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Far-infrared study of impurity local modes in Ni-doped PbTe

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Abstract

In this paper, we present far-infrared reflection spectra of Ni doped PbTe single crystal samples. Crystals of PbTe(Ni) were grown by the Bridgman method. These spectra were analyzed using a fitting procedure based on the modified plasmon–phonon interaction model. Together with the strong plasmon–phonon coupling we obtained three local modes of Ni at about 130, 165 and 190 cm^{-1} which correspond to the impurity atoms in different valence states. The positions of these modes depend on impurity center charge, and their intensities depend on temperature and Ni concentration.
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1. Introduction

The doping of $A^{IV}B^{VI}$ semiconductor compounds with transition metal impurities has significant scientific and practical interest due to the new materials preparing possibilities. Lead telluride (PbTe) belongs to the $A^{IV}B^{VI}$ IR-sensitive narrow-gap semiconductors group, which acquire new properties in consequence of doping [1,2]. Transition metals behave either as donors (Cr, Co, Ni) [3] or neutrals (Mn) [4]. For example, when PbTe is doped with chromium, free carrier concentration n increases to 1.3×10^{19} cm^{-3} and remains unchanged with further increase of the Cr concentration, up to the chromium solubility limit.

The direct proof of mixed valence existence, in the case of third group periodical system elements (B, Ga and In) as dopands in PbTe, is obtained using vibrational spectroscopy methods. In our earlier papers [5,6], we analyzed the far-infrared reflection and Raman scattering spectra. We registered three local modes of impurities. The positions of this modes, depends on sort of impurity and impurity center charge.

In this paper, we present far-infrared reflection spectra of Ni doped PbTe single crystal samples. The Ni concentration in the samples used here were 1×10^{19} and 2×10^{19} at./cm^3. These spectra were analyzed using a fitting procedure based on the modified plasmon–phonon interaction model. Together with the strong plasmon–phonon coupling we obtained three local modes of Ni, which correspond to the impurity atoms in different valence states.

2. Samples and experiment

Single crystals of PbTe(Ni) were grown by the Bridgman method in evacuated sealed 12–15 mm quartz tubes with a cone shaped lower end by moving in the temperature gradient of 15–20 K/cm. The impurity concentration in the starting mixture were 3.3×10^{20} and 4.6×10^{20} at./cm^3. The Ni concentration in the crystals used here were 1×10^{19} and 2×10^{19} at./cm^3, respectively, determined by chemical analysis. Solubility limit and segregation effect for Ni doped PbTe were discussed in more details in Ref. [7].

The structural characteristics were obtained by the XRD powder technique. All the samples were examined under the same conditions, using a Philips PW 1729 X-ray generator, a

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Phylips 1710 diffractometer and the original APD software. The radiation source was an X-ray LLF tube with copper radiation and graphite monochromator. The radiations were λCuKα1 = 0.154178 and 0.154438 nm. The anode tube load was 40 kV and 30 mA. Slits of 1.0 and 0.1 mm were fixed. For product identification, the MPDS program and JCPDS (ASTM) (Joint Committed on Powder Diffraction Standards) cards files were used. The unit cell was calculated by the least square methods. All the registered reflections corresponded to PbTe crystals and gave the parameter of the cubic unit cell of $a = 0.6455(3)$, which are in good agreement with values cited in literature [8]. The dislocation density were (5–7) $\times 10^{-4}$ cm^{-2}, registered by selective etching [9].

The Hall effect and conductivity measurements showed that the both crystals exhibit n-type conductivity with room-temperature electron concentration of 6×10^{16} cm^{-3} (in samples with Ni concentration of 1×10^{19} at./cm^3) and 4×10^{16} cm^{-3} (sample with 2×10^{19} at./cm^3).

The far-infrared measurements were carried out with a BOMEM DA-8 FIR spectrometer. A DTGS pyroelectric detector was used to cover the wave number rang from 40 to 400 cm^{-1}.

3. Results and discussion

The far-infrared reflection spectra of the Ni-doped PbTe single crystal samples are shown in Fig. 1. The experimental data are represented by circles. We can clearly see the difference in shape of reflection spectra for these two samples. For PbTe $+1 \times 10^{19}$ at./cm^3, concentration of free carriers is high, plasma minimum occurs on higher wave numbers and the phonon structure is screened (Fig. 1a). With the Ni concentration increase, the concentration of free carriers decreases [7], plasma minimum shifts to lower wave numbers, and the phonon structure becomes very clear (Fig. 1b).

In principle, the spectra might be interpreted with the help of a frequency dependent dielectric function with not less than four classical oscillators corresponding to the TO modes superimposed by Drude part taking into account the free-carrier contribution. In this way the fit immediately yielded the TO mode frequencies. LO modes are determined by the maximum of the dielectric loss function. In our case the pure LO modes of the lattice are strongly influenced by the plasmon mode (ω_P) of the free carrier. As a result, a combined plasmon–LO phonon mode should be observed [10]. Therefore the determination of LO mode is connected with the elimination of free carrier influence. Impurity local modes have the same behavior. Considering this fact, we have decided that in analysis of experimental results in Fig. 1 we must use a dielectric function that takes into account the existence of plasmon–LO phonon interaction. The lines in Fig. 1 were obtained using a dielectric function:

$$\varepsilon_f(\omega) = \varepsilon_\infty \frac{\prod_{j=1}^{2}(\omega^2 + i\gamma_{jl}\omega - \omega_{lj}^2)}{\omega(\omega + i\gamma_P)(\omega^2 + i\gamma_t\omega - \omega_t^2)} \times \prod_{n=1}^{p} \frac{\omega^2 + i\gamma_{Ln}\omega - \omega_{Ln}^2}{\omega^2 + i\gamma_{tn}\omega - \omega_{0n}^2} \prod_{k=1}^{2} \frac{\omega^2 + i\gamma_{LOk}\omega - \omega_{LOk}^2}{\omega^2 + i\gamma_{TOk}\omega - \omega_{TOk}^2} \quad (1)$$

The ω_{lj} and γ_{lj} parameters of the first numerator should be interpreted as the eigenfrequencies and damping coefficients of the longitudinal plasmons–phonon waves, which arise as a result of the interaction of the initial modes. The parameters of the first denominator correspond to similar characteristics of the transverse (TO) vibrations; the γ_P value describes the plasmon mode damping coefficient in the limit of the zero frequencies and ε_∞ constant denotes the contribution of the excitations at high frequencies relative to the spectral interval of interest. The second term in Eq. (1) represents Ni-impurity local modes, to be

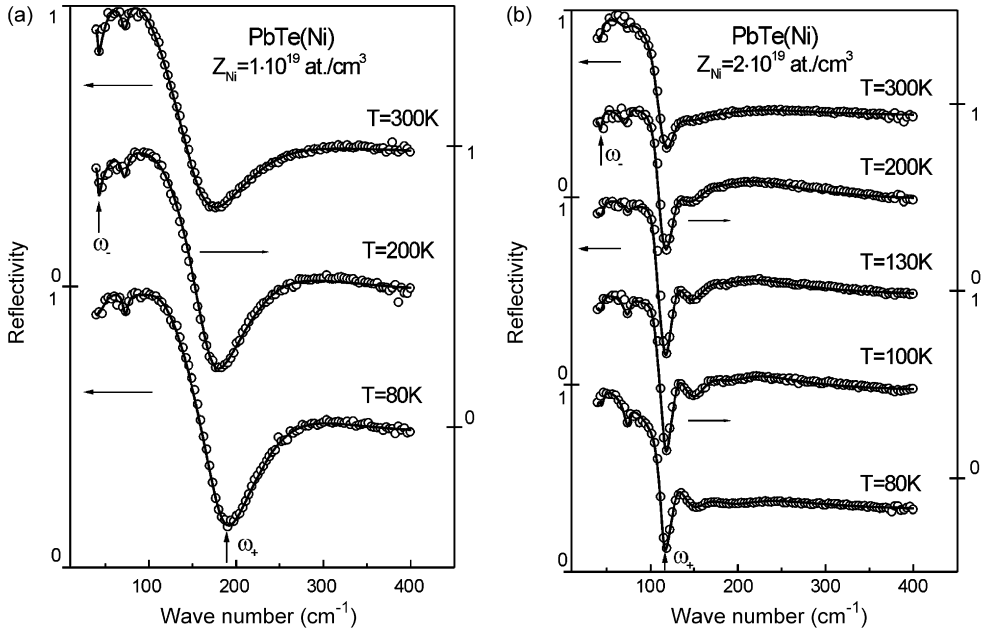

Fig. 1. Far-infrared reflection spectra of PbTe(Ni) single crystal. Experimental spectra are presented by circles. The solid lines are calculated spectra obtained by a fitting procedure based on the model given by Eq. (1). (a) $Z_{Ni} = 1 \times 10^{19}$ at./cm^3, $p = 1$; (b) $Z_{Ni} = 2 \times 10^{19}$ at./cm^3, $p = 3$.

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