Effect of the structure of a crystal and its defects on the relaxation of nuclear spin waves in antiferromagnets

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A parallel microwave pump was used in the wide frequency band $\nu_p = 500$–1200 MHz to investigate the relaxation of nuclear spin waves in antiferromagnets with different numbers of magnetic sublattices, namely, $MnCO_3$ (two sublattices), $CsMnF_3$ (six sublattices), and also in $CsMnF_3$ with deliberately disturbed stoichiometry. A qualitative difference between the relaxation of nuclear magnons due to the different structure of $MnCO_3$ and $CsMnF_3$ was found for the first time. It is shown that, for temperatures $T \gtrsim 2$K, the principal mechanism responsible for the relaxation of nuclear spin waves in $MnCO_3$ is a process involving the participation of two phonons, whereas in $CsMnF_3$ there is an additional appreciable contribution (especially at high frequencies) due to a process involving two electron magnons, which is not present in the two-sublattice antiferromagnet. $CsMnF_3$ with disturbed stoichiometry was found to exhibit a new nuclear magnon relaxation process with linear dependence on temperature and wave vector. A model is proposed in which stoichiometry defects are treated as a set of two-level systems. The model is capable of describing all the observed experimental relationships for this process.

1. INTRODUCTION

Studies of the relaxation properties of spin waves in ferromagnetic crystals yield important information on the different interactions of these waves with each other as well as with elastic lattice vibrations and crystal defects.

The traditional method of investigating spin-wave damping relies on measurements of the amplitude h_c of the threshold microwave field for their parametric excitation, an amplitude known to depend on the relaxation rate. This method is particularly convenient because, as a rule, parametric resonance involves the excitation of spin-wave packets having a narrow spread in k and well-known values of the frequency ω_k and wave vector k, so that the relaxation rate can readily be investigated as a function of these parameters and of temperature T.

The most convenient materials for such studies are antiferromagnets with "easy plane" magnetic anisotropy ($MnCO_3$, $CsMnF_3$, $CsMnCl_3$, and $FeBO_3$) for which there is now a considerable volume of established experimental information[1–6] on the relaxation of electron and nuclear spin waves (ESW and NSW). Moreover, there is a substantial number of theoretical publications on the damping of ESW and NSW in such materials.[7–10] Comparison of experimental data on ESW relaxation with theoretical results is complicated by the fact that, as a rule, the energy of the excited ESW is $\hbar\omega_{fk} \approx k_B T$, whereas theoretical expressions for the rate of relaxation have been given in a simple form only in the limiting cases of high and low temperatures. On the other hand, the NSW energy has been well below the thermal energy in all experiments: $\hbar\omega_k / k_B \lesssim 0.03$ K. The theoretical formulas are sufficiently simple in this limit. It is precisely this consideration, as well as experimental work in a wide range of frequencies, that has been responsible for considerable advances in our understanding of the nature of NSW relaxation in recent years.[11–13]

Since calculations of the spin-wave relaxation rate γ from experimental data rely on the use of a particular formula, we must examine its validity. Considerations of energy balance lead to the following expression for the relaxation rate γ of parametric spin waves:

$$\gamma = h_c |V_{\mathbf{k}}| / 2\hbar = \gamma_{el} + \gamma_{nel}, \tag{1}$$

where V_k is the coupling coefficient between the spin wave and the pump (in fact, $V_{\mathbf{k}} = -\partial\hbar\omega_{\mathbf{k}}/\partial H$ is the effective magneton of the excited spin waves) and γ_{el} and γ_{nel} are the rates of relaxation of elastic (only $\mathbf{k}$ changes) and inelastic processes, respectively. All processes contributing to the resultant relaxation of the spin wave under consideration are assumed to be *additive* in this calculation.

A theoretical modification of (1) has also been proposed,[14,15] based on the assumption that elastic relaxation processes play a special role. When $\gamma_{nel} \gg \gamma_{el}$, this result is identical with (1), but in the opposite limiting case the formula is different:

$$\gamma = h_c |V_{\mathbf{k}}| / 2\hbar = (\gamma_{el}\gamma_{nel})^{1/2}. \tag{2}$$

Analysis of experimental data obtained at low enough temperatures at which the relaxation of electron and nuclear spin waves is dominated by elastic processes (scattering of these waves by paramagnetic nuclear spins and by the specimen boundary)[5,13] supports the validity of (1). In particular, it has been shown experimentally that, at low enough temperatures ($T \lesssim 2$ K), the main contribution to NSW relaxation in all the easy-plane antiferromagnets that have been investigated ($MnCO_3$, $CsMnF_3$, and $CsMnCl_3$) has the form[4,12,13] $\gamma \propto Tk$. Theory shows[9,13] that this contribution to the relaxation process is due to elastic scattering of parametric NSW by fluctuations of the longitudinal component of nuclear magnetization. Analysis of experimental data, based on (1), yields good agreement with the theory[13] not for the dependence on the main parameters (T, k, $\omega_{\mathbf{k}}$), but also for

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the magnitude of the relaxation rate in all the materials investigated. The use of (2), on the other hand, does not result in these functional relationships. Additional evidence in favor of (1) is provided by a recent experiment by Tulin and Govorkov, who performed direct measurements of NSW relaxation in $MnCO_3$, using the spin-echo method with parametric NSW.[16]

Thus, current experimental evidence supports the additivity of all the processes contributing to the resultant spin-wave relaxation and the validity of (1). The technique of parametric excitation of NSW by a parallel microwave pump and the modulation method of recording the threshold h_c are described in detail in our previous paper.[13]

Our task in the present research was to solve the following problems. Firstly, the theory of NSW relaxation proposed in Ref. 13, which relied on the multi-sublattice structure of $CsMnF_3$, required direct confirmation. It had to be demonstrated that this process does not occur in an antiferromagnet with a simple two-sublattice structure. Secondly, there was no experimental confirmation of the frequency dependence proposed in Ref. 10 for the NSW relaxation process with the participation of two phonons. Finally, after detailed studies of intrinsic NSW relaxation processes, it became possible to investigate the effect of defects on the relaxation properties of nuclear magnons.

2. THREE-FERROMAGNON INTERACTIONS IN MULTI-SUBLATTICE ANTIFERROMAGNETS

A detailed study of NSW relaxation in $CsMnF_3$ in a wide range of frequencies and temperatures was reported in our previous paper.[13] We found that the dominating process at relatively low temperatures ($T<2$ K) is elastic scattering of nuclear spin waves by thermal fluctuations of the nuclear-spin magnetization with a relaxation rate[1)] $\gamma_1 \propto Tk$ (see the next section). As the temperature increases, the leading process is that with a strong temperature dependence of the relaxation rate, namely, $\gamma_2 \propto T^5$. Studies of the frequency dependence of this process have shown that it is appreciably different from the theoretical dependence predicted for the coalescence of a nuclear magnon (n) and a phonon (ph) into a single phonon ($n+ph\to ph$) (Ref. 10):

$$\frac{\gamma_{n2ph}}{2\pi}=\frac{\pi^2}{20I(I+1)}(1-\xi^2)^2\left(\frac{k_B\tilde{\Theta}}{Mv_s^2}\right)^2\left(\frac{k_B}{\hbar}\right)^2\frac{T^5}{\Theta_D^3 v_s k}. \qquad (3a)$$

Here $\xi\equiv\omega_k/\omega_n$ is the reduced NSW frequency, $k_B\tilde{\Theta}$ is the energy of second-order magnetoelastic interaction per magnetic ion, $M=\rho V_0$, $V_0=V/\mathcal{N}$, V is the volume of the crystal, $\mathcal{N}$ is the number of magnetic ions in the crystal, V_s is the velocity of sound, $\Theta_D\equiv\hbar w_s/V_0^{1/3}k_B$, and I is the nuclear spin. It was suggested in Ref. 13 that there is an additional relaxation process involving the coalescence of a nuclear magnon (n) and an electron magnon (m) in the ferromagnetic (f) branch of the spectrum into a different f-magnon ($n+m\to m$). The amplitude for this process is not zero because of the multi-sublattice structure of $CsMnF_3$ (six magnetic sublattices). The following expression was obtained for the relaxation rate in this case:

$$\frac{\gamma_{n2m}}{2\pi}=c_{2m}(1-\xi^2)^2\frac{g\mu_B}{\hbar}\frac{H^2}{\alpha k}\left(\frac{T}{\Theta_N}\right)^5, \qquad (3b)$$

where α is the inhomogeneous-exchange constant, $\Theta_N\equiv g\mu_B\alpha/V_0^{1/3}k_B$ and $c_{2m}\sim 10^{-3}$. The combination of these two NSW relaxation mechanisms has enabled us to achieve a reasonably satisfactory description of the relaxation rate γ_2 of $CsMnF_3$ as a function of temperature, NSW frequency, and magnitude of the wave vector.

However, the question naturally arises as to the frequency dependence of the relaxation rate γ_2 that is proportional to T^5 in the two-sublattice antiferromagnet for which theory suggests that the $n+m\to m$ process is not present. We have investigated the two-sublattice $MnCO_3$ in which the process with $\gamma_2\propto T^5$ was first observed in Ref. 3. Figure 1 shows measurements of the relaxation rate γ_2 at $T=4.2$ K in a wide frequency range. The coordinates in the figure are chosen to obtain a straight line passing through the origin for the function $\gamma_{n2ph}(\omega_k)$. It is clear that the experimental data for $MnCO_3$ are satisfactorily described only within the framework of the relaxation process $n+ph\to ph$. At the same time, the frequency dependence of γ_2 in $CsMnF_3$ is essentially nonlinear (see Fig. 1) and can be satisfactorily described only by the sum $\gamma_{n2ph}+\gamma_{n2m}$ of relaxation rate.

Thus, our studies have enabled us not only to observe a qualitative difference between the NSW relaxation in two- and multi-sublattice antiferromagnets $MnCO_3$ and $CsMnF_3$, but also to explain the behavior of NSW relaxation in these two structures.

Using our results together with (3a), we can estimate the constant $\tilde{\Theta}$ for $MnCO_3$. For $v_s=3.4\cdot 10^5$ cm/s, $\Theta_D=66.8$ K, and $Mv_s^2/k_B=1.61\cdot 10^5$ K, we have $\tilde{\Theta}=1.6\cdot 10^4$ K. The corresponding constant for $CsMnF_3$ is[13] $\tilde{\Theta}=1.1\cdot 10^3$ K.

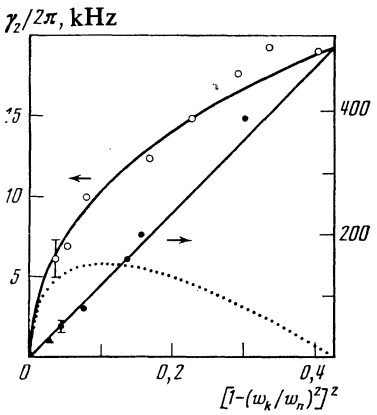

FIG. 1. Frequency dependence of the relaxation rate γ_2 in the two-sublattice antiferromagnet $MnCO_3$ (full points, right-hand scale) and the six-sublattice antiferromagnet $CsMnF_3$ (open circles, left-hand scale) for nuclear spin waves with $k=10^5$ cm^{-1} at $T=4.2$ K. The straight line passing through the origin corresponds to the theoretical frequency dependence (3a) for the process with participation of two phonons, whilst the dashed curve corresponds to the process with participation of two electron magnons (3b). The solid curve drawn through the open circles is the sum of (3a) and (3b). The triangle represents the data from Ref. 4 and demonstrates the spread of the experimental data obtained for different $MnCO_3$ specimens.

In conclusion, we must say something about the dependence of γ_2 on the wave vector in $MnCO_3$. The data reported by Govorkov and Tulin[4] and the results of our own experiments in the frequency range $\omega_k/2\pi > 500$ MHz show that γ_2 is independent of k. At lower frequencies, we have succeeded in observing an increase in γ_2 with decreasing k, but this increase is much weaker than $\gamma_{n2ph} \propto 1/k$. We suggest that this discrepancy is due to the inadequate quality of the $MnCO_3$ crystals. Quasiparticle interactions without quasimomentum conservation are possible because of the presence of lattice defects, so that the $1/k$ dependence may be substantially weakened at $k \lesssim l_d^{-1}$ (l_d is the characteristic separation between defects).

3. NATURE OF THE NSW RELAXATION PROCESS WITH $\gamma_1 \propto Yk$

Before we proceed to investigate interactions between the nuclear spin waves and crystal defects, we must verify that the previously observed relaxation processes are intrinsic, i.e., properties of a perfect crystal. This applies mainly to the process with $\gamma_1 \infty Tk$, since the rate of NSW relaxation by defects may have the same dependence ($\gamma_d \propto Tk$) as for the intrinsic process, i.e., for elastic scattering of nuclear spin waves by fluctuations of the longitudinal component of the nuclear magnetization. This process was first examined by Richards[9] in the limit $\omega_n - \omega_k \ll \omega_n$:

$$\gamma_R = \frac{\omega_n}{8\pi}(V_0^{1/3}k)\frac{TJ_0}{k_B\Theta_N^2}, \tag{4}$$

where J_0 is the Fourier component of the exchange integral. Estimates of the relaxation rate based on (4) are in good agreement with experimental results for the three easy-plane antiferromagnets $MnCO_3$ (Ref. 4), $CsMnF_3$ (Ref. 13), and $CsMnCl_3$ (Ref. 12).

The relaxation rate has been calculated theoretically[10] in second-order perturbation theory for arbitrary ω_k, and the result was a strong frequency dependence:

$$\tilde{\gamma}_R = \xi^3\gamma_R, \quad \xi \equiv \omega_k/\omega_n. \tag{5}$$

However, measurements performed on the three easy-plane antiferromagnets in a sufficiently wide frequency range ($\xi = 0.76$-0.95 in $MnCO_3$—Ref. 4, $\xi = 0.71$-0.93 in $CsMnCl_3$—Ref. 12, and $\xi = 0.45$-0.9 in $CsMnF_3$—Ref. 13) have shown that, to within experimental uncertainty ($\pm 25\%$), the relaxation rate γ_1 was independent of the NSW frequency.

There are two possible ways of removing this contradiction between theory[10] and experiment. Firstly, we could assume that all the specimens that we have investigated contained defects, and elastic scattering of NSW by these defects (whether they were dislocations,[17,18] point defects,[18] or paramagnetic impurity[19]) had the same dependence on Tk as γ_R but an essentially different dependence on the NSW frequency than indicated by (5), namely:

$$\gamma_d = C_d\xi^{-1}(1-\xi^2)^2Tk. \tag{6}$$

Here C_d is a coefficient that depends on the parameter of the interaction between the NSW and defects, and is proportional to the concentration of the latter. It is readily seen that, in principle, a linear combination of (5) and (6) will enable us (because of the presence of the free parameter C_d) to obtain a weak frequency dependence for the relaxation rate γ_1 (Ref. 20). However, the coefficient C_d should then have a definite value in each case. In particular, a least-squares reduction[13] of the experimental data on $CsMnF_3$ yielded the following results:

$$\frac{\gamma_1}{2\pi}[\text{kHz}] = \left[\xi^3A_R + C_d\frac{1}{\xi}(1-\xi^2)^2\right]T[\text{K}]k[10^5\,\text{cm}^{-1}], \tag{7}$$

$$A_R = 3.87\pm0.43, \quad C_d = 2.36\pm0.38, \quad \min\chi^2 = 2.9,$$

where χ^2 is the sum of the squares of the individual deviations of experimental points from the corresponding theoretical values.

An accidental agreement between the values of C_d obtained for three different easy-plane antiferromagnets and the values necessary to "smooth out" the frequency dependence (5) seems most unlikely.

In the second variant, which we have examined in Ref. 13, attention was paid to the fact that calculations of elastic scattering of NSW by fluctuations of the longitudinal nuclear-magnetization component must include higher orders of perturbation theory. However, the diagram technique does not enable us to sum explicitly the contributions of all the fluctuation graphs and hence obtain an exact expression for $\tilde{\gamma}_R$. It was assumed[13] that the higher-order diagrams renormalize differently of the Suhl-Nakamura and the indirect three-spin[21] interactions. This, in turn, alters radically the frequency dependence of $\tilde{\gamma}_R$ and explains satisfactorily the experimental results.

In the present paper we use a different approach to the calculation of $\tilde{\gamma}_R$. The point is that the inclusion of higher-order perturbation theory terms is physically equivalent to the inclusion of the finite lifetime of the virtual electron magnons that effect the indirect coupling between nuclear spins. In other words, ESW relaxation also affects NSW relaxation. This is quite natural since, in addition to their strong dispersion $\omega(k)$, "transformed" from the ESW branch, the nuclear spin waves should also acquire a finite spectrum width $\Delta\omega_k$. In the weak-coupling approximation, we may write

$$\Delta\omega_k = \frac{d\omega_k}{d\omega_{fk}}\Delta\omega_{fk}, \tag{8}$$

where $\Delta\omega_{fk} = 2\gamma_{fk}$ is the ESW spectrum width. In the range of values of the parameters (H,T) in which the experiment[13] was performed, the main contribution to ESW relaxation was made by elastic scattering of f-magnons by fluctuations of the longitudinal component of nuclear magnetization[7] (analogous to the Richards process) with a relaxation rate

$$\gamma_{fk} = \frac{I(I+1)}{12\pi}(V_0^{1/3}k)\frac{\omega_n^2}{\omega_{fk}}\left(\frac{J_0}{k_B\Theta_N}\right)^2.$$

Elastic scattering of the magnons by fluctuations of the nuclear-magnetization longitudinal component [the joint action Richards relaxation (5) and of the contribution introduced by the electron subsystem in accordance with (8)]

leads to the following expression for the NSW relaxation parameter:

$$\frac{\gamma_1}{2\pi}[\text{к}\Gamma\text{ц}] = A_R\left[\xi^3 + \frac{1}{\xi}(1-\xi^2)^2\right] T[\text{К}]k[10^5\text{cm}^{-1}]. \quad (9)$$

It is important to note that (9) does not contain adjustable parameters and the frequency dependence of γ_1 is in good agreement with experiment[13] [for $C_d = A_R$, the frequency dependence is the same as (7)]. The calculated result is $A_R = 2.6 \pm 0.1$.

Thus, our analysis enables us to conclude that the NSW relaxation process with linear dependence on temperature and on the modulus of the wave vector is intrinsic: it is determined by the scattering of electron and nuclear magnons by fluctuations in the longitudinal component of nuclear magnetization.

It is interesting to note that the interdependence of ESW and NSW damping means that we are able, at least in principle, to obtain data on the relaxation of magnons belonging to both branches by studying only one of them.

4. NSW RELAXATION IN A SPECIMEN CONTAINING DEFECTS

The detailed study of intrinsic nuclear-magnon relaxation processes in the ferromagnets $MnCO_3$ and $CsMnF_3$ enabled us to investigate NSW damping in crystals containing defects. The aim of our study was to identify the contributions made to the observed relaxation rate by nonintrinsic processes, and to interpret the resulting data. This analysis can be used, at least in principle, to estimate the concentration and types of defects in the specimens, which is usually a relatively complex problem. The simplest and most common cause of defects is non-stoichiometry of the chemical composition of the crystal, which occurs in any specimen to some extent. We therefore selected a $CsMnF_3$ crystal with deliberately altered stoichiometry for our investigation. The specimen contained a 1% Mn excess over its stoichiometric composition. The uniformity of the distribution of the excess atoms within the specimen was not monitored.[2)] The x-ray diffraction pattern of the $CsMnF_3$ crystal with modified stoichiometry was the same as that of pure specimens.

Examination of NMR and AFMR spectra shows that the frequencies of these resonances and their linewidths for the specimen with modified stoichiometry do not differ to within experimental uncertainty from the corresponding data for the $CsMnF_3$ specimen with nominal stoichiometric composition. This enables us to conclude that the difference between the NSW relaxation rates obtained for these specimens is not due to an essential change in the crystal structure, or an appreciable change in the spectra, but entirely due to interactions between nuclear magnons and crystal defects.

The parametric NSW excitation was "hard" in character, i.e., the parametric instability set in abruptly when the microwave field amplitude reached the value h_{c1}, and then smoothly vanished at $h_{c2} < h_{c1}$. The relaxation rate was calculated from h_{c2} and it was found that, at frequencies $\omega_k/2\pi > 400$ MHz, the relaxation of nuclear spin waves in the specimen with modified stoichiometry was the same to within experimental error as in the specimen with nominal stoichiometric composition: $\gamma = \gamma_1 + \gamma_2$. Further decrease of the frequency of the investigated NSW reveals, against the background of intrinsic processes $\gamma_1 + \gamma_2$, a process with $\gamma_{id} \propto Tk$ and a much stronger frequency dependence than that of γ_d (6) in elastic scattering by defects. This is most clearly seen for temperatures $T < 2$ K, when the contribution of the intrinsic process to total relaxation $\gamma_2 \propto T^5$ is negligible. Figure 2 shows the frequency dependence of the total NSW relaxation $\gamma \propto Tk$. It is clear that the relaxation rate γ increases by a factor of about 15 as the frequency $\omega_k/2\pi$ is reduced from 400 to 150 MHz. This suggests that, as the frequency is reduced, a new relaxation process is "turned on" and is probably connected with the fact that conservation laws cannot be satisfied for this process at high frequencies $\omega_k \gtrsim 2\pi \cdot 400$ MHz.

In addition to the process with the rate γ_{1d}, the sudden appearance of one more relaxation process with relaxation rate $\gamma 2_d$ (see Fig. 2) was observed at lower NSW frequencies $\omega_k < \omega_k^* = 2\pi(358 \pm 5)$. This is also probably due to hindrances imposed by the energy and momentum conservation laws at $\omega_k > \omega_k^*$. To within experimental uncertainties, the relaxation rate $\gamma 2_d$ is independent of the modulus of the wave vector k, and its temperature dependence is $\gamma_{2d} \propto T^3$ (for $R = 4.2$ K we have $\gamma_{2d} = 2\pi(50 \pm 25)$ kHz). An analogous NSW relaxation was observed previously in the antiferromagnetic $CsMnCl_3$ crystal.[12] It was suggested[12] that the similar nuclear-magnon damping mechanism was due to impurities, and certain possible relaxation channels were examined.

Let us now consider the interpretation of the new NSW relaxation process with $\gamma_{id}(T,k,\omega_k)$ observed in the $CsMnF_3$ specimen with modified stoichiometry. None of the processes involving relaxation on defects that were considered above[17-19] is capable of explaining the frequency dependence

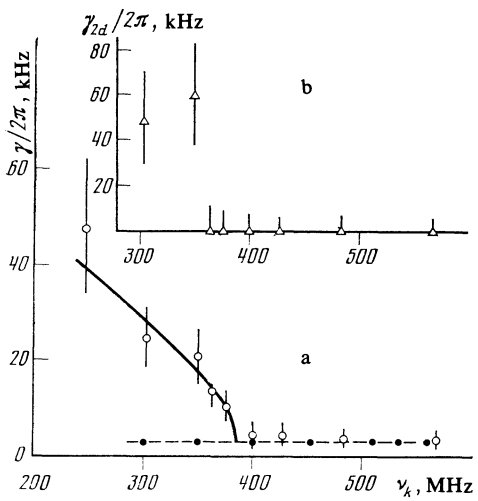

FIG. 2. (a) Frequency dependence of relaxation rate γ of $CsMnF_3$ with nominal stoichiometric composition (full points) and with modified stoichiometry (open circles) for nuclear spin waves with $k = 0.7 \times 10^5$ cm^{-1} at $T = 1.7$ K. Solid curve—theoretical results (12). (b) Frequency dependence of relaxation rate γ_{2d} in $CsMnF_3$ specimen with modified stoichiometry for nuclear spin waves with $k = 0.7 \times 10^5$ cm^{-1} at $T = 4.22$ K.

of γ_{ld}. Satisfactory agreement with experiment is obtained if the defect is looked upon as a two-level system with a transition frequency $\omega_d = 2\pi \cdot 732$ MHz and the following NSW relaxation channel: a nuclear magnon $(\omega_k, \mathbf{k})$ coalesces with another nuclear magnon $(\omega_q, \mathbf{q})$ and gives rise to a transition from the lower level of the defect to an upper level with energy $\hbar\omega_d = \hbar\omega_k + \hbar\omega_q$. If the two-level system is described by the spin operator $\hat{\sigma}(\sigma = \frac{1}{2})$, this process may be determined by an anharmonism of the form

$$\lambda(\Delta l_y \hat{\sigma}^+ + \text{h.c.}), \quad \Delta l_y = l_y - \langle l_y \rangle,$$

where Δl_y is the amplitude (quadratic in the spin deviations), of the oscillations of the antiferromagnetic vector, $\mathbf{l} = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0 = (0, l_y, 0)$, and λ is the characteristic interaction energy. Bearing in mind the connection between electron and nuclear spin oscillations, it can be shown after some algebra (see, for example, Ref. 2) that the amplitude for this process is

$$\Psi_{2n\sigma} \approx \frac{\lambda}{\mathcal{N}} \frac{(g\mu_B H_\Delta)^2 J_0 \omega_n^2}{\hbar^3 (\omega_{fk}\omega_{fq})^2 (\omega_k \omega_q)^{1/2}}. \tag{10}$$

The corresponding relaxation rate is

$$\gamma_{2n\sigma} = n_d \pi V_0 b_\sigma \int \frac{d\mathbf{q}}{(2\pi)^3} \left| \frac{\Psi_{2n\sigma}}{\hbar} \right|^2 \operatorname{sh} \frac{\hbar\omega_k}{2k_BT} \times \left(\operatorname{sh} \frac{\hbar\omega_q}{2k_BT} \operatorname{sh} \frac{\hbar\omega_d}{2k_BT} \right)^{-1} \delta(\omega_k + \omega_q - \omega_d), \tag{11}$$

$$b_\sigma = \frac{\sigma(\sigma+1)}{3} \frac{\hbar\omega_d}{k_BT}.$$

Here n_d is the concentration of the defects, $g\mu_B H_\Delta$ is the hyperfine gap in the electron-magnon spectrum, and the hyperbolic functions describe the equilibrium occupation numbers of the nuclear magnons and of the two-level system.

Direct evaluation of (11) yields

$$\frac{\gamma_{2n\sigma}}{2\pi} = \frac{n_d}{(2\pi)^2} \frac{\sigma(\sigma+1)}{I(I+1)} \left(\frac{k_BT}{\hbar} \right) \frac{\lambda^2 J_0}{(k_B\Theta_N)^3} \frac{(1-\xi^2)^2}{\xi_d - \xi} \frac{g\mu_B \alpha\tilde{k}}{\hbar\omega_n}, \tag{12}$$

where

$$\xi_d \equiv \frac{\omega_d}{\omega_n}, \quad (\alpha\tilde{k})^2 = H_\Delta^2 \left[\frac{(\xi_d - \xi)^2}{1 - (\xi_d - \xi)^2} - \frac{\xi^2}{1-\xi^2} \right] + (\alpha k)^2. \tag{13}$$

Equation (12) gives a near-linear dependence on temperature and wave-vector modulus for frequencies $|\omega_d/2 - \omega_k| \ll \omega_d/2$, in accord with the experimental data. Analysis of the data by the least-square procedure (5 points) yielded $\omega_d = 2\pi(732 \pm 6)$ MHz, $n_d^{1/2}\lambda/k_B = (1.45 \pm 0.03)$ K and $\min \chi^2 = 1{,}1$. Substituting $n_d \sim 10^{-2}$, we obtain $\lambda/k_B \approx 15$ K, which lies within the range of characteristic energies of exchange interaction between the Mn^{2+} ions. The solid line in Fig. 2 shows the theoretical frequency dependence (12), which satisfactorily describes the experimental results.

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[1)] The relationship with the notation used in Ref. 13 is: $\gamma_j \equiv 2\pi\Gamma_j$ $\omega_j \equiv 2\pi\nu_j$.

[2)] All the $CsMnF_3$ crystals that we investigated were grown by S. V. Petrov at the Institute of Physical Problems of the USSR Academy of Sciences.

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