Journal of Zhejiang University SCIENCE A
ISSN 1673-565X (Print); ISSN 1862-1775 (Online)
www.zju.edu.cn/jzus; www.springerlink.com
E-mail: jzus@zju.edu.cn

Hole transport and phonon scattering in epitaxial PbSe films*

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Received June 25, 2007; revision accepted Sept. 30, 2007; published online Dec. 14, 2007

Abstract: The combined characterizations of mobility and phonon scattering spectra allow us to probe hole transport process in epitaxial PbSe crystalline films grown by molecular beam epitaxy (MBE). The measurements of Hall effect show p-type conductivity of PbSe epitaxial films. At 295 K, the PbSe samples display hole concentrations of $(5\sim8)\times10^{17}$ cm^{-3} with mobilities of about 300 cm^2/(V·s), and at 77 K the hole mobility is as high as 3×10^3 cm^2/(V·s). Five scattering mechanisms limiting hole mobilities are theoretically analyzed. The calculations and Raman scattering measurements show that, in the temperatures between 200 and 295 K, the scattering of polar optical phonon modes dominates the impact on the observed hole mobility in the epitaxial PbSe films. Raman spectra characterization observed strong optical phonon scatterings at high temperature in the PbSe epitaxial films, which is consistent with the result of the measured hole mobility.

Key words: Electrical properties of PbSe, Mobility, Scattering mechanism
doi:10.1631/jzus.A071350 **Document code:** A **CLC number:** TN3

INTRODUCTION

Recently, much work has been devoted to the transport of IV-VI materials, which is motivated by the potential applications in quantum devices (Romero and Drndic, 2005; Matsushita *et al*., 2005; Shi *et al*., 2004). Romero and Drndic (2005) reported rich and tunable transport phenomena in closed-packed arrays of PbSe nanocrystals. Matsushita *et al*.(2005) gave the evidence for charge Kondo effect by transport measurements of Tl-doped PbTe single crystals. In spite of this, the scattering of charge carriers with phonon and impurity in the transport processes, which contributes to the mobility of the IV-VI semiconductors, has not been well studied. In early published papers, discrepancies exist for the explanation of mobility behavior (Allgaier and Scanlon, 1958; Zogg and Huppi, 1985), and there was no theoretical justification for mobility behavior in IV-VI materials because of insufficiency of phonon scattering data, which was difficult to obtain from conventional Raman spectra due to high symmetry of crystal structure (Cao *et al*., 2006). Since the scattering mechanisms in epitaxial PbSe films have not been well understood, it is necessary to study transport properties of holes in light of information concerning the LO phonon and deformation potentials for optical and acoustic mode scattering that are available.

The present work aims to study the scattering mechanism of the holes in epitaxial PbSe crystalline films. The PbSe films were grown on freshly cleaved BaF_2(111) substrates in our molecular beam epitaxy (MBE) system. Hall effect and Raman scattering measurements were carried out. To explain the transport properties, five scattering mechanisms are considered theoretically with the aim to determine which of them plays the major role in limiting the intrinsic mobility. For the p-type PbSe epitaxial films, it is found that the scattering of polar optical modes dominates the impact on the observed hole mobilities in PbSe epitaxial films. The Raman spectra charac-

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* Project (No. 10434090) supported by the National Natural Science Foundation of China

terizations confirm the strong scattering by the polar optical mode.

Experiments

The PbSe films were grown on freshly cleaved BaF_2(111) substrates in our MBE system which was described in detail in (Si *et al*., 2005). As an essential prerequisite for the study of the electronic properties, a highly insulating substrate must be used for growing PbSe films. BaF_2(111) has proven to be an appropriate substrate material, which is available in high crystalline quality and no significant degradation of epitaxial layers occurs during thermal cycling because of the well matched thermal expansion coefficients (Springholz *et al*., 1996; Xu *et al*., 2007). Excessive strains that could be built in the PbSe epitaxial layers on cooling, which should changes their electrical properties, are avoided. The strain due to the lattice constant mismatch between PbSe and BaF_2 can be neglected because the film thickness is about 1 μm, therefore complete plastic relaxation of strain can be assumed. The samples listed in Table 1 were grown at the same substrate temperature of 450 °C with variation of film thickness. The film thickness was measured by a TENCOR α-step profiler. The resistivities ρ and Hall coefficients R_H were measured with Van der Pauw geometry using indium as contacts. The samples were directly immersed in liquid nitrogen for the 77 K measurement. Variation of temperature was realized by heating the specimen holder. The mobilities and carrier concentrations given here are derived from $\mu=R_H/\rho$ and $R_H=1/n_e$. Raman scattering measurement was performed to observe phonon scattering in PbSe films. For the measurements of the Raman spectra, an Ar^+ laser (λ=514.5 nm) with a cw output of about 10 mW was used. Laser beam was focused using an Olympus lens and the light spot size was about 1 μm in diameter. A Jobin Yvon LabRAM HR 800 monochromotor and an Ander DU420 classic CCD detector were used. The measured spectral range is from 50 to 500 cm^{-1} with the resolution higher than 3 cm^{-1}. A notch filter was used to eliminate the Rayleigh scattered light, which has low wavenumber cutoff to ~50 cm^{-1}. The measurements of Raman spectra were carried out in a backscattering geometry at temperatures that varied from 77 K to 293 K.

Table 1 The hole concentrations and mobilities of PbSe films at room temperature

Sample No.	Thickness (μm)	Carrier type	Carrier concentrations (300 K) ($\times10^{18}$ cm^{-3})	Hall mobility (300 K) ($cm^2/(V\cdot s)$)
A	0.80	p	0.76	297
B	0.90	p	0.78	278
C	0.94	p	0.76	292
D	1.00	p	0.72	233
E	1.30	p	0.67	316
F	1.38	p	0.71	291
G	1.92	p	0.54	289

Results and discussion

As it is well known that narrow band gap semiconductors usually display relatively high carrier concentration, and a typical hole concentration of $(5\sim8)\times10^{17}$ cm^{-3} for non-intentionally doped PbSe was reproducibly obtained as given in Table 1. The observed p-type conduction in the PbSe is due to the fact that to obtain high crystalline quality the epitaxy of PbSe is usually under VI group which is rich ambient and Se interstitial atoms acting as acceptors in PbSe. Compared to the hole concentration, the concentration of the minority carriers (intrinsic electrons) in PbSe at room temperature is lower than ~10^{15} cm^{-3}. Therefore, the contribution of electrons to conduction process could be negligible.

The measured temperature dependence of Hall mobility for the epitaxial PbSe films is presented in Fig.1a. The hole mobility is found to increase with the decrease of measurement temperature and at 77 K hole mobility is as high as 3×10^3 $cm^2/(V\cdot s)$. To understand the measured intrinsic mobility, the following five scattering mechanisms were studied that include impurity scattering, acoustic mode scattering, piezoelectric activity of the acoustic modes scattering, non-polar and polar optical modes scattering. The measured mobility can be expressed as follows:

$$\frac{1}{\mu}=\frac{1}{\mu_{im}}+\frac{1}{\mu_{ac}}+\frac{1}{\mu_{piezo}}+\frac{1}{\mu_{non\text{-}op}}+\frac{1}{\mu_{op}}, \qquad (1)$$

where μ_{im} is mobility limited by impurity scattering and μ_{ac}, μ_{piezo}, $\mu_{non\text{-}op}$, μ_{op} is the mobility limited by acoustic mode scattering, piezoelectric activity of the acoustic modes scattering, non-polar optical modes

scattering, polar optical modes scattering, respectively. Within the five different scattering mechanisms, it is required to determine which ones dominate the contribution to the observed mobility.

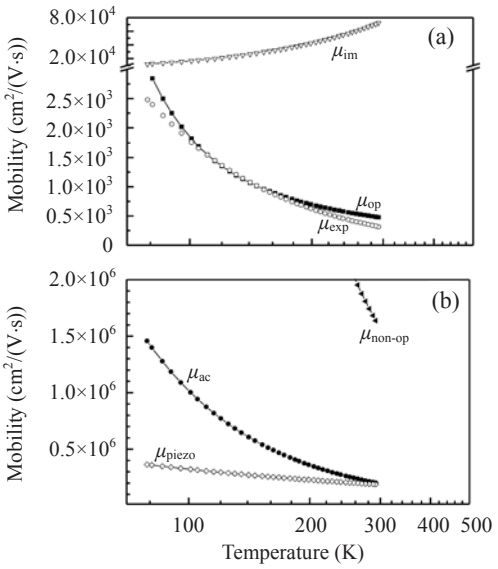

Fig.1 The mobilities of the epitaxial PbSe films versus temperature. (a) The μ_{exp}, μ_{op} and μ_{im} represent the experimental data for PbSe mobility, the calculated polaron mobility, and the calculated ionized impurity scattering mobility; (b) The filled triangle curve $\mu_{non\text{-}op}$, filled circle curve μ_{ac} and open diamond curve μ_{piezo} are the theoretical mobilities calculated from Eqs.(5), (3) and (4), respectively

The impact of impurity scattering on mobility is the first factor to be considered. The expression of mobility for the impurity scattering derived by Brooks and Herring (Chattopadhyay and Queisser, 1981) using the Born approximation applies when the carrier concentrations $n<10^{18}$ cm^{-3} in PbSe films. In this case the mobility formula given by Brooks and Herring is expressed as:

$$\mu_{\text{im}}=\frac{128\sqrt{2}\pi^{1/2}\varepsilon^2(kT)^{3/2}}{m^{*1/2}NZ^2e^3}\left(\ln\frac{24m^*\varepsilon(kT)^2}{ne^2\hbar^2}\right)^{-1}, \quad (2)$$

where $m^*=(m_\perp{}^2\times m_{//})^{1/2}\approx 0.042m_0$ is the effective mass of holes, ε is static dielectric constant, N is the number of impurity atoms, n is the carrier concentration and Z is the charge of the impurity atom. In the non-intentionally doped PbSe, we could take the value of 7×10^{17} cm^{-3} as the number of impurity atom N that is close to the carrier concentration and the values of other parameters used in Eqs.(2)~(7) are given in Table 2.

Table 2 Parameters of the PbSe

Parameters	Value	Parameters	Value
c_{11}	1.3×10^{11} [a]	$\hbar\omega_l$ (eV)	0.0164 [c]
U_{ac} (eV)	10 [a]	U_{op} (eV)	5 [a]
$m^v_\perp$	0.034 [b]	ε_∞	21
$m^v_{//}$	0.068 [b]	ε	218

[a]: Wu *et al.*, 2002; [b]: Martinez and Schluter, 1975; [c]: Yang *et al.*, 1999

The calculated mobilities are presented as μ_{im} curve in Fig.1a. μ_{im} has the values of 1.20×10^4 and 7.23×10^4 cm^2/(V·s) at 77 K and 295 K, respectively. These values exceed the measured ones by 2 orders. It suggests that the impurity scattering does not play the key role in limiting mobility at about 77 K. This result is consistent with the mobilities given by other authors (Allgaier and Scanlon, 1958; Hohnke and Kaiser, 1974). It was shown that the ionized impurity scattering limited the mobility at lower temperatures (T<20 K).

Since the PbSe is partially ionic, the previous studies had suggested that the observed temperature dependence mobility is attributed to the combination of scattering by acoustic and by optical modes (Allgaier and Scanlon, 1958). But no quantitative analysis of the mobility of acoustic mode scattering had been made because of the lack of the details of the band structure of PbSe in early times. They were simply mentioned that combination of both scattering of optical and acoustic phonon affected carrier transport, without giving experimental evidence and without giving temperature range (Tetyorkin *et al.*, 1996; Dashevsky *et al.*, 2004). Here, we propose a theoretical approximation to the mobility of acoustic phonon scattering in terms of presently available PbSe parameters. The mobility of the acoustic mode scattering in a parabolic band where non-degenerate statistics apply is given by (Bardeen and Shockley, 1950):

$$\mu_{\text{ac}}=\frac{2\times(2\pi)^{1/2}\times e\times h^4\times c_{11}}{3\times(m^*)^{5/2}\times(kT)^{3/2}\times U_{\text{ac}}^2}=(m_0/m^*)^{5/2}\times\frac{3.2\times10^{-5}\times c_{11}}{U_{\text{ac}}^2}\times T^{-3/2}, \quad (3)$$

where c_{11} is average longitudinal elastic coefficient,

U_{ac} is the deformation potential of the holes and the value is listed in Table 2. Using the deformation potential of 10 eV, we obtained a value of 1.46×10^6 cm^2/(V·s) for 77 K mobility and 2.03×10^5 cm^2/(V·s) for room temperature mobility. The acoustic-mode scattering limiting mobility estimated from Eq.(3) is shown in Fig.1b. The mobilities determined by this mechanism are 3 orders higher than the measured ones at the temperature range of 77~295 K. It suggests that the contribution from the acoustic phonon scattering should not play a dominant role in PbSe film at the temperature range of 77~295 K.

The piezoelectric scattering occurs in crystals because of polarization associated with the acoustic phonons. The mobility of carriers in parabolic band is given by (Aven and Segall, 1963):

$$\mu_{\text{piezo}} = (m_0/m^*)^{3/2}\times\frac{1.05\times c_{11}\times\varepsilon^2}{e_{14}^2}\times T^{-1/2}, \qquad (4)$$

where e_{14} is piezoelectric coefficient. However, to our knowledge there is no data available on the piezoelectric coefficient for PbSe materials. Here, we could make a rough estimation of the upper limit of piezoelectric coefficient for PbSe using the formula $e_{14}=q^*/d^2$ (d is lattice constant, q^* is the effective charges) (Berlincourt and Jaffr, 1963). Assuming a significantly large value 5 C/m^2 of e_{14}, we estimated room temperature mobility to be 1.89×10^5 cm^2/(V·s) and the mobilities at different temperatures are displayed in Fig.1b. The mobility values are also 2 orders higher than the measured ones, and it can also be neglected as well.

The expression for the hole mobility ($\mu_{\text{non-op}}$) in degenerate p-like bands scattered by the non-polar interaction with the optical phonons can be obtained from the study of Dale Marirus in Ge (Brown and Bray, 1962). It can be shown that

$$\mu_{\text{non-op}} = \mu_{\text{ac}}\int_0^\infty \left\{2x\mathrm{e}^{-x}(\mathrm{e}^{\theta/T}-1)\Big/[(U_{\text{op}}^2/U_{\text{ac}}^2)(\theta/T)\times[(1+\theta/(Tx))^2+\mathrm{e}^{\theta/T}(1-\theta/(Tx))^2]]\right\}\mathrm{d}x, \qquad (5)$$

where U_{op} and U_{ac} are the deformation potentials for optical and acoustic mode scattering respectively. θ is the Debye temperature and $k\theta=\hbar\omega_l$ is the longitudinal optical phonon energy. The values are listed in Table 2. We estimated $\mu_{\text{non-op}}$ to be 1.63×10^6 cm^2/(V·s) at room temperature. The data are plotted in Fig.1b. This mechanism appears to be the least important one in the 5 scattering mechanisms and should not dominate the scattering.

The last scattering mechanism to be considered is the polar optical-mode scattering. The strength of the carrier-phonon scattering is represented by the polar coupling constant α,

$$\alpha = \frac{e^2}{\hbar}\times\sqrt{\frac{m_0}{2\hbar\omega_l}}\times\frac{\varepsilon-\varepsilon_0}{\varepsilon\varepsilon_0}\times\sqrt{\frac{m^*}{m_0}}. \qquad (6)$$

Using the relevant parameters that are listed in Table 2, α is 2.4. In the intermediate coupling region of $1<\alpha<6$, Lee *et al.*(1953) gave the result for the mobility which is valid in the low temperature $T<\theta$,

$$\mu_{\text{op}}(L-P) = \frac{e}{2m^*\omega_l\alpha}\times(m^*/m_{\text{p}}^*)^3\times f(\alpha)\times(\mathrm{e}^{\theta/T}-1), \qquad (7)$$

where $m_{\text{p}}^*=(1+\alpha/6)m^*$ and $f(\alpha)\approx1$ when α takes the value of 2.4. The values obtained from Eq.(7) are shown as the curve μ_{op} in Fig.1a. The result is close to the measured mobilities except for a small discrepancy in $T>200$ K. From the fact that the calculations provide an approximate fitting with the measured values, it appears that the polar optical mode scattering clearly dominates the intrinsic scattering in epitaxial PbSe films.

The small discrepancy at $T>200$ K between the theoretical and the experimental results may be due to non-applicability of Eq.(7) at high temperatures ($T>200$ K). Actually, Eq.(7) only describes the mobility scattered by unique optical phonon mode. However, Raman scattering experiments have usually observed more than one optical phonon scatterings and the intensity of various optical modes increases as measurement temperature rises. To accurately calculate the mobility, the effect of all those optical phonon modes should be included in the theoretical calculation of the scattering at $T>200$ K.

The multiple phonon scattering process is confirmed by our temperature-dependent Raman scattering measurement as shown in Fig.2. The Raman

spectrum contains prominent bands that are at 74 cm^{-1}, 90 cm^{-1}, 123 cm^{-1}, 143 cm^{-1} at all the measured temperatures. The peaks at 90 cm^{-1} and 143 cm^{-1} that are attributed to the modes 2TO(Γ) and LO(Γ) respectively were previously observed by other authors for PbSe films (Chen and Shen, 2006; Ovsyannikov *et al.*, 2004). The peaks at 74 cm^{-1} and 123 cm^{-1} are assigned to the 2TA(X) and the TO(X)+LA(X) or LO(X)+TA(X) respectively, which has been predicted by theoretical calculations (Upadhyaya *et al.*, 2002). The additional mode at 166 cm^{-1} observed at 293 K is assigned to 2LO(X) although several wavenumbers shift from the theoretical value (Ovsyannikov *et al.*, 2004). Fig.2 shows that the relative intensity of these observed optical phonon modes changes significantly as temperature lowers down and LO phonon mode highlights (263 K and 233 K). However, at 203 K the intensity of all the phonon modes is very weak. This Raman scattering observation indicates that at high temperature (T>200 K) the polar optical phonon modes other than LO phonons could be enhanced, which also has effect on the measured hole mobility of PbSe films. Thus, the measured mobility in the temperature range from 200~295 K is lower than the calculated values that only use the single phonon frequency to represent the optical modes of the lattice.

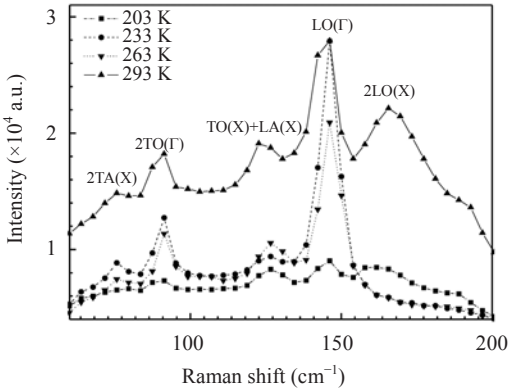

Fig.2 Raman spectra of the epitaxial PbSe films at different temperature

CONCLUSION

From the comparison of the mobility data measured in non-intentionally doped PbSe epitaxial films with the theoretical calculations of five scattering mechanisms, we conclude that polar optical phonon scattering clearly dominates the intrinsic scattering in 200 K~295 K. The scattering from the other mechanisms including impurity scattering, acoustic mode scattering, piezoelectric activity of the acoustic modes scattering, and non-polar optical modes scattering is negligible. We found that the measured mobility is lower than the calculated one and that the discrepancy increases with temperature. The discrepancy between the measured and the calculated mobility that results in the region of 200~295 K could be attributed to the effect of the other observed polar optical phonon mode scattering (except LO phonon) being not included in the scattering terms at high temperature. The observation of Raman spectra scattering intensity from 203 K to 293 K is consistent with the mobility variation.

ACKNOWLEDGEMENTS

The authors would like to thank Jin Qing Shen at Zhejiang University for assistance in the electrical measurements and Jin Chen at Shanghai Jiao Tong University for assistance in Raman scattering measurements.

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