Nonlinear radiation damping of nuclear spin waves and magnetoelastic waves in antiferromagnets

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Parallel pumping of nuclear spin waves in antiferromagnetic $CsMnF_3$ at liquid helium temperatures and magnetoelastic waves in antiferromagnetic $FeBO_3$ at liquid nitrogen temperature in a helical resonator was studied. It was found that the absorbed microwave power is approximately equal to the irradiated power from the sample and that the main restriction mechanism of absorption in both cases is defined by the nonlinear radiation damping predicted about two decades ago. Nonlinear radiation damping is sure to be a common feature of the parallel pumping technique for all normal magnetic excitations and it must be taken into account for interpretation of nonlinear phenomena in parametrically excited magnetic systems.

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I. INTRODUCTION

Normal magnetic oscillations, such as electronic spin waves, nuclear spin waves, and magnetoelastic waves describe deviations of magnetization from the equilibrium in magneto-ordered systems—ferromagnets, antiferromagnets, and ferrites. Microwave parametric resonance of normal magnetic oscillations is a powerful tool to study both material properties and various nonlinear phenomena in magneto-ordered systems [1–15]. The vast majority of studies of supercritical behavior of spin-wave systems were carried out in microwave resonator cavities. The resonators, however, as a rule were excluded from consideration, when processing and discussing the results of measurements. All observed effects were attributed to the intrinsic magnetic properties of single crystals in a model of sample interacting with a homogeneous microwave field. This simplified picture can lead to misinterpreted results and incorrectly determined nonlinear interaction constants. As in laser physics, the dynamics of electromagnetic oscillations in the resonator cavity can play an extremely important role in the process of microwave pumping of normal magnetic excitations.

Parallel pumping of magnetic excitations, in which the microwave magnetic polarization is parallel to the external magnetic field, is one of the most convenient and popular methods of parametric resonance. In this case a microwave magnetic field $\mathbf{h}(t)$ enhanced by a microwave resonator is applied to the sample parallel to its equilibrium magnetization, which is parallel to the steady external magnetic field $\mathbf{H}$. The alternating magnetic field excites the parametric resonance of the form $\omega_p = \omega_\mathbf{k} + \omega_{-\mathbf{k}}$, where ω_p is the pumping field frequency and $\omega_\mathbf{k} = \omega_{-\mathbf{k}}$ are the half-pump frequencies of excited in the sample parametric pair of waves with oppositely oriented wave vectors $\mathbf{k}$ and $-\mathbf{k}$.

The excited-wave amplitudes grow exponentially when the microwave-field amplitude h on the sample exceeds the parametric resonance threshold h_c. According to S theory [5,7], this growth is restricted by the nonlinearities of the magnetic system, which are exhibited by (a) phase mismatching of the forced magnetic oscillations with the microwave field and (b) by the positive nonlinear magnetic relaxation due to nonlinearities of magnetic systems. In this theoretical picture the resonator cavity is considered to be just as an ancillary system that enhances microwave field amplitude on a sample. No other effect associated with the microwave resonator cavity is assumed in this small-sample-approximation approach.

Actually, the process of parallel pumping includes two steps. First, the external microwave source excites the same frequency ω_p microwave magnetic oscillation $h(t)$ of the resonator cavity. Second, this magnetic oscillation excites the parametric pair ($\omega_\mathbf{k}$ and $\omega_{-\mathbf{k}}$) of magnetic excitations of the sample. In principle, one can expect a backward radiation of the parametric pairs and a bunch of associated effects in the system of two interacting in resonance oscillations. However, in the simple picture of the small-sample approximation this backward radiation is assumed to be negligibly small compared to intrinsic absorption; only an energy flow from the microwave pump to the sample is taken into account.

The main focus of the present paper is to demonstrate what was theoretically predicted two decades ago [16,17]: that nonlinear radiation damping, which is an effect due to backward irradiation of parametric pairs to the resonator, is a common and dominant feature in the process of parallel pumping of magnetic excitations. Examples of the nontrivial role of microwave resonators to the process of parallel pumping of magnetic excitations have already been discussed in Refs. [18,19]; however, these facts did not attract attention and the resonator-less approach is still used for the description of parallel pumping of magnetic oscillations [14,20–22]. In this paper we study the role a helical resonator for parallel pumping of (a) nuclear spin waves in an antiferromagnetic $CsMnF_3$ at liquid helium temperatures and (b) magnetoelastic waves in an antiferromagnetic $FeBO_3$ at liquid nitrogen temperatures. It should be noted that these antiferromagnets are the classical objects to study nuclear spin waves and magnetoelastic waves, respectively.

The concept of nuclear spin waves was introduced by de Gennes *et al.* [23]. The most remarkable property of these excitations is that, at liquid helium temperatures, they exhibit the coupled oscillations of two subsystems that are completely different in their magnetic properties. The electronic spins are

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ordered while the state of the nuclear spins is paramagnetic; the polarization is no more than several percent. The longitudinal part of the hyperfine interaction creates a strong effective magnetic field at the nucleus, which determines the Larmor nuclear magnetic resonance frequency ω_n. The nuclear spin polarized by this field creates a perturbation on the electron shell, which leads to a gap $\gamma H_{\Delta,hf}$ in the electronic spin-wave spectrum $\omega_{e,k}$. Neglecting the transverse part of the hyperfine interaction, the electronic spin-wave branch and the free precession of the nuclear spins with the Larmor frequency can be considered as the normal oscillations of the system. The transverse part of the hyperfine interaction leads to mixing of these "pure" modes. The branches are pushed apart, and this separation is larger, when the interaction between the pure modes is stronger. Homogeneous oscillations are most strongly coupled, and the coupling weakens as the wave vector increases. Thus the nuclear magnetic resonance frequency $\omega_{n,0}$ decreases and becomes noticeably lower than the Larmor precession frequency, and the band $\omega_{n,k}$ of nuclear spin waves arises:

$$\omega_{n,k} = \omega_n \left[1 - \left(\frac{\gamma H_{\Delta,hf}}{\omega_{e,k}} \right)^2 \right]^{1/2}. \quad (1)$$

Here, $\omega_{e,k} = \gamma[H(H + H_D) + H^2_{\Delta,hf} + (\alpha k)^2]^{1/2}$ is the frequency of the electronic spin wave, H_D is the Dzyaloshinskii field, $H^2_{\Delta,hf} \propto 1/T$ is the gap due to the hyperfine interaction, α is the exchange constant, and γ is the gyromagnetic ratio. A detailed review of nuclear spin-wave properties in weakly anisotropic antiferromagnets is given in Refs. [24,25].

Magnetoelastic waves describe normal modes of linearly coupled elastic waves and electronic spin waves in magneto-ordered crystals. So far as the magnetoelastic waves contain both elastic and magnetic components, they can be excited both by elastic vibrations and by an alternating magnetic field. One of the most interesting objects to study magnetoelastic waves is the high Néel temperature antiferromagnet $FeBO_3$ ($T_N = 348$ K). Parallel pumping of magnetoelastic waves in this crystal for the first time was observed in Ref. [26]. The spectrum of magnetoelastic waves in iron borate can be written as [19]

$$\omega_{me,k} = c_e k \left[1 - \left(\frac{\gamma H_{\Delta,ef}}{\omega_{e,k}} \right)^2 \right]^{1/2}, \quad (2)$$

where c_e is the sound velocity, $H_{\Delta,ef}$ describes an efficiency of linear interaction between spin and elastic subsystems, $\omega_{e,k} = \gamma[H(H + H_D) + H^2_{\Delta,me} + (\alpha k)^2]^{1/2}$ is the frequency of the electronic spin wave, and $H_{\Delta,me}$ is the field, which corresponds to magnetoelastic gap.

We show that, beyond the small-sample approximation, the resonator oscillation dynamics plays an extremely important role in the process of parametric resonance of nuclear spin waves and magnetoelastic waves and gives the dominant mechanism of parallel pumping restriction in both cases by nonlinear radiation damping.

II. EXPERIMENT

The experimental absorbing cell is shown in Fig. 1. The sample is placed in an open helical resonator, which is a half-wavelength dipole excited by the pulsed microwave pumping field $\mathbf{h}(t)$. The inner diameter of the helix equals 0.5 cm and the diameter of the copper wire is 0.5 mm. To a first approximation the wire length needed to make the helix is $\simeq\lambda/2$, which is about 15 cm for 1 GHz. The effective volume of this resonator is estimated as $\sim$200 mm^3. The effect of microwave absorption is detected by the receiving antenna. This absorbing cell to study parallel pumping of nuclear spin waves and magnetoelastic waves was used at different temperature conditions.

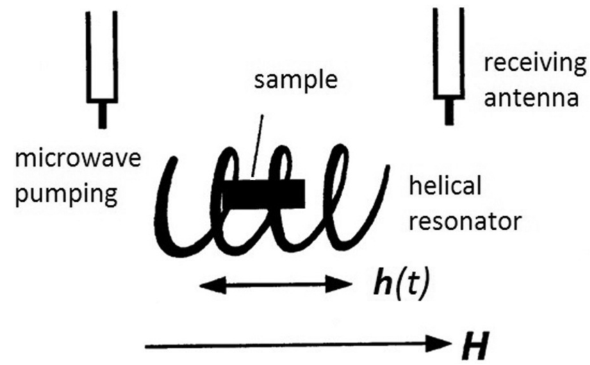

FIG. 1. Schematic diagram of the experimental absorbing cell.

Parametric pairs of nuclear spin waves were excited by a pulsed (300 to 2000 μs) parallel microwave pump with repeating frequency 10 to 100 Hz in the helical resonator with the quality factor $Q \sim 300$ to 500 over a wide range of frequencies $\omega_p = 600$ to 1200 MHz. The measurements were made on single-crystal sample $v_s = 3 \times 3 \times 5$ mm^3 of the easy-plane antiferromagnet $CsMnF_3$ ($T_N = 53.5$ K) at liquid helium temperatures $T = 1.9$ to 4.2 K and magnetic fields $H = 500$ to 2000 Oe. The single-crystal of $CsMnF_3$ was grown in the Kapitza Institute for Physical Problems (Moscow, Russia) in 1980s and has been analyzed in detail in x-ray, neutron, and magnetostatic studies. Our measurements of parametric resonance of electronic and nuclear spin waves confirmed the high quality of this sample. The ratio of the sample volume to the volume of resonator cavity was $v_s/v_R \sim 0.2$. The relaxation rate of parametrically excited spin waves estimated by the threshold amplitude was $\eta_{\mathbf{k}}/2\pi \sim$ 6 to 20 kHz with the accuracy of 25%.

Typical forms of the microwave pump pulse passed through the resonator cavity and microwave irradiation after the pulse are shown in Figs. 2(a) and 2(b), respectively. Figure 2(a) shows the distortion of a rectangular microwave pumping pulse after it has passed the cavity with the sample. The chip on top of the pulse corresponds to the beginning of intensive absorption of incident power, and a general phenomenon, the "tail" behind the rear edge of the pulse demonstrates the microwave radiation from the sample. This tail on the scale increased is shown in Fig. 2(b). Our aim is to study in detail the radiation from the sample, observed after switching off the pump pulse. We could observe this nontrivial radiation at $P/P_c - 1 \gg 1$. Experimentally we observed one peak if the pump frequency was equal to the frequency of the resonator $\omega_p = \omega_R$ and up to three beating peaks if $\omega_p \neq \omega_R$. It should be noted that, below the threshold of parametric resonance, the microwave radiation after the pump pulse demonstrates

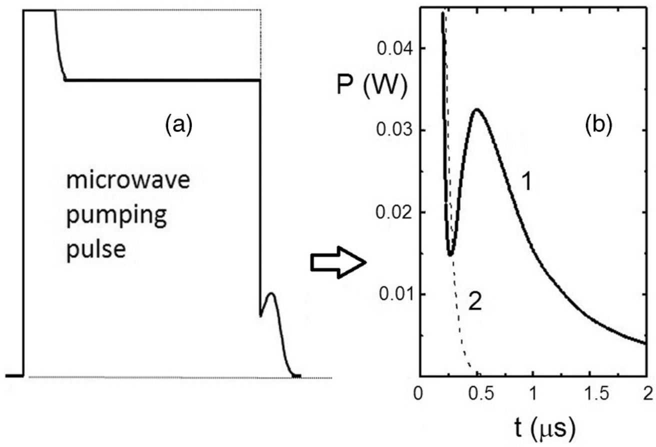

FIG. 2. (a) A typical form of the microwave pumping pulse passed through the helical resonator. One can see a microwave absorption by the sample (upper part) and a nonuniform radiation effect after the end of microwave pumping. (b) Curve 1 demonstrates a nonmonotonic radiation power signal from the sample after the pump pulse was turned off. The pumping power $P \approx 2000 P_c$. Curve 2 demonstrates the case when $P < P_c$, when just an exponentially decreasing radiation from the resonator cavity is observed. The experimental parameters are $T = 2.08$ K, $\omega_p/2\pi = 1094$ MHz, and $H = 1840$ Oe.

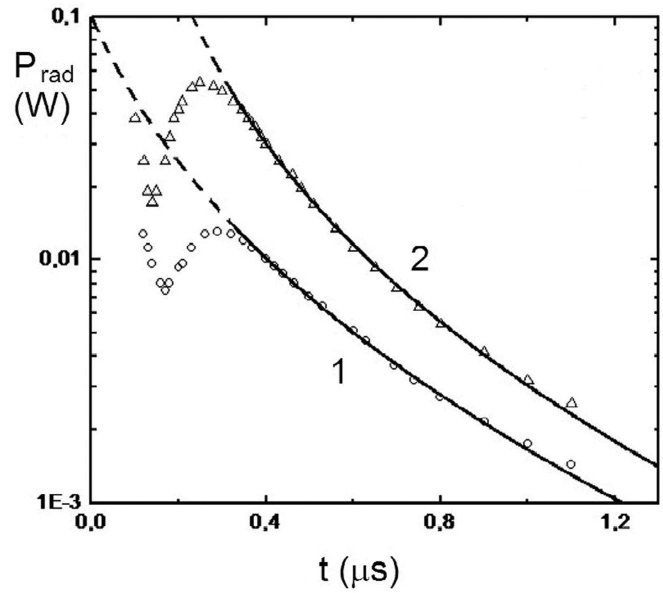

FIG. 3. Irradiation power of magnetoelastic waves (logarithmic scale) versus time after the end of the microwave pump pulse at two overcriticalities: (1) $P/P_c = 13.2$ and (2) $P/P_c = 52.3$ at $T = 77$ K, $\omega_p/2\pi = 1109.7$ MHz, and $H = 231$ Oe. Solid lines describe theoretical fit (see the text).

just an exponential decrease [see curve 2 in Fig. 2(b)], which corresponds to unloaded resonator cavity irradiation.

Qualitatively, nonmonotonic behavior of the radiation can be explained as follows: In the excited state, we have two coupled resonant oscillators. One describes electromagnetic oscillations in the cavity, and the second describes forced oscillations of parametric pairs. After switching off the pump field, both oscillators begin to interfere and relax. The first relaxes quickly, with the rate corresponding to the resonator quality factor [see curve 2 in Fig. 2(b)]. The second oscillation lives much longer; its relaxation rate is mainly determined by the nonlinear radiation damping of parametric pairs. The interference of the two oscillations at the beginning of the radiation results in a deep minimum [see curve 1 in Fig. 2(b)]. Then, the electromagnetic wave in the cavity completely disappears, and we see only the signal emitted by the sample. Exploring this signal, we obtain information about the parametric pair number and relaxation rate.

Parametric pairs of magnetoelastic waves were excited in the $v_s \simeq 20$ mm^3 sample of the "easy-plane" antiferromagnet $FeBO_3$ by the pulsed microwave field of the frequency $\omega_p/2\pi = 900$ to 1200 MHz at magnetic fields $H = 30$ to 500 Oe at liquid nitrogen temperature $T = 77$ K. The single-crystal of iron borate was grown at the Ioffe Physical and Technical Institute (St Petersburg, Russia) in the 1980s and was analyzed in x-ray, optical, and neutron studies. The high quality of this sample was also confirmed by the measurements of parametric resonance of magnetoelastic waves. The ratio of the sample volume to the volume of resonator cavity was $v_s/v_R \sim 0.1$. We observed similar effects of the nonuniform radiation from the resonator-sample system after the end of the microwave pump pulse as in the case of nuclear spin waves. Typical experimental data of irradiation are shown in Fig. 3. Note that the accuracy of the relative measurements at a fixed pump frequency is 5%, i.e., basically it fits in dot size in Fig. 3 (and below, in Fig. 4).

Microwave power applied to the magnetic system of the sample leads to intense forced oscillations of parametric pairs at the frequency of driving force (microwave pumping). The energy of the forced oscillations is absorbed intrinsically by the thermal bath of the crystal. On the other hand, this

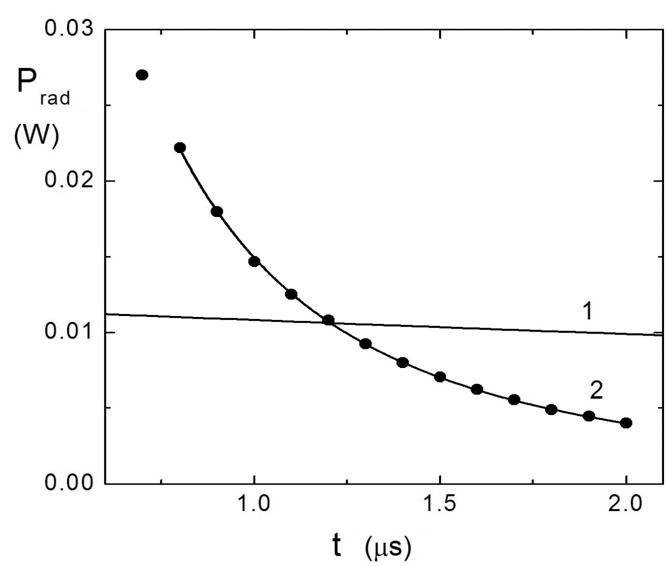

FIG. 4. Radiation power (dots) from the parametrically pumped nuclear spin waves versus time in $CsMnF_3$ at $T = 2.08$ K, $\omega_p/2\pi = 1094$ MHz, and $H = 1840$ Oe. Curve 1 schematically demonstrates the radiation-power slope in the case of linear damping. Curve 2 demonstrates the theoretical fit of formula (4) (see the text).

energy is nonlinearly radiated back to resonator cavity. Our measurements have shown that, at large pumping amplitudes, the nonlinear radiation damping is considerably superior to the intrinsic relaxation in the crystal. It means that almost all energy of parametric pairs abandons the sample backwards to resonator cavity. In other words, the radiated power is approximately equal to absorbed microwave pumping power. Thus, the stationary state of parametric pairs is mainly defined by the radiation from the sample through the "resonator-sample" nonlinear interaction.

III. DATA AND THEORY

Let us consider the monotonically decreasing time dependence of radiated power behind the radiation peak. The decrease of the parametric pairs number $N_k(t)$ is described by the equation $dN_k = -2\eta(N_k)dt$, where $\eta(N_k) = \eta_k + \eta_{nl}N_k$ is the relaxation rate, η_k is the linear part, and $\eta_{nl}N_k$ is the nonlinear part, respectively. Integrating of this equation, one obtains

$$N_k(t) = \frac{\eta_k/\eta_{nl}}{u\exp[2\eta_k(t-t_0)] - 1}, \quad (3)$$

where $u = 1 + \eta_k/\eta_{nl}N_k(t_0)$, t_0 is the starting time ($t \geqslant t_0$).

If we assume that the nonlinear part of the damping is entirely defined by nonlinear radiation damping, then the radiated power $P_{\rm rad}(t)$ can be expressed as

$$P_{\rm rad}(t) = -\hbar\omega_p \frac{dN_k}{dt}\frac{\eta_{nl}N_k(t)}{\eta_k + \eta_{nl}N_k(t)} = \hbar\omega_p \frac{2\eta_k^2/\eta_{nl}}{\{u\exp[2\eta_k(t-t_0)] - 1\}^2}. \quad (4)$$

A. Nuclear spin waves

A typical time slope for radiation power is shown in Fig. 4. Mean-square fitting by using formula (4) with $t_0 = 0.8\ \mu$s gives $\hbar\omega_p 2\eta_k^2/\eta_{nl} = 1.8\times10^{-4}$ W and $\eta_k/\eta_{nl}N_k(0.8\,\mu\text{s}) = 9.05\times10^{-2}$. The linear relaxation rate calculated from the threshold of parallel pumping is $\eta_k = 4.46\times10^4\ \text{s}^{-1}$. Thus we obtain $\eta_{nl} = 1.6\times10^{-11}\text{s}^{-1}$ and $\eta_{nl}N_k(0.8\,\mu\text{s}) = 4.93\times10^5\ \text{s}^{-1}$ which is one order greater than the linear relaxation rate η_k. The number of parametric pairs at $t_0 = 0.8\ \mu$s is equal to $N_k(0.8\,\mu\text{s}) \simeq 3.1\times10^{16}$. This estimate for the number of parametric pairs is in agreement with the estimate obtained in Ref. [27] from the susceptibility in the over-threshold region.

Note that the obtained result is stable to the variation of η_k. For example, if we take linear relaxation rate, say, 40% greater, $\eta_k = 6.24\times10^4\ \text{s}^{-1}$, then from the fit one gets $\eta_{nl}N_k(0.8\,\mu\text{s}) = 4.64\times10^5\text{s}^{-1}$, $\eta_{nl} = 1.4\times10^{-11}\ \text{s}^{-1}$, and $N_k(0.8\,\mu\text{s}) \simeq 3.3\times10^{16}$. We see that the accuracy of the threshold measurement does not seriously affect the nonlinear damping term due to relatively small value of linear damping.

Let us now compare experiment and theory. The theoretical formula for the coefficient of nonlinear radiation damping can be expressed in the form

$$\eta_{nl}^{\rm (theor)} \simeq \xi_R 2\pi\hbar Q \frac{V_k^2}{v_R}, \quad (5)$$

where V_k is the coupling coefficient for the parametric pair with the pump field in the resonator cavity. For antiferromagnetic systems it is proportional to an effective magnetic moment $\hbar\partial\omega_{n,k}/\partial H$ of the excited wave. For nuclear spin waves one has [25,28]

$$V_k = -\frac{1}{2}\frac{\partial\omega_{n,k}}{\partial H} = \frac{\omega_n^2}{4\omega_{n,k}}\frac{\gamma^4(H_{\Delta,hf})^2(2H+H_D)}{\omega_{e,k}^4}. \quad (6)$$

The factor ξ_R in Eq. (5) depends on the geometry of the resonator cavity. For a rectangular resonator cavity one has $\xi_R = 1$. For a helical resonator a compression of half wavelength $\lambda/2$ to the length of helix l occurs and gives $\xi_R \sim \lambda/2l$.

Let us estimate theoretical nonlinear radiation damping (5) for the experiment shown in Fig. 4, using the following parameters: $\omega_n = 2\pi\times666$ MHz, $H_D = 0$, $H_{\Delta,hf}^2 = 6.4/T[K]$ kOe^2, $l \sim 1$ cm. One gets: $\eta_{nl}^{\rm (theor)} \sim 0.6\times10^{-11}$, whose order of magnitude is in a good agreement with the experimental result.

B. Magnetoelastic waves

Let us now consider the experimental results shown in Fig. 3 for magnetoelastic waves. The linear relaxation rate calculated from the threshold of parallel pumping in this case is $\eta_k = 3.2\times10^5\ \text{s}^{-1}$. From the mean-square fit, using formula (4) with $t_0 = 0.4\ \mu$s, one gets (1) $\eta_{nl}N_k(0.4\ \mu\text{s}) = 0.55\times10^6\ \text{s}^{-1}$, $N_k(0.4\ \mu\text{s}) \simeq 2.6\times10^{16}$ for $P/P_c = 13.2$ and (2) $\eta_{nl}N_k(0.4\ \mu\text{s}) = 0.94\times10^6\ \text{s}^{-1}$, $N_k(0.4\ \mu\text{s}) \simeq 4.4\times10^{16}$ for $P/P_c = 52.3$. For both cases we obtain the same experimental coefficient of nonlinear radiation damping $\eta_{nl} = 2.1\times10^{-11}\ \text{s}^{-1}$.

In order to derive a theoretical estimate, we find

$$V_k = -\frac{1}{2}\frac{\partial\omega_{me,k}}{\partial H} = \frac{(c_e k)^2}{4\omega_{me,k}}\frac{\gamma^4(H_{\Delta,ef})^2(2H+H_D)}{\omega_{e,k}^4}. \quad (7)$$

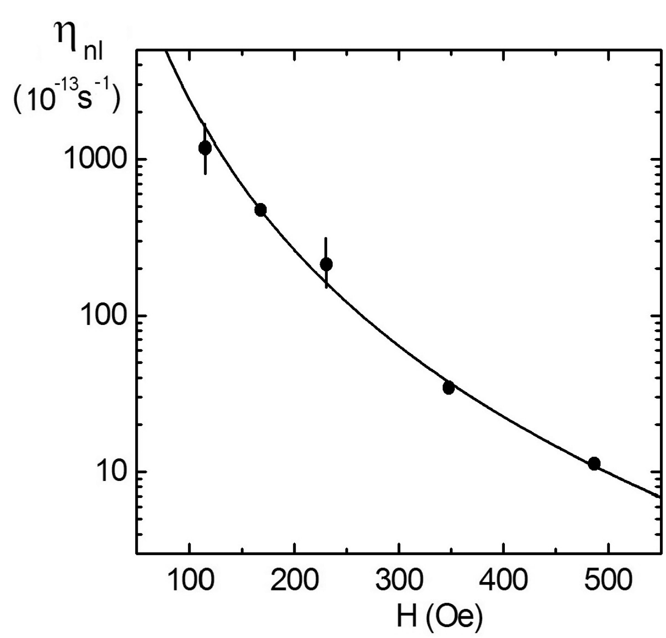

FIG. 5. Magnetic-field dependence for the nonlinear radiation damping coefficient of magnetoelastic waves in $FeBO_3$ at $T = 77$ K and $\omega_p/2\pi = 1109.7$ MHz. Solid line is the theoretical fit.

Thus, by using Eq. (5) and the following parameters for the iron borate: $c_e \simeq 4.8 \times 10^5$ cm/s, $H_{\Delta,ef} \simeq 2$ kOe, $H_{\Delta,me} \simeq 2.2$ kOe, $H_D \simeq 100$ kOe, $\alpha \simeq 0.08$ Oe cm, one gets $\eta_{nl}^{(\mathrm{theor})} \sim 2.9 \times 10^{-11}$ s^{-1}, whose order of magnitude is in a good agreement with the experimental result.

From the theoretical formula (5), it follows that the nonlinear radiation damping is a quadratic function of the effective magnetic moment of the excited waves. For magnetoelastic waves it rapidly decreases with the static magnetic field increase. Dots in Fig. 5 show the magnetic-field dependence of experimentally obtained coefficient of nonlinear radiation damping. The accuracy of measurements is 25%. The solid line represents the theoretical prediction of the field dependence. We see a good agreement with the theory: the nonlinear radiation damping decreased by about 100 times, with increasing field H from 100 Oe to 500 Oe.

IV. CONCLUSION

In this paper we experimentally studied microwave parallel pumping of nuclear spin waves in antiferromagnetic $CsMnF_3$ at liquid helium temperatures and magnetoelastic waves in antiferromagnetic $FeBO_3$ at liquid nitrogen temperature in a helical resonator. From our measurements, it follows that, at large pumping amplitudes, the nonlinear radiation damping is considerably superior to the intrinsic relaxation of parametric pairs in both crystals. The obtained results are in a good agreement with the theory by the field and over-threshold dependencies and are of the order of magnitude of the theoretical prediction. Thus, we demonstrate that the nonlinear radiation damping is the main mechanism of parametric-instability restriction during parallel microwave pumping of two different types of normal magnetic oscillations, nuclear spin waves and magnetoelastic waves in different antiferromagnets. We believe that the nonlinear radiation damping is a common feature of parallel pumping technique and can be detected by purposeful experiments with other types of normal magnetic oscillations in magneto-ordered systems. For example, a specific radiation of parametrically excited spin waves in ferromagnetic YIG after turning off the pump pulse has already been observed in Ref. [29] and has not been explained in the framework of the small-sample approximation.

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