The Properties of the Faraday Effect in $DyAlO_3$ Dysprosium Orthoaluminate

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Abstract: In this article, one of the magneto-optical properties of the rare earth element dysprosium orthoaluminate is studied: the Faraday effect. Additionally, the Faraday constant has been determined, and its wavelength-dependent graph has been plotted. The spectral and temperature dependencies of the Faraday effect (EF) in dysprosium orthoaluminate ($DyAlO_3$) have been measured, which included the measurement of its magnetic sensitivity as a function of temperature. That is, the temperature increased from 90 K to 300 K. Additionally, the Verday constant of dysprosium orthoaluminate was compared with the Verday constant of $Tb_3Ga_5O_{12}$.

Key words: rhombic crystal, Faraday effect, Verde constant, dysprosium orthoaluminate, active environment

INTRODUCTION

The great interest in the study of various rare earth (-RE) compounds is largely due to their potential use as active media in visible-band lasers, operating both in up-conversion and conventional optical circuits. In modern spectroscopy of rare earth elements, particular attention is paid to the problem of studying the energy spectra of trivalent RE-ions R^{3+} doped into crystal structures of lower symmetry (rhombic, monoclinic, etc.), while a number of factors of practical and scientifically important importance remain unclear and controversial to this day.

Therefore, further prospects for the development of applications of low-symmetric RE compounds in optics require a deeper study of the energy spectrum of RE ions formed in low-symmetric crystal environments (CS, D2 symmetry). However, a number of issues related to the microscopic expression of the main mechanisms of light absorption and emission in such RE compounds for a particular optical range and the microscopic expression of the mechanism of magneto-optical activity (MOF) of rare earth ions in them remain relatively poorly studied to this day.

This is a high-level Kramers (having an odd number of electrons in the unfilled 4f shell) RE-ions belong to excited multiplet electronic energy structures called Dy^{3+}, Er^{3+}, and RE- orthoaluminate belongs to calcined perovskite-type non-cubic compound crystals called $RAlO_3$.

Although there is some scattered information in the literature about the magneto-optical, magnetic and optical properties of dysprosium orthoaluminate $DyAlO_3$, It is impossible to estimate the value of magneto-optical activity in a particular range based only on data on the magnetic and optical properties of the RE compound. Therefore, from the point of view of the development and practical application of fundamental ideas about the physical mechanisms responsible for the optical and magneto-optical properties of RE-magnets, the application of microscopic expression of both the fundamental mechanisms of light absorption and emission, and the mechanisms of magneto-optical activity of rare earth ions Dy^{3+} in orthoaluminate crystals is extremely relevant.

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METHODS

The magneto-optical properties of RE orthoaluminates RalO3 are distinguished by a number of characteristic features. First, between the thickness of the orthorhombic crystal RalO3 and the magnitude of the magneto-optical effect, due to the natural birefringence characteristic of such crystals, the angle of rotation θ of the major axis of the polarization ellipse of the light beam passing through the crystal is sufficiently small and in terms of absolute magnitude, the magnetic fields actually achieved in the visible regions of the spectrum (~10kE) do not exceed a few arcminutes in magnitude.

RE-orthoaluminates exhibit a number of characteristic properties in their magneto-optical characteristics. Firstly, due to the lack of proportionality resulting from the natural double refraction characteristic of such crystals, there is a significant difference between the thickness of the orthorhombic crystal $RAlO_3$ and the magnitude of the magneto-optical effect. The angle of rotation θ of the major axis of the polarization ellipse of the light passing through the crystal remains sufficiently small, and in terms of absolute magnitude, it does not exceed several arc minutes in the real magnetic fields achieved in the visible spectrum (∼10 kE). Using the formula $V = 2\left(\frac{\theta}{H}\right) \cdot \frac{\pi}{\lambda}\Delta n$, with characteristic values for this case (n, Δn, λ, V) taken as [2]: $n \approx 2.0$; $\Delta n \approx 3 \cdot 10^{-2}$; $V \approx 1$ (min/cm·E); $\lambda \approx 5 \cdot 10^{-5}$ cm, calculations can be made to show that the value of θ found in a field of $H \approx 5 \cdot 10^3$ E does not exceed ∼3′. Secondly, the presence of large natural double refraction ($\sim 10^{-2}$) complicates precision optical measurements significantly, ultimately leading to strong temperature and wavelength-dependent variations in the angle θ of the major axis of the

light polarization ellipse. Therefore, due to the aforementioned factors, highly sensitive methods are used for dynamically recording the rotation angle of the polarization plane when measuring the angle θ.

In this method, the magnitude of the rotation angle θ of the polarization ellipse is determined by the relative change in the intensity of light passing through an optical system consisting of a polarizer, sample, and analyzer, given by $\delta = \Delta I/I_0$. The magnetization of the sample leads to the rotation angle θ being a periodic function corresponding to the frequency of the alternating magnetic field H. Thus, the light flow I reaching the photodetector has two signals: a variable ΔI related to the change in the rotation of the polarization plane, $\Delta I = (I - I_0)$, (more precisely, the large ellipse of polarization), and a constant I_0 determined by the light flow passing through the polarizer-sample-analyzer system at H = 0. Using the Jones matrix ([5]) to determine the relative amplitudes and phases of the electric field vector components of the light passing through the magnetized orthorhombic sample (see [1,3]), and considering the orientations of the transmission planes of the polarizer (P) and analyzer (A) with respect to the orthorhombic crystal axes (see Figure 5b), it is not difficult to show that the intensity of the light reaching the photodetector I can be expressed as: $I = 1/2I_0 \cdot (1 \pm \sin\chi \cdot \sin\varphi)$

Here, the (±) signs correspond to the two orthogonal states of the polarizer's transmission plane. From this, an expression can be obtained for the relative change in light intensity δ passing through a magnetized orthorhombic crystal in the case of large natural double refraction, where $n \cdot \Delta n >> \varepsilon''$: $\delta = (I - I_0)/I_0 = 2\theta = \frac{\varepsilon_{xy}}{\bar{n} \cdot \Delta n} \cdot \sin\frac{2\pi \cdot \Delta n \cdot l}{\lambda}$

In this case, under the assumption of a small rotation angle, the relative change in light intensity δ\deltaδ determines the double angle of rotation of the polarization plane (when the analyzer azimuth is equal to 45 degrees). Thus, in this geometry of the experiment, the relative change in light intensity δ expresses a coupling that depends on both the wavelength and temperature (where the natural double refraction Δn of the crystal is a functional relationship of these parameters). The amplitude of the coupling is directly determined by the magnitude of the Faraday rotation angle α_ϕ (where $\alpha_\phi \sim \varepsilon_{xy}$), and their period is defined by the magnitude of the natural crystallographic double refraction Δn.

ANALYSIS AND RESULTS

In rhombohedral crystals, the orientation of the polarization ellipse is related to the rotation angle θ of the crystal's dielectric permittivity tensor's non-diagonal component by the following relationship:

$$tg2\theta = \sin\chi \cdot \sin\Phi = \sin\Phi = \frac{\varepsilon_{xy}}{\bar{n} \cdot \Delta n} \cdot \sin\frac{2\pi \cdot \Delta n \cdot l}{\lambda} \quad (1)$$

$\bar{n} \approx \frac{n_{xx} - n_{yy}}{2}$ – the average refractive index of the crystal, l – its thickness, λ – the wavelength of light in vacuum, $\Delta n = n_{xx} - n_{yy}$ – natural crystallographic birefringence. The angle θ depends on both the wavelength and the crystal thickness and has an oscillatory character, where the oscillation amplitude is proportional to ε_{xy}, and its period is proportional to the value of natural birefringence Δn.

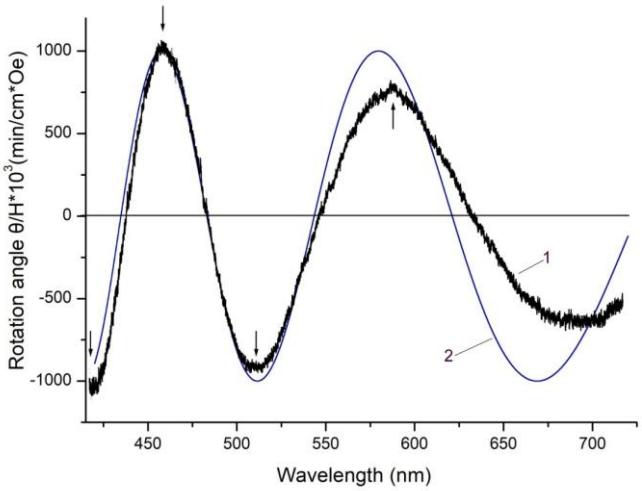

Figure 1. The spectral dependence of the θ/H quantity in TbAlO3 recorded in an external magnetic field (H = 7 kOe) along the crystallographic [100] direction of a rhombohedral crystal with a thickness of l=0.0145 cm. The figure also shows the dependence of the phase factor $\sin\Phi$ on the wavelength.

In the external magnetic field, the optical properties of RE-orthoaluminate serve as a vivid example. In Figure 1, the light polarization of the $TbAlO_3$ crystal is presented along the "light" axis at **a** temperature of 300 K, showing the spectral dependence of the rotation angle θ of the major axis of the polarization ellipse in the wavelength range of 430 - 720 nm (specifically, the dependence of the θ/H quantity).

In the comparison, this image shows the relationship of the calculated phase factor $\sin\Phi = \sin\frac{2\pi \cdot \Delta n \cdot l}{\lambda}$ with the wavelength when the crystal thickness $l = 1.45 \cdot 10^{-2}$ cm and the parameter value $\Delta n = 1.5 \cdot 10^{-2}$. These comparisons indicate that the calculated relationship of the phase factor adequately expresses the dependence of the rotation angle θ of the polarization ellipse in the orthoaluminate on the wavelength (albeit with some difference in the long wavelength part of the spectrum). For $TbAlO_3$ (at the given temperature), processing the results of measuring the rotation angle θ was carried out to restore the spectral relationship of the Verde constant as follows: first, the dependence of the two-fold light refraction Δn on the wavelength is determined in the observed spectral areas. For this purpose, the size θ/H constitutes the phase shift between the nearby extrema of the spectral relationships - between the negative and positive maxima, corresponding to the wavelengths λ1 and λ2, which form π radians (in Figure 1, these extrema are marked with arrows). In that case, from formula (1):

$$\Delta\Phi = \Phi_2 - \Phi_1 = \pi = 2\pi \cdot \Delta n l \cdot \left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1}\right), \quad \text{or}$$

$$\Delta n = \frac{\lambda_2 \cdot \lambda_1}{2l \cdot (\lambda_2 - \lambda_1)} \quad (2)$$

Here, l is the crystal thickness, and λ_1 and λ_2 are the wavelengths of the spectral extrema of the angle θ. According to formula (2), the calculations for Δn based on the results of Figure 1 show that in the long-wavelength part of the spectrum, Δn increases monotonically from 0.013 to 0.016 in the short-wavelength part. This indicates that there is a

sufficiently noticeable dispersion of natural double refraction in the observed crystal.

Now, we use the fact that the relative Faraday rotation angle a_F/l (i.e., the rotation angle of light polarization uniformity calculated per unit length of the sample) is directly related to the off-diagonal component of the dielectric permittivity tensor through the following expression:

$$\frac{a_F}{l} = \frac{\pi}{\lambda} \cdot \frac{\varepsilon_{xy}}{\bar{n}} \qquad (3)$$

By combining equations (1) and (3), and considering that we can express the Faraday rotation angle as $a_F = VlH$ (where V is the Verde constant), we can obtain the final expression for the orthoaluminate Verde constant:

$$V = 2\left(\frac{\theta}{H}\right) \cdot \frac{\pi}{\lambda} \Delta n \qquad (4)$$

This allows for the restoration of the Verde constant of the investigated orthoaluminate at a specified temperature and wavelength. For instance, at a wavelength of λ=632 nm (see Figure 1), in the calculations of the Verde constant of TbAlO3 , the average value of the parameter $\left(\frac{\bar{\theta}}{H}\right) = \frac{1}{2}\left[\left(\frac{|\theta|}{H}\right)_{\lambda_1} + \left(\frac{|\theta|}{H}\right)_{\lambda_2}\right]$ is used, where λ1 and λ2 are the wavelengths of the spectral dependencies of the θ/H parameter corresponding to closely located extrema (negative and positive). The value $\Delta n = 1.3 \cdot 10^{-2}$ is the average value of the double refraction parameter for the investigated wavelength.

Ultimately, at T=300K, a rhombohedral crystal measured along the light axis a achieves a Verde constant of the terbium orthoaluminate that is twice that of the terbium garnet at a wavelength of 632 nm.

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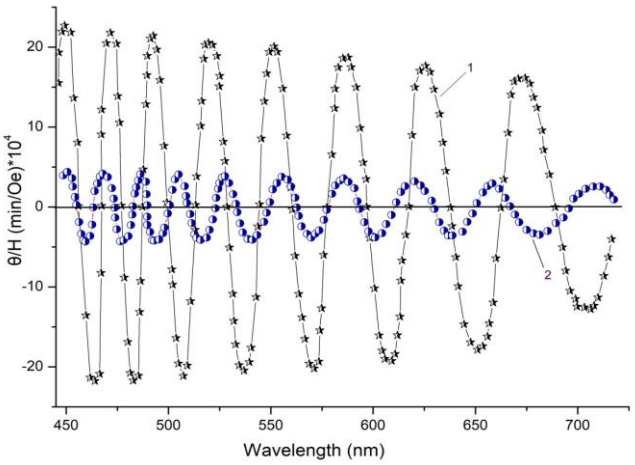

gure 2. The spectral dependence of the θ/H value measured along the b-axis of the rhombic crystal DyAlO3 with thickness l = 0.42 cm at T = 90 K (1) and 300 K (2)."

In an analogous manner to the previous case, the spectral dependence of the θ/H value measured along the 'light' b-axis of the rhombic crystal at T = 90 K and 300 K can be used to determine the Verde constant V and the two-fold optical birefringence magnitude Δn of $DyAlO_3$ (Figure 3). The experimental results presented in this figure show that with decreasing wavelength at T = 90 K, there is a monotonic increase in the Δn value (see the additional graph in the figure). Additionally, it has been determined that with the decrease in sample temperature, the period of the angle θ oscillation decreases, which is attributed to the slight increase in natural crystallographic birefringence with temperature decrease. In this case, the amplitude of the angle θ oscillation significantly increases, which, according to formula (1) and in accordance with formula (3), leads to an increase in the comparative Faraday rotation angle that is directly related to the off-diagonal component of the dielectric permittivity tensor εxy.

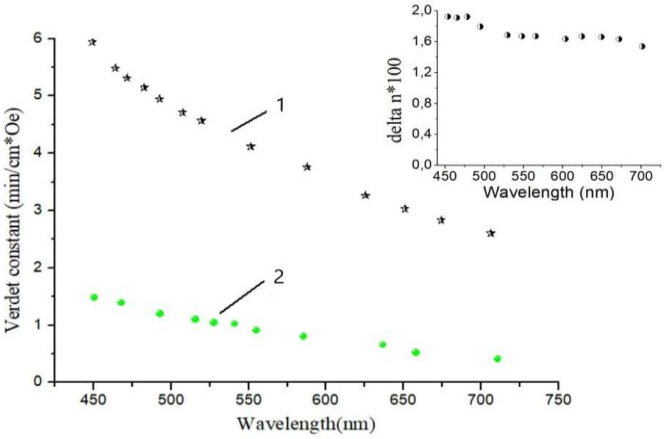

Figure 3. The spectral dependence of the Verde constant of the orthogonal DyAlO3 measured along the b-axis of the rhombic crystal at T = 90 K (1) and 300 K (2). In the additional graph: the spectral dependence of the Δn value found at T = 90 K is presented.

Upon careful examination of the experimental results presented above, it can be noted that at T = 300 K, the Verde constant V measured along the 'light' b-axis of the rhombic $DyAlO_3$ crystal at a wavelength of 632 nm is 0.662 min/sm·E, which is nearly 1.5 times greater than that of the terbium garnet. The primary interest lies in the significant dependence of the $DyAlO_3$ Verde constant on wavelength and temperature (Figure 3). This suggests that the 'paramagnetic' mechanism of magneto-optical activity in the dysprosium orthogonal aluminum crystal FE contributes significantly. The frequency dependence of this mechanism is expressed by the frequency factor $\omega^2/(\omega_0^2 - \omega^2)$. Therefore, it can be said that the paramagnetic mechanism plays a major role in the observed NE mixture in FE, as can be deduced from the frequency dependence of the inverse Verde constant measured at T = 90 K, allowing us to determine the 'effective' frequency of the allowed optical transitions in Dy^{3+} ions, which is $\omega_0 = 99 \cdot 10^{14} s^{-1}$ ($\lambda_0 = 190$ nm).

On the other hand, considering the pronounced temperature dependence of the magneto-optical effect observed in $DyAlO_3$, it is possible to derive the dependence of the Verde constant V measured in the direction of an external magnetic field N along the b-axis of the dysprosium orthogonal aluminum on the magnetic susceptibility χ (see Figure 4). A thorough examination of Figure 4 indicates that this dependence is linear within the experimental error margins, which allows us to express the Verde constant in terms of the frequency dependence of FE in $DyAlO_3$ as follows:

$$V = (C_p\chi + D)\frac{\omega^2}{\omega_0^2 - \omega^2} \qquad (5)$$

Here, V is the Verde constant; ω is the light frequency; Sp is the 'paramagnetic' constant of Verde; D represents the

contribution of the mixing of the ground and excited multiplets of the Dy^{3+} ion in an external magnetic field. The analysis of the experimental results presented in Figure 4, using formula (5), showed that the contribution of D is sufficiently small, allowing it to be neglected in subsequent considerations.

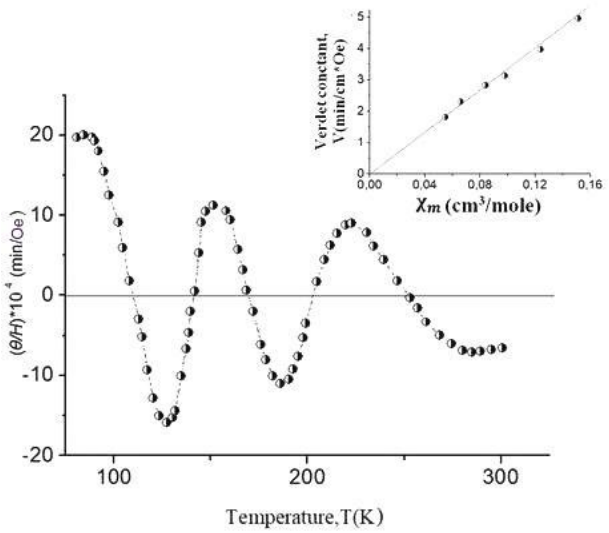

Figure 4. The temperature dependence of the θ/H value measured along the b-axis of $DyAlO_3$ at a wavelength of λ = 510 nm. The thickness of the investigated crystal is l = 0.42 cm. In the additional graph: the dependence of the molar magnetic susceptibility χ on the Verde constant of $DyAlO_3$ measured along the b-axis of the rhombic crystal at the given wavelength.

Conclusion

The spectral and temperature dependencies of the Faraday effect (EF) in dysprosium orthogonal aluminum DyAlO3 are measured, complemented by measurements of its magnetic sensitivity's temperature dependence. At T = 300 K and a wavelength of 632 nm, the measured Verdet constant V along the 'easy' b-axis of the rhombic $DyAlO_3$ crystal is 0.662 min/cm·Oe, which is nearly 1.5 times higher than the Verdet constant of $Tb_3Ga_5O_{12}$, which is 0.46 min/cm·Oe. This suggests that it can potentially be used as an active medium in lasers.

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