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DEPENDENCE OF EXCITON LINEWIDTH ON THE COMPOSITION OF $Zn_xMg_{1-x}Se$ LAYERS GROWN BY MBE[†]

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This work deals with the study of the photoluminescence and reflectivity properties of $Zn_xMg_{1-x}Se$ epilayers grown by molecular beam epitaxy on (001)GaAs and (111)ZnTe substrates. The photoluminescence spectra of $Zn_xMg_{1-x}Se$ layers grown on GaAs and ZnTe substrates are dominated by blue emission bands. The energetical positions and relative intensities of the bands depend on Mg contents in the epilayers. The shift of the maxima of blue emission toward higher photon energies and a simultaneous steep increase in the linewidth with an increase in Mg concentration are observed. A small amount of Mg added to ZnSe leads to a sharp increase in the linewidth from 2 meV in pure ZnSe layer grown on GaAs substrate to about 180 meV in $Zn_{0.78}Mg_{0.22}Se$.

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The usefulness of mixed ternary compounds of Mg halcogenides materials arises from the possibility of tuning of the band-gap energies and lattice constants by adjusting the content of the particular elements [1–3]. Unfortunately, there is a considerable lack of knowledge about the optical parameters of these materials grown on different substrates. Since major defect-related problems associated with the superlattices arise from interfacial lattice mismatch, the use of lattice-matched combinations is essential for constructing light emitting devices [4–6]. A detailed study of the edge luminescence in $Zn_xMg_{1-x}Se$/ZnTe and $Zn_xMg_{1-x}Se$/GaAs heterostructures is thus important to the understanding of the optical properties of the wide gap heterostructures, also in view of their application to blue-green opto-electronic devices.

In this paper we report luminescence and reflectivity properties of ZnSe and $Zn_xMg_{1-x}Se$ epilayers grown by molecular beam epitaxy on (001)GaAs and (111)ZnTe substrates as a function of temperature and Mg composition in the layer $(1-x)$ in the range from 0 to 0.4. These measurements were used to investigate the localization energy of the excitons caused by compositional fluctuations [7, 8].

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The $Zn_xMg_{1-x}Se$ layers were grown by solid source molecular beam epitaxy on GaAs and ZnTe substrates in facility described elsewhere [1]. The substrate temperature during the growth was kept in the range of 550–580 K. Growth rates of ZnSe and $Zn_xMg_{1-x}Se$ layers were in the range between 0.02 nm/s and 0.2 nm/s. The Mg content in the $Zn_xMg_{1-x}Se$ layers was determined from X-ray diffraction measurements assuming a linear dependence of the lattice constant with Mg concentration. The lattice constants for ZnSe and MgSe were assumed to be 0.5668 nm and 0.589 nm, respectively [9].

Photoluminescence (PL) and reflectivity spectra were measured in the temperature range from 7 K to 330 K using a closed cycle cryogenic system DE-202 (APD – Cryogenics Inc), an SPM-2 monochromator and a photomultiplier R-1333 (Hamamatsu). The PL measurements were performed using the 325 nm line of He–Cd laser (Omnichrom) at 4 mW for excitation. Low-temperature reflectance measurements have been performed in backscattering geometry using a 100 W tungsten-halogen lamp as a light source.

PL spectra of ZnSe crystal and $Zn_xMg_{1-x}Se$ epilayers grown on (111)ZnTe and (001)GaAs substrates are presented in Fig. 1A.

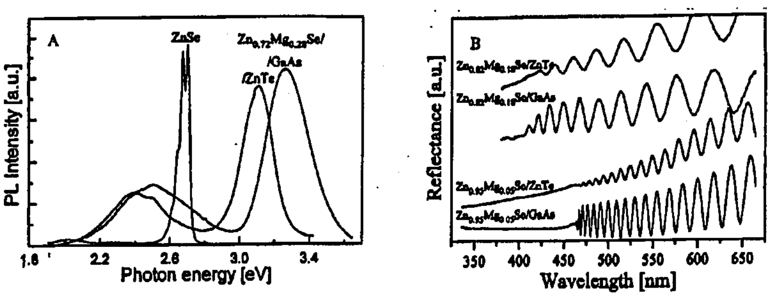

Fig. 1. Photoluminecence spectra of ZnSe crystal and $Zn_xMg_{1-x}Se$ epilayers grown on (111)ZnTe and (001)GaAs substrates (A). The reflection spectra of $Zn_xMg_{1-x}Se$ epitaxial layers grown on GaAs and ZnTe substrates (B).

The highest energetical PL bands in blue emission region observed in $Zn_xMg_{1-x}Se$ layers at low temperatures is associated with radiative recombination of free excitons. The relative intensities and energetical position of this bands depend on the Mg contents in $Zn_xMg_{1-x}Se$ epilayers. The shift of the maxima of PL bands toward higher photon energies with increase in Mg concentration is observed. The intensity of the blue band in luminescence spectra decreases with the increase in Mg content in $Zn_xMg_{1-x}Se$ layers. The decrease in the intensity is accompanied by a simultaneous steep increase in the linewidth. In $Zn_xMg_{1-x}Se$ layers grown on GaAs substrates a small amount of Mg added to $Zn_xMg_{1-x}Se$ layer leads to a sharp increase in the linewidth from 2 meV in pure ZnSe layer to about 190 meV in $Zn_{0.78}Mg_{0.22}Se$. The free exciton luminescence is observed up to the Mg content of $1 - x = 0.12$. At higher Mg content only one broad luminescence band is observed. The band-gap energy and lattice constant of $Zn_xMg_{1-x}Se$ increase with Mg content as well. Because the lattice constant of

GaAs is smaller than that of $Zn_xMg_{1-x}Se$, ($a_{GaAs} < a_{ZnMgSe}$) the biaxial compressive strain appears at the interface causing probably an additional shift of the luminescence band towards higher photon energy. When $Zn_xMg_{1-x}Se$ layer is grown on ZnTe substrate ($a_{ZnTe} > a_{ZnMgSe}$), we observe a shift of PL bands (compared to $Zn_xMg_{1-x}Se$/GaAs structure) in opposite direction due to the presence of tensile strain. These spectra were compared with a spectrum of the $Zn_xMg_{1-x}Se$ layer obtained by thermal diffusion of Mg into ZnSe crystal [1]. With the increase in Mg content in $Zn_xMg_{1-x}Se$ layer grown on ZnTe substrate the tensile strain decreases because the lattice constant of the grown layer increases.

Figure 1B presents reflection spectra of $Zn_xMg_{1-x}Se$ epitaxial layers grown on GaAs and ZnTe substrates. The strong interference fringes occur in the reflectivity spectrum for $h\nu < E_g$ where the epilayer is transparent and disappears abruptly for $h\nu > E_g$. The band-gap energies of all $Zn_xMg_{1-x}Se$ epilayers were estimated from measurements of reflection spectra near to the fundamental absorption edge at different temperatures. The temperature dependence of band-gap energy for $Zn_xMg_{1-x}Se$ epilayers grown on GaAs and ZnTe substrates has been estimated (Fig. 2A). The assignment of the edge luminescence has been confirmed by the coincidence of the high energy luminescence peaks with the free exciton signals in the photoreflectance spectra.

In Fig. 2B the dependence of the exciton linewidth on the composition of $Zn_xMg_{1-x}Se$ layer is shown. The half-width of the luminescence line measured

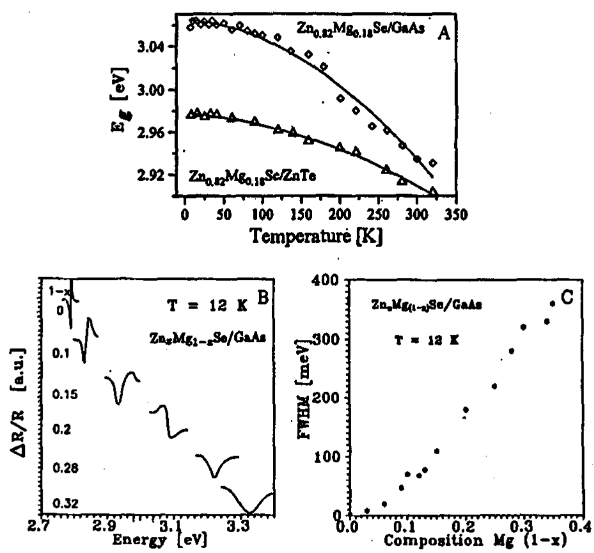

Fig. 2. The temperature dependence of band-gap energy for $Zn_xMg_{1-x}Se$ epilayers grown on GaAs and ZnTe substrates (A). Photoreflectance spectra ($\Delta R/R$) of $Zn_xMg_{1-x}Se$ layers with different Mg contents $(1-x)$ (B). The dependence of the free exciton half-width on the Mg composition $(1-x)$ of $Zn_xMg_{1-x}Se$ layers (C).

at $T = 12$ K increases with Mg content in $Zn_xMg_{1-x}Se$ layer grown on GaAs substrate from 2 meV for $1-x=0$ to 350 meV for $1-x=0.34$ and is considerably larger than that in ZnSe crystal (1 meV). This behavior is probably caused by the broadening due to the alloy disorder and due to the inhomogeneity in Mg concentration across the excited region. We observed a similar behavior of edge luminescence in $Zn_xMg_{1-x}Se$ layers obtained by thermal diffusion of Mg into ZnSe crystals [1].

The free exciton transitions of all $Zn_xMg_{1-x}Se$ epilayers were estimated from measurements of photoreflection spectra (PR) at different temperatures. The band position is determined by the mobility edge of localized excitons at compositional fluctuations [10, 11]. The energy difference between the luminescence band (which is due to localized states), and the free exciton recombination (estimated from PR spectra) can therefore be considered as a localization energy directly related to the compositional fluctuations [8, 9].

Based on the optical measurements it is difficult to perform estimations of Mg concentration in $Zn_xMg_{1-x}Se$ epilayers grown on ZnTe substrates because larger tensile stress modifies optical properties of the strained layers.

The study of the $Zn_xMg_{1-x}Se$ ($0 \leq 1-x \leq 0.4$) system allowed us to identify the growth conditions that yield layers of high optical quality. Along with tuning of the band-gap energy with concentration of Mg, these alloys are expected to provide tuning of the band offsets to ZnSe [2–5].

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