Relaxation of electron spin waves in antiferromagnetic $CsMnF_3$

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The relaxation rate $\Delta\nu$ of electron spin waves (SW) in the antiferromagnet $CsMnF_3$ is investigated. Two principal terms, $\Delta\nu_1$ and $\Delta\nu_2$, which determine the SW relaxation, are set apart in the entire range of the experimental parameters (SW frequency $\nu_k = 11$–22 GHz, temperature $T = 1.2$–2.8 K, magnetic field $H = 1$–5 kOe). Comparison of the results with the theory has shown that the first of these terms, $\Delta\nu_1$, is well described both in magnitude and in all functional dependences by the theory of SW scattering from the magnetization fluctuations of the ^{55}Mn nuclei. The term $\Delta\nu_2$, which has a strong temperature dependence and is proportional to H^2, was observed earlier in several antiferromagnets and was usually ascribed to the contribution of three-magnon relaxation. Our measurements of the frequency dependence of $\Delta\nu_2$ have shown, however, that the three-magnon-relaxation theory does not describe the $\Delta\nu_2$ (ν_k) functional dependence and, in addition, calculation yields for the relaxation a value smaller than experiment by about an order of magnitude.

One of the most important properties of spin waves (magnons) is the lifetime, which is determined by the effectiveness with which they interact with one another, with phonons, and with sample defects. Spin-wave (SW) relaxation in weakly anisotropic antiferromagnets has been the subject of many experimental studies (see Refs. 1–8), whose results yield the following picture of the behavior of SW relaxation: At low temperatures, the SW relaxation is directly proportional to the magnon wave vector **k** and is practically independent of temperature.[7,8] This contribution (which we designate $\Delta\nu_1$) is observed in all the antiferromagnets investigated in Refs. 1–8, viz., $CsMnF_3$,[3–5] $MnCO_3$,[4–6] $CsMnCl_3$,[7,8] and $RbMnCl_3$.[8] As the temperature is raised, a contribution $\Delta\nu_2$ rises above the background of $\Delta\nu_1$, has a strong temperature dependence (T^{6-8}), and is proportional to the square of the magnetic field. This contribution becomes predominant at $T \gtrsim 1.5$ K ($MnCO_3$ and $CsMnF_3$) or $T \gtrsim 4$ K ($CsMnCl_3$).

The values and functional dependences of $\Delta\nu$ (k, T) are well described by the theory of elastic scattering of SW by fluctuations of the nuclear-subsystem longitudinal magnetization.[9] Contributions can be made to $\Delta\nu_1$ also by elastic scattering of SW from sample defects whose relaxation parameters have the same dependences on T and k. The functional dependences of the parameter $\Delta\nu_2$ (H, T) are satisfactorily described by the equation for the three-magnon relaxation process,[10,11] i.e., a process in which the investigated magnon coalesces with a thermal magnon of the same mode into a magnon of a higher mode of the SW spectrum. Besides $\Delta\nu_1$ and $\Delta\nu_2$, various contributions were found to be made to the relaxation by SW interactions with phonons [1,4–8] and impurities,[4,6] but these contributions were as a rule smaller than $\Delta\nu_1$ and $\Delta\nu_2$.

Thus, the experimental results known when our investigation was started were satisfactorily described by the theory and it was assumed that the nature of SW relaxation is in the main well understood. It must be noted, however, that in each detailed investigation[1–8] of the SW relaxation rates the magnon frequency was fixed, and the theoretically predicted strong dependences of the relaxation on the frequency, $\Delta\nu$ (ν_k), were never verified in experiment. The task of the present work was therefore the following: study of the frequency dependence of the SW relaxation, measurements in the widest possible range of temperatures, and comparison of the results with the theories of three-magnon and elastic relaxation of electronic SW in the entire investigated range of frequencies, temperatures, and magnetic fields.

We chose for the investigation a $CsMnF_3$ crystal, in which the magnon-phonon interaction is noticeably manifested only at the points where the SW and sound spectra intersect, and no impurity peaks are observed, so that the experimental results can be reduced with account taken of two contributions to the relaxation, $\Delta\nu_1$ and $\Delta\nu_2$. At helium temperatures these contributions in $CsMnF_2$ are approximately of the same order, so that each can be compared sufficiently accurately with the theory. Moreover, at the end points of the temperature range we have $\Delta\nu_1 \gg \Delta\nu_2$ at $T = 1.2$ K and $\Delta\nu_2 \gg \Delta\nu_1$ at $T > 2.5$ K, a situation likewise favorable for the data reduction.

EXPERIMENT AND REDUCTION OF RESULTS

Spin waves were excited in the investigated sample by the method of parallel microwave pumping. The SW relaxation rate was determined from the threshold parametric-excitation field. A single crystal of $CsMnF_3$ on a teflon stand was placed in the antinode the H_{012} mode of a microwave field on the axis of a cylindrical cavity resonator with loaded $Q \approx 10^4$. The microwave sources were klystron oscillators generating millisecond pulses at frequencies from 22 to 44 GHz. A microwave pulse passing through the cavity was detected and fed to an oscilloscope. The excited parametric magnons were revealed by appearance, on the pulse, of chips corresponding to the onset and cessation of microwave power by the sample. The magnetic field h at the sample, in relative units, was determined with a calibrated attenuator, while its absolute value was calculated from the measured values of the microwave power, of Q, and of the coupling with the cavity. The relaxation rate of parametric electronic SW was calculated using the equation[12]

$$\Delta\nu = \frac{2\gamma^2 H h_c}{\nu_p} \frac{\nu_{20}^2}{\nu_{20}^2 - \nu_p^2}, \tag{1}$$

where h_c is the threshold field, $\gamma = 2.8$ GHz/kOe is the magnetomechanical ratio, ν_p is the pump frequency, and $\nu_{20} = 115$ GHz (Ref. 13).

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The relative accuracy of the measured $\Delta\nu$ was $\sim 10\%$ at all frequencies. The accuracy of the absolute measurement of the microwave field h in the empty cavity is 15% (Ref. 4). The sample introduces some distortion in the microwave-field distribution,[2] as manifested by a change of the field amplitude h. With allowance for distortion, the error of h does not exceed 30%. The experiments were made in the temperature range $T = 1.2$–2.8 K.

The parametric excitation of the SW was "hard" at all pump frequencies: the SW excitation threshold h_{c1} exceeded the extinction threshold h_{c2}. The experimental results were reduced with account taken of two relaxation mechanisms—elastic and three-magnon. Since the turned-off part of the relaxation is not related to either mechanism, we used the field value h_{c2} to calculate the damping of the electronic SW.

The experimental results were reduced using the equation

$$\Delta\nu^{э}=A_1\Delta\nu_{mn}+A_2\Delta\nu_{3m}. \qquad (2)$$

Here A_1 and A_2 are fit parameters, $\Delta\nu_{mn}$ is the parameter of the elastic scattering of magnons by the nuclear-subsystem magnetization fluctuations, and $\Delta\nu_{3m}$ is the rate of three-magnon relaxation. We calculated $\Delta\nu_{mn}$ from the theoretical equation[1)] obtained in Ref. 14:

$$\eta_{mn}=\frac{I(I+1)}{12\pi}\omega_n\left(\frac{J_0}{\Theta_N}\right)\left(\frac{\omega_n}{\Theta_N}\right)\left(\frac{sk}{\omega_{\mathbf{k}}}\right),\quad \Delta\nu_{mn}=\frac{\eta_{mn}}{\pi}. \qquad (3)$$

Here I is the nuclear spin, ω_n the frequency of the unshifted NMR, $J_0 = H_E g\mu_B/S$ the exchange integral, $\Theta_N = s/v_0, v_0$ the volume of the unit cell, $\omega_{\mathbf{k}}$ the magnon frequency $s = 2\pi\alpha\gamma$ the spin-wave velocity, α the exchange constant, μ_B the Bohr magneton, g the Landé factor, and S the electron-shell spin. The value of $\Delta\nu_{3m}$ was calculated using an equation obtained from the amplitude calculated in Ref. 11 for three-magnon interaction[11]:

$$\Delta\nu_{3m}=\frac{1}{4\pi^2}\frac{g\mu_B}{\hbar}\frac{H^2}{\alpha k}\left(\frac{T}{\Theta_N}\right)^3\frac{J_0}{\hbar\omega_{\mathbf{k}}}\operatorname{sh}x_k I_{3m}. \qquad (4)$$

Here

$$I_{3m}=\int_{x_-}^{x_+}dx\,(x+x_k)^2/[\operatorname{sh}x\operatorname{sh}(x+x_k)],\quad x_k=\hbar\omega_{\mathbf{k}}/2k_BT,$$

k_B the Boltzmann constant,

$$x_\pm=\frac{\hbar(\omega_{10}{}^2+\omega_\pm{}^2)^{1/2}}{2k_BT},\quad \omega_\pm=\frac{1}{2}\left(\frac{\omega_{20}}{\omega_{10}}\right)^2\left(\omega_{\mathbf{k}}\pm\frac{g\mu_B}{\hbar}\alpha k\right),$$

and ω_{10} and ω_{20} the antiferromagnetic-resonance frequencies of the two SW modes. The contributions of the two processes to the spin-wave relaxation was assumed to be additive (see the discussion in Ref. 8).

RESULTS AND DISCUSSION

Figures 1 and 2 show the experimental data on the relaxation $\Delta\nu$ of electron spin waves, at a frequency $\nu_p = 30.4$ GHz, vs the squared magnetic field for several temperatures, and vs temperature at a fixed field. The theoretical curves were obtained by least squares for the entire aggregate of the points at the given frequency. It is seen from Figs. 1 and 2 that Eqs. (2)–(4) make it possible to describe, using the fit parameters $A_1 = 1.2 \pm 0.4$ and $A_2 = 7.9 \pm 2.4$, the functional dependence of the relaxation of the electronic SW in

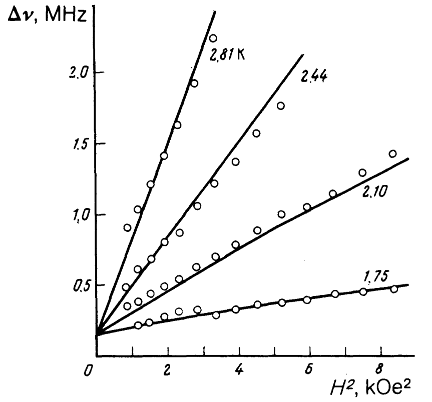

FIG. 1. Magnon relaxation rate $\Delta\nu$ in $CsMnF_3$ at $\nu_p = 30.4$ GHz vs the static magnetic field at various temperatures. Solid curves—least-squares reduction of the experimental results, using Eq. (2) with fit parameters $A_1 = 1.2$ and $A_2 = 7.9$.

$CsMnF_3$ at a given pump frequency. A similar picture was observed also for other pump frequencies; the coefficient A_1, apart from experimental errors, did not vary in the investigated frequency band. It can thus be stated that the experimental data on the relaxation $\Delta\nu_1$ are satisfactorily described by the theory of elastic scattering of SW by fluctuations of the nuclear magnetic subsystem.

The parameter A_2, which fitted the value of $\Delta\nu_2$ to the theory of three-magnon relaxation of electronic SW, ranged in the investigated frequency band from max $A_2 = 19 \pm 6$ at $\nu_p = 22.5$ GHz to min $A_2 = 5 \pm 1.5$ at $\nu_p = 41.4$ GHz. In other words, the theoretical frequency dependence of $\Delta\nu_{3m}(\nu_p)$ does not agree with the experimental $\Delta\nu_2(\nu_p)$ dependence, and the absolute value of $\Delta\nu_2$ is larger than the theoretical prediction for by approximately an order of magnitude. It must be recognized, however, that the experiments were performed on the hexagonal antiferromagnet $CsMnF_3$, whereas the theoretical equations were obtained in the isotropic cubic model and should be correct with accuracy to the coefficients.

As already noted, the absolute value of the SW relaxation is not determined with high accuracy, so that it is more advantageous to compare with the theory the frequency dependence not of each individual contribution $\Delta\nu_1$ or $\Delta\nu_2$, but their ratio $\xi = \Delta\nu_2/\Delta\nu_1$. To determine the frequency dependence we used the values $\Delta\nu_1(H_1 \to 0)$ and $\Delta\nu_2(H_2^2 = 6$ kOe$^2)$ obtained by reducing the experimental results at a

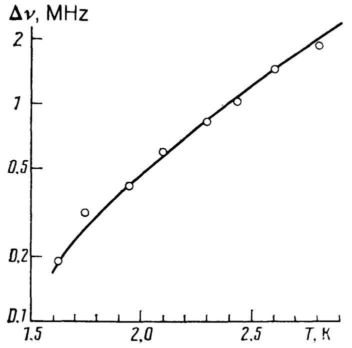

FIG. 2. Temperature dependence of the relaxation parameter $\Delta\nu$ in $CsMnF_3$ at $\nu_p = 30.4$ GHz and $H^2 = 2.8$ kOe^2. Solid curve—calculated from Eq. (2) with fit parameters $A_1 = 1.2$ and $A_2 = 7.9$.

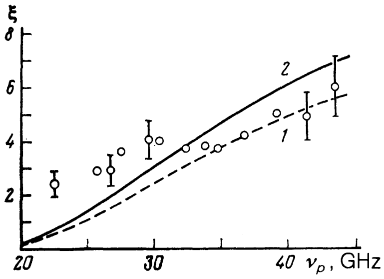

FIG. 3. Dependence of the parameter ξ on the pump frequency ν_p at $T = 2.0$ K. Theoretical curves: 1—calculated using Eqs. (3) and (4) with fit parameter $A_2/A_1 = 4.5$; 2—obtained by least squares using all the experimental points and $A_2/A_1 = 5.5$.

fixed frequency using Eq. (2). The field $H_1 = 0$ was chosen because $\Delta\nu_2 \to 0$ as $H \to 0$. We assumed in the calculation that $\Delta\nu_1 = A_1\Delta\nu_{mn}$ and $\Delta\nu_2 = A_2\Delta\nu_{3m}$. The corresponding $\xi(\nu_p)$ frequency dependences are shown in Fig. 3 from which it can be seen that even curve 2, obtained by least squares with the fit parameter $A_2/A_1 = 5.5$, can describe only the general tendency of ξ to increase with increase of frequency.

Thus, the two relaxation processes—elastic and three-magnon—do not suffice to describe all the experimental results. Since the contribution $\Delta\nu_1$ is described, to within experimental errors, by elastic-relaxation processes, the deviation from the theory is due to the fact that the contribution $\Delta\nu_2$, traditionally attributed to the three-magnon process, is not described by the theory of this interaction.

In our opinion, the disparities between the values and frequency dependences of $\Delta\nu_2$ and $\Delta\nu_{3m}$ require a theoretical search for a new relaxation channel, one yielding $\Delta\nu(H,T)$ dependences simlar to those of $\Delta\nu_{3m}$, but a different frequency dependence. This process in conjunction with $\Delta\nu_{3m}$ (or taken alone) should fit the experimental results for $\Delta\nu_2$. For example, curve 1 of Fig. 3, with a fit parameter $A_2/A_1 = 4.5$, agrees well with the experimental results at $\nu_p > 34$ GHz, but at lower frequencies the experimental points lie noticeably above the curve 1; this may attest to the onset of a new SW-relaxation mechanism. To be sure, the low accuracy with which $\Delta\nu$ is measured prevents us from stating categorically that an inflection actually exists on the $\xi(\nu_p)$ plot vs frequency in the interval $\nu_p = 30$–35 GHz.

It should be noted that calculations for slow SW relaxation on paramagnetic impurities[15] yielded $\Delta\nu(H,T)$ dependences similar to the experimental ones. An accurate comparison of the experimental results with this theory, however, calls for information on the impurity content, on the impurity levels, and on their relaxation times. In our study, the impurity content of the samples was not specially investigated. We can nonetheless expect the impurity content to fluctuate from sample to sample, with variation of the SW relaxation rate as the result. Indeed, in experiments performed on various samples at $\nu_p = 36$ GHz (Refs. 3 and 5, and our present study, the ratios $\xi = \Delta\nu_2/\Delta\nu_1$ obtained differed by about a factor of two, and it is natural to assume that $\Delta\nu_1$ and/or $\Delta\nu_2$ contains a relaxation contribution from crystal defects. In an attempt to answer this question, we investigated to $CsMnF_3$ samples in which the "hardness" and the value of the magnon-phonon peak in the SW relaxation differed strongly, but the values of ξ, $\Delta\nu_1$ and $\Delta\nu_2$ agreed within the limits of experimental error. It is apparently impossible, unfortunately, to determine definitely from the various data which of the relaxation components, $\Delta\nu_1$ or $\Delta\nu_2$, changes from sample to sample, in view of the low accuracy of the absolute measurement of $\Delta\nu$.

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[1)]To obtain the correct dimensionality, all the quantities in the parentheses in Eqs. (3) and (4) must be converted into units that make the parentheses nondimensional.

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