Temperature-independent Faraday effect in rare-earth iron garnets

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We investigate the frequency dependence of the Faraday effect (FE) and of the absorption of ytterbium, dysprosium, terbium gadolinium, europium, samarium, and yttrium iron garnets (IG) and of $Nd_1Y_2Fe_5O_{12}$ in the energy interval $\hbar\omega = 7500$–$20\,000\ \mathrm{cm}^{-1}$ at temperatures 82 and 295 K. The contribution of the rare-earth (RE) sublattice to the total FE of REIG is determined, and the frequency dependence is obtained of the coefficient C_e that characterizes an FE that is proportional to the magnetization of the RE sublattice. The frequency dependence of the FE due to the action of the iron-sublattice exchange field on the RE ions is determined for the first time, and it is shown that this contribution plays a principal role in the IG of ytterbium, gadolinium, samarium, and neodymium-yttrium ($Nd_1Y_2Fe_5O_{12}$).

INTRODUCTION

The Faraday effect (FE) in rare-earth iron garnets (REIG) was systematically investigated in a number of studies.[1-7] The explanation of the experimental results was based on the assumption that the FE is due to electrodipole (e) and magnetodipole (m) transitions and is proportional to the magnetizations of the iron garnet (IG) crystal magnetic sublattices, while the FE is a sum of contributions of all the sublattices to the resultant magneto-optical effect:

$$\alpha_F = \mp (A_e + A_m) |M_{\mathrm{Fe}}{}^a| \pm (B_e + B_m) |M_{\mathrm{Fe}}{}^d| \mp (C_e + C_m) |M_{\mathrm{R}}{}^c|. \tag{1}$$

Here M^a_{Fe}, M^d_{Fe}, M^c_{R} are the sublattice magnetizations; A, B, C are the magneto-optical coefficients for the octahedral (a), tetrahedral (d) and dodecahedral (c) ions. The superscripts and subscripts correspond to the temperatures above and below the compensation point. Equation (1) can also be represented in a form in which the gyromagnetic effect is written out explicitly:

$$\alpha_F = \alpha_e + \alpha_m = \alpha_e{}^{\mathrm{Fe+R}} + \frac{\pi n e}{mc^2} [g_{\mathrm{Fe}} (M_{\mathrm{Fe}}{}^d - M_{\mathrm{Fe}}{}^a) - g_{\mathrm{R}} M_{\mathrm{R}}{}^c], \tag{2}$$

where e and m are the charge and mass of the electron, $n = \varepsilon^{1/2}$ is the refractive index, and g_{Fe} and g_{R} are the spectroscopic splitting factors.

The question of the origin and magnitude of the gyromagnetic contribution to the FE can by now be regarded as answered, and the problem is to determine the physical origin of the gyroelectric FE $\alpha_e^{\mathrm{Fe+R}}$. We point out two present approaches to this problem: 1) The contribution made to the Faraday rotation by the iron ions are assumed to be for all the REIG and to yttrium IG, and the contribution of the RE sublattice is determined from the total Faraday effect of the IG. 2) The contributions of the iron sublattices and of the RE sublattice to FE are determined, using Eq. (1), from the temperature dependence of $\alpha_F^R(T)$ and the magnetizations of the IG sublattices.[3-5] Neither approach describes satisfactorily a number of experimental facts. In the first case, for example, the gyroelectric coefficient C_e turns out to depend strongly on the temperature, and even reverses sign for the erbium and holmium IG with change of temperature.[6] In the second case the contributions of the iron sublattices do not agree with the value of α_F^R of the yttrium IG, and A_e and B_e in $Y_3Fe_5O_{12}$ tend to zero as the Néel point is approached.[3] Nor can the indicated theories explain the new experimental fact,[9] that the FE due to the RE sublattice of the IG differs from zero when the RE sublattice magnetization is zero. An explanation was therefore proposed in Refs. 9 and 10, in which the diamagnetic effect and a mixing mechanism[1] were introduced besides the usually considered FE mechanism due to the different level population of the ground multiplet of the RE ion and proportional to the magnetization of the RE sublattice. Thus, the contribution of the RE sublattice to the FE is represented in the form

$$\alpha_F{}^{\mathrm{R}} = C_e M_{\mathrm{R}} + D H_{eff}, \tag{3}$$

where D is a temperature-independent coefficient, H_{eff} is the effective magnetic field and takes, in the case of mixing of the wave functions of the ground and excited states of the RE ion, the form

$$H_{eff} = H \pm 2H_{ex} = H \pm \frac{g_J}{g_J - 1} H_m,$$

where H, H_{ex} and H_m are respectively the external, exchange, and molecular fields, while g_J is the Lande factor.

The theory developed explained quite well the temperature and field dependences of the FE in terbium, dysprosium, and erbium IG, and yielded numerical values of the coefficients C_e and D. Nonetheless there are still very many questions to be answered. The point is that both in the cited studies and in most other experiments on the FE in REIG the measurements were made at only one wavelength, $\lambda = 1.15\ \mu\mathrm{m}$. The FE at this wavelength has a small absolute value in certain REIG. In addition, the contribution of the RE ions to the FE of the IG crystal can be influenced by ${}^6A_{1g}({}^6S) \to {}^4T_{1g}({}^4G)$ transitions in the Fe^{3+} ions in the region $\hbar\omega = 10\,500\ \mathrm{cm}^{-1}$, as well as by transitions in RE ion absorption lines having natural frequencies in this very same wavelength region.

To determine the temperature-independent contribution DH_{eff} to the FE of REIG and its frequency dependence, we have therefore undertaken the present investigation of

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the frequency dependence of the FE in the transparency region of IG crystals between two transitions in Fe^{3+} ions:

$$ {}^6A_{1g}({}^6S)\rightarrow{}^4T_{1g}({}^4G), \quad {}^6A_1({}^6S)\rightarrow{}^4T_1({}^4G). $$

We investigated typical representatives of three groups of REIG: 1—with the heavy RE elements Tb and Dy, which have a negative specific paramagnetic rotation; 2—with the elements Gd, Sm, and Eu in the middle of the lanthanoid series; and 3—with the light RE element neodymium.

MEASUREMENT PROCEDURE

To investigate the optical absorption and the Faraday effect in IG we used a magneto-optical system that recorded automatically the spectra of the light passing through the sample. The apparatus was based on a DMR-4 spectrometer, and the spectra were recorded with an LKD-4 automatic *x-y* plotter. In the working energy range 7500–20 000 cm^{-1} we used as radiation receivers a PbS photoresistor and FEU-79 and FEU-39 photomultipliers. The optical and magneto-optical spectra were recorded with a resolution 6–8 cm^{-1} in the 8000–13 000 cm^{-1} range and 3 cm^{-1} at 13 000–18 000 cm^{-1}. The Faraday spectra were plotted by a dynamic procedure in which the magnetic field was modulated and the change was measured of the intensity of the transmitted light at an angle 45° between the polarizer and the analyzer. The maximum ac amplitude in the electromagnetic gap in which the stem of the optical cryostat with the sample was placed amounted to 2.5 kOe. The measurements were made at two temperatures, room and 82 K. When the alternating field was not strong enough to magnetize the sample to saturation, the measurements and the calibration of the results were based on data obtained by a static procedure, in which the magnetic field intensity reached 18 kOe. The spontaneous Faraday effect values α_F^s needed for the calculations were obtained by extrapolating a number of values of $\alpha_F(H)$ at each temperature to zero field from regions exceeding the technical-saturation field, i.e., from 5 to 18 kOe. The values of α_F^s obtained in our measurements at $\lambda = 1.15$ μm agree with the analogous data of Ref. 10, where the measurements were made in fields up to 200 kOe, and with data by others.[5–8]

We investigated IG single crystals grown by spontaneous crystallization from the molten solution. The measurements were made on mechanically polished plates 20–100 μm thick oriented in the (111) plane, so that the direction of propagation of the light and the orientation of the external field coincided with the [111] axis, which is the easy-magnetization axis for all the investigated samples at the indicated temperatures. In this experimental geometry, the crystal is uniaxial and there are no magnetic birefringence effects.

EXPERIMENTAL RESULTS AND THEIR DISCUSSION

Figure 1 shows the low-temperature optical-absorption spectra of a number of IG. The room-temperature data, which agree in main outline with ours, can be found in Ref. 11. The values of the refractive index were calculated from the formula

$$ I=I_0(1-R)^2e^{-2\alpha d}, $$

which takes into account the reflection of the radiation from two surfaces of the sample (I_0 and I are the measured intensities of the incident and transmitted light, d is the sample thickness, and the reflection coefficient $R = 0.3$ was obtained under the assumption that the refractive index in the investigated wavelength band is 2.2).

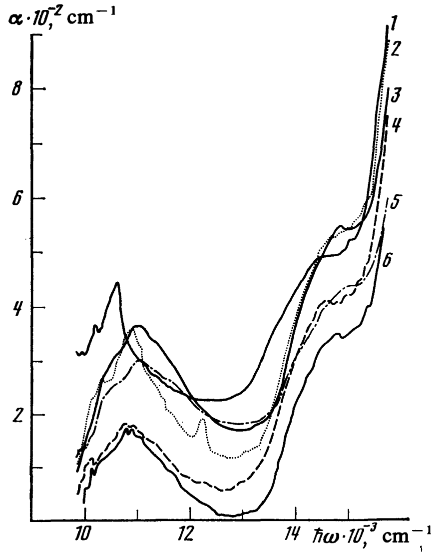

FIG. 1. Absorption spectra of iron garnets: 1—$Yb_3Fe_5O_{12}$, 2—$Dy_3Fe_5O_{12}$, 3—$Eu_3Fe_5O_{12}$, 4—$Y_3Fe_5O_{12}$, 5—$Sm_3Fe_5O_{12}$, 6—$Tb_3Fe_5O_{12}$.

Two absorption maxima are observed in the $\alpha(\omega)$ spectra, corresponding to optical transitions in the Fe^{3+} ions. The first maximum near 10 500 cm^{-1} is usually identified with the ${}^6A_{1g}({}^6S)\rightarrow{}^4T_{1g}({}^4G)$ transition, and the second in the 16 400 cm^{-1} region with the ${}^6A_1({}^6S)\rightarrow{}^4T_1({}^4G)$ transition. The smallest absorption in the investigated energy region is

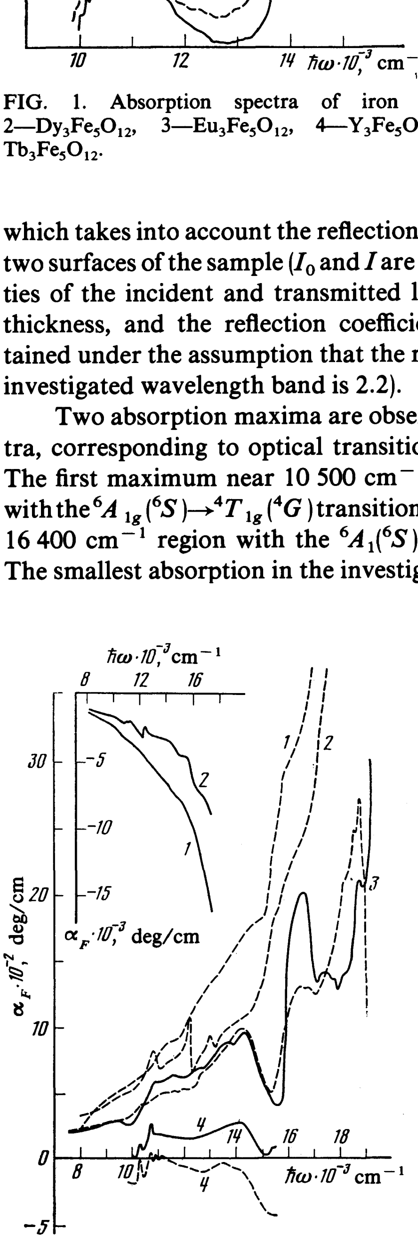

FIG. 2. Frequency dependence of the Faraday effect of iron garnets: 1—$Tb_3Fe_5O_{12}$, 2—$Dy_3Fe_5O_{12}$, 3—$Eu_3Fe_5O_{12}$, 4—$Yb_3Fe_5O_{12}$ (dashed curves—295 K, solie—82 K).

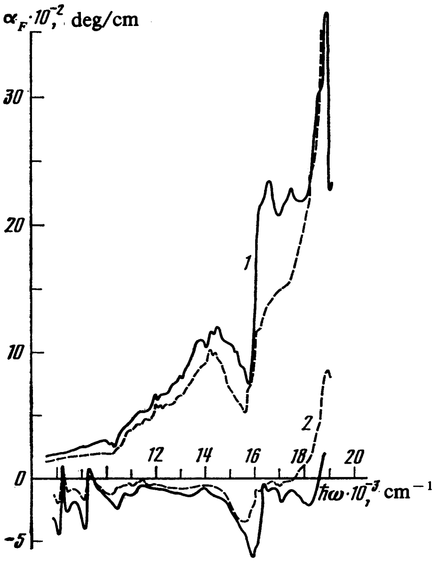

FIG. 3. Frequency dependence of the Faraday effect of iron garnets: 1—$Y_3Fe_5O_{12}$, 2—$Sm_3Fe_5O_{12}$ (dashed line—295 K, solid—82 K).

by the TbIG, and the largest by Yb IG. The absorption of the ytterbium IG near 10 000 cm^{-1} is enhanced by the transition $^2F_{7/2}\rightarrow{}^2F_{5/2}$ in the ytterbium ions. In the DyIG there are also observed absorption lines at energies 10 500 and 12 300 cm^{-1}, due to *f–f* transitions.

Figures 2–4 show the frequency dependences of the Faraday rotation of the polarization plane of linearly polarized light in the investigated IG. It can be seen from the figures that the absolute value of the Faraday rotation of all the IG increases with increasing frequency. The monotonic increase of the FE is due to intense transitions in Fe^{3+} and RE ions, whose natural frequencies lie in the ultraviolet. It is easy to trace against this background the influence of the two aforementioned transitions in the Fe^{3+} ions and of the narrower *f–f* transitions in the RE ions.

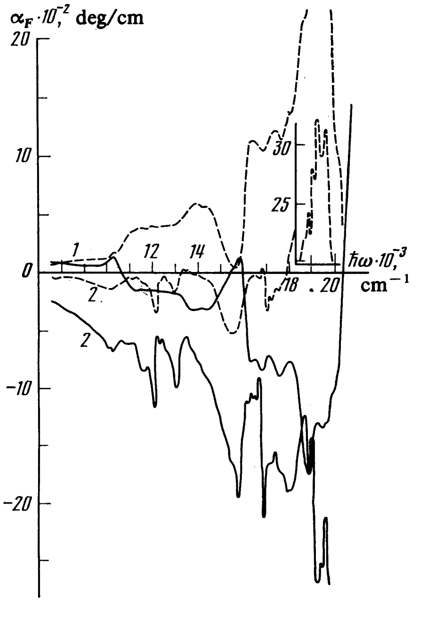

FIG. 4. Frequency dependence of Faraday effect of iron garnets: 1—$Gd_3Fe_5O_{12}$, 2—$Nd_1Y_2Fe_5O_{12}$ (dashed curves—195. K, solid—82 K).

The reversal of the sign of α_F in the garnets $Tb_3Fe_5O_{12}$ and $Dy_3Fe_5O_{12}$ on going from room to nitrogen temperature is explained by the fact that the compensation point is located in this temperature interval. The absolute value of the FE in these crystals at 82 K increases steeply compared with the FE in $Y_3Fe_5O_{12}$.

The Faraday effect in the europium IG at $T = 295$ K is practically the same as that of the yttrium IG in the entire investigated frequency range, i.e., the Eu rare-earth sublattice makes an anomalously small contribution to the combined FE. The values of α_F at 82 K in this crystal exceed the corresponding values at 295 K. In the IG $Sm_3Fe_5O_{12}$, $Yb_3Fe_5O_{12}$, and $Nb_1Y_2Fe_5O_{12}$ the FE decreases drasticaly, reverses sign (even at room temperature in IG with the RE ions Sm^{3+} and Nd^{3+}), although these garnets have no compensation point in the temperature interval 82–295 K. The absolute values of α_F in $Sm_3Fe_5O_{12}$ change little with changing temperature. The complicated frequency dependence of the FE, observed in the IG $Nd_1Y_2Fe_5O_{12}$, is due to a large number of *f–f* transitions in this region of the spectrum. The room-temperature reversal of the FE sign at 18 000 cm^{-1} is due to electronic transitions in the Fe^{3+} ions. These transitions are also the cause of the reversal of the FE sign in samarium IG. Even though the optical transitions in the Gd^{3+} ions begin only in the ultraviolet, complicated frequency and temperature dependences of the FE in the gadolinium IG are observed already in visible light: The sign of the effect reverses at $T = 82$ K near 10 500 cm^{-1}, and at higher energies ($\approx$20 500 cm^{-1}) the sign of the FE changes both at nitrogen and at room temperature.

As already noted in the Introduction, in the analysis of the measurement results we started from the assumption that the gyroelectric contribution to the FE of REIG on account of the Fe^{3+} sublattices is the same in all the garnets and is equal to FE of the yttrium IG, so that the gyroelectric FE of REIG can be represented in the form $\alpha_e^{\rm REIG} = \alpha_e^{\rm Fe} \pm \alpha_e^{\rm R}$. The contribution of α_e^R of the RE sublattice to the total FE is assumed to be proportional not only to the magnetic moment of this sublattice, but includes only the temperature-independent term $DH_{\rm eff} \equiv \alpha_e^d$, which we shall hereafter name diamagnetic. Thus, the angle of rotation of the polarization plane on account of the RE ions is a sum of a paramagnetic and a diamagnetic contributions: $\alpha_e^{\rm R} = \alpha_e^p + \alpha_e^d$. The gyromagnetic contributions to the FE on account of the iron and rare-earth sublattices were taken into account in accord with expression (2).

The magneto-optical coefficients C_e and D are obtained by simultaneous solution of the two equations (3) at $H = 0$ for the temperature 82 and 295 K. The contributions α_e^p and α_e^d to the FE for several fixed values of the incident-light frequency, and the coefficients C_e and D for $\lambda = 1.15\,\mu$m are listed in Table I. The RE-sublattice magnetizations, molecular fields, and g factors used in the calculations, and references to the corresponding sources, are also listed in this table. The magnetizations of the iron sublattices of the REIG were taken from Refs. 12–15. The molecular fields for

TABLE I.

Garnet	T, K	M_R, $\mu_B/2$ mol	H_m, kOe,	g_J	α_e^R, deg/cm				α_e^p, deg/cm				α_e^d, deg/cm				C_e, deg/cm·μ_B	D, deg/cm·kOe
					8696 cm^{-1}	10 000 cm^{-1}	12 000 cm^{-1}	14 000 cm^{-1}	8696 cm^{-1}	10 000 cm^{-1}	12 000 cm^{-1}	14 000 cm^{-1}	8696 cm^{-1}	10 000 cm^{-1}	12 000 cm^{-1}	14 000 cm^{-1}	8696 cm^{-1}	8696 cm^{-1}
$Yb_3Fe_5O_{12}$	82	2,34 [12]	139	8/7	–	500	755	995	–	90	290	230	–	410	465	765	+39 *	0,4 *
	295	0,73	102			348	433	670		28	90	70		320	342	600		
$Dy_3Fe_5O_{12}$	82	22,45 [12]	130	4/3	−1130	−1460	−2150	−2570	−1370	−1556	−2560	−3410	240	96	410	840	−61	0,46
	295	5,29	117		−105	−270	−268	−86	−323	−367	−603	−804	216	93	335	718		
$Tb_3Fe_5O_{12}$	82	23,73 [7]	183	3/2	−1422	−1980	−3570	−5750	−1922	−2660	−4460	−7360	500	680	890	1610	−81	0,9
	295	6,55	165		−90	−133	−450	−600	−534	−733	−1230	−2030	444	600	780	1430		
$Gd_3Fe_5O_{12}$	82	26 [14]	300	2	93	127	114	272	−14	−83	+24	−90	114	210	90	365	−0,55	0,21
	295	7,2	260		90	124	93	296	−4	−23	+8	−25	100	148	83	320		
$Sm_3Fe_5O_{12}$	82	1.85 [13]	1200	2/7	−540	−515	−790	−1000	(36)	(180)	(85)	(440)	−575	−695	−880	−1440	—	–
	295	0	890		−424	−517	−650	−1070	(0)	(0)	(0)	(0)	−424	−517	−650	−1070		
$Nd_1Y_2Fe_5O_{12}$	82	3,26 [13]	225	8/11	−670	−960	−1390	−1630	−425	−600	−940	−690	−245	−360	−450	−940	−130	0,4
	295	1,18	180		−344	−530	−670	−1000	−153	−220	−340	−250	−190	−310	−330	−750		

*The values are given for $\hbar\omega = 10\,000$ cm^{-1}.

REIG at room temperature were taken from Ref. 16, and were determined for 82 K from the formula $H_m = \lambda M_{Fe}$. There are no published data on the molecular fields in neodymium-yttrium garnets, and we determined them under the assumption that the exchange field in this garnet is equal to the exchange field in the gadolinium IG.

The frequency dependences of the coefficients C_e and D for Tb, Dy, Yb, and Nd-Y IG are shown in Fig. 5a. The frequency range of these coefficients is limited to 14 000 cm^{-1}, for at higher light frequencies the contribution made to the FE from the $^6A_1(^6S) \to {}^4T_1(^4G)$ transitions in the Fe^{3+} ions increases. In addition, in the calculations of these coefficients we purposely excluded those sections of $\alpha_F(\omega)$ on which RE-ion absorption lines were superimposed. It can be seen from the figures that the coefficients C_e and D increase substantially in absolute value with increasing frequency.

The coefficients C_e of the terbium, dysprosium, and ytterbium IG agree well both in sign and in absolute value with the parameter C_e obtained for the corresponding ions in paramagnetic glasses.[17] Recalculation and extrapolation of the data to $\lambda = 1.15\,\mu$m using the formula $\alpha_e \sim (\lambda^2 - \lambda_0^2)^{-1}$ gave for the specific rotation of the RE sublattice the values − 75, − 66, and + 40 deg/cm·μ_B for Tb^{3+}, Dy^{3+}, and Yb^{3+}, respectively. In addition, the values of C_e and D obtained by us for the terbium IG agree well with the corresponding coefficients for terbium gallate.[18] The main contribution to the rotation of the light-polarization plane in the Tb and Dy IG is made by the paramagnetic term α_e^p. Estimates of the natural frequencies from the frequency dependence of $\alpha_e^p(\hbar\omega)$ for $Dy_3Fe_5O_{12}$ yielded $\hbar\omega_0 = 30\,800$ cm^{-1} and $\hbar\omega_0 = 42\,000$ cm^{-1} for $Tb_3Fe_5O_{12}$, in satisfactory agreement with the natural frequencies ω_0 for the RE ions in paramagnets.[17] The coefficient C_e recalculated in accordance with our data for stoichiometric $Nd_3Fe_5O_{12}$ at $\lambda = 1.15\,\mu$m was − 390 deg/cm·μ_B, whereas the value calculated for the free Nd^{3+} ion from the data of Ref. 17 is − 205 deg/cm·μ_B at the same wavelength. The values of C_e and D for the gadolinium IG are − 0.55 deg/cm·μ_B and 0.21 deg/cm kOe from our data and − 1.1 deg/cm·μ_B and 0.2 deg/cm·kOe for paramagnetic gallate garnet from the data of Ref. 19.

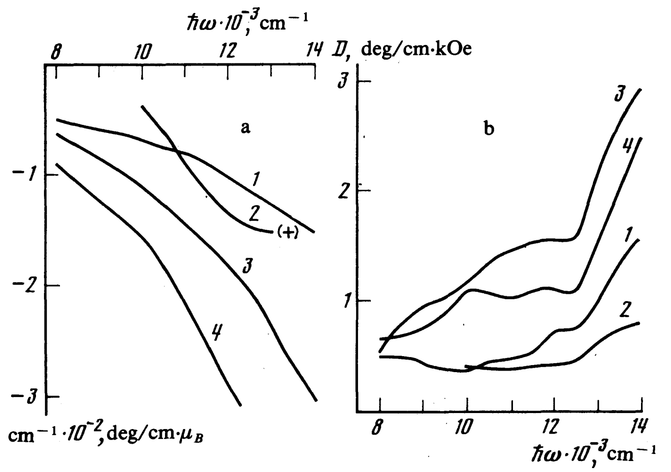

FIG. 5. Frequency dependences of magneto-optic coefficient C_e (a) due to the paramagnetic contribution, and of the temperature-independent coefficient (b): 1—$Dy_3Fe_5O_{12}$, 2—$Yb_3Fe_5O_{12}$, 3—$Tb_3Fe_5O_{12}$, 4—$Nd_1Y_2Fe_5O_{12}$, for which D is multiplied by $-g_J/(g_J - 1)$.

It can be seen from the $\alpha_F(\hbar\omega)$ curves that the octahedral transition in the Fe^{3+} ions in the 10 500 cm^{-1} region manifests itself, albeit not very pronouncedly, in all the IG, whereas the stronger tetrahedral transition near 16 400 cm^{-1} is clearly pronounced.[2)] In terbium and dysprosium IG the contributions of these transitions are less noticeable above the background of the total EF and affect little the values of the coefficients C_e and D, whereas the analogous parameters in the Sm and Gd IG is less reliable, since the values of the FE in these IG in the long-wave region are small both at room and nitrogen temperatures and begin to be overlapped by gyroelectric contributions due to optical transitions in the Fe^{3+} ions.

Examinations of the data in the table show that the absolute values of α_e^d of the ytterbium, samarium, and gadolinium IG exceed substantially the values of α_e^p in these crystals. The diamagnetic contribution is several times higher than the paramagnetic in $Yb_3Fe_5O_{12}$, larger by an order of magnitude in $Gd_3Fe_5O_{12}$, while in $Sm_3Fe_5O_{12}$ the gyroelectric rotation of the RE sublattice is practically completely diamagnetic. The values of α_e^p in the table for the samarium IG are regarded by us as estimates (and are therefore in parentheses). The reason is that the magnetic moment of the Sm^{3+} ions is small and all the determined quantities can vary with the "chosen" values of the RE-sublattice magnetization, although this does not alter the main contribution that the decisive role is played by the temperature-independent contribution DH_{eff} to the FE. We note also that in the entire frequency region the paramagnetic rotation of the ytterbium IG and the diamagnetic contributions of $Sm_3Fe_5O_{12}$ and $Nd_1Y_2Fe_5O_{12}$ are opposite in sign to those of the corresponding contributions for the other investigated crystals.

The similar form of the change of the FE spectra in $Yb_3Fe_5O_{12}$ and $Nd_1Y_2{\cdot}Fe_5O_{12}$ compared with the yttrium IG is explained by two different mechanisms. In the case of the ytterbium IG the magneto-optical coefficient C_e, in contrast to the corresponding coefficients of other IG, is positive. This makes positive the paramagnetic contribution made to the FE by the RE sublattice, and it is this which leads to the experimentally observed $\alpha_F(\hbar\omega)$ curves, if it is recognized that the moment of the RE sublattice is directed in this case opposite to the resultant magnetic moment of the Fe^{3+} sublattices both at room and at nitrogen temperature. In the case of the neodymium-yttrium garnet the situation is different. The magnetic moment of the Nd^{3+} ions, as that of all light element with more than half-filled f shell, aligns itself parallel to the resultant magnetic moment of the iron sublattices. It is this which causes the observed decrease of the FE at negative values of the coefficient C_e in neodymium-yttrium YG.

Thus, the measurements of the frequency dependence of the FE in rare-earth and yttrium IG have revealed the substantial role of the temperature-independent FE due to the action of the exchange field of the iron sublattices on the RE ions. We have shown that this contribution, heretofore completely disregarded, plays the fundamental role in ytterbium, gadolnium, samarium, and neodymium-ytterbium IG. We determined the frequency dependence of the coefficient D that characterizes the temeprature-independent FE. Our analysis has shown that in IG with terbium and dysprosium ions the largest contribution to the gyroelectric FE is made via the paramagnetic mechanism; this agrees with results[1,4,5,11] based on measurements at $\lambda = 1.15\ \mu$m. The obtained values of the coefficients C_e, which characterize the paramagnetic Faraday rotation, agree both in sign and in magnitude with the corresponding coefficients for RE ions implanted in paramagnetic glasses. Nonetheless, the contribution of the temperature-independent FE in these crystals is also appreciable, and this must be taken into account.

In conclusion, the authors thank B. V. Mill for supplying the neodymium-yttrium iron garnet samples.

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1) It should be noted that the exchange mechanism of the FE in RE ions, due to splitting of the excited levels by the molecular field produced at the RE ions by the iron sublattices, was first introduced in Ref. 1.

2) It can be seen from Fig. 4 that the influence of this transition in the gadolinium garnet is very large and depends strongly on temperature precisely near the laser wavelength 0.63 μm. This is probably why the authors of Ref. 20, who investigated yttrium and gadolinium IG at $\lambda = 0.63\ \mu$m, found it necessary to introduce for the explanation of their results a number of additional terms of the type $(d + aM_a^2 + bM_d^2 + cM_c^2)H$, with temperature dependent fit parameters a, b, c, and d.

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