Relaxation of nuclear spin waves in antiferromagnetic $MnCO_3$

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The dependence of the relaxation rate of nuclear spin waves on temperature and the wave vector is investigated by the parallel pumping technique in a broad frequency and temperature range. It is shown that the relaxation rate η may be expressed by the formula $\eta[Hz] = 0.015\ Tk + 0.66\ T^7$, where k is the wave vector. The cause of weakening of the Richards relaxation mechanism responsible for the term $\sim Tk$ is discussed. It is suggested that the second term $\sim T^7$ corresponds to scattering of spin waves by phonons.

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INTRODUCTION

The investigation of parametric excitation of spin waves in magnetically ordered systems, using parallel pumping techniques, is of definite interest. With this method it has proved possible to investigate spin waves over a large range of wave vectors by using standard equipment. The first suggestions of nuclear spin waves due to the Suhl-Nakamura interaction,[1,2] came from de Gennes *et al.*[3] Parametric excitation of spin wave was observed by Adams *et al.*[4] in $CsMnF_3$ and $RbMnF_3$ and by Yakubovskiĭ[5] in $MnCO_3$. These experimental data, taken at low frequencies, did not allow any conclusion to be drawn about the mechanism of the parametric excitation nor about the relaxation of the nuclear magnons.

The object of this investigation is $MnCO_3$, a well-investigated antiferromagnet with weak ferromagnetic properties. $MnCO_3$ has a rhombohedral structure; the plane perpendicular to the threefold symmetry axis is the easy plane for the magnetic system. The low-frequency modes of the electron antiferromagnetic and nuclear resonances are[6–8]

$$\omega_{ek}^2 = \gamma_e^2[H(H+H_D)+H_\Delta^2+\alpha^2k^2], \quad (1)$$

$$\omega_{nk}^2 = \omega_{n0}^2[1-\gamma_e^2 H_\Delta^2/\omega_{ek}^2], \quad (2)$$

where ω_{ek} and ω_{nk} are the frequencies of the electron and nuclear spin waves, γ_e is the gyromagnetic ratio for the electron, H is the external magnetic field, $H_D = 4.4$ kOe is the effective field of the Dzyaloshinskiĭ interaction, H_D^2 is the electron-spectrum gap due to the hyperfine interaction, α^2k^2 is the spatial-dispersion term, and $\omega_{n0}/2\pi = 640$ MHz is the frequency of the unbiased NMR. The restricted NMR frequency spectrum allows the possibility of investigating the entire system of the nuclear spin waves.

In a previous paper[3] we investigated the parametric excitation of nuclear spin waves in $MnCO_3$ at a frequency of 1080 MHz. We found two threshold processes corresponding to the excitation of nuclear spin waves and magnetoelastic oscillations. In this work we investigate in detail the parametric excitation of nuclear spin waves in antiferromagnetic $MnCO_3$ over a broad range of pump frequencies and sample temperatures. We obtain the dependence of the absolute values of the relaxation frequency η_{nk} of the nuclear spin waves on the wave vector for different frequencies and temperatures.

EXPERIMENT

The spectrum of the nuclear spin waves in $MnCO_3$ lies in the frequency region below 640 MHz. For parametric excitation an rf field has to be applied at a frequency equal to twice the spin-wave resonance frequency, with $\mathbf{h} \parallel \mathbf{H} \perp \mathbf{C}_3$, where h is the rf magnetic field, H is the external magnetic field, and C_3 is the threefold symmetry axis perpendicular to the easy plane of the magnetization of the sample. In this work we investigated the susceptibility of $MnCO_3$ parallel to the magnetic field at frequencies below 1280 MHz $= 2\omega_{n0}/2\pi$.

The $MnCO_3$ sample was a well-faceted single crystal plate whose principal surface coincided with the easy plane of magnetization. The sample was placed in a helical resonator made of aluminum wire (in aluminum, in contrast to copper, the magnetoresistance is small).

It was possible by using two input and output attenuators to change the power fed into the resonator, and as a result the amplitude of the rf field, while keeping the output power constant. At the same time the rf signal power, which is proportional to the imaginary part of the parallel susceptibility of the sample, was recorded for different levels of the rf magnetic field in the sample.

EXPERIMENTAL RESULTS

In Fig. 1 we plot the dependence of the transmitted rf power (at $f = 1170$ MHz, $T = 1.24$ K) on the external magnetic field for different levels of rf power in the resonator.

As the rf field approaches some threshold in the field $H = H_{res}$, corresponding to an NMR signal at half the pump frequency, an additional absorption peak appears, due to the excitation of NMR spin waves with wave vectors near $k = 0$. The absorption line has at the very outset a finite width of the order of the NMR line at half the pump frequency. Within a range of rf input powers the width of the line does not change, but the intensity does. When the rf power is further increased, the region of the excess absorption extends to smaller magnetic fields.

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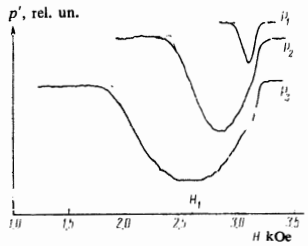

FIG. 1. Dependence of the power transmitted through the resonator on the magnetic field at different levels of rf power: $P_3 > P_2 > P_1$, $f = 1170$ MHz, $T = 1.24$ K. The break on the curve P_3 in the region 3.1–3.2 kOe corresponds to the second threshold.

From the curves displayed in Fig. 1 we can deduce the dependence of the amplitude of the critical field h_c on the magnitude of the magnetic field H. With the help of construction represented in Fig. 1 by the dotted line we find lower limits of the magnetic field which restrict the excess absorption. When the magnetic field is so defined, an error can be caused by the existence of an absorption tail that extends to low magnetic fields. Thus it is difficult to determine the instant of disappearance of the excess absorption. In consequence the determination of the magnetic field has a subjective character. Based on this, the authors have chosen the method of determining the magnetic field outlined above.

The rf magnetic field was estimated from the value of the power fed to the resonator, the coupling of the coaxial line with the resonator, and the Q-value and linear dimensions of the resonator. Since the calculation of the rf field distribution in a helical resonator is very complicated, we assumed for simplicity the rf field to be homogeneous and concentrated inside the helix. The coupling of the input and output coaxial lines with the resonator was maintained constant at all frequencies of interest, as monitored with a panoramic frequency-characteristic meter (FCM). The accuracy with which the absolute value of the rf magnetic field can be determined in such an estimate is very low ($\sim +100, -50\%$). However, the relative error on going from one frequency to another (another helix) is much smaller, because it depends only on Q-value and the coupling, which have been measured with the FCM. The relative error of the rf field determination in one experiment depends only on the calibration of the attenuators.

The dependence of the amplitude of the critical field h_c on the external magnetic field H is shown in Fig. 2

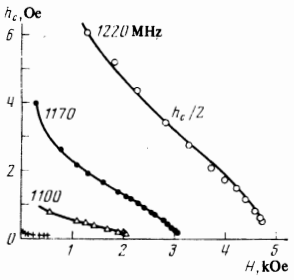

FIG. 2. Dependence of the critical field h_c on H for different frequencies; $T = 1.24$ K. For the frequency 1220 MHz the scale of h_c is doubled. The crosses show the data of Ozhogin and Yakubovskiĭ[10] at 875 MHz.

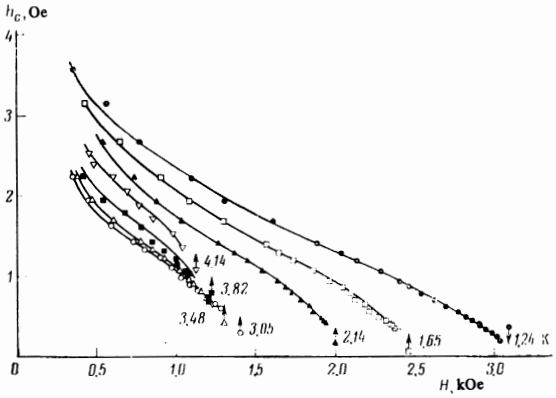

FIG. 3. Dependence of the critical field h_c on H for different temperatures, $f = 1170$ MHz.

for different frequencies and a fixed temperature ($T = 1.24$ K) and in Fig. 3 for a fixed pump frequency (1170 MHz) and different temperatures. As the pump frequency increases the rf field amplitude needed to excite the nuclear spin waves also increases (Fig. 2). As the pump frequency approaches twice the frequency of the undisplaced NMR a rapid increase of h_c is observed. In Fig. 3 we show the dependence of h_c on H found by Ozhogin and Yakubovskiĭ[10] for $MnCO_3$ at 875 MHz. With changing temperature, the resonant field H_{res} (marked by arrows in Fig. 3) and the critical field h_c change. As the temperature increases from the lowest value (1.24 K) to ~3 K the curves shift mainly into the region of weaker magnetic fields and h_c increase slightly as $H \to H_{res}$. When the temperature rises above 3 K, the h_c increases noticeably.

As noted in the Introduction, the frequency limitation of the NMR spectrum makes it possible in principle to investigate the whole system of nuclear spin waves in $MnCO_3$. However, as the pump frequency approaches a value double the unbiased NMR frequency ($2\omega_{n0}/2\pi = 1280$ MHz) the NMR gain tends to unity and therefore the parallel susceptibility at the pump frequency drops. It is therefore impossible in experiment to come arbitrarily close to the frequency 1280 MHz. Consequently the possibility of investigating the nuclear spin wave spectrum is limited to frequencies slightly smaller than $\omega_{n0}/2\pi$. The maximum frequency at which experimental results could be obtained was 1220 MHz. At 1237 MHz the useful signal did not exceed the noise in the system. A closer approach to $2\omega_{n0}/2\pi$ is possible only via substantial improvement of the noise characteristics of the experimental equipment. Figure 4 shows the dependence of the beyond-threshold susceptibility as a function of the rf power inside the resonator at a frequency 1170 MHz and at a field $H = 2.6$ kOe (marked by an arrow in Fig. 1). With increasing power above the critical value, the susceptibility grows sharply in an interval ~5 dB, and subsequently decreases before experiencing another increase (not shown in the figure). This second increase corresponds to the second threshold.

DISCUSSION OF THE RESULTS

Investigation of parametric excitation of nuclear spin waves in a broad range of frequencies, wave vectors,

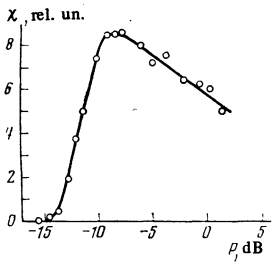

FIG. 4. Dependence of the beyond-threshold susceptibility on the power, $f = 1170$ MHz, $T = 1.24$ K, $H = 2.6$ kOe.

and temperature allows us to draw conclusions concerning the spin-wave excitation mechanism and relaxation rate of these waves as functions of various external parameters. Adams *et al.*[4] and Yakubovskiĭ[5] performed their experiments at low frequencies corresponding to wave vectors $k \approx 10^4$ cm^{-1}. In the present work we have reached $k \approx 10^6$ cm^{-1} at the maximum frequency 1220 MHz. This should allow us to draw more reliable conclusions about the relaxation behavior of the spin waves.

From the experimental dependence of $h_c(H)$ we can extract information concerning the dependence of the relaxation frequency of the nuclear spin waves η_{nk} on the wave vector. Using Ozhogin's[11] formula

$$h^{nn}(H) = \frac{4\pi\eta_{nk}\omega_{ek}^2}{\gamma_e^2(H_D+2H)\omega_{nk}}\left(\frac{\omega_{ek}^2}{\gamma_e^2 H_\Delta^2}-1\right) \tag{3}$$

and Eq. (2) for the NMR frequency, we find

$$h_c^{nn}(H) = \frac{4\pi\eta_{nk}H_\Delta^2\omega_{n0}^2\omega_{nk}}{(\omega_{n0}^2-\omega_{nk}^2)^2(H_D+2H)} \tag{4}$$

or

$$h_c(H_D+2H) = \eta_{nk}f(\omega_p)/T, \tag{5}$$

where

$$f(\omega_p) = \frac{\omega_{n0}^2\omega_{nk}}{(\omega_{n0}^2-\omega_{nk}^2)^2}\cdot 4\pi H_\Delta^2 T \tag{6}$$

is a function of the pump frequency only, because $\omega_{nk} = \omega_p/2$; ω_p is the pump frequency and $H_\Delta^2 T = 5.8$ kOe$^2\cdot$K.

From (1) we can find the relationship between the wave vector of the excited nuclear spin waves and magnetic field that restricts the excess-absorption curve

$$k^2\alpha^2 = \gamma_e^2[H_{res}(H_{res}+H_D) - H(H+H_D)], \tag{7}$$

where H_{res} is the resonant NMR field at half the pump frequency and α is the velocity of the antiferromagnetic

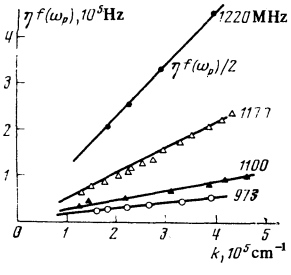

FIG. 5. Dependence of the function $\eta(k)f(\omega_p)$ on k; $T = 1.24$ K. The first four points only are shown for the frequency 1220 MHz, but the straight line goes through all the points and reaches values of $k = 10^6$ cm^{-1}.

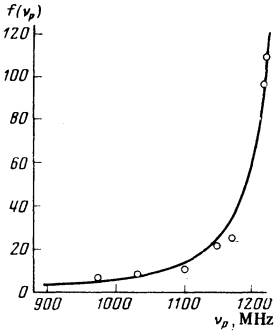

FIG. 6. The function $f(\nu_p) \sim \omega_p/[\omega_{n0}^2 - (\omega_p/2)^2]^2$. The points are the slopes of the straight lines from Fig. 5.

spin waves ($\alpha = 1.07\times10^5$ cm/sec).

In this way the $h_c(H)$ plots (Figs. 2 and 3) can be recalculated into the functions $\eta(k)$. In Fig. 5 plots of $\eta f(\omega_p) \sim h_c(H_D+2H)$ against the wave vector k are shown for various frequencies at $T = 1.24$ K. It can be seen that experimental points fall on straight lines, thus attesting to a linear dependence η on k. In Fig. 6 the slope of the straight lines of Fig. 5 are shown; the function $f(\omega_p)$ is represented by the continuous line. It follows that the different slopes of the straight lines of Fig. 5 correspond to changes of the coefficient $f(\omega)$ and not to variation of η with frequency.

Figure 7 shows plots of $\eta(k)/T$ for different temperatures at 1170 MHz. Within the limits of experimental error, the points fall on a series of parallel straight lines displaced vertically from one another. From this fact it follows that η has a linear dependence on Tk. To account for the vertical displacement without change of slope we must introduce in $\eta(k, T)$ a term that depends on temperature only and not on k. Extrapolating the experimental lines in Fig. 7 to $k = 0$ we find the dependence of η_{n0}/T on temperature, as shown in Fig. 8.

Thus, the experimental results demonstrate that the relaxation rate of the nuclear magnons is given by the formula

$$\eta_{nk} = 0.015\,Tk + 0.66\,T^2 \text{ [Hz]}. \tag{8}$$

The accuracy of the determination of the constants (+100%, −50%) is connected with the estimate of the critical field h_c. Though the absolute accuracy is small, we can improve it substantially by comparing our results with spin-echo measurements of the nuclear spin-spin relaxation in $MnCO_3$.[12]

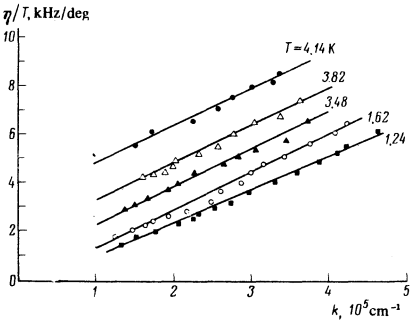

FIG. 7. Dependence of the product of the relaxation rate and the reciprocal temperature on the wave vector k; $f = 1170$ MHz.

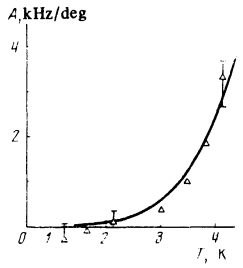

FIG. 8. Dependence of the function $A=\eta(0)/T$ on temperature. The solid curve is proportional to T^5.

In the nuclear-magnon relaxation theory proposed by Richards[13] the relaxation rate for $MnCO_3$ is given by the expression $\eta=0.4\,Tk$.[5] The similar dependence on temperature and wave vector found experimentally suggests that the Richards[13] relaxation mechanism is realized in the $MnCO_3$ nuclear system but is greatly weakened. The reason for this weakening is apparently spin-lattice relaxation. The relaxation rate can be estimated from the integral measurements of the NMR saturation[8] at $\eta\approx 200$ Hz for $MnCO_3$ ($\tau\approx 1$ msec). Parametrically excited spin waves, characterized by frequency and wave vector, are transformed by dipole interactions[12] into isoenergetic magnons. Since the relaxation to the lattice is slow, the magnons are long-lived and in turn can be transformed into the initial spin-wave state. Thus, spin-lattice relaxation is the bottleneck in the attenuation of nuclear magnons and can essentially affect the values of the coefficients in (8).

The second term in (8) constitutes a mechanism that does not depend on the wave vector of the nuclear magnons and depends strongly on temperature. The accuracy of the determination of the exponent is small in this case. Within the limits of the experimental error, the dependence can range from T^6 to T^8. From the form of the temperature dependence we can assume that this term in the relaxation corresponds to scattering of spin waves by phonons.[14] The experimental results show that the linear dependence of the nuclear spin wave relaxation rate on the wave vector holds for $k>10^5$ cm^{-1}. Assuming that the Richards relaxation mechanism takes place, we can conclude that the inhomogeneous broadening of the spin-wave wave-vector spectrum of our $MnCO_3$ sample is equal in value to the broadening $\Delta k(k)$ due to the dipole interaction in the Richards model, at $k_0\approx 10^5$ cm^{-1}. The nuclear spin wave relaxation rate determined by the true value of Δk does not depend on k below k_0. This corresponds in Figs. 5 and 7 to the horizontal parts of the plots of η versus k in the region from the minimum value of η to $k=0$. To simplify the figures we do not show these sections in Figs. 5 and 7 (see[9]).

It is thus clear that in investigations at low frequencies, corresponding to excitation of spin waves with $k<10^5$ cm^{-1}, we cannot expect a linear dependence of the relaxation rate on the wave vector. Preliminary results show that k_0 is greater for $RbMnF_3$ than for $MnCO_3$, while for $CsMnF_3$ we can expect smaller values of k_0.

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