Anomalous field dependence of the Faraday effect in paramagnetic $Gd_3Ga_5O_{12}$ at 4.2 K

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A nonmonotonic magnetic field dependence of the Faraday effect (Faraday rotation as a function of the magnetic field passes through a maximum) in paramagnetic $Gd_3Ga_5O_{12}$ is observed at 4.2 K at a wavelength of 0.47 μm. A theoretical explanation of this effect is given.

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It is usually assumed that the contribution of the rare-earth (RE) sublattice to the Faraday effect (FE) α_F in ferrite-garnets is proportional to its magnetization. This proposition can be checked by studying FE in isomorphic paramagnetic RE compounds (garnets-gallates and aluminates). In the infrared region of the spectrum the Faraday effect in gadolinium-gallium garnet $Gd_3Ga_5O_{12}$ at 4.2 K can be assumed to be proportional to the magnetization.[1]

However, in the visible region of the spectrum at wavelength $\lambda = 0.47$ μm, the Faraday effect in $Gd_3Ga_5O_{12}$ exhibits unusual behavior at $T = 4.2$ K (Fig. 1): First α_F increases in absolute magnitude with increasing field and then it begins to decrease in the range of fields where the magnetization (indicated by the dashed curve) approaches saturation. In addition, as our measurements have shown, the Faraday effect at the wavelength investigated changes sign with increasing temperature: It is negative at 4.2 K and positive at room temperature (see Fig. 2). This likewise cannot be explained by assuming that the Faraday effect is proportional to the magnetization. The observed characteristics of the Faraday effect can be understood if the Faraday rotation α_F is represented as a sum of two terms comparable in magnitude and having opposite signs,
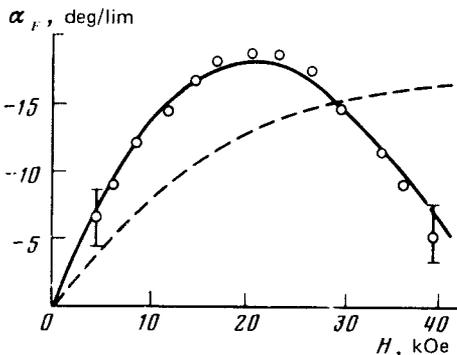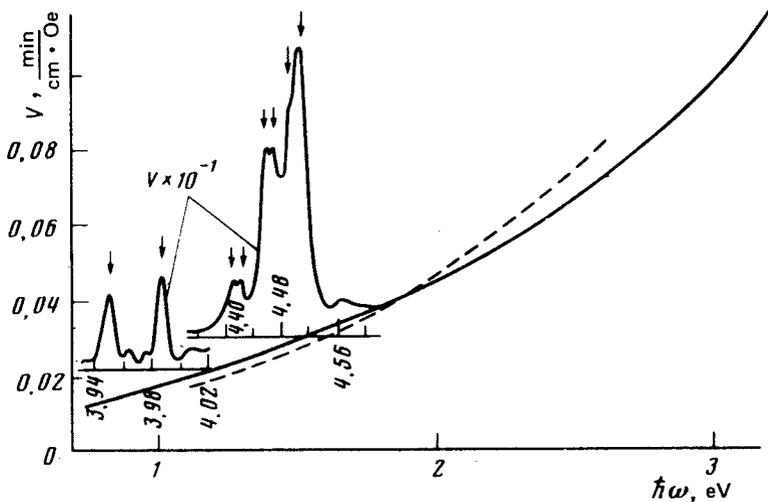

FIG. 1.

FIG. 2.

one of which is proportional to the magnetization m, while the other is proportional to the field H.

In general, the Faraday effect consists of the frequency-independent gyromagnetic Faraday effect $\alpha_m = C_m m$ (m is the reduced magnetization) and a frequency-dependent gyroelectric Faraday effect α_e. Usually, far from absorption lines, the expression for α_e is represented as a sum of three terms: diamagnetic α_A, paramagnetic α_C and α_B, due to mixing of ground-state and excited-state wave functions in a magnetic field (see, for example, Ref. 2) However, for the S-ion Gd^{3+} ($L = 0$) α_C and α_B vanish.[3] In this case, together with the diamagnetic contribution, the contribution to the Faraday effect from mixing of wave functions belonging to different multiplets of the excited term becomes important (for RE ions with $L \neq 0$, this contribution is neglected compared to α_C). Calculation of the Faraday effect assuming that the energy of the external magnetic and crystalline fields is much smaller than the energy of the spin orbital interaction leads to the following expression:

$$\alpha_e = \sum_n A_n \omega^2 \omega_n (\omega_n^2 - \omega^2)^{-2} \{ q_n \mu_B H - \xi_n m \}, \tag{1}$$

where ω_n is the transition frequency to the nth allowed term, A_n is a constant proportional to the oscillator strength of the transition, ξ_n is the L-S coupling constant in this term, and q_n is a numerical constant, which depends in a complicated manner on the spectroscopic parameters.[1)] The sum of the first terms, which have a weak temperature dependence and are proportional to H, describes the contribution of α_h to the Faraday effect, which arises as a result of mixing of wave functions that belong to different multiplets. The sum of the second terms, proportional to m, describes the diamagnetic contribution α_A to the Faraday effect, arising due to splitting of excited multiplets in a magnetic field (an analogous expression for α_A is presented in Ref. 3). We emphasize two facts that distinguish the Faraday effect in magnets with S ions ($L = 0$) from the Faraday effect in magnets with LS ions ($L \neq 0$). First, both contributions—diamagnetic

and mixing—have the same frequency dependence and, second, the diamagnetic term is proportional to the magnetization, i.e., it depends on the temperature (for $L \neq 0$, α_A does not depend on temperature). Thus, the expression for the Faraday effect in $Gd_3Ga_5O_{12}$ can be represented in the form

$$\alpha_\phi = \alpha_m + \alpha_e = \alpha_m + \alpha_A + \alpha_h = (C_m + C_A)m + V_h H$$
$$= (C_m + C_A)B_{7/2}(\mu H/kT) + V_h H \qquad (2)$$

(here $\mu = 7\mu_B$ is the moment of Gd^{3+} and we include the fact that the magnetization of paramagnetic Gd^{3+} ions is well described by a Brillouin function). Competition between different contributions to α_F leads both to the unusual behavior of $\alpha_F(H)$ at 4.2 K and to the change in α_F with increasing temperature.

The continuous curve in Fig. 1 shows the dependence (2) for $C_m + C_A = -60$ deg/cm and $V_h = 1.2$ deg/cm kOe. Using the experimental value of C_m and including the dispersion in the index of refraction, we obtain $C_A = -390$ deg/cm. If it is assumed that α_F is determined by the efficient transition with $q = 3$, then from these quantities it is possible to estimate the effective spin-orbital coupling constant $\xi \approx 10^6\mu_B$ Oe. This value agrees in order of magnitude with data from optical measurements for RE ions, but it is somewhat too low, since in performing the calculation we ignored the contribution of diamagnetic ions present in the matrix to α_F.

Taking into account the spectral dependence of C_A, $V_h \sim \omega^2(\omega_0^2 - \omega^2)^{-2}$, where $\omega_0(2\pi c/\omega_0 = 0.2\,\mu m)$ is the effective resonant absorption frequency, it is possible to estimate the Faraday effect over a wide spectral range for $T \geqslant 4.2$ K. It is easy to see that by choosing the experimental conditions carefully, it is possible to obtain, after compensating for two of three contributions, different "exotic" dependences on the wavelength of light, temperature, and magnetic field.[2]

For example, at $T \approx 70$ K, α_A and α_h cancel out each other and the Faraday effect is nearly independent of ω, while α_m and α_A cancel out each other at $\lambda \approx 0.5\,\mu m$ and the Faraday effect in this case is nearly independent of temperature. The solid curve in Fig. 2 shows the spectral dependence of the Verdet constant $V = \alpha_F/H$ at 295 K and the dashed curve is for 635 K. As is evident from the figure, for $\lambda \cong 0.67\,\mu m$, the Faraday effect does not depend on the temperature. C_m, C_A, and V_h can be determined from the spectral data. The values obtained differ from the estimates made from measurements at 4.2 K at a single wavelength by not more than a factor of 2. Keeping in mind the roughness of the spectral estimates and the fact that in this calculation we ignore the temperature and field dependence of V_h, the agreement is good.

In the region of transitions $^8S \rightarrow {}^6P$, ^8I (Fig. 2, $\hbar\omega \cong 4$ and $\hbar\omega = 4.5$ eV, the positions of the absorption maxima are indicated by arrows), the dependence $\alpha_F(\hbar\omega)$ indicates that the Faraday effect, because of these transitions, has a diamagnetic spectral dependence, which is valid if the mechanism permitting the transition involves mixing of states with different parity to the excited multiplets, while the transitions occur from an orbital singlet.

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[1]For example, for transitions to the first excited level $L = 1$, $S = 7/2$ of the configuration $4f^65d$ we have $q_1 \cong 3$. We note that the quantities q_n have a weak temperature dependence for $kT \lesssim 10\,\mu_B H$, which is attributed to the change in the population of Zeeman components of the principal multiplet.

[2]Such dependences are much more difficult to observe in other compounds of RE ions. It is either necessary to perform the measurements in megagauss fields or at high temperatures. Thus, using the data in Ref. 4, we can estimate that in $Tb_3Ga_5O_{12}$ a_e must change sign at $T = 2000$ K.

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