Change of magnetization following parametric excitation of magnons in antiferromagnetic $FeBO_3$

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We measured the change $\Delta\mathscr{M}$ of the magnetic moment in the easy-plane antiferromagnet $FeBO_3$ following parametric excitation of magnons of frequency $\omega_{\mathbf{k}} = 17.7$ GHz. The measurements were performed at $T = 4.2$ K using a superconducting quantum magnetic-flux meter. The measured $\Delta\mathscr{M}_{\rm meas}$ exceeds by at least an order of magnitude the magnetic-moment change $\Delta\mathscr{M}_p$ due to paramagnetic magnons. The observed discrepancy indicates apparently that a significant contribution to the relaxation of parametric magnons is made by their interactions with phonons.

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The elementary excitations in magnetically ordered substances are magnons. From the known magnon spectrum $\varepsilon_{\mathbf{k}}(\mathbf{k})$ one can calculate (see, e.g., the book by Vonsovskii[1]) the equilibrium values of the thermodynamic quantities that determine the magnetic properties of a substance, and in particular the contribution to the temperature dependence of the magnetization. In these calculations it is taken into account that the magnons obey Bose statistics: their average number in a state with energy ε_k is given by

$$\bar{n}_{\mathbf{k}} = \left(\exp\frac{\varepsilon_{\mathbf{k}}}{k_BT} - 1\right)^{-1}$$

to each magnon corresponds an energy $\varepsilon_{\mathbf{k}} = \hbar\omega_{\mathbf{k}}$, and upon excitation of one magnon the projection of the magnetic moment of the sample on the direction of the magnetic field $\mathbf{H}$ changes by an amount

$$\Delta\mathscr{M} = -\partial\varepsilon_{\mathbf{k}}/\partial H. \tag{1}$$

Depending on the magnetic structure, which determines the spectrum of the magnons, the thermodynamic quantities vary with temperature in accordance with different laws. Thus, for example, the change of the magnetization in ferromagnets obeys the known Bloch law $\Delta M(T) \propto T^{3/2}$ In antiferromagnets with magnetic anisotropy of the easy axis type, at temperatures $T \ll T_{AE}$ ($T_{AE} = (\mu/k_B)(2H_AH_E)^{1/2}$ is the temperature corresponding to the gap in the magnon spectrum, H_E is the exchange field, $\mu = g\mu_B$, μ_B is the Bohr magneton, and g is the spectroscopic splitting factor) the temperature dependence of the sublattice magnetization $\tilde{M}$ is described by an exponential law. For antiferromagnets with easy-plane anisotropy, as shown by Borovik-Romanov,[2] the magnon spectrum has a gapless branch and the sublattice magnetizations vary quadratically with temperature:

$$\tilde{M}/\tilde{M}_0 = 1 - \xi(T/T_N)^2, \tag{2}$$

where

$$\xi = \xi_1 = \frac{\mu\tilde{M}_0}{12a^3H_E^2}\left(\frac{k_BT_N}{\mu H_E}\right)^2$$

at $T_H, T_D \ll T \ll T_{AE}$ and $\xi = 2\xi_1$ at $T_{AE} \ll T \ll T_N$; here a is the lattice constant. The foregoing laws were verified by numerous experiments, a review of which is contained in Ref 1.

It is also of interest to investigate the change of the magnetization upon excitation of nonequilibrium magnons, since this permits a study of the mechanism of energy transfer from the excited magnons to the vibrations of the crystal lattice.

The change ΔM of the magnetization following excitation of homogeneous precession ($\mathbf{k} = 0$) was investigated by Bloembergen and Damon[3] in a nickel ferrite and by Borovik-Romanov, Zhotikov, Kreĭnes, and Pankov in antiferromagnetic $CoCO_3$. We know of only one study, that of le Gall,[5] which is devoted to the study of the onset of ΔM upon excitation of inhomogeneous oscillations of the magnetic moment. He measured the change of the Faraday rotation of the plane of polarization of light following parametric excitation of the magnons in ferrimagnetic yttrium iron garnet.

The purpose of the present study was to measure directly the changes of the magnetization of an antiferromagnet when magnons are excited in it parametrically. It is advantageous in this case to investigate weak ferromagnets with easy-plane anisotropy and with a strong Dzyaloshinskiĭ field H_D, for which the change of the magnetization upon excitation of one magnon can be substantially more than one Bohr magneton μ_B. The low-frequency branch of the magnons in such substances is described according to Ref. 2 by the expression:

$$(\omega_{\mathbf{k}}/\gamma)^2 = H(H+H_D) + H_\Delta^2 + \alpha_\parallel^2 k_\parallel^2 + \alpha_\perp^2 k_\perp^2, \tag{3}$$

where γ is the magnetomechanical ratio, H_Δ^2 is a spectrum parameter due to the magnetoelastic and hyperfine interactions while $\alpha_\parallel$ and $\alpha_\perp$ are exchange constants. Thus we have in accordance with (1) for one magnon

$$\Delta\mathscr{M} = -\frac{g\gamma}{2}\frac{2H+H_D}{\omega_{\mathbf{k}}}\mu_B. \tag{4}$$

The object of the investigation was chosen to be $FeBO_3$, for which $\gamma = 2\pi\cdot 2.8$ Ghz·kOe^{-1} and H_D $(T=0) = 108$ kOe.[6]

EXPERIMENTAL PROCEDURE

The magnons were parametrically excited by parallel pumping at a frequency $\omega_p = 2\pi\cdot 35.5$ GHz. We used a rectangular cavity comprising a short-circuited segment of a standard 8-mm-band waveguide tuned to the H_{104} mode. One of the end faces of the cavity was made of copper foil 0.1

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mm thick. The investigated sample, measuring $4\times1\times1$ mm, was secured to this wall at the antinode of the microwave magnetic field $\mathbf{h}$.

The measurements were performed in a zero constant magnetic field. The microwave magnetic field $\mathbf{h}$ was applied to the basal plane of the crystal along its long side. The cw microwave magnetron oscillator operated in the long-pulse regime ($\tau_p = 1$ msec) at a repetition frequency 50 Hz. The microwave power was measured with a thermistor wattmeter with absolute accuracy 20%. The experiments were performed at $T = 4.2$ K. To keep the sample from overheating, the cavity was filled with liquid helium.

The change of the magnetic moment of the sample was measured with a film-type superconducting quantum magnetic-flux meter (SKIMP). The receiving loop of the meter had an area ~ 10 mm^2 and was placed outside the cavity in such a way that its plane was perpendicular to the long side of the crystal. The meter was calibrated against a magnetic field produced by a current-carrying coil of 0.07 mm wire wound to have the shape and size of the investigated sample, and placed symmetric to the sample relative to the receiving loop. The bandwidth of the amplifier of the meter was sufficient for measurements at a pulse duration 1 msec.

The output signal of the meter and the detected microwave pulse that passed through the cavity was recorded with a two-beam oscilloscope. The error in the measurement of the magnetic moment was determined mainly by the error of the setting of the receiving loop relative to the crystal and to the calibration coil, and by the possible deviation of the sample magnetization from the direction of sample's long side. It is estimated not to exceed $\pm 50\%$.

RESULTS OF EXPERIMENT

When the threshold value h_{c1} of the microwave field at the sample is reached, a "step" is observed on the pulse passing through the cavity and corresponds[7] to hard excitation of magnons of frequency $\omega_k = \omega_p/2$. Simultaneously with the appearance of the "step", an abrupt change of the magnetic moment of the sample was recorded.

Since a field $H = 0$ was used in the experiment, domains with different magnetization directions could exist in the sample. These directions should coincide with the directions of easy magnetization in crystal of the basal plane, directions that repeat every 60° in a sample of regular shape in accord with the symmetry. The parametric excitation of the magnons is due to the microwave field component $\mathbf{h}$ which is parallel to the magnetization, and this excitation should therefore start in different domains at different microwave-field values whose ratio is

$$\frac{h_{c2,3}}{h_{c,1}}=\frac{2\cos\varphi}{|\cos\varphi\pm\sqrt{3}\sin\varphi|}, \tag{5}$$

where φ is the minimum angle between $\mathbf{M}_{1,2,3}$ and $\mathbf{H}$; the subscripts 1, 2, and 3 designate the different noncollinear types of domain.

With further increase of h to a value $h_{c2} = 2h_{c1}$ we observed a second step, corresponding to magnon excitation in domains rotated 60° relative to the first ones. From this fact, with allowance for the threshold-field measurement accuracy and for the analysis of (5), it follows that $\varphi < 5°$, i.e., in the field h_{c2} the magnons were excited in domains with $\mathbf{M}$ direction close to the $\mathbf{h}$ direction.

We note also that since we have observed in the fields h_{c1} and h_{c2} one step each, rather than a sequence of small steps that follow one another, there are grounds for assuming, in view of the nonuniformity of the field at the sample, that the number of domains is small and their size is comparable with the sample size.

From the power ΔP absorbed in the sample and from the magnon lifetime $\tau = 0.5\times10^{-7}$ sec known from Ref. 7 we can calculate the total number N_p of the magnons parametrically excited in the sample:

$$N_p=\Delta P\tau/\hbar\omega_k=1.4\cdot10^{13}. \tag{6}$$

The change of the magnetic moment of the sample, corresponding to this number of magnons, can be calculated by using Eq. (4). In $FeBO_3$, excitation of one magnon with $\omega_k = 17.7$ GHz should be accompanied by a ≈ 17-μ_B decrease of the magnetic moment. Thus, the parametric magnons should correspond to a magnetic-moment change

$$\Delta\mathscr{M}_p=-\gamma H_D\Delta P\tau\mu_B/\hbar\omega_k=2.2\cdot10^{-6}\ \mathrm{G\cdot cm^3}. \tag{7}$$

The magnetic-moment change measured by the SKIMP meter, $\Delta\mathscr{M}_{\rm meas} = 2\times10^{-5}$ G·cm^3, exceeds $\Delta\mathscr{M}_p$ by an order of magnitude. We note that the weak ferromagnetic moment of a saturated sample amounts to 2×10^{-1} G·cm^3.

DISCUSSION OF RESULTS

The observed discrepancy means that under parametric excitation the number of magnons in the crystal exceeds substantially the number of paramagnetic magnons with $\omega_k = \omega_p/2$. This discrepancy may turn out to be even larger if account is taken of the possible existence of 180° domains in the sample. These excess magnons can be the result of relaxation of the parametric magnons or of the heating of the sample by the microwave power. The sample heat rise needed to explain the observed decrease of the magnetic moment can be easily estimated from (2), using for $FeBO_3$ the value $\xi = 0.15$ known from Ref. 8. It amounts to more than 10 K, thereby excluding this possibility.

It can thus be concluded that the excess magnons are due to relaxation of the parametric ones. The most probable are three-particle relaxation processes such as: merging of a parametric magnon with a thermal magnon to produce a magnon of the high-frequency branch of the spectrum, decay of a parametric magnon into two phonons or into a magnon and a phonon, and coalescence with a thermal phonon to produce a magnon.[7] The first two processes decrease the total number of magnons. In addition, the magnons that interact with the parametric ones have in the first process a frequency exceeding ω_k by an order of magnitude, and correspondingly shorter lifetimes than the parametric magnons, and in accordance with (4) a smaller magnetic moment. Therefore these processes cannot account for the result.

In the third and fourth processes the number of magnons is conserved. Moreover, the third process results in magnons having a frequency lower than $\omega_{\mathbf{k}}$, and in accord with the foregoing the change of the magnetic moment of the sample will increase rather than decrease. We assume thus that our experiment indicates that the relaxation of magnons of frequency ~ 17 GHz at $T = 4.2$ K in $FeBO_3$ is determined to a considerable degree by their interaction with phonons.

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