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Photoexcitation screening of the built-in electric field in ZnO single quantum wells

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ZnO/Mg0.22Zn0.78O quantum wells were studied by excitation-intensity-dependent luminescence at 10 K. The samples were grown by laser molecular-beam epitaxy on ScAlMgO4 substrates to evaluate the well width dependence (1 to 10 nm) of exciton transition energies. Under weak excitation, the photoluminescence shows a quantum-confined Stark effect for the wide wells. The well width dependences of the experimental transition energies are compared with previously reported calculations to evaluate the electric field due to spontaneous and piezoelectric polarizations. The internal electric field is comparable with 650 kV/cm. With an increase in excitation intensity, blueshift of the luminescence was observed, suggesting photoexcitation screening of electric fields. © 2008 American Institute of Physics. [DOI: 10.1063/1.2981523]

Wide band-gap semiconductors have attracted much attention due to their potential applications for optoelectronic devices operating in the blue and ultraviolet (UV) regions of the spectrum. Recently, ZnO and related oxides have been proposed as promising wide-gap semiconductors for short wavelength applications. The binding energy of ZnO excitons is relatively large at 60 meV so that the excitonic recombination could be permitted even at room temperature. Several groups reported the growth of the p-type doped ZnO films, which is very important for realizing practical application of light-emitting diodes. In addition, introduction of magnesium in ZnO plays a key role in band-gap engineering. Growth of ZnO/Mg0.22Zn0.78O quantum wells (QWs) using various experimental methods has been reported: laser molecular-beam epitaxy (MBE), metal-organic chemical vapor deposition, and MBE. In such a system grown along the c-axis, its optical properties are affected by both spontaneous and piezoelectric polarization effects. These effects result from the lattice mismatch and wurtzite symmetry. So far, the impact of strain in the ZnO well layer on the electric field has not been extensively studied. Bretagnon et al. pointed out the importance of lengthening a barrier layer thickness ($L_B$) to precisely evaluate the built-in electric field, which is 900 kV/cm for ZnO/Mg0.22Zn0.78O single QWs (SQWs). In this work, we report on optical properties of ZnO QWs and a screening effect of the built-in electric field with photocarriers studied by estimating the excitation intensity dependence of the photoluminescent spectra. The blueshift of the emission is observed under stronger excitation, probably due to the screening of the electric field by the photocarriers.

Our SQW samples were grown by laser MBE on high-temperature annealed MgZnO templates (being 100 nm in thickness) deposited on the c-plane of the ScAlMgO$_4$ (SCAM) substrates. The top barrier layer is composed of 50-nm-thick Mg$_x$Zn$_{1-x}$O ($x=0.22$). Thus, it could be regarded as a ZnO well sandwiched between Mg$_{1-x}$Zn$_x$O barriers. We have grown samples having a continuously spread variation in the well widths on a substrate using a moving mask during the deposition. The technique was adopted so as to suppress variation in the crystal growth conditions, which is suited for systematic study on the material properties. Details of the growth conditions and experimental setup have been given elsewhere. These samples were characterized by photoluminescence (PL) spectroscopy at temperature of 10 K. Two QW samples, having nominally same specifications, were studied. The well width of the first sample ranges from 1 to 10 nm, while the other one ranges from 2.7 to 8.3 nm. A continuous-wave He–Cd laser (emitting wavelength is 325 nm) was used as an excitation source.

Figure 1(a) shows near-band edge PL spectra in ZnO/Mg$_{0.22}$Zn$_{0.78}$O SQWs for eleven different well thicknesses (2.7 nm = $L_w$ = 8.3 nm). The excitation intensity is ~160 kW/cm$^2$. For wide well widths ($L_w$ ≥ 3.9 nm), the QW spectra exhibit PL peaks below the exciton resonance of bulk ZnO (3.37 eV, a vertical dashed line in Fig. 1).

Stronger excitation yielded in different PL features, as shown in Fig. 1(b). The strong excitation condition was performed by the frequency-tripled pulsed beam from a Q-switch yttrium aluminum garnet laser (3.49 eV, ~1.6 kW/cm$^2$). Now, the PL peak energies are higher than or nearly equal to the resonance energy of the bulk. Second, the width of the peak became sharper under such stronger excitation.

Figure 2 shows the emission energy (triangles and squares) as a function of $L_w$ (1–10 nm) under weaker excitation. The PL energies obtained for the two specimens are in reasonable agreement with respect to each other. As expected, the emission from ZnO QWs exhibits strong dependence on $L_w$. Due to the quantum confinement effect, the blueshift of the emission was observed when $L_w$ is decreased. When $L_w$ ≥ 3.9 nm, strong redshift observed under weak excitation is a typical phenomenon of the existence of a quantum-confined Stark effect (QCSE), as illustrated in the...
Two sets of experimental spectra are shown under weak excitation conditions. Three sharp lines around 3.1 or 3.2 eV in samples taken at 10 K: the well widths are from Ref. 8. Dashed line corresponds to behavior of the energy shift with the presence of the built-in electric field caused by the excitation-induced carriers. The energy difference between experiments and the well thickness, as shown by a dashed line. The linearity is valid as long as the exciton binding energy is independent of the well thickness as later discussed in detail. The slope of the dashed line leads to an internal electric field of \( \sim 560 \text{ kV/cm} \).

The experimental data under stronger excitation \((\sim 1.6 \text{ kW/cm}^2)\) were also plotted against \( L_w \) (open circles, \( L_w = 2.7 \) to 8.8 nm). It is obvious that the emission energies became close to the calculation result of the excitonic transition energies (blue solid line) neglecting an electric field \((F=0 \text{ kV/cm})\) theoretically deduced by Bretagnon et al.\(^8\). It can be understood as a result of the screening of the internal electric field caused by the excitation-induced carriers. The energy difference between experiments (open circles) and calculation (solid line), for \( L_w = 6.6 \text{ nm} \), is about 40 meV. This energy difference could correspond to a localization energy of the QW excitons. The localization of excitons occurs due to fluctuations of the well width and/or barrier heights.\(^9\)

Here, we try to evaluate the built-in electric field of our samples. Bretagnon et al.\(^8\) calculated the exciton transition energies as a function of Mg composition \((x)\) of the barrier layers including the effects of internal electric field. The quenching of the excitonic binding energies \((E_b^0)\) due to the presence of the field is also taken into account in the calculation. For sufficiently wide wells, where the quantum-confinement effects are negligible, it can be regarded as an impact of electric field \((F)\) on the transition energies. Our experimental results are again plotted in Fig. 3 with the results of calculation abovementioned\(^8\) for \( F = 300, 650, \) and 900 kV/cm.

As it is understood from the comparison in Fig. 3, the experimental data are in reasonably good agreement with the calculated results for the field of 650 kV/cm. The value is slightly greater than that obtained in Fig. 2 \((=560 \text{ kV/cm})\),

\[ E_b^0(F=0) < E_b^0(F) \]
inherent to a ZnO almost same Mg composition. It is explained in terms of the slightly smaller than the previously reported value for the greater for narrow wells, energy difference is smaller than those at.

The field in the well is approximately proportional to geometrical effect. The field in the well is approximately proportional to geometrical factor. The photocarrier screening effect of interwell might be similar to that of the latter QWs. 

A self-consistent theoretical approach predicted the screening effect of the built-in polarization by Bretagnon and co-workers. 

For weakly and strongly photoexcitations, a QCSE is revealed. We deduced an internal function of well width. This tendency on function of well width is different from that of the latter QWs.

The magnesium composition of our SQWs is similar to Mg0.22Zn0.88O single QWs grown by laser MBE. The barrier layer thickness was evaluated in Ref.8, whereas the barrier layer thickness is slightly greater in magnitude than our value. 

It is reasonable because the Eb/LB = 200 nm.

Evidenced by the luminescence blueshift with an increase in the built-in electric field on the luminescence energy has been also shown. This tendency on the blueshift with an increase in the built-in electric field is different from that of the latter QWs. 

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