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Emissions from single localized states observed in ZnCdS ternary alloy mesa structures

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Sharp and discrete emission lines from single localized states in ZnCdS ternary alloys were clearly observed from selectively grown mesa structures. This is a demonstration that compound semiconductor alloy systems, which usually show broad emission spectra due to alloy fluctuations, are able to exhibit emissions from discrete energy levels with quasi-zero-dimensional density of states in limited mesa areas where limited number of deeper localized states will contribute. Introduction of ZnCdS/MgS short-period superlattices is found to play a significant role for the exciton migration enhancement from shallower to deeper localized states, which makes the observation of the emission lines from the single localized states possible in the ZnCdS alloy layers.

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Studies on quantum information technology, which is closely related to future quantum communication, computation, or cryptography, have made great strides in recent years.¹ It is essentially based on the principles of quantum mechanics, and controls of single electrons or single photons and of their mutual interactions are the issues of primary importance. Toward this direction, single-photon emitters (SPEs), which can provide single photons on demand, are key devices and have been actively studied. Single-photon emissions have been observed with such as single atoms,² single colloidal quantum dots (QDs),³ or single nitrogen-vacancy color centers.⁴ For the near-future realization of practical devices, however, SPE based on semiconductors will be preferable,⁵ and QDs with discrete density of states have been intensively investigated on InAs and its related semiconductor materials.^{6–8} From the viewpoint of oscillator strengths, II–VI semiconductors are more attractive to enhance the exciton-photon interactions,⁹ and single-photon emission has been studied with CdSe QDs^{3,10} and impurity states in ZnSe.¹¹ Preparation of such single-energy states in semiconductors forms the basis to realize SPE.

In this letter, a possibility to explore single energy states in II–VI semiconductors is demonstrated. Compound semiconductor alloys usually exhibit broad luminescence by the inhomogeneous broadening due to their compositional fluctuations. It will be shown that sharp and discrete emission lines are observable from single localized states in ZnCdS ternary alloy mesa structures, which were selectively grown in restricted surface areas. Furthermore, the role of ZnCdS/MgS short-period superlattices (SPSL) is discussed on their exciton migration enhancement from shallower to deeper localized states, which leads to the observation of the sharp and discrete emission lines from ZnCdS compound semiconductor layers.

The samples studied were prepared by metalorganic molecular-beam epitaxy on n^+ -GaAs (001) substrates. Precursors used were diethyl zinc, dimethyl cadmium,

bismethyl-cyclopentadienyl magnesium, and ditertiarybutyl sulfide. The growth temperature was 380 °C. Prior to the growth, carbonaceous masks with opening windows ranging from 100×100 to 900×900 nm² were patterned on a GaAs substrate. Further details on the mask patterning procedures are given in Ref. 12. In this study, 80 periods of ZnCdS/MgS SPSL with the respective ZnCdS and MgS layer thicknesses of 0.8 and 0.7 nm, determined from the growth time, were selectively grown in the mask openings. This resulted in mesa structures composed of four self-formed smooth {034} side facets and a (001) top surface. The Cd composition in the ZnCdS alloy is 0.58, which is closely lattice-matched to the GaAs substrate.¹³

Spatially resolved microphotoluminescence (μ -PL) measurements were carried out at the temperature of 35 K. A He-Cd laser line at the wavelength of 325 nm was focused on a sample surface employing a 100× microscope objective lens with a numerical aperture of 0.5, and this resulted in a probe beam diameter below 1 μ m ϕ . In order to pick up the luminescence exclusively from a single mesa structure, an orifice was placed just before the collection lens for the coupling into an optical fiber. The μ -PL signal was dispersed by a 0.55-m spectrometer and was detected by a liquid-nitrogen-cooled CCD detector. The spectral resolution of the whole measurement setup is approximately 540 μ eV near the wavelength of 400 nm.

In Fig. 1, the solid line shows the μ -PL spectrum measured from the ZnCdS/MgS SPSL mesa structure grown on a 600×600-nm² mask opening. This mesa structure is 70 nm high and has a \sim 400×400-nm² (001) top surface. Several distinct sharp emission lines denoted as P1–P4 are clearly observed. Additional lines at around 3.168 and 3.205 eV, labeled with crosses, are also observed on the carbonaceous mask surface and are identified as laser stray lines. The full width at half-maximum (FWHM) of the sharp emission lines is down to 800 μ eV at 35 K in the present measurements and will be narrower at liquid-He temperature. Macroscopic PL (macro-PL) spectrum measured from the uniformly grown planar ZnCdS/MgS SPSL is also shown by the dashed line in

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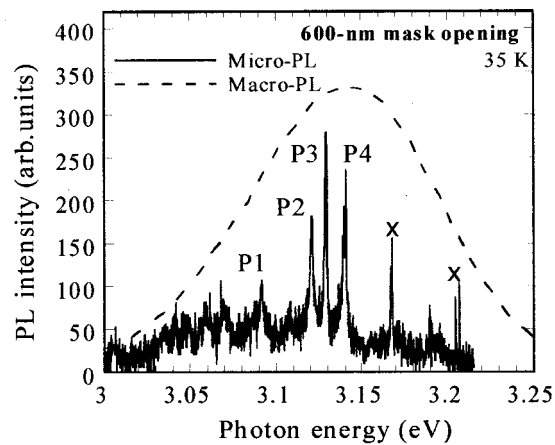


FIG. 1. Macro- (dashed line) and micro- (solid line) PL spectra of ZnCdS/MgS SPSL. In micro-PL, a single mesa structure grown on $600 \times 600 \text{ nm}^2$ mask opening was detected. Several sharp emission lines (P1–P4) are clearly observed. Additional lines labeled with crosses indicate laser stray lines.

Fig. 1. The broad spectrum with its peak energy of 3.14 eV and FWHM of 130 meV is observed, the spectrum linewidth of which is mostly determined by the compositional fluctuations in the ZnCdS alloy.¹⁴ It is noted that the energy range within which the sharp emission lines are observed is almost overlapped with that of the macro-PL spectrum. This correspondence shows that the observed sharp lines are originated from the radiative recombinations in the ZnCdS layers, where the emissions from individual localized states formed by the compositional fluctuations in the ZnCdS layers are revealed with the measurements in the restricted surface area.

Excitation power dependence of the three dominant sharp emission lines (P2–P4) was investigated, and the measured PL spectra are shown in Fig. 2(a). As a reference, the spectrum measured from the mask surface with the excitation power of $20 \mu\text{W}$ is also shown by the dotted line at the bottom of Fig. 2(a). Except for the unresolved subsidiary

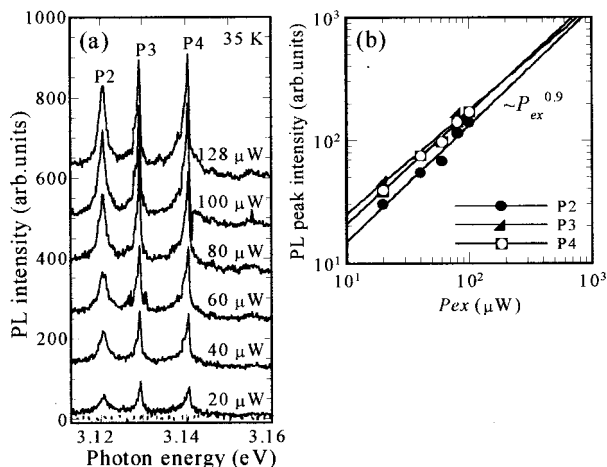


FIG. 2. Excitation power dependence of PL spectra of three dominant sharp lines (a), and peak intensities derived from the spectra (b). In (a), spectra are vertically shifted for clarity and the spectrum measured from the masked surface area with the excitation power of $20 \mu\text{W}$ is also shown by the dotted line at the bottom.

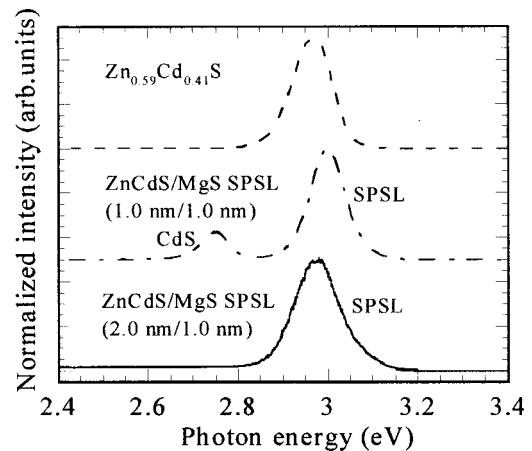


FIG. 3. Macro-PL spectra of uniformly grown $\text{Zn}_{0.59}\text{Cd}_{0.41}\text{S}$ ternary alloy (dashed line), ZnCdS (1.0 nm)/MgS(1.0 nm) (dot-dashed line) and ZnCdS(2.0 nm)/MgS(1.0 nm) SPSL structures (solid line) at 20 K. CdS layers (2 nm thick) were grown on the top of the samples for the optical probe.

sidebands, the FWHM of the sharp emission lines did not show any noticeable increase for the higher excitations. This demonstrates the quasi-zero-dimensional nature of the P2–P4 emission lines. As is clear from Fig. 2(a), the relative intensity ratio of the P2, P3, and P4 peaks remains almost constant. This shows that each emission lines are independent with each other most probably due to their spatial isolation. The excitation power dependences of the peak intensities are plotted in Fig. 2(b). It is found that all the observed sharp lines exhibit almost linear dependence to the excitation power. This demonstrates the excitonic nature of the observed sharp emission lines⁷ originated from single localized states in the ZnCdS alloy.

The sharp emission lines discussed here were only observed with the ZnCdS/MgS SPSLs selectively grown in the limited area. The PL spectra observed from ZnCdS single layers selectively grown in the similar structure showed only broad spectra. To clarify the difference, understanding of the transport and recombination processes of photoexcited carriers in ZnCdS alloys is the key issue. For this purpose, ZnCdS ternary alloys with the Cd composition range of 0.14–0.70 were prepared as uniform layers. On these sample surfaces, 10 stacks of 2-nm-thick CdS optical probing layers with 2-nm-thick ZnCdS barrier layers were grown, where the CdS layers have the lower transition energy than that of the ZnCdS alloys and will function as an optical probe of photoexcited carriers. As an example of such measurements, the PL spectrum of the ZnCdS ternary alloy with the Cd composition of 0.41 is shown in Fig. 3 by the dashed line. The peak at around 2.97 eV is the contribution from the ZnCdS alloys, but the luminescence from the CdS probing layers was almost absent. It is noted that the additional luminescence from the CdS probing layers is observable when the Cd compositions in the ZnCdS alloys is less than 0.23. Statistical theory¹³ predicts that the localization effect due to alloy potential fluctuations in ZnCdS shows maximum around the Cd composition of 0.32 and is steeply reduced toward the ZnS binary limit. Therefore, the observed Cd dependence will be reasonably interpreted with the change of the migration length in the ZnCdS alloys.

To study the role of the SPSLs for the migration of photoexcited carriers, similar 2-nm-thick single CdS probing layer was grown on top of the uniformly grown ZnCdS/MgS SPSL samples. The macro-PL spectrum measured on the SPSL in which both the ZnCdS wells and the MgS barriers were 1 nm thick is shown by the dot-dashed line in Fig. 3. In this sample, the luminescence from the surface CdS probing layer at around 2.75 eV was clearly observed in addition to the emission from the SPSL at around 3.0 eV. The observation of the clear luminescence peak from the CdS probing layer, in spite of the single layer rather than the 10 stacks of layers discussed earlier, demonstrates the distinguished migration enhancement of the photoexcited carriers in this ZnCdS/MgS SPSL.

However, when the ZnCdS well layer thickness is increased from 1 to 2 nm in the SPSL, the luminescence from the probing layer was quenched as shown by the solid line in Fig. 3. The observed well thickness dependence was examined with a simple Kronig–Penny-type model calculation and the miniband width in the SPSL was found to be critically dependent on the well width. Most of the materials parameters for this calculation are given in Ref. 15. It was negligibly small with the 2-nm-thick ZnCdS well layers, whereas it amounts to 10 meV for the SPSL with 1-nm-thick ZnCdS well layers. Tunneling time corresponding to the 10-meV miniband width is estimated to be on the subpicosecond range,¹⁶ which is much faster than the previously reported exciton radiative recombination lifetime of ~ 310 ps.¹⁴ Under this situation, excitons will tunnel through the 1-nm-thick MgS barrier layers rather than recombine in the 1-nm-thick ZnCdS well layers.

The localized states discussed in this letter are formed by the alloy potentials which will be extended generally in “long-range.” Although the earlier discussion on the tunneling time is based on conventional SL states, the discussion similar to that given here will be applicable to the present observations, that is, the presence of the barrier layers will terminate the spatial extension of the localization potentials to 1 nm or ~ 4 monolayers and will effectively increase the coupling of the neighboring states. This will contribute to increase the tunneling rate between the states and the enhanced migration will delocalize the excitons in the SPSL. The localization of excitons observed in the 2-nm-thick ZnCdS layers will be reasonably interpreted following the earlier discussions on the tunneling rate. Since the ZnCdS/MgS SPSL samples below the total thickness of 60 nm showed very weak luminescence, the exciton migration length will be on this order and the surface recombination will dominate the recombination processes. This observation can be also interpreted in terms of a mobility edge,¹⁷ at which the exciton transfer rate and the radiative recombina-

tion rate are balanced. The introduction of the SPSL will increase the exciton transfer rate by the induced tunneling processes and enhance the delocalized nature of excitons, which will lower the mobility edge.

In the case of the ZnCdS/MgS SPSL mesa grown on the 600-nm mask opening, the top (001) plane area will be about 400×400 nm². Assuming that the exciton migration length is ~ 60 nm from the earlier discussion and that one deepest localized state contribute to the sharp emission line in the respective area with the radius of 60 nm, ~ 14 localized states will contribute to the observed sharp emission lines. This estimation gives reasonable interpretation of the observed sharp lines number of ~ 15 .

In summary, sharp and discrete exciton emission lines with well-suppressed background luminescence were clearly observed from single localized states in ZnCdS ternary alloys. ZnCdS/MgS short-period superlattices were found to be very effective to enhance the exciton migration and to concentrate into single luminescence centers within the migration distance. The possibility to emit sharp lines from alloy semiconductors will be beneficial to tune the emission lines to cavity resonance wavelengths for the achievement of semiconductor single-photon emitters.

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