Magnetization of a garnet film through a change in its multidomain structure under circularly polarized light

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An effect of photomagnetization of a $(Tm_{2.4}Bi_{0.6})(Fe_{3.9}Ga_{1.1})O_{12}$ garnet film at $T = 300$ K caused by a change in the film multidomain structure is discovered. The photomagnetization (ΔM) is induced by the action of a circularly polarized pulsed lasing (power $P < 600$ kW/cm^2, pulse duration $\tau_i \sim 7$ ns) in a wavelength range of 450 nm to 600 nm. The effect is measured vs the power and polarization of light and vs the magnetic field perpendicular to the film surface. Without a magnetic field the value of ΔM is the maximum for the circular polarization of light (0.1 G $< \Delta M <$ 1 G at a power of 300 kW/cm^2). With a change of the polarization direction ΔM changes sign (for linear polarization $\Delta M = 0$). Our study of photomagnetization in external magnetic fields and observation of the film domain structure behavior led us to the conclusion that the effect is unambiguously attributed to a change in the multidomain structure of film by the action of light.

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I. INTRODUCTION

High intensity optical radiation is known to change the magnetic properties of both nonmagnetic and magnetic crystals. The effects arising from such an impact are conventionally distinguished as optomagnetic (OM) and photomagnetic (PM) effects. The latter are essentially related to absorption of the optical radiation energy, this being their principal difference from the OM effects (see Refs. 1 and 2 and references therein). A characteristic example of OM is the inverse Faraday effect (IFE), basically, the action of a circularly polarized light (CPL) propagating in a *transparent medium* on this medium, such as would be induced by an effective magnetic field, i.e., causing a change in the medium magnetization. A theoretical analysis of IFE was done by Pitaevsky[3] (see also Ref. 4) and Pershan.[5] At present the IFE is being actively and successfully explored as a technique for nonthermal ultrafast (by femtosecond pulse laser) modification of the magnetization of films (orthoferrites, ferrimagnetic garnets)[2,6–10] (see also Refs. 11 and 12), ferrimagnetic amorphous alloys,[13] and antiferromagnetics[14] (see also Refs. 15 and 16. According to Refs. 2 and 6–9, this method shows good promise for practical applications such as magnetic storage technology, spin electronics, and quantum computing.

As for the PM effects in magnetic crystals, the research interest here is apparently not limited to mere heating of a crystal by optical radiation. A fairly complete analysis of the theoretical and experimental work carried out on these PM effects by mid-1980s is offered in the overview.[1] At present the research in this area is quite active. Of keen interest, in our opinion, are the works[17,18] that report observation of the domain walls displacement in $(YCa)_3(FeCoGe)_5O_{12}$ epitaxial films under the impact of linearly polarized laser light; the laser beam (light spot radius of 50 μm) was focused on the film surface in the spatial region containing a domain wall. The authors attribute this displacement effect to the photoinduced changes in the magnetic anisotropy.

We believe that the most interesting PM effects occur in magnetics under CPL. An example of such phenomena is the experimentally studied magnetization of a film of ferrimagnetic amorphous alloy CdFeCo by CPL[19] (see also the references therein). The authors explain this effect by a complex impact of the CPL on a test sample: the CPL both heats up the illuminated magnetized area and also affects it as a magnetic field corresponding to IFE.

Among the PM effects in magnetics of particular interest are those arising in polydomain magnetics by CPL (PMPM by CPL), when a beam aperture largely exceeds the dimensions of the magnetic domains. We will further denote the PM effects in polydomain magnetics as PMPM. It is worth remembering here that in the absence of an external magnetic field a magnetic at thermodynamic equilibrium has, due to its polydomain structure, zero total magnetization. In this situation the polydomain structure in PMPM by CPL can, in principle, play a double role. On one hand, the CPL-induced changes of magnetization in the adjacent domains may be different, which is likely to lead (given sufficiently strong pinning of the domain walls) to overall magnetization of the sample. On the other hand, the CPL may cause a *change* in the polydomain structure of sample, such that the volumes of the adjacent domains differing in the magnetization direction become unequal. This may also lead to overall magnetization of the sample. The PMPM by CPL was observed in ferromagnetic semiconductors EuS[20,21] and $CdCr_2Se_4$.[22,23] In Ref. 24 a discussion of these effects in EuS ended up in a suggestion, apparently for the first time in accordance with Ref. 1, that this effect may arise from a *change* in the polydomain structure of sample. The idea essentially is that in illumination of this ferromagnetic semiconductor with CPL, due to circular dichroism in the domains with oppositely directed magnetizations, creates different concentrations of photoelectrons. Hence, the intensity of exchange interaction that depends on photoelectron concentration (see Ref. 1) and, consequently, the exchange energy density become different in the domains with the oppositely directed magnetizations. This causes an increase in the volume of the domains with a higher intensity of exchange interaction and, correspondingly, a decrease in the volume of the domains in which this intensity is lower. In other words the CPL works here as some effective magnetic field. Note, though, that the nature of this field

differs from the magnetic field related to the IFE. A detailed theoretical study into this mechanism (PMPM by CPL), which is likely to take place not only in ferromagnetic semiconductors but also in ferrimagnetic garnets (for example, Yttrium Iron Garnet (YIG)), was conducted in Refs. 25–28. In ferrimagnetic garnets, as shown in Ref. 27, the change in the intensity of exchange interaction is conditioned by the light-induced modification of the electron structure of magnetic ions Fe^{3+} (according to Ref. 1, here we deal with excitation of Fe^{3+} related excitons). V. F. Kovalenko *et al.*[1] provide a thorough discussion of these works, specifying the validity criteria for the method employed. Besides, in Ref. 1 they consider another scenario of PMPM by CPL, which was proposed in Ref. 29 (see Ref. 26 as well). By this mechanism (also involving the circular dichroism) the factors taken into account are the changes in the constant of only the inhomogeneous exchange by CPL. It should be noted for completeness that a *change* in the polydomain structure may also occur through IFE (i.e., without absorption of CPL) for a magnetic in the polydomain state.[30] In Ref. 30 this effect was evaluated for the ferromagnetic semiconductor EuO. We would like to point out that the approach used therein can be modified, taking into account the absorption process. Strictly speaking, in this case the CPL-induced magnetization cannot be classified as an IFE effect. They do, however, use the term "inverse Faraday effect" in a broader sense (to define the effect of magnetization by CPL in nontransparent media as well). Note that, as follows from Ref. 30, a *change* in the multidomain structure through IFE is, in principle, possible for any magnetic being in the multidomain state and featuring a good mobility of the domain walls.

Which of the previous mechanisms is responsible for the effect of PMPM by CPL and to what extent remains an open question. According to the theory in Refs. 25–30, the maximum value of the effect would be expected from the scenario described in Refs. 25–27. It is obvious, though, that prior to answering the previous question we have to establish, through specific experiments, the mere fact that PMPM by CPL is related exactly to the change in the polydomain structure. However, interpreting of the experimental results[20–23] as manifestation of PMPM by CPL that is solely related to a *change* in the sample polydomain structure was only tentative for the lack of data on the latter. Thus, we might suggest that the changes in the magnitude of magnetization in the domains with oppositely directed magnetizations are different because of the circular dichroism. Such changes could, as mentioned previously, lead to the CPL-induced PMPM by a mechanism other than a change in the polydomain structure.

So far there have been no reports on observation of PMPM effects by CPL that can without ambiguity be related with a *change* in the polydomain structure of samples. Our paper deals with exactly this issue and provides a description of such effects. For a sample we used a magnetic film.

II. EXPERIMENTAL RESULTS AND DISCUSSION

It is obvious that the effect of PMPM by CPL when the light is directed perpendicular to a film surface (see Ref. 25) is best observed in a thin film with a large perpendicular anisotropy (such as the bubble garnet film[31]). In this case

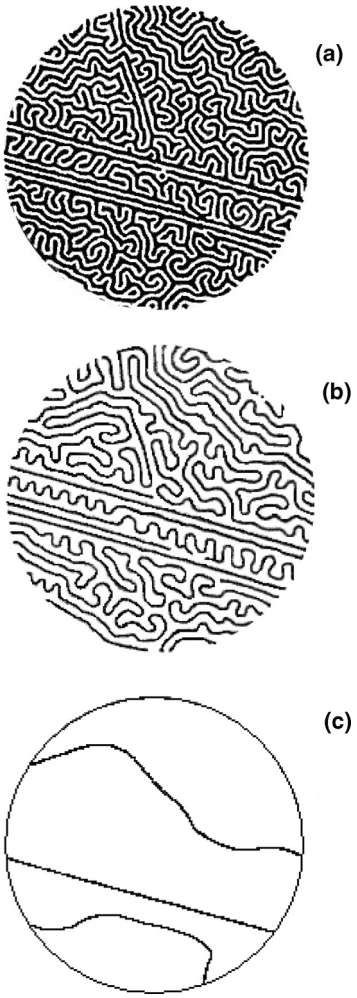

FIG. 1. Micrographs of the magnetic domain of a $(TmBi)_3(FeGa)_5O_{12}$ garnet film in various dc magnetic fields perpendicular to the film plane (a) $H_Z = 0$ Oe, (b) $H_Z = 50$ Oe, (c) $H_Z = 100$ Oe.

the domain magnetizations are directed with and counter to the direction of CPL propagation, hence, the circular dichroism manifests itself most effectively. Therefore we used, in particular, a $(Tm_{2.4}Bi_{0.6})(Fe_{3.9}Ga_{1.1})O_{12}$ garnet film: this film of thickness 7 μm with a large perpendicular anisotropy exhibits a typical mazelike domain pattern (the width of the domain is 10 μm) at $H_Z = 0$. The film was grown on a (111) $Gd_3Ga_5O_{12}$ substrate, Neel temperature $T_N = 430$ K, magnetic compensation temperature $T_C = 120$ K. The experiments were carried out at a temperature of 300 K at which the saturation magnetization $4\pi M_S \cong 70$ G (the measurements were performed with a tangent magnetometer). In Fig. 1 we present micrographs of the magnetic domain of a $(Tm_{2.4}Bi_{0.6})(Fe_{3.9}Ga_{1.1})O_{12}$ garnet film in various dc magnetic fields perpendicular to the film plane. Figure 2 illustrates the magneto-optic Faraday effect vs H_Z (the dc magnetic field perpendicular to the film plane), measured at $\lambda = 630$ nm. Since the aperture of the light beam is roughly 1.5 mm, which largely exceeds the width of the domain at $H_Z = 0$, the curve in Fig. 2 is nothing but the magnetization curve. One can see that its shape is actually a narrow hysteresis loop (the coercivity by our estimation is $\leqslant 1$ Oe at $T = 300$ K). Note that such a small

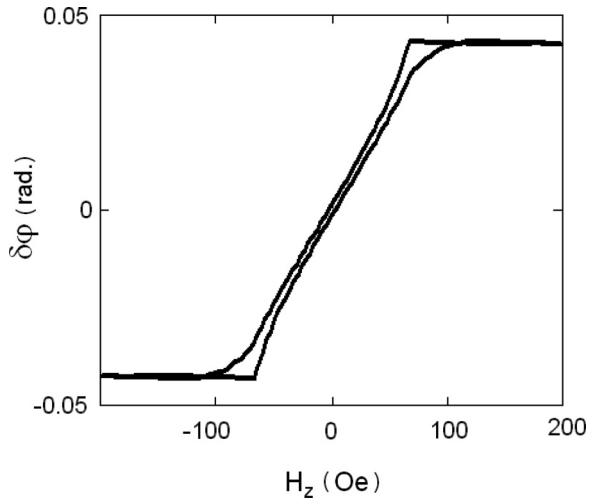

FIG. 2. Dependence of the magneto-optic Faraday effect $(TmBi)_3(FeGa)_5O_{12}$ ($\lambda = 630$ nm) on H_Z-dc magnetic field perpendicular to the film plane.

value of coercivity contributes to a *change* in the polydomain structure.

A source of polarized radiation was provided by a tunable $\lambda = 420 - 2500$ nm laser (pulse duration $\tau_i \approx 7$ ns, pulse repetition rate 10 Hz, power flux density $P < 600$ kW/cm^2). The sample was placed in the H_Z-dc magnetic field of a solenoid so that the field was perpendicular to the sample surface.

We observed the PMPM by CPL signal by successively using (i) the induction method and (ii) the magnetooptical method.

(i) The former technique involves an induction coil of 1.5-mm-inner diameter, placed directly on the sample surface. This coil is part of the measuring circuit whose oscillations period $T_c \approx 2 \times 10^{-7}$ s, $Q_c \leqslant 10$. A polarized radiation pulse passing along the induction coil axis z was incident at normal to the plane of the sample, causing a change in its magnetization ΔM_Z. The characteristic time of the ΔM_Z variation is determined by the relaxation time of magnetization τ_{im}; in our case $T_c \gg \tau_{im} > \tau_i$. A directly measured quantity corresponding to ΔM_Z was the e.m.f. $E_{emf}(t)$ signal picked by the measuring circuit and observed on the oscillograph. At $t > \tau_{im}$ the signal $E_{emf}(t)$ manifested itself as damping oscillations. We have found out that the phase of the $E_{emf}(t)$ signal is unambiguously dependent on the direction of the circular polarization of radiation. It means that it is this direction that determines the sign of ΔM_Z (see Figs. 3 and 4).

First, we studied the effect of PMPM by CPL for the case $H_Z = 0$ when an unilluminated sample was in the polydomain state, its total magnetization being zero. Note that with the linear polarization of radiation there was no photomagnetization effect in this case. The results of the experiment are given in Figs. 3–5. Figure 3 shows oscillograms of the $E^R_{emf}(t)$, $E^L_{emf}(t)$ signals for the right- and left-hand circular polarizations, respectively ($\lambda = 525$ nm, $P \approx 200$ kW/cm^2. Here and below the upper indices, R and L designate the right- and left-hand circular polarizations, respectively. It is seen that the phases of signals $E^R_{emf}(t)$ and $E^L_{emf}(t)$ are opposite, so $\Delta M^R_z = -\Delta M^L_z$.

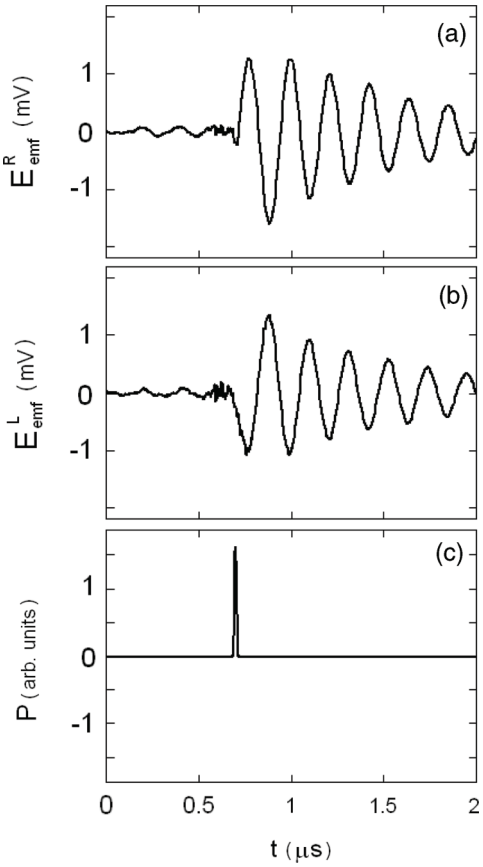

FIG. 3. (a), (b) Oscilloscope traces of $E^{R(L)}_{emf}(t)$ obtained with the right- and left-hand CPL at $H_Z = 0$ and $\lambda = 525$ nm; (c) monitor of the CPL laser pulse intensity ($P \approx 200$ kW/cm^2).

In further experiments we took measurements of the initial (at $t \cong \tau_{im}$) amplitudes of the signals and their phases, i.e., $E^{R(L)}_{emf}(\tau_{im})$; these particular quantities were chosen because $E^{R(L)}_{emf}(\tau_{im}) \sim \Delta M^{R(L)}_z$. In the following diagrams the quantities $\Delta M^{R(L)}_z$ are given in arbitrary units; at the same time, by our estimate the value of the PMPM-by-CPL effect for $\lambda = 525$ nm and $P = 300$ kW/cm^2 at $H_Z = 0$ is of the order of 1 G. It should be viewed as an approximate value because it was obtained from the induction measurements (in processing of the results we had to use a simplified model describing

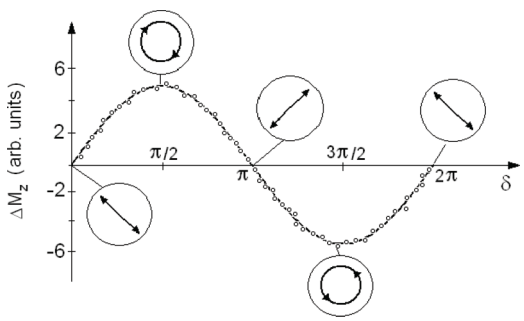

FIG. 4. Dependence of the photomagnetization ΔM_z on the light polarization, with $\lambda = 525$ nm at $H_Z = 0$. δ is the phase difference between the ordinary and extraordinary rays of the Babinet compensator.

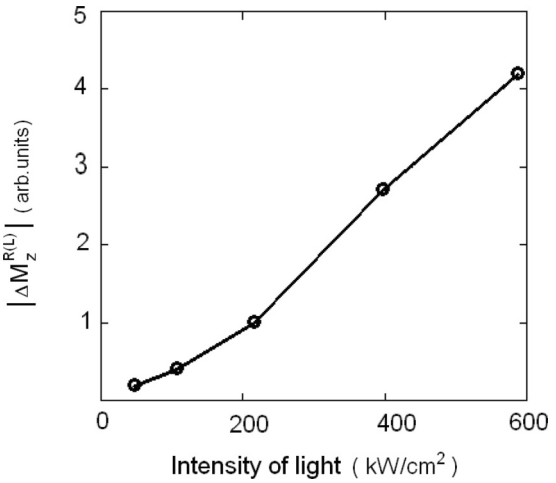

FIG. 5. Dependence of photomagnetization $|\Delta M_z^{R(L)}|$ on the power of CPL ($\lambda = 525$ nm) at $H_Z = 0$.

the coupling between the CPL magnetized area of sample and the measuring circuit). In order to specify the value of $\Delta M_z^{R(L)}$, we have carried out magneto-optical measurements (to be discussed in Sec. II).

Figure 4 is the dependence of ΔM_z on the polarization of light ($\lambda = 525$ nm, $P \approx 600$ kW/cm^2) at $H_Z = 0$. The polarization was varied using a quartz Babinet compensator.

Figure 5 demonstrates the dependence of $|\Delta M_z^{R(L)}|$ on the power of CPL ($\lambda = 525$ nm). It is seen that the value of photomagnetization is practically proportional to the radiation power, which is in agreement with the predictions in Refs. 25–28. Also note that by our estimate the temperature increase during a laser pulse in the laser spot area is $\leqslant$10 K at $\lambda = 525$ nm, $P \cong 600$ kW/cm^2.

We also studied the effect of PMPM by CPL vs magnetic field strength; the results are shown in Fig. 6. On the dependence corresponding to $\lambda = 525$ nm, $P = 600$ kW/cm^2, and magnetic field H_Z (0 Oe $\leqslant H_Z \leqslant 200$ Oe), the circles stand for ΔM_z^R and the crosses for ΔM_z^L. When interpreting this dependence one should bear in mind that at fields $0 \leqslant H_Z < 100$ Oe the sample is in the polydomain state, while at 100 Oe $\leqslant H_Z \leqslant 200$ Oe, it is single domain (see Figs. 1 and 2). It allows one to distinguish between the contributions in PMPM, which come from the changes in the magnetization

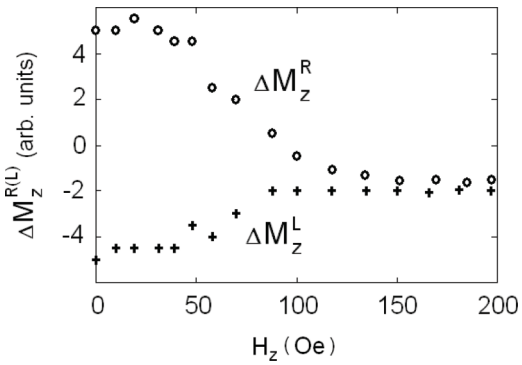

FIG. 6. Photomagnetization ($\lambda = 525$ nm, $P \approx 600$ kW/cm^2) vs strength of magnetic field $H_Z > 0$.

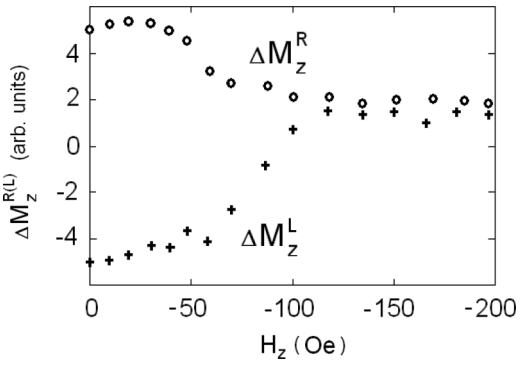

FIG. 7. Photomagnetization ($\lambda = 525$ nm, $P \approx 600$ kW/cm^2) vs strength of magnetic field $H_Z < 0$.

in the domains proper and from the *change* in the domain structure.

Specifically, since at magnetic fields 100 Oe $\leqslant H_Z \leqslant 200$ Oe a sample is apparently in the single-domain state, the change in the sample magnetization by CPL in these fields occurs only through a change in the magnitude of spontaneous magnetization of the formed single domain, which may be induced by both the sample heating and the heating-unrelated change in the state of magnetoactive ions. These changes in the single domain magnetization, being different through dichroism, we denote as $(\Delta M_z^R)^m$ and $(\Delta M_z^L)^m$ for the right- and left-hand polarizations, respectively. It is also seen from Fig. 6 that the characteristic value for the quantity $|(\Delta M_z^R)^m - (\Delta M_z^L)^m|/|(\Delta M_z^R)^m + (\Delta M_z^L)^m| \leqslant 0.2$. Besides, one can see here that the quantities $\Delta M_z^{R(L)}|_{H=0}$, describing the effect of PMPM by CPL at $H_Z = 0$, and $\frac{1}{2}|(\Delta M_z^R)^m - (\Delta M_z^L)^m|$ relate as

$$\Delta M_z^R\big|_{H=0} = -\Delta M_z^L\big|_{H=0} \gg \tfrac{1}{2}\left|\left(\Delta M_z^R\right)^m - \left(\Delta M_z^L\right)^m\right|.$$

However, $\frac{1}{2}|(\Delta M_z^R)^m - (\Delta M_z^L)^m|$ is the quantity that would correspond to the photomagnetization of sample in the polydomain state if the PM caused a change only in the magnetization of the adjacent domains, their volumes remaining equal (i.e., there was no change in the polydomain structure of sample). Such a situation could take place given strong pinning of the domain walls. So it is obvious that the effect of PMPM by CPL at $H_Z = 0$ relates to the *change* in the polydomain structure of sample. With the field increasing from $H_Z = 0$ to values high enough to trigger transition to the single domain state of the sample ($H_Z \approx 100$ Oe, see Fig. 2), the mechanism of photomagnetization through a *change* in the polydomain structure is gradually replaced by that through a change of magnetization in the single domain, which is shown in Fig. 6.

The results presented in Fig. 7 are similar to the data in Fig. 6. They differ in that the Fig. 6 diagram corresponds to the positive values of H_Z, whereas the one in Fig. 7 is for the negative H_Z. From comparison of these diagrams for 100 Oe $\leqslant |H_Z| \leqslant 200$ Oe it follows that the value of photoinduced magnetization at $H_Z > 0$ for the right (left)-hand polarization of light is opposite in sign to that at $H_Z < 0$ for the left (right)-hand polarization. It is quite consistent with the fact

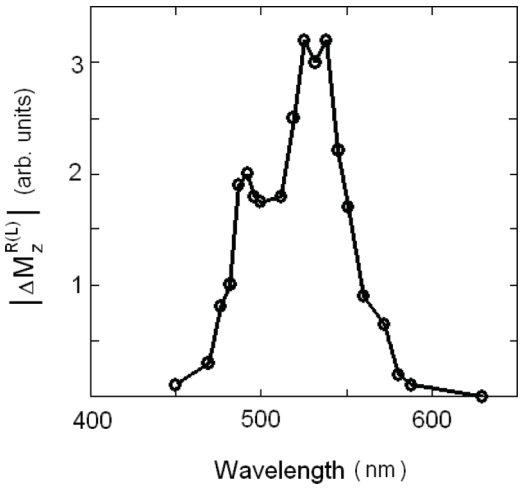

FIG. 8. Photomagnetization $|\Delta M_z^{R(L)}|$ vs CPL wavelength in the wavelength range of 450 nm–600 nm ($P = 500$ kW/cm^2, $H_Z = 36$ Oe).

that spontaneous magnetization in a single-domain state has opposite directions at $H_Z > 0$ and at $H_Z < 0$.

We have studied the dependence of this effect on a CPL wavelength. Figure 8 illustrates the dependence of $|\Delta M_z^{R(L)}|$ on CPL wavelength in the range λ =450 nm − 600 nm ($P =$ 500 kW/cm^2, $H_Z = 36$ Oe). It reveals a high "sensitivity" of the effect to wavelength.

(ii) We also used a well-known magnetooptical technique (the pump probe method) for investigating the effect of PMPM by CPL with $\lambda = 525$ nm at $H_Z = 0$. Together with the PMPM-inducing radiation, a probe beam (linearly polarized radiation from a He-Ne laser, $\lambda = 630$ nm) was passed at a small angle to normal through the illuminated area of the sample. We observed a Faraday rotation of the probe beam polarization plane simultaneously with the CPL impact. It is obvious that it relates to the PMPM effect: the direction of rotation of the probe beam polarization plane changed for the opposite with a change in the polarization direction of the PMPM producing radiation. Based on the results from magneto-optical observation of the hysteresis loop (see Fig. 2)

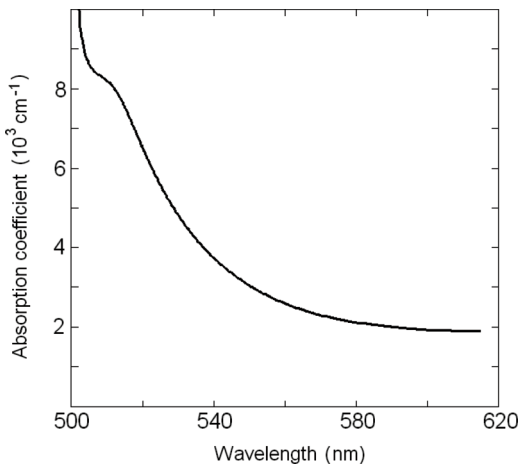

FIG. 9. Absorption spectra between 505 nm and 615 nm.

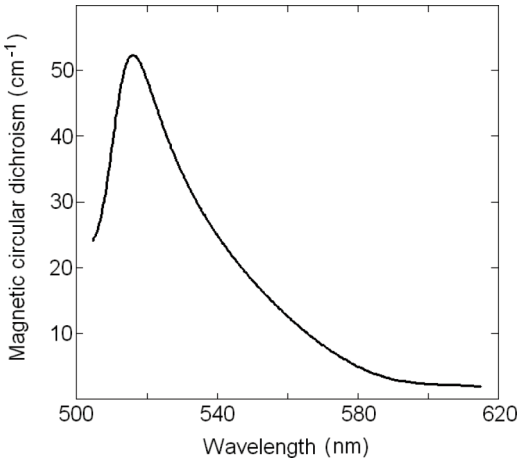

FIG. 10. Magnetic circular dichroism spectra between 505 nm and 615 nm.

we have made the following conclusion: the action of CPL with $\lambda = 525$ nm, $P \cong 300$ kW/cm^2, $\tau_i \approx 7$ ns causes magnetization of sample ≈0.1 G, such that would be induced by application of a magnetic field of ≈1.5 Oe. Considering this result jointly with the induction measurements data, we can conclude that CPL with the same characteristics causes sample magnetization of 0.1 G–1 G.

Besides the experiments for observation of photoinduced magnetization, we have measured the absorption and magnetic circular dichroism spectra and obtained reliable data for the wavelengths' range of 505–615 nm (the sample was in a single-domain state at $H_Z = 200$ Oe). These results are shown in Figs. 9 and 10. At wavelengths below 505 nm the transmitted light was practically undetectable because of a strong absorption. It is seen from Fig. 10 that in the 505–615 nm range of interest the value of the magnetic circular dichroism has a maximum at 515 nm. Comparison of the diagrams in Figs. 10 and 8 reveals a correlation between the magnetic circular dichroism and photomagnetization spectra.

III. CONCLUSIONS

We believe that our results provide sufficient evidence that the effect of PMPM by CPL reported in this paper is unambiguously related to a *change* in the polydomain structure of sample. To prove this fact was the goal of this work, which has been achieved. However, to establish the microscopic nature of the observed effect, or else to verify the theories proposed in Refs. 25–27, 29, and 30, will require further investigations. We think that an important part of this research would be a visualization of the domain restructuring process.

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