Quantum confined Pockels effect and optical polarized spectroscopy of interfaces in type-II heterostructures

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Abstract. We report on a strong enhancement of the Pockels effect in direct-gap type-II ZnSe/BeTe heterostructures. The effect has been found experimentally and analyzed theoretically. In quantum well structures with equivalent interfaces, say the Zn-Te and Te-Zn interfaces, the unbiased sample is optically isotropic in the plane perpendicular to the growth direction. In the presence of an electric field the electron and hole wavefunctions are pushed towards one of the interfaces and off from the other, as a result the balance is broken and the photoluminescence polarization becomes nonzero and reaches 70–80 in moderate electric fields. To describe the field-induced in-plane anisotropy in type-II heterostructures, a theoretical model has been developed which is based on the representation of optical matrix elements in the microscopical tight-binding model.

For zinc-blende heterostructures CA/C'A' grown along the [001] principal axis, the point symmetry, C_{2v} , of a single interface is reduced as compared with the point group T_d for a bulk material and D_{2d} for a quantum well (QW) with symmetrical interfaces. It leads to new optical phenomena, in particular, to the in-plane optical anisotropy. The latter can be observed in QW structures if the normal and inverted interfaces are nonequivalent as realized on heteropairs with no-common atom or by distorting the free carrier wavefunction in an electric field and making different its values at the opposite interfaces.

Up to now, the in-plane optical anisotropy in QW structures induced by normally applied electric field, or the quantum confined Pockels effect, has been observed for two heterosystems both of them having a type-I band alignment. Kwok et al. [1] studied photoluminescence (PL) of biased GaAs/AlGaAs multiple QWs (MQWs), no noticable anisotropy was found for allowed interband transitions whereas the nominally forbidden PL peak e^{2-hh1} showed a substantial difference reaching 20–30% between [110] and [110] polarizations. Krebs et al. [2] reported the observation of a dichroism up to 11% in In-GaAs/InP MQWs inserted into a *p-i-n* diode. The quantum confined Pockels effect in type-I heterostructures has been described [2, 3] in terms of the generalized envelope function approximation [4]. In type-I QWs, where the interband optical transitions are spatially direct, the overlap between electron and hole wave functions extends over the whole well layer which results in averaging and considerably reducing the effect of interface-induced and electric-field affected heavy-light hole mixing. On the contrary, in type-II structures the spatially-indirect transitions arise due to the electron-hole overlap only within an extremely narrow region containing the interface. Therefore, the transition oscillator strength has to be strongly correlated with anisotropic orientation of the interface chemical bonds. This is confirmed by observations of high linear polarization of the PL in InAs/AlSb [3] and ZnSe/BeTe [6] MQWs with nonequivalent normal and inverted interfaces.

In the present work we have studied the Pockels effect in type-II ZnSe/BeTe doublebarrier heterostructures. Modification of the band diagram under external electric fields



Fig. 1. The polarized PL spectra measured at different bias voltages for the analyzer orientation along the x' and y' axes of the sample A with $Zn_{0.9}Mn_{0.1}Se$ in the middle layer (a) and sample B with the pure ZnSe middle layer (b).

allowed us to detect signal from an individual interface and to determine orientation of chemical bonds from the induced optical anisotropy effect. A set of resonant tunneling ZnSe/BeTe double-barrier structures (DBS) were grown on GaAs:Si(001) substrates by molecular-beam epitaxy. The layer sequence is symmetric and comprises lattice matched BeZnSe:I (300 nm) contact layers, undoped ZnSe (25 nm) spacer layers, which act as electron emitters under bias voltage applied, and the BeTe/ZnSe/BeTe (4 nm/5 nm/4 nm) double-barrier structure. In sample B the central layer was of pure ZnSe while in sample A it was substituted by a $Zn_{0.9}Mn_{0.1}Se$ (5 nm) layer. For the both structures the normal (BeTe on ZnSe) and inverted (ZnSe on BeTe) interfaces are grown respectively under Zn and Te termination and contain the Zn-Te chemical bonds.

The PL spectra detected in linear polarizations along [110] and [110] axises are shown in Fig. 1 for the two studied structures. The emission lines under study originate from the spatially indirect transitions involving electrons from the ZnSe emitters (E) or quantum wells (W) and holes from the BeTe layers. The both samples demonstrate very similar behavior. At zero electric field pairs of PL lines at 1.84 eV for the sample A and at 1.87 eV for the sample B are due to recombination of electrons from the left- and right-hand side emitters and photoholes in BeTe (the lines EI and EN). Additionally, in the sample B a pair of the WI and WN lines at 1.95 eV involving electrons from the well layer is observed. Splitting between the EN and EI lines can be attributed to small charge-asymmetry of the emitters. Positive electric fields push off electrons from the rightmost Zn-Te interface and form a triangular potential near the leftmost Te-Zn interface pressing electrons towards the



Fig. 2. Effect of the electric field on the position (a), intensity (b) and polarization (c) of the PL lines due to optical transitions from the ZnSe emmitters to the adjacent BeTe layers in the sample A.

latter. As a result the relative intensity of the EN line contributed by transitions at the Zn-Te interface increases and that for the EI line related to the Te-Zn interface vanishes. The field reversal leads to exchange of the role played by the interfaces. The bias-induced PL intensity variations and energy shifts for the EN and EI lines in the sample A are presented in Fig. 2(a) and (b). The field-induced optical anisotropy, i.e. the Pockels effect, is visualized in Fig. 2(c) as a degree of PL linear polarization in the axes $I_{1\bar{1}0}$, I_{110} . For both directions of the bias voltage the absolute value of P_l rapidly increases, saturates to 0.7–0.8 at the voltage $U \ge 0.25$ V and is almost invariable for higher U up to ± 2 V. Under the same conditions the linear polarization detected in the axes [100] and [010] was absent.

In the ZnSe/BeTe system the conduction- and valence-band offsets amount to 2 eV and 1 eV respectively and the penetration depth for an electron into the BeTe layer or for a hole into the ZnSe layer is of the order of one monomolecular layer. Therefore in type II direct-gap ZnSe/BeTe heterostructures the wavefunctions of an electron and a hole participating in the spatially indirect transition overlap remarkably only over few atomic planes. In this case calculation of the interband matrix elements presupposes the knowledge of microscopic behavior of the wavefunctions at the interfaces which can be obtained by using pseudopotential or tight-binding models. From the polarization degree of 0.7–0.8 we obtain for the ratio between the indirect-transition matrix elements: $|M_{x'}/M_{y'}| = 2.4-3$ for one and the inverse value for the other interface. This means a strong in-plane anisotropy for the interband matrix elements of the velocity operator, \mathbf{v}_{cv} . We express the matrix elements in terms of the tight-binding parameters by using representation given in [7] and based on the quantum mechanical definition of the velocity operator $\mathbf{v} = (i/\hbar)[H\mathbf{r}]$, where *H* is the tight-binding Hamiltonian.

In the nearest-neighbor sp^3 model the scalar product of \mathbf{v}_{cv} and the light polarization unit vector \mathbf{e} can be expanded for the electron and hole states with zero in-plane wavevector as

$$\frac{ia_0}{4\hbar} \sum_{l} \left[V(sa, pc) C_{2l,s}^* \left(C_{2l-1,x'} e_{x'} \pm i C_{2l+1,y'} e_{y'} \right) + V(pa, sc) C_{2l-1,s}^* \left(C_{2l,x'} e_{x'} \pm i C_{2l-2,y'} e_{y'} \right) \right].$$
(1)

Here the sign \pm corresponds to the transitions from the $\pm 3/2$ heavy-hole valence states to the spin-up and spin-down conduction states which are written in terms of the *s*-like planar orbitals ϕ_{ns} with the expansion coefficients C_{ns} and *p*-like planar orbitals with the expansion coefficients C_{np_j} ($j = x' \parallel [1\bar{1}0]$], $y' \parallel [110]$), the integer *n* is even and odd respectively for the anion and cation planes, a_0 is the lattice constant, V(sa, pc) and V(pa, sc) are the two linearly independent *sp*-interaction parameters. The terms in the sum (1) describe contributions to the optical matrix element due to particular interatomic optical transitions. It should be emphasized that the transitions $2l \leftrightarrow 2l - 1$ and $2l \leftrightarrow 2l + 1$ are polarized orthogonally. The experimental high degree of linear polarization can be understood taking into account that only a finite number of terms in Eq. (1) are important, say those arising due to the Se(s) \rightarrow Zn(p), Zn(s) \rightarrow Te(p), Te(s) \rightarrow Be(p) interatomic transitions and that the product $C_{-1,s}C_{0,y'}$ exceeds $C_{-2,s}C_{-1,x'}$ and $C_{0,s}C_{1,x'}$, where n = 0 is assigned to the Te interface plane.

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