Faraday effect in Tl_3AsS_4 crystals

[1] Adamenko D., [1] Kushnirevych M., [2] Kokhan O. and [1] Vlokh R.

[1] Vlokh Institute of Physical Optics, 23 Dragomanov Street, 79005 Lviv, Ukraine, vlokh@ifo.lviv.ua
[2] Department for Inorganic Chemistry, Uzhgorod National University, 46 Pidgirna Street, 88000 Uzhgorod, Ukraine

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Abstract. We have studied experimentally the Faraday effect in Tl_3AsS_4 crystals. The effective Faraday tensor coefficient and the Verdet constant are determined under normal conditions and $\lambda = 632.8\,\mathrm{nm}$. They are equal to $0.06F_{11} + 0.94F_{22} = (9.23 \pm 0.21)\times 10^{-13}\,\mathrm{m/A}$ and $V_F = (82.2 \pm 1.8)\,\mathrm{rad/T\times m}$, respectively. We have shown that the Tl_3AsS_4 crystals are one of the most efficient magnetooptic materials among magnetically non-ordered ones.

Keywords: Faraday effect, Tl_3AsS_4 crystals, Verdet constant

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1. Introduction

Thallium arsenic sulfosalt Tl_3AsS_4 is a fangite mineral [1] that belongs to the point symmetry group *mmm*. Its structure is characterized by four formula units per cell [2]. The crystal is optically biaxial, with the principal refractive indices equal to $n_a = 2.829$, $n_b = 2.774$ and $n_c = 2.825$ at $\lambda = 632.8\,\mathrm{nm}$ [3]. Tl_3AsS_4 is transparent in a wide spectral range (0.6–12 μm [3]). It is also known that its elastooptic coefficients [4, 5] are $p^r_{31} = 0.92$, $p^r_{32} = 1.09$ and $p^r_{33} = 1.33$ (at $\lambda = 632.8\,\mathrm{nm}$) [3] in the relative units expressed with respect to the coefficient $p_{31} = 0.29$ for the quartz crystals at $\lambda = 589.3\,\mathrm{nm}$ [6].

It is also known that the Tl_3AsS_4 crystals are characterized by a very slow shear acoustic wave propagating with the velocity ~ 600 m/s [7]. A notable acoustic-wave slowness and high enough elastooptic coefficients result in high acoustooptic figures of merit, which are equal to $453\times 10^{-15}\,\mathrm{s^3/kg}$, $555.7\times 10^{-15}\,\mathrm{s^3/kg}$ and $792.8\times 10^{-15}\,\mathrm{s^3/kg}$ for some of propagation and polarization directions [7]. These figures are comparable with those typical for the best acoustooptic materials (e.g., TeO_2 crystals [8]). Moreover, as shown in our recent work concerned with the acoustic anisotropy [9], the efficiency of the acoustooptic effect in Tl_3AsS_4 can become even higher if the anisotropic interaction of optical wave with the slowest transverse acoustic wave is provided. Under such conditions, the acoustooptic figure of merit can become extremely high (~ $3\times 10^{-12}\,\mathrm{s^3/kg}$). As a matter of fact, the Tl_3AsS_4 crystals are efficiently used as working elements of tunable acoustooptic filters for the infrared spectral range [10].

In spite of some information available on a number of physical properties of Tl_3AsS_4, these crystals have not been studied in sufficient detail and the corresponding literature data is still scarce. As an example, one can suppose that the thallium arsenic sulfosalt can be efficiently used in the other branches of optoelectronics different from acoustooptics. Being centrosymmetric, these crystals cannot reveal the optical effects described by the second-order optical nonlinearities (e.g., the linear electrooptic effect or the second harmonic generation). On the other hand, the effects of externally induced optical rotation such as the Faraday effect or electrogyration are allowed by the symmetry of Tl_3AsS_4 and still await their studies. The aim of the present work is to characterize the Faraday rotation in these crystals.

2. Experimental conditions and procedures

As mentioned above, the Tl_3AsS_4 crystals are optically biaxial. At the room temperature and the light wavelength of $\lambda = 632.8\,\text{nm}$, the plane containing the optic axes coincides with the crystallographic plane *ab*, where the *b* axis represents the acute bisector of the angle $2V$ between the optic axes ($2V =$ $28.46 \pm 0.1\,\text{deg}$) [9]. The Faraday rotation becomes free of accompanying optical effects whenever the light propagates along one of the optic axes and the magnetic field is applied in this direction.

Under these conditions the Faraday effect is described by the relations

$$B_{jk} = B^0_{jk} + ie_{jkl}F_{lm}H_m,\ \Delta\rho_l = \frac{\pi n_c^3}{\lambda}F_{lm}H_m,\ V_F = \frac{\pi \bar{n}^3}{\lambda}F_{lm}, \tag{1}$$

where $\Delta\rho_l$ implies the specific optical rotation angle, B_{jk} the actual (magnetically perturbed) optical-frequency dielectric impermeability tensor, B^0_{jk} the impermeability tensor in the absence of external magnetic field H_m, e_{jkl} the unit Levi-Civita tensor, n_c the refractive index for the light propagating along the optic axis, $\bar{n}$ is the mean refractive index along the direction of light propagation, V_F the Verdet constant, and F_{lm} the Faraday tensor. For the case of point symmetry group *mmm*, the latter tensor acquires the following form:

$$\begin{array}{c|ccc} & H_1 & H_2 & H_3 \\ \hline \Delta\rho_1 & \frac{\pi\bar{n}^3}{\lambda}F_{11} & 0 & 0 \\ \Delta\rho_2 & 0 & \frac{\pi\bar{n}^3}{\lambda}F_{22} & 0 \\ \Delta\rho_3 & 0 & 0 & \frac{\pi\bar{n}^3}{\lambda}F_{33} \end{array}, \tag{2}$$

where the crystallographic axes *a*, *b* and *c* correspond respectively to the principal axes *X*, *Y* and *Z* of the Fresnel ellipsoid (abbreviated as the axes 1, 2 and 3). Application of the magnetic field along the optic axis induces the two nonzero components, $H_1 = H\sin V = 0.2458H$ and $H_2 = H\cos V = 0.9693H$.

Let the light wave vector and the magnetic field directions be parallel to the optic axis. Then the magnetically induced rotation of the polarization plane reduces to

$$\Delta\rho_{Y'} = \frac{\pi n_c^3}{\lambda}F'_{22}H_{Y'}, \tag{3}$$

where F'_{22} denotes the effective Faraday component corresponding to the rotated coordinate system, of which Y' axis coincides with the optic axis:

$$F'_{22} = F_{11}\sin^2 V + F_{22}\cos^2 V = 0.06F_{11} + 0.94F_{22} = \Delta\rho_{Y'}\lambda / \pi n_c^3 H_{Y'}. \tag{4}$$

Hence, one can determine the combined Faraday coefficient $0.06F_{11}+0.94F_{22}$ for the Tl_3AsS_4 crystals, using a simple and direct technique for measuring the optical rotatory power for the light propagating along one of the optic axes.

To measure the Faraday rotation, we have employed a single-ray polarimetric technique. In our experiment, a He-Ne laser with the wavelength of $632.8\,\text{nm}$ is used as a light source. The longitudinal magnetic field is applied using an electromagnet. A crystal sample thickness is $d=2.83\,\text{mm}$. As stressed before, the change in the polarization plane rotation imposed by the Faraday effect is measured in the geometry when the light propagates along the optic axis and the magnetic field is applied along the same direction. A plane-parallel crystalline plate is placed in between the poles of electromagnet. Finally, the sample is oriented such that the centre of the conoscopic rings is aligned with the light beam centre.

3. Results and discussion

The dependence of the specific optical rotation upon the external magnetic field is presented in Fig. 1. This dependence is exactly linear, as it should be for the case of Faraday rotation. The combined Faraday coefficient calculated using a standard linear fitting of the experimental data is equal to $0.06F_{11}+0.94F_{22}=(9.23\pm0.21)\times10^{-13}\,\text{m/A}$, while the appropriate Verdet constant amounts to $V_F=(82.2\pm1.8)\ \text{rad}/\text{T}\times\text{m}$.

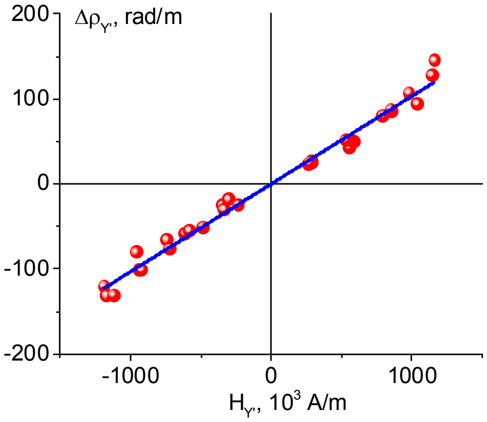

Fig. 1. Experimental dependence of specific rotatory power on the magnetic field applied to Tl_3AsS_4 crystals, and its linear fitting.

The latter values derived in our experiments should be considered as very high and so the Tl_3AsS_4 crystals can be referred to highly efficient magnetooptic materials. This is readily confirmed by a comparison with a number of well-known magnetooptic materials. For example, the Verdet constant is equal to 115 rad/T×m for $Sn_2P_2S_6$ crystals [11], $V_F=187\ \text{rad}/\text{T}\times\text{m}$ for ZnTe, and $V_F=147\ \text{rad}/\text{T}\times\text{m}$ for Cu_2O [12]. One of the best magnetooptic materials, $Tb_3Ga_5O_{12}$ crystal, is characterized by the Verdet constants $134\ \text{rad}/\text{T}\times\text{m}$ at the wavelength 632.8 nm and $36.4\ \text{rad}/\text{T}\times\text{m}$ at 1053 nm [13, 14]. They are comparable to that obtained for our crystals. Hence, Tl_3AsS_4 can be reliably classified as one of the most efficient magnetooptic materials, at least among magnetically non-ordered crystalline materials.

4. Conclusion

We have experimentally determined the Faraday coefficient and the Verdet constant for Tl_3AsS_4 under the normal conditions and at the laser wavelength 632.8 nm. Our experimental geometry, which corresponds to the light propagation and magnetic field directions parallel to one of the optic axes, is free of systematic errors. Therefore our data are reliable enough. The parameters mentioned above are equal to $0.06F_{11} + 0.94F_{22} = (9.23 \pm 0.21)\times 10^{-13}\,\mathrm{m/A}$ and $V_F = (82.2 \pm 1.8)\ \mathrm{rad/T\times m}$, respectively. Following from these values, the Tl_3AsS_4 crystals can be regarded as one of the best magnetically non-ordered materials for magnetooptic applications, with such competitors as $Sn_2P_2S_6$, ZnTe, Cu_2O, and $Tb_3Ga_5O_{12}$.

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Анотація. *У цій роботі експериментально вивчено ефект Фарадея в кристалах* Tl_3AsS_4. *Визначено ефективний коефіцієнт Фарадея та сталу Верде за нормальних умов на довжині хвилі* $\lambda = 632{,}8\ \text{нм}$. *Вони дорівнюють відповідно* $0{,}06F_{11} + 0{,}94F_{22} = (9.23 \pm 0.21)\times 10^{-13}\,\mathrm{m/A}$ *і* $V_F = (82{,}2 \pm 1{,}8)\ \text{рад/Тл}\times\text{м}$. *Показано, що кристали* Tl_3AsS_4 *можна віднести до найефективніших магніто-невпорядкованих матеріалів для магнітооптичних застосувань.*