Mn^{55} nuclear acoustic resonance in antiferromagnetic $RbMnF_3$ near the Néel temperature*

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The frequency-temperature relation for nuclear resonance of Mn^{55} has been studied in the antiferromagnetically ordered phase of $RbMnF_3$, using ultrasonic methods. For $T \lesssim 60$ °K molecular-field theory and for $60\,°K \lesssim T \lesssim T_N$ (Néel temperature $T_N = 83$ °K) a modified molecular-field theory is found to be consistent with the experimental results. Evidence is found for a strong effect of coupling between nuclear and electronic spin-wave modes in $RbMnF_3$ near T_N.

We have studied the propagation of ultrasonic waves in the antiferromagnetically ordered phase of $RbMnF_3$, with special emphasis on absorption peaks due to antiferromagnetic and nuclear acoustic resonances. In previous communications[1-3] we reported on various aspects of antiferromagnetic resonance, particularly on its temperature and magnetic field dependence in the vicinity of the Néel temperature T_N. This note is concerned with nuclear acoustic resonance (NAR) of Mn^{55} below the Néel temperature. NAR in antiferromagnetically ordered phases has been studied in recent years in MnTe,[4] $RbMnF_3$,[5] and $CsMnF_3$.[6] However, to date, the observation of resonant interactions of ultrasonic waves with both electronic (antiferromagnetic) and nuclear-spin-wave modes has been reported only for $RbMnF_3$.[2,7] In $RbMnF_3$ the two magnetic modes couple strongly to each other and to elastic modes, which makes the study of the magnetic modes of this material by acoustic waves particularly interesting.

The experiments were carried out using the pulse-echo ultrasonic technique[8] and the frequency range covered was 100–800 MHz. All the data shown here were obtained using transverse ultrasonic waves propagating along the [110] and polarized along the [001] directions. This mode is particularly well suited to the study under consideration for the following reasons: (i) absence of "critical" attenuation, (ii) small resonant absorption, and (iii) low-sound velocity (the sample can be thinner).

Figure 1 shows the resonant absorption as a function of temperature, at several frequencies, obtained in the course of this investigation. Two sets of peaks appear, those labeled A_1, A_2, A_3 have been identified as due to acoustic antiferromagnetic resonance (AAFMR) and those labeled B_1, B_2 have been identified as due to nuclear acoustic resonance (NAR) of the Mn^{55} nucleus. Figure 2 shows the frequency-temperature (FT) relation of the NAR peaks in the absence of an externally applied magnetic field. Merry and Bolef's[9] results on the Mn^{55} NAR and Ince's[10] results on Mn^{55} NMR (using electromagnetic waves), both at lower temperatures ($T \lesssim 36$ °K), and Moran and Lüthi's[11] results near T_N, are also included in Fig. 2.

For purposes of comparison with the experimental results, the FT relation of the NMR modes is calculated by the four-sublattice model[10] and shown by the solid lines in Fig. 2. The parameters necessary for the calculation are obtained by applying the results of the molecular-field theory for $T \lesssim 60$ °K. For the temperature range $60\,°K \lesssim T \lesssim T_N$ a modified molecular-field theory was used, in which the temperature dependence of the sublattice magnetization M is assumed to be of the form $M \propto (T_N - T)^{1/3}$, in accordance with experimental observations.[12,13] The applicability of this empirical modification has been studied in the case of AAFMR and discussed at some length in a previous publication.[3] The agreement obtained between the experimental and theoretical results seen in Fig. 2 shows that the modified molecular-field

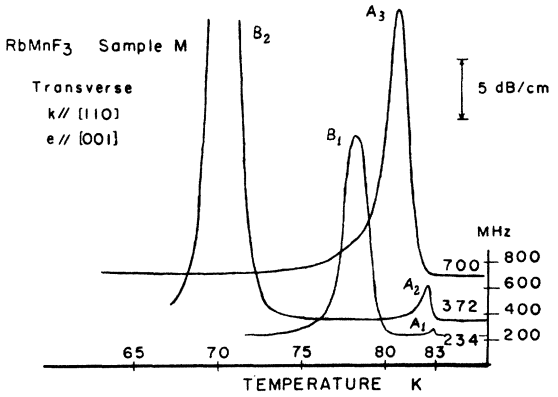

FIG. 1. Resonant absorption as a function of temperature, at several frequencies. Two sets of peaks appear, those labeled A_1, A_2, A_3 are due to acoustic antiferromagnetic resonance (AAFMR) and those labeled B_1, B_2 are due to nuclear acoustic resonance (NAR) of the Mn^{55} nucleus.

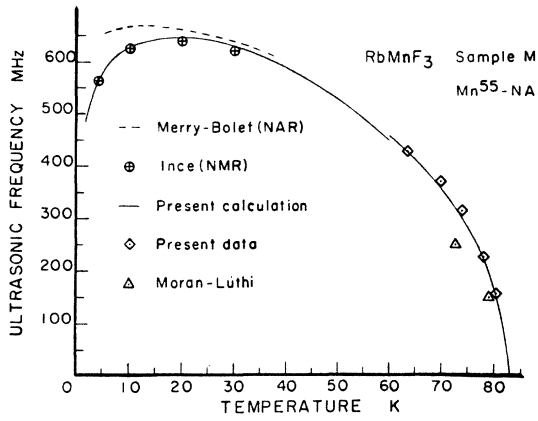

FIG. 2. Frequency-temperature relation for NAR in the absence of an externally applied field.

theory accounts well for the temperature dependence of NAR frequencies between ~60 °K and T_N, obtained in this study. It must be emphasized, however, that the observed NAR frequencies appear to depend somewhat on the samples used.[11] The differences noted between the results of Moran and Lüthi and ours, and between the results of Ince, and Merry and Bolef are probably examples of the sample dependence mentioned. The agreement shown in Fig. 2 should be considered, however, in the context of the fact that the modified molecular-field theory accounts for the temperature dependence of both the NAR frequencies and the AAFMR frequencies discussed previously.[3]

It is interesting, furthermore, to note that the temperature dependence of the observed Mn^{55} NAR frequency, ω_n, is

$$\omega_n \propto (T_N - T_n)^{0.50 \pm 0.02} \quad (60\ ^\circ\mathrm{K} < T_n < T_N)\ , \qquad (1)$$

where T_n is the temperature corresponding to the peak for a particular ω_n.

The exponent 0.5 in relation (1) is also expected to arise if the sublattice magnetization were to follow the Brillouin-function temperature dependence of the molecular-field theory. This is because in the molecular-field theory the temperature dependence of sublattice magnetization M is $M \propto (T_N - T)^{1/2}$ near the Néel temperature, and the hyperfine field H_{NN}, which is responsible for the nuclear precession, is related to the magnetization by $H_{NN} \propto M$. As mentioned previously, for $RbMnF_3$, $M \propto (T_N - T)^{\beta}$ where $\beta \sim \frac{1}{3}$. It is concluded, therefore, that the exponent 0.50 in relation (1) does not come from the Brillouin-function temperature dependence of M, but from a large "frequency-pulling"[14] effect on the nuclear modes owing to their coupling with electronic modes. This effect is particularly pronounced in $RbMnF_3$ because of the rather low frequencies of the electronic modes. The conclusion is substantiated by the agreement between experimental and calculated results seen in Fig. 2, the latter being based on $M \propto (T_N - T)^{1/3}$.

For the other antiferromagnetic materials in which the NARs have been observed to date, i.e., MnTe[4] and $CsMnF_3$,[6] the reported exponents in the form of Eq. (1) are 0.35 and 0.30, respectively, near the Néel temperature, in close relation to the one-third-power law of sublattice magnetization. We note in this connection, that in $CsMnF_3$ there are two antiferromagnetic modes,[15] one of which has a lower frequency than that for $RbMnF_3$ and the other which is much higher ($\sim 10^{11}$ Hz at 4.2 °K). Corresponding to these antiferromagnetic modes there are two nuclear modes. In the absence of an external magnetic field one of these modes has a very low frequency (~40 MHz at 4.2 °K), whereas the other is at a higher frequency (~670 MHz at 4.2 °K). The latter mode corresponds to the one for which the exponent 0.3 was determined.[6] This nuclear mode however, is coupled to the high-frequency antiferromagnetic mode and therefore the "frequency-pulling" effect is negligible.[14,15] The exponent (0.3) would thus reflect the temperature dependence of M, hence of H_{NN}, as expected in the absence of coupling.

The similarity in the magnetic character of MnTe and $CsMnF_3$ suggests that the temperature dependence of NAR found in MnTe can also be accounted for by a negligible effect of the coupling between the nuclear and antiferromagnetic modes.

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they only tentatively attributed to NAR. The close relation between their results and those obtained in the present study supports their conjecture).
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