Direct observation of photoinduced change in magnetocrystalline anisotropy in $CdCr_2Se_4$:Ga

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The effect of pulsed infrared laser radiation on magnetization of the magnetic semiconductor $CdCr_2Se_4$ was investigated. Exposure to a laser pulse of 0.01 J/cm^2 was found to result in a photoinduced increase in the cubic magnetocrystalline anisotropy energy by 10^3 erg/cm^3. It is suggested that illumination results in the formation of uniaxial anisotropic centers. When averaged over the volume, these centers contribute to the cubic magnetocrystalline anisotropy of the specimen. The anisotropy energy of the photoinduced anisotropic centers is estimated, and the axes of easy magnetization are identified.

INTRODUCTION

Light is known[1,2,3] to have an effect on the magnetic parameters of the ferromagnetic semiconductor $CdCr_2Se_4$. It has been shown[4,5] that exposure of $CdCr_2Se_4$ specimens at 78 K to light with $h\nu \sim 1$–1.3 eV is accompanied by a reduction in the dynamic magnetic permeability and an increase in the dynamic coercive force of the specimen. This has been referred to as the photoferromagnetic effect (PFE) and has been observed in specimens doped with a donor impurity (Ga) and in undoped specimens with growth defects. Doping of $CdCr_2Se_4$ with silver (acceptor) resulted in the suppression of PFE even for silver concentrations of about 10^{19} cm^3. Spectroscopic investigation[6] of PFE have shown that this effect is due to transitions of electrons from the valence band to impurity levels lying below the bottom of the conduction band. It is suggested that the photoexcitation of electrons to these levels results in the formation of photoinduced uniaxial anisotropic centers capable of efficient interaction with domain walls. It is therefore interesting, from the point of view of explaining the nature of photomagnetic defects in $CdCr_2Se_4$, to perform a direct examination of the effect of light on magnetocrystalline anisotropy in single-crystal specimens of this compound. An investigation of this type is reported below.

EXPERIMENTAL METHOD

One of the direct methods of examining magnetocrystalline anisotropy in ferromagnets is to measure the component of the magnetization vector **M** of the specimen at right angles to the magnetic field **H** for different directions of the latter relative to the crystallographic axes of the ferromagnet. The semiconductor $CdCr_2Se_4$ is a cubic ferromagnet ($T_c \sim 130$ K) and its easy magnetization axes lie along the [100] crystallographic axes. It can be shown that, for different orientations of the external field **H** relative to the crystallographic directions in the (001) plane of the crystal, the component of magnetization perpendicular to **H** for $k_1/MH \ll 1$ is $M_\perp = (k_1/2H)\sin 4\psi$, where ψ is the angle between **H** and the [100] axis and k_1 is the first cubic anisotropy constant of the ferromagnet. Hence, if the specimen is magnetized to saturation (i.e., $|\mathbf{M}| = M_s$, where M_s is the saturation magnetization) and M_s is constant, a variation in $M_\perp$ in a fixed external field can only be due to a variation in anisotropy, i.e., the constant k_1. One the other hand, $M_\perp$ increases with increasing k_1 and decreases with decreasing k_1 (for $k_1 > 0$, which is the case for $CdCr_2Se_4$). Thus, $M_\perp$ is a direct measure of the magnetocrystalline anisotropy energy of the ferromagnet.

We have investigated the effect of pulsed laser radiation on the perpendicular ($M_\perp$) and longitudinal ($M_\parallel$) components of magnetization in $CdCr_2Se_4$ specimens for different orientations of the external field **H** relative to the crystallographic [100] directions in the (001) plane of the crystal. The source of light was a Q-switched neodymium-glass laser producing a first-harmonic pulse ($\lambda = 1.064\,\mu$m) of length 20 ns and power up to 0.5 MW. The repetition frequency of the light pulses was 12.5 Hz. The second-harmonic power ($\lambda = 0.53\,\mu$m) did not exceed 130 kW. The laser beam was incident at right angles to the specimen surface. The specimens were in the form of disks with (001) base plane. Their diameter and thickness were 1.5–0.3 and 0.15–0.25 mm, respectively. The external magnetic field was applied in the plane of the disk. The variation in magnetization under the influence of the light pulse was examined with a search coil (3–10 turns) placed next to the specimen. The geometry of the experiment is illustrated in Fig. 1. The variation in the longitudinal component $M_\parallel$ was examined by arranging the axis of the search coil parallel to the external field **H**. The perpendicular component of the magnetization $M_\perp$ was examined with the coil axis perpendicular to the field. The

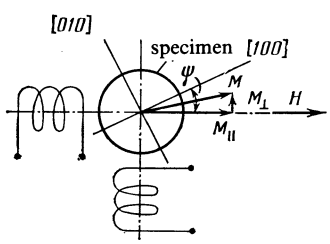

FIG. 1. Mutual orientation of the specimen, search coil, and magnetic field.

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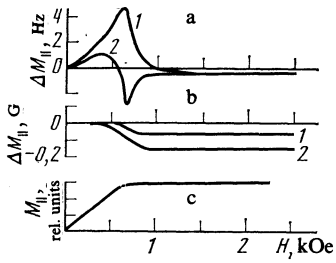

FIG. 2. Photoinduced change in the longitudinal component of magnetization $\Delta M_{\parallel}$ as a function of the external magnetic field ($T = 78$ K): a—$CdCr_2Se_4$:$Ga_{0.0015}$, $\lambda = 1.064$ μm, curve 1—**H** along the [100] direction, 2—along the [110] direction; b—curve 1—$CdCr_2Se_4$:$Ga_{0.0015}$, $\lambda = 0.53$ μm, 2—$CdCr_2Se_4$:$Ag_{0.002}$, $\lambda = 1.064$ μm; c—longitudinal magnetization of the specimen as a function of the external magnetic field.

amplitude of the emf generated in the search coil was fed into a strip chart recorded through an integrating synchronous pulse detector.

EXPERIMENTAL RESULTS

We have examined two types of specimen: I—specimens of $CdCr_2Se_4$ and $CdCr_2Se_4$:Ga, in which we observed the effects of light on the dynamic magnetic permeability and coercive force and II—specimens of $CdCr_2Se_4$ in which this effect was not observed.

Figures 2a and b show the photoinduced change $\Delta M_{\parallel}$ in the longitudinal component of magnetization. When the $CdCr_2Se_4$:Ga_x specimen ($x = 0.0015$) was exposed to the first harmonic from the neodymium-glass laser for H less than the saturation field, i.e., the specimen was divided into the domains, the magnitude and sign of $\Delta M_{\parallel}$ were found to depend on the direction of the external magnetic field relative to the crystallographic axes. When the field was in excess of the saturation value, it was found that $\Delta M_{\parallel} < 0$ and was not very dependent on the orientation and strength of the external magnetic field. On the other hand, when the specimen was exposed to the second harmonic of the neodymium-glass laser, the variation in the longitudinal magnetization was observed only in saturating fields and $\Delta M_{\parallel} < 0$ (Fig. 2b, curve 1). Moreover, $\Delta M_{\parallel}$ was independent of the orientation of **H** relative to the crystallographic axes. This effect is due to thermal variations in spontaneous magnetization in the specimen. When the domain structure is present, $M_{\parallel}$ depends only on the external field strength and the demagnetizing factor of the specimen, but is independent of the spontaneous magnetization M_s. This means that, when the domain structure is present, $M_{\parallel}$ will not vary in the course of thermal variations in M_s, and the signal proportional to the thermal variation in spontaneous magnetization will appear only in saturating fields. When the $CdCr_2Se_4$:Ag_x with $x > 0.002$ was exposed to the first harmonic of the neodymium-glass laser, we observed only the thermal signal with negative phase, the dependence of which on the magnetic field (Fig. 2b, curve 2) is similar to that shown in Fig. 2b (curve 1). The magnitude of this variation in $M_{\parallel}$ does not depend on the direction of **H** relative to the crystallographic axes of the crystal.

Figure 3 shows the measured photoinduced variation in the perpendicular component of magnetization $\Delta M_{\perp}$ as a

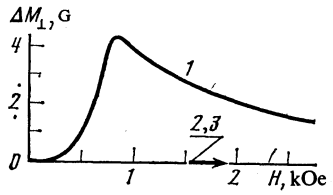

FIG. 3. Photoinduced transverse magnetization as a function of the external magnetic field ($T = 78$ K, $\lambda = 1.064$ μm: curve 1 corresponds to the angle $\psi \simeq 22°$ between **H** and [100]; 2, 3—**H** parallel to [100] and [110], respectively.

function of the external field for the $CdCr_2Se_4$:Ga_x specimen ($x = 0.0015$). $\Delta M_{\perp}$ is practically zero for **H** lying along the [100] and [110] axes in saturating fields. The maximum variation in $M_{\perp}$ is observed in external fields approaching the saturation value for **H** lying at an angle $\psi = 22°$ to the [100] axis. It was found that $\Delta M_{\perp}$ decreased with external field as $1/H$. The angular dependence of $\Delta M_{\perp}$ on the orientation of the external field relative to the [100] axis is shown in Fig. 4.

When the $CdCr_2Se_4$:Ga_x specimen ($x = 0015$) was exposed to the second harmonic of the neodymium-glass laser, the results were similar to those obtained for $CdCr_2Se_4$:Ag_x in that only the thermal variation of the longitudinal component of magnetization was recorded (Fig. 2b) and there was no photoinduced change in the perpendicular component for any orientation of **H** relative to the crystallographic axes.

The photoinduced variation in the perpendicular component of magnetization, $\Delta M_{\perp}$, is very dependent on the concentration of gallium in the $CdCr_2Se_4$ specimens. $\Delta M_{\perp}$ increases with increasing concentration in the range $0 < x < 0.002$. The maximum change in this component in $CdCr_2Se_4$:$Ga_{0.002}$ is $\Delta M_{\perp} \simeq 4$–6 G, i.e., of the order of 2% of the spontaneous magnetization of the specimen.

We have carried out experiments to elucidate the spectral range of the radiation that is responsible for the photoinduced increase in $M_{\perp}$ in $CdCr_2Se_4$. The MDR-2 monochromator and the pulsed light source ISSh-100-3M were used to produce monochromatic radiation in the form of 2-μs pulses. Examination of the spectral characteristics of the photoinduced increase in the perpendicular component of magnetization showed that this effect was due to light pulses with $h\nu \sim 1$–1.2 eV, which corresponded to the spectral range responsible for PFE.[6] The photoinduced increase in the perpendicular component of magnetization was observed only in the $CdCr_2Se_4$ specimen exhibiting PFE, and the spectra of the two effects were the same. It may therefore be concluded that the two effects are due to the same pho-

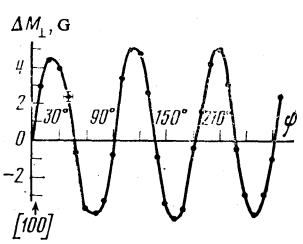

FIG. 4. Angular distribution of the photoinduced change in perpendicular magnetization $\Delta M_{\perp}$ ($T = 78$ K, $\lambda = 1.064$ μm, $CdCr_2Se_4$:$Ga_{0.0015}$, $H = 1.5$ kOe, ψ is the angle between **H** and [100].

toinduced centers. The increase in $M_\perp$ under the influence of light is due to the increase in the magnetocrystalline anisotropy energy. In fact, if the increase in $M_\perp$ were due to the increase in M_s under illumination (this effect was predicted and discussed in Refs. 7 and 8), the increase in $M_\perp$ would be zero at low temperatures, whereas experiments show that $\Delta M_\perp$ increases monotonically with decreasing temperature down to 4.2 K in a fixed external field. We also note that the increase in $M_\perp$ when the $CdCr_2Se_4$ specimens were illuminated was not due to the possible heating of the specimens because the quantities k_1 and M_s decreased monotonically with increasing temperature in $CdCr_2Se_4$. Hence, heating could only lead to an increase in $M_\perp$ in a constant external field.

DISCUSSION OF EXPERIMENTAL RESULTS

The currently available microscopic model of photoinduced centers[1,4,6] is based on the hypothesis of photoinduced change in the valence of the Cr ion. This model was further developed in Ref. 9 in which it was necessary to assume that the centers had uniaxial magnetocrystalline anisotropy in order to explain the induced magnetic aftereffects in $CdCr_2Se_4$:Ga. When the photoinduced uniaxial centers are averaged over the volume of the cubic crystal, they contribute to the cubic anisotropy if the centers occupy certain definite positions in the unit cell and their easy magnetization axes lie along equivalent directions, for example, either [100] or [111].

Without going into the nature of the photoinduced anisotropic centers in $CdCr_2Se_4$, let us estimate the concentration N and the anisotropy energy of such centers that are necessary to explain the observed change $\Delta M_\perp \sim 10^{-2} M_s$ under illumination of the $CdCr_2Se_4$ specimen. We shall suppose that the photoinduced centers are distributed uniformly throughout the specimen with concentration N and, when they are formed under illumination, their easy magnetization axes are uniformly distributed along the [111], [11$\bar{1}$], [$\bar{1}$11], and [$\bar{1}$1$\bar{1}$] directions in the crystal. We shall also assume that the centers are distant from one another so that the interactions between them can be neglected. The anisotropy energy of a region of volume N^{-1} adjacent to a photoinduced center is then given by

$$E_a^i = N^{-1}\{k_1(m_i^2n_i^2+m_i^2p_i^2+n_i^2p_i^2)+K_1\sin^2\beta_i+K_2\sin^4\beta_i\}, \quad (1)$$

where k_1 is the first cubic anisotropy constant of the crystal, K_1 and K_2 are the uniaxial anisotropy constants of the anisotropic center, β_i is the angle between **M** and the easy-magnetization axis of the ith center, and m_i, n_i, p_i are the direction cosines of the magnetization vector in the ith region. For centers with easy-magnetization axis along the [111] [11$\bar{1}$], [$\bar{1}$11], and [$\bar{1}$1$\bar{1}$] axes, we find that $\cos\beta_i$ are respectively equal to $(m_1+n_1+p_1)$, $(m_2+n_2-p_2)$, and so on. Averaging over the volume of the crystal, we find that, in the first approximation (assuming that the crystal is uniformly magnetized), the expression for the average anisotropy energy per unit volume of the crystal is

$$E_a \approx (k_1 + {}^4/_9 K_2 N)(m^2n^2+m^2p^2+n^2p^2). \quad (2)$$

Thus, the change in the anisotropy of the crystal as a whole during the formation of the photoinduced centers with easy-magnetization axes along [111] directions corresponds to the change $\Delta k_1 = 4/9(K_2N)$ in the first anisotropy constant. The photoinduced change in the perpendicular component of magnetization is then given by

$$\Delta M_\perp = \frac{2}{9}\frac{K_2N}{H}\sin 4\psi. \quad (3)$$

If we suppose that the easy-magnetization axes are distributed over the [100], [010], [001] directions, similar calculations show that the change in the first anisotropy constant is $\Delta k_1 = -2/3(K_2N)$. Experiment shows that $\Delta K_1 > 0$, so that the easy-magnetization axes of the photoinduced centers are distributed over the [111] directions. Expression (3) satisfactorily describes experimental results (Figs. 3 and 4) for $H > H_s$, where H_s is the saturation field. The quantity $\Delta M_\perp H$ amounts to about 1000 erg/cm^3, and hence $K_2N \sim 10^3$–10^4 erg/cm^3. This change in the magnetocrystalline anisotropy of the specimen is possible for $K_2 \sim 10^{-13}$–10^{-14} erg and $N \sim 10^{16}$–10^{17}. Such densities are attainable in practise when the specimen is pumped by short ($\tau \sim 10^{-8}$ s) and powerful ($W \sim 10^6$ W) laser pulses in the narrow spectral band in which photomagnetic effects are observed in $CdCr_2Se_4$.

When $\Delta M_\perp$ was determined, we neglected inhomogeneities in magnetization that arose around the photoinduced anisotropic centers. However, more accurate calculations (Ref. 10) yield a similar expression for $\Delta M_\perp$ for $[k_1 + 4/9(K_2N)]/MH \ll 1$, which again does not contain terms that depend on K_1.

The photoinduced change in the longitudinal component $M_\parallel$ includes contributions due to both K_1 and K_2. When H is greater than the saturation value, so that we have a single-domain specimen, we find that, in the first approximation in which only K_2 is taken into account, we have

$$\Delta M_\parallel \approx -M_s(K_2N/18MH)(k_1/MH)(1-\cos 8\psi). \quad (4)$$

Comparison of (3) with (4) shows that $\Delta M_\parallel \simeq (k_1/4MH)\Delta M_\perp$, i.e. $\Delta M_\parallel \sim 10^{-2}$–$10^{-3}\Delta M_\perp$, even for $H \simeq 1$ kOe. This explains why the $\Delta M_\parallel$ signal, which is periodic in the angle ψ, was not detected against the background of, for example, possible thermal signals, whereas the positive change in the component $M_\perp$ exceeds the possible thermal signal that should lead to a reduction in $\Delta M_\perp$ in constant H.

When the external field H is less than the value necessary to saturate the magnetization of the specimen, and the specimen splits into domains, the calculation of $\Delta M_\perp$ and $\Delta M_\parallel$ becomes much more complicated because the local field acting on each particular domain depends on the domain structure and varies from point to point in the crystal. The quantities $M_\parallel$ and $M_\perp$ vary both in the rotation of the vector **M** in each of the domains and when the domain boundaries are displaced or their thickness varies. We shall not, therefore, examine the experimental results obtained for $\Delta M_\parallel$ and $\Delta M_\perp$ in fields that were small in comparison with the saturating field. We merely note that the positive change in $M_\parallel$ observed under illumination of a single-domain specimen cannot be explained without taking into account the displacement of domain boundaries. It is important to allow

for the fact that, when H is close to the saturation field, the ratio of the domain volumes corresponding to different orientations of the magnetization vector in the crystal depends not only on the external field and the shape of the specimen, but also on the anisotropy energy. Hence, a change in the magnetocrystalline anisotropy under illumination should lead to a shift in domain walls.

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