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Thermoreflectometry of single crystals of mercury selenide in the range of 35–300 K in a fiber–optic „pump-probe“ scheme with a Fabry–Perot interferometer

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A study of the reflection of infrared radiation at a wavelength of 1530 nm from the surface of HgSe single crystals in a two-beam pump-probe fiber–optic scheme was carried out using the thermoreflectometry (TR) method with a Fabry–Perot interferometer (FP). Along with the „high-temperature“ anomaly of the probe laser signal (at $T > 100$ K) in the relaxation region, a „low-temperature“ anomaly was revealed in the heating region, consisting in a change in signal polarity at $T < 50$ K. A qualitative interpretation of the observed features of the relative intensity of the reflected signal in the region of heating and relaxation is proposed, based on the hypothesis of two types of energy barriers, separating the bulk chiral states from the Fermi-arc surface states.

Keywords: thermal reflection, interferometer, mercury selenide, Weyl nodes.

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The pump–probe method consists in using a probe beam to monitor the variation of optical characteristics of a sample irradiated with a high-power laser pump pulse. An implication of interferometer makes such measurements more sensitive and informative [1,2]. Owing to the time response of the observed phenomena, the photoreflectance effect [2] is neglected in the present study. We have described in detail in [1] an affordable compact two-beam fiber-optic device designed for the examination of near-surface layers of solids by thermoreflectometry (TR) with a Fabry–Perot (FP) interferometer at low temperatures. In the present study, we report the results of experiments with a vacuum measurement cell of an updated design that allows one to perform measurements at $T = 300$ K as well as with the temperature sweep from 35 K to 180 K. Temperature was controlled using a miniature germanium thermometer arranged close to the sample. Pulsed pump and continuous probe radiation was transported to the sample within a single fiber-optic waveguide at wavelengths of 1470 and 1530 nm, respectively (Fig. 1).

The duration of a heating laser pulse (1470 nm, the power was 0.1 W) was 10 μs (the energy was 1 μJ). Continuous radiation of the probing laser ($\lambda = 1530$ nm, the power was 1 mW) had initial intensity I_0 at the waveguide output. The end of the emitting fiber and the reflecting sample plane were separated by a gap of approximately 100 μm, forming a Fabry–Perot interferometer that acts as a phase sensor for a reflected wave of probe radiation. The lateral size of the studied region on the sample surface was 20–30 μm. Relaxation of the temperature inhomogeneity induced by pulsed heating was monitored at the interferometer output by measuring the variation of amplitude of the reflected probe signal (synchronous with a heating pulse) as a function of time $\Delta I(t)$ relative to mean value I_m at the operating point of the interferometer:

$$\Delta I(t) = I_0 R(T)\Big(1 - \cos\big(4\pi L(T)/\lambda + \varphi(T,t)\big)\Big) - I_m,$$

where T is the mean temperature, $R(T)$ is the mean reflection coefficient, $L(T)$ is the interferometer gap in vacuum, and $\varphi(T,t)$ is the phase shift of the probe beam upon reflection from the sample. We assume that $R(T)$ and $L(T)$ vary subtly at the heating pulse power used ($\Delta T \approx 5$ K); in this case, within the linear part of interferometer response an output signal variation $\Delta I(t) \propto \varphi(T,t)$. The essential stabilization of the I_m interferometer signal at the operating point in the middle of the linear section of the metering characteristic was effected by adjusting voltage U at a piezoelectric actuator. The magnitude of the voltage variations measured at the photodetector, being proportional to $\Delta I(t)$, did not exceed 0.1 V, which is inferior to the linear range of 3 V. The emergence of a non-equilibrium density of free surface charges under pulsed heating may induce a change in both absorption k of radiation on the sample surface involving refraction index n and a phase shift φ of the reflected beam, since $\mathrm{tg}\,\varphi = 2k/(1 - n^2 - k^2)$ for radiation reflected normally from a strongly absorbing medium [3]. Such variations of the phase of reflected radiation can lead to the negative values of the observed variation of signal intensity $\Delta I(t)$ [1,2].

The TR-FP method was tested in [1] during investigations performed at $T = 300$ and 77 K on several semiconductor materials with different electron spectra (ZnSe, n-InSb, and HgSe). Of the utmost interest in [1] is the observation

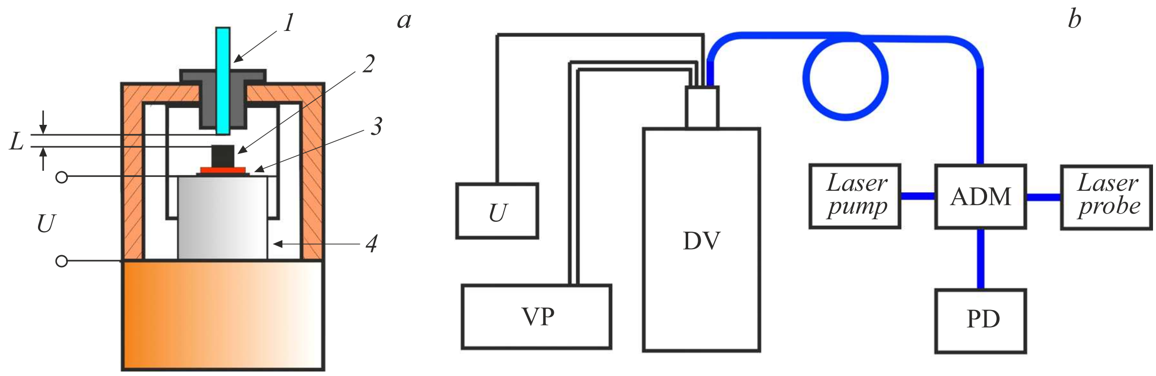

Figure 1. Measurement cell (*a*) and overall view of the setup (*b*). *1* — Waveguide, *2* — sample, *3* — heater, *4* — piezoelectric transducer, L — Fabry–Perot interferometer gap, U — piezoelectric element control voltage, VP — vacuum pump, DV — cryostat, ADM — add-drop multiplexer for pump and probe radiation, PD — photodetector of the thermoreflection signal, *Laser pump* — heating laser (0.1 W, 1470 nm), *Laser probe* — probing laser (1 mW, 1530 nm).

of a deep minimum at $T = 77$ K with a sign change in the time dependence of relative intensity of signal $\Delta I(t)$ in single crystals of gapless mercury selenide (candidate to Weyl semimetals (WSM) with broken inversion symmetry) [4,5]. A qualitative interpretation of this anomaly was constructed in reliance on the fundamental feature of WSMs: the presence of an energy barrier separating bulk chiral states of Weyl fermions from Fermi arc surface states [6]. Additionally the nontrivial shape of the spectral dependence of absorption of HgSe nanocrystals near the utilized wavelengths should be noted [7].

The present study is a continuation of the previous investigation of the near-surface region of HgSe single crystals aimed at identifying new features of the time behavior of the reflected signal. Two HgSe samples for study (samples Nos. 1 and 2) were cut from a homogeneous part of single-crystalline ingots grown by the Bridgman method. Both samples had the form of rectangular parallelepiped $1 \times 2 \times 6$ mm in size. Sample No. 2 was doped with Ga donor impurity. The parameters of samples were evaluated at $T = 77$ K. Electron density n_e, conductivity in zero magnetic field σ_0, and Hall mobility μ_{H} were determined. The obtained values were $n_e = 4 \cdot 10^{17}\,\mathrm{cm}^{-3}$, $\sigma_0 = 4.25 \cdot 10^3\,\Omega^{-1} \cdot \mathrm{cm}^{-1}$, and $\mu_{\mathrm{H}} = 6.6 \cdot 10^4\,\mathrm{cm}^2/(\mathrm{V} \cdot \mathrm{s})$ for sample No. 1 and $n_e = 4.2 \cdot 10^{18}\,\mathrm{cm}^{-3}$, $\sigma_0 = 1.53 \cdot 10^3\,\Omega^{-1} \cdot \mathrm{cm}^{-1}$, and $\mu_{\mathrm{H}} = 2.3 \cdot 10^4\,\mathrm{cm}^2/(\mathrm{V} \cdot \mathrm{s})$ for sample No. 2. These parameters agree well with the data from [8] for HgSe, thus indicating that the studied samples were of a reasonably high quality. In contrast to polished unetched HgSe samples used in [1], samples Nos. 1 and 2 were etched in a 5% solution of bromine in isobutyl alcohol for ~ 3 min prior to measurements. This was performed in order to reduce the influence on the reflection of laser radiation of surface contamination and oxide films.

Figure 2 presents dependences $\Delta I(t)$ for samples Nos. 1 and 2 at different temperatures. It can be seen that the signal decay in relaxation region II becomes more pronounced as T decreases from 300 to 110 K, and a fairly deep minimum accompanied by an inversion in $\Delta I(t)$ sign forms eventually. Alongside with the formation of a drop in $\Delta I(t)$, the value of $\Delta I_{\max}$ (maximum positive intensity for a given T recorded at $t = \tau_p$ within heating section I) decreases with T descending. As was already noted, these $\Delta I(t)$ dips in relaxation region II were observed for HgSe samples at $T = 77$ K [1]. The curves measured in the present study differ fundamentally from the ones obtained earlier in that they exhibit a $\Delta I(t)$ feature in heating region I. It follows from Fig 2 that the sign of $\Delta I(t)$ changes in region I when T is reduced to 85 K. As T decreases to 35 K, this „low-temperature“ anomaly evolves in such a way that the polarity of the entire thermoreflection signal changes.

Let us analyze the experimental results relying on the concepts of nontrivial topological nature of the electron spectrum of HgSe outlined in [1]. To interpret both „high-temperature“ and „low-temperature“ (being more sensitive to T variation) anomalies, we assume that the Brillouin zone of HgSe features Weyl nodes of type $W1$ (the distance between nodes of opposite chirality is $\Delta\kappa_{W1}$) alongside with pairs of Weyl nodes of type $W2$, which have $\Delta\kappa_{W2} < \Delta\kappa_{W1}$. Such configuration is fairly common upon well-known WSMs with broken inversion symmetry, such as TaAs [9], TaP [10], NbAs [11], and NbP [12]. Thus, the results of TR-FP experiments should be interpreted by generalizing the approach [1] with a single barrier to the case of two tunnel barriers $\mathscr{W}_1^{\pm}$ and $\mathscr{W}_2^{\pm}$ corresponding to pairs of Weyl nodes of different types with overbarrier activation energy $\varepsilon_{a1} > \varepsilon_{a2}$. It is also important to bear in mind that Fermi arcs are retained in WSMs above the Lifshitz transition (i.e., when zero chirality is achieved by varying, e.g., the Fermi energy) [13]. This implies that the overbarrier activation energy is probably nonzero even at $n_e > 10^{18}\,\mathrm{cm}^{-3}$, which is fairly high for HgSe.

The assumed presence of two different tunnel barriers between bulk and surface chiral states provides an opportunity to interpret qualitatively the „high-temperature“ and „low-

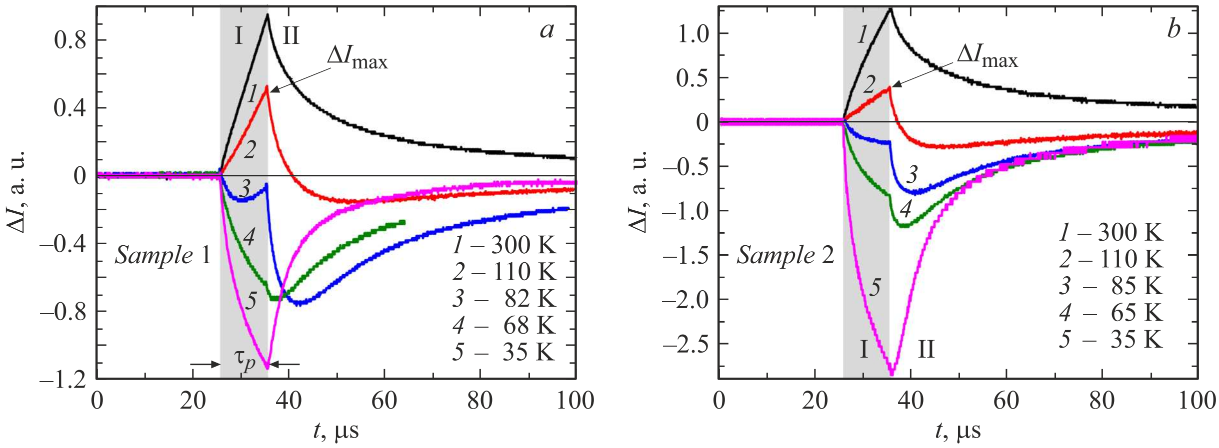

Figure 2. Time dependences of the intensity change of thermoreflection signal ΔI for HgSe single crystals in the pump–probe setup with a Fabry–Perot interferometer at different temperatures. *a* — Sample No. 1, *b* — sample No. 2. I — Heating region, II — relaxation region.

temperature" $\Delta I(t)$ anomalies. At relatively high temperatures, when condition $\varepsilon_{a2} \leqslant k_B T \leqslant \varepsilon_{a1}$ is satisfied, a $\Delta I(t)$ dip observed against the background of relaxation of the temperature inhomogeneity may be associated with barrier $\mathscr{W}_1^{\pm}$. This scenario was examined in detail in [1]. Barrier $\mathscr{W}_2^{\pm}$ is „activated" at lower T when condition $k_B T \leqslant \varepsilon_{a2}$ is satisfied, providing overbarrier activation with probability $k_2 = A_2 \exp\left(-\frac{\varepsilon_{a2}}{k_B T}\right)$ and subsequent tunnel relaxation of free surface electrons. Barrier $\mathscr{W}_2^{\pm}$ opens up an additional (new) channel of influence on the phase of the reflected wave via an increase in the non-equilibrium surface charge density. Therefore, the results of our experiment suggest that a heating pulse may induce a sharp increase in the non-equilibrium surface carrier density in HgSe due to the parallel operation of two channels of overbarrier activation with subsequent tunnel relaxation of non-equilibrium Dirac 2D electrons.

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Conflict of interest

The authors declare that they have no conflict of interest.

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