Multicolour laser from a single bandgap-graded CdSSe alloy nanoribbon

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Abstract: Multicolour lasing with wavelength varying from 578 nm to 640 nm is realized from a single bandgap-graded CdSSe alloy nanoribbon, by selecting the excited spot at room temperature. Though reabsorption is a serious problem to achieve lasing at short wavelength, multiple scatters on the nanoribbon form localized cavities, and thus lasing at different wavelengths is realized. By increasing the excitation area, multicolour lasing from the same nanoribbon is also observed simultaneously.

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1. Introduction

One-dimensional semiconductor micro-nanoscale waveguide (MNW), including nanowire and nanoribbon, is a promising building block for integrated photonic applications with compact size, strong optical confinement, sufficient optical gain, and easy synthesis [1]. Various kinds of devices based on single MNW have been realized, including lasers [2], light-emitting diodes [3], photodetectors [4], and photonic biosensors [5]. However, the factor that the majority MNW devices are made of homogeneous materials, thus mono-bandgaps, has limited many applications. In the past decade, the emerging of spatial bandgap-graded alloy semiconductor MNWs with composition-tunable gain has attracted intense attentions [6–10]. More recently, the realization of wide-range bandgap tenability in a single CdS$_{1-x}$Se$_x$ MNW structure has expended the application horizon greatly [11–13]. In such single crystalline structure, the composition along the length of MNW can be continuously varied from pure CdSe at one end to pure CdS at the other end, and consequently the bandgap is tuned from 1.74 eV to 2.44 eV. The broad emission/absorption spectra make it possible to realize novel devices, such as wide-wavelength tunable nanolaser, white light emitting diode, and full-spectrum solar cell, from a single nanowire structure.

However, due to the monotonous change of the bandgap along the bandgap-graded alloy MNW, such structure usually has the property that PL generated in the middle section of the MNW can only propagate freely to one of the ends, where the bandgap is widest. While along the bandgap-decreasing direction, due to the strong confinement of the optical mode, the guided PL light will be efficiently reabsorbed, and the reemission shows a red shift [13]. Thus one may think that, other than at long wavelength range, lasing based on axial Fabry–Pérot (FP) waveguide modes would be difficult to be realized from a single bandgap-graded alloy MNW structure, due to the huge loss caused by reabsorption. As a result, though multicolour lasers have been demonstrated based on semiconductor multi-nanowires [7, 14], however, we haven’t find such reports from single MNW.

In this work, we present multicolour lasers from a single bandgap-graded CdSSe alloy nanoribbon (BGCAN). By utilizing multiple scatters, several localized cavities are formed on the BGCAN, thus by exciting different sections of the nanoribbon, lasing behaviors are observed with wavelengths varying from 578 nm (yellowish green) to 640 nm (red). By increasing the size of excitation spot, simultaneous multicolour laser emission is observed from the same nanoribbon.

2. Sample preparation and experimental setup

The CdSSe BGCANs are synthesized on silicon wafer via source-moving thermal evaporation method [11]. In the method, the source is prepared by filling two boats in CdS and CdSe powders respectively, which are separated by another two empty boats. To vary the difference of temperature between two powder boats, a motor is employed to push the source along a horizontal quartz tube leading to position changing with respect to the heating zone. Hence the component x of the CdS$_x$Se$_{1-x}$ vapor can be continuously changed, and the chemical vapor is deposited on a silicon wafer placed downstream. For a more detailed description of the process, please refer to [11]. Figure 1(a) shows the scanning electron microscopy (SEM) image of a single BGCAN we use in our experiment. Other than two visible defects and one indent, the BGCAN shows a smooth surface and uniform thickness as 200 nm, even though the ingredients vary continuously along the nanoribbons. It has a length of 127 µm, and an average width of 3 µm.
The BGCAN is first transferred from the grown silicon wafer by a fiber taper to a glass slide. A 532 nm pulsed Nd:YAG (yttrium aluminum garnet) laser with 5 ns pulse duration, 2 kHz repetition rate is used as excitation source. The laser is focused via a 20 X object lens (NA = 0.4), and irradiates the BGCAN perpendicularly. The photoluminescence (PL) emission of the BGCAN is then collected by the same object and directed to a charge-coupled device (CCD) and a spectrometer (HORIBA Jobin Yvon iHR550) simultaneously. A notch filter with blocking wavelength centered at 532 nm is used to block the excitation laser pulses when taking images and spectra. The details of the optical excitation-collection experimental setup can be found in [15]. Figure 1(b) shows a PL image of the BGCAN taken by the CCD camera, where the pump laser spot can cover the whole nanoribbon. From this image, we can find the colour of the PL varies continuously from green to red along the nanoribbon, which indicates the gradual change of the composition, as well as the bandgap structure (schematically illustrated in Fig. 1(c)). The spectrum from the whole nanoribbon is shown in Fig. 1(d).

3. Results and discussion

We focus the laser beam to a spot of 25 µm in diameter, so that only one fifth of the nanoribbon is excited. When excitation intensity is increased above the threshold, several sharp lasing peaks emerge from broad PL background. Interestingly, depending on which section of the BGCAN being excited, lasing with different colours can be realized. As shown in Fig. 2(b), lasing peaks centered around 640 nm, 621 nm, 598 nm, and 578 nm are observed by moving the excitation spot along the nanoribbons from narrow bandgap end to the wide bandgap end (Fig. 2(a)). The lasing wavelength tuning range $\Delta \lambda$ is 61 nm, which covers most of the PL spectrum range of the BGCAN (plotted in Fig. 1(d)). Considering the central wavelength of the gain is around 610 nm, we have achieved a large tunability $\Delta \lambda / \lambda$ of 10% from a single semiconductor nanoribbon [7].
Fig. 2. (a) Schematic diagram of moving the focused excitation spot along the nanoribbon. (b) The normalized lasing spectra (shown in green, yellow, orange, red lines) collected by changing excitation spot along the nanoribbon. The insets are the corresponding lasing images at different wavelengths.

To ensure the lasing behavior, the dependences of laser intensity on the pumping power at different wavelength ranges are studied. As shown in the inset of Fig. 3, when the laser is focused on the narrow bandgap end of the BGCAN, sharp peaks around 633 nm emerge and enhance dramatically with a small increasing of pump power. This clearly indicates the onset of lasing behavior, and the threshold is measured as 10 MW/cm², as shown in Fig. 3.

Fig. 3. The relationships between output laser intensity and excitation power density. The red line and the green line represent the laser intensities centered at 633 nm and 579 nm, respectively. The inset shows the evolution of the lasing spectra centered at 633 nm with increasing excitation power density.

When the excitation laser is focused on the wide bandgap end, lasing is still realized around 578 nm, though the threshold is measured as 68 MW/cm² (shown in Fig. 3) and higher than that of the lasing mode at 633 nm. As we exam the corresponding lasing image, we find the laser mode does not extend to the whole nanoribbon as an ordinary single nanowire FP laser does, where the partial reflectivity at the two endfaces forms the cavity. As shown in the left inset of Fig. 2(b), the bright yellowish green light is mainly localized within a section terminated by one nanoribbon end and a bright spot where a visible defect exists (indicated in Fig. 1(a)). Beyond this section, the yellowish green light fades quickly, and the PL colour changes to red due to the strong reabsorption and reemission. Thus any reflection from the other endface won’t help to build up the green laser. Though defect is not favorable for a waveguide, however in this case, it acts as a mirror. In this way, the back scattering from defect provides the necessary feedback to build up a localized FP laser, and prevents the
stronger absorption due to the bandgap decreasing along the nanoribbon. Correspondingly, as shown in Fig. 4(a), we find that the spectrum peaks around 578 nm show mostly regular spacing similar to those of FP cavities, except the mode numbered as 5 and a missing mode (numbered as 7)—likely due to the imperfection of the nanoribbon end. The free spectrum range is measured close to 0.93 nm. This is further confirmed by plotting these peaks in frequency domain with nearly equal-spacing.

![Fig. 4. (a) Lasing spectrum centered around 578 nm. The peaks indicating individual optical modes in the spectrum are labeled with numbers. The inset shows the equal-spacing of the optical modes in frequency domain. (b) The lasing spectrum around 621 nm shows optical modes with irregular intervals.](image)

The several defects, together with two endfaces, may form many cavities on the BGCAN. This is the main reason we can tune the lasing wavelength as we move the excitation spot along the BGCAN. When the excitation laser is focused on the center part of the BGCAN, lasing peaks around 598 nm are observed. As shown in the middle inset of Fig. 2(b), mainly three bright yellow spots exist on the BGCAN, and their positions sit on one indent, one defect, and one endface. However, red colour PL is the leading one in the section above the indent. It is noted that for the red lasing mode peaks around 640 nm, several bright spots can be observed on the BGCAN other than two endface, as shown in the right inset of Fig. 2(b). Actually, the light scattered from the lower endface is even weaker than those from other scatters. This indicates that the laser cavity at this wavelength is also different from ordinary FP nanowire lasers cavity formed by the two endfaces. Namely, multi-scattering plays the main role in the forming of laser cavity at long wavelengths. As a result, as we can find in Fig. 4(b), the lasing peaks around 621 nm show irregular spacing, more or less similar to that from random lasers [16].

By using the multi-scattering effect, lasing modes at different wavelength can be localized at different sections of the nanoribbon. Thus the reabsorption problem of the BGCAN is solved, though not in a perfect way. Consequently, by moving the focused pump beam along the nanoribbon, the colour of laser can be controlled. Moreover, we are also able to achieve multicolour lasing simultaneously from one BGCAN. By replacing the 20 X object lens with a 10 X one, we increase the excitation area to 90 µm in diameter, which covers more than half of the BGCAN. Two groups of lasing peaks can be observed at the same time, and they are generated from different cavities (Fig. 5). While we move the excitation position, different lasing groups can be turned on and off. We believe that with a larger excitation area and a higher pump power, lasing from most groups within the gain range of BGCAN can be realized simultaneously.
By employing the bandgap-graded ZnCdSSe alloy nanowire [17], or organic photonic heterostructure nanowire [18], together with a proper pumping source (e.g. UV pulsed laser), single BGCAN white-light emitting laser with wavelength tuning range covering the whole visible spectra is able to be realized. This is favorable for many integrated photonic applications such as miniature tunable laser, ultrafast nanolasers, high resolution full colour display and sensing, and introduction of an external cavity with much higher quality factor may be helpful to reach this goal [19].

4. Conclusion

In summary, we have presented a multicolour laser based on single CdSSe bandgap-graded alloy nanoribbon. The existence of the multiple scatters on the BGCAN plays important role for realization of lasing at different wavelengths. By increasing the excitation area, multicolour laser emissions are observed simultaneously from the same nanoribbon. This work paves the way for broad BGCAN applications such as miniature tunable laser, high density colour display, and sensing.

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