Nuclear magnetic resonance mode interaction in $RbMnF_3$

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Nuclear magnetic resonance was investigated in the cubic antiferromagnet $RbMnF_3$ under strong frequency pulling conditions. A deviation of the NMR spectrum from the standard dependence is observed in the region of the nuclear magnetic resonant mode intersection. The deviation increases with increasing angle between the direction of the magnetic field and the [001] axis of the sample. The complicated behavior of NMR near the intersection point is discussed.

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At the present time $RbMnF_3$ is one of the widely investigated antiferromagnets. It is one of the known antiferromagnets with cubic symmetry. This results in the low value of the effective anisotropy field and in the low value of the sublattice-flipping field above which the magnetic structure can be adequately described. On the other hand, the magnetic properties of this crystal are the result of the Mn^{++} ion, the nucleus of which has nuclear magnetism. This leads to a coupling between the oscillations of the electron and nuclear magnetic systems, which causes in turn, owing to the joint oscillations of the electron and nuclear magnetizations, a peculiar NMR spectrum known as "dynamic frequency shift" or "pulling." An analysis of this phenomenon was presented by de Gennes *et al.*[1]

The antiferromagnetic resonance spectrum in $RbMnF_3$ was investigated by Teaney and Fraiser.[2,3] For a magnetic field stronger than its sublattice-flipping field and directed along the [100] axis, the spectrum is described by the formulas

$$\omega_{e1}^2 = \gamma_e^2 \left[H^2 - \frac{3}{2} H_A H_E + 2 H_E H_N \right], \tag{1}$$

$$\omega_{e2}^2 = \gamma_e^2 \left[3 H_A H_E + 2 H_E H_N \right]. \tag{2}$$

Nuclear resonance in this substance was investigated by Witt and Heeger,[4] and agrees well with the formulas of the theory of[1]:

$$\omega_{n1}^2 = \omega_{n0}^2 \left(1 - \frac{\omega_{eN}^2}{\omega_{e1}^2} \right) \tag{3}$$

The second nuclear-resonance branch, which corresponds to interaction with the field-independent AFMR branch, was observed in an investigation[5] of the damping of sound in $RbMnF_3$, and is described by formula (3) with index 2. Here γ_e is the gyromagnetic ratio for the electron, H is the external magnetic field, H_A is the crystalline anisotropy field, H_E is the exchange field, H_N is the effective field produced by the nuclei at the electrons as a result of the hyperfine interaction, ω_{n0} is the NMR frequency in the hyperfine field of the electrons, and $\omega_{eN}^2 = \gamma_e^2 2 H_E H_N$.

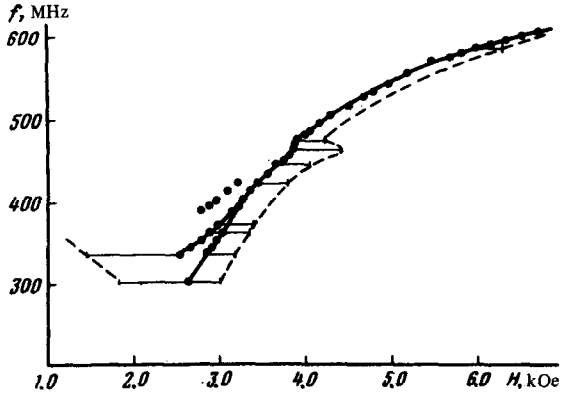

FIG. 1. Dependence of the NMR frequency in $RbMnF_3$ on the magnetic field. The solid curve corresponds to the absorption maxima and the dashed curves limit the line width.

We have investigated the field-dependent branch of the NMR—Eq. (3) with index 1. The nuclear resonance in $RbMnF_3$ is observed at frequencies on the order of 500 MHz, which called for the use of unique high-frequency circuitry. The sample was placed in a four-turn copper coil connected to a 75-Ω cable, which together with a phase shifter (line of variable length) forms a resonator. By varying the length of the resonator with the aid of the phase shifter it is possible to vary the resonant frequency in a wide range. It is thus easy to cover the frequency band that calls for investigation. The sample was placed in the coil of the tank circuit between the poles of an electromagnet. The NMR line was recorded at a fixed frequency and the magnetic field was scanned. The experiment was performed at 1.24 K. The sample was so oriented that the axis of the coil coincided with the [001] axis. The magnetic field could be rotated in the (010) plane.

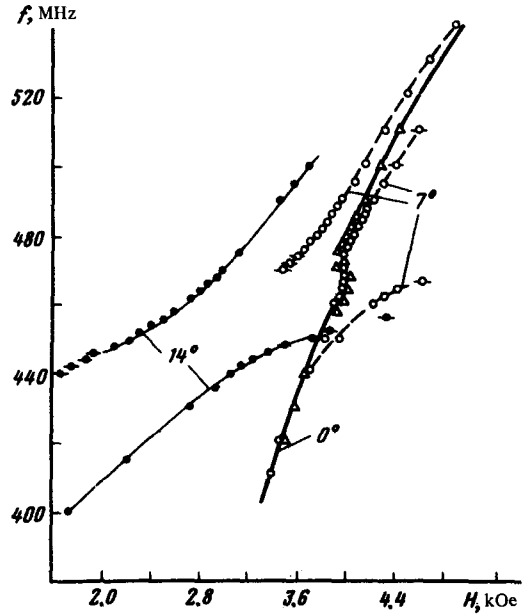

FIG. 2. Influence of the deflection of the magnetic field away from the [001] axis of the sample on the nuclear magnetic resonance spectrum.

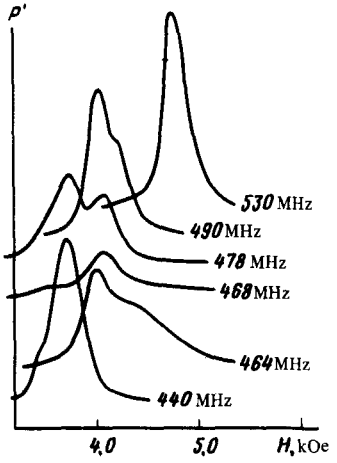

FIG. 3. Nuclear magnetic resonance line shape on going through the interaction point. The angle between the direction of the magnetic field and the [001] axis is 7°.

Figure 1 shows a general picture of the spectrum of the nuclear magnetic resonance with the external magnetic fields parallel to the [001] axis of the sample. At a magnetic field somewhat less than 4 kOe, a step-like distortion of the spectrum is observed. When the magnetic field is deflected away from the [001] direction, the distortion of the spectrum increases until splitting is observed, as shown in Fig. 2 for an angle of 7°. With further increase of the angle, the NMR spectrum shifts in accordance with the anisotropy, and the average frequency of the distortion of the spectrum decreases (14°). The splitting acquires in this case a similar form. Figure 3 shows plots of the absorption line for NMR in the splitting region at an angle of 7°, showing the transfer of the intensity from one branch to another, a rather long coexistence of the two branches in parallel, and the presence of a certain third branch (at a frequency 464 MHz).

The experiment demonstrates thus that in a certain frequency region (near 450 MHz) a coupling exists between the oscillations of the field-dependent NMR branch and some internal vibrational system. The parameters of this system correspond to the field-independent NMR branch [Eq. (3) with index 2]. This coupling increases with increasing deviation of the constant magnetic field from the [001] direction of the crystal. The coupling picture, however, is much more complicated than that expected for a doubly connected system of NMR modes. In the case of a doubly connected system, the asymptotic behavior of the branches far from the intersection region should tend to the behavior of noninteracting oscillations, but this is evidently not observed in the experiment.

The character of the splitting for a field directed at an angle 7° to the [001] axis (Fig. 2) shows the presence of two additive interaction mechanisms, one of which corresponds to splitting and the other to distortion of the spectrum in the region of 465 MHz. The complicated behavior of the NMR branches near the splitting, which can be partially ascribed to the domain structure of the sample, indicates nevertheless that some third vibrational system takes part in the interaction.

As to the interaction mechanisms, two of them can be indicated. Both are connected with the violation of the symmetry. Inasmuch as under dynamic

coupling conditions[1] the NMR spectrum reflects the antiferromagnetic resonance spectrum, we can assume that the AFMR modes exhibit a similar behavior. The perturbation of the AFMR spectrum near the intersection point was pointed out by Hinderks.[6] The influence of the deflection of the direction of the magnetic field from symmetrical on the coupling between the AFMR modes was investigated by Borovik-Romanov and Prozorova[7] in $MnCO_3$. Owing to the symmetry violation caused by the oblique direction of the field, the noninteracting AFMR branches are no longer natural frequencies of the antiferromagnet. The calculation in[7] led to good agreement with experiment. Similar results are produced also by internal stresses in the crystal, due to the defect structure and to the magnetoelastic properties. These stresses distort the cubic structure of the sample and lead to new natural oscillations.

The complicated behavior of the spectrum near the intersection point does not lead to the clear picture obtained in[7]. On the other hand, this complexity may indicate that a third system, most probably phonons, takes part in the interaction.

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