Observation of the 'size effect' at the linear excitation of spin-waves in antiferromagnetic $MnCO_3$

1991 J. Phys.: Condens. Matter 3 9253

(http://iopscience.iop.org/0953-8984/3/46/025)

J. Phys.: Condens. Matter 3 (1991) 9253–9256. Printed in the UK

LETTER TO THE EDITOR

Observation of the 'size effect' at the linear excitation of spin-waves in antiferromagnetic $MnCO_3$

B Ya Kotyuzhanskii†, L E Svistov† and H Benner
Institut für Festkörperphysik, Technische Hochschule Darmstadt, D-6100 Darmstadt, Federal Republic of Germany

Received 16 April 1991, in final form 7 July 1991

Abstract. The antiferromagnetic resonance in thin single-crystal plates of $MnCO_3$ was studied at $\omega_p/2\pi = 9.3$ GHz and $T = 4.2$–10 K for large values of the angle θ between the static magnetic field $\boldsymbol{H}$ and the basal plane of the crystals. A set of quasi-equidistant absorption lines was observed for a wide range of magnetic fields H, which can be attributed to the size effect: the resonances of plane spin-waves with different wavevector values, $|k| \simeq 10^5$ cm^{-1}, standing perpendicularly to the crystal plate. The relaxation of the spin-waves was evaluated from the width of individual resonance lines, and found to amount to $\Delta\omega_k/2\pi \simeq 0.03$ MHz at $T = 4.2$ K.

Experiments on the parametric excitation of spin-waves with a frequency $\omega_k/2\pi \simeq 10^{10}$ Hz and a wave vector $|\boldsymbol{k}| \simeq 0$–$10^6$ cm^{-1} in antiferromagnetic materials with easy-plane anisotropy have shown that, at liquid helium temperatures, the relaxation of the spin-waves, $\Delta\omega_k$, can in some cases be rather slow: $\Delta\omega_k/2\pi \simeq 0.1$–1 MHz. Therefore, the mean free path, $l = s/\Delta\omega_k$, of the corresponding magnons, with propagation velocity $s_k = |\partial\omega_k/\partial k| \simeq 10^5$–$10^6$ cm s^{-1} for $k \simeq 10^5$ cm^{-1}, can reach the order of the sample size: $l \simeq 1$ mm [1]. Moreover, it has been found that magnons do not fully relax on the crystal boundaries, but can be reflected elastically for a high enough reflection coefficient, r (for example, $r \simeq 0.99$ in FeF_3 [2]).

The properties described above made it possible to observe the 'size effect' under parametric excitation of spin-waves on the $FeBO_3$ samples—thin ($d = 0.1$–0.5 mm) naturally faceted single-crystal plates, the larger facets of which were coincident with the basal plane of the crystal [3]. The size effect is related to the additional conditions imposed by the sample boundaries on the excited spin-waves. It is revealed in a dip of the microwave threshold field, h_c, of the parametric excitation and in a corresponding increase of the microwave power absorbed by the sample. The dips occur at those magnetic fields, $H = H_N$, where standing plane waves (with a wavevector $\boldsymbol{k}$ directed perpendicularly to the plate) are excited:

$$N\lambda = 2d \qquad k = k_z = 2\pi/\lambda \tag{1}$$

where N is integer and z is parallel to the principal axis, C_3, of the crystal. Earlier, an analogous size effect for parametric excitation of spin-waves had also been observed in ferrimagnetic YIG [4].

† Permanent address: Institute of Crystallography, Academy of Sciences of the USSR, Moscow 11733, USSR.

Under the parallel-pumping conditions with $\boldsymbol{h} \parallel \boldsymbol{H} \perp \hat{z}$ [3], spin-waves of the quasi-acoustic branch of the spectrum with frequency $\omega_k = \omega_p/2$ (where ω_p is the frequency of the microwave pumping field) are excited [5]. For the more general case of arbitrary field direction with respect to the crystal axes, this branch [6] can be described by

$$(\omega_k/\gamma)^2 = H\cos\theta(H\cos\theta + H_D) + H_\Delta^2 + \alpha_\parallel^2 k_\parallel^2 + \alpha_\perp^2 k_\perp^2. \tag{2}$$

Here γ is the gyromagnetic ratio, H_D is Dzyaloshinskii's field, H_Δ is a parameter governed mainly by the hyperfine and magnetoelastic interactions, $\boldsymbol{\alpha}$ is the stiffness tensor (the indices $\perp$ and $\parallel$ show the direction of $\boldsymbol{k}$ with respect to the principal axis) and θ is the angle between $\boldsymbol{H}$ and the basal plane.

From (1) and (2), one can obtain expressions for H_N and the interval ΔH between the neighbouring dips:

$$\Delta H = (2\pi\alpha_\parallel/d)[H_0(H_0\cos\theta + H_D) - H(H\cos\theta + H_D)]^{1/2}/(2H\cos\theta + H_D)(\cos\theta)^{1/2} \tag{3}$$

where $H_0 = H_0(\theta)$ is the field where spin-waves with $k = 0$ are excited at a given θ; its value can be obtained from (2).

From (1)–(3), it can easily be calculated that in experiments with $\omega_p/2\pi \simeq 10^{10}$ Hz and typical values of $\gamma = 2\pi \times 2.8$ MHz Oe^{-1} and $\alpha = 10^{-2}$–10^{-3} Oe cm, the maximum value of k amounts to 10^5–10^6 cm^{-1} and the corresponding number of wavelengths, N, should be more than 10^3. We wanted to find out whether one can also observe standing spin-waves with large k directly for linear excitation of a spin system by the microwave field.

It is well known that for an infinite sample in a uniform microwave field, $\boldsymbol{h}$, and a static magnetic field, $\boldsymbol{H}$, due to conservation of energy and quasi-momentum,

$$\omega_p = \omega_k \qquad k = k_p = \omega/c \simeq 0 \tag{4}$$

so it follows that linear excitation of only uniform spin oscillations is possible—i.e., in our case, excitation of only an antiferromagnetic resonance (AFMR). But the presence of any non-uniformity, in principle, avoids this restriction. In particular, the excitation of spin-waves with $k \neq 0$ is possible in a sample of a finite size; however, the effectiveness of this excitation, $\mathcal{H} = (a_k/h)^2$ (a_k is the spin-wave amplitude), drops rapidly with increase of k. Thus, for the excitation of plane spin-waves in a plate, as considered above, $\mathcal{H}$ decreases as k^{-1}.

For the observation of the size effect, we have chosen crystals of the well-investigated antiferromagnetic $MnCO_3$ (space group of symmetry D_{3d}^6, $\gamma/2\pi = 2.8$ MHz Oe^{-1}, $H_D =$ 4.4 kOe, $H_\Delta^2 = (5.8/T + 0.3)$ kOe^2, $\alpha_\parallel = 0.8 \times 10^{-2}$ Oe cm and $\Delta\omega_k/2\pi \simeq 0.1$ MHz at $T = 4.2$ K [1]). As in [3], the samples were naturally faceted plates of dimensions $\simeq 2 \times 2 \times (0.1$–$0.5)$ mm^3, the plane of which coincided with the basal plane. We used a standard X-band EPR spectrometer ($\omega_p/2\pi = 9.3$ GHz) with a rectangular cavity. The conditions of excitation were as usual for the observation of uniform AFMR—$\boldsymbol{H} \perp \boldsymbol{h} \perp C_3$ axis—but the angle θ could be varied in the experiment. The error in measuring θ was about 3°. The experiments were carried out in a helium gas flow cryostat with a minimum temperature $T = 4.2$ K. At small angles, θ, we observed the usual antiferromagnetic resonance line, which moved to higher resonance fields H_0 with increasing θ in perfect agreement with (2). At large enough values of θ (>70°), $T = 4.2$ K and at maximum sensitivity, we have observed a large number of quasi-equidistant resonance lines in fields, H, smaller than AFMR field. The intensity of these lines was only about 10^{-5} of the intensity of the AFMR line. As an example, two records representing a set of resonance lines in a wide field range are shown in figure 1(a). One can see from figure 1(a) that the

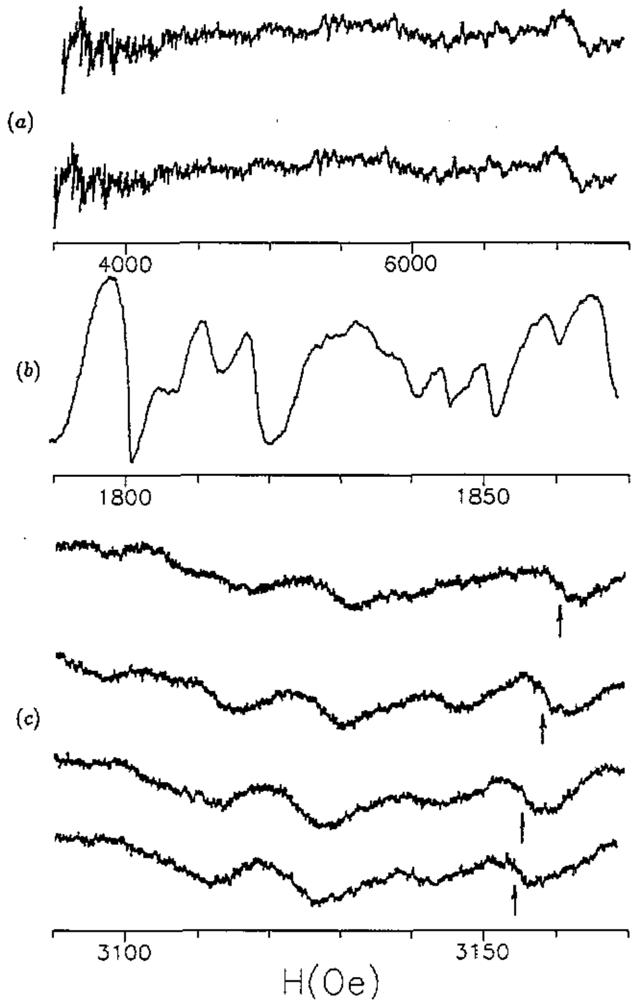

Figure 1. Records of the derivative of the microwave signal reflected from a cavity with a sample at $T = 4.2$ K, $d = 0.13$ mm: (*a*) two spectra recorded for the same conditions to prove the reproducibility, $\theta = 80°$; (*b*) extended field scale, $\theta = 70°$; (*c*) effect of slight temperature changes, $\theta = 80°$. T increases by $\simeq 0.01$ K to the upper side of the figure.

whole set of lines, or resonance spectrum, is reproducible. A small part of the resonance spectrum is shown in figure 1(*b*) for an extended field scale.

The spacing, ΔH, between neighbouring lines in a crystal with $d \simeq 0.1$ mm at $\theta = 80°$ and $H \simeq 1$ kOe was about 10 Oe. This value corresponds to the interval, ΔH, between resonance fields, H_N, of standing spin-waves differing by half a wavelength across the sample, calculated from (3) using known value of $\alpha_\parallel$ and taking the AFMR field for H_0. At constant H and θ, the measured spacing, ΔH, increased with decreasing crystal thickness d. The value of ΔH also increased with increasing angle θ in agreement with (3). The total number of half wavelengths, N, can be obtained from (1) and (2). Its value increases with decreasing H and amounts to about 1.5×10^3 at $H = 0$. We have also investigated the changes of the observed effect with temperature. These experiments can be separated into two groups. At small changes of about 0.01 K it was possible to follow the shift of some chosen absorption line (see figure 1(*c*)). With increasing T, it moved to larger H and the shift agrees to an order of magnitude with that calculated from (1) and (2), assuming that all changes in (2) only result from the temperature dependence of H^2_Δ. Obviously, the changes of H_N caused by thermal expansion of the crystal and by the temperature dependencies of the other parameters in (2) are much smaller.

For larger variations of T, it was only possible to follow a qualitative change of the spectrum. With increasing T, the separate lines broadened and decreased and the spectrum disappeared between 7 and 10 K. Such behaviour is quite natural if one takes into account that the relaxation, $\Delta\omega_k$, of spin-waves increases with temperature and the mean free path becomes smaller than the smallest sample dimension.

From the width of the individual resonance lines (figure 1(b)) one can evaluate the relaxation parameter of the excited spin-waves to be $\Delta\omega_k/2\pi = 0.03$ MHz. This value is somewhat smaller than the value obtained from the threshold field, h_c, of parametric excitation for the waves with $\omega_k = 2 \times 10^{10}$ Hz. This difference is quite natural because, in accordance with our present understanding, the relaxation of spin-waves in $MnCO_3$ at $T = 4.2$ K is determined by a three-magnon process, the effectiveness of which decreases with decreasing ω_k [1].

The question can arise as to why the observed resonance lines are not identical (see figure 1(a)). Obviously, condition (1) is strictly valid only for ideal thin crystal plates. Of course, the samples used in the experiments differ from the ideal. Since we used the naturally faceted crystals, their surfaces can be considered as parallel, on average. The finite side dimensions of a sample and growth steps on the surface which are larger than the spin-wave wavelength, λ, must cause the additional groups of resonance lines. Surface defects smaller than λ can lead to interaction between different groups of standing spin-waves and, consequently, to a change in shape and a broadening of some of the resonance lines.

The results obtained indicate that the observed phenomenon can, in fact, be attributed to the size effect. Our experiments once again confirmed results obtained earlier on crystals of $FeBO_3$ and FeF_3 that magnons with $k = 10^5$ cm^{-1} can be reflected elastically without considerable relaxation from the boundaries of a crystal. It is also essential that in similar experiments one can determine independently the relaxation parameter, $\Delta\omega_k$, and stiffness coefficient, α, from the width of individual resonance lines and from the spacing ΔH between them. The latter has already been determined in such a way on $FeBO_3$ in parallel-pumping experiments [3]. We have used known value of $\alpha_\parallel$ to prove that we observe the size effect. Obviously, we could conversely estimate $\alpha_\parallel$ from our experimental results, assuming that we have observed the size effect and substitute the spacing between resonance lines (averaged in some small field range) as ΔH into equation (3). From our point of view, this method is much simpler than performing neutron experiments.

References

[1] Kotyuzhanskii B Ya and Prozorova L A 1990 *Sov. Sci. Rev.* A **13** 1–131

[2] Kotyuzhanskii B Ya and Prozorova L A 1980 *Izv. Akad. Nauk* **44** 1409–12

[3] Kotyuzhanskii B Ya and Prozorova L A 1980 *Pis. Zh. Eksp. Teor. Fiz.* **32** 254–7 (Engl. Transl. 1980 *Sov. Phys.–JETP Lett.* **32** 235–8)

[4] Jantz W and Schneider J 1971 *Solid State Commun.* **9** 69–71

[5] Ozogin V I 1970 *Zh. Eksp. Teor. Fiz.* **58** 2079–89 (Engl. Transl. 1970 *Sov. Phys.–JETP* **31** 1121–6)

[6] Borovik-Romanov A S 1959 *Zh. Eksp. Teor. Fiz.* **36** 766–81 (Engl. Transl. 1959 *Sov. Phys.–JETP* **9** 539)