Spectroscopic investigations of photoinduced changes of the spatial distribution of charge carriers in modulation-doped quantum-well structures

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We investigated the photoinduced changes of the spatial distribution of the free holes in *p*-type modulation-doped GaAs/$Al_xGa_{1-x}As$ heterostructures by measuring the energy of the quantum-well luminescence, which is conditioned by the many-particle effect of band-gap renormalization and the excitonic interactions between the electrons and holes. A conventional chopper technique with millisecond time resolution permitted us to study the dynamics of the hole density variations in the quantum wells. The strength of excitonic effects on the luminescence signals could be estimated by application of a magnetic field oriented parallel to the growth direction of the samples. [S0163-1829(96)08219-7]

I. INTRODUCTION

During the last years the investigations of the electrical and optical properties of modulation-doped quantum-well structures have attracted considerable interest both from the scientific and the technological points of view. These studies were decisively driven by the progresses in the fabrication of the structures leading to crystals with nearly perfect two-dimensional characteristics. The optical properties such as, e.g., the energy of the fundamental luminescence, are mainly determined by the spacings of the corresponding electrical subbands in the quantum wells resulting from the quantization effect perpendicular to the interfaces. For the first time Pinczuk *et al.*[1,2] observed a strong impact of the two-dimensional charge carrier concentration on the energy of the band gap in *n*-type modulation-doped quantum-well structures. Since then much work has been spent on this subject both experimentally and theoretically in order to explain this shrinkage of the band gap due to an increased density of charge carriers in the wells.[3–9] This effect, usually referred to in the literature as band-gap renormalization (BGR), is a consequence of the many-body exchange and correlation corrections due to the presence of the free carriers in the system.

It is known that photons with energies above the barrier band gaps of modulation-doped quantum-well structures cause a transfer of the free charge carriers from the wells into the barriers and reduce very efficiently the two-dimensional carrier gas density in the wells.[8,9] By using Kleinman's equation[3] for the calculation of the BGR as a function of the quantum-well carrier density measurements of the position of the quantum-well luminescence permit us to estimate the size of the carrier density reduction in the wells. On the other hand, the energy of the luminescence is additionally affected by excitonic interactions between the electrons and the holes.[10–14] These excitonic effects, whose strengths depend on the free carrier gas density due to the screening of the Coulomb interaction between the electrons and the holes, are not considered in Kleinman's theoretical expression. Information about the excitonic character of the recombination process is obtained by magnetic-field-dependent luminescence measurements.

In the following we report on our results of the photoluminescence measurements carried out on *p*-type modulation-doped quantum-well structures. In our experimental setup above-band-gap excitation alternated in time with the detection of the luminescence signal. This was realized by a conventional chopper technique with millisecond time resolution (Fig. 1). We used the 514-nm line of an Ar^+ laser (pump beam) to obtain an efficient reduction of the two-dimensional hole gas and a tunable dye laser with photon energies below the barrier band gap (probe beam) to provide the optical excitation for the measurements of the quantum-well photoluminescence. By changing the delay time between the illumination and the detection of the optical signal insights into the dynamics of the variations of the charge carrier distribution can be gained. The excitonic character of the photoluminescence signal was studied by experiments with an applied magnetic field. The field-dependent (diamagnetic) shift revealed a pronounced excitonic contribution to the quantum-well photoluminescence in our *p*-type doped structures.

II. EXPERIMENTAL DETAILS

The presented results of photoluminescence measurements were achieved using molecular-beam epitaxially grown *p*-type modulation-doped GaAs/$Al_xGa_{1-x}As$ multiple quantum-well structures. The sample that will be discussed in detail in the following consists of 10 periods of GaAs wells of widths $L_z=11$ nm, separated by spacers of a thickness of 24.5 nm from 15-nm-wide Be-doped layers ($n_{Be}=3\times10^{17}$ cm^{-3}) in the center of the $Al_{0.32}Ga_{0.68}As$ barriers. From Raman spectroscopic data of a similar sample we derive a hole density of $p=1.7\times10^{11}$ cm^{-2} ($T=2$ K, sample in the dark).[15]

The experiments were performed with the sample immersed in superfluid liquid helium ($T=2$ K) in a bath cry-

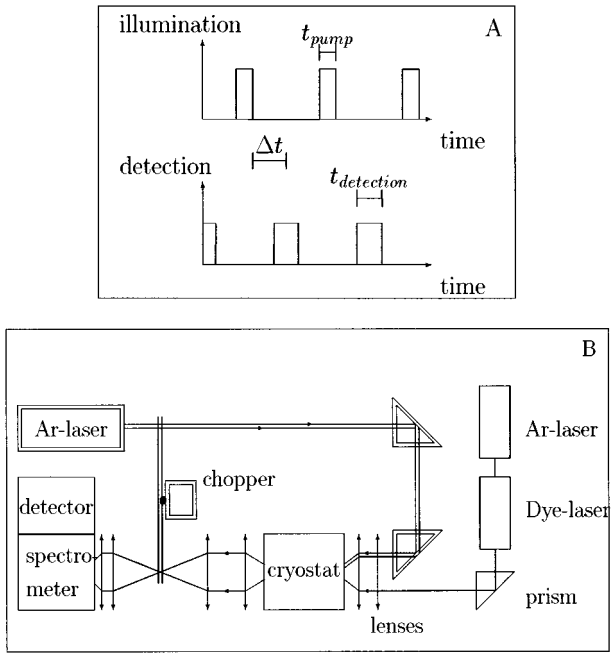

FIG. 1. Schematic illustration of the experimental setup. The interchange between sample illumination by an Ar^+ laser (pump laser period t_{pump}) and detection of the photoluminescence signal (period $t_{detection}$) is realized by a chopper wheel (*A*). The devices needed for the pump laser irradiation are drawn with double lines (*B*). The delay time Δt is defined as the time interval between the end of the Ar^+ pump laser pulse and the middle of the detection period $t_{detection}$.

ostat or in a split coil superconducting magnet system ($B \leqslant 8$ T). The light of an Ar^+ laser pumped dye laser (pyridine) (Fig. 1) with photon energies below the $Al_{0.32}Ga_{0.68}As$ barrier band gap served as the probe beam by providing the optical excitations of the charge carriers in the wells. The optical signals of the electron-hole recombination processes were dispersed with a computer controlled triple spectrograph (DILOR XY) and analyzed by an optical multichannel detector. The use of a Raman spectrometer permits us to perform also inelastic light scattering experiments under conditions realized by our experimental setup. The 514-nm line of a second Ar^+ laser emitting photons with energies far above the barrier band gap was used to vary the two-dimensional hole concentration in the wells. The pump beam and the detection of the signals were separated from each other on the time scale (Fig. 1) by utilizing a conventional chopper technique. Changing the rotation frequency of the chopper allowed a variation of the mean delay time Δt defined as the period between the end of the sample illumination t_{pump} and the middle of the detection time $t_{detection}$. Delay times Δt between approximately 0.3 and 200 ms could be attained by our technique. The detection time $t_{detection}$ should be chosen as short as possible in order to attain a defined relation between the characteristics of the signal and the quoted delay times. In the case of photoluminescence measurements this can be done easily due to the strong intensities of these signals. For the detection of weaker signals (e.g., Raman signals) the time window $t_{detection}$ can be enlarged at the expense of losing gradually the clear definition of the delay time Δt.

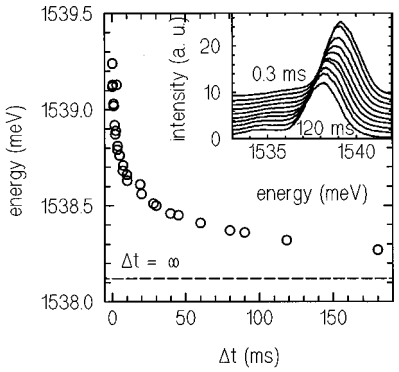

FIG. 2. Energy of the fundamental quantum-well photoluminescence vs delay time Δt. The dashed line represents the energetic position for $\Delta t = \infty$; i.e., no Ar^+ laser is used in this case. The inset displays the luminescence spectra for varying Δt.

Cylindrical lenses used in the experimental setup produced line focuses of the laser lights on the sample keeping the pump laser power densities below 200 mW/cm^2 in order to avoid sample heating. A reduction of the dye laser intensity by positioning a filter into the optical path is demanded by the fact that photons with energies below the barrier band gap also contribute to a hole transfer out of the quantum wells.[16,17] Dye laser power densities below 0.2 mW/cm^2 used here ensure that the effect of the carrier redistribution is mainly caused by the high-energy Ar^+ laser light as can be shown easily.

III. PHOTOINDUCED VARIATION OF THE HOLE DENSITY IN THE QUANTUM WELLS

By changing the frequency of the chopper wheel we detected the signals of the GaAs quantum-well photoluminescence at different delay times Δt. It has been observed that for growing chopper frequency the position of the signal is shifted towards higher energies (inset of Fig. 2). Increasing the delay time from about 0.3 ms to about 120 ms manifests itself in a redshift of about 1 meV in our sample. In Fig. 2 the energetic positions of the main photoluminescence peaks obtained by fitting the line shapes with sums of Gaussian and Lorentzian curves are plotted versus the mean delay time Δt. The horizontal dashed line at an energy of 1538.1 meV represents the signal position in the case without the high-energy Ar^+ laser illumination. With increasing Δt the luminescence signal will asymptotically approach the position of the dashed line. The effect of the additional sample irradiation can be observed for delay times even larger than 120 ms. Furthermore, it is worth mentioning that for high-speed chopper rotations small changes in Δt bring about large changes of the energetic positions while for slow chopper rotations only small shifts of the luminescence signals occur even if the delay time Δt is varied drastically.

The time-dependent effect of the additional pump laser beam becomes obvious from these results. We interpret the observed energy shifts in terms of a variation of the two-dimensional quantum-well hole density. The absorption of photons with energies above the barrier band gap generates electron-hole pairs in the barrier regions.[8,9] As illustrated in Fig. 3(a) the ionized acceptor atoms in the doped barrier

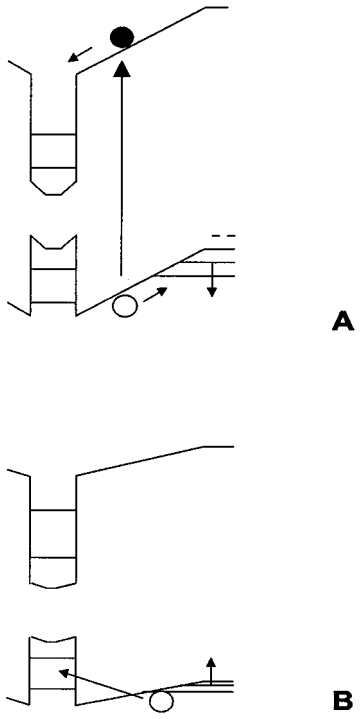

FIG. 3. Schematic presentation of the physical processes involved in our experiments. (a) Photons with energies above the barrier band gap create pairs of electrons and holes in the barriers, which are then separated by the built-in electric field leading to a decrease of the hole densities in the wells and an enrichment of photoinduced holes in the barrier regions. At the same time the shape of the potential approaches the flat band case. (b) The transfer of the barrier holes back into the wells occurs by tunneling and comes along with a new increased bending of the band structure.

areas of p-type modulation-doped quantum-well structures attract the holes whereas at the same time the electrons following the slope of the conduction band drop back into the wells. The recombination of these photoinduced electrons with the hole gas results in a reduction of the two-dimensional hole concentration in the quantum wells. As a consequence of the whole process a hole transfer from the wells into the barriers is achieved, which is observable by measuring the fundamental quantum-well luminescence due to the charge-density-dependent effects. A reduced hole concentration in the wells is equivalent to a reduction of many-body exchange and correlation effects and an increase of the excitonic interaction meaning that the luminescence energy is finally determined by the subband structure and an excitonic shift. Because of the lowered significance of many-body contributions, which reduce the band gap, the signals are blueshifted if the sample is illuminated by the Ar^+ laser in advance. At the same time this energy shift is partly compensated by an increasing exciton binding energy (redshift) when the screening of the Coulomb interaction between the electrons and holes by the two-dimensional hole gas is reduced by the pump laser irradiation. It will be shown below by magnetic-field-dependent measurements that the luminescence signal of our sample bears in fact a finite excitonic character even at a hole density of $p=1.7\times10^{11}$ cm^{-2}.

Redistributing the holes in the manner described above, the quasi Fermi levels in the wells and barriers are shifted

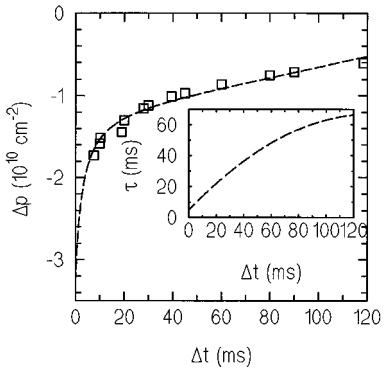

FIG. 4. Calculated reduction of the two-dimensional hole concentration Δp as a function of the delay time Δt. A hole density of $p=1.7\times10^{11}$ cm^{-2} is assumed in the case of the exclusive illumination by the dye laser. For a satisfying description of $\Delta p(\Delta t)$ a time-dependent tunneling time $\tau(\Delta t)$ must be supposed. The inset gives the fit results of $\tau(\Delta t)$.

relative to each other. To re-achieve equilibrium the photoinduced holes in the barriers return into the wells by a tunneling process through the spacer regions[9] [Fig. 3(b)]. The refilling of the wells with the hole gas increases the many-body corrections and causes again a shrinkage of the quantum-well band gap due to the effect of BGR. This dynamical process of balancing the quasi Fermi levels can be observed in our experiments because the relaxation times come up to orders that are accessible with our simple chopper technique.

Neglecting for the moment the effect of screening of excitonic interaction by the hole gas and interpreting the shift of the luminescence signal only as an effect of the BGR, the measurements of the fundamental band-gap luminescence enable us to estimate the momentary hole density in the quantum wells by using Kleinman's[3] exponential equation

$$\Delta E_{\mathrm{BGR}}\ (\mathrm{meV})=2Cp\ \ (\mathrm{cm}^{-2})^{\alpha}, \tag{1}$$

where $\alpha=0.32$ and $C=2.2\times10^{-3}$. Under the assumption of a two-dimensional hole density of $p\approx1.7\times10^{11}$ cm^{-2} in the case of no additional Ar^+ laser irradiation[15] we converted the experimental data of BGR into concentration values of transferred quantum well holes Δp. Figure 4 displays the reduction of the hole concentration in the quantum wells as a function of the delay time Δt. It can be seen from this plot that even for $\Delta t=180$ ms there are still more than 4×10^9 cm^{-2} photoinduced holes kept in the barriers, indicating a strong localization effect of the holes in the doped barrier centers of our sample. In the case of the shortest delay time of $\Delta t=0.3$ ms as many as 18% of the original quantum well holes have been transferred into the $Al_xGa_{1-x}As$ barriers.

In order to get more insight into the back transfer mechanism of the photoinduced holes in the barriers we made an ansatz for the time-dependent hole density variation with an exponential form

$$\Delta p(\Delta t)=\Delta p_0\exp[-\Delta t/\tau(\Delta t)], \tag{2}$$

where the tunneling time τ was assumed to depend on the delay time Δt. A delay-time-independent tunneling time does not fit the experimental data. Fitting a second-order polynomial to $\tau(\Delta t)$, the experimental data agree very well with this

simple formula (Fig. 4). We infer from this fitting that the tunneling time τ indeed depends on the number of holes in the barriers. As can be seen from the inset of Fig. 4, which displays the fitting result for $\tau(\Delta t)$, the tunneling probability decreases with the proceeding back transfer of the holes into the wells. This is consistent with our observations of Fig. 2 that during the first milliseconds after the Ar^+ laser incidence the back transfer of the holes is most striking. We interpret this in correlation with the band bending, which depends on the number of holes in the barriers as indicated in Fig. 3. Immediately after the pumping pulse the valence-band structure comes close to the flat-band case [Fig. 3(b)] leading to rather short tunneling times due to small effective barrier heights and thicknesses. The steady back transfer of the holes reestablishes an increasingly bended valence-band structure [Fig. 3(a)]. As a consequence the tunneling probabilities are decreasing, i.e., the tunneling times are increasing.

Our simple experimental setup for the observation of the dynamics of the charge carrier relaxation process can only be used if the strength of the localization of the photoinduced holes within the barrier regions is strong enough to extend the back transfer into the quantum wells over a sufficiently long period in the range of a few milliseconds. The lifetime of the photoexcited holes in the potential minimum of the doped barrier center regions is mainly determined by the thickness of the spacers and the acceptor concentration in the doped barrier regions. The corresponding parameters of the sample described here exhibit the strongest illumination effect of all the modulation-doped heterostructures we have examined in the frame of this work.

The model given above (Fig. 3) of the charge transfer mechanism is also supported by measurements of the temperature dependence. An increase of the sample temperature strongly reduces the detectable effects of the pumping by the Ar^+ laser. At temperatures above 40 K no blueshift of the photoluminescence signal could be observed even at the smallest achievable delay time $\Delta t = 0.3$ ms. We interpret these findings in terms of an increasing population of the higher subbands in the barrier potential (Fig. 3) at elevated temperatures. This decreases both the effective thickness and height of the barriers, which the holes have to tunnel through to return into the quantum wells. The lifetimes of the photoexcited holes in the potential minimum of the barrier center regions become too short to be detected by our simple experimental setup.

So far we have interpreted the observed shifts of the photoluminescence signal in terms of the BGR only. This approach does not take into account the finite contribution of excitonic effects to the photoluminescence signal and ignores the variation of the exciton binding energy when the hole concentration in the wells is changed. The application of a magnetic field parallel to the growth direction was used to gain insight into the excitonic character of the recombination process in our sample. A diamagnetic shift of the electron-hole recombination line is a clear indication for an incompletely screened excitonic interaction. The magnetic-field-dependent shift of the heavy-hole exciton ground state in three dimensions has been calculated numerically by Cabib, Fabri, and Fiorio[18] and was experimentally investigated on a large number of undoped quantum-well samples (e.g., Ossau *et al.*[10]). From the observed energy shifts the reduced effective masses μ of the heavy-hole excitons were deduced, which are a function of the well width L. In doped semiconductors the exciton binding energy is reduced because of the screening of the Coulomb interaction by the free carriers. In the limit of a complete screening the magnetic-field-dependent shift corresponds to the Landau shift of the band edge state ($\Delta E = \frac{1}{2}\hbar eB/\mu$). This case seems to be realized by the heavily n-type modulation-doped GaAs/$Al_xGa_{1-x}As$ sample ($n = 4.8\times10^{11}$ cm^{-2}) investigated in Ref. 17).

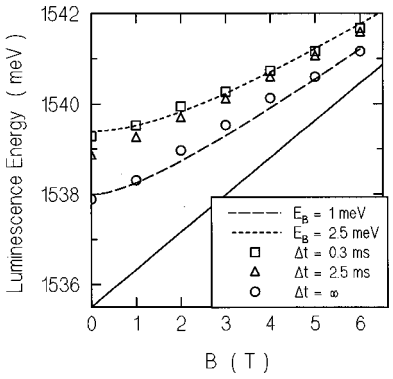

FIG. 5. Shift of the luminescence energy as a function of the magnetic flux density B for different delay times Δt. The dashed and dotted lines represent the diamagnetic shifts of excitons with binding energies E_B and an effective reduced mass $\mu = 0.77m_0$ calculated according to Ref. 19. The solid line corresponds to the linear Landau shift of the band states and serves for comparison.

If the screening of the Coulomb interaction between electrons and holes is incomplete, the slope of the diamagnetic shift depends on the exciton binding energy E_B, i.e., on the actual carrier concentration in the quantum well. As can be seen in Fig. 5 the photoinduced changes of the hole concentration in our p-type-doped structure affect the diamagnetic shift of the luminescence line. With increasing delay time Δt the slope of the magnetic-field-dependent shift increases. With increasing hole concentration the field dependence of the luminescence line approaches the linear Landau shift of the band states (solid line in Fig. 5). Making use of the theoretical results of Ref. 18 we have calculated the diamagnetic shifts of excitons with binding energies E_B to describe our experimental results displayed in Fig. 5 keeping the value of the reduced effective exciton mass $\mu = 0.07m_0$ ($L_z = 12$ nm, from Ref. 10) fixed. This permits us to give an estimate of the variation of E_B with Δt. The photoinduced reduction of the hole concentration in the quantum wells at a delay time $\Delta t = 0.3$ ms results in an increase of the exciton binding energy of about 1.5 meV compared to the value of E_B without sample illumination. This means that in the range of hole concentrations realized in our sample ($p \leq 1.7\times10^{11}$ cm^{-2}) the energy of electron-hole recombination is significantly conditioned by BGR as well as by excitonic interaction. For decreasing hole concentrations in the quantum wells the blueshift caused by decreasing many-particle interaction effects is partly compensated by a redshift due to the increasing excitonic binding energy E_B. To reach an estimate of the real size of the photoinduced change of the BGR the measured blueshift of the photoluminescence signal and the variation of the exciton binding en-

ergy have to be added. This gives a factor of about 2 for the size of the BGR compared to the value deduced from the shift of the luminescence line only. So the value of Δp displayed in Fig. 4 can be seen as a lower limit of the true variation of the hole density, which might be larger by the same factor.

Our observations of a finite excitonic electron-hole interaction in our p-type sample are in agreement with measurements of Akiyama, Matsusue, and Sakaki[19] who deduced a weak efficiency of the screening of Coulomb interaction in p-type modulation-doped structures from the observation of well-resolved excitonic peaks in photoluminescence excitation spectra.

IV. CONCLUSION

In summary, the many-body effect of the BGR was used to control the photoinduced variation of the hole density in the quantum wells of p-type modulation-doped heterostructures. After the depopulation of the wells achieved by illumination of the sample with an Ar^+ laser ($\lambda = 514$ nm) we observed the charge relaxation process by measuring the fundamental quantum-well photoluminescence making use of a very simple experimental setup. By changing the delay between the transfer of the holes into the barrier regions and the detection of the luminescence signal the dynamics of the refilling of the wells could be illustrated. Hereby, the success of our experimental setup is mainly based on the long lifetime of the photoinduced holes in the barrier regions in the range of some milliseconds. Using a theoretical expression of the BGR the density of the two-dimensional hole gas could be derived. The relaxation velocity decreases with time, which is consistent with our model describing the back transfer of the holes into the wells by a tunneling process. Finally we studied the excitonic effects involved in the luminescence process by applying a magnetic field along the growth direction. It turned out that excitonic effects are present in our doped sample with the consequence that the photoinduced changes of the density of the two-dimensional hole gas are considerably larger than calculated under neglect of excitonic contributions. Finally we want to mention that our experimental setup is also well suited to study effects of the photoinduced variations of the spatial distribution of the holes on Raman signals.[20,21]

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[1] A. Pinczuk, J. Shah, R. C. Miller, A. C. Gossard, and W. Wiegmann, Solid State Commun. **50**, 735 (1984).

[2] A. Pinczuk, J. Shah, H. L. Störmer, R. C. Miller, A. C. Gossard, and W. Wiegmann, Surf. Sci. **142**, 492 (1984).

[3] D. A. Kleinman and R. C. Miller, Phys. Rev. B **32**, 2266 (1985).

[4] S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).

[5] M. H. Meynadier, J. Orgonasi, C. Delalande, J. A. Brum, G. Bastard, M. Voos, G. Weimann, and W. Schlapp, Phys. Rev. B **34**, 2482 (1986).

[6] C. Delalande, G. Bastard, J. Orgonasi, J. A. Brum, H. W. Liu, M. Voos, G. Weimann, and W. Schlapp, Phys. Rev. Lett. **59**, 2690 (1987).

[7] C. Weber, C. Klingshirn, D. S. Chemla, D. A. B. Miller, J. E. Cunningham, and C. Ell, Phys. Rev. B **38**, 12 748 (1988).

[8] E. A. Meneses, F. Plentz, and C. A. C. Mendonca, Superlattices Microstruct. **5**, 11 (1989).

[9] A. S. Chaves, A. F. S. Penna, J. M. Worlock, G. Weimann, and W. Schlapp, Surf. Sci. **170**, 618 (1986).

[10] W. Ossau, B. Jäkel, E. Bangert, and G. Landwehr, Surf. Sci. **174**, 188 (1986).

[11] J. C. Maan, Surf. Sci. **196**, 518 (1988).

[12] A. B. Henriques, E. T. R. Chidley, R. J. Nicholas, P. Dawson, and C. T. Foxon, Phys. Rev. B **46**, 4047 (1992).

[13] E. X. Ping and H. X. Jiang, Phys. Rev. B **47**, 2101 (1993).

[14] R. Stepniewski, W. Knap, A. Raymond, and B. Etienne, Phys. Rev. B **50**, 11 895 (1994).

[15] C. Schüller, J. Kraus, G. Schaack, G. Weimann, and K. Panzlaff, Phys. Rev. B **50**, 18 387 (1994).

[16] B. Jusserand, J. A. Brum, D. Gardin, H. W. Liu, G. Weimann, and W. Schlapp, Phys. Rev. B **40**, 4220 (1989).

[17] F. Plentz, F. Meseguer, J. Sánchez-Dehesa, N. Mestres, and E. A. Meneses, Phys. Rev. B **48**, 1967 (1993).

[18] D. Cabib, E. Fabri, and G. Fiorio, Nuovo Cimento **10**, 185 (1972).

[19] H. Akiyama, T. Matsusue, and H. Sakaki, Phys. Rev. B **49**, 14 523 (1994).

[20] J. Kraus, R. Hartmann, and K. Panzlaff, in *Proceedings of the 14th International Conference on Raman Spectroscopy*, edited by N. -T. Yu and X. -Y. Li (Wiley, New York, 1994), p. 338.

[21] J. Kraus, G. Weimann, and K. Panzlaff, J. Phys. Condens. Matter **7**, 7761 (1995).