Two-photon pumped lasing in a single CdS microwire

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We report the two-photon absorption induced lasing performance in a single CdS microwire under the excitation of a femtosecond pulse laser with a wavelength of 800 nm. Sharp lasing peaks are centered at 523 nm with an average linewidth of 0.4 nm, indicating a high quality factor of 1300. The lasing emission is highly dependent on the polarization direction of the excitation light and the optimized lasing threshold is estimated to be 3.3 mJ/cm² as the polarization direction is parallel to the microwire axis. Furthermore, the polarization-dependent lasing effect is confirmed by performing the finite-difference time-domain method.

Micro/nanolasing based on one-dimensional semiconductor micro/nanostructures has attracted much interest due to its great potentials for developing the miniaturized optoelectronic components. CdS is an excellent II-VI semiconductor material with a direct band gap of 2.42 eV at room temperature, which is a good candidate for optoelectronic applications in the visible spectrum range. The stimulated emission and lasing have been observed in CdS nanowires and nanoribbons, indicating that one-dimensional CdS structures can be used as promising micro/nanolasers for integrated photonics. Generally, the lasing mechanisms in the semiconductor microwire are attributed to the whispering-gallery mode (WGM) and Fabry-Pérot (F-P) cavity. For CdS wire structure, it is reported that the lasing mechanism is almost all assigned to F-P resonance, where two flat end facets serve as two reflecting mirrors. From the aspect of intrinsic physical processes, there are mainly three effects for laser actions: exciton-exciton (ex-ex) scattering, exciton-longitudinal optic (ex-LO) phonon scattering, and electron-hole plasma (EHP).

Previous efforts about CdS laser mostly focus on the lasing effect under one-photon excitation (OPE). Compared with OPE, the two-photon excitation (TPE), where the excitation photon energy below the band gap of CdS, can be achieved through the simultaneous absorption of two photons. The advantages of TPE are that it can provide a larger penetration depth, which reduces the nonradiative recombination due to surface defects; TPE produces the frequency upconverted emission and offers three-dimensional resolution for imaging, data storage, and microfabrication. In addition, the near-infrared excitation source is needed for TPE, which is more suitable for optical communications and biological/medical applications.

Furthermore, TPE based on the two-photon absorption (TPA) process is highly dependent on the polarization of the incident laser due to the optical anisotropy in an individual micro/nanowire, leading to the polarization-dependent TPE efficiency. Therefore, it gives us an impetus to study the polarization-related two-photon pumped lasing properties in a single CdS microwire.

In this letter, we report the two-photon pumped lasing in an individual CdS microwire at room temperature under a femtosecond laser with a wavelength of 800 nm. The emission signal is collected from the end facet of the microwire by a fiber and the green emission is obtained around 523 nm. The dark-field microscope images visually present the spontaneous emission (SE), amplified SE (ASE), and laser oscillation as the excitation energy densities increase. Further study shows that the lasing emission is highly dependent on the polarization direction of the excitation light and the optimized threshold (3.3 mJ/cm²) is obtained as the excitation polarization is parallel to the microwire axis, which shows a decrease than that of perpendicular case (4.8 mJ/cm²). The finite-difference time-domain (FDTD) simulations agree with experiment results well.

CdS microwires were synthesized in a horizontal tube furnace via the chemical vapor deposition method. Figure 1(a) shows the conventional confocal microscope configuration. A tungsten halogen lamp with a continuous spectrum (3200 K) was used as testing and illuminating source. The femtosecond laser system consisted of a mode-locked Ti/sapphire oscillator (Spitfire, Spectra-Physics, 800 nm, 50 fs, 1 kHz) was introduced as the linearly polarized light excitation source. For linear optical measurement, the beams were focused and re-collimated by a pair of objectives (40×, Olympus). For lasing measurement, a lens with a focus length of 30 cm was used as a condenser to generate the focused spot with a diameter of ~100 μm. The laser emission from the end facet of the sample was first collected by a fiber and then coupled to a monochromator equipped with a photomultiplier and a lock-in amplifier (SR830, SRS). The polarization angle with respect to the long-axis, θ, was defined, which was controlled by rotating a half-waveplate at 800 nm. The single CdS microwire with a diameter around 1 μm and length of ~28 μm is shown in Fig. 1(b), which is further used for the upconverted lasing study. Figure 1(c) shows the micro-absorption spectrum of the sample, which indicates the absorption edge around 511 nm and the
bandgap of 2.42 eV. Therefore, under the excitation of 800 nm ($E_p = 1.55$ eV), the simultaneous absorption of two-photons occurs. The very weak peaks located at 650 nm and 730 nm are attributed to defect-related absorption, indicating the high crystal quality and low defects concentrations inside the microwire. It has been reported that high defects concentrations can lead to excitons annihilation and suppress the photoluminescence intensity.24 Because of the very low defects in our sample, the defect effect on lasing action can be neglected in our experiment.

Figure 2 shows the emission spectra of the single CdS microwire at different excitation energy densities. At a relatively low excitation density of 2.7 mJ/cm$^2$, the spectrum exhibits a broad SE band centered at 523 nm with a full width at half maximum (FWHM) of $\sim$14 nm. Then some weak shoulders gradually appear on the broad band at 3.3 mJ/cm$^2$. When the energy density reaches to 3.4 mJ/cm$^2$, some discrete sharp peaks emerge on the SE band with a dramatic decrease of the emission bandwidth ($<1$ nm), indicating the outset of lasing action. Only some of resonant modes are initially presented in the output emission spectrum due to the gain-induced mode selection process near the threshold ($3.4$ mJ/cm$^2$). However, both the emission intensity and the number of lasing modes rapidly increase as the energy densities increase ($4.0$ mJ/cm$^2$), indicating the post-threshold behavior of the lasing emission in a waveguide resonant cavity. Because the lasing is obtained at room temperature and no obvious red-shift of the spectrum is observed as the pumping energy increases, the lasing mechanism can be attributed to the ex-LO scattering in the range of the excitation intensity in our experiment.9,13 The insets of Fig. 2 exhibit dark-field images of the sample under different energy densities, corresponding to SE ($2.2$ mJ/cm$^2$), ASE ($3.3$ mJ/cm$^2$), and lasing oscillation ($4.0$ mJ/cm$^2$), respectively. Furthermore, the sharp peaks observed in Fig. 2 (red curve) can be ascribed to the longitudinal F-P modes. The mode quality factor, $Q$, is defined as $Q = \lambda/\Delta\lambda$, where $\lambda$ and $\Delta\lambda$ are the peak wavelength and FWHM, respectively. In our experiment, the average mode spacing and linewidth of the laser modes are about 0.85 nm and 0.4 nm, respectively. Thus, the $Q$ factor is estimated to be 1300 at the center wavelength of 523 nm. It is worth noting that the mode spacing at 3.4 mJ/cm$^2$ is larger than that at 4.0 mJ/cm$^2$. Because the microwire length ($\sim$30 $\mu$m) is comparable to the size of excitation beam spot, the inhomogeneous excitation of Gauss beam along the microwire becomes dominant as the excitation density decreases. The energy density on both ends of the microwire is obviously lower than that of the central part, leading to relatively low excitation efficiency. Thus, some modes cannot be excited due to different mode losses, resulting in the larger mode spacing at 3.4 mJ/cm$^2$.

For a F-P cavity mode, the mode spacing $\Delta\lambda_m$ and $Q$ factor can be expressed by the following equations:\n
\[ \Delta\lambda_m = \frac{\lambda^2}{2L[n - \lambda (dn/d\lambda)]}, \]
\[ Q = 2nL\pi/\lambda(1 - \sqrt{R_1R_2}), \]

where $\lambda$ is the resonance wavelength, $n$ is refraction index at wavelength $\lambda$, $L$ is the cavity length, $dn/d\lambda$ is the chromatic dispersion, and $R_1$ and $R_2$ are the reflectivities of the two facets. For CdS microwire, in normal incidence condition, it can be calculated that $R_1 = R_2 = 21\%$, according to the equation $R = (n - 1)^2/(n + 1)^2$. The deduced mode spacing and $Q$ factor according to the equations are about 0.83 and 1163, respectively, at 523 nm, which is in good agreement with the experimental values of 0.85 and 1300.

Since the bandgap ($E_g = 2.42$ eV) of the sample is less than twice of a single photon energy ($E_p = 1.55$ eV), the energy of two incident photons is sufficient to produce excitons or electron-hole pairs via the virtual state-assisted band-edge transition, which indicates that the lasing is probably induced by TPA. The excitation mechanism of the lasing can be further understood by the curve of integrated emission intensity versus the pump energy density. As illustrated in Fig. 3, the s-shaped courses of the experimental values clearly confirm lasing action of the CdS microwire. The lasing thresholds are $\sim3.3$ mJ/cm$^2$ ($\theta = 0^\circ$), $\sim4.3$ mJ/cm$^2$ ($\theta = 45^\circ$), and $\sim4.8$ mJ/cm$^2$ ($\theta = 90^\circ$), respectively, which is further confirmed by a simplified rate equation analysis.10 The emission intensity shows a nearly quadratic dependence.

FIG. 1. (a) Experimental set-up schemes for optical measurements. For micro-absorption measurement, the lens ($f = 30$ cm) is replaced by an objective ($40 \times$), (b) The bright field image, and (c) the micro-absorption spectrum of a single CdS microwire.

FIG. 2. Two-photon pumped room-temperature emission spectra from the end facet of the microwire as excitation energy density increases, keeping the polarization direction parallel to the sample axis ($\theta = 0^\circ$). The insets show the dark-field images corresponding to spontaneous emission, amplified spontaneous emission, and lasing oscillation, respectively. The scale bar represents 10 $\mu$m in length.
on the excitation energy below the threshold, which confirms the dominant contribution of TPA to the emission. It adds further evidence to the presence of the TPA-induced lasing in our sample, which in turn suggests that the TPA process can produce the net optical gain necessary for the lasing process in CdS microwire lasers. Meanwhile, it can be seen that the incident light’s polarization direction has an effect on the threshold for lasing and the optimized lasing threshold is obtained at $\theta = 0^\circ$, which is 1.45 times less than that of perpendicular situation.

Furthermore, the dependence of lasing emission on the polarization direction of the excitation light in the single CdS microwire is also investigated. Figure 4 shows the integrated emission intensity versus the different polarization angles at the excitation density of 5.0 mJ/cm$^2$. The emission intensity exhibits a periodic ($\cos^2\theta$) dependence on the polarization angle and the strongest lasing emission can be observed when the polarization direction of excitation light is parallel to the microwire axis ($\theta = 0^\circ$). The intensity ratio at $\theta = 0^\circ$ and $\theta = 90^\circ$ is about 4. This phenomenon can be attributed to the large dielectric constant mismatch between the CdS microwire and the air surroundings. At one-dimensional micro/nanowire system with the diameter comparable to the wavelength of excitation light, the optical field component perpendicular to the wire axis is attenuated inside the wire due to confinement caused by the large dielectric mismatch, while the component along the axis is not reduced.$^{25}$

The polarization-related lasing emission is further analysed by performing the FDTD simulations. In our simulation, a single CdS microwire ($D = 1\, \mu m$, $L = 28\, \mu m$) was situated in the $y$-$z$ plane with the microwire axis parallel to the $z$ axis. The plane wave (800 nm) was introduced as the excitation source and propagated along $x$-axis with different polarization angles. Figure 5 shows the cross section field distribution of the $y$-$z$ plane at $\theta = 0^\circ$, $45^\circ$, and $90^\circ$, respectively. At $\theta = 0^\circ$, the optical field is almost coupled into the microwire due to the negligible confinement effect along the axis. The dielectric mismatch becomes important at $\theta = 45^\circ$ and the field component perpendicular to the axis is attenuated, leading to the decrease of coupling efficiency. At $\theta = 90^\circ$, the strong confinement effect results in a dramatic drop of optical field inside the wire and the field largely distributes on the surface instead. It indicates that the transform from SE to laser emission occurs under a lower excitation energy with the parallel polarization and under a given pump energy the emission intensity is the strongest at the parallel case, which agrees with our experimental results well.

In summary, the TPA-induced longitudinal F-P mode lasing in a single CdS microwire is obtained. The sharp lasing peaks centered at 523 nm with an average linewidth of 0.4 nm indicate a high $Q$ factor of 1300. The polarization-dependent lasing emission exhibits the strongest emission intensity as the polarization direction of incident laser is parallel to the microwire axis. It indicates that the single CdS microwire has potential applications in frequency upconversion lasers and polarization-sensitive laser devices.

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