Enhancement of the Pockels component in the electroreflectance spectra of quantum wells

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In this paper we present a study of the electroreflectance spectra of several quantum wells. Using polarized light we are able to separate the contributions related to the linear electro-optical tensor (Pockels effect) and the quadratic electro-optical tensor (Kerr effect). We have observed an enhancement of the linear electro-optical contribution to the electroreflectance spectra in the light-hole region of the fundamental transition. Such enhancement is not observed in the fundamental transition of the III-V compounds and is due to the mixing of the atomiclike orbitals induced in a quantum well.

The electric-field effects on the optical properties of semiconductor quantum wells (QW's) or superlattices (SL's) differ from those induced in bulk material. Most of these differences are due to the confinement of electrons and holes. For instance, in the case of QW's, the electric field produces strong energy variations in the fundamental transitions which are quadratic functions of the applied field [quantum confined Stark effect (QCSE)].[1] For multiple quantum wells (MQW's) or SL's the modification induced by the electric field depends not only on the magnitude of the field but also on the barrier width.[2,3] As a consequence of these differences the electro-optical properties of QW's or SL's are not easily related to those of the bulk constituent compounds.

For moderated fields the changes in the dielectric constant induced by the electric field can be split in terms of two complex tensors:[4] the linear electro-optical tensor (LEO) and the quadratic electro-optical tensor (QEO), which, respectively, describe the linear variations (LEO, Pockels effect) and quadratic variations (QEO, Kerr effect) with the electric field. The Pockels effect is linked to the influence of the electric field on the lattice periodic part of the Bloch functions in the crystal,[5,6] whereas the Kerr effect results from the modification on the envelope wave function due to the electric field.

The confinement changes the spectral dependence and the magnitude of these tensors, and increases the number of independent components due to the reduction of the symmetry. For example, near the fundamental transition there is a strong increase of the Kerr effect,[7–9] due to the QCSE. In bulk material, this enhancement is also present, but is less pronounced[10,11] and is related to the low-field limit of the Franz-Keldysh effect.[12,13] For energies below the band gap, the Pockels effect is dominant, both in bulk and in QW's or SL's, and its magnitude is nearly constant up to the band-gap energy.[9,14] Only recently have there been reports of a Pockels effect enhancement near a forbidden transition in MQW's structures.[15]

In this paper, we present experimental evidence of an enhancement of the Pockels effect in QW's. Due to the confinement, the light-hole (LH) QW states have an s component, which does not appear in bulk states. Such a change in the weight of the atomiclike orbitals allows field-induced couplings that are linear functions of the applied electric field, thereby increasing the LEO coefficient. In order to investigate the modification induced in the Pockels effect by the reduction of dimensionality, we have performed polarized electroreflectance (ER) measurements on (001) oriented GaAs quantum wells, with the electric field applied along the growth direction, and the incident and reflected light normal to the growth plane. We have observed an increase in the LEO coefficient in the $n=1$ LH transition.

Due to the lower symmetry in the QW as compared to the bulk, the number of independent components of the electro-optical tensors increases: the LEO tensor has now two independent components instead of one, and the QEO tensor has six instead of three. In our experimental configuration only one LEO and one QEO component must be considered. For light polarized along the $[1\bar{1}0]$ direction the reflectivity can be written as[16,17]

$$R(\omega,F)=R(\omega,0)[1-L(\omega)F+Q(\omega)F^2] ,$$

where ω is the frequency of the light, F is the electric field inside the QW, and L (respectively, Q) is a combination of the real and imaginary part of the r_{63} component of the LEO tensor (respectively, the R_{13} component of the QEO tensor). If the light is polarized along the [110] direction we have

$$R(\omega,F)=R(\omega,0)[1+L(\omega)F+Q(\omega)F^2] .$$

Then,

$$\frac{\Delta R_{[1\bar{1}0]}}{R_{[1\bar{1}0]}} \propto (2QF_0-L)\left[\frac{\partial F}{\partial V}\right]_{F_0}\Delta V , \quad (1)$$

$$\frac{\Delta R_{[110]}}{R_{[110]}} \propto (2QF_0+L)\left[\frac{\partial F}{\partial V}\right]_{F_0}\Delta V\ , \qquad (2)$$

where F_0 is the static electric field inside the QW and ΔV the modulating voltage. Therefore, the difference between the two spectra is related to the Pockels effect, and the sum to the Kerr effect.

The samples used in this work where grown by molecular beam epitaxy on (001) GaAs substrates. The thickness of the GaAs QW has been varied between 50 and 40 Å and the barrier material is either $Al_xGa_{(1-x)}As$ with $x=0.35$, or AlAs. All samples have a 50-Å GaAs cap layer. A thin Au film was deposited on the surface to create a semitransparent Schottky diode. ER measurements at 80 or 300 K were performed with a standard experimental setup. In addition to the ac modulating voltage we have applied a dc voltage, which was used to change the electric field inside the well. In an ideal Schottky diode, the relation between the electric field at a distance x from the surface and the applied voltage is given by[18] $F=F_{\max}(1-x/W)$, where W is the depletion width and $F_{\max}\sim(C-V)^{1/2}$, V being the applied voltage, and C a constant that depends on the Schottky barrier height and on the temperature. In our samples, the GaAs QW's are located at distances between 600 and 1600 Å from the surface, depending on the sample, and for the dc voltages used here, we have $x/W\ll 1$ and hence $F=F_{\max}$.

In Figs. 1 and 2, we show the experimental spectra corresponding to a 40-Å GaAs QW with $Al_xGa_{1-x}As$ barriers and a 45-Å GaAs QW with AlAs barriers, respectively. Only the energy region of the fundamental transitions is presented; no other features are observed in the spectral range studied here (1.9–1.5 eV). Similar spectra are obtained for the other samples.

To simplify the notation, $\Delta R_{1\bar{1}0}/R_{1\bar{1}0}+\Delta R_{110}/R_{110}$ spectra will be called from now on "+" spectra, and $\Delta R_{1\bar{1}0}/R_{1\bar{1}0}-\Delta R_{110}/R_{110}$ spectra will be called "−" spectra. The + spectra have two features corresponding to transitions between the $n=1$ electron state and the $n=1$ heavy-hole state (e-HH) or $n=1$ light-hole state (e-LH). The − spectra have only one feature in the energy region of the e-LH transition. The square of the intensity ratio of the e-LH transition of the + spectra to the e-LH feature of the − spectra increases linearly as we increase the reverse voltage (i.e., as the electric field inside the QW is increased). This field dependence of the intensity ratio agrees with the expected behavior in the quadratic and linear electro-optical components of the electroreflectance spectra.

The shapes of the + and − spectra are quite different. The + spectra display a first derivative line shape for both heavy-hole and light-hole transitions, implying that the main mechanism of modulation in these spectra is the energy modulation of the transition (QCSE).[19] On the contrary, the shape of the − feature suggests an intensity modulation mechanism for this transition.

From all the above mentioned results we conclude that the features observed in the − spectra are related to the Pockels effect which is enhanced in the energy region of the light-hole transition. The spectral dependence of the − spectra suggest that the intensity of the transition has a strong linear dependence on the electric field, whereas in the case of the Kerr effect (+ spectra), it is the energy of the transition that has a strong quadratic dependence on the field.

The confinement produces not only changes in the localization of the particles, but also modifies the relative weights of the different components $[u_{\xi k}(r)]$ of the lattice

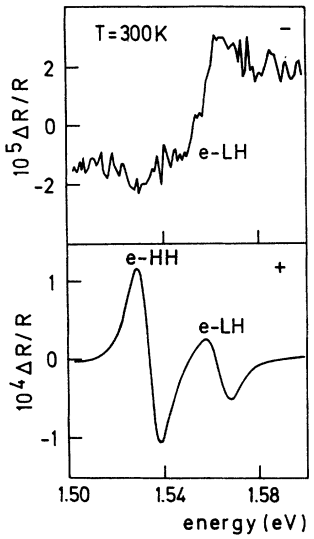

FIG. 1. $\Delta R_{1\bar{1}0}/R_{1\bar{1}0}-\Delta R_{110}/R_{110}$ spectrum "−" and $\Delta R_{1\bar{1}0}/R_{1\bar{1}0}+\Delta R_{110}/R_{110}$ spectrum "+" of a 40-Å GaAs QW with $Al_xGa_{(1-x)}As$ barriers taken at 300 K. Note the different shape of the spectra. The − spectrum suggest an intensity modulation mechanism and the + spectrum an energy modulation one.

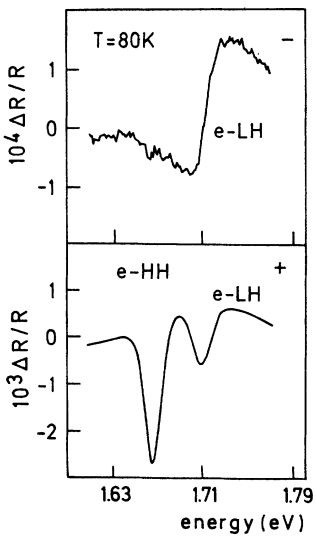

FIG. 2. $\Delta R_{1\bar{1}0}/R_{1\bar{1}0}-\Delta R_{110}/R_{110}$ spectrum "−" and $\Delta R_{1\bar{1}0}/R_{1\bar{1}0}+\Delta R_{110}/R_{110}$ spectrum "+" of a 45-Å GaAs QW with AlAs barriers taken at 80 K. Note the different shape of the spectra. The − spectrum suggests an intensity modulation mechanism and the + spectrum an energy modulation one.

periodic part of the Bloch function of the crystal needed to build a particular QW state. Within the formalism of the $\mathbf{k}\cdot\mathbf{p}$ theory, the wave function of a QW state can be written as

$$\Psi(r)=\sum_{\xi} F_{\xi}(r)u_{\xi 0}(\mathbf{r}) , \tag{3}$$

where $u_{\xi 0}(\mathbf{r})$ is the $u_{\xi k}(\mathbf{r})$ function at $\mathbf{k}=\mathbf{0}$ and F_{ξ} is the ξ component of the envelope wave function. In a four-band $\mathbf{k}\cdot\mathbf{p}$ model, ξ runs through the conduction band (CB), light-hole (LH), heavy-hole (HH) and spin-orbit (SO) bands. For a (001) oriented QW, the wave functions of the QW LH states have components on the u_{CB}, u_{LH}, and u_{SO} functions, contrary to what happens to the HH states which only have components on the u_{HH} function. This difference is the origin of the different behavior in the e-HH and e-LH transitions observed in the present study. In the $\mathbf{k}\cdot\mathbf{p}$ formalism, the intensity of the transition between state 1 (i.e., electron) and the state 2 (i.e., hole) can be expressed as follows:

$$|\langle\Psi_1|H|\Psi_2\rangle|^2 \propto \left| \sum_{\xi\xi'} \langle F_{1\xi}(r)|F_{2\xi'}(r)\rangle\langle u_{1\xi}|\nabla_{\chi}|u_{2\xi'}\rangle + \sum_{\xi\xi'} \langle u_{1\xi}|u_{2\xi'}\rangle\langle F_{1\xi}(r)|\nabla_{\chi}|F_{2\xi'}(r)\rangle \right|^2 , \tag{4}$$

where χ represents the direction of the polarization of light; in our case, χ is contained in the (001) plane. For a perfect QW there is full translational symmetry along the in-plane direction and thus

$$F_{j\xi}(r)=F_{j\xi}^{k_{j\parallel}}(z)e^{ik_{j\parallel}r_{\parallel}} , \tag{5}$$

where $j=1,2$; and z and $r_{\parallel}$ indicate the growth and the in-plane directions, respectively. The notation clearly indicates that the wave function has a well-defined in-plane $k_{\parallel}$ value. Therefore, (4) becomes

$$\left| \sum_{\xi\xi'} \langle F_{1\xi}^{k_{1\parallel}}(z)|F_{2\xi'}^{k_{2\parallel}}(z)\rangle\delta_{k_{1\parallel},k_{2\parallel}} \times[\langle u_{1\xi}|\nabla_{\chi}|u_{2\xi'}\rangle + ik_{2\chi}\delta_{\xi\xi'}] \right|^2 . \tag{6}$$

The first factor in this expression gives rise to the usual parity selection rules for the envelope wave function. For states at the Γ point, which are the ones to be considered here, $k_{1\parallel}=k_{2\parallel}=0$ and the last term is zero for a perfect QW. However, in a real QW there are always in-plane fluctuations (interface roughness, impurities, etc.) which could make this term no longer zero if states 1 (electron) and 2 (hole) have components on the same wave-function basis ($\xi=\xi'$). For the transitions studied here, states 1 and 2 have components on the same wave-function basis only if state 2 is a light hole.

An estimation of the amount of in-plane fluctuations needed to observe the present phenomenon can be made assuming that they relax the $\Delta k_{\parallel}=0$ selection rules. For the e-LH transition the expression (6) turns to

$$\left| \sum_{\xi\xi'} \langle F_{1\xi}|F_{2\xi'}\rangle\langle u_{1\xi}|\nabla_{\chi}|u_{2\xi'}\rangle + ik\{\langle F_{1\mathrm{LH}}|F_{2\mathrm{LH}}\rangle + \langle F_{1\mathrm{CB}}|F_{2\mathrm{CB}}\rangle + \langle F_{1\mathrm{SO}}|F_{2\mathrm{SO}}\rangle\} \right|^2 . \tag{7}$$

Due to the selection rule for the ∇_{χ} matrix elements, only $\langle s|\nabla_i|p_i\rangle=(m_0^* E_p/2)^{1/2}$ must be considered (where $i=x,y,z$; m_0 is the electron-free mass and $E_p\sim 23$ eV (Ref. 20).

As an example, we present in Fig. 3 the envelope functions of the $n=1$ LH-like state and $n=1$ conduction-band state of a 45-Å GaAs QW with AlAs barriers, calculated using a four-band (CB, HH, LH, and SO) $\mathbf{k}\cdot\mathbf{p}$ model (for clarity the envelope of the $u_{\xi 0}(\mathbf{r})$ function of the SO band maximum is not depicted).

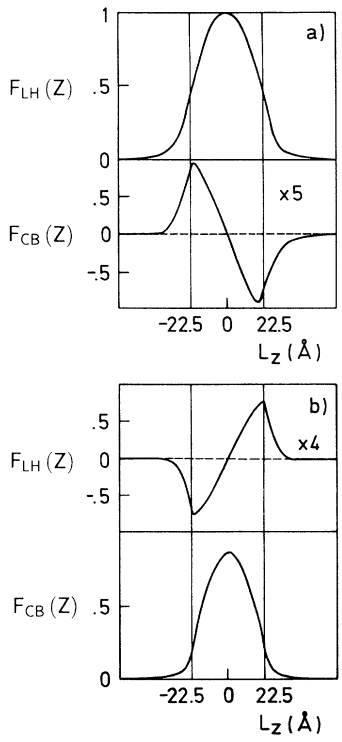

FIG. 3. Light-hole-band and conduction-band envelope wave functions components of a 45-Å GaAs QW with AlAs barriers (a) $n=1$ light-hole-like state and (b) $n=1$ conduction-band state.

As can be observed in this figure, for the $n=1$ LH-like state the CB component of the envelope wave function F_{CB} has odd parity and the LH component of the envelope wave function $F_{2\mathrm{LH}}$ has even parity. The SO com-

ponent, not shown, also has even parity. The corresponding components of the $n=1$ conduction-band state have opposite parity. Let us consider the transition between the $n=1$ conduction-band state and the $n=1$ LH-like state. For zero electric field, due to the different parity of the envelope wave functions, there is no contribution of the k terms in expression (7) to the intensity of the transition. When an electric field is applied, the envelope wave functions are modified and, in particular, the k terms of (7) are proportional to the electric field. Therefore, the total intensity of the transition, which is proportional to the square of the sum of the two terms of expression (7), should have a linear term with the electric field. In our samples, the k needed to explain the observed intensity of the present phenomena can be estimated to be about 0.005–0.002 times the $k_{\max}$ of the first Brillouin zone.

The present behavior is quite different from that observed in bulk material. The lattice periodic part of the Bloch function at the valence-band maximum of bulk III-V compounds has only p_x, p_y, and p_z atomiclike orbital components, with no s component, which results in a small coupling of these functions to the electric field. Therefore, there has been no experimental observation of a linear component in the ER spectra of the fundamental E_0 transition.[21] In order to observe such a component we must go to higher transitions, i.e., E_1.[22] A linear component in the ER spectra of the E_0 transition has been observed in crystals where the functions of the valence-band maximum has a strong mixing of d and p atomiclike orbitals (i.e., CuCl).[6]

In conclusion, we have studied the ER spectra of several QW's. In the energy region of the fundamental transition the field-induced modification of the optical properties of the LH transitions has a linear component with the electric field. Such a contribution to the ER spectra has not been observed in the fundamental transition of the bulk material and is explained in terms of the mixing between conduction-band and valence-band wave functions induced in the QW.

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