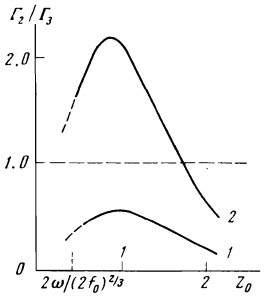

FIG. 3. Ratio of the probability of two-photon to three-photon ionization for the process (37) as a function of the parameter Z_0, calculated from (35). Curve 1—$F = 10^7$ V/cm, curve 2—$F = 5\times10^6$ V/cm.

which shows Γ_2/Γ_3 as a function of $Z_0 = 2\omega/(2f)^{2/3}$ for the process

$$Na^- + n\omega \to Na + e^- \qquad (37)$$

($|E_i| = 3\omega$, $\omega = 947$ cm^{-1}, which corresponds to the CO_2 laser radiation) for different wave fields. When $F \sim 10^6$ V/cm, the increase in n_f occurs in an electric field f still weak enough in comparison with the characteristic atomic field $f_0 = |E_i|^{3/2}$. The effect becomes more appreciable as the ratio $|E_i|/\omega$ increases. The nonmonotonic dependence of n_f on f will probably be found for neutral atoms as well.

We are indebted to V. S. Vinogradov for discussions of impurity EA in solids.

[1)]We note that Prodan and Rozneritsa[15] considered two-photon EA, but their results are subject to computational errors.

[2)]It is well known that this quantity determines the van der Waals constants in systems containing the ion. When $f = 0$, the formula given by (33) becomes identical with the expression for the polarizability of a negative ion given in[17].

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Translated by S. Chomet

Optical orientation in ferromagnetic europium sulfide

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(Submitted April 27, 1976)
Zh. Eksp. Teor. Fiz. **71**, 2068–2076 (December 1976)

Results are presented of a study of optical spin orientation in a ferromagnetic semiconductor. The experimental results are interpreted in terms of a unified model which takes into account the effect of illumination on both the temperature and the Curie point of the spin system. Experiment shows that the Curie temperature of the magnetic semiconductor is raised by illumination.

PACS numbers: 75.50.Dd, 75.30.La, 78.20.Ls

1. Optical spin orientation has by now been observed in a large number of paramagnetic semiconductors.[1] The effect of unpolarized light on a ferromagnetic medium has been investigated by several workers (see, for example,[2]). In this case, illumination gives rise to a change in the magnetic permeability of the medium. In contrast to previous experiments,[2] illumination by circularly polarized light produces optical magnetization of a demagnetized ferromagnet.[3,4] In this paper, we present the results of an investigation into the optical spin orientation in europium sulfide.

Europium sulfide is a magnetic semiconductor. Studies of the optical spin orientation in such compounds are of particular interest. From the standpoint of optical spin pumping, a ferromagnetic is the very opposite of a

gaseous medium (in which optical pumping was observed first). In gases, the spins of the individual atoms interact with one another only at the time of collision, whereas, in a ferromagnetically ordered spin system, the spins of the individual electrons are coupled by a strong exchange interaction. A system of this kind may exhibit an essentially new effect in which magnetization by light is not due to the direct momentum transfer from a photon to the spin system. Circularly polarized light may act as an external disturbance defining only a special direction along which the spins will be oriented as a result of the exchange interaction. The optical pumping effect may turn out to be quite substantial when light is used as a controlling agent in this way.

2. Let us consider the possible orientation mechanisms in europium sulfide. The energy spectrum of the electron states in europium sulfide is the sum of delocalized and localized states.[6] The former comprise states in the conduction and valence bands, formed as a result of the strong overlap of s, d, and p shells of the europium and sulfur atoms. The localized states include the states of electrons in the $4f$ shells of the europium ions Eu^{+2} ($^8S_{7/2}$). The overlap of the wave functions for the $4f$ shells of neighboring ions is small and, consequently, the bound-electron approximation provides a sufficiently acceptable description of the state.[6] The exchange interaction leads to the ordering of the spins of the $4f$ electrons so that, for temperatures $T < T_c \approx 16.2$ °K, the europium sulfide becomes a ferromagnet.

The edge of the optical absorption band of europium sulfide is connected with the transition of electrons from the $4f$ shells to the conduction band, i.e., $4f^7(^8S_{7/2}) \rightarrow 4f^6(^7F_J)5d(t_{2g})$.[7] Analysis of the selection rules for this process[9] has shown that the probability of the optical transition depends on the polarization of the absorbed light and the orientation of the electron spin in the initial and final states. In particular, the most probable transitions under illumination by circularly polarized light are those from levels for which the component of the resultant angular momentum ($J = \frac{7}{2}$) in the direction of propagation of the incident radiation is opposite in sign to the momentum of the photon. The depletion of these levels should lead to partial orientation in the spin system of the $4f$ shells of the Eu^{+2} ions. At the same time, there should be partial orientation of the spins of electrons released into the conduction band. The stationary values of the spin orientation of $4f$ shells and band electrons are determined by the ratio of pumping and relaxation rates. Thus, in paramagnetic europium sulfide, the orientation of localized and delocalized electrons by light is analogous to the optical pumping of the ground and excited states in a gas.

In ferromagnetic europium sulfide, the spins of the $4f$ shells of neighboring Eu^{+2} ions are practically parallel so that, on the microscopic scale, the spin orientation is nearly complete. However, in a demagnetized specimen, whose volume is divided into a large number of differently oriented domains, the macroscopic average spin orientation is zero. If we proceed from the individual paramagnetic atoms to the ferromagnetic domains, we readily see that, because of strong circular dichroism in the domain, the absorption of circularly polarized light should lead to partial macroscopic spin orientation in the initially demagnetized specimen. However, in addition to the above direct effect of light on level populations in the ferromagnet, there are certain additional optical magnetization mechanisms. They are connected with the strong exchange interaction between the $4f$ shells of the Eu^{+2} ions.

When the energy of an absorbed photon is low enough, the transfer of an electron to the conduction band may result in a transition of the $4f$ shell from the magnetic to the nonmagnetic state ($^8S_{7/2} \rightarrow {}^7F_0$). The reduction in the concentration of magnetic ions leads to a weakening of exchange (the effective Weiss field is proportional to the concentration of magnetic ions). The simultaneous increase in the concentration of electrons in the conduction band leads to an increase in the molecular field (indirect exchange[10]). Since the Curie temperature is proportional to the Weiss field, we may conclude that the appearance of electrons in the conduction band and of holes in the $4f$ level gives rise to a change in T_c. Because of strong dichroism, the absorption of circularly polarized radiation depends on the orientation of the magnetization of the domain and, consequently, the change in T_c will be different in differently oriented domains. In the neighborhood of the Curie point, the magnetization of a domain is very dependent on the difference between the temperature T and T_c. This is responsible for the fact that illumination of a demagnetized multidomain specimen by circularly polarized light should produce a resultant magnetization.

We note that the recombination of a conduction-band electron with an unoriented hole in the $4f$ level may give rise to the appearance of an unoriented Eu^{+2} ion with a certain finite probability. An unoriented spin of this kind in a ferromagnetic lattice is a packet of magnons. Hence, the size of this packet is close to the lattice constant, and the characteristic wave vector of the spin excitations is large, so that the packet rapidly spreads throughout the volume of the crystal. The creation of a large number of high-energy magnons should lead to an increase in the temperature T of the system. The magnon temperature will also increase as a result of the energy relaxation of electrons in the conduction band. The resultant effect of light on magnetization is thus determined both by the change in T_c and the change in T.

Finally, one further specific orientation mechanism connected with the redistribution of domain volumes is possible in a multidomain specimen. Thus, as soon as the absorption of light gives rise to a change in exchange energy, and, in the case of circularly polarized incident radiation, this change depends on the domain orientation, the free-energy density in differently oriented domains turns out to be different. The domain structure is determined by the free energy minimum condition.[5] This means that the volumes of domains with high exchange energy should increase, whereas the volumes of domains with lower energy should decrease. The orientation-dependent change in the domain volume should, when averaged over a large number of domains, result in an additional orientation of the spin system.

3. The magnetization produced by light can be detected in various ways.[3,4,12] The results reported below were obtained by measuring optically induced circular dichroism (OICD), using the apparatus described previously.[3] The specimens were in the form of europium sulfide films deposited on a transparent substrate. The specimen thickness d was 5×10^{-5} cm with $d\sim1/K$, where K is the light absorption coefficient. The specimen was placed in a helium cryostat, and the estimated temperature in the illuminated region was found to be 10–12 °K. Under these conditions, and for a pumping rate of 10^{25} cm$^{-3}\cdot$ sec^{-1}, the signal due to the optically induced circular dichroism was found to be equal in magnitude to the dichroism in an external field of 1 Oe (~0.01%).

An increase in the substrate temperature by 4 °K led to a reduction in the effect by not less than two orders of magnitude (below the limit of sensitivity of the apparatus). Additional measurements at liquid-nitrogen temperatures, using digital signal storage, showed reliably that the magnitude of the effect under these conditions was lower by at least three orders of magnitude than the initially observed effect. This enables us to conclude that the observed strong OICD is connected with ferromagnetic ordering of the spins of the Eu^{+2} ions. We also investigated experimentally the effect of an external magnetic field ($H_{ext}\lesssim2$ kOe) on the optical pumping process. The magnetic field was perpendicular to the direction of propagation of the pump radiation, and was found to weaken the effect, but the interpretation of this phenomenon in the case of a ferromagnetic specimen was essentially different from the usual explanation of the Hanle effect.[15] The dependence of the induced circular diochroism on a magnetic field parallel to the pump axis is shown in Fig. 1. It is clear from this graph that the effect decreases with increasing external magnetic field and, when the latter is of the order of 1.3 kOe, the effect changes sign. Further increase of H_{ext} gives rise to a rapid increase in the effect. It is important to note that the external magnetic field did not exceed 2 kOe, i.e., the magnetization of the film did not exceed 20% of the saturation value. The dependence of the sign of the effect on the sign of H_{ext} is connected with the symmetry of the experimental arrangement which was designed to measure the difference between circular dichroism due to illumination by right- and left-handed circularly polarized radiation. The change in the sign of the OICD in a relatively weak magnetic field has no analog in the optical orientation of paramagnetic media.

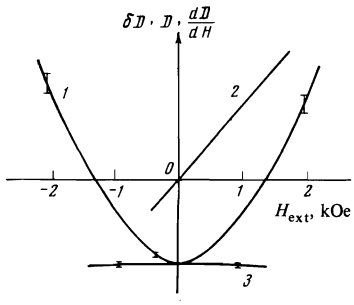

FIG. 1. Optically induced circular dichroism $\delta D(H)$ (1), magnetic circular dichroism $D(H)$ (2), and the derivative $dD(H)/dH$ (3) as functions of the external magnetic field H parallel to the light beam. Vertical bars on experimental curves indicate the noise level.

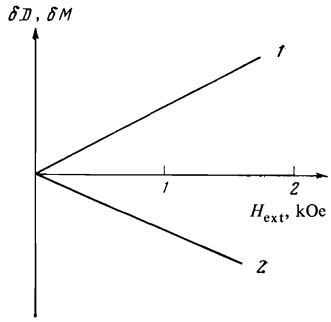

FIG. 2. Change in magnetic circular dichroism (1) and magnetization (2) produced by unpolarized illumination as functions of the external magnetic field.

The same experimental arrangement and the same wavelength of the helium-neon laser (0.63 μ) were used to determine the magnetic circular dichroism and its magnetic-field derivative as functions of the external magnetic field (curves 2 and 3). Comparison of the curves in Fig. 1 shows that the observed field dependence of optically induced circular dichroism is specific for optical magnetization.

4. To explain the experimental results, we must first establish the relationship between optically induced circular dichroism and optically induced magnetization. To do this, we investigated the effect of unpolarized radiation on magnetization and circular dichroism of ferromagnetic europium sulfide in a constant magnetic field (Fig. 2). It is clear from these results that the magnetization decreases under illumination but, despite this, circular dichroism is found to increase. This apparently paradoxical result is explained by the particular spectral dependence of circular dichroism in europium sulfide.

The spectral dependence of magnetic circular dichroism established for our specimens was similar to that reported by Ferre.[8] For wavelengths near 0.63 μ, dichroism falls sharply as the photon energy is increased. At the same time, it is known that, as the temperature is reduced, the optical absorption edge of europium sulfide undergoes a "red shift" connected with the increase in ferromagnetic order in the domain. An increase in the microscopic magnetization (in a domain) is thus found to be accompanied by a reduction in dichroism for $\lambda=0.63$ μ although the magnetization of the specimen as a whole (due to the reorientation of the domains) leads to an increase in dichroism.

The change in the macroscopic mean dichroism $\bar{D}$ and magnetization M observed experimentally can be written in the form

$$\delta\bar{D}=\bar{D}\left[\frac{1}{D}\frac{\partial D}{\partial M_0}\delta M_0+\frac{\delta n}{n}\right],\quad \delta M=M\left[\frac{\delta M_0}{M_0}+\frac{\delta n}{n}\right], \tag{1}$$

where D and M_0 are, respectively, circular dichroism and magnetization of an individual domain, and n is the average projection of the unit vectors lying along the

magnetization in each domain along the direction of propagation of the incident light. It is clear that an increase in n (magnetization of the specimen on the macroscopic magnetization curve) leads to a simultaneous increase in both M and D. At the same time, when $\partial D/\partial M_0 < 0$, the increase in the microscopic magnetization M_0 leads to an increase in M and a reduction in D. In our case, stationary application of unpolarized light reduces largely to the heating of the specimen. We then have $\delta n \approx 0$, $\delta M_0 < 0$, so that $\delta M < 0$ and $\delta D > 0$, and this is, in fact, observed in practice.

5. Let us now analyze the results on the effect of a longitudinal magnetic field on optical pumping (Fig. 1). The positive sign of the OICD in strong magnetic fields corresponds to an increase in the light absorption coefficient for light with right-handed circular polarization in the case of a circularly polarized pump. For an individual domain, this means that the absorption of light is accompanied by an increase in the dichroism, and this is in complete agreement with the results of experiments on unpolarized illumination (Fig. 2). As shown in Sec. 4, the magnetization of the domain is then reduced.

The negative sign of OICD in a weak magnetic field, which corresponds to a reduction in dichroism and an increase in the domain magnetization under illumination, would appear to be in conflict with the results of experiments with unpolarized illumination. It will be shown below that, in low fields, the effect of light with modulated polarization is not reduced to heating. In this case, it is possible to isolate experimentally the nonthermal effect of the pump, i.e., the increase in domain magnetization connected with the change in Curie temperature. The increase in T_c when light excites electrons into the conduction band was first considered in[10] but, so far, this effect has not been observed experimentally.[11]

Let us illustrate the foregoing discussion by a simple calculation. Let us suppose that the film is subdivided into a large number of domains, the magnetization of which is perpendicular to the plane of the specimen. Because of strong circular dichroism, the light absorption coefficient K^i is strongly dependent on polarization and the orientation of domain magnetization relative to the direction of propagation of the incident radiation:

$$K^{(i)} = K_1 - K_2 n_i \sigma,$$

where K_1 is the polarization-independent part of the absorption coefficient, K_2 is the coefficient representing the magnitude of circular dichroism, $n_i = +1$ when the direction of magnetization is parallel to the direction of propagation, $n_i = -1$ in the opposite case, and $\sigma = \pm 1$ represents right- and left-handed circular polarizations. In the case of unpolarized or plane-polarized light, we have $\sigma = 0$. For a demagnetized specimen, the dichroism averaged over a large number of domains is zero and $K = K_1$. For a partially magnetized specimen,

$$K(\sigma) = K_1 - K_2 n \sigma,$$

where n is the mean domain orientation, which can be simply expressed in terms of the mean magnetization M, i.e., $n = M/M_0$, and M_0 is the limiting value of the domain magnetization. Thus, dichroism turns out to be proportional to magnetization:

$$K_{(+1)} - K_{(-1)} = -2K_2 M/M_0.$$

We shall assume below that the absorption of light in a domain is uniquely related not only to the direction but also to the magnitude of magnetization. We shall also assume that the absorption of light leads only to a change in the magnetization and this change may be connected both with a change in the temperature of the system and a change in the exchange Weiss field. In the neighborhood of the Curie point, the magnetization depends on the difference between T and T_c, i.e., $M_0 = M_0(T - T_c)$, so that the rate of change of magnetization under illumination is given by

$$\frac{dM_0}{dI} = \frac{dM_0}{dT}\left(\frac{dT}{dI} - \frac{dT_c}{dI}\right) \tag{2}$$

where I is the intensity of the pump. The change in T_c is connected with the transport of electrons from the $4f$ levels of ions to the conduction band, and the increase in T_c due to indirect exchange predominates over the reduction in the Curie temperature due to the weakening of direct exchange. The Coulomb attraction of electrons to holes produced by light in the $4f$ shells of europium (Eu^{+3}) ensures that the excited electrons are localized near the holes at distances of the order of the screening length r_D. Under the conditions prevailing in our experiment, in which the concentration of electrons in the conduction band was up to 10^{18} cm^{-3}, we had $r_D \sim 10^{-6}$ cm, which was much less than the linear dimensions of a domain ($l \sim 10^{-4}$–10^{-5} cm[13]), and we may therefore conclude that the change in the Curie temperature had a local character and was subject to discontinuous variations between the domains. Under these conditions, we have for each domain illuminated by light

$$\frac{dT_c}{dI} = \delta\tau(K_1 - K_2 n_i \sigma), \tag{3}$$

where τ is the electron recombination lifetime, $\delta = J_{sf}S/2$ represents the enhancement of direct exchange resulting from the increase in the electron concentration in the conduction band, J_{sf} is the exchange constant, and S is the mean spin of the $4f$ shells of europium ions.

The temperature of the spin system is determined by the relationship between the heat released as a result of light absorption and the heat-transfer conditions. Since the linear dimensions of the domains are very small ($l \sim 1$ μ), one may suppose that, very approximately, the specimen is at a temperature T which is average over all the domains and is determined by the mean absorption coefficient K:

$$dT/dI = aK, \tag{4}$$

where $a = B/C$ is the ratio of the heat released by the absorption of one photon to the thermal conductivity of the wall separating the system from the ambient medium.

Using (3) and (4), we can readily obtain the expression for the rate of change in the light absorption coefficient of the ferromagnetic film under the action of incident radiation:

$$\frac{dK(\sigma_2)}{dI(\sigma_1)}=\frac{dK_1}{dM_0}\frac{dM_0}{dT}(K_1-K_2n\sigma_1)\left(\frac{B}{C}-\delta\tau\right)$$
$$-\frac{dK_2}{dM_0}\frac{dM_0}{dT}\sigma_2\left[\left(\frac{B}{C}-\delta\tau\right)K_1n-\left(\frac{B}{C}n^2-\delta\tau\right)K_2\sigma_1\right]; \quad (5)$$

where n is the mean orientation of the domain, M_0 is the limiting magnetization in the domain, K_1 and K_2 are the polarization-dependent and polarization-independent parts of the absorption coefficient, respectively, σ_1 is the polarization of the pump, and σ_2 is the polarization of the probing radiation.

Let us begin by considering how unpolarized illumination affects the absorption of unpolarized light.[14] It is readily seen that

$$\frac{dK(0)}{dI(0)}=\frac{dK_1}{dM_0}\frac{dM_0}{dT}\left(\frac{B}{C}-\delta\tau\right)K_1. \quad (6)$$

Because of the red shift of the absorption band, we have $dK_1/dM_0>0$. It is well known that an increase in temperature is accompanied by a reduction in the saturation magnetization, so that $dM_0/dT<0$. Thus, the sign of the change in the absorption coefficient for unpolarized light in the case of unpolarized illumination turns out to be unambiguously related to the ratio of the changes in T and T_c. When heating predominates over the increase in the Curie temperature, so that $B/C>\delta\tau$, the absorption coefficient is reduced by illumination. On the other hand, when the increase in T_c predominates ($B/C<\delta\tau$), there is an increase in the absorption: $dK(0)/dI(0)>0$. In the experiments with stationary (or modulated at low frequency $\omega\sim100$ Hz) unpolarized illumination, the absorption coefficient is found to be reduced by illumination, i.e., under these conditions, the temperature increases more than T_c.[1)]

The situation is qualitatively different when linearly polarized pump radiation is modulated in intensity at a high frequency ($\omega\sim10^6$ sec^{-1}). In this case, the thermal effect of light is substantially weakened by inertia, and experiment shows the presence of an enhancement (at the frequency of the perturbation) of absorption of light from the probing laser. We note that delayed effects (connected, for example, with the recombination lifetime of the electron) cannot produce a shift in the response to the perturbation by more than $\pi/2$, i.e., they cannot give rise to a change in the sign of the effect. This experiment thus has a unique interpretation, i.e., the enhancement in absorption of the probing light is connected with the "red" shift of the absorption edge under the action of the pump.

Let us now consider the effect of an unpolarized pump on the magnitude of circular dichroism:

$$\frac{dK(+1)}{dI(0)}-\frac{dK(-1)}{dI(0)}=-2\frac{dK_2}{dM_0}\frac{dM_0}{dT}\left(\frac{B}{C}-\delta\tau\right)K_1n. \quad (7)$$

Proceeding by analogy with the above discussion and noting that, for the wavelengths which we have used in our experiments, $dK_2/dM_0<0$, we find that dichroism should increase under illumination by unpolarized light provided $B/C>\delta\tau$. Comparison of (6) and (7) will show that the signs of the changes in absorption and dichroism are always opposite under illumination. This is, in fact, confirmed by experiment. Absorption is found to decrease and dichroism to increase under steady illumination. When the pump is modulated at high frequency, so that the thermal effect is reduced, it is found that absorption is enhanced by illumination whereas dichroism is reduced.

Finally, let us consider the experiment on optically induced circular dichroism. In this experiment, one detects the difference between dichroisms induced by light with right- and left-handed circular polarization:

$$\left(\frac{dK(+1)}{dI(+1)}-\frac{dK(-1)}{dI(+1)}\right)-\left(\frac{dK(+1)}{dI(-1)}-\frac{dK(-1)}{dI(-1)}\right)$$
$$=4\frac{dK_2}{dM_0}\frac{dM_0}{dT}\left(\frac{B}{C}n^2-\delta\tau\right)K_2. \quad (8)$$

We recall that n is the mean orientation of the domains in the magnetic field. It is clear from this expression that the experiment on OICD can be used to separate the contribution of the heating of the system from the change in the Curie temperature. In the case of a highly magnetized specimen $n>(\delta\tau C/B)^{1/2}$ and heating is the predominant effect whereas, for a demagnetized specimen $n\approx0$ and the entire effect is determined by the change in the Curie temperature in differently oriented domains. Thus, OICD in a demagnetized specimen can be used to isolate the change in magnetization connected with the change in the Curie temperature. Analysis shows that T_c increases under illumination, i.e., the increase in indirect exchange predominates over the reduction in direct exchange in the Weiss field. Experimental estimates show that T_c increases by an amount of the order of 0.1 °K, which corresponds to an electron concentration in the conduction band of the order of 10^{18} cm^{-3}.

To check the conclusion that the parabolic dependence of OICD on the external magnetic field is connected with the average heating of the specimen, and that the negative sign of OICD in the case of low n is due to localized effects in the region of the light spot, we carried out an experiment in which the pump and probing beams were separated in space (Fig. 3b). Curve 1 in this figure corresponds to coincident beams and curve 2 to a partial overlap of the light spots. Curve 3 corresponds to the case where the pump and probing beams were separated in space by 1 mm. It is clear that OICD in a fully demagnetized specimen is observed only when the light spots associated with the pump and probing beams overlap. For a magnetized specimen, the effect is observed even when the spots are separated to distances exceeding their linear dimensions by a substantial factor. We note that a simple reduction in pump intensity (Fig. 3a) does not affect the overall situation.

6. Let us now summarize briefly the foregoing discussion. All the experimental data can be explained in terms of a unified model. The observed optical spin pumping in a ferromagnetic semiconductor is connected with the orientation of the ground state of the Eu^{+2} ions

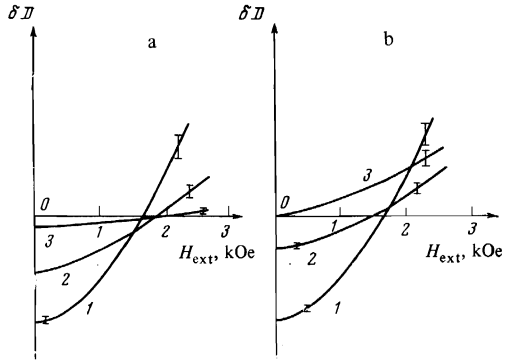

FIG. 3. Optically induced circular dichroism as a function of external magnetic field for: a—successive reduction (1, 2, 3) of the intensity of the pump (light spots of the magnetizing and probing beams coincident), b—successive increase in the separation between the light spots (beam intensities constant). 1—light spots overlap ($l = 0$), 2—light spots partially overlap ($l = 0.1$ mm), 3—light spots separate ($l = 1$ mm). Vertical bars indicate noise levels.

responsible for the absorption of light. The possibility of optical magnetization is connected with the different absorption of polarized light in differently oriented domains. The effect of the light reduces to both the heating and the reduction in the Curie temperature of the spin system.

It has been shown experimentally that the steady application of unpolarized light reduces largely to the heating of the spin system. In the case of high-frequency modulation of the light intensity, the experimental data can be unambiguously interpreted as the enhancement in magnetization under illumination (the increase in the Curie temperature due to indirect exchange predominates over the increase in the temperature of the spin system because of the inertia of thermal effects).

The method of double polarization modulation which was used to observe optically induced circular dichroism can also be used to isolate the nonthermal contribution of the effect of the illuminating radiation. The change in the sign of OICD when the magnetic field is parallel to the light beam shows that the increase in magnetization accompanying the enhancement of indirect exchange through photoexcited electrons predominates over its reduction associated with the transition of a proportion of the Eu^{+2} ions to the nonmagnetic Eu^{+3} state during the absorption of light. Thus, two independent experiments have confirmed the enhancement of magnetization by light predicted by Vonsovskiĭ, Samokhvalov, and Bersyshev.[10]

The authors are indebted to B. P. Zakharchenya and V. G. Fleĭsher for their constant support and attention, to I. Ya. Korenblit and V. I. Perel' for valuable suggestions and discussions, and to I. A. Smirnov who stimulated our interest in optical orientation in magnetic semiconductors and provided us with the specimens for these experiments.

[1)]It might have been thought that the reduction in the absorption coefficient is connected not with the red shift and the change in the temperature of the spin system but with the reduction in the concentration of electrons in the 4*f* shells. However, estimates show that the magnitude of this effect is smaller by at least an order of magnitude than that of the effect discussed above.

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Translated by S. Chomet