Ferromagnetic resonance and the phase diagram of a 2D ferromagnet $(CH_3NH_3)_2CuCl_4$

S. O. Demokritov, N. M. Kreĭnes, V. I. Kydinov, and S. V. Petrov
Institute of Physics Problems, Academy of Sciences of the USSR

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The spectrum of the ferromagnetic resonance of a 2D ferromagnet $(CH_3NH_3)_2CuCl_4$ is measured in a magnetic field perpendicular to the easy plane. The *H-T* phase diagram is determined. The data obtained experimentally cannot be described in terms of the standard model for an easy-plane ferromagnet.

The study of 2D magnetic systems with a two-component order parameter (*XY* ferromagnets or antiferromagnets) has recently been the focus of considerable attention.[1] The unusual phase transition in these magnetic systems, which was predicted by Berezinskiĭ[2] and Kosterlitz and Thouless[3,4] (the BKT transition), has attracted special attention.

In the present letter we report the results of a study of the compound $(CH_3NH_3)_2CuCl_4$, which is a typical 2D ferromagnet with an easy-plane anisotropy.[5,6] This crystal, which has a face-centered orthorhombic structure with unit-cell parameters $a = 7.30$ Å, $b = 7.54$ Å, and $c = 18.55$ Å, consists of $CuCl_4$ layers separated by two (CH_3NH_3) layers.[7] The ferromagnetic exchange interaction beween the magnetic copper ions Cu^{2+} ($S = 1/2$) in the layer has a nearly Heisenberg nature with $J = 19.6$ K, and the corresponding exchange field is[5] $H_E = 530$ kOe. The weak anisotropy ($\sim 1\%$) of this exchange tends to line up the spins in the basal plane, which corresponds to the effective field[5,8] $H_A \approx 1.5$ kOe. This easy-axis anisotropy accounts for the fact that at low temperatures the compound $(CH_3NH_3)_2CuCl_4$ behaves as a 2D-*XY* ferromagnet. Because of the presence of a very weak uniaxial anisotropy in the plane, $H_a \approx 80$ Oe, and because of a small exchange between the spins in the adjacent planes,[5,8] $H_e \approx 50$ Oe, below[5,6] $T_c = 8.9$ K this crystal develops ferromagnetic order.

The field of the easy-plane anisotropy H_A of this material was determined experimentally in large fields: from the field dependence of the projection of magnetization along the hard-magnetization axis,[5] $M_{\parallel}(H)$, and from the measurement of the ferromagnetic resonance at the frequency of 9 GHz. The analysis of the results of Ref. 8 is based on the well-known formulas describing the dependence of the resonance frequency of the ferromagnet on the external field **H** which is perpendicular to the easy plane:

$$\nu = \gamma \sqrt{H_a H_A \left[1 - \left(\frac{H}{H_A}\right)^2\right]}, \quad \text{for } H \lesssim H_A \tag{1a}$$

$$\nu = \gamma \sqrt{(H - H_A)(H + H_a - H_A)} \approx \gamma (H - H_A), \quad \text{for } H > H_A \tag{1b}$$

It can be seen from Eq. (1) that at $H = 0$ the ferromagnetic resonance spectrum has a gap $\Delta = \gamma\sqrt{H_a H_A}$, which vanishes in the external field $H = H_A$. This feature of the spectrum is attributable to the spin-flip phase transition: In the field $H = H_A$ the magnetic moment lines up perpendicular to the easy plane; $M_{\parallel}$ in this case becomes saturated and the projection in the plane $M_{\perp}$ vanishes.

In our experiment we measured the ferromagnetic resonance frequency ν of $(CH_3NH_3)_2CuCl_4$ versus the external magnetic field H applied perpendicular to the easy plane (within 0.5°) at the temperatures 1.5–20 K in the frequency range 100–4500 MHz. The crystal, $2\times2\times0.15$ mm in size, was placed into a stripline. The rf field **h** was directed perpendicular to **H**. The field dependences $\nu(\mathbf{H})$ which we measured for several temperatures are illustrated in Fig. 1. For comparison, we have included in this figure several curves calculated from Eq. (1) for the same temperatures.

We see that at low frequencies and hence low fields, the ferromagnetic resonance spectrum cannot be described by the standard equations [Eq. (1)] for an easy-plane ferromagnet. Only at a rather large magnetic field (at the ferromagnetic resonance frequencies $\nu \gtrsim 3.5$ GHz) does the behavior of the crystal correspond to an easy-plane ferromagnet which is magnetized along the hard-magnetization axis. The ferromagnetic resonance spectrum in this case is described by the curve $\nu = \gamma\ (H - H_A)$ [see Eq. (1)], where the anisotropy H_A is consistent with the measurements of Refs. 5 and 7. In the magnetic field H_A^* (see Fig. 1) the ferromagnetic resonance spectrum reveals a structural feature which we attribute to the phase transition of the system. This transition, however, occurs in a field much lower than that of the spin-flip transition, H_A, of an ordinary easy-plane ferromagnet.

Figure 2 shows the temperature dependences $H_A^*(T)$ and $H_A(T)$ found from an analysis of the experimental spectra. The field H_A^* corresponds to maximal damping of the resonance frequency in our experiments. The anisotropy field H_A was found by extrapolating the rf part ($\nu \gg 3.5$ GHz, $H \gg H_A^*$) of the measured spectrum on the basis of Eq. (1).

It can be seen in Fig. 2 that H_A^* vanishes at $T \cong 9$ K. This temperature is the same as the temperature at which the ferromagnetic order appears. At $T = T_c$ the field H_A

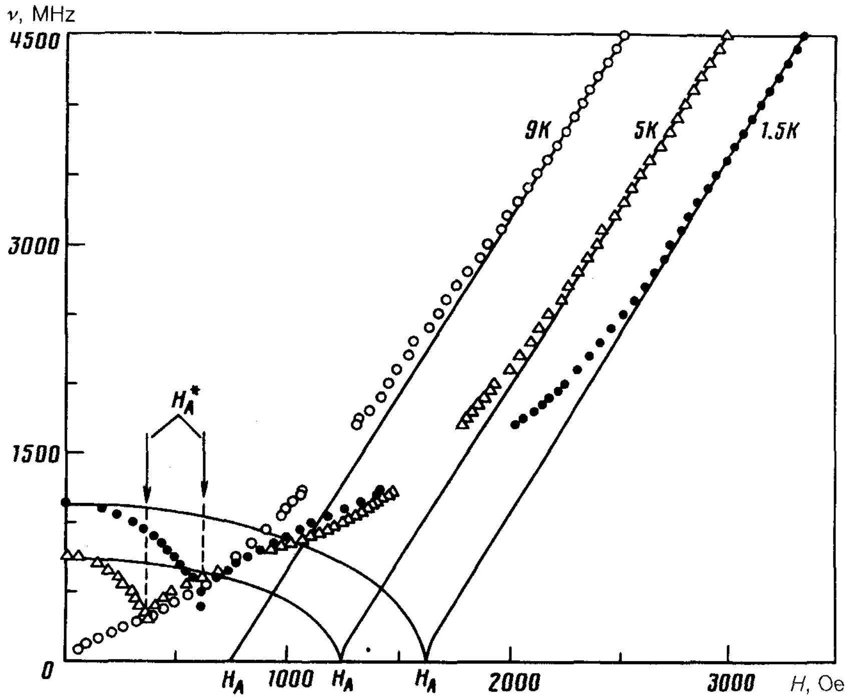

FIG. 1. The ferromagnetic resonance spectrum of a 2D ferromagnet $(CH_3NH_3)_2CuCl_4$ in a magnetic field perpendicular to the easy plane at various temperatures. Solid lines—the ferromagnetic resonance spectra calculated from Eq. (1).

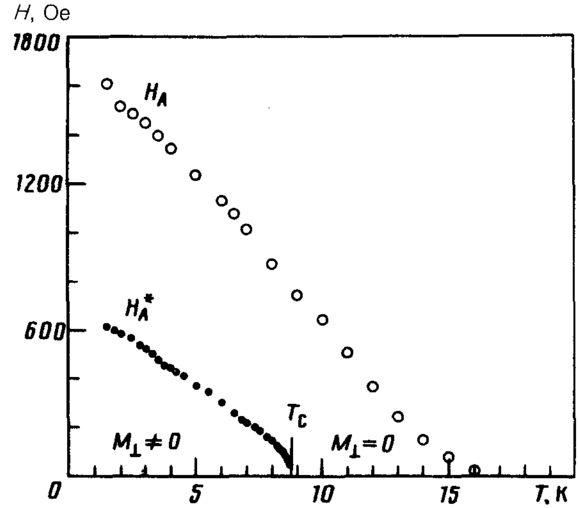

FIG. 2. Temperature dependence of the easy-plane anisotropy field $H_A(T)$ and the phase-transition field $H_A^*(T)$ of $(CH_3NH_3)_2CuCl_4$ in a magnetic field perpendicular to the easy plane. The values of $H_A(T)$ and $H_A^*(T)$ were found from the analysis of the ferromagnetic resonance spectra.

has no structural features and it vanishes only at $T \approx 16$ K, which is approximately equal to the temperature of the exchange in the layer, J. The values of $H_A^*(T)$ and $H_A(T)$, which were extrapolated to $T=0$, differ by nearly a factor of three $(H_A^* \ll H_A)$.

The line $H_A^*(T)$ tends to the point $T = T_c$ as $H \to 0$. This point separates on the $H=0$ axis the phases with a spontaneous magnetic moment in the $-M_\perp$ plane from these without it. The single phase transition which is seen in the structural feature of the ferromagnetic resonance spectrum at $H = H_A^*(T)$ can therefore be attributed to the disappearance of the magnetic-moment projection $M_\perp$ at a given temperature. Figure 2 can then be regarded as a phase diagram of a 2D ferromagnet $(CH_3NH_3)_2CuCl_4$ in a magnetic field perpendicular to the easy plane and the line $H_A^*(T)$ can be regarded as a line describing the phase transitions, whose order parameter is $M_\perp$. The line $H_A(T)$ in Fig. 2 does not correspond to an actual phase transition. H_A characterizes the scale of the fields above which the system begins to behave in a manner similar to an ordinary easy-plane ferromagnet.

On the basis of the given hypothesis the magnetization of the crystal should therefore behave in the following manner: when $H < H_A^*$ we have $M_\perp \neq 0$ and when $H \geqslant H_A^*$ we have $M_\perp = 0$. On the other hand, according to the magnetic measurements of Ref. 5, and according to our measurements of the samples, the component of the magnetization along the hard-magnetization axis, $M_\parallel$, changes linearly when $H < H_A$: $M_\parallel = M_0(H/H_A)$ and becomes saturated only in the field $H = H_A \gg H_A^*$ (M_0 is the saturation magnetization). $M_\parallel$ has no structural features in the phase-transition field, $H = H_A^*(T)$.

The fact that there is a single phase transition in a magnetic field, which is much smaller than the anisotropy field, even at $T \ll T_c$, is inconsistent with the standard model for an easy-plane ferromagnet. On the other hand, the experimental data in the literature are fully described by this model. There is yet no explanation for this discrepancy. Such an unusual behavior, however, apparently stems from the two-dimensional nature of this magnetic system.

We note in conclusion that these studies were stimulated by the work of V. L. Pokrovskiĭ and G. V. Uĭmin on the low-dimensionality magnetic materials, for which we are deeply indebted to them.

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