Nonlinear magnetic resonance in $(CH_3NH_3)_2Mn_{0.5}Cu_{0.5}Cl_4$ crystals

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A nonlinear microwave absorption similar to a classical nonlinear resonance of an anharmonic oscillator has been observed in $(CH_3NH_3)_2Mn_{0.5}Cu_{0.5}Cl_4$ crystals. The characteristics of the microwave absorption curve have been found as a function of the pump power. The temperature dependence of the critical power has been found.

The magnetic compounds of the family $(C_nH_{2n+1}NH_3)_2BX_4$, where $n=1, 2, \ldots$, B is a transition-group ion, and X is a halogen, exhibit a rich array of physical properties. Being quasi-2D crystals, they can serve as models for studying specific reduced-dimensionality properties. By replacing either the magnetic ions or the halogen (or both) and by varying n, one can control the type of magnetic order, the magnetic-ordering temperature, and the anisotropic properties. With $n=1$, for example, in the $C_2(MnCu)(ClBr)_4$ system $[C\equiv(CH_3NH_3)]$, various substitutions make it possible to switch from a ferromagnetic order to an antiferromagnetic order and from an easy-plane anisotropy to an easy-axis anisotropy.[1]

For this study we selected $C_2Mn_{0.5}Cu_{0.5}Cl_4$ crystals, which are at the middle of the Mn–Cu series. The compound C_2MnCl_4, at one end of this series, is an antiferromagnet with intraplane and interplane antiferromagnetic interactions, an easy anisotropy axis, and $T_N=45$ K. Along with the uniform mode of magnetic oscillations, the antiferromagnetic-resonance spectrum of this crystal contains a nearby exchange mode.[2]

The compound C_2CuCl_4, at the other end of this series, is an easy-plane ferromagnet with $T_c=8.9$ K. In this compound the behavior of the ferromagnetic resonance in the geometry in which the magnetic field is perpendicular to the ferromagnetic planes is associated with the presence of a (Berezinskiĭ–)Kosterlitz–Thouless phase.[3] It has also been observed that a bifurcation and a transition to a chaotic dynamic region occur at high microwave power levels.[4]

Crystals containing any single type of magnetic ion (either Mn or Cu) are quite interesting in themselves. One might thus expect that the mixed crystals $C_2(MnCu)Cl_4$ would exhibit an unusual combination of properties.

According to static magnetic measurements,[1] the $C_2Mn_{0.5}Cu_{0.5}Cl_4$ crystal exhibits a behavior corresponding to an easy-plane ferromagnet with $T_c \simeq 8$ K. However, the temperature dependence of the reciprocal paramagnetic susceptibility, which obeys a Curie–Weiss law at $T>110$ K, yields a negative value $\theta=-140$ K. At temperatures $T<110$ K the susceptibility varies in a nonmonotonic way. This behavior has tentatively

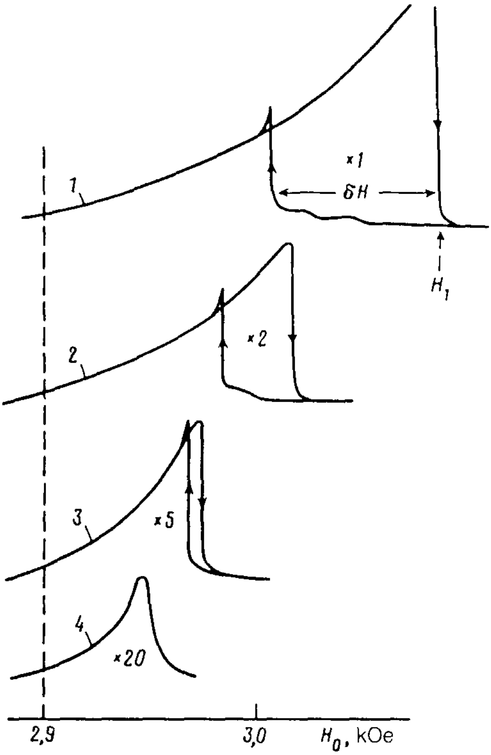

FIG. 1. Shape of the microwave absorption line at various power levels as the magnetic field is swept in different directions. 1—$q=0.75$; 2—$q=0.4$; 3—$q=0.15$; 4—$q=0.05$; ($q=P/P_{\max}$). The numerals are the gain values.

been attributed to regions in which a short-range magnetic order arises in the course of competing antiferromagnetic and ferromagnetic interactions with anisotropies of different signs.

The magnetic resonance was studied on the spectrometer described in Ref. 5. The source of electromagnetic radiation was a tunable Gunn-diode microwave source with a maximum power $P_{\max}=200$ W. We used a rectangular cavity for the frequency $f=10$ GHz and the TE_{102} mode with $Q\sim 1000$.

The test samples were platelets with a thickness $t\simeq 0.3$ mm and with dimensions 2×2 mm in the plane. The C axis of the crystal was oriented perpendicular to the plane of the platelet.

The external magnetic field was oriented in the plane of the sample. The magnetic component of the microwave field was oriented along the C axis.

At temperatures below the magnetic ordering temperature we observed resonant-absorption curves of unusual shape. At low microwave power levels (a few milliwatts) the line is slightly asymmetric, and its shape is independent of the direction in which the magnetic field is swept (curve 4 in Fig. 1). As the microwave power is raised, the height of the resonant peak increases. At a certain power level P_c, we find a hysteresis as the magnetic field is swept in the different directions (curve 3 in Fig. 1). With a further increase in the microwave power, the absorption intensifies, the absorption

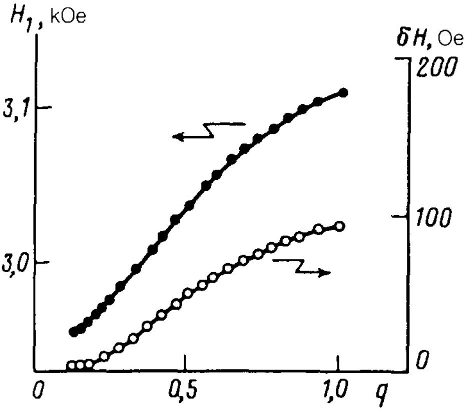

FIG. 2. The critical field H_l and the width of the hysteresis loop, δH, versus the microwave power level. $T=6.5$ K.

peaks shift to higher magnetic fields, and the hysteresis region becomes broader (curves 3 and 4 in Fig. 1).

In light of these results we would like to point out the following. On the one hand, we know[6] that a lineshape of this sort can be observed for an ensemble of absorbing centers for which the splitting between resonating levels has a Gaussian distribution. For such systems, however, one does not observe a difference in the shape of the curves as the magnetic field is swept in different directions. On the other hand, hysteresis effects occur in disordered systems of the spin-glass type. In such systems, there is a shift of the resonant peak when the direction of the magnetic-field sweep is changed, but there is essentially no change in the maximum absorption level.[7] In all these cases, an increase in the microwave power is accompanied by simply an increase in the absorption intensity, without any significant distortion of the absorption line.

All the formal, superficial indications in our case thus make the situation look very similar to a classical nonlinear resonance for an anharmonic oscillator.[8]

With this thought in mind, we studied some characteristics of the shape of the nonlinear-resonance line for various levels of the microwave power and for various temperatures. Figure 2 shows the field at which the microwave absorption is cut off, H_l, and the width of the hysteresis loop, δH, versus the power of the electromagnetic radiation. It was found that at power levels $P/P_{\max}=q>0.25$ the relationship between q and H_l is $q=q_0+bH_l^m$ $(m=3/2)$, where q_0 and b are adjustable quantities. Such a behavior is known[8] to characterize the stiffness of a system and to be determined by the nature of the nonlinearity.

Figure 3 shows the critical power $q_c=P_c/P_{\max}$ (P_c separates the regions with and without hysteresis) versus the temperature. An unexpected result here is that there is a hysteresis all the way to $T^*\simeq 15$ K, which is well above T_c. The hysteresis disappears above $T^*=15$ K, but the line remains asymmetric. The absorption line becomes symmetric again only above 30 K.

The data presently available on the $C_2Mn_{0.5}Cu_{0.5}Cl_4$ crystal are definitely insufficient for an unambiguous determination of the magnetic structure and the nature of

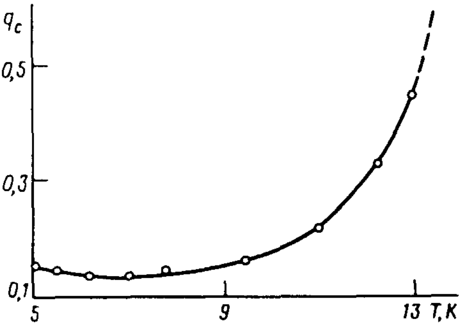

FIG. 3. Temperature dependence of the critical power $q_c = P_c/P_{\max}$.

the interactions between magnetic ions. However, the temperature dependence of the resonant absorption and a comparison with the purely manganese and copper compounds suggest that a coupling between the manganese and copper subsystems is playing an important role in this nonlinear behavior of the magnetic resonance.

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