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## Effect of light and an electric field on ferromagnetic resonance and photoconductivity in $\text{CdCr}_2\text{Se}_4$

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The effect of an electric field on the photo-induced changes of the ferromagnetic-resonance parameters and on the photoconductivity of  $\text{CdCr}_2\text{Se}_4$  is investigated. It is shown that an increase of the photo-induced change of the ferromagnetic resonance field is observed in electric fields  $E > E_{\text{thr}}$  ( $E_{\text{thr}}$  is the threshold field) and in this case the current-voltage characteristics become nonlinear. The behavior of the current-voltage characteristics of the photocurrent and the photo-induced shift of the ferromagnetic-resonance field are significantly different for samples exposed to "white" and monochromatic light. This is ascribed to the infrared photoconductivity quenching observed in  $\text{CdCr}_2\text{Se}_4$ . The experimental results are discussed on the basis of existing theories and concepts. It is shown that the most satisfactory interpretation of the experimental data can be obtained by assuming that the superexchange interaction integral is independent of the hole density produced by light or an electric field in the nonmagnetic sublattice.

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### 1. INTRODUCTION

The interaction of magnetic and electronic states in ferromagnetic semiconductors turns out to be in a number of cases quite strong and can lead to appreciable mutual changes of these states when the crystals are externally excited (by an electric field, by illumination, injection, ...).<sup>[1-3]</sup> The investigation of these changes of the magnetic and electronic states uncovers new possibilities for a more detailed study of carrier transport processes and the phenomenon of magnetic ordering.

The effects connected with the change of the magnetic state due to illumination of a sample were recently observed in the ferromagnetic semiconducting crystals  $\text{CdCr}_2\text{Se}_4$ .<sup>[4-6]</sup> In<sup>[4]</sup> it was shown, that the initial magnetic susceptibility of  $\text{CdCr}_2\text{Se}_4$ , doped with Ga decreases to approximately one-third when exposed to light. At helium temperatures, after the light is turned off, the initial susceptibility is not fully restored, and at nitrogen temperatures a slow return to the initial value is observed. In other studies<sup>[5,6]</sup> of the high-frequency magnetic permeability of  $\text{CdCr}_2\text{Se}_4$  (Ga) exposed to light it was shown that the photo-induced changes of the magnetic permeability have low inertia and are observed at millisecond durations of the illumination.

In the cited studies<sup>[4-6]</sup> the photo-induced effects were investigated in the absence of an external magnetic field, when the sample was in a demagnetized state. In<sup>[7]</sup> some of us investigated the photo-induced changes in the parameters of the ferromagnetic resonance (FMR) of  $\text{CdCr}_2\text{Se}_4$ , crystals in the magnetic-saturation state. It was observed that illumination of the samples with "white" light leads to a change in the FMR field, and this effect, just as in the work of Veselago *et al.*,<sup>[5]</sup> has low inertia, the relative magnitude of the effect being  $\delta H_r/H_r \sim 10^{-4}$  ( $H_r$  is the resonant field 2000 Oe,  $\delta H_r = H_{111} - H_d$ , where  $H_{111}$  is the resonant field in the presence of light and  $H_d$  is the resonant field in darkness).

We have investigated in this study the photo-induced change of the FMR field using additional excitation of electronic states of  $\text{CdCr}_2\text{Se}_4$  with the aid of an electric field.

### 2. EFFECT OF ELECTRIC FIELD ON PHOTO-INDUCED CHANGES OF THE FERROMAGNETIC-RESONANCE PARAMETERS

The FMR was excited in the  $\text{CdCr}_2\text{Se}_4$  crystals at a frequency 9000 MHz at  $T = 77^\circ\text{K}$ . The samples for the measurements were cut out of bulky single crystals in the form of disks  $\sim 100 \mu$  thick with the basal plane

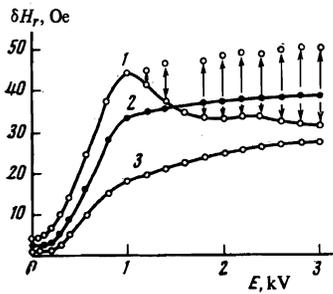


FIG. 1. Dependence of the change of the FMR field on the electric field at light intensities  $I/I_0$  equal to 1.0, 0.5, and 0.25 (curves 1, 2, and 3, respectively), with  $I_0 \sim 10^{-2}$  W/cm<sup>2</sup>. The sample is CdCr<sub>2</sub>Se<sub>4</sub> (0.037% Ga),  $T=77$  K (the arrows between the points correspond to the change of the resonant field in the instability region).

(110). Silver pointlike microcontacts were deposited on opposite ends of the disks by vacuum sputtering, and the external electric potential  $E$  was applied through these contacts. During the measurements, the samples were in a vacuum cryostat with optically transparent windows in the antinode of the microwave magnetic field of the resonator. The external magnetic field, perpendicular to the microwave field, was applied in the plane of the disk, and could make an angle  $0 < \varphi < 90^\circ$  with the electric field. Light of intensity  $\sim 10^{-2}$  W/cm<sup>2</sup> was incident normal to the surface of the sample through an opening in the resonator wall. Provision was made for irradiating the samples simultaneously with "white" and monochromatic light from an incandescent lamp of 500 W power using a UM-2 monochromator (the monochromator slits were  $\approx 1$  mm wide). The resonant field and the FMR absorption line width were measured with accuracy  $\pm 0.5$  Oe.

Simultaneously with the investigation of the FMR we investigated the photoconductivity (PC). To this end, a resistor was connected in series with the sample and with the power source, and the signal from the resistor was fed to a measuring amplifier or to the input of an  $x$ - $y$  recorder.

In the absence of an electric field, the photo-induced shift of the FMR field ( $\delta H_r$ ) at the maximum employed light intensities reached 5–10 Oe.<sup>[7]</sup> Application of an external electric field above a certain threshold value,  $E > E_{thr}$ , increased the shift  $\delta H_r$  to 50 Oe.

Figure 1 shows the values of the shift of the resonant field as a function of the external electric field for light intensities  $I/I_0$  equal to 1.0, 0.5, and 0.25 ( $I_0 \sim 10^{-2}$  W/cm<sup>2</sup>). We see that for each intensity, when a certain threshold field  $E_{thr}$  is reached, equal to 100, 180, and 250 V (curves 1, 2, and 3), an increase of  $\delta H_r$  is observed, which tends to saturation with further increase of the field. At intensities  $I \geq I_0$  in fields  $E \sim 1$  kV, the magnitude of the effect decreases somewhat, and in strong fields an instability is observed, connected with the spontaneous jumplike change of  $\delta H_r$ . No dependence of  $\delta H_r$  on the angle between the electric and magnetic fields was observed. Nor did turning on an electric field in darkness change the value of  $\delta H_r$ .

Figure 2 shows the current-voltage characteristics of the photocurrent at intensities  $I/I_0$  equal to 1.0, 0.5, and 0.25. The dark current of these crystals in maximum electric fields is  $\sim 1$   $\mu$ A, and their current-voltage characteristic is linear in the entire range of electric fields. In the presence of light, a deviation from linearity is observed at each value of the intensity after  $E_{thr}$  is reached. At  $I \geq I_0$ , on the section of the current-voltage characteristic at  $E \sim 1$  kV, a decrease of the photocurrent and a jumplike instability are observed. From a comparison of Figs. 1 and 2 it is seen that a definite correlation exists between the changes of the magnetic and electric properties in CdCr<sub>2</sub>Se<sub>4</sub>. The main increase  $\delta H_r$  is observed in fields  $E$  at which a deviation of the current-voltage characteristic from Ohm's law takes place. The section with the photocurrent instability coincides with the region of saturation of  $\delta H_r$  and with its instability. In fields  $E \sim 3$  kV, an abrupt increase of the current and a breakdown are observed. The breakdown occurs as a rule on the surface of the crystal and damages the latter. After the breakdown, the samples lose their sensitivity to light, but the magnetic parameters (line width and FMR field) do not undergo substantial changes.

Thus, when light or an electric field are separately applied to the sample, there are either no changes whatever in the magnetic properties (when only the electric field is applied) or insignificant changes occur (when the light is applied). On the other hand, the simultaneous action of the light and of the electric field under the conditions described above lead to appreciable changes of both the magnetic and the electric properties. Recognizing that the change of the magnetic characteristics under the influence of the light and of the electric field is apparently a consequence of excitation of electronic states, we shall investigate in greater detail the electric parameters of the crystals under different experimental conditions.

### 3. SINGULARITIES OF THE PHOTOCONDUCTIVITY OF CdCr<sub>2</sub>Se<sub>4</sub> UNDER MONOCHROMATIC ILLUMINATION

Let us examine the singularities in the behavior of the PC under monochromatic illumination in strong electric fields. As shown in<sup>[6]</sup>, for the CdCr<sub>2</sub>Se<sub>4</sub> crys-

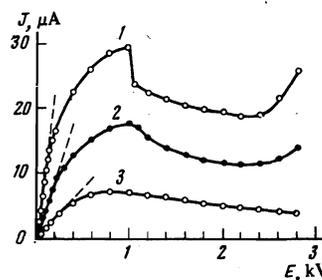


FIG. 2. Current-voltage characteristics of photocurrent  $J$  following illumination with "white" light with intensities  $I/I_0$  equal to 1.0, 0.5, and 0.25 (curves 1, 2, and 3, respectively). Sample CdCr<sub>2</sub>Se<sub>4</sub> (0.037% Ga).

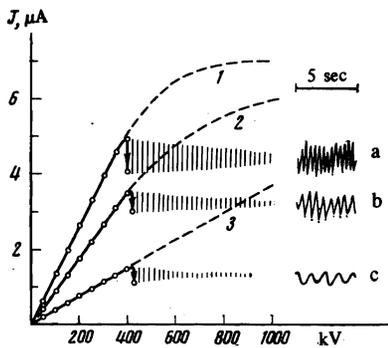


FIG. 3. Current-voltage characteristics of the photocurrent  $J$  following illumination with "white" light (dashed curve) and monochromatic light (solid curve) of intensity  $I/I_0$  equal to 0.25 (curve 1), 0.125 (curve 2), and 0.05 (curve 3). The dashed sections correspond to regions of self-oscillations of the photocurrent (a, b, c).

tals the maximum of the spectral sensitivity of the PC lies in the range  $E = 1.2-1.4$  eV. Thus, for example, for the sample  $\text{CdCr}_2\text{Se}_4$  (0.037% Ga) the maximum of the spectral sensitivity corresponds to an energy  $E = 1.4$  eV (8800 Å). Figure 3 shows the current-voltage characteristics for this crystal under monochromatic illumination  $\lambda = 8800$  Å for different intensities. The dashed lines in this figure correspond to the photocurrent following illumination with "white" light of the same intensity as the monochromatic light. It is seen that in contrast to the "white" light the monochromatic light leads to an abrupt jumplike interruption of the photocurrent in fields  $E \sim 400-420$  V, and self-oscillations of the photocurrent take place, the amplitude and frequency of which depend on the intensity of the light and on the magnitude of the electric field. The regions of the self-oscillations at different intensities are shown in Fig. 3 (a, b, c). The frequency of the self-oscillations varied from crystal to crystal and averaged  $\sim 10$  Hz.

To ascertain the causes of the differences in the behavior of the current-voltage characteristic under "white" and monochromatic illumination, we investigat-

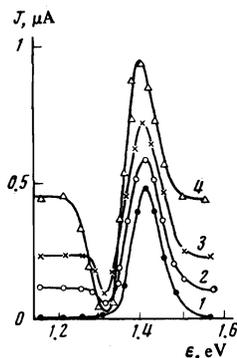


FIG. 4. Spectral dependence of the photocurrent  $J$  on the energy in the presence of constant monochromatic background illumination ( $\lambda = 8800$  Å) of intensity  $I_{\text{thr}}$  ( $U_{\text{thr}}/I_0$  is equal to zero (curve 1), 0.05 (curve 2), 0.125 (curve 3), and 0.25 (curve 4)). Sample  $\text{CdCr}_2\text{Se}_4$  (0.037% Ga). Field  $E \sim 50$  V,  $I_0 \sim 10^{-2}$  W/cm<sup>2</sup>,  $T = 77$  K.

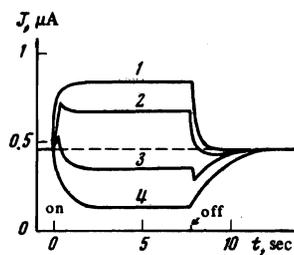


FIG. 5. Time dependences of the photocurrent  $J$  following turning on and off light with wavelengths  $\lambda = 8800, 8950, 9100,$  and  $9300$  Å (curves 1, 2, 3, and 4, respectively) at a constant background illumination  $\lambda = 8800$  Å.

ed the PC with simultaneous illumination of the samples from two independent sources of monochromatic light. One source served as constant illumination at a wavelength  $\lambda = 8800$  Å coinciding with the maximum of the spectral sensitivity of the PC. The wavelength of the second source varied in a wide range. We investigated the spectral dependence of the photocurrent on the intensity of the constant illumination. In the absence of illumination, the PC dispersion curve has the usual form with a maximum spectral sensitivity near 1.4 eV (Fig. 4, curve 1). In the presence of the illumination, we observed in addition to the maximum of the spectral sensitivity, an anomalous decrease of the illumination photocurrent near 1.3 eV (9400 Å) (curves 2, 3, 4). Similar dispersion curves were observed also in the case when "white" background illumination was used in place of the monochromatic illumination. Effects of similar type are known in the literature as effects of infrared quenching of the photoconductivity.<sup>[6]</sup> In the investigation of this phenomenon it was noted that the stationary state is established following turning the light on or off after a rather long time interval (tens of seconds).

Figure 5 shows the time dependences of the photocurrent following turning on and off the light of the second sources with fixed wave lengths equal to 8800, 8950, 9100, and 9300 Å (curves 1, 2, 3, 4) at a constant background illumination  $\lambda = 8800$  Å,  $I/I_0 = 1$  ( $I_0 \sim 10^{-2}$  W/cm<sup>2</sup>).

We see that the shapes of the photocurrent curves differ significantly after light of different wavelengths is turned on and off. The time necessary to establish the stationary state is as a rule several times smaller for the positive photocurrent than for the negative photocurrent. At the wavelength at which the polarity of the photocurrent is reversed, the curves have a complicated character and the transient curves have both fast and slow components.

Without going into details of the explanation of the effective infrared quenching of the photoconductivity, we note that it is explained by resorting to various representations of the local disturbances to the energy band structure of the medium, wherein the fundamental band scheme is supplemented with a large set of various levels (donor, acceptor, sticking centers for the photocarriers, traps, excitons, etc.). Optical excitation of several levels causes these levels to become active

centers of photocarrier recombination in a number of cases. The increase in the recombination rate, which leads to a decrease of the effective lifetime of the carriers, serves as the basis of the notions concerning the infrared quenching of the photoconductivity.

It is possible that the observed infrared quenching of the photoconductivity in  $\text{CdCr}_2\text{Se}_4$  can be connected with analogous notions, but its detailed description calls for a detailed study of the energy band structure of  $\text{CdCr}_2\text{Se}_4$ . Investigations of the photo-induced change in the FMR parameters under monochromatic illumination were hindered by the low intensity of the light. The above-described increase of the shift  $\delta H$ , in the presence of an electric field had made it possible to observe photo-induced effects also under monochromatic illumination. The investigations have shown that the region of the spectral sensitivity of the PC and of the photo-induced changes of the FMR field for the investigated  $\text{CdCr}_2\text{Se}_4$  (0.037–0.19% Ga) samples lie in the same wavelength interval with a maximum sensitivity near 1.4 eV at  $T = 77^\circ\text{K}$ .

Just as in the case of the PC, the illumination of the samples with light of wavelengths 9440 Å leads to a decrease of the photo-induced shift of the FMR field, due to the constant illumination  $\lambda = 8800$  Å (infrared quenching of the photo-induced shift of the FMR field).

We note, however, that in contrast to "white" light, illumination with monochromatic light of the same intensity leads to a much smaller increase of the shift  $\delta H$ , in an electric field. The apparent reason is that the interruption of the photocurrent under monochromatic illumination limits the region of the existence of the nonlinear section of the current-voltage characteristic (Fig. 3) on which, as shown above, the main increase of  $\delta H$ , takes place.

#### 4. DISCUSSION OF EXPERIMENTAL RESULTS

Real changes in the carrier density from  $\sim 10^8 \text{ cm}^{-3}$  to  $\sim 10^9 \text{ cm}^{-3}$ , which took place in the present study, do not make it possible to use the ideas of<sup>[3,10,11]</sup> for the explanation of the results presented above. The low inertia of the observed effects does not agree with the assumption concerning the photoactive centers ( $\text{Cr}^{2+}$  ions).<sup>[4]</sup>

An analysis of the interaction between the magnetoactive atoms of magnetic semiconducting crystals via the nonmagnetic sublattice has shown (see also<sup>[9]</sup>) that various types of elementary site excitations (magnons, polarons, conduction electrons, and holes) can greatly influence the value of the superexchange interaction integral in individual cases. In particular, it was shown that the appearance of holes at the anions in the valence band hinders the electron transition between the magnetoactive atoms through the nonmagnetic sublattice, and can lead to a decrease of the integral of the superexchange interaction. In this case the character of the behavior of the magnetic subsystem should be determined by the character of the behavior of the hole carriers.

Let us consider from this point of view the possible mechanism whereby the electric field influences the increase of the photo-induced changes in the FMR field.

As shown above, the main increase of  $\delta H$ , is observed in electric fields  $E > E_{\text{thr}}$ , in which case the behavior of the photocurrent deviates from Ohm's law. This deviation of the current-voltage characteristics from Ohm's law and the simultaneous appearance of low-frequency oscillations of the photocurrent under monochromatic illumination are similar in character to the analogous anomalies of the photocurrent in the nonmagnetic superconductors  $\text{CdSe}$ .<sup>[12]</sup> It has been shown that the onset of these anomalies in  $\text{CdSe}$  in the field region  $E > E_{\text{thr}}$  is due to injection of holes from the anode, which are captured by the traps in the region near the anode to form additional recombination centers for the photoelectrons. A section with increased recombination rate is thus produced in the region of the anode and leads to an increase of the resistance in this part of the sample and to a nonlinearity of the current-voltage characteristic. Limitation of the current by the anode causes the electrons to accumulate in this part of the section and to form a negative space charge (an electric domain). The dimension of such a domain is determined by the electric field. With increasing field  $E$ , the produced domain increases, and at the instant when it reaches the cathode, low-frequency oscillations of the photocurrent are produced.

As a result of the additional recombination of the photoelectrons with the injected localized hole recombination centers, an increase takes place in the effective lifetime of the holes in the valence band, and consequently the hole-connected change of the integral of the superexchange interaction and the photo-induced shift of the FMR field increase.

For a more detailed interpretation of many results, for example the effect of infrared quenching of the photoconductivity, the behavior of the current-voltage characteristics following illumination with white and monochromatic light, etc., additional information is needed on the dependence of the integral of the superexchange interaction on the concentration of the holes in the valence band, on the presence of injection of holes from the anode in fields  $E > E_{\text{thr}}$ , and on the existence of an electric domain.

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## Size effects in parametric excitation of spin waves in ferrites

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The nonlinear properties of yttrium iron garnet (YIG) ferrite spheres with dimensions comparable with the mean free path of parametrically excited spin waves are investigated at a pumping frequency of 9370 MHz, using a dielectric cavity made of polycrystalline rutile. The damping constant of the spin waves is found to be dependent on the wave vector and on the sample diameter. It is also observed that the imaginary part of the nonlinear susceptibility, the threshold for the appearance of self-modulation of the magnetization, and the hard excitation of spin waves all depend significantly on the efficiency of scattering of the waves by surface inhomogeneities.

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### INTRODUCTION

Most experiments on parametric excitation of spin waves in ferrites were performed on samples whose dimensions greatly exceeded the free path  $l$  of the excited waves.<sup>[1]</sup> As a result, the boundaries of the ferrite did not exert a noticeable influence on the nonlinear properties; in particular, it has been established that the threshold of the parametric excitation of the short-wave spin waves and the associated field damping parameter  $\Delta H_h$  do not depend on the state of the surface, although if we disregard the condition  $l \ll r$ , where  $r$  is the radius of the investigated ferrite sphere, then  $\Delta H_h$  should be strongly influenced by the quality of the surface finish.<sup>[2]</sup>

This paper is devoted to an investigation of the nonlinear properties of a ferrite sphere made of yttrium iron garnet (YIG), the initial radius of which was 0.51 mm. The radius of the sphere was then decreased to 0.26 mm, and ultimately to 0.09 mm. In the latter case, the condition  $l \approx r$  was satisfied. The quality of the surface finish was in all cases the same—the surface was polished with diamond paste with grain dimension smaller than  $l \approx r$ . The measurements were performed at a pump frequency 9370 MHz, in both the pulsed regime (pulse duration 200  $\mu$ sec, pulse repetition frequency 50 Hz) and in the cw regime. To increase the sensitivity of the experimental setup, since we measured nonlinear properties of such small ferrite samples, smaller in volume by more than two orders of magnitude than the customarily employed ferrites, the cavity resonator<sup>[3]</sup> was replaced by an open dielectric resonator of rectangular form, made of polycrystalline rutile.<sup>[4]</sup> This circumstance led also to a considerable lowering of the threshold power of the excitation of the spin waves; for example, in the field  $H_c$ —

the field of the minimal threshold of the spin-wave instability, at which spin waves are excited with a wave vector  $k$  close to zero—the threshold power was of the order of several hundred microwatts. The samples were oriented in such a way that the direction of the easy-magnetization axis [111] coincided with the direction of the constant magnetic field  $H_0$ .

### EXPERIMENTAL RESULTS AND DISCUSSION

1. The results of an investigation of the threshold of the parametric instability in parallel pumping are shown in Fig. 1. For short-wave spin waves in constant magnetic fields  $H_0 < H_c = 1540$  Oe, up to a sample diameter 0.52 mm inclusive, within an experimental accuracy limit  $\pm 0.5$  dB the instability threshold does not depend on the sample diameter. The threshold is appreciably increased, however, for the sample of 0.18 mm diam-

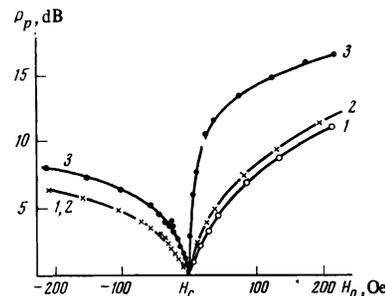


FIG. 1. Dependence of the threshold of parametric-instability excitation threshold on the constant magnetic field  $H_0$  in the case of parallel pumping  $P_p$ . Curves 1, 2, and 3 correspond to YIG spheres 1.02, 0.52, and 0.18 mm in diameter. The absolute value of the threshold field at the point  $H_c$  amounts to 0.27 Oe for curves 1 and 2 and 0.34 Oe for curve 3;  $H_0 \parallel [111]$ .