Behavior of Raman modes in InPBi alloys under hydrostatic pressure

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Behavior of Raman modes in InPBi alloys under hydrostatic pressure
AIP Advances, 9(3)
http://dx.doi.org/10.1063/1.5085132

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Cite as: AIP Advances 9, 035120 (2019); https://doi.org/10.1063/1.5085132
Submitted: 09 December 2018. Accepted: 01 March 2019. Published Online: 13 March 2019

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Submitted: 9 December 2018 • Accepted: 1 March 2019 • Published Online: 13 March 2019

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ABSTRACT
Raman spectra of InPBi alloys with bismuth amount 0.3%-2.0% were measured under hydrostatic pressure in diamond anvil cell up to ∼4 GPa at room temperature. Two bismuth related Raman modes were identified and their evolutions under pressure were studied. The linear pressure coefficients of these two modes are determined to be 1.292 and 2.169 cm⁻¹/GPa, respectively. The different behaviors of these two modes under pressure suggest that they may have distinct origins. InP related Raman modes were also investigated including two InP related modes caused by Bi doping.

INTRODUCTION
As a typical III-V narrow band gap semiconductor material, InP has various applications in infrared optoelectric devices. By doping with Bi, InPBi alloys could be formed and it was predicted to be one of the most stable candidates in fabricating infrared optoelectric devices.1 Recently, high quality InPBi alloys were successfully synthesized by gas source molecular beam epitaxy (MBE) method at low temperature.4 The high crystal quality of InP1-xBix has been confirmed by X-ray diffraction and Rutherford Backscattering Spectrometry (RBS) measurements.2,3 It is very important to build up a full understanding of the physical properties of this kind of material in order to realize all its potential applications. Since optical means are powerful and nondestructive ways for investigating material properties, photoluminescence and Raman spectra are used to study the emission bands and vibration modes in InP1-xBix, respectively.2,5 It was found out that incorporation of Bi inside InP substrate would reduce its band gap which made it more suitable for applications in near infrared region.2,5 At the meantime, Bi doping also caused additional Raman vibration modes compared with InP layer grown at the same temperature on InP substrate.4,6 Two new bismuth related modes were identified at ∼150 and ∼170 cm⁻¹, and were attributed to InBi vibration modes.4,6 Furthermore, compared with InP layer, two more InP related Raman modes at ∼312 and ∼337 cm⁻¹ were also observed,4,6 possibly caused by lattice distortion in InP due to Bi doping. Although these additional modes were discussed in recent reports,4,6 here we would like to provide further evidence for identifying these modes from Raman spectra under hydrostatic pressure.
Evolution of optical properties, especially photoluminescence (PL) and Raman spectra, of semiconductors and nanostructures under pressure provides important information for investigating their band structure, lattice vibration nature, etc. The linear pressure coefficients of LO and TO phonon modes from InP were derived as 5.6 and 6.3 cm⁻¹/GPa, respectively.

In this letter, we report the first experimental Raman study of InPBi alloys under hydrostatic pressure (up to ~4 GPa). Both Bi and InP Raman modes are analyzed against pressure. Features including peak position, full-width at half maximum (FWHM), integrated peak intensity are investigated.

**EXPERIMENTAL**

InPBi layer with thickness ~420 µm was grown on InP substrate at ~320 °C by V90 gas source molecular beam epitaxy (MBE). Elementary Bi source was used and its flux was controlled by adjusting cell temperature. Bi incorporation in InP layers was estimated from RBS measurements. More details about the growth process can be found in Ref. 2. In this work, InPBi alloys with Bi concentrations of 0.3%, 1.0%, 2.0% were studied (named as InPBi_1, InPBi_2, and InPBi_3, respectively) with comparison from a InP layer (named as InP LT) grown under similar conditions. Room temperature Raman spectra were taken under the back-scattering geometry by a confocal micro-Raman system (LabRAM HR Evolution system) with 514.5 nm excitation from a Ar⁺ laser. For measurements under pressure, the sample was thinned into tens of micrometers mechanically from the substrate, and then loaded into the diamond anvil cell (DAC). Condensed argon was introduced as the pressure-transmitting medium in the DAC. PL peaks from Ruby powders were used for calibrating the pressure value in the DAC.

**RESULTS AND DISCUSSION**

Figure 1 shows the Raman spectra of these four samples in ambient environment at room temperature. Two InP vibration peaks at 306.46 and 343.20 cm⁻¹ are observed from undoped InP layer, which are attributed to InP TO(Γ) and LO(Γ), respectively (denoted as InP TO-1 and InP LO-2). All peak features, such as peak position, FWHM and integrated peak intensity, are obtained by fitting the Raman spectrum with Lorentz functions. The low energy shoulder of the InP TO-1 peak was attributed to the lower branch of the coupled plasmon-LO mode. These two Raman peaks are also observed from three InPBi samples. Additionally, two Bi related Raman peaks are identified at ~150 and ~170 cm⁻¹ (denoted as Bi-1 and Bi-2, respectively), together with two more InP related Raman modes at ~312 and ~337 cm⁻¹ (denoted as InP TO-2 and InP LO-1, respectively) in InPBi samples. The peak positions of these two newly emerged InP related vibration modes in InPBi samples are very close to 313 cm⁻¹ (TO(L)) and 336 cm⁻¹ (LO(X)) modes from InP. This is probably caused by the lattice distortion in InP due to Bi doping. The lattice distortion in InP will lead to an effective polarizability derivative for the TO(L) and LO(X) modes, which results in the previously inactive modes being detected. This effect is reversed to a certain degree under external hydrostatic pressure which will be discussed later.

To develop a deeper understanding of these observed vibration modes, Raman spectra of InPBi alloys under hydrostatic pressure were measured. Generally, Raman signal from samples in DAC are much weaker than those in ambient environment due to the scattering in pressure transmission medium (condensed argon).
argon in our experiments) and the diamond window and also the
decrease in collection efficiency for using a long focus range objec-
tive lens. Signals from InPBi_1 and InPBi_2 were obtained suc-
cessfully in DAC, although the two Bi related modes were hard to
distinguish from InPBi_1 sample. Here the Raman spectra from
InPBi_2 in DAC under various pressures are depicted in Fig. 2.
We should mention that after careful analysis, the InP related
Raman peaks demonstrate the same behavior under pressure based
on observations from InPBi_1 and InPBi_2. Pressure value inside
the DAC was calibrated by the peak position of Ruby PL R1 line based on the empirical formula developed from Murnaghan equation,\(^{11-13}\)

\[
P = \frac{1904}{B} \left( \frac{\lambda}{\lambda_0} \right)^B - 1
\]

where \(P\) is the pressure in GPa, \(\lambda_0\) the peak position of Ruby
R1 line at ambient pressure, \(\lambda\) the peak position of Ruby R1 line
at certain pressure (\(\lambda_0\) and \(\lambda\) values were obtained by fitting the
Ruby R1 emissions with Lorentz function), and \(B\) is a constant
of 7.665.

Figure 2 shows the Raman spectra of InPBi_2 under vari-
ous hydrostatic pressures. The same data process was performed.

As expected, all Raman modes demonstrate a blue-shift as environ-
mental pressure increases. Additionally, the ratio of peak intensity
of InP LO-1 to that of InP LO-2 decreases when the pressure goes
up. As mentioned earlier, the InP LO-1 Raman mode is probably
induced by lattice distortion of InP caused by Bi doping. The appli-
cation of an external hydrostatic pressure can effectively reduce
the distortion of the InP lattice, which therefore weakens the polarizabil-
ity derivatives and suppresses the doping-induced mode, leading to
the intensity drop of the InP LO-1 peak, while the InP LO-2 Raman
mode is less affected. Based on the fitting results, the Raman peak
positions versus pressures of different modes are summarized in
Fig. 3. A linear fitting was performed for the data in Fig. 3 for each
Raman vibration mode and the linear pressure coefficients are listed
in Table I.

For the two InP related Raman vibration modes observed in
InP layer and also all other samples, InP TO-1 and InP LO-2, their
linear pressure coefficients are found to be 4.404 and 5.377 cm\(^{-1}\)
GPa\(^{-1}\), respectively. The two coefficients are close to those reported
experimental values,\(^{8,9}\) and also comparable to theoretical calcula-
tions.\(^{10}\) It should be noted that a parabolic function was used for
obtaining the pressure coefficients, although the second order coef-
cient was very small.\(^{9}\) Interestingly, two newly emerged InP related

\[
\begin{array}{c}
\text{InPBi}_2 \text{ Raman Peak Position vs. Pressure} \\
\begin{array}{c}
\text{Bi-1} \\
\text{Bi-2} \\
\text{InP TO-1} \\
\text{InP TO-2} \\
\text{InP LO-1} \\
\text{InP LO-2}
\end{array}
\end{array}
\]

\[
\begin{array}{c}
\text{FIG. 3. Peak positions of various Raman vibration modes} \\
\text{in InPBi measured from InPBi}_2 \text{ in DAC at different hydro-
static pressures. Red lines are the linear fitting results,} \\
\text{respectively.}
\end{array}
\]
CONCLUSION

To summarize, based on Raman spectrum measurements of InPBi alloys under hydrostatic pressure, linear pressure coefficients of various Raman modes are obtained by fitting experimental results. The two Bi related Raman modes (~150 and ~170 cm⁻¹) are identified and tentatively assigned to α-Bi₂O₃ A₃g Raman mode and overtone of Bi cluster Raman mode, respectively. Investigation of these Raman modes in doped InP is certainly helpful for understanding the nature of impurity incorporations in it and also provides useful information for realizing its potential applications.

ACKNOWLEDGMENTS

CCZ acknowledges financial support from an open project from State Key Laboratory of Functional Materials for Informatics, Shanghai Institute of Microsystem and Information Technology, and partial support from Natural Science Foundation of China (No.: 11504299). JQN acknowledges the financial support from Natural Science Foundation of China (11874390) and Hundred Talents Program of Chinese Academy of Sciences.

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