Nonlinear Radiation Damping of Nuclear Spin Waves and Magnetoelastic Waves in Antiferromagnets

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Abstract

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. We believe that the nonlinear radiation damping is a common feature of parallel pumping technique of all normal magnetic excitations and it can be detected by purposeful experiments.

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I. INTRODUCTION

Microwave parametric resonance of normal magnetic oscillations, such as electronic spin waves, nuclear spin waves and magnetoelastic waves is a powerful tool to study linear and nonlinear properties of magnetooordered systems (ferromagnets, antiferromagnets and ferrites) [1–6]. 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. A microwave magnetic field $h(t)$ enhanced by a microwave resonator is applied to the sample parallel to its equilibrium magnetization, which is parallel to the steady external magnetic field $H$. The alternating magnetic field excites the parametric resonance of the form $\omega_p = \omega_k + \omega_{-k}$, where $\omega_p$ is the pumping field frequency and $\omega_k = \omega_{-k}$ are the half-pump frequencies of excited in the sample parametric pair of waves with oppositly oriented wave vectors $k$ and $-k$.

The excited waves amplitudes grow exponentially when the microwave field amplitude $h$ exceeds the parametric resonance threshold $h_c$. According to S theory [3, 4], 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 system. 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 $\omega_p$ microwave magnetic oscillation of the resonator cavity. Second, this magnetic oscillation is absorbed by the parametric pair ($\omega_k$ and $\omega_{-k}$) 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 small sample approximation this backward radiation is assumed to be negligibly small compared to absorption; only an energy flow from the microwave pump to the sample occurs.

Notwithstanding that examples of non-trivial role of microwave resonator to the process of
parallel pumping of magnetic excitations have already been discussed in Refs. [7–11], these facts did not attract much attention and the small sample approximation approach is still in wide use for the description of parallel pumping of magnetic oscillations. The main focus of the present paper is to demonstrate that theoretically predicted two decades ago [7, 8] nonlinear radiation damping, 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.

In this paper we studied parallel pumping in a helical resonator 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.

The concept of nuclear spin waves was introduced by de Gennes et al [12]. The nuclear spin wave denotes the magnetic excitation of mixed electronic and nuclear spin oscillations that is situated in the nuclear magnetic resonance frequency range. The most remarkable property of these excitations is that, at liquid helium temperatures, they exhibit the coupled oscillations of two completely different in their magnetic properties subsystems. The electronic spins are ordered while the state of the nuclear spins is paramagnetic; the polarization is no more than several percent. As a result of mixing of these two subsystems by hyperfine interaction, the frequency of the electronic spin waves increases, and the nuclear magnetic resonance frequency $\omega_{n,0}$ decreases and becomes noticeably lower than the Larmor precession frequency $\omega_n$ of nuclear spins. In other words, there arises the so-called dynamic nuclear magnetic resonance pulling and the band of nuclear spin waves $\omega_{n,k}$:

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

(1)

where $\omega_{e,k} = \gamma[H(H + H_D) + H_{\Delta hf}^2 + (\alpha k)^2]^{1/2}$ is the frequency of electronic spin wave, $H_D$ is the Dzyaloshinskii field, $H_{\Delta hf}^2 \propto 1/T$ is the gap due to hyperfine interaction, $\alpha$ is the exchange constant and $\gamma$ is the gyromagnetic ratio. The detailed review of nuclear spin wave properties in weakly anisotropic antiferromagnets is given in Ref. [13].

Magneetoelastic waves describe normal modes of linearly coupled elastic waves and electronic spin waves in magnetooordered crystals. So far as the magneetoelastic waves contain both elastic and magnetic components, they can be excited both by elastic vibrations and by alternating magnetic field. One of the most interesting objects to study magneetoelastic
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.[14]. The spectrum of magnetoelastic waves in iron borate can be written as [11]:

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

(2)

where $c_ e$ is the sound velocity, $H_{\Delta,ef}$ describes an efficiency of linear interaction between spin and elastic subsystems, $\omega_{e,k} = \gamma [H(H + H_D) + H_{\Delta,me}^2 + (\alpha k)^2]^{1/2}$ is the frequency of 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 the 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 $h(t)$. 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.

Parametric pairs of nuclear spin waves were excited by a pulsed (300 – 2000 $\mu$s) parallel microwave pump with repeating frequency $10 – 100$ Hz in the helical resonator with the quality factor $Q \sim 300 – 500$ over a wide range of frequencies $\omega_p = 600 – 1200$ 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 $T = 1.9 – 4.2$ K and magnetic fields $H = 500 – 2000$ Oe. The ratio of the sample volume to the volume of resonator was $v_s/v_R \sim 0.2$. The relaxation rate of parametrically excited spin waves estimated by the threshold amplitude was $\eta_k/2\pi \sim 6 – 20$ kHz with the accuracy of 25 %.

A typical form of the microwave pump pulse passed thorough the resonator is shown in
Figure 1: Schematic diagramm of the experimental absorbing cell.

the left side of Fig.2. There is a microwave absorption by the parametric pairs (upper part of the pulse) which is demonstrated by the decrease of the pump pulse. At the end of the pulse one can see a general phenomenon, a non-uniform time dependence with a peak of the microwave radiation after the pump pulse, a typical oscillation of the microwave pulse which appears above the threshold of parametric resonance. We could observe this non-trivial radiation at \( P/P_c - 1 \gg 1 \). The peak demonstrates a beating of magnetic oscillation of the resonator cavity mode with the parametric pair \([9]\). In this case the lineshape of the cavity-sample system becomes splitted into two humps \([10]\) which is a direct indication that the small sample approximation is not valid any more. 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 just an exponential decrease (see, curve 2 in Fig.2), which corresponds to unloaded resonator cavity irradiation.

Parametric pairs of magnetoelastic waves were excited in the \( v_s \simeq 20 \text{ mm}^3 \) sample of the “easy-plane” antiferromagnet FeBO\(_3\) by the pulsed microwave field of the frequency \( \omega_p/2\pi = 900 - 1200 \text{ MHz} \) at magnetic fields \( H = 30 - 500 \text{ Oe} \) at liquid nitrogen temperature \( T = 77 \text{ K} \). The ratio of the sample volume to the volume of resonator was \( v_s/v_R \sim 0.1 \). We observed similar effects of the non-uniform radiation from the cavity-sample system after the end of microwave pump pulse as in the case of nuclear spin waves. Typical experimental data of irradiation are shown in Fig.3.

We found very important feature of experiments with irradiation: the radiation power is approximately equal to the absorption power. Thus, the stationary state of parametric pairs is defined by the radiation from the sample through the “sample-resonator” nonlinear
Figure 2: **LEFT:** 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 non-uniform radiation effect after the end of microwave pumping. **RIGHT:** Curve 1 demonstrates a non-monotonic 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.

III. DISCUSSION

Let us consider the monotonically decreasing time dependence of radiated power behind the beating 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, $\eta_k$ is the linear and $\eta_{nl}N_k$ is the nonlinear parts, respectively. Integrating of this equation, one obtains

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

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

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

$$P_{rad}(t) = -\hbar\omega_p \frac{dN_k}{dt} \frac{\eta_{nl}N_k(t)}{\eta_k + \eta_{nl}N_k(t)} = \hbar\omega_p \frac{2\eta_k^2/\eta_{nl}}{u \exp[2\eta_k(t - t_0)] - 1}.$$
Figure 3: Irradiation power of magnetoelastic waves versus time after the end of the microwave pump pulse at two overcriticalities: $P/P_c = 13.2$ and $P/P_c = 52.3$ at $T = 77$ K, $\omega_p/2\pi = 1109.7$ MHz and $H = 231$ Oe. Solid lines describe theoretical fit (see the text).

Figure 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 demonstrate the radiation power slope in the case of linear damping. Curve 2 demonstrate the theoretical fit of formula (4) (see, the text).

A. Nuclear Spin Waves

A typical time slope for radiation power is shown in Fig. 4. Mean-square fit using formula (4) with $t_0 = 0.8$ $\mu$s gives: $\hbar \omega_p \cdot 2\eta_k^2/\eta_{nl} = 1.8 \cdot 10^{-4}$ W and $\eta_k/\eta_{nl}N_k(0.8 \mu s) = 9.05 \cdot 10^{-2}$. The linear relaxation rate calculated from the threshold of parallel pumping is $\eta_k = 4.46 \cdot 10^4$ s$^{-1}$. Thus we obtain $\eta_{nl} = 1.6 \cdot 10^{-11}$ s$^{-1}$ and $\eta_{nl}N_k(0.8 \mu s) = 4.93 \cdot 10^5$ s$^{-1}$ which is one
order greater than the linear relaxation rate $\eta_k$. The number of parametric pairs at $t_0 = 0.8 \mu s$ is equal to $N_k(0.8 \mu s) \simeq 3.1 \cdot 10^{16}$. This estimate for the number of parametric pairs is in agreement with the estimate obtained in Ref.\[15\] from the susceptibility in the overthreshold region.

Note that the obtained result is stable to the variation of $\eta_k$. For example, if we take linear relaxation rate, say, 40% greater, $\eta_k = 6.24 \cdot 10^4 \text{ s}^{-1}$, then from the fit one gets $\eta_{nl}N_k(0.8 \mu s) = 4.64 \cdot 10^5 \text{ s}^{-1}$, $\eta_{nl} = 1.4 \cdot 10^{-11} \text{ s}^{-1}$ and $N_k(0.8 \mu s) \simeq 3.3 \cdot 10^{16}$. We see that the accuracy of the threshold 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:

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

where $V_k$ is the coupling coefficient for the parametric pair with the pump field in the resonator cavity, in other words, it is proportional to an effective magnetic moment $\hbar \partial \omega_{n,k} / \partial H$ of excited wave. For nuclear spin waves one has \[13\] \[16\]:

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

The factor $\xi_R$ in Eq.(5) depends on the geometry of 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 it can result in $\xi_R \sim \lambda/2l$.

Let us estimate theoretical nonlinear radiation damping, Eq.(5) for the experiment shown in Fig.4, using the following parameters: $\omega_n = 2\pi \cdot 666$ MHz, $H_D = 0$, $H_{\Delta,hf}^2 = 6.4/T[K]$ kOe$^2$, $l \sim 1$ cm. One gets: $\eta_{nl}^{(\text{theor})} \sim 0.6 \cdot 10^{-11}$ which of the order of magnitude is in a good agreement with the obtained experimental result.

**B. Magnetoelastic Waves**

Let us 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 $\eta_k = 3.2 \cdot 10^5 \text{ s}^{-1}$. From the mean-square fit using formula (4) with $t_0 = 0.4 \mu s$ one gets: 1)
Figure 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.

$$\eta_{nl} N_k(0.4 \mu s) = 0.55 \cdot 10^6 \text{ s}^{-1}, \quad N_k(0.4 \mu s) \simeq 2.6 \cdot 10^{16} \text{ for } P/P_c = 13.2 \text{ and } 2) \eta_{nl} N_k(0.4 \mu s) = 0.94 \cdot 10^6 \text{ s}^{-1}, \quad N_k(0.4 \mu s) \simeq 4.4 \cdot 10^{16} \text{ for } P/P_c = 52.3.$$ For both cases we obtain the same experimental coefficient of nonlinear radiation damping $\eta_{nl} = 2.1 \cdot 10^{-11}$ s$^{-1}$.

In order to derive theoretical estimate, we find:

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

Thus, using Eq.(5) and the following parameters for the iron borate: $c_e \simeq 4.8 \cdot 10^5$ 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: $\eta_{nl}^{(theor)} \sim 2.9 \cdot 10^{-11}$ s$^{-1}$, which of the order of magnitude is in a good agreement with the obtained experimental result.

Dots in Fig. 5 show the magnetic field dependence of experimentally obtained coefficient of nonlinear radiation damping. The solid line represents the theoretical prediction of the field dependence. We see a perfect fit within two orders of magnitude of experimental data for nonlinear radiation damping.

IV. CONCLUSION

In this work we have experimentally confirmed 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. The obtained results are in a good agreement with the
theory by the field and overthreshold dependencies and are of the order of magnitude of
the theoretical prediction. We believe that the nonlinear radiation damping is a common
feature of parallel pumping technique and it can be detected by the purposeful experiments
with other types of normal magnetic oscillations in magnetooordered systems. For example, a
specific radiation after turning off the pump of spin waves in YIG has already been observed
in Ref.\textsuperscript{17} and it was not explained in the framework of small sample approximation.

[1] H. Suhl, J. Phys. Chem. Solids \textbf{1}, 209 (1957).
[2] E. Schoemann, Phys. Rev. \textbf{116}, 827 (1959).
[3] V. E. Zakharov, V. S. L’vov, and S. S. Starobinets, Usp. Fiz. Nauk \textbf{114}, 609 (1974) [Sov.
Phys.- Usp. \textbf{17}, 896 (1975)].
[4] V. S. L’vov, Wave Turbulence under Parametric Excitation (Springer-Verlag, Berlin, 1994).
[5] A. G. Gurevich and G. A. Melkov, Magnetization Oscillations and Waves (CRC Press, Boca
Raton, 1996).
[6] V. L. Safonov, Nonequilibrium Magnons (Wiley-VCH, Weinheim, 2013).
[7] V. L. Safonov, J. Magn. Magn. Mater. \textbf{97}, L1 (1991).
[8] V. L. Safonov and H. Yamazaki, J. Magn. Magn. Mater. \textbf{161}, 275 (1996).
[9] A. V. Andrienko and V. L. Safonov, Pis’ma Zh. Eksp. Teor. Fiz. \textbf{60}, 787 (1994) [JETP Lett.
\textbf{60}, 800 (1994)].
[10] A. V. Andrienko and V. L. Safonov, Pis’ma Zh. Eksp. Teor. Fiz. \textbf{62}, 147 (1995) [JETP Lett.
\textbf{62}, 162 (1995)].
[11] A. V. Andrienko, V. L. Safonov, and H. Yamazaki, J. Phys. Soc. Jpn. \textbf{67}, 2893 (1998).
[12] P. G. de Gennes, P. A. Pincus, F. Hartmann-Boutron, and J. M. Winter, Phys. Rev. \textbf{129}, 1105
(1963).
[13] A. V. Andrienko, V. I. Ozhogin, V. L. Safonov, and A. Yu. Yakubovskii, Usp. Fiz. Nauk, \textbf{161},
1 (1991) [Sov. Phys.- Usp. \textbf{34}, 843 (1991)].
[14] A. V. Andrienko and L. V. Podd’yakov, Zh. Eksp. Teor. Fiz. \textbf{95}, 2117 (1989) [Sov. Phys.-
JETP \textbf{68}, 1224 (1989)].
[15] A. V. Andrienko, Zh. Eksp. Teor. Fiz. \textbf{101}, 1644 (1992) [Sov. Phys.- JETP \textbf{74}, 876 (1992)].
[16] V. I. Ozhogin and A. Yu. Yakubovskii, Zh. Eksp. Teor. Fiz. \textbf{67}, 287 (1974) [Sov. Phys.- JETP
40, 144 (1975)].

[17] V. S. Zhitnyuk and G. A. Melkov, Zh. Eksp. Teor. Fiz. 75, 1755 (1978) [Sov. Phys.- JETP 48, 884 (1978)].