Modified spontaneous emission properties of CdS quantum dots embedded in novel three-dimensional microcavities

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Abstract:

Modified spontaneous emission properties in the presence of confined photon modes inside the three-dimensional (3-D.) optical microcavities are demonstrated. Self-formed pyramidal-shaped semiconductor structures fabricated by selective-area growth technique are utilized as an optical microcavity in which discrete photon modes are generated. Noticeable modification of spontaneous emission from active layers embedded in microcavity structures is clearly observed in μ-PL spectra at room temperature. Almost perfect coincidence between enhanced photoluminescence peak wavelengths and the resonance modes inside microcavity is observed. Furthermore, Purcell factors calculated from the obtained Q values reach ~9, which is inaccessible in the planar microcavities with only one-dimensional photon confinement normal to the layers. These results indicate that the effective coupling between electronic system and 3-D. confined optical fields is realized by the achievement of present low-loss 3-D. microcavities with small cavity volume V_c whose dimension is comparable to λ^3.

Keywords:

three-dimensional microcavity, spontaneous emission, photonic dot, ZnS

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1. Introduction

Spatial confinement of photon modes and their coupling to electronic systems enable us to control a spontaneous emission rate artificially and will introduce a new degree of freedom in electron-photon interaction [1,2]. A number of attempts to give the efficient confinement of radiation field have been made towards the realization of practical devices with attractive functions such as a thresholdless laser [3,4] or a single photon source [5] as well as a strong interest of basic physics. Up to now, most studies are based on planar microcavities with a pair of distributed Bragg reflectors (DBR)[6,7]. In these structures, the photonic confinement is only one-dimensional normal to the DBRs, thus the predicted enhancement factor of spontaneous emission rate is limited to below 3 [8]. In order to realize further enhancement of this value, fabrication of three-dimensional (3-D.) optical microcavity structures is an essential issue. For an ideally coupled modes, which is spatially and spectrally overlapped between atomic states and photonic modes, the enhancement factor of the spontaneous emission rate in the optical cavities is given by the Purcell factor $F_p = (3/4\pi^2)(Q\lambda^3/V_c)$[2], where Q is the mode's quality factor, λ is the wavelength in the material, and V_c is the volume of the cavity. Up to now, some structures for three-dimensional optical confinement are proposed. For example, pillar structures are among them [9-12]. However, these structures require rather elaborate post–growth processes such as dry etching to diminish the lateral size of semiconductor planar microcavities. In these structures, enhancement [9-12] and inhibition [12] of spontaneous emission has been reported. In terms of the future device applications, on the other hand, mechanical structural stability of these structures will be questionable. While microparticles and microdisks [13-17] have generally high Q values up to

17,000 [17], their cavity volumes are much larger than the respective optical wavelengths to keep the condition for the total internal reflections, and the resultant enhancement factor of the spontaneous emission rates tends to be low due to their large cavity volumes.

Recently, we have fabricated self-organized ZnS pyramidal structures on the window areas of carbonaceous masks using low-temperature selective-area growth technique[18] as shown in Fig. 1. In the reflectance spectrum of these structures, the resonance peaks corresponding to the photon modes inside the pyramidal structures showed up clearly, which indicates that the fabricated pyramidal structures function as 3-D. microcavities with high Q value of ~4900 [19]. It is also confirmed that the pyramidal-size dependence of the observed resonance wavelength was well explained by the approximate calculation of the resonance modes [19]. In order to study an electron-photon interaction, however, introducing the electronic system that will interact with photon modes inside the cavity is essential. In this paper, active layers are embedded in ZnS microcavity structures and their luminescence properties were investigated towards the future fabrication of novel light-emitting devices using artificially controlled electron-field interaction. Modified spontaneous emission properties in the presence of 3-D. confined radiation field will be clearly demonstrated, which becomes possible by the achievement of present low-loss, novel 3-D. microcavities with small cavity volume V_c of which dimension is comparable to λ^3.

2. Experimental

Three-dimensional ZnS optical microcavities have been selectively grown using metalorganic molecular-beam epitaxy (MOMBE) method on DBR layers based on a ZnSe/MgS superlattices (SLs). The

DBR structure was grown by metalorganic vapor-phase epitaxy (MOVPE) and was consisted from 5-pairs of ZnSe and ZnSe/MgS SLs quarter-wavelength layers. The SL structure was formed with about 13 periods of ZnSe (9.5Å) / MgS (32 Å) alternating layers. It gives the quite large refractive index difference of 0.55 in the blue-green wavelength region. The highest reflectivity of the DBR was about 92% at the center wavelength of 510 nm, which was in close agreement with the theoretical calculation [20]. The further details of the fabrication and characterization of the DBRs are discussed elsewhere [21]. Carbonaceous masks were used for the selective-area growth, which were patterned using scanning electron microscope (SEM) on the DBR surface. 15 kV acceleration voltage and 6-8 pA beam current were used for the electron beam irradiation. The window size in the carbonaceous mask to grow the pyramids was 900 nm x 900 nm of which sides were parallel to [100] and [010] directions. This alignment minimizes an anisotropy of the grown structures [22]. Precursors used to grow selective-area ZnS pyramids were diethyl zinc (DEZn) and ditertiarybutyl sulfide (DtBS). CdS thin layers were grown as active layers and were embedded inside the ZnS pyramidal structures. Dimethyl cadmium (DMCd) was also used for this purpose. The CdS/ZnS structures were growth at 350ºC to 380ºC. Structural properties of the pyramidal structures were characterized by an atomic-force microscope (AFM), and optical properties of these structures were characterized by both μ-reflection measurements using a Halogen lamp and μ-photoluminescence (μ-PL) measurements using the 325 nm-line of He-Cd laser. The incident light has been arranged to focus on one of the pyramids by an objective lens with x 80 magnification. After the identification of the pyramidal image and the luminescence spot observation, the area was limited to just one pyramid using an orifice. Then the

signals from a single microcavity were introduced into a optical fiber coupled with a 50-cm monochromator equipped with the liquid nitrogen cooled multi-channel CCD detector. All the measurements were performed at room temperature throuout this work and the results are discussed in detail.

3. Results and discussion

AFM observations revealed that the grown ZnS pyramidal structures had approximately 340 nm height and were surrounded by four equivalent smooth {034} facets, which form 37º to the (001) plane. Root-mean-square (RMS) value of the {034} side facets of the pyramid is about 1.8 nm and is much smoother than those of etched sidewalls. This smooth {034} ZnS facet will contribute to keep the optical scattering losses at the air-semiconductor interfaces minimum. Since the ZnS pyramidal-shaped structures were proved to function as the 3-D. microcavities or as the photonic dots in our previous work [19], the modulation of the spontaneous emission properties within the pyramidal 3-D. microcavities was studied. For this purpose, CdS layers were embedded as active layers inside the ZnS pyramidal structures and underlying DBRs were prepared for the effective optical confinement at the boundary between ZnS pyramidal structures and the substrates. In this photonic dot sructure, three 2 nm-thick CdS active layers separated with 6 nm-thick ZnS barrier layers were placed at 240 nm in height from the base plane of the ZnS pyramidal-shaped microcavity. Figure 2 shows the PL spectrum of a single CdS/ZnS photonic dot and for comparison, a spectrum from non-patterned area where the only CdS/ZnS uniform layers are also plotted. Distinguished modification of spontaneous emisson at around 538.5 nm and 539.5 nm are unambiguously observed from the microcavity area in contrast to the non-patterned area where the only broad luminescence

from the underlying DBR layer was dominated. Because the bandgap energy of CdS is 2.36 eV at room temperature [23], the origin of these luminescence might not be intrinsic. It is interpreted in this case that CdS-related luminescence, possiblely deep-level emission of CdS layers is effectively coupled to the discrete photon modes and then enhanced inside the ZnS microcavity. For the further elucidation of the origin of this modification, PL spectrum was compared with photon resonance modes revealed by the μ-reflection measurements and is shown in Fig. 3. In μ-reflection spectrum, high Q values as large as 1000 are shown. It should be noted that PL peak wavelengths are almost perfectly coincided with the resonance modes inside the microcavity. This result indicate that only the electronic transitions whose frequencies are coincident with the resonant modes in the cavity are allowed to give luminescence, otherwise the spontaneous emission is strongly suppresed. Furthermore, Purcell factors calculated from the obtained Q values reach ~9, which is inaccessible in the planar microcavities with only one-dimensional photon confinement. These results indicate that the spontaneous emission rate is effectively alterd by the electron-photon interaction, in which the emitter is spatially and spectrally coupled to the discrete radiation field in the presence of three-demensionally confined spaces. In order to realize the higher coupling efficiency of the electron-photon interaction, spatial overlap between CdS active layers and the anti-node position of the electric field is essential. In this sense, we are now preparing the numerical calculations with FDTD method in order to understand the modal distribution inside the pyramid. Apart from the coupling efficiency between electric system and radiation field, however, observed modification properties of the spontaneous emission inside the ZnS pyramidal-shaped microcavity are not affected essentially by the positioning of active layers

inside the cavity. These results are believed to be very encouraging to the future achievements of practical devices utilizing controlled electron-field interaction.

4. Conclusion

In conclusion, modified spontaneous emission properties of pyramidal-shaped CdS/ZnS photonic dot structures were studied. The clear modification properties of the spontaneous emission at room temperature was observed from the ZnS pyramidal-shaped structure embedded with the CdS active layers. These results show that the spontaneous emission is efficiently modulated by the discrete photon modes inside the cavity. It can be shown that the present novel pyramidal photonic dot structures are promising system in terms of the possibility of the strong modification of electron-photon interaction due to high quality factor and small cavity volume. This work was partly supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture.

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Figure captions

Fig. 1: (a) Atomic force microscope image of a selectively grown ZnS pyramidal-shaped 3-D. microcavity structure. (b) The schematic drawing of (a). Selective growth was clearly observed on 700-nm square opening of carbonaceous mask. Four pyramidal sidewalls are composed of atomically flat {034} facets. In this paper, optical characterizations are performed of the samples with CdS active layers embedded in this structure, in which the intermediate ZnS layers are replaced by three 2 nm-thick CdS active layers separated with 6 nm-thick ZnS barrier layers.

Fig. 2: μ-PL spectra of both 3-D. microcavity area (thick curve) and non-patterned area (thin curve) measured at room temperature. Clear resonant structure at around 539 nm is observed by proving 3-D. microcavity area. On the other hand, only featureless luminescence from underlying DBR layer was observed from non-patterned area.

Fig. 3: μ-PL (upper curve) and μ-reflectance (lower curve) spectra obtained from 3-D. microcavity area. PL peak positions are well coincident with photon resonant modes with high Q values ~1000. Calculated Purcell factor F_p reaches ~9, which indicates the effective coupling between electronic system and 3-D. confined radiation field.

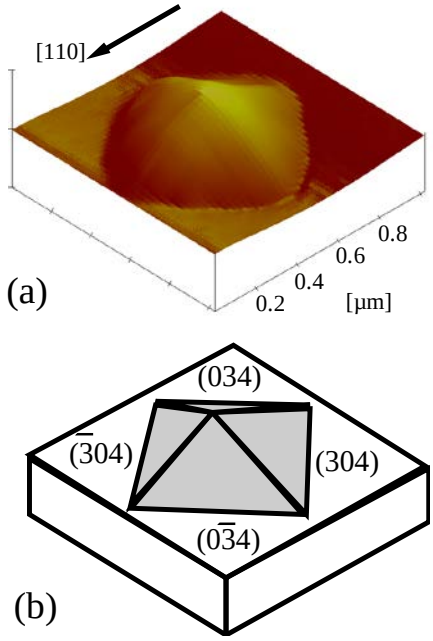

Fig. 1

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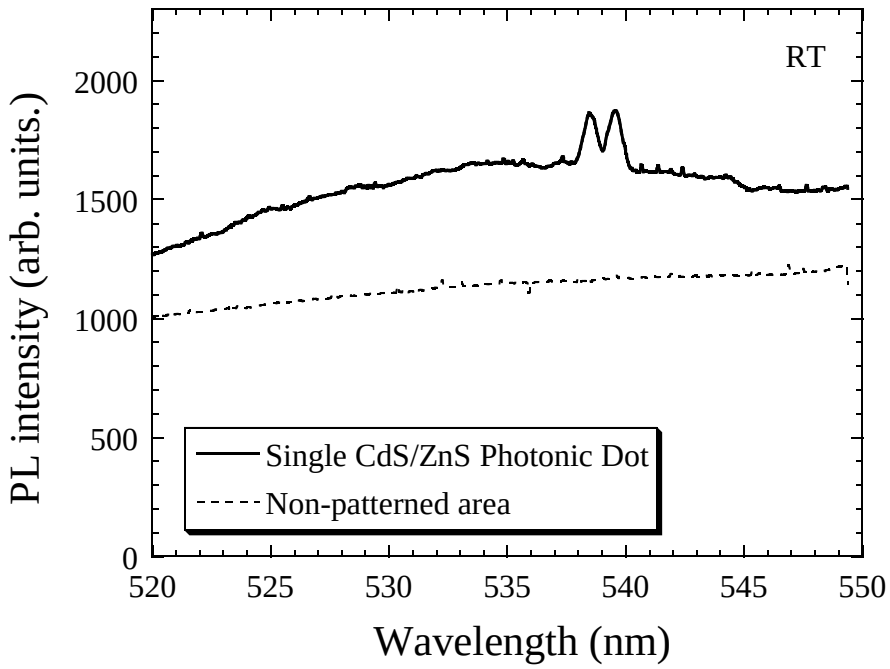

Fig. 2

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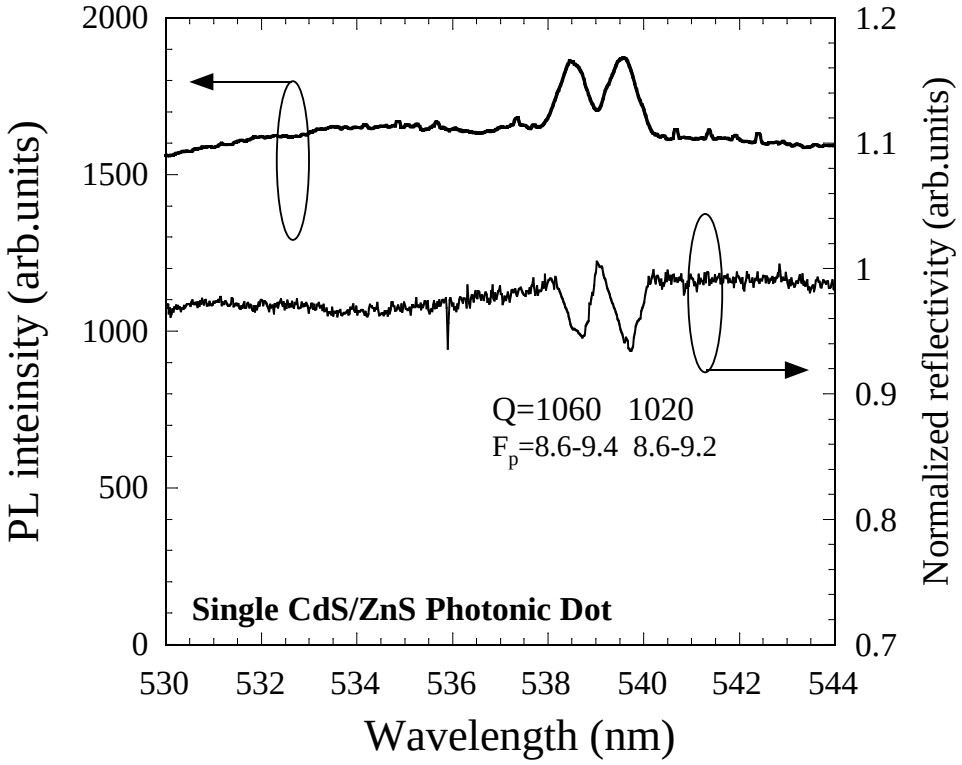

Fig. 3

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