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Far infrared properties of iron doped single crystal PbTe

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Far infrared reflectivity spectra of single crystal PbTe samples doped with 0.2 at% Fe were measured in the temperature range between 10 K and 300 K. These spectra show very sharp and narrow dips below 120 cm^{-1}. The experimental data were numerically analyzed using a fitting procedure based on the plasmon-phonon interaction model. Two local modes were observed. The first one was noted between 130 cm^{-1} and 170 cm^{-1} depending on the temperature and the second more frequency stable one was noted at about 230 cm^{-1}.

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1. Introduction

Lead telluride has been intensively investigated experimentally and theoretically as it is a very interesting material for infrared optoelectronics [1]. Its applications were restricted because it grows with a high deviation from stoichiometry with defects which are electrically active. Doping lead telluride with various elements improved its properties a lot. For instance, when PbTe is doped with some group III impurities such as In, Ga or B it has some very interesting properties: Fermi level pinning and persistent photoconductivity [2, 3]. Introduction of indium, for instance, leads to the formation of an impurity energy level. If the concentration is high enough then it leads to Fermi level pinning of the impurity level. Similar behavior was observed in PbTe doped with Cr and Mn [4,5] concerning Fermi level pinning in the conduction band. But besides that effect produced by the impurity states, a negative magnetoresistance at low temperature was observed for PbTe doped with Mn and Cr.

A giant negative magnetoresistance effect in PbTe doped with Yb and Mn was also observed [6] and since then more attention has been paid to PbTe doped with rare earth elements [7, 8, and 9]. Besides Mn, some other transition elements were recently used to dope PbTe (Cr, Ni and Fe) [10, 11, 12]. There it was shown that an optimal concentration of dopant Ni can decrease the free carrier concentration even below 10^{17} cm^{-3} with their increased mobility to about 15000 cm^2/Vs at 80 K. Also the position of its plasma frequency in infrared reflection spectra decreases to about 115cm^{-1}. But the plasma frequency damping factor was relatively small. At room temperature it was 70 cm^{-1} and it decreased to about 10 cm^{-1} at 50 K. Only one preliminary paper on optical properties of PbTe doped with 0.04 A% iron gave a rather high free carrier mobility of about 5500 cm^2/Vs at 300 K. So, we decided to increase the percentage of Fe dopant.

In this work we present far infrared reflection spectra of single crystal PbTe doped with 0.2 at% Fe measured in the temperature range between 10 and 300 K. The experimental results were numerically analyzed.

2. Experimental

Single crystal ingots of PbTe doped with a starting composition of 3 At% Fe was synthetized using the standard Bidgman method. High purity elements (6N) were used as the source material. The composition of Fe was decreased from the top to the bottom along the length of the ingot; so samples with different iron contents could be cut and prepared for measurements. The content of Fe in each sample was determined using either ICP or EDS analysis. It was generally between 0.02 at% and 0.3at%. Sample doped with 0.2 at% had the smallest plasma frequency and also the smallest values of the plasma frequency damping factor. We measured far infrared reflectivity of PbTe samples doped with various quantity of Fe and determined the optimal concentration of this dopant as 0.2 at%Fe. Freshly cleaved sample was used for reflectivity measurements. Far infrared reflectivity measurements were made at temperatures from 10 K to 300 K using a Brucker IFS-113V spectrometer equipted with an Oxford Instruments cryostat.

3. Results and discussion

The measured far infrared reflectivity spectra of single crystal PbTe doped with 0.2 at% Fe are shown in Fig. 1. A sharp dip at wave numbers between 115 cm^{-1} and 120 cm^{-1} was observed for each temperature, which is less exposed only for room temperature.

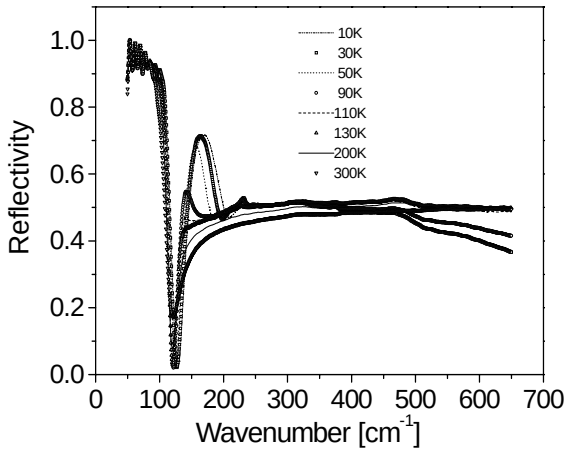

Fig. 1. FIR reflectivity spectra of PbTe doped with 0.2at%Fe at temperatures 10 K-300 K.

The room temperature plasma minimum is at the lowest wave number and it moves slightly towards higher wave numbers when the temperature decreases. A local impurity mode was clearly exposed for the reflectivity diagrams measured at 110 K and lower temperatures. It was the strongest at 10 K, seen at about 170 cm^{-1}, and the weakest at about 130 cm^{-1} for 110 K. The change of its position is a consequence of strong phonon interactions. At about 230 cm^{-1} another mode can be seen in the same temperature range and it did not change so much its position with temperature changes.

All experimental diagrams were numerically analyzed using a modified four parameter model for the dielectric function [13] which takes into account the presence of plasmon-phonon interactions [14]:

$$\varepsilon(\omega)=\varepsilon_\infty \frac{\prod_{j=1}^{2}(\omega^2+i\gamma_{lj}\omega-\omega_{lj}^2)}{\omega(\omega+i\gamma_p)(\omega^2+i\gamma_t\omega-\omega_t^2)}\prod_{n=1}^{p}\frac{(\omega^2+i\gamma_{Ln}\omega-\omega_{Ln}^2)}{(\omega^2+i\gamma_{On}\omega-\omega_{0n}^2)} \quad (1)$$

$$\prod_{k=1}^{q}\frac{(\omega^2+i\gamma_{LOK}\omega-\omega_{LOK}^2)}{(\omega^2+i\gamma_{TOK}\omega-\omega_{TOK}^2)}$$

where ω_{lj} and γ_{lj} are parameters of the first numerator which represent the eigenfrequencies and damping factors of plasmon-longitudinal phonon waves, respectively. First denominator parameters correspond to transversal (TO) vibrations; γ_p is the damping factor of plasma; ε_∞ represents high frequency dielectric permittivity compared to the measured FIR range. The oscillators denoted by j=1, 2 are dominant structures and represent the position of the coupled plasmon-LO phonon modes. The second term in equation (1) represents the iron impurity local modes. Finally, ω_{LOK} and ω_{TOK} are the longitudinal and transverse frequencies and γ_{LOK} and γ_{TOK} are damping factors of the host crystal uncoupled modes. 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 [2].

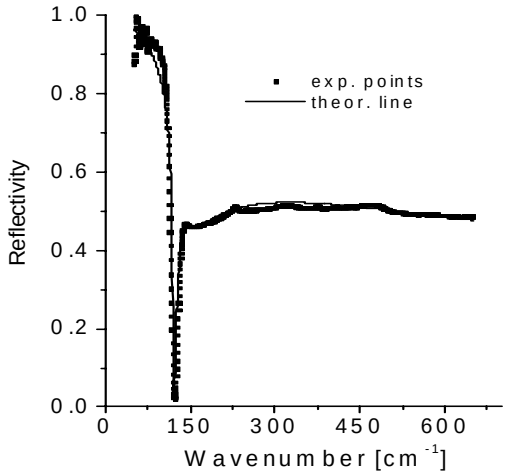

Fig. 2. Far infrared spectrum of PbTe doped with 0.2at%Fe as a function of the wavenumber, measured at T=110 K.

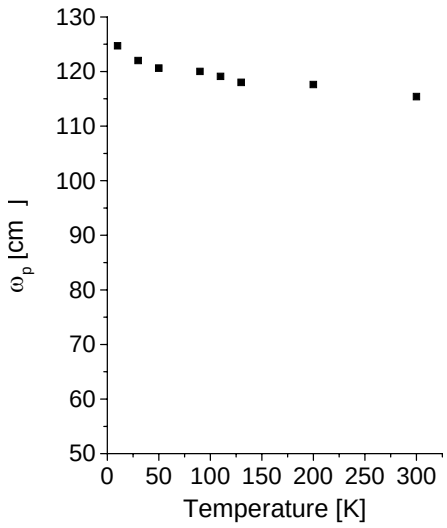

Fig. 3. Plasma frequency ω_p versus temperature.

The values for ω_p were calculated using the Kukharski [14] method. The values of all parameters were calculated from a least squares fit to the experimental reflectivity data. As an example a fitted curve for FIR spectrum of PbTe doped with iron measured at T=110 K is given in Fig. 2.

In Fig. 3 the calculated values of plasma frequencies versus temperature are given and Fig. 4 shows change of plasma frequency damping factor versus temperature. The plasma frequency is relatively stable and it slightly increases from room temperature to 10 K. Its damping factor given in Fig. 4 decreases fast starting from room

temperature until about 130 K and then stabilizes at the value of about 3cm^{-1}. Knowing that the plasma frequency for pure PbTe is about 300 cm^{-1} one can conclude that iron, as a dopant, has a strong influence because it shifts the plasma frequency to much lower values.

Judging by the low value of plasma frequency, one can expect that free carrier mobility is very high and that it increases when the temperature decreases. The free electron room temperature concentration and mobility were calculated using the method of Moss at al [15] which was slightly modified by Dionne and Wooley [16]. The calculated values of the mobility (Fig. 5) are very high compared with the carrier mobility of pure PbTe. Only for PbTe doped with 0.2 at%Cr higher values for mobility but at a lower temperature, 4.2 K was reported [10]. The plasma frequency damping factor for our sample is about five times smaller compared with our recent study when PbTe was doped with Ni [11].

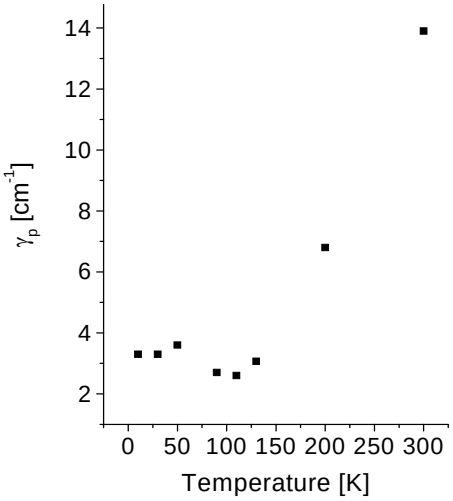

Fig. 4. Plasma frequency damping factor vs. temperature.

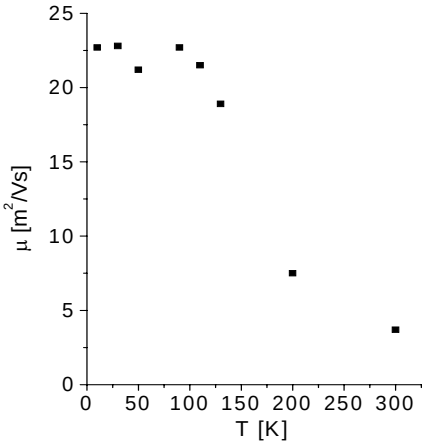

Fig. 5. Optical electron mobility vs. temperature.

Concerning local modes for Fe we have observed two local modes clearly at temperatures below 110 K (Fig. 1). At 10 K a strong phonon can be seen at about 170 cm^{-1} whose frequency decreases to about 130 cm^{-1} at 110 K (Fig. 6). The second mode is much weaker but much more stable at about 230 cm^{-1}. A third local mode is possibly at about 470 cm^{-1}, but judging by the fact that the reflectivity decreases in the range above 500 cm^{-1} for sample temperatures below 110 K, the observed decrease could be associated with a phase transition produced by the impurity, i.e. Fe. The Fe ions could behave as if they are off-centers.

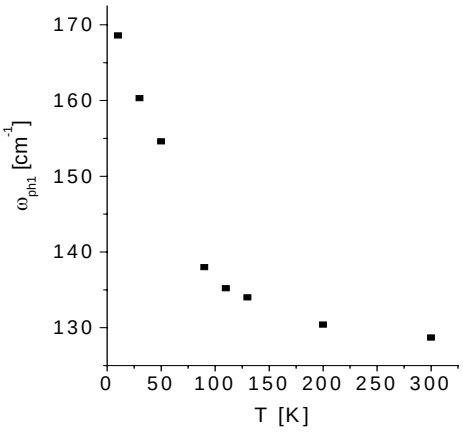

Fig. 6. First local mode frequency vs. temperature.

Since lead telluride alone always has a rather high concentration of defects, Fe impurity ions substitute for Pb atoms. All these ions in PbTe are not at the center of inversion symmetry. When Fe ions with a lighter mass, substitute heavier Pb one gets two modes. We believe that Fe^{+2} enters as neutral or Fe^{+1} as an acceptor and they change the energy states of impurity centers influencing on the local mode frequency. We assume that the mode about 170 cm^{-1} at 10 K corresponds to Fe^{+2} and the other mode at about 230 cm^{-1} belongs to Fe^{+1}.

4. Conclusion

In this work far infrared reflection spectra of single crystal PbTe doped with 0.2 at% Fe at temperatures between 10 K and 300 K have been measured and numerically analyzed using a least squares fit. These spectra show very sharp and narrow dips at wave numbers between 115 cm^{-1} and 120 cm^{-1}. The calculated values of those plasma frequency damping factors were very small as a consequence of very high free carrier optical mobility. Two well exposed local modes were observed. The first one was in the range between 130 cm^{-1} and 170 cm^{-1}, while the second one was more frequency stable at about 230 cm^{-1}.

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