Spherical cavity-mode laser with self-organized CuCl microspheres

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We report lasing of exciton–polaritons in semiconductor microspheres with spherical cavity modes under subpicosecond ultraviolet laser excitation at low temperature. Small bulk crystals of CuCl are melted and formed into fused microspheres with diameters ranging from a few to several tens of micrometers. The threshold for lasing is 1 nJ/pulse, which corresponds to 10^9 photons/pulse. We found that the biexciton-to-longitudinal-exciton transition is responsible for the lasing. © 1997 Optical Society of America

A dielectric sphere whose size is several times that of the optical wavelength acts as a high-Q (quality factor) optical cavity. The resonant modes are called whispering-gallery modes (WGM's) or morphology-dependent resonant modes. Such microcavity structures can be used for efficient optical devices such as low-threshold lasers and low-power nonlinear optical switching devices. Efficient lasing has been observed in liquid droplets,[1] polymer microspheres,[2] and Nd-doped glass microspheres.[3] Semiconductor WGM cavity lasers were pioneered by McCall *et al.* and Mohideen *et al.*[4] Low-threshold lasing in single-mode operation was demonstrated.[5] The small size and the high quality factor invoke phenomena that are characteristic of the granular nature of the radiation field. For example, the output noise of a microdisk laser shows peculiar features near threshold.[6] Braginsky *et al.* pointed out that microspheres made of nonlinear materials could show optical bistability at input power levels as low as that of a single photon.[7] Semiconductor micropheres have a strong potential for use as such quantum optical devices. Although some attempts have been made to fabricate semiconductor microspheres,[8] neither lasing nor optical bistability has yet been reported.

CuCl is a direct-gap semiconductor with a cubic lattice structure. The material has a stable biexciton state with 32-meV binding energy. At the biexciton two-photon resonance, $\chi^{(3)}$ is very large and the linear absorption coefficient is only of the order of 10 cm^{-1}. At a detuning of 0.1 meV, $\text{Re}[\chi^{(3)}]$ is 10^{-4} esu, and we can neglect induced absorption.[9] These values meet the Braginsky criterion for photonic bistability.[7] The optical gain associated with the transition between the biexciton state and the Z_3 exciton state[10] is also important. Lasing in CuCl has been reported in bulk crystals[11] and microcrystals.[12] In this Letter we present a method for fabricating microspheres of CuCl. Laser oscillation under photopumping is also demonstrated.

High-purity zone-refined polycrystalline CuCl ingots and a fused-quartz plate were inserted in a Pyrex tube with 200-Torr argon gas. We heated the tube above the CuCl melting point of 422 °C and then slowly cooled it. The temperature of the substrate was lower than that of the rest of the sample. Small drops of liquid-phase CuCl appeared upon the substrate. During the cooling process the drops froze into solid microspheres. To release the strain, we cooled the tube to room temperature at a slow rate of 100 °C/h.

We examined the shapes of the spheres with a scanning electron microscope (SEM) from the top and from the side. We observed various sizes of spheres with diameters of less than 40 μm upon the substrate. Figure 1 shows an example of a front-view SEM image of CuCl microspheres. From side-view SEM images we found that the microspheres are not perfectly spherical but rather ellipsoidal. The contact angle was ~145°. On the surfaces of the microspheres, all the imperfections were smaller than 100 nm.

To examine the quality of the polycrystalline samples we measured the photoluminescence at low temperature (2 K) by using the second harmonic of the pulses from a cw mode-locked Ti:sapphire. Figure 2 shows the spectra for a single crystal and for a fused

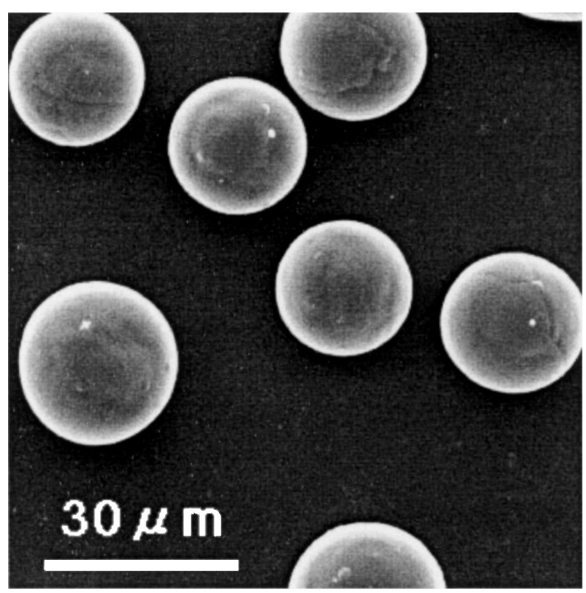

Fig. 1. SEM image of typical self-organized CuCl spheres. These spheres are ~20 μm in diameter.

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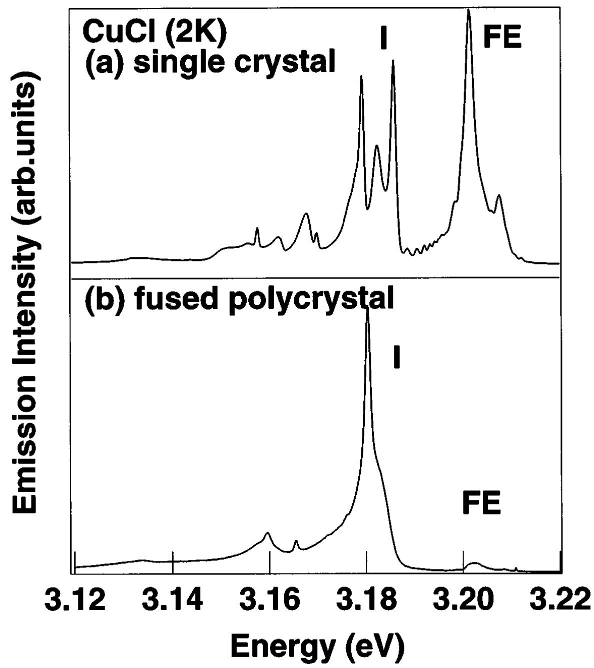

Fig. 2. Luminescence spectra of CuCl crystals at 2K under weak excitation: (a) a single crystal grown by vapor phase transport, (b) fused polycrystalline sample made by heat treatment as described in the text.

polycrystalline sample. Emissions associated with free excitons (FE's) and bound excitons (I's) were observed for both samples. Although the linewidth of the I line of the polycrystalline sample was slightly broader, no emission associated with the donor–acceptor pair recombination was observed, which implies that the sample quality was not degraded by the heat treatment.

Subpicosecond pulses of a Ti:sapphire oscillator amplified by a regenerative amplifier with 1-kHz repetition rate were frequency doubled by a β-BaB_2O_4 crystal. The pulse duration for the generated blue light was ~300 fs FWHM. We set the center wavelength to 383 nm, which is just above the Z_3 exciton resonance. As the absorption coefficient at this wavelength is $\sim 10^3$ cm^{-1}, we could excite the entire volume of the spheres. The substrate with the microspheres was mounted upon a cold finger of a conduction-type helium cryostat (Oxford Microstat) designed for low-temperature microscopy. The pump beam was focused onto one sphere with a spot size of 20-μm diameter by a microscope (Nikon EPI Plan 10×) objective with a superlong working distance. The objective also collected the emission from the individual sphere. We placed the expanded image of the sphere onto an iris. By monitoring the image with a charge-coupled device (CCD) camera, we positioned the iris and adjusted the hole size to select the emission from a particular part of the sphere. This part of the emission was directed onto a spectrometer (focal length, 50 cm) with a liquid-nitrogen-cooled CCD optical multichannel detector.

Figure 3 shows the emission spectra of a CuCl microsphere under various excitation power levels. At the excitation level of 0.8 mJ/cm^2 [curve (a)], two emission bands appeared; one corresponds to the transition from the biexciton to the longitudinal exciton (band M_L at 3.163 eV); the other corresponds to the transition from the biexciton to the transverse exciton (band M_T at 3.168 eV). The resonance frequencies of these bands match those of single crystals. When we increased the excitation level to 1.2 mJ/cm^2, two new emission lines grew nonlinearly [curve (b)]. At the higher excitation levels of 4 mJ/cm^2 two more emission peaks appeared [curve (c)]. All these emission lines were in the energy range of band M_L. Figure 4 shows plots of the emission peak intensities as a function of the pump pulse energy. At low-density excitation the intensity of the line at 3.163 eV is proportional to the square of the excitation pulse energy. At low excitation the biexcitons are created by the fusion of two excitons and emit photons spontaneously. Thus the emissions show a square dependence. Above the power level of ~1 mJ/cm^2, which corresponds to the injection of $\sim 10^9$ photons into the sphere, the emission showed salient growth. The emission line at 3.156 eV is linear to the excitation power in the weak-excitation region because the emission at 3.156 eV is ascribed to the spontaneous emission of bound excitons. Above an excitation of 2 mJ/cm^2 the line also rises. At high density, hot biexcitons with large wave vectors are created and emit photons with lower energy. As a result, the lasing occurs at a lower energy.

Above the lasing threshold level the rim of the sphere becomes brighter than the center and the sharp lines are pronounced in the spectra from the emission at the rim. These features indicate that lasing occurs with the WGM's in the CuCl microspheres.

We can estimate the threshold value from the linear loss and gain coefficient. We can evaluate the linear loss from the Q value. From the line widths of WGM

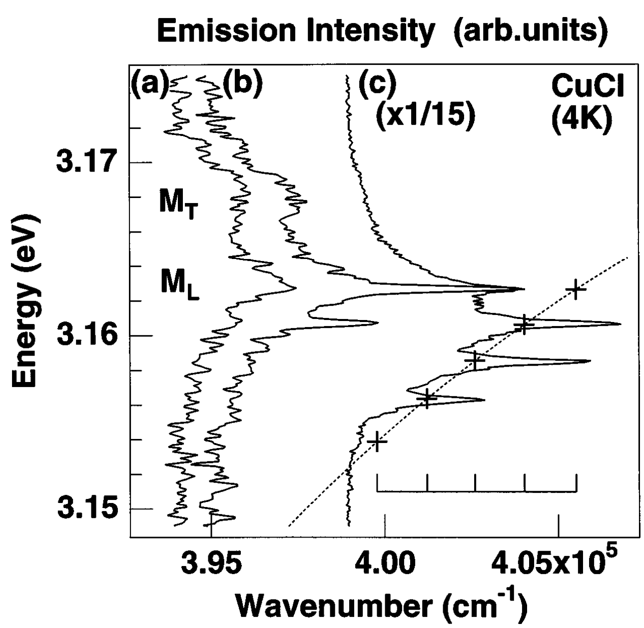

Fig. 3. Emission spectra of a CuCl microsphere with a diameter of 10 μm for three pump levels: (a) 0.8 mJ/cm^2, (b) 1.2 mJ/cm^2, (c) 4.1 mJ/cm^2. The dashed curve is a calculated curve of polariton dispersion with the parameters of bulk CuCl.

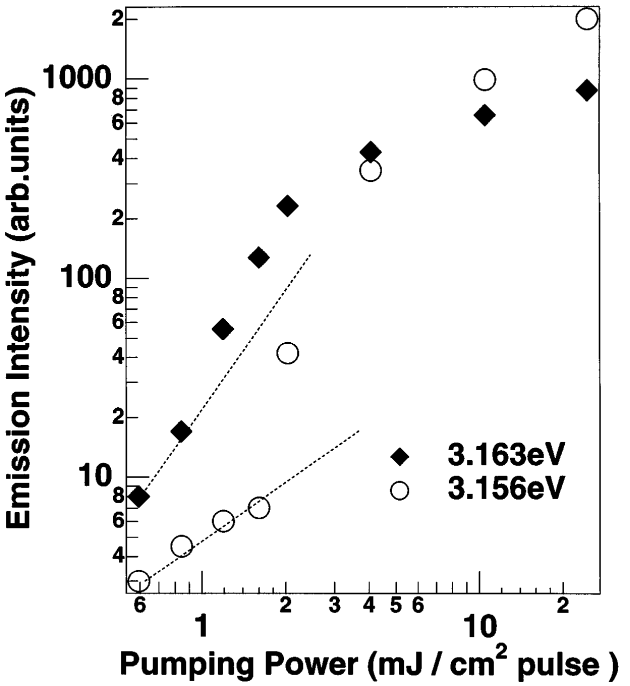

Fig. 4. Pump power dependence of the emission intensities of the lines at 3.156 and 3.163 eV. Dashed lines show linear and square dependence.

emission lines just below the threshold, the Q value of the spheres is 5×10^4, which corresponds to the linear loss of 10 cm^{-1}. The observed Q value is limited by the light scattering at the surface. To obtain the gain value at the M_L band we performed pump–probe spectroscopy, using a bulk single crystal under similar excitation conditions. The gain coefficient was ~200 cm^{-1} for excitation of 0.4 mJ/cm^2. The threshold value estimated from these parameters is 0.1 mJ/cm^2. The observed threshold level is 10 times higher than this value because of the nonuniform biexciton distribution in the sphere.

In Fig. 3 the mode separation between adjacent modes is narrower on the high-energy side. This feature can be explained by the exciton–polariton dispersion of CuCl. The dashed curve in Fig. 3 shows a calculated polariton dispersion curve with the parameters of bulk CuCl.[13] On this curve the WGM resonances at the pump power of 4 mJ/cm^2 are indicated. The wave numbers of the resonance modes align at an even spacing of $\Delta k = 1450\ cm^{-1}$, which implies that all these lasing modes have the same order number. From the value of Δk we can estimate the diameter of the sphere. The obtained value is 14 μm. From the image on the CCD camera we find that the size of the sphere is ~10 μm in diameter. Modification of the WGM resonance by the imperfect spherical shapes or the change in the polariton parameters in polycrystals could be the causes of the discrepancy.

In conclusion, we have fabricated CuCl microspheres by self-organization. Under photoexcitation, biexcitonic lasing is clearly observed in the WGM's of the spherical cavity by exciton–polaritons.

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