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Effect of 'hardness' at the parametric excitation of spin-waves in antiferromagnetic $MnCO_3$

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Abstract. The parametric excitation of spin-waves was studied in the easy-plane antiferromagnet $MnCO_3$ at a microwave pumping frequency $\omega_p/2\pi = 33.8$ GHz and $T = 1.6$ K. The experiments were performed for oblique orientation of the magnetic field H with respect to the basal plane of the crystal. We found that the two different microwave threshold fields for the appearance (h_{c1}) and disappearance (h_{c2}) of parametric excitation as well as the *'coefficient of hardness'* $\kappa = (h_{c1} - h_{c2})/h_{c1}$ strongly depend on the angle ϑ between H and the basal plane, showing a decrease with increasing ϑ. Our experimental results can be explained by the assumption that some part of the spin-wave relaxation and hardness effect arise from an interaction of spin-waves with 'orthogonal' magnetic impurities.

1. Introduction

Parametric excitation of spin-waves in antiferromagnetic materials has already been studied for more than 20 years [1–3]. These investigations were mainly performed on antiferromagnetic crystals with easy-plane anisotropy containing 3d ions. the latter basically include crystals with the high-order symmetry axes C_3 or C_6, e.g. $MnCO_3$ (space symmetry group D^6_{3d}), $CsMnF_3$ (D^4_{6h}) and others. Theoretical consideration of this effect revealed that it is based — like the previously known parallel pumping in ferro- and ferrimagnetic materials [4, 5] — on the parametric instability of spin-waves with a frequency ω_k equal to half the pumping frequency ω_p, which occurs as soon as the amplitude of the pumping microwave field h exceeds a certain threshold value h_c. But the nature of this effect in ferro- and antiferromagnetic materials is different. In ferro- and ferrimagnetic materials the parametric instability is based on dipolar interactions. In antiferromagnetic materials dipolar interaction remains somewhat unimportant and, instead, an exchange enhanced non-linear interaction between spin oscillations, corresponding to different branches of the spin-wave spectrum, is playing the dominating role [6].

In the simplest easy-plane two-sublattice antiferromagnets — including the material $MnCO_3$ which was investigated in this work — the spin-wave spectrum consists of two branches, a quasi-acoustic and a quasi-optic one [7]. In the case of arbitrary orientation of the sample with respect to the magnetic field H the corresponding dispersion

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relations are given by

$$(\omega_{1,\boldsymbol{k}}/\gamma)^2 = H\cos\vartheta(H\cos\vartheta + H_{\mathrm{D}}) + H_{\Delta}^2 + \alpha^2 k^2 \tag{1}$$

$$(\omega_{2,\boldsymbol{k}}/\gamma)^2 = 2H_{\mathrm{A}}H_{\mathrm{E}} + H_{\mathrm{D}}(H\cos\vartheta + H_{\mathrm{D}}) + H^2\sin^2\vartheta + \alpha^2 k^2. \tag{2}$$

Here, H_{E} and H_{A} are the exchange and anisotropy fields, H_{D} is the Dzyaloshinskii field, α^2 denotes the 'stiffness constant' (which is proportional to the exchange field) and H_{Δ} represents a small gap in the spectrum, usually determined by hyperfine and/or magnetoelastic interactions, ϑ is the angle between the static magnetic field and the easy basal plane of the crystal, and γ is the gyromagnetic ratio. For $\mathrm{MnCO_3}$ these parameters are given by $H_{\mathrm{A}} = 3$ kOe, $H_{\mathrm{E}} = 320$ kOe, $H_{\mathrm{D}} = 4.4$ kOe, $\alpha_{\parallel} = 0.79 \times 10^{-2}$ Oe cm ($\alpha_{\parallel} \equiv \alpha_z$, where $\hat{z} \parallel C_3$), $H_{\Delta}^2 = (5.8/T + 0.3)$ kOe^2 and $\gamma = 2\pi \times 2.80 \times 10^6$ s^{-1} Oe^{-1}. Since the in-plane anisotropy of $\mathrm{MnCO_3}$ is small, the corresponding terms have been neglected in (1).

In the optimum case for parametric excitation, $h \parallel H \perp C_3$, spin-waves of the quasi-acoustic branch are excited and the threshold field of this process is defined by the expression [6]:

$$\Delta\omega_{\boldsymbol{k}} = V_{\boldsymbol{k}} h_{\mathrm{c}} = \frac{\gamma^2(2H + H_{\mathrm{D}})}{\omega_{\mathrm{p}}} h_{\mathrm{c}}. \tag{3}$$

Here, $\Delta\omega_{\boldsymbol{k}}$ is the relaxation parameter of the excited pair of spin-waves with wavevectors $\boldsymbol{k}$ and $-\boldsymbol{k}$, the value of which is determined by equation (1) from the condition $\omega_{1,\boldsymbol{k}} = \omega_{1,-\boldsymbol{k}} = \omega_{\mathrm{p}}/2$. The lifetime of corresponding magnons is given by $\Delta\omega_{\boldsymbol{k}}^{-1}$. $V_{\boldsymbol{k}}$ is the so-called bond parameter describing the coupling between microwave pumping field h and excited spin-waves. (Henceforth, we will only consider the acoustic branch and omit the index 1.) By means of relation (3) the relaxation parameter $\Delta\omega_{\boldsymbol{k}}$ can be directly obtained from the experimental threshold field h_{c}. Experiments performed within a wide range of external conditions ($\omega_{\mathrm{p}}, T, H$) on different crystals and with different magnetic ions have allowed the determination of the main interactions responsible for spin-wave relaxation (see e.g. the review [8]).

One of the characteristic features of parametric excitation, which are not yet fully understood, is the so-called 'hard excitation' or 'hardness effect'. This effect is revealed as a hysteresis of parametric excitation with respect to the microwave amplitude: the process only starts at a threshold field h_{c1}, but breaks off at the lower field h_{c2}. Such a hysteresis was first discovered by Joseph *et al* [9] in a parallel pumping experiment on ferrimagnetic YIG and then, simultaneously, by Ozhogin and Yakubovskii [10] and by Kveder *et al* [11] on antiferromagnetic $\mathrm{MnCO_3}$. Later on the hardness effect was observed on all investigated crystals and, thus, represents a common feature which is characteristic for parametric excitation of spin-waves.

In the literature different assumptions are discussed about the possible nature of the hardness effect, but the corresponding theoretical estimates of the 'coefficient of hardness' $\kappa = (h_{\mathrm{c1}} - h_{\mathrm{c2}})/h_{\mathrm{c1}}$ disagree with experiment. The only point, in which all considerations are consistent, is that the hardness effect is connected with the sudden switching off of some part of the spin-wave relaxation, which occurs at increasing spin-wave amplitude $a_{\boldsymbol{k}}$. Usually one supposes that this non-linear relaxation occurs due to some saturation effect decreasing the rate $\Delta\omega_{\boldsymbol{k}}$ by some amount $\delta\omega_{\boldsymbol{k}}(a_{\boldsymbol{k}})$ which depends on the amplitude $a_{\boldsymbol{k}}$ of the spin-waves. That means, $\delta\omega_{\boldsymbol{k}}(a_{\boldsymbol{k}})$ represents the negative non-linear relaxation. If this assumption is correct, then the question about the nature of the hardness effect reduces to the understanding of the nature of negative non-linear relaxation.

The main features of the hardness effect discovered on easy-plane antiferromagnets ($MnCO_3$, $CsMnF_3$, $CoCO_3$, $FeBO_3$ and $CsMnCl_3$) can be summarized as follows:

(i) The hardness coefficient κ grows rapidly with decreasing temperature reaching values of $\kappa = 0.4$–0.7 in $MnCO_3$ and $CsMnF_3$ and $\kappa \approx 10$ in $CoCO_3$ and $FeBO_3$ at $T = 1.2$ K.

(ii) κ decreases with increasing field H.

(iii) In at least three of the investigated crystals the field and temperature dependences of $\Delta\omega_k$, determined from the threshold field h_{c2} by means of equation (3), is in accordance with theoretical predictions for intrinsic (i.e. occurring in the ideal crystals) relaxation processes: the three-magnon coalescence process in $MnCO_3$ and $CsMnF_3$ [11, 12], and a three-particle magnon–phonon process in $FeBO_3$ [13].

In a work [14] especially devoted to the investigation of the hardness effect in $FeBO_3$, the influence of one group of parametrically excited spin-waves on the hardness of parametric excitation of another group of waves with the same or considerable different frequencies has been studied. The results of this investigation allowed the conclusion that at least in this crystal hardness is connected with the saturation of some non-intrinsic relaxation process (e.g. caused by defects in the crystal). The resemblance between manifestations of hardness phenomena in various antiferromagnetic crystals suggests that the nature of this effect is the same in other materials, too.

The present investigation aims at the nature of hard excitation of spin-waves in easy-plane antiferromagnets. As an appropriate system for this investigation we have chosen $MnCO_3$, the magnetic properties of which are well known. Its crystallographic and magnetic parameters are listed in [8].

2. Experimental set-up and samples

Our investigations were performed under CW conditions with a Q-band EPR spectrometer ($\omega_p/2\pi = 33.8$ GHz). The sample was fixed on the bottom of a cylindrical high-Q microwave cavity at the position of maximum microwave field $\boldsymbol{h}$, and a static magnetic field up to 18 kOe was applied. The geometry was chosen in such a way that $\boldsymbol{h}$ was applied within the easy plane of the crystal ($\boldsymbol{h} \perp C_3$) and $\boldsymbol{H}$ could be rotated in the $(\boldsymbol{h}, C_3)$-plane. The frequency of the static field modulation was chosen to be low enough (30 Hz) to prevent any influence of this modulation on the experimental results. The microwave power applied to the cavity was varied in steps of 1 dB. The absolute value of h at the sample position was not measured. The uncertainty of the change of h was about 10%.

When sweeping the static field the parametric excitation of spin-waves is manifested by the onset of absorption at some field H_c. Since both thresholds h_{c1} and h_{c2} of this process depend on H, the hardness effect and the corresponding hysteresis in microwave power give rise to a hysteresis with respect to the static field: on varying H absorption appears at H_{c1} and disappears at a different value H_{c2} (see figure 1). The hardness effect was discovered exactly in this way for the first time [10]. By means of the relation

$$h_{ci}(H_{ci}) = h \qquad i = 1, 2 \tag{4}$$

the experimental data of $H_{c1}(h)$ and $H_{c2}(h)$ allow the field dependences $h_{ci}(H)$ to be evaluated.

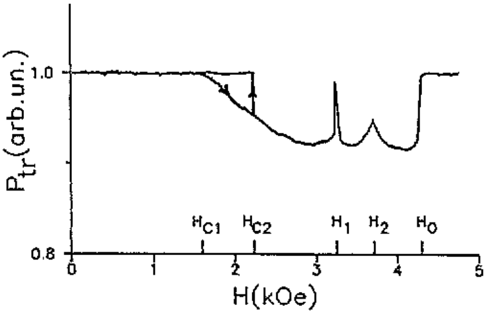

Figure 1. Field dependence of the transmitted microwave signal above threshold ($H \perp C_3$). Bistability between H_{c1} and H_{c2} is related to the hardness effect. H_0 denotes the upper limit of parametric spin-wave excitation i.e. for larger fields $\omega_p/2$ remains under the lowest spin-wave frequency $\omega_{1,k=0}$. The meaning of H_1 and H_2 is explained in the text.

The $MnCO_3$ single crystals investigated were obtained from two different growth experiments (denoted later by I and II). The crystals had the shape of naturally faceted plates, from which we broke off pieces with a linear dimension of about 0.3 mm. Crystals from group II had a deeper pink colour and evidently (see section 3) a larger impurity concentration.

3. Results

On variation of the field direction relative to the easy plane and at a fixed pumping frequency ω_p, spin-waves with same frequency ω_k and wavenumber k are excited at a field H_k the angular dependence of which — in accordance with (1) — is determined by the condition $H_k \cdot \cos\vartheta = \text{constant}$. In figure 2 we have shown the angular dependence of H_0, which corresponds to the upper limit of the field range where parametric excitation is possible ($k = 0$). The same angular dependence was observed for another characteristic field H_2, which is defined by the position of a narrow peak in the above-threshold susceptibility (see figure 1) originating from the intersection of the dispersion curves of spin-waves and transverse sound-waves ($k = \text{constant} \approx 2\times10^5$ cm s^{-1}) [15]. Thus, variation of angle ϑ seems to result only in a general rescaling of the external field confirming the above condition.

The field dependences of the threshold fields h_{c1} and H_{c2} obtained for different ϑ on a crystal from group I are shown in figure 3. For convenience of comparison the horizontal axis was chosen in such a way that the presented threshold fields correspond to spin-waves with equal k. Both threshold fields and the hardness coefficient decrease with increasing H — and the larger angle ϑ is, the faster this decrease will be. For large enough ϑ ($> 30°$)h_{c1} and h_{c2} asymptotically approach the same value (about 0.15 arbitrary units). The slight increase of both threshold fields observed for $\vartheta = 80°$ and $H_\perp \geq 2$ kOe belongs to a peak occurring only for large angles ϑ at higher field. The nature of this peak is not yet known.

Analogous dependencies have also been measured for crystals from group II. The threshold fields and, consequently, the relaxation rates of spin-waves in these crystals

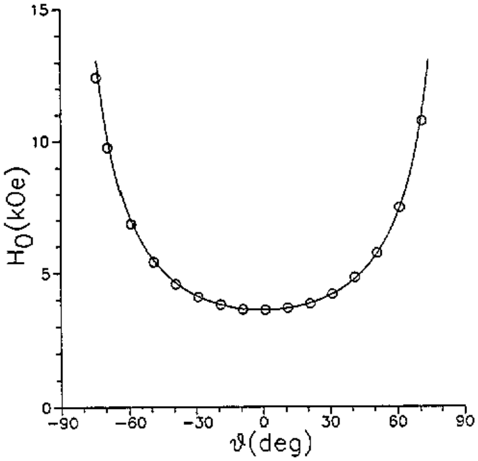

Figure 2. Experimental dependence of H_0 on angle ϑ. The curve shows the theoretical dependence calculated from equation (1) with $\omega_{1,k} = \omega_p/2$ and $k = 0$.

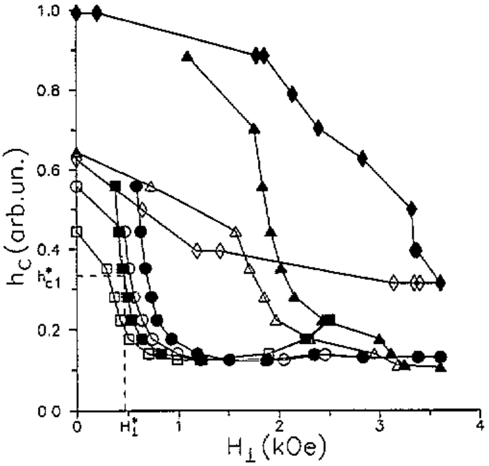

Figure 3. Microwave threshold fields h_{c1} (full symbols) and h_{c2} (open symbols) as a function of the in-plane component of the external magnetic field $\boldsymbol{H}$ for different orientations $\vartheta = 0°$ (◊), 50° (Δ), 75° (○), 90° (□). Full curves are only guides for the eye. According to equation (1), the horizontal axis corresponds to the same values of k. As an example $H^*_\perp = H^* \cos\vartheta$ is shown for $\vartheta = 80°$.

are more than twice larger than in group I and their decrease with increasing angle ϑ is considerably less. To avoid a possible error when comparing the threshold fields, both crystals were simultaneously mounted in the cavity. It is worth noting that crystals from group II showed a several times smaller above-threshold susceptibility.

For directly recording the decrease in h_{c1} with increasing ϑ in crystals from group II we have presented in figure 4 the ϑ-dependence of the ratio H_0/H_{c1} measured at constant h and increasing H. If for spin-waves with a given wavenumber h_{c1} does not depend on ϑ, this ratio should be constant. In figure 4, however, we observe a monotonic increase in this ratio with ϑ. For comparison, in the same figure we have shown the ϑ-dependence of H_0/H_2, and this ratio is really constant with an accuracy of about 5%. The increase of H_0/H_{c1} indicates that H_{c1} moves with increasing angle to a smaller field than expected. That means, due to the negative sign of $\partial h_{c1}/\partial H$ at these fields, h_{c1} also decreases with increasing ϑ in this crystal.

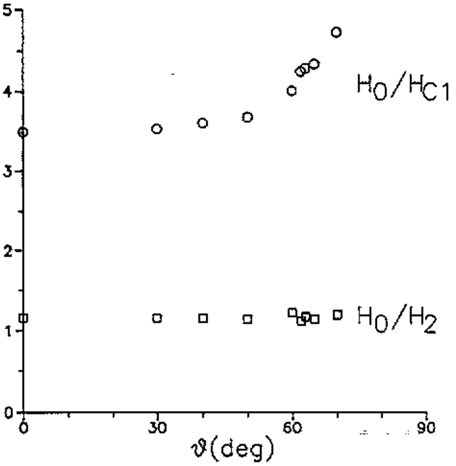

Figure 4. Dependences of ratios H_0/H_{c1} and H_0/H_2 on angle ϑ, measured on a crystal from group II.

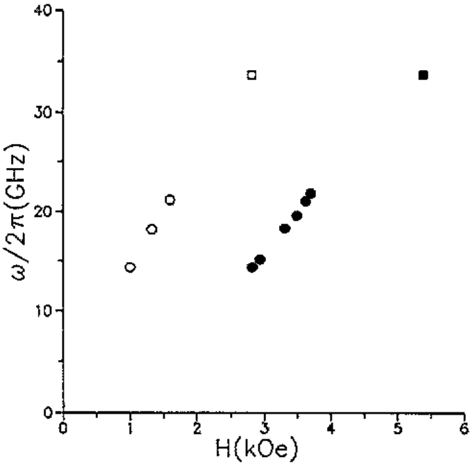

Figure 5. Spectrum of impurity excitation. Circles were obtained from peak in the above-threshold susceptibility at H_1 in [15], rectangles from EPR resonance field. Full symbols, $\boldsymbol{H} \perp C_3$; open symbols, $\boldsymbol{H} \parallel C_3$.

4. Discussion

The decrease in the threshold fields h_{c1} and h_{c2} with increasing angle ϑ, observed on crystals from group I, strictly speaking, does not mean a corresponding decrease of the spin-wave relaxation. The point is that the calculations of coupling coefficient V_K in (3) only refer to the special case $\vartheta = 0$, where in linear approximation the components of the vectors $\boldsymbol{l} = \boldsymbol{S}_1 - \boldsymbol{S}_2$ and $\boldsymbol{m} = \boldsymbol{S}_1 + \boldsymbol{S}_2$ ($\boldsymbol{S}_{1,2}$ denoting the sublattice magnetizations), described by $\boldsymbol{l} = \boldsymbol{l}_0 - \boldsymbol{\lambda} \cdot \exp(\mathrm{i}\omega t - \mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r})$ and $\boldsymbol{m} = \boldsymbol{m}_0 - \boldsymbol{\mu} \cdot \exp(\mathrm{i}\omega t - \mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r})$, respectively, can be separated into two uncoupled groups, one of which $(\lambda_x, \mu_y, \mu_z)$ oscillates with the frequency $\omega_{1,\boldsymbol{k}}$ and the second one $(\mu_x, \lambda_y, \lambda_z)$ with the frequency $\omega_{2,\boldsymbol{k}}$. For $\vartheta \neq 0$ the oscillations of all six components become coupled and the calculation of $V_{\boldsymbol{k}}$ is strongly complicated. From common considerations, however, it is hard to expect that $V_{\boldsymbol{k}}$ will increase with increasing ϑ.

As has already been mentioned in the introduction, there are different assumptions about the nature of the hardness effect. Already in one of its first studies [11] performed on $MnCO_3$ crystals, it was supposed that hardness is related to the saturation of a relaxation mechanism due to magnetic impurities. This assumption was based on the occurrence of a resonance-type peculiarity showing up in the field dependences of both the threshold field $h_{c1}(H)$ and of the above-threshold susceptibility $\chi(h)$ [15]. At a certain field H_1, where this peculiarity occurs (see figure 1), the threshold $h_{c1}(H)$ has a local maximum and $\chi(H)$, consequently, has a minimum. The relation between the spin-wave frequency ω_k and the field H_1 is shown in figure 5. The typical shape of $\omega_k(H_1)$ and the independence of H_1 on wavenumber k allowed the authors of [15] to suggest that there occurs a level-crossing at H_1 between spin-waves and elementary excitations of magnetic impurities coupled by exchange with the Mn^{2+} host ions. It is clear that the interaction between spin-waves and impurity excitations is strongly enhanced at the intersection point, but it can also be observed far away from this point.

Later on, the assumption of the impurity nature of hardness was supported by the experimental result of Andrienko [16] that in presence of parametric excitation of nuclear spin-waves the hardness coefficient in $CsMnF_3$ increases with impurity concentration, and also by special investigations of the nature of hardness in $FeBO_3$ crystals [14]. The latter results have clearly shown that at least in $FeBO_3$ the hardness of the parametric excitation is determined by a non-intrinsic relaxation process of the excited spin-waves.

For clearing up the nature of the hardness effect in $MnCO_3$ we deemed it important to probe the amount of impurities in the crystals investigated. We carried out an EPR study of our crystals with the aim of recording directly the microwave absorption spectrum corresponding to the supposed magnetic impurity excitations. We observed a few weak absorption lines. The position of one of them corresponds to the dependence $\omega_k(H_1)$ (see figures 5 and 6). We take this as an additional proof that the peak in $\chi(H)$ at H_1 is connected with the presence of some defects, which to all appearance are magnetic impurities. We therefore assume that the hardness effect in $MnCO_3$ is connected with the presence of impurities and will try to prove this hypothesis by our experimental data.

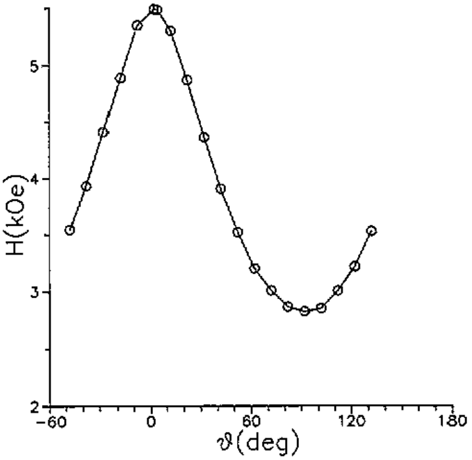

Figure 6. Angular dependence of EPR resonance field of the impurity line.

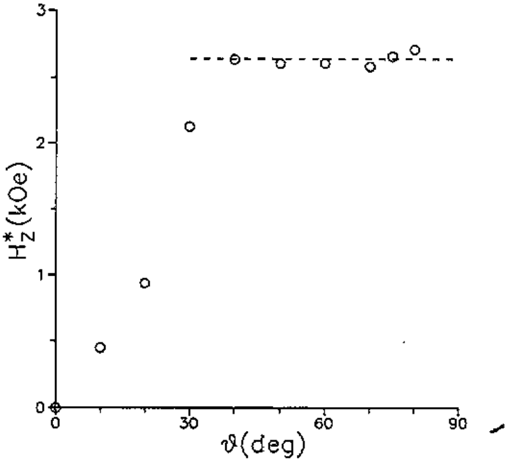

Figure 7. Angular dependence of $H_z^* = H^* \sin\vartheta$ as defined in figure 3.

Ivanov *et al* have shown [17] that the so-called 'orthogonal magnetic impurities', i.e. impurities with a large single-axis anisotropy with a sign opposite to that of the host ions, have an anomalously low excitation frequency, even when they are coupled to the host ions by strong exchange interactions. Such impurity spins are oriented nearly perpendicularly to the spins of the undisturbed host matrix. In the case of

$MnCO_3$ such a role could be played, for example, by Fe^{2+} impurities. Unfortunately, the excitation spectrum of Fe^{2+} in $MnCO_3$ was not calculated, but the calculated spectrum of Fe^{2+} in the isomorphous crystal $CoCO_3$ [17] is in good accordance with experimental results [18].

The magnon relaxation due to magnetic impurities in antiferromagnetic materials was theoretically investigated by Michailov and Farzetdinova [19]. Expressions were obtained for the negative non-linear relaxation resulting from a one-magnon relaxation process by the impurity ions and from the so-called 'slow relaxation process', showing that negative non-linear relaxation can arise from these mechanisms. Unfortunately, numerical estimates were only obtained for the latter process. However, slow relaxation occurs when the excitation frequency of the impurities is much higher than the spin-wave frequency and, within the frame of our model, this condition is not satisfied in $MnCO_3$. Moreover, the temperature dependence of the non-linear relaxation rate calculated in [19] for the slow-relaxation process does not correspond to the experimental data [11, 12, 20]. Cherepanov [21] has considered the interaction of parametrically excited spin-waves with defects, the excitation frequency of which is close to that of the spin-waves. He found that this interaction also leads to a negative non-linear damping. Unfortunately, it is impossible to compare his theoretical result with experimental data, since apart from the impurity concentration some other parameters of unknown magnitude also enter the respective expression.

Since we are not aware of other theoretical studies connecting the hardness effect with magnetic impurities, we will present some qualitative arguments for the decrease in hardness with increasing ϑ.

The first, trivial possible reason can be connected with the shape of the impurity spectrum $\omega_{\mathrm{imp}}(H)$. ω_{imp} grows with increasing magnetic field the stronger, the larger is ϑ. Due to this, the frequency distance between ω_{k} and ω_{imp} for fields $H < H_1$ is also growing, and the interaction of the spin-waves with this impurity mode decreases. Then, the linear and non-linear parts of the spin-wave relaxation rate, corresponding to this interaction, must also decrease with increasing ϑ.

The second possible reason, which seems more likely to us, is connected with the orthogonality of the magnetic impurities. An impurity spin directed nearly perpendicularly to the easy plane has a strong exchange interaction with the neighbouring spins of the host matrix. As a consequence, these spins are disturbed in some volume around the impurity and are tilted out of the easy plane. The existence of such disturbances should lead to a considerable enhancement of the interaction between spin-waves and impurity centre.

The latter explanation is supported by a Mössbauer study of the magnetic phase c–T diagram of $MnCO_3$:Fe^{2+} by Wiltshire and Price [22]. Measuring the direction of the hyperfine field at the ^{57}Fe nuclei they observed that at temperatures below T_N and Fe^{2+} concentrations $c < 0.009$ there exist two distinct magnetic phases, a collinear one at higher T and smaller c with the Fe^{2+} spins nearly parallel to the easy plane ($\vartheta_{\mathrm{hf}} \approx 0°$) and a mixed one at smaller T and higher c with $\vartheta_{\mathrm{hf}} \approx 60°$. For small concentrations up to $c = 0.009$ the transition temperature T_c between these phases is proportional to c. This means that the temperatures (4–6 K) where the hardness effect disappears [11] correspond to the transition temperature $T_c(c)$ of an Fe^{2+} impurity concentration $c \approx 10^{-3}$. This estimate of concentration seems to be realistic for the crystals investigated [15]. In this case hardness exists only in the mixed phase. The strong dependence of the transition temperature T_c on the impurity concentration c indicates a considerable interaction between impurity spins even at low concentration $c \leq 10^{-3}$. Since the interaction between impurities is effected by long-wave magnons [17], this result yields an additional indication that the interaction between spin-waves and impurity centres is large enough to give rise to an extra part of relaxation as proposed in our hypothesis.

In absence of a field component H_z perpendicular to the easy plane there are two equivalent directions for the 'orthogonal impurity' spins in the crystal, differing by the sign of S_z. In this case the impurity spins distribute among both directions, and the directions of the matrix spins show wavy distortions. The occurrence of a non-zero field component H_z for $\vartheta \neq 0$ lifts the degeneracy of both directions. At a sufficiently high value of H_z the impurity spins are oriented preferably in one direction, and the amplitude of the wavy distortions diminishes. Clearly these distortions give rise to an additional spin-wave relaxation, which decreases with increasing H_z.

Within the frame of this hypothesis, the typical value of H_z leading to impurity ordering is determined by the energy barrier to be exceeded for reversing an impurity spin. Then, a qualitative change of the relaxation rate should occur at some nearly constant value $H_z = H \cdot \sin\vartheta$. Figure 3 shows that for large ϑ there occurs a drastic change in the slope of $h_{c1}(H)$ and $h_{c2}(H)$ at some field H^* which depends on ϑ. To characterize this field we have chosen some constant value h_{c1}^* markedly above the asymptotic value of $h_{c1} \approx 0.15$ arbitrary units and define H^* by $h_{c1}(H^*) = h_{c1}^*$. Because of the strong slope of $h_{c1}(H)$ the value of H^* is nearly independent of the special choice of h_{c1}^*. The magnetic field components $H_z^* = H^* \cdot \sin\vartheta$ are shown in figure 7. It is evident that $H_z^*(\vartheta)$ becomes constant at large ϑ, in accordance with our hypothesis. Note that there is no reason to expect H_z^* to be constant at small ϑ, since the increase of H^* for decreasing ϑ is limited by H_0. We should also mention that on some crystals we have observed a considerable asymmetry of this curve and, in particular, a peak at $\vartheta \approx 25°$ which can be attributed to a preferred orientation of the impurity spins, resulting, for example, from elastic strain when cooling the crystal in a glued state. Thus, we believe that all results obtained are consistent with our assumption that the nature of the hardness effect is related to the relaxation of spin-waves caused by orthogonal magnetic impurities.

5. Conclusions

By studying the parametric excitation of spin-waves in $MnCO_3$ under oblique pumping conditions we found that the threshold fields h_{c1} and h_{c2} and the coefficient of hardness k depend strongly on orientation. For spin-waves with the same wavenumber k these properties decrease with increasing angle ϑ. Simultaneously, on the same crystal we observed an EPR absorption line, the position of which fits to the spectrum of a certain excitation registered earlier via its influence on spin-wave excitation thresholds and attributed to magnetic impurities. Moreover, the temperatures (4–6 K) where the hardness disappears correspond to the transition temperature T_c of a reorientational phase transition observed in the impurity-doped system $MnCO_3$:Fe^{2+} with an impurity concentration of 10^{-3}. All these results support the conclusion that some part of the spin-wave relaxation at low temperature as well as the hardness effect are caused by 'orthogonal' magnetic impurities always present in available crystals. Since the hardness of spin-wave excitation in $FeBO_3$ and of nuclear spin-wave excitation in $MnCO_3$ are also related to defects of the crystal, we conclude that the origin of the hardness effect common to all investigated materials is the presence of defects and, in particular, of magnetic impurities. Therefore, future investigation of parametric spin-wave excitation should be performed on crystals with different and well controlled impurity dopings in order to obtain quantitative data on the interaction between spin-waves and impurities.

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