Miniband effect on optical vibrations in short-period In\textsubscript{x}Ga\textsubscript{1-x}As/InP superlattices

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The formation of the miniband electron energy structure and its effect on optical vibrational modes were explored in doped In\textsubscript{x}Ga\textsubscript{1-x}As/InP superlattices with different periods. The analysis of the high resolution x-ray diffraction, Raman and magnetotransport data allowed us to conclude that in spite of the defect structure of the layers constituting the superlattices, their superperiodicity was well defined. The blueshift of the coupled plasmon-LO phonon modes was observed with decreasing superlattice period consistent with the development of the minibands. The coherence lengths of the coupled modes were found to be considerably longer than those of the optical phonons. This provided a quantitative proof of the conditions for breakdown of the Raman selection rules. Due to the defect structure of the layers no Raman selection rules were observed for the longitudinal optical phonons in long-period superlattices. In contrast, the selection rules of the coupled plasmon-longitudinal optical phonon vibrations observed in short-period superlattices were demonstrated to occur due to the increase of the coherence lengths of the coupled modes with respect to the coherence lengths of the optical phonons.

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I. INTRODUCTION

In\textsubscript{x}Ga\textsubscript{1-x}As/InP heterostructures present an important class of semiconductor materials with applications in infrared optoelectronics. Moreover, a large g factor found in these materials\textsuperscript{1} makes them promising in the field of spintronics and quantum information processing. A manipulation of the electronic properties (including g-factor engineering) important in device applications is possible in superlattices (SLs)—structures consisting of periodic sequences of the layers made of two different semiconductor materials. In such structures the dynamic electron properties can be tuned in the direction perpendicular to the layers by a variation of their thicknesses or compositions. However, to the best of our knowledge evidence of the formation of the superlattice band structure in In\textsubscript{x}Ga\textsubscript{1-x}As/InP SLs has been obtained only recently by high-field magnetotransport measurements.\textsuperscript{2} The photoluminescence data presented in Ref. 2 also indicated the existence of the minibands which result from the SL periodicity.\textsuperscript{3} In Ref. 2 no effects of the miniband dispersion were detected in long-period SLs with the narrow minibands (with a width of a few meV).

In this paper we present an extensive investigation of doped In\textsubscript{x}Ga\textsubscript{1-x}As/InP SLs with different periods by x-ray diffraction, Raman scattering and high-field magnetoresistance. The miniband widths of these SLs varied in the interval from 250 meV to the flat isolated electron levels. Therefore, the effects of the miniband dispersion were possible to distinguish. We explored the Raman scattering by the coupled plasmon-LO phonon modes propagating perpendicular to the layers of the In\textsubscript{x}Ga\textsubscript{1-x}As/InP SLs. Such modes emerge due to the coupling between the longitudinal optical (LO) lattice vibrations and the collective electron excitations (plasmons) resulting in significant blueshifts of the frequencies of the LO modes. We found no evidence of such modes in the long-period SLs. The manifestation of the coupled modes in the short-period SLs was assigned to the formation of the miniband energy structure caused by the SL periodicity. It should be mentioned that the effects of the plasmon-LO phonon coupling on the Raman scattering of doped In\textsubscript{x}Ga\textsubscript{1-x}As/InP single quantum wells has been discussed in Refs. 4 and 5. We also found an additional indication of the miniband formation by high-magnetic field magnetoresistance measurements. Moreover, a considerable effect of the superperiodicity on the Raman selection rules of the coupled modes was observed: the longitudinal lattice vibrations detected in the long-period superlattices did not reveal any selection rules because of the defect structure of the bulk InGaAs and InP layers, while the coupled plasmon-LO phonon modes showed evidence in the superlattices of the expected Raman selection rules. Our data reveal that these selection rules came out due to the increase of the spatial coherence length of the coupled vibrations. These results provide additional evidence for the modification of the Raman selection rules induced by atomic-scale roughness in semiconductor SLs.\textsuperscript{6}

II. EXPERIMENTAL

A total number of 30 periods of the lattice-matched (In\textsubscript{0.53}Ga\textsubscript{0.47}As)\textsubscript{m} (InP)\textsubscript{n} SLs with m=6, 7, 10, 15, and 68 ML (except the SL with m=68 which had 20 periods), where n is the thickness of the layers expressed in monolayers (1 ML = 2.9 Å), were grown on semi-insulating (001) InP substrates by molecular beam epitaxy. The barriers of the SLs were
were carried out at transport measurements parallel to the surface of the samples. Ohmic contacts were fabricated by depositing indium. The cooling by liquid nitrogen. The 5145 Å line of an Ar+ laser triple grating spectrometer supplied with a CCD detector for nonresonant excitation.

The SLs were characterized by high-resolution x-ray diffraction (HRXRD) rocking curves of the (004) reflection using the Bede D1 system. Each SL structure was determined by comparing the measured rocking curves to simulated curves to determine the degree of interface roughness and variation in SL periodicity. The simulated curves were obtained by using the Bede RADS Mercury (version 3.88) x-ray rocking curve software based on the Takagi-Taupin equations of dynamical diffraction theory.

The magnetoresistance measurements were performed on samples patterned into an active area of 4 × 4 mm. The Ohmic contacts were fabricated by depositing indium. The transport measurements parallel to the surface of the samples were carried out at T = 4.2 K in the Van der Pauw geometry using a standard low-frequency lock-in technique in a liquid He cryostat with the magnetic fields directed perpendicular to the current. The Raman scattering was collected from the surface of the samples at T = 10 K in the backscattering configuration with an "Instruments S.A. T64000" triple grating spectrometer supplied with a CCD detector cooled by liquid nitrogen. The 5145 Å line of an Ar+ laser was used for nonresonant excitation.

III. RESULTS AND DISCUSSION

The HRXRD rocking curves of the SLs are shown in Fig. 1, which exhibit the usual satellite peaks (order ± 1, ± 2, ...) centered around the average composition peak (order 0). In terms of the consistency in the period of the SLs, the quality is quite good. This is apparent from the small amount of satellite peak broadening that occurs with satellite order. The uniformity of the satellite peak full width at half-maximum (FWHM) within each sample can be seen in Fig. 1, especially in the m = 68 ML sample. A quantitative analysis of the peak broadening of the 68 ML sample shows that the FWHM of the satellite peaks increased by only about 1.5 arcseconds with each consecutive order. Clear broadening with increasing satellite order is a sign of significant variation in the SL period. When compared to data from other studies the variation in the period of our SLs can be estimated at about 0.3%. Excess broadening of each SL peak in comparison to the simulated peak widths (not shown) was consistent with an interface roughness of approximately 2 MLs.

The Raman spectra of the optical vibrational modes propagating along the growth direction measured in z(′x′y′)z and z(x′x′)z parallel-polarized configurations of the backscattering, where x′, y′, z are the crystallographic directions [110], [110], and [001], respectively, are shown for selected SLs in Fig. 2. In such a case, according to the Raman selection rules derived for the deformation-potential electron-phonon interaction (the Fröhlich interaction gives vanishing contribution at the zone center, which is negligible in nonresonant conditions), the longitudinal optical lattice vibrations (LO) are active in the cross-polarized configuration, while no optical modes contribute to the parallel-polarized Raman scattering. For comparison, InAs-like (1), GaAs-like (2), and InP-like (3) "restrahlen" bands determined by experimental room temperature frequencies of the corresponding TO and LO phonons, available from the literature (Refs. 11 and 12) are also shown in Fig. 2 as shaded areas. As can be seen in this figure, we observe the following vibrational modes for the long-period InG GaAlxAs/InP SL with both layers’ thicknesses equal to 68 ML: the allowed LO and the forbidden transverse optical (TO) vibration modes of the InGaAs wells (the InAs-like modes with the indices 1 and the GaAs-like modes with the indices 2, respectively) and the InP barriers (the modes with the indices 3). Moreover, the
The InP-like interface modes found to remain unchanged with the variation of the period. The electrons propagating in the growth direction of a SL are affected by the superperiodicity. The dynamic properties of the miniband structure in the electron energy spectrum of the InP lattice vibrations at a period of the miniband dispersion increases and as a consequence, the blueshift of the corresponding longitudinal modes denoted as \( L_1 \) mode, the GaAs-like coupled \( T_z \) mode, and the InP-like coupled \( T_x \) mode. Their dependence on the plasma frequency is shown in Fig. 4. In all of these modes were found in the Raman spectra of the short-period \( \text{InGaAs/InP SLs} \) except the low-frequency \( L^- \) mode, which is difficult to detect due to its low frequency. The hatched area of Fig. 4 indicates the region of the dielectric function tensor of a SL in the long wavelength approximation according to Ref. 16:

\[
\epsilon_1(\omega) = \frac{d_1 + d_2}{d_1/\epsilon_1(\omega) + d_2/\epsilon_2(\omega)} - \frac{\omega_{\text{LO}}^2}{\omega_{\text{TO}}^2},
\]

with the dielectric function of the InGaAs layers determined as

\[
\epsilon_1(\omega) = \epsilon_{\text{GaAs}} \left( \frac{\omega_{\text{GaAs}}^2 - \omega_{\text{LO}}^2}{\omega_{\text{GaAs}}^2 - \omega_1^2} + \frac{\omega_{\text{LO}}^2 - \omega_2^2}{\omega_{\text{GaAs}}^2 - \omega_2^2} \right)
\]

and the dielectric function of the InP layers determined as

\[
\epsilon_2(\omega) = \epsilon_{\text{InP}} \left( \frac{\omega_1^2 - \omega_{\text{LO}}^2}{\omega_1^2 - \omega_3^2} \right),
\]

where \( d_1 \) and \( d_2 \) are the thicknesses of the corresponding layers, \( \epsilon_{\text{GaAs}} \) and \( \epsilon_{\text{InP}} \) are the dielectric constants of the GaAs and InP, respectively, and \( \omega_{\text{LO}} \) and \( \omega_{\text{TO}} \) are the frequencies of the corresponding longitudinal and transverse phonons.

According to Eqs. (1)–(3) the spectrum of the longitudinal optical vibrations of the doped \( \text{InGaAs/InP SL} \) consists of four modes: the low-frequency plasmonlike coupled \( L^- \) mode, the InAs-like coupled \( L_1 \) mode, the GaAs-like coupled \( L_2 \) mode and the InP-like coupled \( L_3 \) mode. Their dependence on the plasma frequency is shown in Fig. 4. All of these modes were found in the Raman spectra of the short-period \( \text{InGaAs/InP SLs} \) except the low-frequency \( L^- \) mode, which is difficult to detect due to its low frequency. The hatched area of Fig. 4 indicates the region (the mode frequencies and the wave number expansion) where these modes were observed. It follows from the data presented in Fig. 4 that the \( L_1 \) and \( L_2 \) modes reveal no significant shift in the observed region. Moreover, these modes comprise the complex mixed character and they are hindered by the contributions of the TO and IF vibrations. Besides, the energy of the \( L_3 \) mode is affected by the spatial quantization of the GaAs-like phonons in the wells, while the effect of the quantization of the InP-like \( L_3 \) mode is insignificant due to the weak dispersion of the corresponding LO phonons. The...
The coupled modes usually reveal the Raman selection rules of the plasmon-LO phonon modes in the short-period SLs. According to our calculations, the longitudinal lattice vibrations transform to the miniband effect as seen in Figs. 2 and 3. No selection rules are probable due to the defect structure of the short-period SLs. As demonstrated in Ref. 18, in the presence of disorder a finite coherence length of the excitations involved in Raman scattering leads to the asymmetric Raman line shapes, which are distinctly different for the optical phonons and the coupled plasmon-like modes. Then, the shape of the Raman lines reproduces the density of states of the excitations that contribute to Raman scattering. In such a case, the coherence length of the relevant excitations may be obtained. We found the well pronounced asymmetries of the Raman lines of the LO3 phonon of InP in the long-period SL (m=68 ML) and of the L3 coupled mode in the short-period SL (m=6 ML). The asymmetries of the phonon and the coupled lines are different due to their different dispersions [shown in Figs. 6(a) and 6(b)]. The fitting of the Raman intensities calculated as in Ref. 18 to the experimental spectra shown in Figs. 6(c) and 6(d) allowed us to determine the corresponding coherence lengths (Lc): 0.5 nm and 12 nm for the InP longitudinal optical phonon and for the coupled L3 mode, respectively. Obviously, the obtained phonon coherence length is of the order of the crystal unit cell size and, therefore, the phonon Raman lines did not reveal the selection rules. In contrast, the much stronger coherence of the plasmon-LO phonon modes resulted in their symmetrical properties determined by the crystal lattice and, as a consequence, in the corresponding selection rules. The dispersions of the excitations analyzed above, which are indispensable for the calculations of the Raman intensities are shown in Figs. 6(a) and 6(b). The dispersion of the coupled mode was calculated in the random phase approximation, while the dispersion of the InP LO phonon was taken from Ref. 12.

Finally, an additional confirmation of the formation of the miniband energy structure was obtained by the high-field magnetoresistance measurements. The Shubnikov-de Haas oscillations measured with different orientations of the magnetic field (Fig. 7) demonstrate a formation of the quantized Landau levels and show a well pronounced anisotropy of the...
magnetoresistance as expected in SLs due to the different parallel and vertical effective masses. The fitting of the magnetoresistances calculated according to Ref. 19 to the experimental magnetoresistance traces allowed us to obtain the Fermi energies, the vertical effective masses $m_{\parallel}$ and $m_{\perp}$, and the ratios of the parallel to vertical relaxation times $\tau_{\parallel}/\tau_{\perp}$ shown in Figs. 7(a) and 7(b). The calculations were performed with the parallel effective mass of 0.05 $m_0$ corresponding to In$_{0.53}$Ga$_{0.47}$As.15 The difference found between the parallel and vertical effective masses once again reveals the effect of the formation of the superlattice miniband.

IV. CONCLUSION

Doped In$_{x}$Ga$_{1-x}$As/InP SLs with different periods were explored by x-ray diffraction, Raman scattering and high-field magnetoresistance measurements. Clear evidence of the formation of the miniband structure in the electron energy spectrum was observed. No Raman selection rules were detected for the LO phonon modes in the long-period In$_{x}$Ga$_{1-x}$As/InP SLs, while the distinct selection rules were found for the coupled plasmon-LO phonon modes propagating in the growth direction of the short-period SLs. We demonstrated that the selection rules of the coupled modes appear due to the increase of their coherence length. The analysis of the diffraction and Raman data allowed us to conclude that in spite of the defect structure of the layers constituting the SLs, their superperiodicity was well determined. The magnetoresistance measured in the In$_{x}$Ga$_{1-x}$As/InP SLs also demonstrated the anisotropy of the miniband effective electron mass due to the formation of the miniband structure.

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