem to be less known. To our knowledge there is only a brief report of a 35-GHz measurement by Petrov and Kizhaev[3] analyzed by Curatella *et al.*[4]

The magnetic Hamiltonian in $KNiF_3$ can be written as[2]

$$H=-J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-k\sum_{i,\alpha=1}^{2}[S_{ix}^2(\alpha)S_{iy}^2(\alpha)+S_{iy}^2(\alpha)S_{iz}^2(\alpha)+S_{iz}^2(\alpha)S_{ix}^2(\alpha)]+g\mu_B\sum_{i,\alpha=1}^{2}\mathbf{S}_i(\alpha)\mathbf{H}\ ,$$

where $\langle ij\rangle$ labels nearest-neighbor pairs and i,α runs over all "one" up and "two" down sublattice ions. J measures the Heisenberg antiferromagnetic exchange interaction ($J<0$) and k is related to the single-ion anisotropy ($k<0$)[2]. The last term is the Zeeman interaction.

In order to consider the temperature dependence of the antiferromagnetic resonance (AFMR) properties, this system can be described classically in terms of the sublattice magnetizations $\mathbf{M}_1(T)$, $\mathbf{M}_2(T)$ through a phenomenological T-dependent free energy F, which includes contributions from exchange, anisotropy, and Zeeman interactions. This free energy is given by

$$F=-\lambda(T)\mathbf{M}_1\cdot\mathbf{M}_2-\frac{K(T)}{2}\sum_{i=1}^{2}(M_{ix}^2M_{iy}^2+M_{ix}^2M_{iz}^2+M_{iy}^2M_{iz}^2)/M_i^4-\mathbf{H}\cdot(\mathbf{M}_1+\mathbf{M}_2)\ , \tag{1}$$

where $\lambda(t)$ is the molecular field constant, $K(T)$ is the cubic anisotropy constant, and H is the applied magnetic field. We will use the standard approximations[5] $|M_{1z}|-|M_{2z}|=\chi_{\parallel}H$ and $\lambda=1/\chi_{\perp}$ to relate these parameters with the static susceptibilities $\chi_{\parallel}$ and $\chi_{\perp}$. For $H=0$, $M_1=M_2=M_0(T)$, which is the spontaneous sublattice magnetization.

At temperatures below the critical temperature T_N, three types of antiferromagnetic domains have been identified in $KNiF_3$ associated with the three cubic directions $\langle 100\rangle$.[6] When the system is excited by electromagnetic radiation in the presence of an external magnetic field, resonant absorption will take place in the different domains as long as some of the characteristic frequencies of the oscillatory motion of the magnetizations around their equilibrium positions equals the frequency ω_0 provided by the spectrometer.

Choosing the simple cubic base to define the xyz axis these characteristic frequencies for low-anisotropy antiferromagnets are approximately given by[5]

$$\omega^{\pm}\cong\mp\gamma H\left[1-\frac{\chi_{\parallel}}{2\chi_{\perp}}\right]+\gamma\left[2\lambda K+\left[\frac{\chi_{\parallel}H}{2\chi_{\perp}}\right]^2\right]^{1/2} \tag{2}$$

for the d_z domain, and

$$\omega\cong\gamma(2\lambda K+H^2)^{1/2} \tag{3}$$

for the d_x,d_y domains.

Here $\gamma=g\mu_B/\hbar$, and the external magnetic field has been assumed to lie along one of the cubic axes of the crystal. $\omega^{\pm}$ in (2) are associated with the positive and negative circularly polarized modes, respectively. Both modes are degenerate for $H=0$ and their roles are interchanged for $H\neq 0$ if the magnetic field orientation is reversed (Fig. 1).

In the absence of external field it follows from Eqs. (2) and (3) that all modes are degenerated and the frequency associated with the energy gap $g\mu_B\sqrt{2\lambda K}$, which depends on temperature through $\lambda(T)$ and $K(T)$, is given by

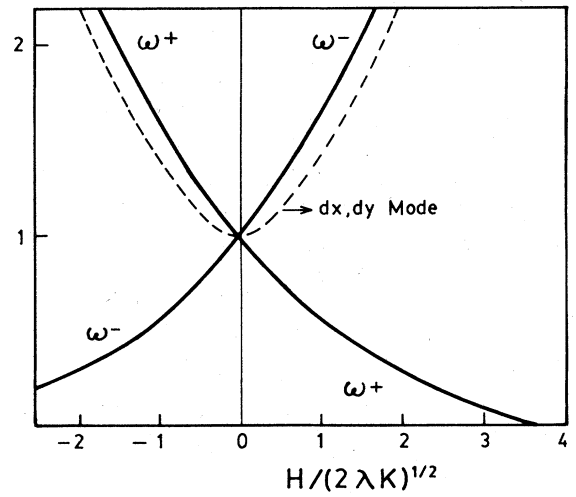

FIG. 1. Resonance frequency modes for the dz (full line) and dx-dy (dashed) domains vs H normalized to the energy gap $(2\lambda K)^{1/2}$. $\chi_{\parallel}/\chi_{\perp}$ of Ref. 10 corresponds to $T/T_N=0.95$.

$$\omega(H=0,T)=\gamma\sqrt{2\lambda K}\ . \tag{4}$$

At $T=0$ the phenomenological parameters can be approximated by[2]

$$K(0)\cong 2k/(2a^3),\quad \lambda(0)\simeq(6J/g^2\mu_B^2)(2a^2)\ ,$$

where a is the next-neighbor distance.

Ganot *et al.*[2] have performed measurements of $\lambda(T)$ and $K(T)$ by one-magnon Brillouin-scattering techniques within the range $5\leq T\leq 235$ K. They have shown that within the experimental uncertainty the exchange field

$$H_E(T)=-\lambda(T)M_0(T)$$

follows a Brillouin law up to 150 K. Assuming this behavior continues for $H_E(T)$ and $M_0(T)$ up to T_N [which implies that $\lambda(T)$ is practically independent of temperature], they found that $K(T)$ decreases rapidly from its maximum value at $T\cong 0$ to 0 at T_N. It follows from Eq. 4 that $\omega(H=0,T)$ is a decreasing function of T, its maximum being attained at $T=0$ and equals $\sqrt{24kJ}/\hbar$ which in the present case is about 110 GHz.

By working at a fixed frequency ω_0, zero-field resonance would be observed at a temperature T if

$$\omega(H=0,T)=\omega_0\ . \tag{4'}$$

By changing the operating frequency ω_0 it would be possible to measure $\omega(H=0,T)$ as a function of T and the higher the frequency ω_0, the lower the required temperature will be to reach condition (4'), provided that in all cases

$$\omega_0\leq\frac{1}{\hbar}\sqrt{24kJ}\ ,$$

otherwise no resonance could occur.

The presence of the magnetic field modifies these conditions according to Eqs. (2) and (3). In this case we expect that by lowering T from T_N the fields at which resonances occur [Eqs. (2) and (3)] will decrease with T up to a temperature in which $\omega_0\sim\gamma\sqrt{2\lambda K}$. Below that temperature the $dx-dy$ resonances disappear and only resonances from the dz domain will be observed. Above that temperature the sweeping of the magnetic field will give a spectrum having contributions from the three domains. In principle, these will appear at different values of H but the possibility of resolving them into individual peaks will obviously depend on the linewidths as compared with their separation in field.

In this paper we present a study of the AFMR in $KNiF_3$ performed at $\omega_0\sim 9.3$ GHz (X band). The spectrum has been measured as a function of temperature in the neighborhood and below $T_N=247$ K, covering the short range in T at which it is recognizable, for under our experimental conditions the spectrum is not visible below 230 K.

EXPERIMENTAL

A single crystal of $KNiF_3$ was mounted at the end of a copper rod ($\phi\sim 2$ mm) held at vacuum while the other end of the rod was maintained in contact with a liquid-nitrogen bath. A heater was added in order to perform measurements at different temperatures, the stability in temperature being better than 1 K. The intensities of the spectra at each temperature were normalized using the spectrum of a second $KNiF_3$ sample maintained at constant 300 K. For this purpose the variable temperature setup and the reference sample were inserted in an X band dual sample cavity (Varian Associates V-4531 dual) and the first derivative of the spectrum was obtained by the use of field modulation techniques. In order to discard the possibility of a dispersive component contribution we checked the spectrometer tuning by means of the simultaneous recording of the $KNiF_3$ spectrum and a standard one of P-doped silicon with known line shape.

The measured spectra at $T=244.5$, 242.5, and 232.5 K are shown in Fig. 2. The g factor and the line width in the paramagnetic phase ($T>T_N$) are coincident with those reported for a polycrystalline sample.[7] In the antiferromagnetic phase each spectrum consists of a very broad line with line widths which are of the same order or even greater than the resonance central field H_r as can be observed in Fig. 2. The variations of the line shape with T are caused by the large line widths and the strong dependence of the resonance field H_r (Fig. 3) with temperature implying that strong contributions of the line centered at $H<0$ (Fig. 1) over the recorded spectrum are to be expected.

In order to analyze the spectra we started with the phenomenological Landau-Lifshitz equation, which for an antiferromagnetic system can be written as[8]

$$\frac{d\mathbf{M}_i}{dt}=\gamma(\mathbf{M}_i\times\mathbf{H}_{Ti})-\frac{\alpha\gamma}{M_i}\mathbf{M}_i\times(\mathbf{M}_i\times\mathbf{H}_{Ti}),\quad i=1,2\ . \tag{5}$$

Here, α is the Landau-Lifshitz parameter, closely related to the intrinsic relaxation time[8] $\tau_i=(\alpha\gamma M_i)^{-1}$, and

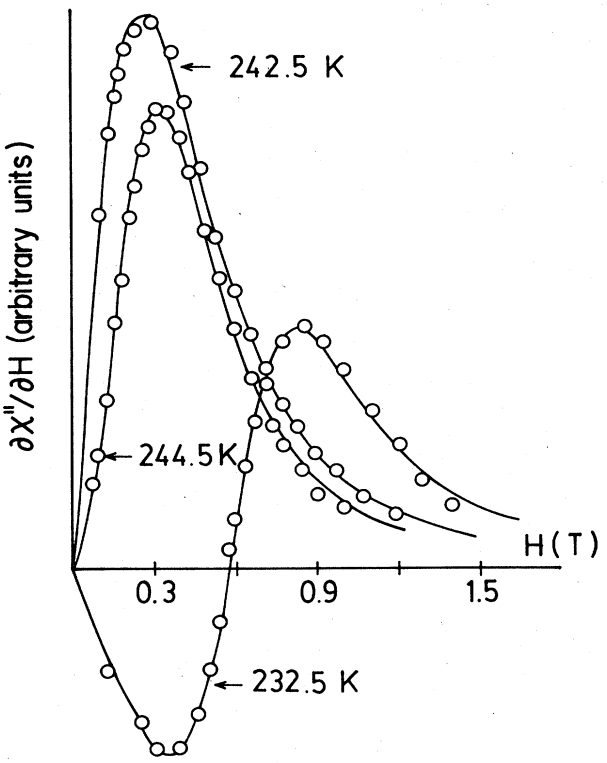

FIG. 2. AFMR spectra of $KNiF_3$: open circles correspond to experimental points taken from the spectrum, full lines represent the best fitting obtained with Eq. (7). The amplitude of the spectrum at 232.5 K was magnified by a factor of 3.

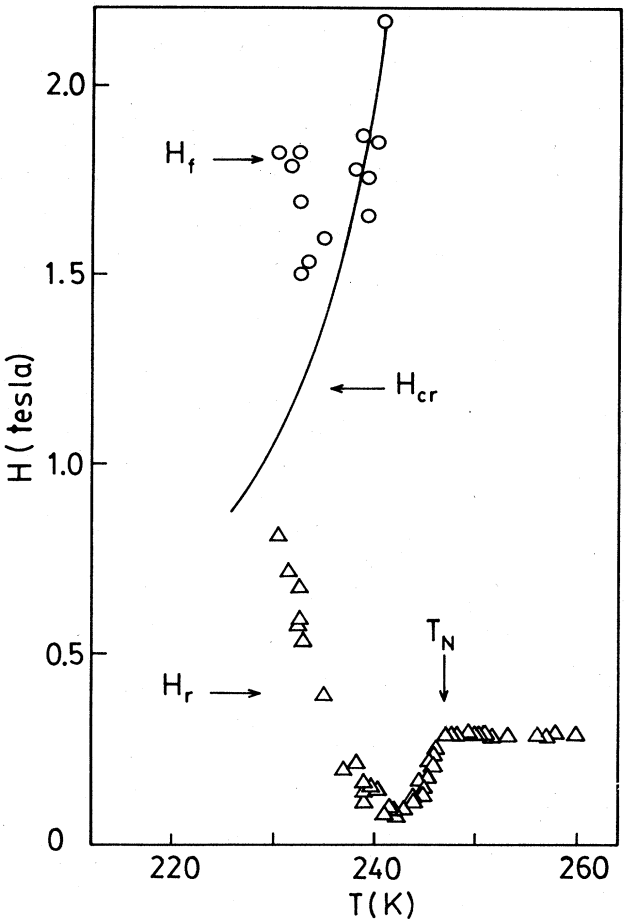

FIG. 3. Resonance fields (open triangles) obtained by fitting the spectra to Eq. (7), against temperature. H_f (open circles) and H_{cr} (full line) are also shown and were obtained as described in the text.

$\mathbf{H}_{Ti} = -\partial F/\partial \mathbf{M}_i$ is the total field on the i sublattice. The imaginary part of the rf susceptibility χ'', which is proportional to the absorption spectra, was calculated from Eq. (5) for H applied parallel to an easy axis of magnetization and is given by

$$\chi''^{(\pm)} = \frac{\omega}{\gamma}\alpha M_0 \frac{[H(1-\chi_{||}/\chi_\perp)\pm\omega/\gamma]^2}{[(H\pm a_1)^2 - a_2^2]^2 + a_3^4} , \tag{6}$$

where

$$a_1 = \frac{\omega}{\gamma}\frac{1-\chi_{||}/2\chi_\perp}{1-\chi_{||}/\chi_\perp} ,$$

$$a_2 = \left\{ \frac{2\lambda K\left[1-\frac{\chi_{||}}{\chi_\perp}\right] + \left[\frac{\omega/\gamma}{\chi_{||}/2\chi_\perp}\right]^2}{\left[1-\frac{\chi_{||}}{\chi_\perp}\right]^2} \right\}^{1/2} ,$$

$$a_3 = (2\lambda M_0 \alpha\omega/\gamma)^{1/2} .$$

In Eq. (6) we have performed the usual approximation $\alpha^2 << 1$.[8] The expression for $\chi''^{(\pm)}$ of Eq. (6) coincides in the limit $T\to 0$ with the line shape calculated by Heller *et al.*[9]

The line shape for the derivative spectrum is

$$\frac{\partial\chi''}{\partial H} = \frac{\partial\chi''^{(+)}}{\partial H} + \frac{\partial\chi''^{(-)}}{\partial H} . \tag{7}$$

Our experimental data were fitted to this expression (Fig. 2) using values of $\chi_{||}$ and $\chi_\perp$ deduced from Refs. 10 and 11.

RESULTS AND DISCUSSION

The least-square fitting procedure provides estimates for anisotropy constant K and the intrinsic line width $\alpha\cdot M_0$. The values obtained are shown in Fig. 4. Within our limited temperature range we found that our results for K are in good agreement with the corresponding values deduced from the data of Ganot *et al.*[2] The dependence of H_r versus T is shown in Fig. 3. It can be observed that between the Néel temperature (247 K) and 242.5 K, H_r decreases with T to nearly zero, where the driving frequency ω_0 equals the energy-gap $\omega(H=0)$

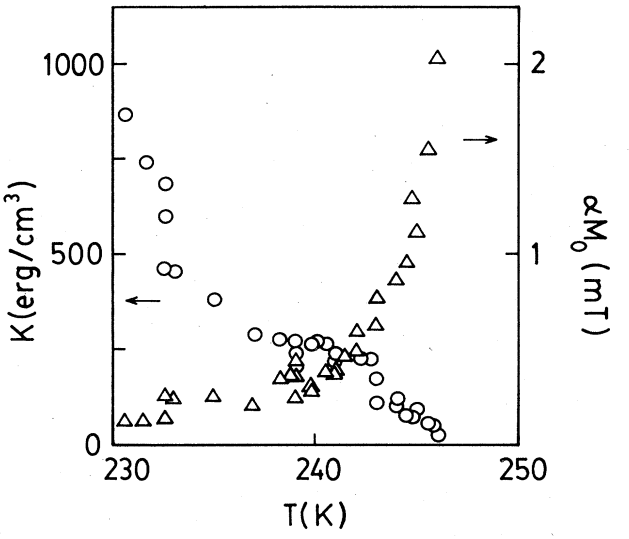

FIG. 4. Variation of the anisotropy (open circles) and intrinsic line width (open triangles) parameters with temperature; obtained by fitting the spectra to Eq. (7).

$=\gamma\sqrt{2\lambda K}$ and increases again by further diminishing T. Above that point the spectrum contains contributions from both the dz and the $dx-dy$ resonances, which are not resolved due to the broad line width; while for $T<242.5$ K only the dz resonance contributes. This is in contrast with the situation in the isomorphic compound $RbMnF_3$ where X band AFMR has been observed up to 4.2 K. In this case the energy gap at $T=0$ is about an order of magnitude smaller than the corresponding one in $KNiF_3$ and therefore the disappearance of the $dx-dy$ will be produced at temperatures which are much lower than 242 K.[12] A further analysis of the experimental data above 242.5 K would require the inclusion of the $dy-dx$ resonance contributions to Eq. (7), and the knowledge of the $M_0(T)$ function.

Petrov *et al.*[3] have performed AFMR measurements in $KNiF_3$ working at $\omega_0\sim 35$ GHz (Q band) within 230 $K<T<T_N$ for an arbitrary orientation of the crystal with reference to the external field. Although it is difficult to establish quantitative comparisons with our data they also observed that the resonance field H_r decreases from 1.2 T at $T=T_N$ about 0.75 at $T=230$ K. This behavior is coincident with our observation in the X band. By the use of the data of Ref. 2 an estimate for the temperature at which $\gamma\sqrt{2\lambda K}$ will reach a value of ~ 35 GHz gives 225 K. Therefore it is expected that within the temperature range covered in their experiment no minimum of H_r versus T will be observed as would have been our case if we had limited ourselves to measurements within 242.5 $K<T<T_N$.

Concerning the line width $\Delta H_{1/2}$ of the AFMR spectrum, this is in general a complicated function of the "intrinsic" line width αM_0.[8] It is worthwhile to point out that $\Delta H_{1/2}$ presents a minimum at $T=T_N(\Delta H_{1/2}\sim 0.3$ T) and increased rapidly as T decreases. A similar behavior has been found recently from measurements of the line width in the isomorphic compound $RbMnF_3$ at C band (~ 5.4 GHz).[13] However, it is difficult to identify the mechanisms which are responsible of the dependence of $\Delta H_{1/2}$ with T in the neighborhood of T_N. For compounds such as $RbMnF_3$ or others involving S-state ions estimates of $\Delta H_{1/2}$ based on magnon-phonon relaxation processes give results which are usually too small[14] as compared with the experimental ones. Rezende and White[14–16] have proposed a different mechanism based on multimagnon-relaxation processes which allow for a better agreement with experiments. If in our case we assume that the phenomenological parameter $(\gamma\alpha M_0)^{-1}$ can be identified with an effective relaxation time, an estimate of the number of magnons required to fit the data gives a number of around 20. This indicates that the picture based on multimagnon relaxation loses its significance[17] at least within the very limited range of $0.92\leq T/T_N\leq 1$ covered by our experiment.

On the other hand, from the behavior of $\Delta H_{1/2}$ with T in the paramagnetic region[7] we have evidence that processes involving the spin-phonon coupling can be much more important in $KNiF_3$ than in compounds based on S-state ions. This suggests that with reference to $KNiF_3$ more than a single mechanism could contribute to the AFMR line width at the observed temperatures.

With reference to the decrease in the spectrum intensity below 242.5 K up to its virtual disappearance at around 230 K we relate this with the rising up of H_r. In fact, for the antiferromagnetic case $\chi_{\parallel}\leq\chi_{\perp}$,[5] so when H reaches a certain magnitude the spin configurations with $\mathbf{M}_1-\mathbf{M}_2=\mathbf{M}\|\mathbf{H}$ become unstable and $\mathbf{M}$ reorients itself perpendicular to H, causing the extinction of the dz spectrum. Such reorientation can be attained by means of any of the two following mechanisms:

(1) The critical spin-flop transition observed in uniaxial compounds. In this case $\mathbf{M}$ would be reoriented if H equals H_f, given by[5]

$$H_f\left[\frac{2\lambda K}{1-\chi_{\parallel}/\chi_{\perp}}\right]^{1/2}. \tag{8}$$

In Fig. 3 we show H_f versus T deduced from the corresponding K values given in Fig. 4.

(2) The antiferromagnetic wall motion.[18] Through this mechanism the dx and dy domains grow at the expense of the dz domains as H increases. The wall motion starts at a threshold field $H_{\rm th}$ and ceases at a critical field $H_{\rm cr}$ when the displacement equals the spacing L between wall and the dz domain disappear. $H_{\rm th}$ and $H_{\rm cr}$ are given by[19,20]

$$H_{\rm th}=\left[\frac{2\lambda B}{1-\chi_{\parallel}/\chi_{\perp}}\right]^{1/2}, \tag{9}$$

$$H_{\rm cr}=\left[H_{\rm th}^2+\frac{2\lambda CL}{1-\chi_{\parallel}/\chi_{\perp}}\right]^{1/2}, \tag{10}$$

where B and C are constants associated to coercive and restoring terms which are added to F [Eq. (1)]. The values of $H_{\rm cr}$, also plotted in Fig. 3, were calculated using the determination of CL by Safa and Tanner[20] and assuming $B\ll CL$.

In choosing between both mechanisms for the spectra disappearance below 230 K we can make the following comments:

(a) We have observed a gradual loss of the *dz* spectrum intensity. This fact points against a critical process like the spin-flop transition.

(b) From Fig. 3 it can be seen that the disappearance of the *dz* spectrum occurs precisely at the temperature in which both curves H_r and H_{cr} cross over.

From these arguments, we conclude that the wall-motion mechanism very likely causes the extinction of the *dz* spectrum. Besides, by choosing H_r as a mean magnetic field applied to the sample this experiment confirms indirectly the T dependence of H_{cr} of Eq. (10) proposed by Petit *et al.*[19] and the *CL* value determined by Safa and Tanner.[20] As additional evidence in favor of the domain-reorientation mechanism, Engelsberg[21] has shown that the strong effects in the FNMR line shape[19] of $KNiF_3$, observed when the resonance field is increased from 0.5 to 0.74, can be explained by assuming that the $dx-dy$ domains grow at the expense of the *dz* domain. It is, however, worthwhile to mention that it would be difficult to argue additionally in favor of domain reorientation on the grounds of a less costly energy process, for in the present case $H_{cr} \sim H_f$.

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