Antiferromagnetic resonance in an intermediate state of $CuCl_2{\cdot}2H_2O$

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Experiments show that when the external magnetic field is aligned precisely along the easy axis of the $CuCl_2{\cdot}2H_2O$ crystal the frequency-field dependence of the antiferromagnetic resonance has a discontinuity at the field corresponding to a flipping of the magnetic sublattices. The results thus show that the magnetic oscillations in the domains of different phases occur independently in the intermediate state.

In antiferromagnetic (AFM) crystals of finite dimensions, the spin-flip transition occurs through an intermediate state which is a stable structure comprised of domains of the AFM and spin-flip phases.[1,2] An antiferromagnetic resonance (AFMR) has been studied previously[3,4] in the intermediate state in $CuCl_2{\cdot}2H_2O$. Aside from Refs. 3 and 4, there has been only a single study of the AFMR in the intermediate state: in MnF_2 crystals.[5] In Refs. 3 and 4 the AFMR frequencies were calculated, and the experimentally observed continuous frequency-field dependence of the AFMR in the intermediate state was interpreted, under the assumption of an "averaging" of the uniform oscillations of the magnetization in a sample partitioned into domains. When the external magnetic field H was aligned precisely along the easy axis of the crystal in Ref. 5, however, a discontinuity was observed in the frequency-field dependence of the AFMR in the intermediate state of MnF_2.

In the present letter we suggest that the AFMR occurs independently in the domains of each phase, since the magnetization oscillations in different domains cannot be strongly coupled because of the low magnetic susceptibility of an antiferromagnet. Since the internal magnetic field in an antiferromagnet in the intermediate state remains constant,[2] the AFMR frequencies in the intermediate state should also be essentially independent of H. The difference between the frequencies before and after the spin flip is far greater than the width of the AFMR lines; this circumstance is responsible for the discontinuity on the frequency-field dependence.

The results found in Refs. 3 and 4 are inconsistent with the results of Ref. 5, so that further experiments are required to refine the mechanism for magnetic excitations in the AFMR in the intermediate state. We have studied the AFMR in $CuCl_2{\cdot}2H_2O$ in an external magnetic field oriented precisely along the easy axis of the crystal.

The spectrometer used in the experiments has been described elsewhere.[6] The working temperature of about 1.8 K is attained by pumping helium vapor from a cryostat holding the sample. The wavelength is measured within 0.2% by a resonator wavemeter. By simultaneously detecting the absorption signal and the magnetic susceptibility signal dM_a/dH, we can determine the difference between the magnetic field at the spin flip from the calibration point H_c within 10^{-3} T. The calibration point is

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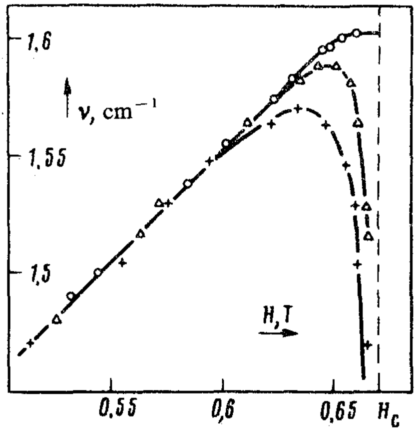

FIG. 1. Frequency-field dependence of the antiferromagnetic resonance for various angular deviations. O—$\psi = 0$; Δ—$\psi = 10'$; $+$—$\psi = 60'$.

the maximum of the dM_a/dH signal. The nonuniformity of H in the sample—a plate with dimensions of $0.3\times3\times3$ mm—does not exceed 0.1% of the magnitude of the field. The sample is initially oriented to maximize the amplitude of the dM_a/dH signal, and then it is oriented to put the AFMR absorption at a frequency as high as possible near H_c. The precision of the orientation in the **ab** plane is $\pm 3'$.

Figure 1 shows the experimental dependence of the AFMR frequency on H for three values of the angle (ψ) which the field **H** makes with the **a** axis in the **ab** plane: $\psi = 0$, $\psi = 10'$, and $\psi = 60'$. In the case $\psi = 0$ there is a horizontal region on the frequency-field dependence as H approaches H_c: The absorption is detected (within the AFMR line, with its width of 0.008 cm^{-1}) only at the frequency 1.604 cm^{-1}. At $H = H_c$ there is a discontinuity on the dependence: Below this frequency, essentially no absorption is observed. The value $H_c = 0.67 \pm 0.01$ T agrees well with the known values of the sublattice flipping field, when allowance is made for the finite sample temperature, $\cong 1.8$ K. In the case of a slight deviation of **H** from the **a** axis ($\psi = 10'$) we see a descending region on the frequency-field dependence near H_c (Fig. 1), but below a certain frequency there is again no absorption (this frequency is determined within 0.05 cm^{-1}). At $\psi = 60'$ the absorption line at $H \cong H_c$ is observed only down to the lower boundary of the working range of the spectrometer, 1.3 cm^{-1}.

Figure 2 shows the initial spectrograms of the AFMR lines, along with the dM_a/dH signal. Within a frame the magnetic field increases (from left to right) essentially linearly. Spectrogram a corresponds to the upper point in Fig. 1 for $\psi = 0$. The absorption line is asymmetric. Its left wing corresponds to the AFMR in the AFM phase, while its right wing reflects the decrease in the intensity of this oscillation on the horizontal part of the dependence in the intermediate state with decreasing volume of the AFM phase. The observed width at half-maximum of the dM_a/dH signal at $\psi = 0$ is 0.006 T and is consistent with the estimate[3,4] of 0.004 T of the width of the region of the intermediate state. Spectrograms b and c were obtained at a lower frequency, at $\psi = 0$ and $\psi = 10'$, respectively. We see quite clearly that there is no absorption line at

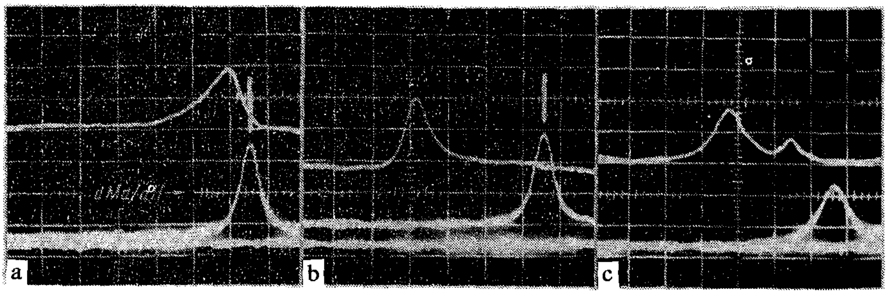

FIG. 2. Spectrograms of the antiferromagnetic resonance lines in $CuCl_2{\cdot}2H_2O$ (upper curves) at the following frequencies and angular deviations: a—1.604 cm^{-1}, $\psi = 0$; b—1.583 cm^{-1}, $\psi = 0$; c—1.583 cm^{-1}, $\psi = 10'$. The arrow shows the value $H_c = 0.67 \pm 0.01$ T.

$H = H_c$ for $\psi = 0$ and that this line appears when the orientation of the sample is changed ($\psi = 10'$).

The existence of an interval of magnetic fields in which the AFMR frequency remains constant and the existence of the discontinuity on the frequency-field dependence indicate the existence of an intermediate state in $CuCl_2{\cdot}2H_2O$. In this intermediate state the internal field in the sample remains constant, and the magnetic oscillations in the domains of different phases occur independently.

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