Bulk properties of semiconductors

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Photoluminescence and photoreflectance of annealed HgCdTe films with high CdTe content

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Abstract. The results of a study of photoluminescence and photoreflectance of $Hg_{1-x}Cd_xTe$ films with CdTe molar fraction $x = 0.5 - 0.7$ are presented. The films were grown by molecular beam epitaxy and annealed under mercury deficiency conditions to convert them to hole-type conductivity by generating intrinsic acceptors, mercury vacancies. Annealing resulted in the significant blue-shift of both the photoluminescence and photoreflectance spectra. Energy levels within the bandgap with ionization energies from 44 to 125 meV were detected in the films, and their nature is discussed.

Keywords: HgCdTe, annealing, photoluminescence, photoreflectance

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Фотолюминесценция и фотоотражение отожженных пленок HgCdTe с большим содержанием CdTe

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Аннотация. В статье представлены результаты исследования фотолюминесценции и фотоотражения пленок $Hg_{1-x}Cd_xTe$ с мольной долей CdTe $x = 0.5 - 0.7$. Пленки были выращены методом молекулярно-лучевой эпитаксии и подвергнуты отжигу в условиях дефицита ртути, что позволило преобразовать их в дырочный тип проводимости за счет генерации собственных акцепторов – вакансий ртути.

Показано, что отжиг привел к существенному высокоэнергетическому сдвигу спектров и фотолюминесценции, и фотоотражения. В запрещенной зоне пленок обнаружены энергетические уровни с энергией ионизации от 44 до 125 мэВ и обсуждается природа выявленных уровней.

Ключевые слова: HgCdTe, отжиг, фотолюминесценция, фотоотражение

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Introduction

$Hg_{1-x}Cd_xTe$ (MCT) solid solutions with a high ($x \geq 0.5$) CdTe content (“composition”) are required in the development of near-infrared (IR) photodetectors for astronomical observations [1], and for fabrication of barrier layers in lasers [2] and *nBn* photodetectors [3]. For the fabrication of both photodetectors and injection lasers, a hole-type conductivity material is required; however, approaches to acceptor doping of MCT with $x \geq 0.5$ are just being sought [4, 5]. An obvious (as in the case of MCT with $x < 0.5$) option there could be the introduction of intrinsic acceptors, mercury vacancies V_{Hg}, by annealing the material under mercury deficiency conditions. We carried out such annealing at temperatures T from 330 to 440 °C for a series of MCT films with $x = 0.5 - 0.7$, and in [6], the results of the studies of the structural properties of the films, their optical transmittance at $T = 294$ K, and photoluminescence (PL) at $T = 103$ K and $T = 294$ K were reported. In the present paper, we report the results of the detailed studies of the optical properties of the annealed films carried out with PL and photoreflectance (PR) methods in a wide temperature range, 30 to 300 K.

Materials and Methods

The 3 to 4 µm-thick films were grown by Molecular Beam Epitaxy (MBE) on (013)GaAs substrates [7] and had a constant composition (determined with *in situ* ellipsometry) across their thickness. Optical studies were performed using the setup described in [8] and based on a VERTEX 80 Fourier-transform IR spectrometer with cooled InGaAs, InSb or HgCdTe photodetector. PL/PR studies were carried out under excitation/modulation with an 809 nm cw laser diode with 2.5 kHz modulation frequency, which provided power density up to 4.0 W/cm^2. During the measurements, the films were placed in a Janis CCS-150 closed-cycle cryostat.

Results and Discussion

Fig. 1, *a* shows low-temperature ($T = 30$ K) PL spectra of the films before and after annealing. With x increasing, a shift in the PL spectra towards higher energies was observed. All spectra generally consisted of two bands: an intense narrow high-energy (HE) peak and a less intense broad low-energy (LE) band. For the as-grown films, the full widths at half maximum (FWHM) of the HE peaks were 22, 31, and 34 meV for films with $x = 0.50$, 0.60, and 0.70, respectively; these values and the increase in FWHM with increasing x were consistent with the PL data obtained earlier at $T = 103$ K [6]. For the as-grown films with $x = 0.50$ and 0.60, the broad LE bands were weakly pronounced and separated from the HE peaks by ~111 meV ($x = 0.50$) and ~76 meV ($x = 0.60$), respectively. For the as-grown film with $x = 0.70$, the LE band was separated from the HE peak by ~115 meV.

As a result of annealing, the HE peaks for the films with $x = 0.50$, $x = 0.60$ and $x = 0.70$ narrowed to 17 meV, 20 meV and 22 meV and shifted towards higher energies by 6 meV, 37 meV and 40 meV, respectively. For a film with $x = 0.50$, a LE peak with FWHM ~79 meV appeared, separated from the HE peak by ~44 meV. A broad, low-intensity LE feature was observed in the spectra of the annealed films at a distance of ~128 meV from the HE peak for the film with $x = 0.60$ and ~105 meV for the film with $x = 0.70$, respectively.

Fig. 1, *b* shows the PL spectra of the annealed film with $x = 0.70$ recorded at excitation laser power $P_{las} = 81.2$ mW at different temperatures. It is seen that the PL signal intensity for both the main HE peak and the LE band decreased with T increasing.

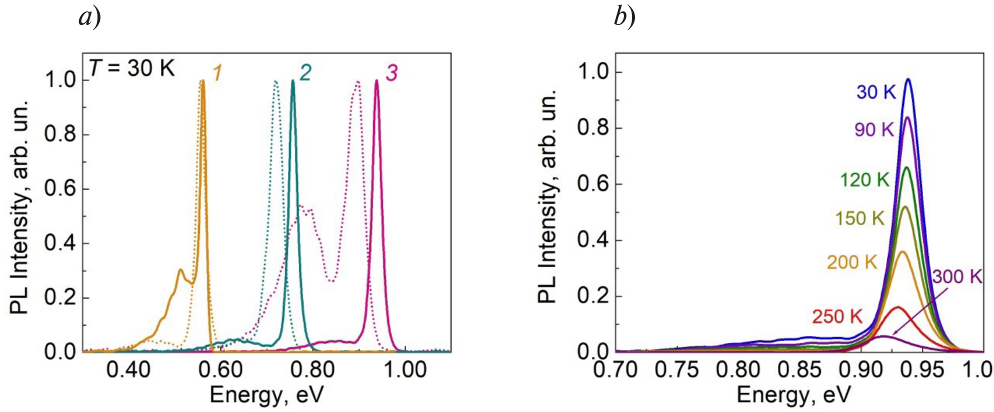
a) *b*)

Fig. 1. Normalized PL spectra of the films with $x = 0.50$ (*1*), 0.60 (*2*) and 0.70 (*3*) at $T = 30$ K (dots, as-grown films; solid curves, annealed films) (*a*), and PL spectra of the film with $x = 0.70$ at different temperatures (*b*)

Fig. 2 shows the PL spectra of the film with $x = 0.70$ at $T = 30$ K and different values of P_{las}. For all the as-grown films including this one (Fig. 2, *a*), with P_{las} increasing the intensity of the HE peaks increased more rapidly than that of the LE bands. A similar effect was observed for the annealed film with $x = 0.70$ (Fig. 2, *b*). The energy of the HE peaks and their FWHM changed with P_{las} insignificantly.

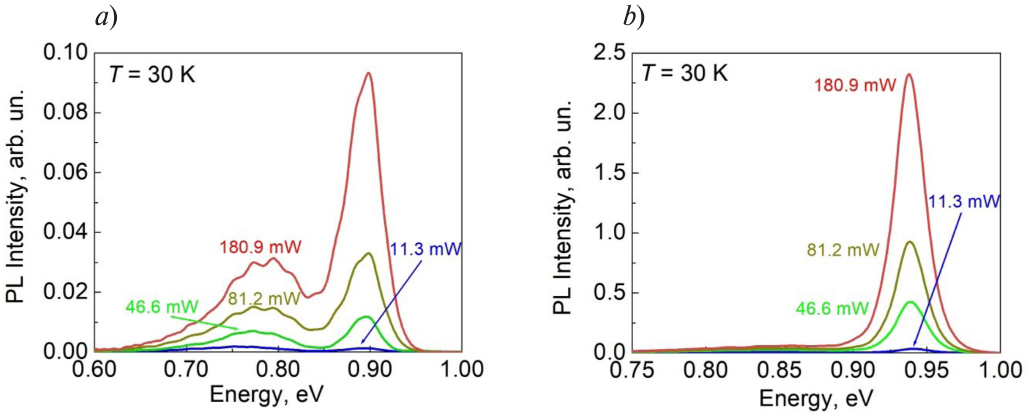
a) *b*)

Fig. 2. PL spectra of the film with $x = 0.70$ recorded at $T = 30$ K under different excitation power values: as-grown film (*a*), annealed film (*b*)

The temperature dependence of the HE PL peak positions for the studied films (see Fig. 3, *a* for the detailed data for the film with $x = 0.70$ and Fig. 3, *b* for the data for all films) did not follow the temperature dependence of the bandgap $E_g(T)$. The latter was calculated using the well-known empirical formula from [9]. A similar effect was previously observed for MCT films with $x = 0.6 - 0.7$ grown both by MBE and liquid-phase epitaxy, and is believed to be caused by the effect of carrier localization in potential wells induced by compositional fluctuations [10].

Fig. 4, *a* shows PR spectra of the film with $x = 0.70$ recorded at $T = 300$ K before and after annealing. Both spectra exhibited a superposition of the Fabry–Pérot interference and the differential signal near the interband transition. The energy of the latter, i.e., the optical bandgap E_g' was extracted from the spectra using the approach developed in [11]. However, the overlap of the signal from the interband transition and the interference in the PR spectra limits the accuracy of determining E_g'. It is seen that the spectrum shifted after annealing towards higher energies by ~31 meV, similar to the shift of the PL HE peaks. A similar effect was observed for the PR

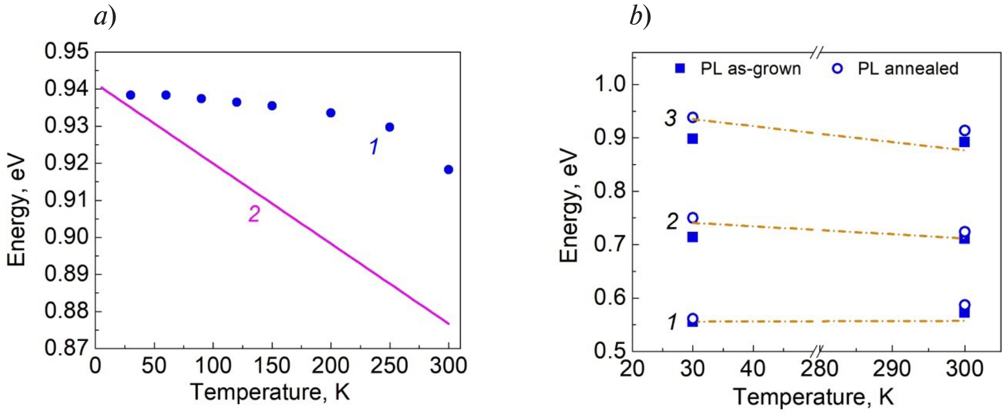

Fig. 3. Temperature dependence of the position of the HE peak in the PL spectra of the film with $x = 0.70$ (symbols *1*) and calculated $E_g(T)$ dependence for MCT with $x = 0.70$ (curve *2*) (*a*), and a review of the positions of the HE peaks for the films with $x = 0.50$ (*1*), 0.60 (*2*) and 0.70 (*3*) in respect to the calculated $E_g(T)$ dependences (lines) (*b*)

spectra of the film with $x = 0.60$. Considering the accuracy of determining the values of E_g' from the PR spectra, they reasonably corresponded to the positions of the HE peaks of the PL for all the films. Since the PR method is sensitive only to interband transitions, it could be concluded that the HE peaks in the PL spectra, including those at low temperatures, also reflected the value of E_g' and were not associated with transitions to energy states within the bandgap.

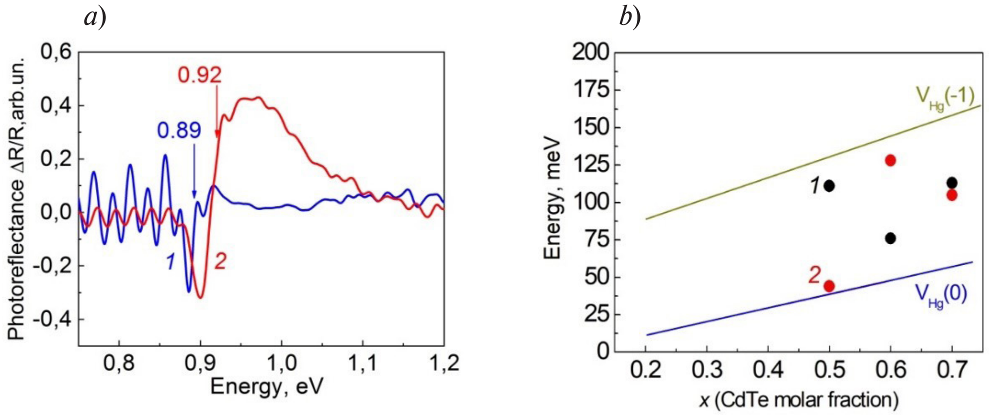

Fig. 4. PR spectra of the film with $x = 0.70$ before (*1*) and after (*2*) annealing, with arrows showing the optical bandgap (*a*), and the values of acceptor ionization energy E_a according to our PL data (symbols: *1*, as-grown films, *2*, annealed ones) and the extrapolation of empirical $E_a(T)$ dependences from [5] to large *x* values (*b*)

The latter gave us a reference point, allowing to proceed to the analysis of energy levels in the bandgap. According to the data of the Hall effect study performed at $T = 77$ K, the as-grown films had *n*-type conductivity with electron concentration $(3 - 60)\cdot10^{14}$ cm^{-3}; as a result of annealing, the films were converted into *p*-type with hole concentration $(2 - 7)\cdot10^{15}$ cm^{-3} [6]. Results of carrier lifetime measurements also suggested that annealed films were converted into *p*-type conductivity: the lifetime dropped ~1000 times in the annealed films with respect to its value of ~50 μs in the as-grown ones. According to these data, we could expect to observe band(s) related to V_{Hg}-induced energy states in the PL spectra of the annealed films. At the same time, as follows from Fig. 1 and 2, the PL spectra of these films already contained a number of LE features related to levels in the bandgap. We plotted the experimental data on the tentative positions of the LE band maxima in Fig. 4, *b* along with the composition dependences of the ionization energies of neutral and single-charged V_{Hg} as adapted from [5]. It is seen in Fig. 4, *b* that our experimental data do not exactly follow the dependences for the two charge states of V_{Hg}. Still, for the annealed film with $x = 0.50$ ionization energy E_a ~ 44 meV can be related to the energy level formed by the neutral V_{Hg}. The experimental value E_a ~ 113 meV for the as-grown film with $x = 0.50$ is in good agreement with a similar value measured for a vacancy-doped MCT sample with the same x in [5] (115 meV). For the films with $x = 0.60$ and $x = 0.70$, however, the observed energy levels could not be directly associated with V_{Hg} as a point defect. Extrapolation of the extrinsic acceptor ionization energy $E_a(x)$ from [12] towards $x = 0.70$ yields a maximum value $E_a \approx 20$ meV. The energy of donor levels in MCT with $x = 0.70$ according to the hydrogen-like model is even lower, ~7 meV [12]. Thus, the observed energy levels should be attributed to native acceptors, or to deep centers that are not described by the hydrogen-like model. For example, a strong LE band in the as-grown film with $x = 0.70$, which disappeared after annealing, was similar in behavior to the band with $E_a = 25$ meV observed in films with $x = 0.30$, where, according to the findings in [13], it was associated with defects caused by the presence of excess tellurium [14]. All these observations suggested the presence of a number of intrinsic defects/complexes in the studied films with the evolution of the defect pattern under annealing. Possibly, V_{Hg} manifestation in the PL spectra of vacancy-doped MCT films with $x > 0.5$ is not as straightforward as in the low-x material.

Conclusion

Using photoluminescence and photoreflectance, we studied optical properties of $Hg_{1-x}Cd_xTe$ films with a high ($x = 0.5 - 0.7$) CdTe content before and after thermal annealing. The annealing resulted in a shift of the spectra towards higher energies, which was indicative of the increase in the CdTe molar fraction of the films, possibly due to the reduction in the compositional fluctuations. Despite the fact that electrical and lifetime measurements showed conversion of conductivity type from *n*- to *p*- in all the annealed films, the photoluminescence studies revealed clear signatures of generated mercury vacancies only in the film with $x = 0.5$. Still, clear signs of the changes in the defect pattern of all the films were observed after annealing. Further studies are required for the clear identification of optical signatures of mercury vacancies in $Hg_{1-x}Cd_xTe$ films with $x > 0.5$.

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