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Nonlinear Absorption of Radio Waves in a Noncollinear Antiferromagnet

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The nonlinear absorption of radio waves (200–800 MHz) in a noncollinear cubic antiferromagnet $Mn_3Al_2Ge_3O_{12}$ in an external magnetic field $\mathbf{H} \parallel [001]$ has been studied in the temperature range of 1.2–4.2 K. We attribute the observed dissipation of the electromagnetic energy to the parametric excitation of inhomogeneous surface waves at the boundaries of antiferromagnetic domains.

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Owing to the multiple degeneration of the ground state of a noncollinear cubic antiferromagnet $Mn_3Al_2Ge_3O_{12}$, a multidomain structure stable in a wide range of magnetic fields can exist in it. In this work, we detect the nonlinear absorption of ultrashort radio waves in the manganese garnet $Mn_3Al_2Ge_3O_{12}$ and attribute it to the parametric excitation of inhomogeneous oscillations of the boundaries of antiferromagnetic domains.

Garnet transits to an antiferromagnetic state at a temperature of about 6.8 K [1]. According to neutron-diffraction studies, a planar 12-sublattice noncollinear structure (crystal symmetry group O_h^{10}) is implemented in it: the magnetic moments of Mn^{2+} are coplanar to the (111) plane and collinear to the [211], [121], and [112] directions (see Fig. 1) [2, 3]. When the external magnetic field H is applied along the [001] direction, the rotation of the spin plane occurs and ends when the external field reaches the critical value $H_c \approx 2.4$ T [4, 5].

In the exchange approximation, the magnetic structure of garnet is described by a pair of antiferromagnetic vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ ($\mathbf{l}_1 \perp \mathbf{l}_2$ and $\mathbf{l}_1^2 = \mathbf{l}_2^2 = 1$) [6]. Analysis shows that the ground state is fourfold degenerate in the directions of the vector $\mathbf{n} = [\mathbf{l}_1\mathbf{l}_2]$, which can be collinear to the [111], $[\bar{1}11]$, $[1\bar{1}1]$, and $[\bar{1}\bar{1}1]$ directions. In the magnetic field $\mathbf{H} \parallel [001]$, this degeneration holds up to the phase-transition field H_c, above which $\mathbf{n} \parallel [001]$.

Thus, four types of antiferromagnetic domains with different orientations of the vector $\mathbf{n}$ can coexist in a garnet crystal. The domain structure holds at a small deviation of the field H from the fourth-order axis until the field in the (110) plane is ≤700 Oe [4].

The magnetic field along the [001] direction with relative inhomogeneity less than 0.1% in the size of the sample was created by a superconducting solenoid.

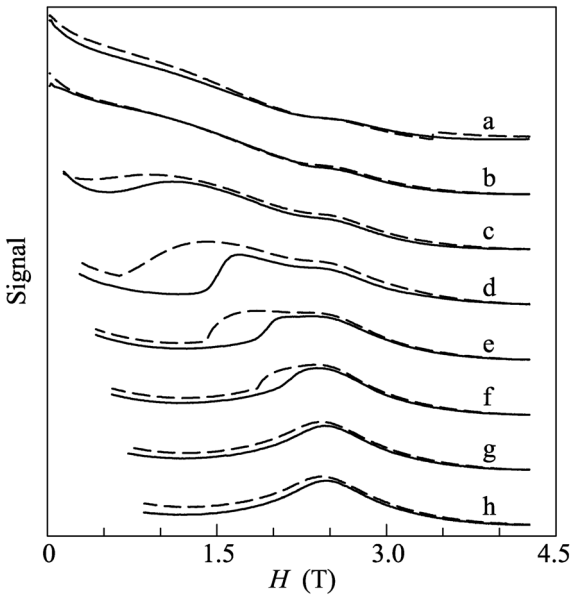

Fig. 1. Power dependences of the shape of the detector signal at a frequency of 685 MHz in $Mn_3Al_2Ge_3O_{12}$ in an external magnetic field $\mathbf{H} \parallel [001]$ for the polarization of the radio-frequency field $\mathbf{h} \perp \mathbf{H}$ and $T = 1.2$ K. The solid and dashed lines correspond to an increase and a decrease in the magnetic field of the solenoid. The letters mark the shapes of the signal corresponding to various radio frequency powers supplied to the resonance system: (a) 20, (b) 19, (c) 18, (d) 17, (e) 16, (f) 15, (g) 13, and (h) 10 dBm.

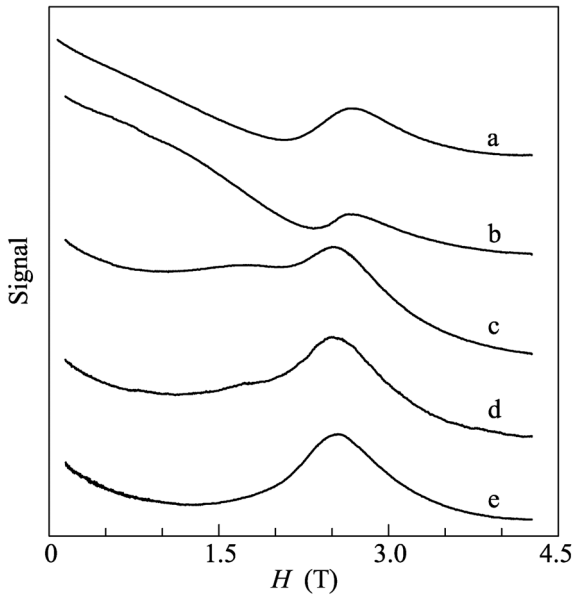

Fig. 2. Temperature dependences of the shape of the detector signal at a frequency of 530 MHz in $Mn_3Al_2Ge_3O_{12}$ at a constant power and $\mathbf{h} \perp \mathbf{H}$ in the temperature range of 1.2–4.2 K. The letters mark the shapes of the signal corresponding to the temperatures of (a) 4.2, (b) 3.3, (c) 2.0, (d) 1.6, and (e) 1.2 K.

The single-crystal sample was either directly immersed in a bath with liquid helium or placed in a vacuum chamber with a heat-exchange ^{4}He gas, which was in a bath with liquid helium. The temperature $T =$ 1.2–4.2 K was controlled in the experiment by the pressure of saturated helium vapor in the bath. In our experiments, we used the broadband resonance system of the "split-ring" type [7]. The transmitted power of the radio frequency field h was detected by a planar diode when the magnetic field was varied at a fixed frequency ω (its stability in the experiment was $\Delta\omega/\omega \sim 10^{-5}$).

We previously studied the magnetic structure of the noncollinear antiferromagnet $Mn_3Al_2Ge_3O_{12}$ by analyzing the spectra of nuclear magnetic resonance (NMR) of ^{55}Mn in the linear absorption regime [5]. In those experiments, three NMR lines were observed in a narrow frequency range near 30 MHz in fields $H < H_c$ only when the radio frequency field had the polarization $\mathbf{h} \parallel \mathbf{H}$.

In this work, we observe the absorption of radio waves with the polarization $\mathbf{h} \perp \mathbf{H}$ in the continuous frequency range of 200–800 MHz when the amplitude of the radio frequency field is above a certain threshold value. Such a spectrum cannot be explained by the resonance properties of the nuclear system in $Mn_3Al_2Ge_3O_{12}$ at $H < H_c$.

Figure 1 shows the power dependences of the shape of the detector signal at a frequency of 685 MHz in $Mn_3Al_2Ge_3O_{12}$ for the polarization of the radio-frequency field $\mathbf{h} \perp \mathbf{H}$ and a temperature of 1.2 K. The letters mark the shapes of the signal corresponding to various radio frequency powers supplied to the resonance system: (a) 20, (b) 19, (c) 18, (d) 17, (e) 16, (f) 15, (g) 13, and (h) 10 dBm (in decibels with respect to 1 mW). The solid and dashed lines correspond to an increase and a decrease in the magnetic field of the solenoid. The resonance absorption corresponding to a branch of low-frequency electron–nuclear oscillations in the high-field phase of manganese garnet ($H > H_c$) is observed in all curves in a field of about 2.5 T. When the pump power is above 15 dBm, the shape of the signal changes qualitatively: additional absorption appears at $H < H_c$. In a range from 15 to 18 dBm, hysteresis is also observed in the magnetic field scans. Such phenomena are not observed for the radio frequency field with the polarization $\mathbf{h} \parallel \mathbf{H}$.

Figure 2 shows the temperature dependences of the shape of the detector signal for the polarization $\mathbf{h} \perp \mathbf{H}$ in the temperature range of 1.2–4.2 K. The letters mark the shapes of the signal corresponding to the temperatures of (a) 4.2, (b) 3.3, (c) 2.0, (d) 1.6, and (e) 1.2 K. At a temperature of 1.2 K and a power of 10 dBm, only resonance absorption is observed near H_c. With an increase in the temperature, an additional absorption signal appears. At a temperature of 4.2 K, it is observed in all fields $H < H_c$. Such qualitative changes in the signal shape do not occur with an increase in the temperature at $\mathbf{h} \parallel \mathbf{H}$.

The parametric excitation of nuclear spin waves in the bulk of $MnCO_3$ and $CsMnF_3$ crystals was studies in [8, 9] under the conditions of longitudinal radio frequency pumping and double resonance with the use of the dependence of the position of the antiferromagnetic resonance line on the temperature of the nuclear magnetic system. Such phenomena can also apparently be observed in manganese garnet, but only in a narrow range near H_c and at the polarization $\mathbf{h} \parallel \mathbf{H}$ different from that used in this work. Furthermore, at $H < 0.8H_c$, in view of the features of the spectrum of electron–nuclear oscillations in $Mn_3Al_2Ge_3O_{12}$, the frequency range of nuclear magnons contracts strongly to the range of 10–40 MHz near 620 MHz [5]. Thus, the corresponding frequency of the parametric pumping should be >1200 MHz, which is noticeably higher than radio frequencies in our experiment. The observed phenomenon also cannot be explained by the resonance creation of acoustic phonons. Indeed, the cross section for this process should be independent of H. Therefore, absorption would be observed at $H > H_c$. However, this was not observed in the experiment.

In the first approximation, the radio frequency field $h(t)$ for the electronic system in $Mn_3Al_2Ge_3O_{12}$ can be considered as quasistatic, because its frequency ($\omega/2\pi < 1$ GHz) is much lower than the frequencies of antiferromagnetic resonance whose branches are above 20 GHz [6]. The magnetic energy density

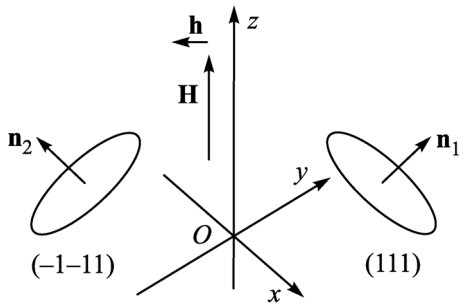

Fig. 3. Static magnetic field **H** is oriented along the fourth-order axis of garnet ([001] direction or the Oz axis), whereas the alternating field component **h** is applied in the (110) or (xy) plane. The force $(\chi_{\parallel} - \chi_{\perp})Hh(t)\sin 2\alpha$ acts on a unit area of the boundary of domains with $\mathbf{n}_1 \parallel [111]$ and $\mathbf{n}_2 \parallel [\bar{1}\bar{1}1]$.

depending on the orientation of the magnetic field $\mathbf{H} + \mathbf{h}$ with respect to **n** has the form

$$\frac{1}{2}(\chi_{\perp} - \chi_{\parallel})(\mathbf{H} + \mathbf{h}, \mathbf{n})^2, \qquad (1)$$

where $\chi_{\perp}$ and $\chi_{\parallel}$ are the components of the susceptibility tensor in the spin plane and parallel to the direction **n**, respectively. In our experiment, $\mathbf{H} \parallel [001]$ and the linearly polarized field **h** lies in the (110) plane. Thus, the difference between the magnetic energy density of domains with $\mathbf{n}_1 \parallel [111]$ and $\mathbf{n}_2 \parallel [\bar{1}\bar{1}1]$ is (see Fig. 3)

$$(\chi_{\parallel} - \chi_{\perp})Hh(t)\sin 2\alpha, \qquad (2)$$

where α is the angle between the vectors **H** and $\mathbf{n}_{1,2}$. Expression (2) specifies the force that acts on a unit area of the boundary between domains with $\mathbf{n}_1$ and $\mathbf{n}_2$ and induces its homogeneous oscillations.

The anomalous absorption of radio waves can be considered as the parametric excitation of inhomogeneous oscillations under the action of force (2) [10]. If $\omega_s(\mathbf{k})$ is the frequency of oscillations of the surface with the wave vector **k**, the absorption of a radio frequency field quantum can be accompanied by the resonance creation of two quanta of surface waves: $\omega = \omega_s(\mathbf{k}) + \omega_s(-\mathbf{k})$ ($\hbar\omega_s(\mathbf{k}) \approx \hbar\omega/2 \sim 10^{-2}$ K).

We emphasize that Fig. 2 demonstrates a decrease in the threshold power of the parametric excitation of domain boundaries with an increase in the temperature T. With an increase in the temperature, driving force (2) also decreases because of a decrease in the difference between the susceptibilities $\chi_{\perp}$ and $\chi_{\parallel}$ [4]. Thus, with an increase in the temperature T, the effective relaxation in the system decreases sharply.

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