# Change in magnetic moment of the easy-plane antiferromagnet $MnCO_3$ by parametric excitation of magnons

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The change in the magnetic moment of the easy-plane antiferromagnet $MnCO_3$ by parametric excitation of magnons with frequencies $\omega_k/2\pi = 18$ GHz has been measured. The magnetic-moment change obtained, associated with the parametric excitation of a single magnon, is $(5.0 \pm 1.5)\mu_B$. In addition, a reduction in the magnetic moment of the specimen, linear in the microwave power, was found. It is suggested that this reduction in magnetic moment is associated with heating of the impurity magnetic subsystem by the microwave power.

## INTRODUCTION

To describe the static and dynamic properties of magnetically ordered crystals it is essential to know the magnon spectrum and also their relaxation properties, i.e., the mechanisms for the interaction of magnons with one another and with other excitations of the magnetic material—phonons, impurity vibrational modes, nuclear magnons etc. One of the classes of magnetic materials which have been thoroughly studied theoretically and experimentally are antiferromagnets with an easy plane of magnetization (AFEPM). The magnon spectrum of materials of this class, as shown by Borovik-Romanov,[1] has two branches: the quasiacoustic $\omega_{1k}$ and the quasioptical $\omega_{2k}$;

$$(\omega_{1k}/\gamma)^2=H(H+H_D)+\alpha_\perp{}^2k_\perp{}^2+\alpha_\parallel{}^2k_\parallel{}^2+H_\Delta{}^2, \tag{1}$$

$$(\omega_{2k}/\gamma)^2=2H_AH_E+H_D(H+H_D)+\alpha_\perp{}^2k_\perp{}^2+\alpha_\parallel{}^2k_\parallel{}^2, \tag{2}$$

where $\gamma$ is the gyromagnetic ratio, $H_D$ the Dzhaloshinskiĭ field, $H_\Delta^2$ the spectrum parameter due to magnetoelastic and hyperfine interactions, $H_A$ the anisotropy field, $H_E$ the exchange field, $H$ the dc magnetic field lying in the basal plane of the crystal, $\alpha_\parallel$ and $\alpha_\perp$ are exchange constants. This magnon spectrum has been confirmed experimentally for a great number of AFEPM ($MnCO_3$, $CsMnF_3$, $FeBO_3$, $CoCO_3$,...), $k_\parallel$ and $k_\perp$ are the components of the wave vector along the crystallographic $C_3$ axis and in the basal plane.

The relaxation properties of magnons in an AFEPM have also been studied both theoretically and experimentally. A lot of information about the relaxation properties of the spin system can be obtained by studying the phenomenon of parametric excitation of magnons (see, for example, the review by Kotyuzhanskiĭ and Prozorova[2]). The action on a specimen by a high-frequency magnetic field $h$ of sufficient amplitude ($h > h_c$) can excite an appreciable number of non-equilibrium magnons of the quasiacoustic branch within a narrow frequency interval. The frequency of the excited magnons $\omega_k$ is equal to half the frequency of the external high-frequency influence $\omega_p$ and the wave vector $k$ is determined by the value of the field $H$. The value of the threshold magnetic field $h_c$ of the parametric process is proportional to the relaxation frequency $\Delta\omega_k$ of the excited group of waves:[3]

$$h_c=\Delta\omega_k\omega_p/\gamma^2(2H+H_D). \tag{3}$$

The relation (3) enables the lifetime of parametrically excited magnons with frequencies $\omega_k$, associated with the pumping field, $T_{2\omega k} = 1/\Delta\omega_k$, to be determined experimentally.

The excitation of a non-equilibrium magnon leads to a reduction in the projection of the magnetic moment **M** of the specimen along the direction of the external dc field **H**. The change in magnetic moment, associated with the excitation of a single magnon of the lower branch of the AFEPM spectrum, is equal to[4]

$$\mu_{\omega k}=-\partial\varepsilon_k/\partial H=-h\gamma^2(2H+H_D)/2\omega_k, \tag{4}$$

where $\varepsilon_k$ is the energy of a magnon with wave vector **k**. It is natural to introduce a time $T_{1\omega k}$, the relaxation time of the specimen's magnetic moment to its equilibrium value upon excitation of a group of magnons with frequency $\omega$ and wave vector **k**.

The change in magnetic moment $\Delta M$ upon excitation of a homogeneous impurity ($\omega/2\pi = 36$ GHz) in the AFEPM $CoCO_3$ was studied by Borovik-Romanov *et al.*,[5] and upon excitation of inhomogeneous oscillations in the AFEPM $FeBO_3$ by Kotyuzhanskiĭ and Prozorova.[4] It was shown in these works that the time $T_{1\omega k}$ in the materials studied exceeds $T_{2\omega k}$ by at least an order of magnitude. The authors explained such a difference by the fact that the most probable relaxation processes determining $T_{2\omega k}$ do not lead to relaxation of the magnetic moment. Thus, in $FeBO_3$ the most probable decay of a parametric magnon is into a magnon and a phonon.[6] The magnon obtained as a result of the decay has a lower frequency and, in accordance with Eq. (4), a larger value of $\mu_{\omega k}$ is associated with it than the original and consequently this process does not lead to relaxation of the magnetization. In so far as spin-lattice relaxation in $FeBO_3$ has a multistep character, the theoretical determination of $T_{1\omega k}$ for comparison with experiment is not a simple problem. Calculation of the time $T_{1\omega k}$ for $CoCO_3$ is still more complicated since in this material there is a more probable magnon scattering process, the scattering by a magnetic impurity.[7]

The aim of the present work is a study of the change in magnetic moment $\Delta M$ of a specimen on parametric excitation of magnons, in such an AFEPM in which the determination of $T_{1\omega k}$ is not such a complicated problem and thus enables an estimate to be made of the change in magnetic moment upon excitation of a single magnon. We chose as material for the study the AFEPM $MnCO_3$, the static and

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dynamic properties of which are at present sufficiently well known.[1,2] The values of the constants determining the magnon spectrum[1] in $MnCO_3$ are:

$$\gamma=2\pi\cdot 2{,}8\,\text{GHz/kOe},\; H_E=320\text{ kOe},\; H_D=4{,}4\text{ kOe},$$
$$H_A=3{,}04\text{ kOe},\qquad H_\Delta{}^2=(5{,}8/T_n+0{,}3)\text{ kOe},$$
$$\alpha_\parallel=0{,}7\cdot 10^{-5}\text{ kOe}\cdot\text{cm}.$$

Here $T_n$ is the temperature of the nuclear spin subsystem (in K).

According to Kotyuzhanskiĭ and Prozorova[8] and to Bar'yakhtar *et al.*,[9] the most probable process in $MnCO_3$ determining $T_{2\omega k}$ is the merging of two magnons of the quasiacoustic branch into a magnon of the quasioptical branch. The most probable relaxation process of magnons of the upper branch is decay into a low-frequency magnon and a phonon. It should be expected from the analysis of the magnon spectrum [Eqs. (1) and (2)] and in accordance with Eq. (4), that the magnetic moments of all secondary quasiparticles are small compared with that of the parametrically excited group of magnons. It would thus be expected that in $MnCO_3$ the time $T_{1\omega k}$ will be close to $T_{2\omega k}$; the latter can be determined from measurements of the threshold for parametric excitation of magnons.

## EXPERIMENTAL METHOD

An apparatus was designed for measuring the change in magnetic moment $\Delta M$ of a specimen under the action of a microwave field, which combined a microwave direct amplification spectrometer with a SQUID magnetometer. Unlike Kotyuzhanskiĭ *et al.*[6] the apparatus enables measurements to be carried out in a range of dc fields 0–150 Oe.

Parametric excitation of magnons was achieved by the method of parallel pumping at a frequency $\omega_p/2\pi = 36$ GHz. A cylindrical cavity tuned to the $H_{012}$ mode in the loop of the microwave magnetic field **h** of which the specimen under study was placed, was used. The dc magnetic field **H** (**h**||**H**) was produced by a superconducting solenoid operating in the short-circuited mode. A cw Gunn-diode microwave generator was used. The microwave power passing through the resonator, proportional to the square of the field at the specimen $h^2$, was measured by a square-law detector which was calibrated in terms of a thermistor power meter. The field $h$ on the specimen was calculated from the incident power and the cavity parameters, with an absolute uncertainty of 20%.

The $MnCO_3$ specimen studied was grown from the same batch as the specimens investigated by Kotyuzhanskiĭ and Prozorova[8] and was a 3 mm diameter disk of thickness 2 mm. The crystallographic $C_3$ axis of the specimen was perpendicular to the plane of the disk. The pickup loop of the flux transformer was wound on the outer wall of the vhf resonator (diameter 18 mm) so that the current induced in the transformer was produced by a change in magnetization of the specimen in the direction of the dc field **H**. The orientations of the fields **H** and **h** and of the crystallographic $C_3$ axis and the measured projection of $\Delta M$ are shown in Fig. 1a. The second arm of the flux transformer was inductively coupled to the measuring cell of the Zimmerman type of standard high-frequency SQUID magnetometer.[10] The microwave radiation in the waveguide channel and in the

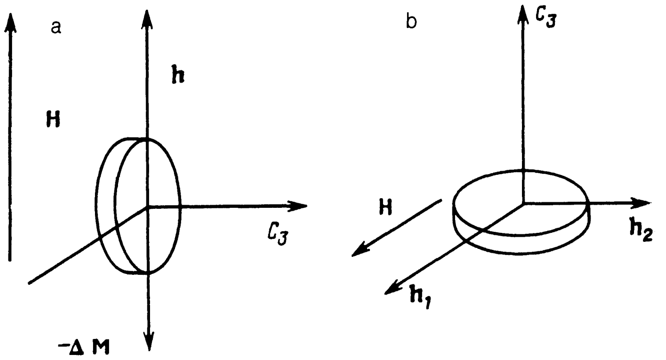


FIG. 1. Mutual orientations of the dc field **H**, the microwave magnetic field, the crystallographic $C_3$ axis, and the measured change in magnetic moment of the specimen $\Delta M$ in the experiments: a) when determining the change in magnetic moment on parametric excitation of magnons; b) when determining the heating of the specimen under the action of microwave power.

low-temperature part of the apparatus was sufficiently well screened to avoid a direct influence of the radiation on the magnetometer operation. The calibration of the SQUID magnetometer was carried out with the help of a small current coil imitating the magnetic moment of the specimen. The uncertainty in the calibration was not more than 10% of the measured quantity $\Delta M$. The chief source of noise was vibration of the flux-transformer loop in the solenoid field, and the sensitivity of the apparatus was therefore strongly dependent on the magnitude of $H$. The values of the uncertainties in the determination of $\Delta M$ are shown below in Fig. 1. The apparatus with the specimen was housed in a liquid-helium Dewar vessel. The temperature was determined from the helium vapor pressure and was 1.2 K.

## EXPERIMENTAL RESULTS

The dependence of the microwave power $P_{\text{inc}}$ incident on the cavity on the power $P_{\text{trans}}$ received at the cavity exit is shown in Fig. 2. For small values of $P_{\text{inc}}$ the dependence is linear. The observed break in the $P_{\text{inc}}$ ($P_{\text{trans}}$) dependence corresponds to the threshold of parametric excitation of magnons. The threshold was additionally determined, on operating the microwave oscillator in the pulsed mode, from the characteristic distortion on the $P_{\text{trans}}$ ($t$) oscillogram corresponding to the development of temporal instability.[2] In a field $H = 100$ Oe the critical field $h_c$ was 0.1 Oe. A crystal of larger dimensions can produce appreciable distortions of the high-frequency field in the resonator, but the good agreement between the measured value of $h_c$ with the value obtained by Kotyuzhanskiĭ and Prozorova[8] allows us to assume that the error in determining $h_c$ is negligible. From the results shown in Fig. 2 and from the measured cavity parameters it was possible to determine the power absorbed $P_{\text{abs}}$ by the specimen on parametric excitation of magnons.

The dependence of the change in the specimen's magnetic moment on $P_{\text{trans}}$ is also shown in Fig. 2. In the region of small powers this dependence falls linearly and on reaching the parametric instability threshold a break is observed and the relation becomes steeper. It is natural to assume that the nonlinear part of the magnetic moment $\delta M$ is associated with parametric excitation of magnons. The dependence of

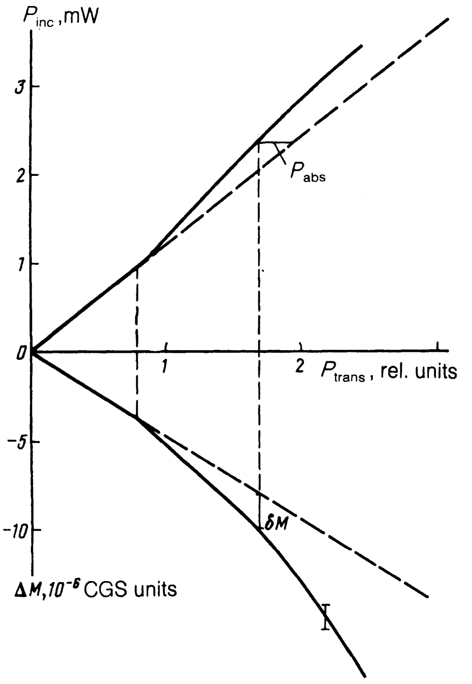


FIG. 2. Dependences of the microwave power incident on the cavity and of the change in magnetic moment of the specimen on the power passing through the cavity. $H = 100$ Oe, $T = 1.2$ K.

$\delta M$ on the power absorbed by the specimen $P_{abs}$ is shown in Fig. 3.

A family of $\Delta M(P_{trans})$ curves is shown in Fig. 4 for different values of the dc field $H$. The break in the relations for all $H$ corresponds to the start of the parametric process. We note that the $\delta M(P_{abs})$ curves for values of the field 50, 100, and 150 Oe agree within the limits of experimental error.

To avoid complications in the interpretation of experimental results it was necessary to determine the characteristic field at which the $MnCO_3$ specimen becomes single-domain. Unfortunately we do not know of work in which the transformation processes of the specimen to single-domain have been studied under the action of a dc field, by direct optical methods in the AFEPM $MnCO_3$. We have thus had to use the following indirect method. The magnetic hf susceptibility of an AFEPM depends on the angle between the weak ferromagnetism vector and the direction of **h**, so that as the specimen becomes single-domain its magnetic susceptibility and consequently the natural frequency of the resonator change. We observed experimentally the shift at low fields in the natural frequency of the resonator with specimen for $\omega/2\pi = 20$ MHz. In fields $70 < H < 300$ Oe the natural frequency of the resonator did not change to an accuracy of 3 MHz, from which we conclude that for $H > 70$ Oe the specimen is practically single-domain.

## DISCUSSION OF THE RESULTS

From the $\delta M(P_{abs})$ dependence shown in Fig. 3 the value of $\mu_{\omega k}$ can be estimated. The number of parametrically excited magnons associated with microwave pumping is

$$n = 2\pi P_{abs}/\hbar\omega_k\Delta\omega_k.$$

For an absorbed power $P_{abs} = 0.1$ mW the number of

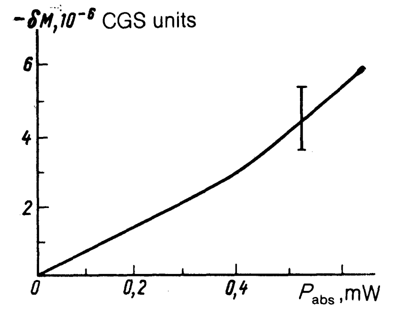


FIG. 3. Dependence of $\delta M$ on the microwave power absorbed by the specimen. $H = 100$ Oe, $T = 1.2$ K.

magnons in the specimen is $n = 1.7 \times 10^{13}$. On the assumption that $T_{1\omega k} \approx T_{2\omega k} = 1/\Delta\omega_k$ we obtain for $P_{abs} = 0.1$ mW an estimate $\mu_{\omega k} = \delta M/n = 4.8\ \mu_B$. This value is at least six times greater than the value $\mu_{\omega k} = 0.68\ \mu_B$ calculated from Eq. (4).

To estimate the heating of the specimen and the reduction in its magnetization associated with it, we used the results of the following control experiment carried out on the same $MnCO_3$ single-crystal. The specimen was attached to the cavity at the output coupling aperture. A dc magnetic field was applied so that the condition for parallel pumping should be satisfied. In the region adjacent to the output coupling aperture of the resonator another microwave magnetic field of lower frequency ($\omega_{p2} = 2\pi \times 17.7$ GHz) was applied with polarization $\mathbf{h}_2 \perp \mathbf{H}$. The mutual orientation of the fields is shown in Fig. 1b. In this way parametric magnons could be excited in the specimen and antiferromagnetic resonance (AFMR) could be observed simultaneously at a frequency $\omega_{p2}$.

The resonant magnetic field in $MnCO_3$ due to hyperfine interaction depends on the temperature of the nuclear spin subsystem, so that the heating of the specimen can be judged from the shift in this field. The shift in the resonance curve was observed only after the threshold power for parametric excitation of magnons was reached. For a power five times greater than the threshold the shift in resonance field was 35 Oe. Such a shift corresponds to heating the nuclear spin system by $\Delta T = 0.075$ K. Since the spin-lattice relaxation time of the nuclear subsystem is much greater than the electron spin and elastic subsystem relaxation time,[4] the heating of the nuclear spin subsystem obtained can only ex-

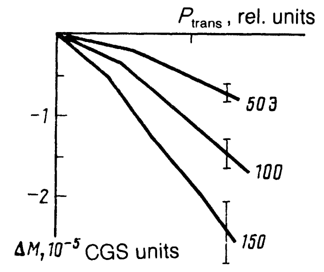


FIG. 4. Dependence of the change in magnetic moment of the specimen on the microwave power passing through the resonator for different values of the dc magnetic field; $T = 1.2$ K.

ceed the heating of the specimen. The temperature dependence of the magnetic moment of $MnCO_3$ at low temperatures has, according to Borovik-Romanov,[1] the form

$$M=M_0(1-\eta T^2/T_N^2), \tag{5}$$

where $M_0 = 180$ CGS units/mole is the spontaneous ferromagnetic moment, $T_N = 32$ K is the Néel temperature and the experimental value of the constant is $\eta = 0.3$. Starting from Eq. (5) and the experimental value of $\Delta T$ we obtain an estimate of the change in magnetic moment $\Delta M$ due to heating of the specimen in the course of the experiment: $\Delta M = 0.5\times 10^{-6}$ CGS units, i.e., considerably less than the observed effect.

The trivial heating of the specimen thus cannot explain the disagreement between the calculated and measured values of $\mu_{\omega k}$. Evidently for $MnCO_3$, as also for $FeBO_3$ and $CoCO_3$,[4,5] the assumption that $T_{1\omega k} \approx T_{2\omega k}$ is incorrect and the relaxation mechanism of the nonequilibrium magnetic moment of the specimen produced by parametric excitation of magnons is more involved than the mechanism described in the introduction and proposed as the basis for existing ideas about the magnon relaxation mechanisms in $MnCO_3$. It is possible that the disagreement is associated with the neglect of an impurity magnon subsystem parametrically excited by the magnons.

We now pass to a discussion of the linear part of the $\Delta M(P_{\text{trans}})$ dependence shown in Figs. 2 and 4. We immediately note that we have no unique interpretation of the observed influence of microwave radiation on the magnetization. The strong dependence of the observed effect on the dc magnetic field (see Fig. 4) evidently indicates an impurity character of the observed effect, since the magnon spectrum [Eq. (1)] in this field range hardly changes. Numerical estimates of the change in magnetization due to nonresonant excitation of AFMR and to resonant excitation of inhomogeneous oscillations at the specimen boundaries also do not correspond to the experimental values obtained.

We associate the reduction in magnetization with heating of the impurity magnon subsystem by the microwave field. To verify such an interpretation of the observed effect we studied the influence of microwave action on the magnetic moment of the well-studied AFEPM $CoCO_3$ with $Fe^{2+}$ magnetic impurity.[12] A linear $\Delta M(P_{\text{inc}})$ dependence was also observed in this material and the value of the slope of the curve in $CoCO_3$ depends strongly on the frequency. As the pumping frequency approaches the frequency of the impurity level (47 GHz) the slope increases by not less than two orders of magnitude, which is evidence in favor of the hypothesis put forward. However, the conclusion about the effect of excitation of an impurity on the spontaneous magnetic moment in $MnCO_3$ requires further investigation.

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