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Lasing of $CdSe_xS_{1-x}$ quantum dots in a glass spherical microcavity

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Abstract

A glass spherical microcavity only a few microns in diameter embedded with $CdSe_xS_{1-x}$ quantum dots (QDs) was fabricated using a physical method; it exhibited good optical stability under continuous-wave laser excitation with high power. We investigated the excitation power dependences of the emission intensity and the linewidth of both transverse electric and transverse magnetic resonance peaks of whispering gallery modes. Stimulated emission behaviour of multi-frequency modes is observed at room temperature. The low threshold value and large mode separation makes QD-containing microspheres promising for visible microlaser applications.

Spherical microcavities are attractive systems for both fundamental physics research in the field of cavity quantum electrodynamics (CQED) and potential applications in optoelectronics. They have good symmetry characteristics due to the confinement in three dimensions. A number of discrete, sharp, spectrally well-separated resonance modes, the so-called whispering gallery modes (WGMs), exist in such spherical microcavities [1, 6]. Several research groups have investigated such optical systems, and different methods have been used to fabricate microspheres to realize WGM and optical microdevices, e.g., for spherical microresonators made from bulk semiconductor spheres of CuCl [2], and glass spheres doped with rare-earth ions or covered with nanocrystals of single molecules on the surface [3–5]. However, these spherical microcavities do not allow one to vary the emission energy of the embedded species. Only the size of the microsphere can be changed [6]. To the best of our knowledge, work on glass spherical microcavities embedded with semiconductor quantum dots (QDs) which allow one to tune both the modes of the microcavity and the luminescence centres in the cavity is scarce. If the QDs are embedded into a spherical microcavity, the emission spectrum of the QD

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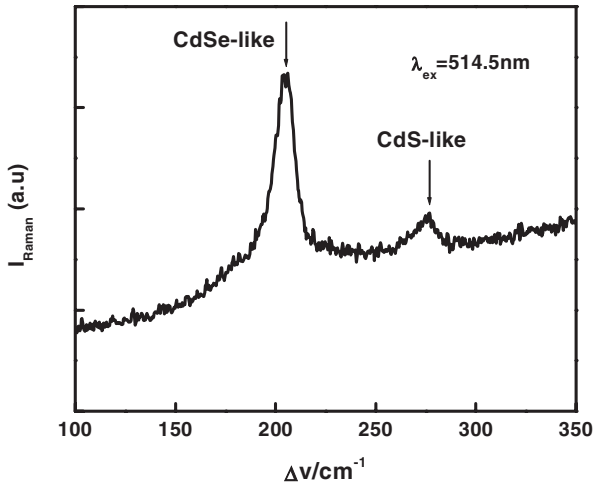

Figure 1. The Raman spectrum of a glass microsphere measured using the T64000 Laser Raman system with a typical spectral resolution of 1 cm^{-1}. λ_{ex} represents the excitation laser line at 514.5 nm.

ensemble drastically changes into a discrete number of sharp modes due to the coupling with the WGMs. Artemyev and Woggon [6–8] prepared spherical microcavities embedded with QDs by using a chemical method. Recently, we have fabricated $CdSe_xS_{1-x}$ QDs in glass microspheres, using a physical method [9, 10], which have better optical stability than the polymer microspheres as the latter are often degraded under strong illumination due to the photosensitive surface reactions. The strong coupling between photonic and electronic states makes it possible to realize stimulated emission behaviour in the case of low threshold. However, the challenge for this method is to fabricate perfect microspheres with small enough diameters to reduce the threshold. In addition, the detailed theory of stimulated emission processes in the optical microsphere is still not well established [8, 14, 15]. Hence, many research groups have concentrated on research into stimulated emission and its potential utilization.

In this work, we have fabricated perfect microspheres only a few microns in diameter. Very strong and sharp resonance modes were apparent in the emission spectrum. We investigated the excitation power dependence of the emission intensity of the resonance modes of $CdSe_xS_{1-x}$ QDs in a glass spherical microcavity. With the increase of the excitation intensity, the intensity of the resonance emission modes increases and the linewidth becomes narrower. From a double-logarithmic plot of the excitation power dependence of the emission intensity of the resonance modes, we found that stimulated emission behaviour of the multi-frequency resonance modes in the lower-energy spectral range of QD luminescence can be achieved at room temperature.

The method of preparation of optical microcavities containing semiconductor QDs was similar to that described in the literature [9] except that a H–O gas burner was employed to replace compressed air to soften the glass quickly. The main processing technique is based on melting Cd-, Se-, and S-ion-containing glass particles using a gas burner and then allowing cooling. In this process, the glass particles formed glass microspheres due to the surface tension. The glass microspheres obtained were annealed at 600 °C for 12 h to form $CdSe_xS_{1-x}$ QDs. The Raman scattering spectrum of a sample was measured using the T64000 Laser Raman system with a typical spectral resolution of 1 cm^{-1}, as shown in figure 1. The excitation laser line is 514.5 nm. Figure 1 shows the strong Raman signals for both CdSe-

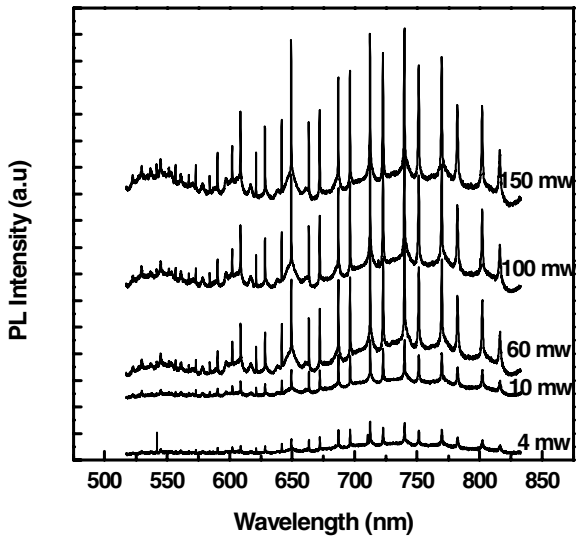

Figure 2. PL spectra of a single spherical microcavity embedded with $CdSe_xS_{1-x}$ QDs under different excitation powers, pumped with the excitation line at 488 nm.

like longitudinal optical (LO) phonon and CdS-like LO phonon modes, located at 208 and 280 cm^{-1}, respectively [11]. It confirms that the $CdSe_xS_{1-x}$ alloy nanocrystals were formed by the annealing process.

Figure 2 shows the photoluminescence (PL) spectra of a single microsphere measured with different excitation powers. The PL spectra were measured using the Dilor Super Labram MicroRaman system with an excitation wavelength of 488 nm. The spatial resolution of <2 μm was achieved using a microscope with a $100\times$ objective lens to ensure that the measurement was on a single microsphere. The PL spectra on the higher-energy side and the lower-energy side are assigned to excitonic transitions of QDs and surface-defect-state-related luminescence, respectively [6, 9]. It is found that a number of very sharp and clearly distinct resonance modes were superimposed on the background broad PL band of the QDs—the so-called WGMs. With increasing excitation power, the wavelength location of the resonance modes is unchanged, which is a sign that there is no photo-induced modulation of the refractive index [12]. The refractive index of the spherical microcavity does not change even with continuous exposure to high excitation power, which implies that the glass spheres resist degradation under strong excitation, showing good optical stability which would be beneficial for fabricating microspherical lasers.

Figure 3 shows the PL spectrum on the lower-energy side for the spherical microcavity, obtained by subtracting the Gaussian background QD emission, at an excitation power of 120 mW. As can be seen from the figure, there are sharp lines with large mode spacing and fewer modes, indicating that the spherical microcavity is small enough in diameter. From the image in the CCD camera of the Super Labram system, we can observe that the diameter of the spherical microcavity is about 4.7 μm. In addition, using Mie scattering theory [13], we can also deduce the size of the microsphere from the mode separation. For the transverse electric (TE_l^n) or transverse magnetic (TM_l^n) field modes, the separation between the adjacent peaks of the WGMs with the same angular quantum number n and adjacent radial quantum number l, $\Delta\lambda$, can be expressed as

$$\Delta\lambda = \frac{\lambda^2 \tan^{-1}\{(n_1/n_2)^2 - 1\}^{1/2}}{\pi n_2 d\{(n_1/n_2)^2 - 1\}^{1/2}}$$

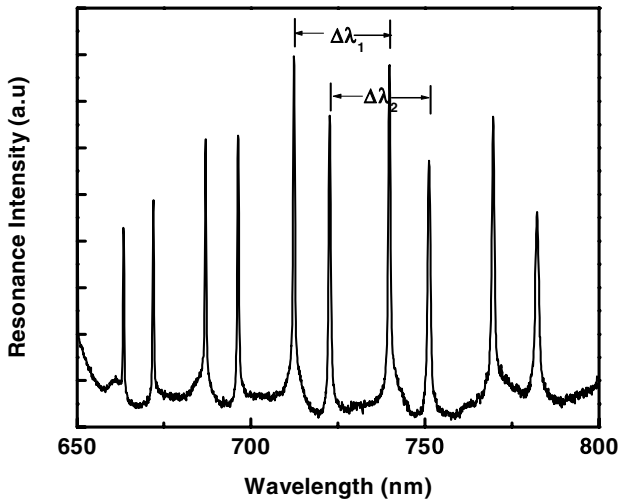

Figure 3. The emission spectrum obtained by subtracting the Gaussian background emission line of the PL of the QDs in the spherical microcavity.

where λ and d are the emission wavelength and the diameter of the spherical microcavity; n_1 and n_2 are the refractive indices of the sphere ($n_1 \approx 1.523$) and the surrounding air ($n_2 \approx 1.0$), respectively. In figure 3 two series of periodic resonance peaks with nearly the same mode separation are observed. They form paired sharp lines in the spectrum. According to the polarization analysis, the right-hand mode of each pair is the TM mode. In figure 3, $\Delta\lambda_1 = 26$ nm, which represents the separation between adjacent TE modes, is the same as $\Delta\lambda_2$, the separation between the adjacent TM modes. In our calculation, at $\lambda = 725$ nm, the diameter of the sphere can be deduced to be about 4.63 μm, which is in excellent agreement with the value from the image in the CCD camera. This demonstrates that we have obtained a spherical microcavity small enough in size; this is beneficial to achieving practical stimulated emission due to there being fewer frequency modes.

Since we have found that the $CdSe_xS_{1-x}$ QDs in the spherical microcavity have good optical gain, it looks promising to operate stimulated emission action under optical excitation. In fact, the resonance modes on the lower-energy side of the spectrum are more suitable for possible stimulated emission operation, where the light emission is induced by the surface-state-related transitions of QDs [9], as the small absorption coefficient can be helpful for reaching the high Q-values of the WGMs. On the higher-energy side of the spectrum, on the other hand, there is high intrinsic absorption of QDs, and hence large damping coefficients are expected, which may suppress the enhancement of the resonance intensity. As can be seen from figure 2, on the lower-energy side of the spectrum, the intensity of the resonance modes is strong enough to emerge from the Gaussian background emission, and the intensities of the resonance modes increase with increasing excitation power, while the background emission intensity does not follow this trend; they became saturated at high excitation power. This makes stimulated emission behaviour easily achievable even under lower excitation power. We examined the Q-values of the resonance modes and found the value to be 1162 at 769 nm and to reach 2015 at 663 nm. The Q-values were obtained by using the equation $Q = \hbar\omega/2\hbar\gamma$, where $\hbar\omega$ is the photon energy and $2\hbar\gamma$ is the Lorentzian fitting the linewidth of the cavity modes. Figure 4 shows a double-log plot of the emission mode intensities as a function of the pump energy after subtracting the Gaussian background line shape. In figure 4, solid

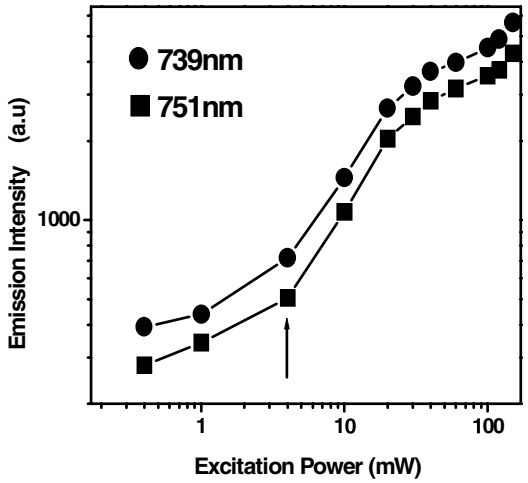

Figure 4. Excitation power dependences of the emission intensities of the 739 and 751 nm resonance modes, indicating the onset of stimulated emission at the arrowed threshold value.

circles and squares show the dependences of the resonance modes at 739 and 751 nm of the TE and TM modes, respectively. All resonance modes on the lower-energy side are examined and it is found that they have nearly the same threshold behaviour as indicated by the plot of figure 4. It is found that below the excitation power of 4 mW, the emission intensities vary slowly with increase of excitation. However, the emission intensities increase steeply beyond the excitation power of 4 mW, which indicates that the threshold is about 4 mW, corresponding to the power density of 31.2 kW cm^{-2}. The slope is not close to 1, no matter whether one looks beyond or below the threshold excitation power. In addition, from figure 4, it is noted that at a high excitation power, the intensity of the resonance modes becomes saturated and changes slowly with increasing pump power. Similar phenomena were also found in other experimental results [16, 17]. This may be attributed to the trapping effect of surface-related defect states of QDs and enhanced non-radiative recombination in glass microsphere under high excitation. More research work is needed to make further progress.

In figure 5 typical changes of linewidths for the resonance modes at 739 and 751 nm under increase of the excitation power from 0.4 to 10 mW are plotted in an expanded view. It is found that the resonance modes narrow above the threshold pump power of 4 mW, but the variation is not very pronounced. And the spectral line is asymmetric. This result may be explained by the fact that every resonance peak consists of more than one resonance mode [14]. It is noted that non-ideal spherical shape of the spheres can lift the degeneracy with respect to the azimuthal mode indices for a given angular quantum number n and radial quantum number l, giving rise to broadening and asymmetry of the resonance modes [15]. It is noted that the linewidth of the single luminescence peak of a single QD is very narrow, being 120 μeV at 10 K [18] narrower than the linewidth of the WGMs. Therefore, a WGM resonant peak will contain not the luminescence line from just one QD, but luminescence lines from many QDs with slightly different sizes due to a large number of QDs present in each single glass microsphere in our samples. When the gain factors of the neighbouring lines are comparable with each other, all of these lines are enhanced in the stimulated emission process and will be superimposed on each other in a small wavelength range. The energy separation between adjacent lines is very narrow and may exceed the spectral resolution of the spectrometer. Therefore these lines are indistinguishable. Thus every resonance peak profile is an envelope curve of several

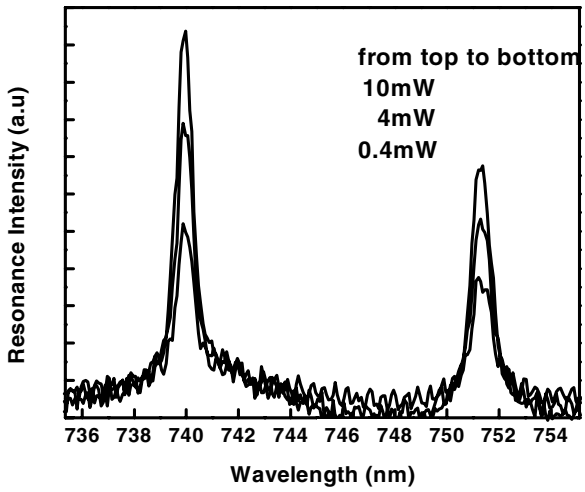

Figure 5. Typical changes of linewidths of the resonance modes at 739 and 751 nm in an expanded view with excitation power increasing from 0.4 to 10 mW.

superimposed line shapes and looks like a single resonance peak. The line shape will remain broadened or asymmetric after stimulated emission. This will lead to the linewidth of the resonance mode being not as narrow as expected in the case of a single $CdSe_xS_{1-x}$ QD embedded in a microsphere. From the above-mentioned evidence, we can confirm that these optical microcavities can realize stimulated emission at the resonance modes under relatively low pump intensity. Also, reduction in the number of modes could make this optical system more practical.

In conclusion, the intensity of the resonance emission modes of QDs in a spherical microcavity with a small diameter increases faster than the background luminescence and the linewidth becomes narrower. The excitation intensity dependence of the resonant modes is not linear on a logarithmic excitation intensity scale. The stimulated emission is realized at relatively low excitation intensity. However, the suppression effect of the neighbouring resonance modes is not clear, showing a multi-longitudinal mode behaviour. The high optical stability and low threshold of the spherical microcavity embedded with QDs makes it promising for visible microlaser applications.

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