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Linearly-polarized photoluminescence from type II ZnSe/BeTe quantum wells with atomically-flat interfaces

A. A. Toropov†‡, S. V. Sorokin†§, S. V. Ivanov†§, N. Hori‡, M. Ichida‡,
A. Nakamura‡, A. Waag¶ and G. Landwehr§

† Ioffe Physico-Technical Institute, St Petersburg, Russia

‡ Nagoya University, Chikusa-ku, Nagoya 464-8603, Japan

¶ Abteilung Halbleiterphysik, Universität Ulm, 89081 Ulm, Germany

§ Physikalisches Institut der Universität Würzburg, D-97074 Würzburg, Germany

ZnSe/BeTe multiple quantum well (MQW) heterostructures have recently attracted attention due to the unique electronic and optical properties [1]. Since the hetero-interface between ZnSe and BeTe is of a strong type II, electrons are localized in ZnSe layers and holes in BeTe. Both electron- and hole-band offsets are extremely high (~ 2 eV for electrons and ~ 1 eV for holes). As a result, the photoluminescence (PL) of the MQW samples originates in spatially indirect transitions. Usually, one or two emission bands are observed in a 1.9–2.2 eV region, depending on a thickness of the constituent layers and experimental conditions [2, 3]. The higher-energy PL peak originates from band-to-band, spatially indirect transitions, whereas the lowest-energy one has been attributed to emission of carriers localized by fluctuations of the QW thickness or some other interface imperfections [3]. The reported spectral width of the peaks was rather large (10–100 meV), which prevented accurate identification of the lowest-energy peak origin. Another specific feature of the PL bands in such samples is frequently observed strong linear polarization of the emission with respect to $[110]$ and $[1\bar{1}0]$ in-plane crystal axes. The polarization has been explained by breaking down the QW roto-inversion symmetry due to either application of external electric field (the quantum confined Pockels effect [2]) or occurrence of chemically non-equivalent left- and right-QW interfaces [3].

In this paper we report on molecular beam epitaxy (MBE) growth and PL studies of ZnSe/BeTe MQW samples with improved quality of interfaces. Narrow linearly polarized lines have been observed in cw PL spectra, which has been attributed to recombination of carriers localized by mono-layer (either *mono-molecular* or *mono-atomic*) fluctuations of the QW thickness.

A typical sample studied was grown by MBE in a Riber 2300 two-chamber system on a GaAs(001) substrate. A GaAs epitaxial buffer was used to improve the quality of the GaAs surfaces. Elemental Be, Te, and Zn effusion cells as well as a Se valve cracking cell were used as molecular beam sources. The substrate temperature was kept constant at T_S of 300°C. The ZnSe and BeTe layers were grown in a group VI-rich regime as confirmed by the $(2 \times 1)\text{Se}$ and $(2 \times 1)\text{Te}$ RHEED reconstructions at $\text{Se}/\text{Zn} \sim 1.5$ and $\text{Te}/\text{Be} \sim 2$ ($P_{\text{Te}}/P_{\text{Be}} \sim 20$) flux ratios, respectively. One should stress that for these experiments the Te/Be flux ratio has been lowered by 1.5 times as compared to that used for growth of the structures studied in [1–3], which has been found to result in much smoother layer-by-layer BeTe growth characterized by long lasting RHEED oscillations at T_S of 300°C. This agrees well with recent independent findings of the optimum MBE growth conditions of flat surface and high structural quality BeTe films [4].

The structure contains a 20-period ZnSe(5 nm)/BeTe(1.8 nm) MQW sandwiched between two thick $\text{Zn}_{1-x}\text{Be}_x\text{Se}$ layers nearly lattice-matched to a GaAs substrate ($x \sim$

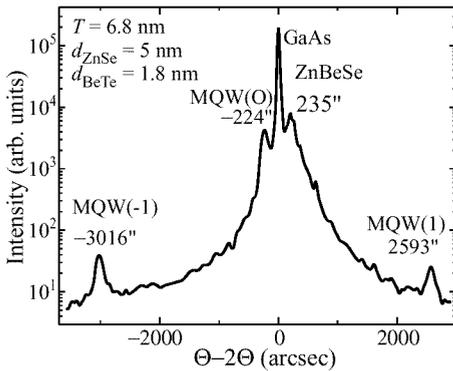


Fig. 1. (004) $\Theta-2\Theta$ XRD rocking curve of the structure.

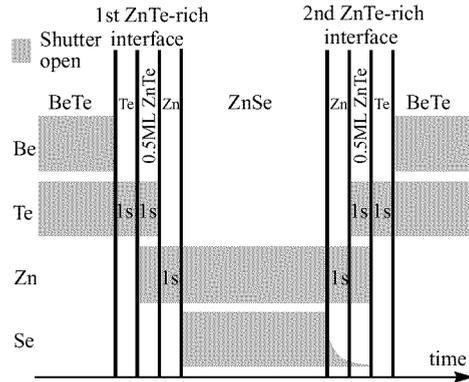


Fig. 2. Shutter sequence applied for obtaining ZnTe-rich interfaces.

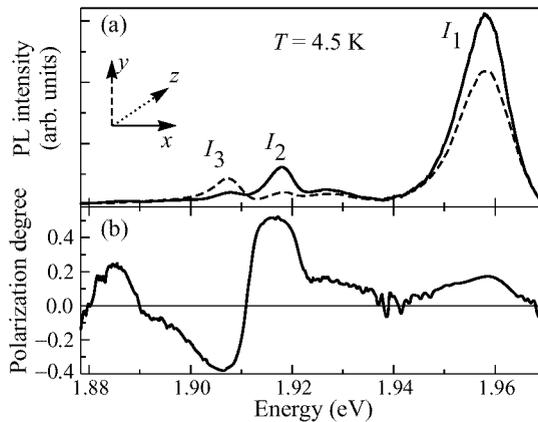


Fig. 3. (a) Photoluminescence spectra measured for two orthogonal linear polarizations along $[110]$ and $[1\bar{1}0]$ in-plane crystal axes (x and y directions); (b) the spectrum of the degree of linear polarization with respect to the $[110]$ and $[1\bar{1}0]$ axis.

0.03–0.04). Figure 1 shows an X-ray diffraction (XRD) rocking curve measured in the sample near a (004) reflection. Besides the peak originating from the GaAs substrate (at 0 arcsec), there are three narrow peaks attributed to 0-, +1-, and -1-order satellite reflections from the MQW structure and a peak at $235''$ relevant to the ZnBeSe claddings. Small width of the peaks as well as close to GaAs symmetrical arrangement of the 0-order MQW satellite and the peak related to the claddings indicate perfect structural quality and pseudomorphic nature of the structure.

The BeTe/ZnSe interfaces were grown as containing predominantly ZnTe bonds with a shutter sequence shown in Fig. 2. The first interface of the ZnSe QW (epitaxy of ZnSe on BeTe) was fabricated by a deposition of 0.5 ML ZnTe in MBE mode under Te-stabilized growth conditions on a Te-terminated surface of BeTe. Then the Te shutter was closed, providing a 1 s exposure of the surface to the Zn flux, which corresponds to deposition of the rest ~ 0.5 ML of Zn atoms, assuming the ZnSe growth rate to be 0.6 ML/s as measured by a RHEED oscillation technique. Although this Zn coverage can be slightly less than 0.5 ML due to some expected reduction of a Zn incorporation coefficient at the absence of

the Se flux, by analogy with the case of CdSe growth [5], it is expected that the additional Zn atoms mostly occupy ZnTe-free sites on the BeTe surface to complete the ZnTe ML. The deposition of BeTe on ZnSe was done by the opposite shutter sequence. A 1 s exposure of ZnSe surface to the Zn flux was followed by a 0.5 ML ZnTe deposition. Then the Zn shutter was closed for 1 s and BeTe growth was initiated on a Te-terminated surface of ZnTe. However, formation of the second ZnTe interface was done at a relatively high Se background pressure due to a finite time of Se valve closing (about 2 s). The latter growth peculiarity is expected to result in essential chemical bond type fluctuations (BeSe instead of ZnTe) at the second interface of ZnSe QWs (or at the first interface of BeTe QW), especially when taking into account the higher binding energy of ZnSe as compared to ZnTe. The latter is additionally reduced by a strain induced Gibbs free energy due to $\sim 7\%$ lattice-mismatch of ZnTe with respect to GaAs and the higher Be incorporation coefficient in comparison with that of Zn. Nevertheless, the roughness of the consequent interfaces of the MQW structure does not increase, as follows from a nearly constant RHEED specular spot intensity observed during the whole growth run, which is probably due to a strong surface smoothing effect of the BeTe growth under the growth conditions used.

Photoluminescence (PL) spectra were measured at 4.5 K in a bath He cryostat in a back-scattering geometry. A 325 nm laser line of a He-Cd laser was used for the PL excitation. The excitation spot size was about 0.5 mm, whereas the excitation power was kept below 1 mW. So the excitation power density in these experiments was below 0.4 W/cm^2 . The spectra were measured for two orthogonal linear polarizations along $[110]$ and $[\bar{1}\bar{1}0]$ in-plane crystal axes. Figure 3(a) demonstrates the two spectra of linearly polarized PL. There are three main peaks in the spectra, labeled as I_1 , I_2 and I_3 .

The peak intensities clearly depend on the detected light polarization, which is illustrated by the spectrum of linear polarization degree shown in Fig. 3(b). The degree of polarization P is defined as

$$P = \frac{I_{[110]} - I_{[\bar{1}\bar{1}0]}}{I_{[110]} + I_{[\bar{1}\bar{1}0]}}$$

where $I_{[110]}$ and $I_{[\bar{1}\bar{1}0]}$ are the PL intensities measured using a linear polarizer oriented along $[110]$ and $[\bar{1}\bar{1}0]$ directions, respectively. The polarization of I_1 peak is rather small (in the range 12–25%, depending on the experimental conditions), whereas I_2 and I_3 emission bands are linearly polarized in opposite directions at the maximum degree in the range of 40–60%. Note that FWHM value of the I_2 and I_3 peaks is as small as 3–5 meV. To our knowledge, this is the smallest value ever reported for PL peaks in ZnSe/BeTe samples, which confirms an extraordinary quality of the sample.

The observed peaks are located in the range of 1.9–2.0 eV, which is usual for observation of the PL related to spatially indirect excitons [1–3]. Keeping in mind that all interfaces in the sample are expected to be almost equivalent (ZnTe-like) one can anticipate relatively small in-plane linear polarization anisotropy of the excitonic emission [3]. From this point of view, it seems natural to attribute the most intensive, in spite of its higher energy position, smaller polarized peak I_1 to emission of indirect excitons in the sample regions with almost equivalent ZnTe-like interfaces.

To explain the two narrow oppositely polarized peaks I_2 and I_3 we assume occurrence of discrete mono-layer fluctuations of the QW thickness, by analogy with the well-known explanation of multiple PL peaks in GaAs/Al(Ga)As multiple QWs. The principle difference is that a ZnSe/BeTe multiple QW structure consists of binary materials which share neither common cations nor anions. For such structures there is a possibility of a mono-atomic (or half-mono-molecular) fluctuation of the QW effective thickness as well. This

imperfection can be regarded as a replacement of one atomic layer (either cation or anion) at the hetero-boundary between the two binary compounds with no common atoms, which results in a single-atomic shift of the interface, accompanied by a change of the interface chemical bond.

The energy gap between I_1 and I_2 is comparable with the carrier confinement energy corresponding to a mono-molecular fluctuation of the hole (BeTe) QW thickness. The respective energy is $\sim 35\text{--}40$ meV for heavy holes, as estimated within the effective mass approximation. For electrons this energy is much smaller (4–8 meV) because of a larger thickness of the ZnSe electron QWs. Therefore we attribute the two peaks (I_2 and I_3) to recombination of holes, localized in plane of the BeTe QW by a mono-layer thickness fluctuation, with electrons in left and right ZnSe layers. This interpretation also explains the opposite polarizations of I_2 and I_3 bands in agreement with the orthogonal polarizations of the interface bonds at the direct and inverted interfaces of an ideal QW with atomically abrupt interfaces [2]. The splitting between the two transitions can result from the inherent difference in the structure of the “left” and “right” interfaces of the BeTe layer, which is in agreement with the above sample growth analysis. In particular, only mono-molecular terraces are expected at the “left” (direct) ZnTe-like interface, whereas mono-atomic terraces can take place at the “right” (inverted) interface of the BeTe QW. The former terraces do not change the chemical type of the interface bond. The latter ones induce the change of the interface bond type and presumably the energy of the transition.

In conclusion, we have reported on improvements in the MBE growth of ZnSe/BeTe QW heterostructures with ZnTe-like interfaces, resulting in observation of two narrow oppositely polarized PL peaks. The peaks are attributed to recombination of holes localized by mono-layer fluctuations of the BeTe QW thickness with electrons from either left or right ZnSe layer. This interpretation is in qualitative agreement with the estimation of the carriers confinement energies, performed within the effective mass approximation for a mono-molecular QW thickness fluctuation, as well as with the symmetry analysis of the QW interface bonds. On the other hand, accurate quantitative description of the energy splitting between the peaks and their degree of linear polarization require additional studies.

Acknowledgements

The authors are thankful to E. L. Ivchenko for useful discussions. This work was supported in part by RFBR Grants, the Program of the Ministry of Sciences of RF “Physics of Solid State Nanostructures”, Volkswagen Stiftung and INTAS Grants No 97-31907 and No 00-02-16997a.

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