Far infrared properties of PbTe doped with Hg

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Single crystal samples of PbTe doped with Hg were grown using the Bridgman method. Far infrared reflectivity spectra were measured at room temperature for samples with 0.5 at. % Hg; 0.9 at. % Hg and 1.4 at. % Hg. The plasma frequency decreased when PbTe was doped with Hg and it was lowest for the PbTe sample doped with 0.5 at. % Hg. The values of the determined optical free carrier mobility increased and was the highest for PbTe doped with 0.5 at. % Hg.

1. Introduction

Some time ago it was shown that alloying PbTe with HgTe resulted in a decrease of the free carrier concentration of PbTe [1, 2]. This is because Hg atoms occupy the sites of Pb vacancies during crystal growth and also during sample annealing in Hg atmosphere. Photovoltaic detectors of $Pb_{0.97}Hg_{0.03}Te$ were produced [3] by Sb^+ ion implantation and the mode photodiodes showed a greater sensitivity than PbTe diodes produced in the same way. The sample composition cannot be determined using X-ray methods as the lattice parameters of PbTe and HgTe are practically identical, both having a value close to 0.646 nm. So the composition change can be determined either by EDS or ICP (inductively-coupled plasma) methods. The HgTe compound is a well-known zero-gap semiconductor [4] and its dielectric function has been studied. Lead telluride doped with Hg gave rise to a strong resonant state near the bottom of the conduction band [5] and suppressed the electronic density of states (DOS) near the top of the valence band making this system a promising n-type thermoelectric.

2. Experimental

Single crystals of PbTe doped with Hg were grown using the standard Bridgman method in an evacuated sealed quartz tube using a mixture of high purity elements (6N) of Pb and Te and 5 at. % of HgTe powder. The obtained ingot was cut into slices, about 2 mm thick, which were polished using diamond paste. The content of Hg, Pb and Te, in each sample, was determined using EDS analysis. Far infrared reflectivity measurements were made using a Bruker IFS-113V spectrometer. The microcrystallinity of the samples and their orientation (close to plane (200)) was checked by X-ray diffraction method. The hot point method was used to establish that all samples were of the p-type.

3. Results and discussion

The room temperature reflectivity diagrams, as a function of the wave number for single crystal PbTe samples doped with 0.5 at. % Hg, 0.9 at. % Hg and 1.4 at. % Hg are given in Fig. 1.
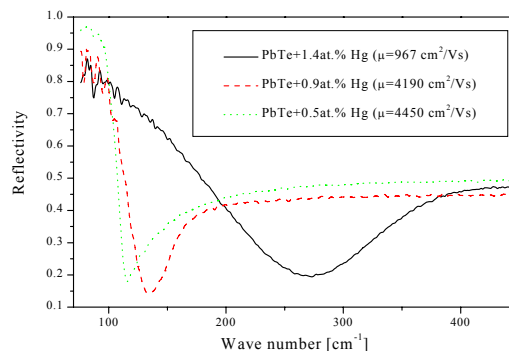

Fig. 1 Room temperature FIR spectra of PbTe doped with 0.5; 0.9 and 1.4 at. % Hg.

For these three spectra plasma minimums were observed at about 110 cm^{-1}, 130 cm^{-1} and 260 cm^{-1}, respectively. It is obvious that the plasma minimum moves towards the higher wave number when the content of Hg was increased from 0.5 at. % to 1.4 at. % of Hg. These FIR spectra were numerically analyzed and each spectrum was interpreted with a frequency dependent dielectric function.

A four-parameter model [6] was used where a combined plasmon-LO phonon mode was observed [7]. The determination of the LO mode is connected with elimination of the free carrier influence, so the dielectric function takes into account the presence of a plasmon-LO phonon interaction:

$$\varepsilon(\omega) = \varepsilon_\infty \frac{\prod_{j=1}^{2}\left(\omega^2 + i\gamma_{lj}\omega - \omega_{lj}^2\right)}{\omega\left(\omega + i\gamma_p\right)\left(\omega^2 + i\gamma_t\omega - \omega_t^2\right)} x \prod_{n=1}^{p}\frac{\left(\omega^2 + i\gamma_{Ln}\omega - \omega_{Ln}^2\right)}{\left(\omega^2 + i\gamma_{On}\omega - \omega_{On}^2\right)} \prod_{k=1}^{q}\frac{\left(\omega^2 + i\gamma_{LOk}\omega - \omega_{LOk}^2\right)}{\left(\omega^2 + i\gamma_{TOk}\omega - \omega_{TOk}^2\right)} \quad (1)$$

γ_{lj} and ω_{lj} from the first numerator should be interpreted as the damping coefficients and eigenfrequencies of the longitudinal plasmon-phonon waves resulting from the interaction of initial modes. The parameters, which belong to the first determinator, correspond to similar characteristics of transverse (TO) vibrations, while γ_p describes the plasmon mode damping coefficient in the limit of zero frequencies. ε_∞ denotes the contribution of excitations at high frequencies relative to the spectral interval of interest. The second term in Equation (1) represents the impurity local modes. Finally ω_{TO} and ω_{LO} are the transverse and longitudinal frequencies, and γ_{LO} and γ_{TO} the damping factors of uncoupled modes of the host PbTe crystal. Best-fit values of these parameters were calculated. The reflectivity spectra were measured down to 50 cm^{-1}, so the value at 32 cm^{-1} for the transverse phonon frequency ω_t was taken from literature [8]. The values of plasma frequency were calculated using the following equation [7]:

$$\omega_p = \frac{\omega_{l1} \cdot \omega_{l2}}{\omega_t} \quad (2)$$

The starting values of all parameters used in the fitting procedure were first determined using Kramers-Krönig analysis [9, 10]. As an example of the fitting procedure Fig. 2 shows the far infrared reflectivity spectrum of a PbTe sample doped with 0.5 at. % Hg where the measured values are given with dashed line and the calculated values are given with a solid line. Values of the optical parameters calculated using equations (1) and (2) are given for all three samples in Table 1.

Table 1. Optical parameters determined for PbTe doped with Hg (all values are given in cm^{-1}, except μ_p which is in cm^2/Vs and ε_∞).

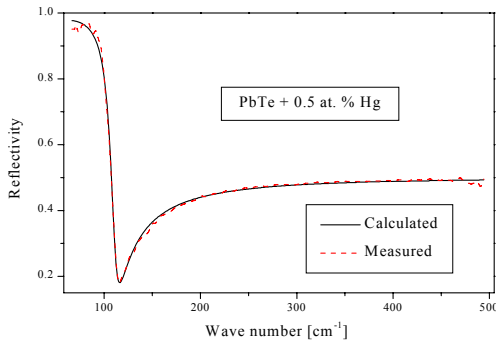|	ω_{l1}	γ_{l1}	ω_{l2}	ω_{O1}	γ_{O1}	ω_{L1}	γ_{L1}	ω_p	ε_∞	μ_p
PbTe+0.5 at. % Hg	111.5	11	30	340	629	460	382	105	31.2	4450
PbTe+0.9 at. % Hg	129	15.2	30	387	528	496	582	121	26	4190
PbTe+1.4 at. % Hg	259.8	102	30	422	144	521	695	243	30.4	967

Fig. 2 Measured (dashed line) and calculated (solid line) room temperature reflectivity diagram of a PbTe sample doped with 0.5 at. % Hg.

The optical free carrier mobility was calculated using the method of Moss et al. [11]. It was the highest for PbTe doped with 0.5 at. % Hg. The position of the plasma frequency for PbTe doped with only 0.5 at. % Hg was the lowest (105 cm^{-1}) and it increased to above 240 cm^{-1} when the content of Hg was increased to 1.4 at. % Hg. Obviously there is an optimal at. % of Hg which could give the best properties of PbTe doped with Hg. Judging by the position of the plasma minima and the values of the optical free carrier mobility, given in Table 1, one can expect it to be about 0.5 at. % of Hg or even less. Here one should take into account that for pure PbTe the position of plasma frequency is about 400 cm^{-1} [12].

4. Conclusions

In this work far infrared reflectivity spectrum of PbTe crystal sample doped with Hg were measured and analyzed. Looking at the position of the plasma frequency for PbTe doped with Hg in Fig. 1 one can see that plasma frequency decreased from about 240 cm^{-1} for the sample with 1.4 at. % Hg to the value of only 105 cm^{-1} when the content of Hg was decreased to 0.5 at. % Hg. Obviously the influence of Hg was positive and its optimal concentration is expected to be around 0.5 at. %. For the optimal concentration of doped Hg, the properties of PbTe

can be significantly improved. That could be important for the production of IR detectors in modern astronomy.

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