Longitudinal pumping of spin waves in ferromagnetic chromium chalcogenide spinels

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Spin waves are excited parametrically by parallel pumping in the ferromagnets $CdCr_2Se_4$ and $CdCr_2S_4$. Study of the spin-wave damping in the crystals, which differ considerably from the yttrium iron garnet, shows that divalent and tetravalent chromium ions play a significant role. The contribution of the intrinsic spin-wave relaxation is so appreciable that it can be easily measured even at liquid-helium temperatures. The value of the contribution and its dependence on the wave number agree well with contemporary theories. In some cases, "anomalies" in the threshold curves are observed and can apparently be attributed to excitation of spin waves with $\theta_k \neq \pi/2$.

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1. INTRODUCTION

Spin-wave damping in magnetically ordered substances has been investigated theoretically and experimentally for more than 20 years. The most thoroughly studied theoretically (see, e.g., Refs. 1–5) where the relaxation processes inherent in an ideal crystal and determined by the fundamental interactions within the magnetic system and between this system and the lattice. Also investigated were different extrinsic properties, which are determined by scattering of spin waves from defects of the crystal[6] and by the interaction of the magnetic system with certain centers such as impurity ions with strong spin-orbit coupling (see, e.g., Refs. 7 and 8).

In experiment, most attention was paid to damping of homogeneous magnetization oscillations (to the limiting case of spin waves with wave number $\mathbf{k} = 0$), i.e., to the linewidth of homogeneous ferromagnetic or antiferromagnetic resonance. Measurement of the damping of spin waves with $\mathbf{k} \neq 0$ entails much greater difficulties. The most widely used method at present is based on observing the threshold of parametric excitation of spin waves. It is customary to use here longitudinal pumping (by an alternating field parallel to the constant field $\mathbf{H}_0$, Ref. 8), for in this case it is possible to produce excitation (and consequently determine the damping) of spin waves with values of k adjustable by varying H_0 in a sufficiently wide range—approximately 10^4–10^6 cm^{-1}.

Measurements of the thresholds of parametric excitation of spin waves with longitudinal pumping (see, e.g., Refs. 9–13) were aimed at determining the spin-wave relaxation parameter ΔH_k and comparing its value as a function of temperature and of wave number with the theoretical one, as well as at verifying the premises and conclusions of the theory of parametric excitation of spin waves, which was developed in greatest detail precisely for the case of longitudinal pumping. In most such studies single crystals of ferrimagnets were used, principally yttrium-iron garnets (YIG)[12]; attention was paid also to antiferromagnets.[13–15] At the same time, ferromagnets were considered in most theoretical studies devoted both to the damping mechanisms (relaxation processes) of spin waves[1–7] and to their parametric excitation.[9,16]

An experimental investigation of the damping of spin waves and of high-grade YIG single crystals by longitudinal pumping[12] has shown, in particular, that the intrinsic relaxation processes, namely three-magnon dipole coalescence[1–3] and four-magnon exchange scattering[4,5] make at sufficiently high temperatures (above ~ 100 K) substantial contributions to ΔH_k and determine for the most part the dependence of ΔH_k on k at $k \gtrsim 10^5$ cm^{-1}. The theories[3,5] describe quantitatively well these contributions, despite the fact that they were developed for ferromagnets. A similar situation obtains also with the main premises of the theory of parametric excitation of spin waves; while developed with a ferromagnetic model, they were compared with the results of numerous experiments for ferrimagnets, mainly YIG.

It follows therefore that there is undoubted interest in comparing the premises of the theories of parametric excitation and of the theories of spin-wave relaxation processes, primarily of the intrinsic processes, with experiments on ferromagnets, especially with parameters that differ from YIG. Suitable objects for this are single crystals of chromium chalcogenide spinels $CdCr_2Se_4$ and $CdCr_2S_4$. These substances are ferromagnets with Curie temperatures 130 K for $CdCr_2Se_4$ and 86 K for $CdCr_2S_4$ with magnetic moment $\sim 6\mu_B$ (μ_B is the Bohr magneton) per formula unit, corresponding to spontaneous magnetizations M_0 that are approximately twice as large as for YIG (at liquid helium temperature, $4\pi M_0 = 4450$ G for $CdCr_2Se_4$ and 5130 G for $CdCr_2S_4$. Most importantly, the inhomogeneous exchange constants D in the chromium chalcogenide spinels have (see below) values that are smaller by more than one order of magnitude than YIG. This should lead to a considerable increase of the contribution of the intrinsic processes to the spin-wave damping.

In the investigation of the damping of spin waves in iron garnets, certain attention[9,12,17,18] was paid to the influence of rapidly relaxing impurity ions with strong spin-orbit cou-

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pling, or to the so-called ionic relaxation process, which was investigated earlier in detail (see, e.g., Ref. 19) for homogeneous magnetization oscillations. In the case of garnets such ions are rare-earth ions and also ions of divalent and tetravalent iron. Calcogenide spinels constitute a very convenient object for the investigation of such processes, since the rapidly relaxing ions Cr^{2+} and Cr^{4+} are produced in them[20] in controllable amounts in the course of the growth by suitably doping the crystals, and also in the course of the heat treatment as a result of the appearance of vacancies. The influence of these ions manifests itself quite strongly because the basic magnetic system is made up in these systems of Cr^{3+} ions with quenched orbital momenta. This influence was investigated in detail earlier for the case of homogeneous oscillations of the magnetization (see, e.g., Ref. 20).

2. MEASUREMENT PROCEDURE

The spin waves were parametrically excited by pumping with a longitudinal alternating field of frequency 9.4 GHz. The threshold field in such pumping, for an isotropic ferromagnet or when the constant field is oriented along the axis ⟨100⟩ or ⟨111⟩ of a cubic crystal, is determined by the expression[9]

$$h_c=\min_{\theta_k}\left[\frac{\omega_p}{\omega_M}\frac{2\Delta H_k}{\sin^2\theta_k}\right]. \quad (1)$$

Here ω_p is the pump frequency, $\omega_M = 4\pi\gamma M_0$ (γ is the magneto-mechanical ratio), θ_k is the angle between the direction of propagation of the excited spin waves and the constant magnetization $\mathbf{M}_0$ (which has in this case the same direction as $\mathbf{H}_0$). The value ΔH_k can depend on the spin-wave frequency $\omega_k = \omega_p/2$, on the values of H_0, M_0, and k, and also on the polar angle θ_k and generally speaking on the azimuthal angle φ_k of the vector $\mathbf{k}$. If ΔH_k has a weak (or no) dependence on θ_k, a threshold is observed for spin waves with $\theta_k = \pi/2$, and their relaxation parameter can be determined from the value of the threshold field using (1). In our case this took place at sufficiently large values of k and not always (see below) at small k.

The value of k is defined by the expression

$$Dk^2=H_{ci}(\theta_k, \varphi_k)-H_{0i}, \quad (2)$$

which is a consequence of the dispersion relation for spin waves in a ferromagnet. Here H_{0i} is the internal constant magnetic field, H_{ci} is its value at which $k = 0$ for the given θ_k and φ_k. If the constant field is oriented along the axes ⟨100⟩ and ⟨111⟩, there is no dependence of H_{ci} on φ_k and

$$H_{ci}(\theta_k)=[(\omega_p/2\gamma)^2+(2\pi M_0\sin^2\theta_k)^2]^{1/2} -2\pi M_0\sin^2\theta_k-H_A, \quad (3)$$

where $H_A = 2K_1/M_0$ for the orientation ⟨100⟩ with account taken of only the first anisotropy constant K_1, and $H_A = -4/3K_1/M_0$ for the orientation ⟨111⟩. Thus, if the angle θ_k is known (in particular, if $\theta_k = \pi/2$), then k is uniquely determined by the value of the external constant magnetic field.

To decrease the necessary generator power and to increase the sensitivity of the indication of the threshold, we

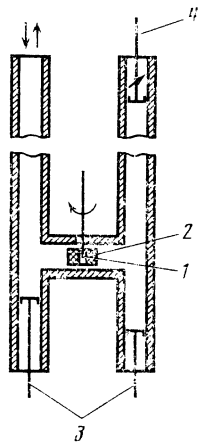

FIG. 1. Diagram of measuring section: 1—sample; 2—dielectric resonator; 3, 4—pistons.

used an dielectric resonator[21] made of the thermally stabilized TBNB[1)] with $\epsilon\approx 80$. The resonator was rectangular with dimensions $3.10\times 3.22\times 3.85$ mm and had a cylindrical bore of diameter ≈ 0.75 mm for the sample. Its intrinsic Q was ~ 1500. The resonator was located in the waveguide measurement section shown schematically in Fig. 1. The pistons 3 served for preliminary (at room temperature) matching of the section—the maximum standing-wave ratio was obtained at its input in the absence of a dielectric resonator and with the piston 4 replaced by a matched load. The piston 4, situated outside the cryostat, was used to regulate the coupling of the dielectric resonator to the waveguide.

The measurement section was placed in a helium cryostat. The samples were polished spheres of ≈ 0.5 mm diameter and were oriented with an x-ray diffractometer and glued to holders in the form of short quartz tubes secured in special bushings. These bushings were secured at a fixed angle to the shaft of an angle-reading dial. Such an arrangement made it possible not only to superimpose plane perpendicular to the sample rotation axis (in which the external magnetic field was located) on a specified crystallographic plane (in our case ⟨110⟩), but also to fix the directions of the crystallographic axis in this plane. The accuracy of this fixing, as well as the accuracy of superimposing the ⟨110⟩ plane on the plane perpendicular to the sample rotation axis, was not worse than 1°.

The use of a dielectric resonator made it possible to measure values of $2\Delta H_k$ up to 20 Oe, by using a low-power (~ 10 W) tunable magnetron. It was modulated by long (~ 100 μsec) pulses, making it possible to fix the threshold with sufficient accuracy. The threshold was determined from the change of the dependence of the power reflected by the resonator on the incident power when the latter was varied with a precision polarization attenuator. Recognizing that the damping of the attenuator in the measurements was not less than 10 dB, the off-duty cycle was ~ 200, and we fixed only the moment of the threshold up to which, under longitudinal pumping, the sample did not absorb any power; there was no noticeable heating of the sample at all.

The calculation of the connection between the power P_0 incident on the radiator and the field h_0 at the sample is

difficult, in the case of a dielectric resonator. To determine this connection we compared the threshold powers of the parametric excitation in a calibration sample (YIG sphere of ≈ 0.5 mm diameter) under identical conditions in the dielectric and cavity resonators. For the cavity resonator we used the relation (the sample was in the antinode of the magnetic field)

$$h_0^2 = (8\pi Q_0/\alpha\omega V_0) P_0 (1 - |\Gamma|^2), \tag{4}$$

which follows from the definition of the resonator Q and from the expression for the energy stored in it. Here Q_0 is the intrinsic Q of the resonator, V_0 is its volume, Γ is the reflection coefficient, and

$$\alpha = \int_{V_0} h^2\, dV / h_0^2 V_0, \tag{5}$$

where h is the amplitude of the alternating magnetic field inside the cavity volume. The value of α is easily calculated for hollow resonators of simple shape (see, e.g., Ref. 22); for the rectangular resonator used by us, made from a waveguide with cross section 10×23 mm, we obtained $\alpha = 0.48$ when the sample was placed near the center of its end wall.

It should be noted that the inhomogeneity of the alternating magnetic field in the dielectric resonator is larger than in the usually employed cavity resonators. However, the sample dimensions (≈ 0.5 mm) in our case were smaller than those used in the case of cavity resonators (2–3 mm), and the inhomogeneity of the field over the sample did not exceed 3%. According to the theory,[23] inhomogeneity of the alternating field increases h_c by a factor $1 + l_k/L$ times, where l_k is the mean free path of the spin waves and L is a parameter that characterizes the inhomogeneity of the field. For the inhomogeneity indicated above we have $L \sim 0.1$ cm. The mean free path $l_k \approx 2Dk/2\Delta H_k$, as follows from the results presented below, was 10^{-3}–10^{-4} cm. Thus, the error in the determination of ΔH_k, due to the inhomogeneity of the alternating field and of the dielectric resonator, could in our case be neglected.

The inhomogeneous exchange constant D was calculated by us from the temperature dependences of the spontaneous magnetization,[24,25] which corresponded in a rather large temperature interval to the 3/2 law. These values were $3.35\cdot 10^{-10}$ Oe·cm^2 and $2.02\cdot 10^{-10}$ Oe·cm^2 for $CdCr_2Se_4$ and $CdCr_2S_4$, respectively.

We used in the present study $CdCr_2Se_4$ single crystals grown by two methods, by spontaneous crystallization from the solution in the melt[26] and by gas transport.[27] The $CdCr_2S_4$ single crystals were grown by the gas transport method.[28]

3. SHAPE OF THRESHOLD CURVES

The dependences of the threshold field h_c on the external constant magnetic field H_0, which we shall call hereafter the threshold curves, are shown in Fig. 2 for several samples. The arrows mark the values of $H_0 = H_c(\pi/2) = H_{ci}(\pi/2) + 4\pi M_0/3$. It can be seen from the figure that for curves 1–3 the minimum of the threshold field is located near $H_0 = H_c(\pi/2)$, as is the case in YIG at room temperature. This shape of the threshold curve ("butterfly") is assumed to be typical for longitudinal pumping. Its left branch corresponds to excitation of spin waves with $\theta_k = \pi/2$ and with k that depends on H_0 in accordance with (2), while the right-hand branch (in the approximation of plane spin waves) corresponds to excitation of waves with $k \approx 0$ and with angle θ_k that depends on H_0.

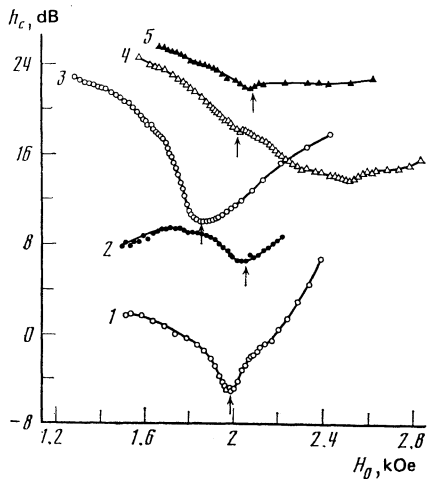

FIG. 2. Threshold curves: 1—$CdCr_2Se_4$, ⟨100⟩, 4.2 K; 2—$CdCr_2Se_4$, ⟨111⟩, 4.2 K; 3—$CdCr_2Se_4$, ⟨100⟩, 77 K; 4—$CdCr_2S_4$, ⟨100⟩, 4.2 K; 5—$CdCr_2S_4$, ⟨111⟩, 4.2 K. The arrows show the calculated values of $H_c(\pi/2)$. The zero of the ordinate axis corresponds to $h_c = 1$ Oe.

This treatment of the branches of the threshold curve, however, is based on the assumption that if the $\Delta H_k(\theta_k)$ dependence is nonexistent or weak, so that in expression (1) the decisive factor is $\sin^{-1}\theta_k$. It was indicated in Ref. 29 that this assumption may not be satisfied; this was demonstrated theoretically for two relaxation mechanisms (scattering of spin waves by dislocations and by interstitial impurities) and was used to explain the "anomalous" course of the right-hand side of the threshold curve in YIG at low temperatures. The dependence of ΔH_k on the angles θ_k and φ_k was used to explain the shapes of the experimental threshold curves also in Ref. 30.

As can be seen from Fig. 2, in our case the threshold curve in $CdCr_2S_4$ at 4.2 K for the easy direction ⟨100⟩ has a broad minimum at a field value exceeding $H_c(\pi/2)$ by more than 500 Oe. For the hard direction ⟨111⟩ in the same crystal, the minimum of h_c is located near $H_c(\pi/2)$, but the right-hand side of the threshold curve is almost flat. It is quite probable that the cause of these anomalies is the strong angular dependence of ΔH_k. As can be seen from Fig. 2, in the investigated $CdCr_2S_4$ sample there exist much larger values of h_c than in $CdCr_2Se_4$, probably owing to the larger contribution made to ΔH_k by the scattering of the spin waves from the defects. The principal role is played here apparently not by scattering from those defects which were considered in Ref. 29, but by scattering which can also give a strong angular dependence of ΔH_k.

The influence of the $\Delta H_k(\theta_k)$ dependence on the threshold curve is not confined to its right-hand branch; this dependence can influence the form of the threshold curve also

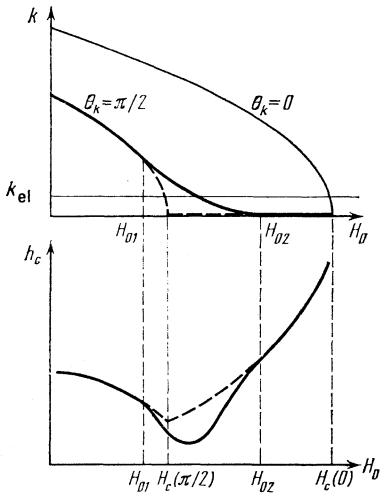

FIG. 3. Dependences of the wave number k of the excited spin waves and of the threshold field on the external magnetic field H_0. Dashed lines—there is no dependence of ΔH_k on θ_k, solid—there is a sufficiently strong dependence; k_{el}—wave number, independent of H_0, of the elastic waves of the same frequency as the excited spin waves.

at $H_0 < H_c(\pi/2)$. Figure 3 shows schematically the change of the wave number k of the excited spin waves and of the threshold field H_c with changing H_0. The dashed lines show the curves $k(H_0)$ and $H_c(H_0)$ in the case when ΔH_k does not depend on θ_k (and increases with increasing k), while the solid lines show the case when there is a sufficiently strong $\Delta H_k(\theta_k)$ dependence. In the latter case, in a certain field interval from H_{01} to H_{02}, the minimization of h_c with respect to θ_k can yield $k \neq 0$ and $\theta_k \neq \pi/2$. In this region, without resorting to some supplementary data, we can neither determine ΔH_k from the measured value of h_c nor find the values of k of the excited spin waves. It is easily seen that by using in this case Eqs. (1) and (2) and assuming that $\theta_k = \pi/2$ we, first, overestimate ΔH_k and, second, underestimate the k, as a result of which the $\Delta H_k(k)$ dependence can be substantially distorted.

With increase of k, however, an increase takes place in the contribution made to ΔH_k by the intrinsic relaxation processes, for which the dependence on θ_k should not be strong. Therefore, at sufficiently large k, the relative contribution of the processes that depend on θ_k in ΔH_k should decrease, and the minimization of h_c should lead to excitation of the spin waves with θ_k equal to (or close to) $\pi/2$. In our case, owing to the low values of D, the contributions of the intrinsic relaxation processes are large, and the $k(H_0)$ dependence is strong. This gives grounds, in those cases when the position of the minimum of the threshold curve differs little from the calculated value $H_c(\pi/2)$ (as for the curves 1–3 on Fig. 2), to assume that $\theta_k = \pi/2$, on the left hand side of the threshold curve and construct, by starting from this curve, the $\Delta H_k(k)$ dependence. The errors that can arise in this case will be significant only in the region of relatively small k.

It must be noted that in the presence of a large number of scattering centers, i.e., precisely when a strong dependence of ΔH_k on θ_k arises, Eq. (1) for the threshold field, owing to the non-additivity of the scattering contribution, becomes strictly speaking incorrect[6] if ΔH_k in it is taken to mean a relaxation parameter that is the sum of the parameters determined by different elementary processes. However, at a small number of scattering centers (when the "butterflies" are not anomalous) the contribution of the scattering can be regarded as additive[6] and Eq. (1) can be used to determine the true relaxation parameter.

4. TEMPERATURE DEPENDENCES AND CRYSTALLOGRAPHIC ANISOTROPY OF THE SPIN-WAVE DAMPING

Figure 4 shows the temperature dependences of the relaxation parameter $\Delta H_{k\to 0}$ of the spin waves with $k \approx 0$ and $\theta_k \approx \pi/2$ (at $H_0 = H_c(\pi/2)$) for a $CdCr_2Se_4$ crystal grown by the gas-transport method. As can be seen from Fig. 4, at low temperatures one observes a strong crystallographic anisotropy of the relaxation parameter, which decreases with increasing temperature and practically vanishes at $T \gtrsim 30$ K. In the temperature range 30–90 K the quantity $\Delta H_{k\to 0}$ remains practically unchanged, and then begins to increase rapidly as a result of the influence of the critical fluctuations of the magnetization. Such a temperature dependence is similar to the temperature dependences of the linewidth ΔH_0 of homogeneous ferromagnetic resonance in the same crystal.[31] There are, however, rather significant quantitative differences: the anisotropy of ΔH_0 vanishes at higher temperatures (≈ 60 K), the fluctuation growth also sets in at higher temperatures (≈ 120 K), there is no decrease of ΔH_0 in the $\langle 100 \rangle$ direction with decreasing temperature at $T < 30$ K. The actual causes of these differences are not clear to us. The low-temperature anisotropy of $\Delta H_{k\to 0}$, just as of ΔH_0 (see Ref. 31), however, is due to the contribution of the ionic relaxation mechanism, namely, to the appearance of Cr^{2+} ions whose density in these crystals is quite high.

The dependences of $\Delta H_{k\to 0}$ on the angle θ between the $\langle 100 \rangle$ axis and the direction of $\mathbf{M}_0$ in the $\{110\}$ plane at 4.2 K, for $CdCr_2Se_4$ crystals grown by the gas-transport method and by spontaneous crystallization from a solution in the melt, are shown in Fig. 5. It can be seen from the figure that for both crystals one observes in the $\langle 111 \rangle$ direction a maximum of $\Delta H_{k\to 0}$, and its value is several times larger for a

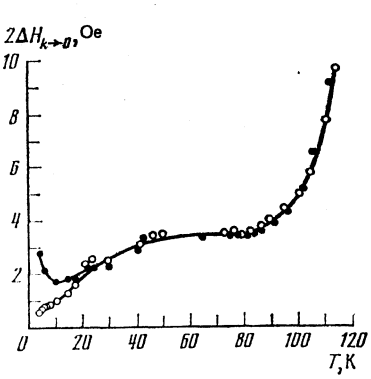

FIG. 4. Temperature dependences of 2 $\Delta H_{k\to 0}$ in $CdCr_2Se_4$: ○—$\langle 100 \rangle$; ●—$\langle 111 \rangle$.

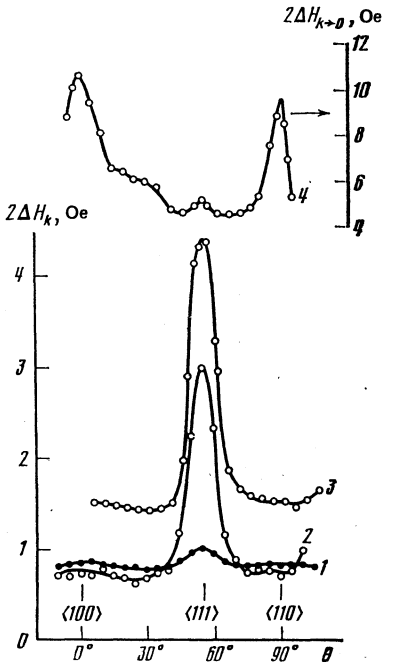

FIG. 5. Angular dependences of $2\Delta H_k$ in $CdCr_2Se_4$ at liquid helium temperature: 1—$CdCr_2Se_4$ grown from a solution in the melt, $k \approx 0$; 2—$CdCr_2Se_4$ grown by the gas-transport method, $k \approx 0$; 3—the same crystal, $k \sim 10^6$ cm^{-1}; 4—$CdCr_2Se_4$: 0.5% Ag, $k \approx 0$.

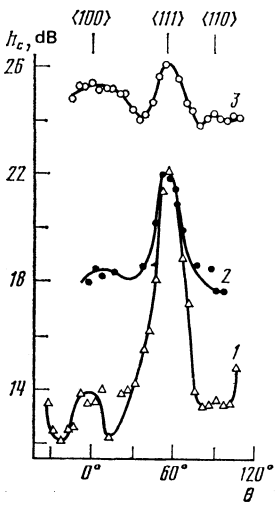

FIG. 6. Angular dependences of the threshold field in $CdCr_2S_4$ at liquid-helium temperature; 1—$CdCr_2S_4$, curve plotted for the minimum threshold field; 2—$CdCr_2S_4$, $H_0 = H_c(\pi/2)$; 3—$CdCr_2S_4$: 2.5% Ag, $H_0 = H_c(\pi/2)$.

crystal grown by the gas transport. At the same time, the quantity ΔH_k far from the ⟨111⟩ direction for this crystal is less, the minimum value $2\Delta H_{k\to 0}$ for it amounts to 0.6 Oe.[31] It follows therefore that in the $CdCr_2Se_4$ crystal grown by gas transport there are many more Cr^{2+} ions because of the large number of Se vacancies and because atoms of the transporting substance (in this case, chlorine) enter into the lattice, but there are some fewer defects, primarily geometric (cracks or voids).

As can be seen from Fig. 5, small maxima are observed for both crystals in the ⟨100⟩ and ⟨110⟩ directions; these crystals were not observed earlier on the angular dependences of ΔH_0 of these crystals.[20,31] The maxima of ΔH_0 in these directions appear for $CdCr_2Se_4$ doped with silver,[20] and are due to the influence of the Cr^{4+} ions produced by this doping. Figure 5 shows the angular dependence of $\Delta H_{k\to 0}$ also for a $CdCr_2Se_4$ crystal doped with silver. It contains strong maxima in the ⟨100⟩ and 110 directions, just as the angular dependence of ΔH_0 for this crystal. One can also see clearly in the ⟨111⟩ direction, however, a small maximum which was not observed earlier on the angular dependences of ΔH_0.

It follows from the foregoing angular dependences that the $CdCr_2Se_4$ crystal contains as a rule, besides the fundamental-valence ions Cr^{3+}, also Cr^{2+} and Cr^{4+} ions, but the density ratio of the Cr^{2+} and Cr^{4+} ions depends on the growth and doping conditions. A change of this ratio such that the density of the Cr^{2+} is increased is achieved by annealing the $CdCr_2Se_4$ crystals in vacuum.[32] Measurements of the angular dependences $\Delta H_{k\to 0}$ have shown that after such an annealing (20 hours at a temperature 620°C) the peaks in the ⟨111⟩ directions have increased more than in the ⟨100⟩ and ⟨110⟩ directions, owing to the larger probability of formation of Se vacancies than of Cd vacancies by this annealing. We note that the possibility of the coexistence of Cr^{2+} and Cr^{4+} ions is due apparently to the fact that these ions are in potential wells near the defects that stimulate their formation.

For the $CdCr_2S_4$ crystal, the threshold curves, as noted above, have an "anomalous" character, due apparently to the fact that, at any rate, $\theta_k \neq \pi/2$ on a considerable section of these curves. This does not enable us to determine ΔH_k, and Fig. 6 shows for this crystal the angular dependences of the directly measured quantity—the threshold field, and furthermore for two cases: at fields $H_0 = H_c(\pi/2)$ (where, however, in this case $k \neq 0$ and $\theta_k \neq \pi/2$), and at the minima of the threshold curves (at fields considerably exceeding H_c $(\theta/2)$ where, as can be seen from Fig. 3, we also have generally speaking $k \neq 0$, $\theta_k \neq \pi/2$). It can be seen from Fig. 6 that despite the uncertainty in the values of k and θ_k, the crystallographic anisotropy of the threshold, i.e., of ΔH_k, manifests itself distinctly on both curves. This anisotropy corresponds in the main to the anisotropy of ΔH_0 in this crystal[28]: large maxima of h_c, as well as of ΔH_0, are observed in the ⟨111⟩ direction and are due to the Cr^{2+} ions. However, weak maxima appear also in the ⟨100⟩ and ⟨110⟩ directions, as can be particularly clearly seen from the $(h_c)_{\min}(\theta)$ curve. Thus, coexistence of the Cr^{2+} and Cr^{4+} ions takes place also in the $CdCr_2S_4$ crystals. We note that it was observed for these crystals also on the angular dependences of ΔH_0.[33]

Figure 6 shows also the angular dependence of h_c (at $H_0 = H_c(\pi/2)$) for a $CdCr_2S_4$ crystal doped with silver. It can be seen that the doping increases substantially the maxima of h_c in the ⟨100⟩ and ⟨110⟩ directions. However, in contrast to the $CdCr_2Se_4$ crystals doped with silver, these maxima remained smaller than for the maxima in the ⟨111⟩ directions. One of the causes of this may be the larger degree

of localization of the Cr^{2+} and Cr^{4+} ions near the corresponding defects in $CdCr_2S_4$, as was noted earlier[34] on the basis of measurements of the ferromagnetic resonance.

5. DEPENDENCE OF THE SPIN-WAVE DAMPING ON THE WAVE NUMBER k

As follows from the preceding arguments, the dependences of ΔH_k on k can be obtained with reasonable error from the threshold curves for $k \gtrsim 2\times 10^5$ cm^{-1}, when their minima are close to $H_c(\pi/2)$. The angular dependence of ΔH_k at $k \approx 10^6$ cm^{-1} for one of the samples is shown together with the angular dependence of $\Delta H_{k\to 0}$ in Fig. 5. It can be seen that the increase of k has led to an increase of the damping due mainly to the contribution of the intrinsic relaxation processes. This increase, however, is anisotropic, namely it has a maximum in the ⟨111⟩ direction. This is most likely due to the k-dependence of the ionic contribution to ΔH_k.

Figure 7 shows plots of ΔH_k against k for one of the $CdCr_2Se_4$ samples at different temperatures and orientations of $\mathbf{M}_0$. Also shown are the calculated dependences of the contributions of the dominant intrinsic relaxation processes: three-magnon dipole coalescence 3c (Ref. 3) and four-magnon exchange scattering 4sc (Ref. 5) (the three-magnon dipole splitting is forbidden by the conservation laws at $k < 1.27\times 10^6$ cm^{-1}). The figure shows first of all that the indicated relaxation processes make a rather substantial contribution to the spin-wave damping. It must be noted that at 4.2 K the approximations assumed in Ref. 3 in the calculation of the contribution of the process 3c in ΔH_k are poorly satisfied, and the calculation is rough. Nonetheless, it can be concluded that this temperature the process 3c makes a substantial contribution to the dependence of ΔH_k on k (especially for the ⟨100⟩ direction, in which the ionic contribution is a minimum). At 77 K, as seen from Fig. 7, the intrinsic processes make the principal contribution to ΔH_k —not less than two-thirds (with the exception of the region of small k). The excess of the experimental values over the calculated sum of the contributions of the two intrinsic processes taken into account for large k at 4.2 K (at this temperature only the process 3c is effective in practice) can be attributed to the k-dependence of the contributions of the ionic process. Such a dependence does not follow from the theory of Ref. 7, but it was observed in experiment for rare-earth ions in YIG.[12,18] At 78 K, the cause of this excess is not clear—one cannot exclude the influence of other intrinsic processes or carriers.

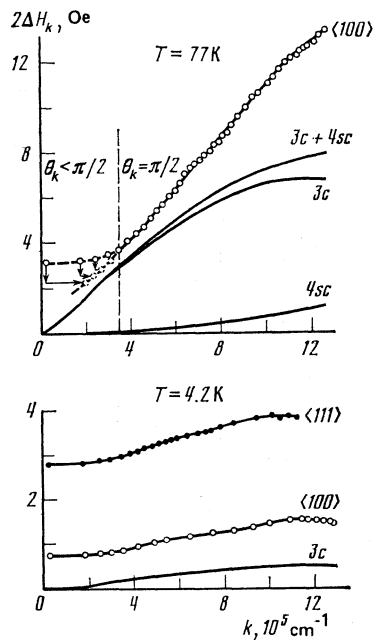

FIG. 7. Dependences of 2 ΔH_k on k in $CdCr_2Se_4$ (sample grown by the gas-transport method). 3c and 4sc—theoretical contributions of the processes of three-magnon coalescence and four-magnon scattering. The arrows show how the experimental dependence should be corrected when account is taken of the fact that $\theta_k < \pi/2$.

As for the excess of the experimental points over the sum of the contributions of the aforementioned intrinsic processes at small k, it must be noted above all that there are always intrinsic processes, other than 3c and 4sc, which contribute to ΔH_k as $k\to 0$. These include, in particular, the Kassuya-LeCraw magnon-phonon processes.[2,35] The dependence of their contribution to ΔH_k on k is not clear (in Ref. 2 it is assumed to be nonexistent), but it cannot be strong enough, and furthermore decrease with increasing k, to explain the aforementioned discrepancy between experimental and calculated curves in the region of small k. One of the causes of this discrepancy may be the error, discussed above, in the determination of both ΔH_k and k, due to the failure to satisfy the assumption $\theta_k = \pi/2$ all the way to $k = (2\text{–}3)\times 10^5$ cm^{-1}. The plot for 77 K in Fig. 7 shows schematically how the experimental curve should be corrected to take this circumstance into account; however, without knowing the actual values of θ_k no such correction can be made.

Another cause of the discrepancy between the experimental curves and the sum of the contributions of the processes 3c and 4sc at small k may be the increase in the damping of the spin (or more accurately magnetoelastic) waves in the region of the intersection of the spectra of the spin waves and of the elastic waves (see, e.g., Ref. 36). Using the values of the rates of the transverse and longitudinal elastic waves in $CdCr_2Se_4$ (Ref. 37), it is easy to verify that these crossings will lie in the regions of values $k \sim 10^5$ cm^{-1} (at $k = 0.7\times 10^5$ cm^{-1} and 1.3×10^5 cm^{-1}). Upon excitation of spin waves with $\theta_k \neq \pi/2$, these peaks (see Fig. 3) are stretched out along H_0 and will not be noticeable on the threshold curves, but their influence on the $\Delta H_k(k)$ dependence can be substantial.

CONCLUSION

The main premises of the theory of parametric excitation and of the theories of the relaxation processes of the spin waves in ferromagnets, which were compared earlier with experiments on ferrimagnets (principally on one substance—YIG) and were formed to a considerable degree under the influence of these experiments, are compared in the present paper with experiments performed on ferromagnets with parameters considerably differing from those of YIG. No substantial discrepancies were observed in this case between experiment and the developed premises.

The singularities of the damping of the spin waves in chromium-chalcogenide spinels $CdCr_2Se_4$ and $CdCr_2S_4$, on which these experiments were performed, consist in the following.

1. Large values of contributions of the intrinsic relaxation processes to ΔH_k, due to small values of the inhomogeneous-exchange constant.

2. The inevitable presence of strongly anisotropic contributions of ionic relaxation processes, due to the rapidly relaxing Cr^{2+} and Cr^{4+} ions that are always present in these crystals.

3. The substantial, in many cases, influences of the dependence of the damping of the spin waves on the angle θ_k on the conditions of parametric excitation, and consequently on the character of the threshold curves, which makes it difficult to obtain on the basis of these curves the dependence of ΔH_k on the wave number k.

The results of the present study have also shown that measurement of the damping of the spin waves in longitudinal pumping, despite the restriction noted above, is (at any rate when dielectric resonators are used) a more sensitive method of revealing anisotropic contributions of rapidly relaxing ions than an investigation of homogeneous ferromagnetic resonance.

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