Antiferromagnetic resonance in the intermediate state of MnF_2

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Experimental data indicate the fact that near the intermediate state of an antiferromagnet the internal magnetic field does not change and that an antiferromagnetic resonance is excited in macroscopic domains of the antiferromagnetic and spin-flop phase independently of each other.

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If a magnetic field is oriented along the antiferromagnetism vector, then in a number of antiferromagnets, when the critical field is attained, a first-order phase transition occurs: reversal of the magnetic sublattices (spin-flop). In addition, if the specimen is an infinitely long cylinder, the transition occurs by a jump out of the antiferromagnetic phase (AP) into the reversed, spin-flop phase. In a specimen with finite dimensions, this transition occurs through the intermediate state,[1,2] which exists in the range of magnetic fields determined by the demagnetization factor of the specimen. The intermediate state is a thermodynamically stable, periodic structure made up of domains of the anti-

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ferromagnetic and spin-flop phases. The first experimental proof of the existence of an intermediate state in an antiferromagnet was obtained in Ref. 2 while studying the magnetization of tetragonal double-sublattice antiferromagnet: MnF_2.

The intermediate state in MnF_2 was then studied by using optical spectroscopy[3,4] and NMR.[3] It could be expected that the antiferromagnetic resonance (AFMR) will also show a special behavior, characteristic of the intermediate state. The indications of such behavior can be established by using the theoretical results in Ref. 1, wherein, in particular, it is shown that when the magnitude of the external magnetic field changes, the internal magnetic field in the antiferromagnetic in the intermediate state remains constant. Since the coupling between fluctuations in the domains of the antiferromagnetic and spin-flop phases cannot be strong due to the small value of the susceptibility of the antiferromagnet, it should be expected that the AFMR frequencies in the vicinity of the intermediate state will remain essentially constant as the external magnetic field is varied. Fluctuations in antiferromagnetic and spin-flop phases must occur independent of one another, while their intensities must be proportional to the fraction of the substance of the corresponding phase. However, in $CuCl_2 \cdot 2H_2O$[5] in the intermediate state the AFMR frequency was observed to vary smoothly with increasing field, which was interpreted theoretically as the appearance of collective oscillations of the coexisting phases in the intermediate state. The study of AFMR in MnF_2, for which the existence of an intermediate state was shown most convincingly,[2-4] was of greatest interest.

Single-crystal specimens of MnF_2 consisted of plates with an extended (001) plane with dimensions 3 X 3 mm and thickness from 0.05 to 0.3 mm. For specimens with this geometry, in accordance with the demagnetization factor, we could expect a width of about 0.1 T for the intermediate-state region. The experiment was performed using an arrangement[4] that allowed recording the susceptibility dM_z/dH along the C_4 axis, changing the orientation of the specimen to within 0.5′, and determining reliably the intensity of absorption lines. Observation of dM_z/dH simultaneously with the observed resonance lines allowed following the antiferromagnetic state, its orientation, and the behavior of AFMR, and it also gave a good reference point, namely, H_c, corresponding to the maximum dM_z/dH, relative to which the magnitude of the resonant field in the intermediate-state region was measured to within 10^{-2} T. The inhomogeneity of the magnetic field at the location of the specimen did not exceed 0.1% of the value of the field.

As mentioned above, it was expected that in the region of the intermediate state the AFMR frequencies would be independent of H [horizontal segments on the characteristic $\nu=\nu(H)$], which could be easily observed, if the spectrum is scanned in frequency $J(\nu)|_{H=\text{const}}$. However, existing microwave generators do not allow us to do this. Nevertheless, if we take into account the finite width of the resonance lines, then even with the usual scan performed in the experiment with respect to the field $J(H)|_{\nu=\text{const}}$, it is possible to investigate reliably horizontal sections of $\nu=\nu(H)$. Moreover, if spectra are recorded with generator frequencies differing from each other by an amount that is much smaller than the absorption line width, then it is possible to reproduce the shape of the absorption lines $J(\nu)|_{H=\text{const}}$, having the data $J(H)|_{\nu=\text{const}}$. Following this procedure, we were able to record the horizontal sections and to observe the broadening of the lines of the high frequency (HF) AFMR modes in the intermediate-state region from 0.1 cm^{-1} to 0.5 cm^{-1}. This broadening is apparently related to the fact that the resonance in the in-

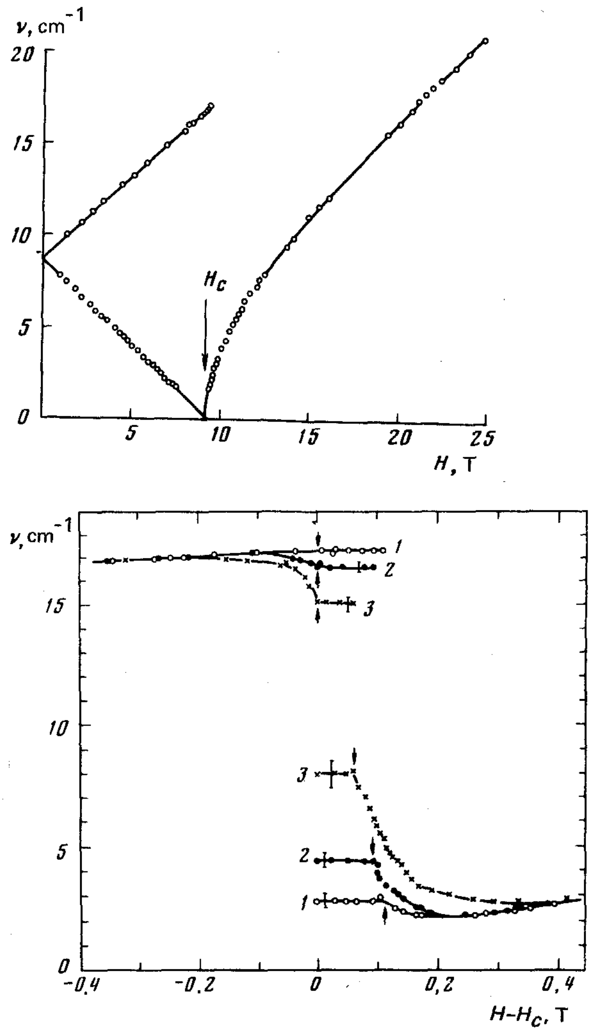

FIG. 1. The frequency-field dependence of AFMR in MnF_2 at $\psi = 0 \pm 1'$; the continuous lines represent the theory.[7,8]

termediate state for the specimen as a whole is not homogeneous, and different domains of each of the phases contribute to the line width.

Figure 1 shows the frequency-field dependence of AFMR for MnF_2 at a temperature of 4.2 K with H oriented exactly ($\pm 1'$) along the tetragonal C_4 axis. The specimens were roughly ($\pm 10'$) oriented according to the maximum dM_z/dH and exactly according to the absorption at the highest possible frequency for the HF AFMR mode. The experimental points follow closely the theoretical curves[7,8] with $g = 2.00$ and yield the frequency $\nu_{H=0} = 8.7\ \text{cm}^{-1}$, as in Ref. 7. Extrapolation of the characteristics of the low-frequency and post spin-flop mode to $\nu = 0$ allows us to determine the width of the metastable region: 0.1 ± 0.05 T, which agrees well with the theoretical estimates[8]: $H_1 - H_2 \simeq H_A\sqrt{2H_A/H_E} = 0.15$ T ($H_A = 0.88$ T; $H_E = 55.6$ T[7]).

Figure 2 shows on an enlarged scale the frequency-field dependence of AFMR in the region of the intermediate state for different ψ angles of inclination of the field rela-

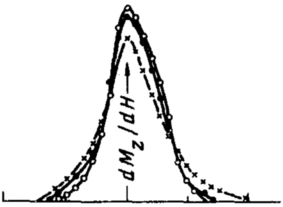

FIG. 2. The frequency-field dependence of AFMR in the vicinity of the intermediate state. The arrows indicate the boundaries of the horizontal sections, while the numbers next to the arrows correspond to the angles of inclination: 1) $\psi = 0'$; 2) $\psi = 10'$; 3) $\psi = 20'$, ○ are experimental points for $\psi = 0'$; • - $\psi = 10'$; × - $\psi = 20'$.

tive to C_4. The value of H_c, for which dM_z/dH reaches a maximum, is equal to 9.21 ± 0.05 T. On the horizontal parts of the characteristics $\nu = \nu(H)$, the intensity of the antiferromagnetic phase in the substance decreases with increasing field, while the spin-flop phase increases. For $\psi = \psi_{cr} \simeq 30'$ (Ref. 2), the horizontal sections disappear and the absorption lines exist in the frequency range from 2 to 17 cm^{-1}, while for $\psi < \psi_{cr}$, transparency windows are observed: Absence of absorption in some interval of frequencies and fields ΔH, corresponding to the intermediate state. The presence of horizontal sections on the characteristics $\nu = \nu(H)$ shows that when the external magnetic field varies, the internal field in MnF_2, in an intermediate state, remains constant, while the fluctuations in the antiferromagnetic and spin-flop phases occur independently.

In addition, it is evident from Fig. 2 that the horizontal sections of the characteristics $\nu = \nu(H)$ narrow with increasing ψ, which agrees with the narrowing of the intermediate-state region because of the decrease in the magnetization jump with an increase in ψ^2.

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