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Dynamical Color-Controllable Lasing with Extremely Wide Tuning Range from Red to Green in a Single Alloy Nanowire Using Nanoscale Manipulation

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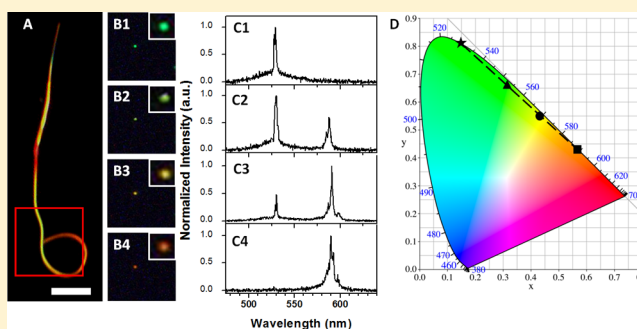
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S Supporting Information

ABSTRACT: Multicolor lasing and dynamic color-tuning in a wide spectrum range are challenging to realize but critically important in many areas of technology and daily life, such as general lighting, display, multicolor detection, and multiband communication. By exploring nanoscale growth and manipulation, we have demonstrated the first active dynamical color control of multicolor lasing, continuously tunable between red and green colors separated by 107 nm in wavelength. This is achieved in a purposely engineered single CdSSe alloy nanowire with composition varied along the wire axis. By looping the wide-gap end of the alloy nanowire through nanoscale manipulation, two largely independent (only weakly coupled) laser cavities are formed respectively for the green and red color modes. Our approach simultaneously overcomes the two fundamental challenges for multicolor lasing in material growth and cavity design. Such multicolor lasing and continuous color tuning in a wide spectral range represents a new paradigm shift and would eventually enable color-by-design and white-color lasers for lighting, illumination, and many other applications.

KEYWORDS: Cadmium sulfide selenide, alloy composition-graded nanowire, nanowire manipulation, multicolor laser, dynamic color tuning



Multicolor laser sources have an extremely wide range of applications including color display,¹ general lighting,² biological detection,³ holographic imaging,⁴ and three-dimensional (3D) projection.⁵ Color display itself is important in many aspects of technology and daily life. One important advantage of multicolor lasers for color display is the more widely available color range, or color gamut, compared to the currently existing display technologies based on incoherent sources, such as cathode ray tube (CRT) and organic light emitting diode (OLED). The large spectral line width of incoherent light sources degrades the color purity and leads to a small color gamut.^{6,7} As coherent light sources, multicolor lasers render high-purity monochromatic colors and thus extend the color range significantly if proper wavelengths are chosen. For lighting and illumination applications, it is interesting to note the recent research² showing that the combination of four separate lasers with specifically selected wavelengths can achieve the large chromaticity range^{1,8} and similar color rendering ability as state-of-the-art LEDs or phosphors, even though lasers contain only a series of narrow emission lines. For many of the above applications that require high power output, multicolor lasers offer great advantages due to the much higher wall-plug efficiency^{9–11} than incoherent LED sources, thus

leading to greater energy efficiency. While the importance of multicolor lasers and dynamical color control has been well-recognized for a long time, the realization of such sources has been challenging due to several technology barriers.

Multicolor lasers that are necessary for all these critical applications mentioned above require color ranges with widely separated wavelengths, or even across the entire visible spectrum. Such multicolor lasers are fundamentally different from multimode lasers. The latter is made of a given semiconductor and relies on cavity structures to generate multiple lasing wavelengths corresponding to various cavity modes. Since these multiple modes are all supported by the same gain material, their separation is limited within the gain bandwidth of a semiconductor, which typically ranges in 1–30 nm.^{12,13} Such a wavelength range is too small for display or lighting applications. Thus multicolor lasers require integration or monolithic growth of multiple gain materials, or semiconductor alloys of different alloy compositions.^{14,15} This requirement poses a formidable challenge for the traditional planar epitaxial technology due to the large lattice mismatch

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typically involved. In addition to material challenges, cavity design is also a crucial issue when multiple gain materials are involved in a single integrated structure. This is because the light emitted by the wide-gap materials will be absorbed by the narrow-gap materials. Thus lasing is typically achieved only in the longest wavelength of the structures. Innovative ideas and designs are needed to achieve multicolor lasing.

With the advent of nanotechnology in the past decades, many novel ideas have been pursued for multicolor lasing. Various small-size coherent and incoherent multicolor light sources based on nanowires,^{16,17} quantum dots,^{13,18,19} and microfluidic droplet dye-lasers^{20–22} have been demonstrated. However, in most of these designs the multicolor lasers were obtained by combining several single-wavelength lasers with additional waveguides to guide and mix the multicolor emissions,^{17,20–22} thus inevitably increasing the dimension of the devices and cost of the technology. Recently, spontaneous emission of different colors was demonstrated from different sections of a single ZnCdSse alloy nanowire.²³ Very recently, we demonstrated two-color lasing from a single nanosheet.²⁴ Multicolor emission and lasing were also demonstrated on a single substrate where different nanowires with different alloy compositions graded along the substrate show different emission colors.^{14,15,25,26} Despite such progress, multicolor lasing with active color control has not been realized.

In this paper, we attempt to address these challenges by using a novel combination of gain materials and cavity design to demonstrate room-temperature lasing at two visible colors (red and green) from a single CdSse alloy nanowire. The wavelength separation of 107 nm between two colors is much larger than the gain spectra bandwidth limitation of typical semiconductor materials.^{27–31} More importantly, we show that the color of the total laser output can be controlled dynamically between the two fundamental colors by changing the relative pumping strength of the two segments. This allows the laser generation of any color between the two fundamental colors according to the color map of International Commission on Illumination (CIE). Our novel dual-color nanowire laser is based on CdSse alloy nanowires with purposely engineered bandgap variation along a single nanowire.³² The continuously tuned alloy composition along the wire allows the bandgaps to be eventually varied widely enough to generate the two distinct colors and at the same time assures the high quality of semiconductor crystal due to the gradual composition change. By looping the wide-gap end of the nanowire, two relatively isolated (or weakly coupled) cavities are created, reducing the absorption of the short wavelength light in the narrow-gap section and allowing both segments (cavities) to lase simultaneously. This way the two fundamental (material and cavity structure) challenges faced with multicolor lasing are solved at the same time.

Our approach is schematically shown in Figure 1. The sulfur composition, x , of $\text{CdS}_x\text{Se}_{1-x}$ alloy can be changed continuously from 1 at one end to 0 at the other end, resulting in a color variation of bandgap emission from green to red along the nanowire (Figure 1A).³² It is interesting to see whether such a nanowire could lase at different colors simultaneously since it contains different gain materials for different colors in one cavity. Because the wide-gap (CdS-rich) segment is transparent to the red light emitted by the CdSe-rich part, red color can easily reach the threshold and start lasing. However, the shorter wavelength emission from the CdS-rich section will experience a strong absorption in the narrower-gap (CdSe-rich) section,

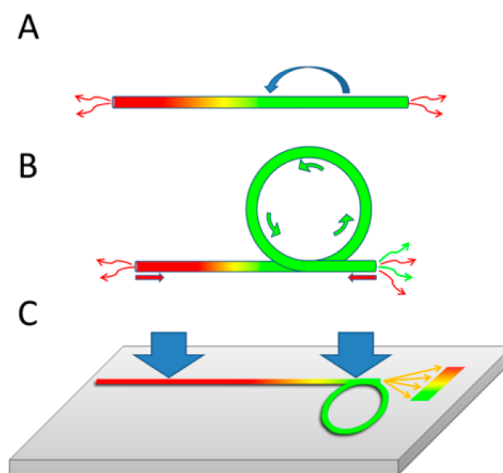


Figure 1. (A) Sketch of a straight CdSse alloy nanowire. The color variation along the wire represents the continuously changing composition. The straight wire can only support red lasing due to the strong absorption of green light by the red segment. (B) Sketch of the looped nanowire, which can support both green and red lasing. (C) Design of the color-tunable laser using this looped nanowire by changing the relative strengths of pumping of the two segments, indicated by the two blue arrows. The total output color at the loop junction can change from red to green continuously.

thus green lasing becomes nearly impossible without an extremely long segment (see Sections S2 and S3 in Supporting Information for detailed analysis). In order to achieve simultaneous lasing at both long and short wavelengths, light emitted from the wide-gap region needs to be well-confined within its own segment. One convenient but critically important way to create distinct cavities in a single nanowire structure is to loop the wide-gap section to form a relatively isolated cavity for the green light as illustrated by Figure 1B. It has been demonstrated that the junction coupling efficiency of such a ring cavity is very high,^{33–35} providing strong feedback for the green emission and reducing the strong absorption in the straight part (narrow-gap part). As a result, the green mode can oscillate in the ring cavity with enough gain and low loss to achieve lasing. At the same time, the red emission from the CdSe-rich section can still propagate in the entire nanowire with a cavity defined by two end-facets (labeled by red arrows in Figure 1B). The merit of the looped structure and its comparison with straight wire are presented in detail in Section S4 of Supporting Information.

The unique feature of our looped-cavity design enables the active tuning of the output color. Figure 1C shows the concept of color-tunable laser via the looped CdSse alloy nanowire. The excitation laser output is split into two beams to pump the green looped part and red straight part separately, allowing for the control of the relative intensities of each part by changing its corresponding pumping strength. As a result, the overall laser output from the looped nanowire would continuously change color from green to red corresponding to the change in relative mixture of the two fundamental colors.

Figure 2 shows the results of optical characterization of the straight CdSse alloy nanowire before looping. A nanowire of 200 μm in length and 400 nm in diameter was placed on a glass substrate. Because of the composition grading along the length direction, the real color photoluminescence (PL) image shown in Figure 2B under 405 nm laser excitation displays the corresponding color changes from green to red. To

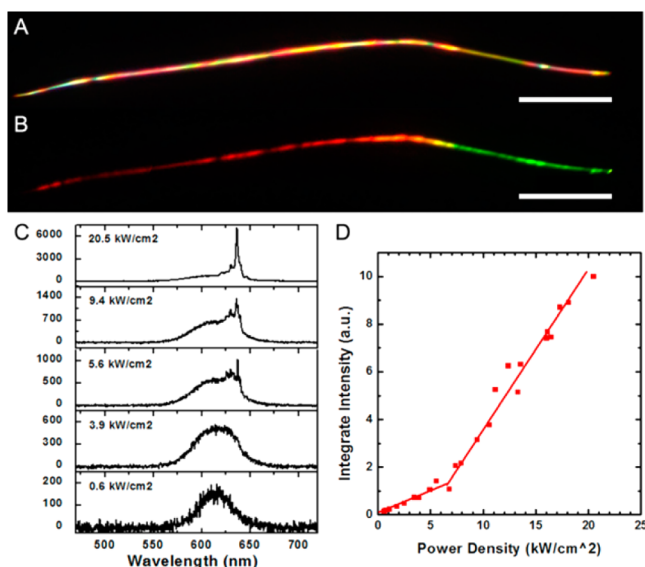


Figure 2. Optical characterization and red color lasing of a straight CdSe alloy nanowire. (A) Dark-field image of the nanowire. (B) The real color PL image of the nanowire. Scale bars in A and B are 30 μm . (C) Emission spectra under a single excitation pulse with increasing peak power density. (D) The integrated intensity of the lasing peak at 637 nm versus the excitation power density.

demonstrate lasing, the nanowires were then pumped by a 355 nm pulsed laser with repetition rate of 10 Hz and pulse width of 9 ns (from the third harmonic of the 1064 nm YAG laser). The excitation laser is focused to 250 μm in diameter spot to uniformly pump the entire nanowire. The emission spectra under different pumping powers are shown in Figure 2C. Consistent with the previous prediction, only the red lasing peak at 637 nm appears even up to high pumping level. The integrated intensity of the lasing peak versus the excitation power is plotted in Figure 2D, which shows a clear lasing threshold with peak pumping power density at 6.7 kW/cm².

Figure 3 illustrates the dual-color lasing result after looping the green end of the same nanowire as in Figure 2. A tapered fiber was used to manipulate and loop the green end of the wire into a 15 μm diameter circle, which is shown in Figure 3A. Figure 3B shows the real color image of the dual-color lasing emission under uniform excitation of the entire structure. The bright spot at the junction of the loop (labeled by green arrow) is a good demonstration of the waveguiding behavior. Because this junction is the output point of both green and red lasing, the color of this bright spot appears yellowish, as a result of color mixture of these two colors. In addition, a red spot (labeled by the red arrow) is observed on the straight segment of this wire. We performed scanning electron microscopy on this nanowire after lasing experiment and found that it is due to scattering from a small particle attached to the nanowire (see Section S1 in Supporting Information). Contrary to the case of the straight nanowire in Figure 2C, the lasing spectra of the looped wire exhibits a significant green lasing peak at 530 nm under high excitation power (Figure 3C), while the center of the red lasing peak remains at 637 nm, consistent with the lasing wavelength before looping. The output intensities of the two lasing peaks are plotted in Figure 3D as a function of total pumping power density, showing clear threshold behavior for both colors, consistent with the spectral change in Figure 3C. The threshold power density of the red and green lasing are 6.9

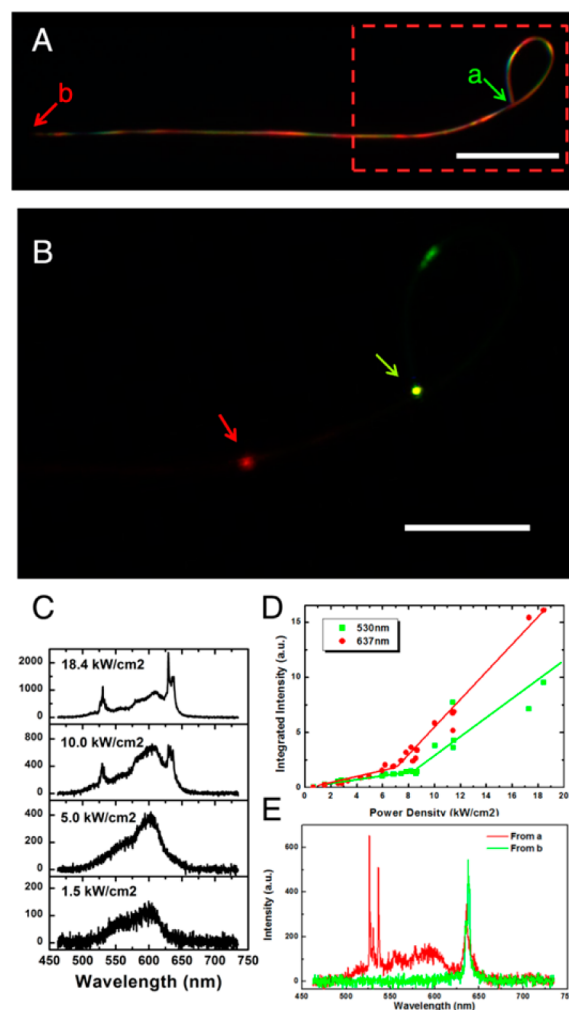


Figure 3. Optical properties of the looped nanowire. The wire is the same one as in Figure 2. (A) Dark-field image of the looped nanowire under white light illumination. Labels *a* and *b* indicate the junction of looping and the end point of the narrow-gap end, respectively. Scale bar is 30 μm . (B) Real color image under more intense pumping showing dual-color lasing. The image area corresponds to the red dashed box in (A). Scale bar is 15 μm . Green arrow indicates the junction of the loop, corresponding to the position *a* in (A). Both red and green lasing can emit from this point thus it appears in yellow color. Red arrow points to a scatter point on the wire body. (C) Spectra under a single pulse pumping with different peak power density, showing two sets of lasing peaks around 530 and 637 nm, respectively. (D) Integrated intensities of the green and red lasing versus the pumping power density. (E) Spectra obtained using confocal measurement with green curve measured at point *a* and red curve at point *b*.

and 8.7 kW/cm², respectively. The clear threshold is also evidenced by the well-known “S”-shaped curve when the laser intensity is plotted against pumping intensity on log–log scales, as shown in Figure S10 of the Supporting Information. The wavelength separation of the green and red lasing modes is 107 nm, much larger than the gain bandwidth of the typical II–VI semiconductors of a single composition.^{27–31} Because of the largely separated optical cavities, the wavelength spacing of the two-color lasing is determined only by the composition distribution along the nanowire. More details about the analysis of the average gain spectra for looped and straight cavities can be found in Sections S2 and S3 of Supporting Information. For

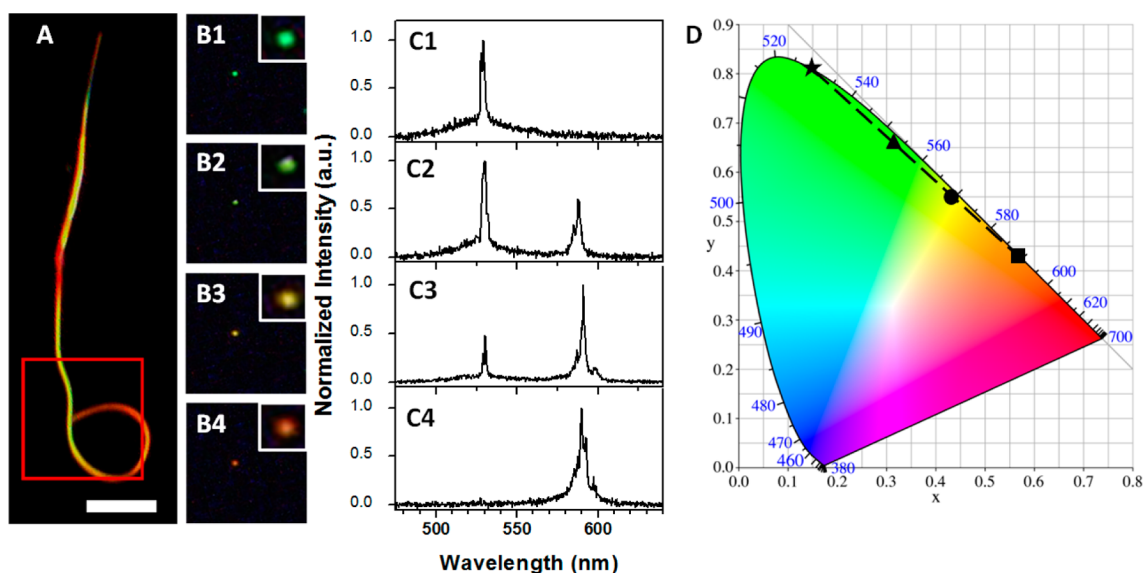


Figure 4. Color control of a looped wire dual color laser. (A) Dark-field image of the looped nanowire, where the scale bar is 10 μm . Two excitation beams are focused to less than 40 μm spot size to the looped and straight sections respectively. (B1–B4) Real color images of the lasing under different pumping for the two cavities. The images are taken from the red box in A. The bright spots correspond to the junction of the loop. The insets show the zoom-in images of the junction lasing spots. (C1–C4) Normalized lasing spectra under different controlled pumping intensities, collected simultaneously with the images of C1–C4. The intensity ratios of the green and red modes in C1–C4 are 10:0, 6:4, 3:7, and 0:10, respectively. (D) The calculated colors from the spectra in C1–C4 plotted on CIE1931 color space. Star (★), triangular (▲), circle (●), and square (■) labels correspond to C1–C4, respectively.

the geometry and composition gradient of the wire shown in Figure 3, the two wavelengths with maximum gain are separated by 123 nm, which is consistent with the experimental result of 107 nm. With further optimization of material growth, the wavelength separation can be eventually extended to around 200 nm in such CdSSe alloy nanowires, as discussed in the Supporting Information.

The confocal PL measurement in Figure 3E demonstrates the output position of the two-color lasing. By inserting a pinhole into the image plane, we were able to collect the emission spectra only from an area less than 5 μm at both the loop junction and the CdSe-rich end-facet (position *a* and *b* in Figure 3A respectively). The spectrum at the junction shows both green and red lasing peaks while the spectrum at CdSe-rich end only exhibits longer wavelength lasing, consistent with the previous discussions. Because of the spatial filtering effect of the confocal system, the relative intensity of the green peaks is stronger than that obtained with a large collection area in Figure 3C. The line width of a single lasing peak at 526 nm is 0.9 nm, limited by the resolution of the spectrometer. The longitudinal mode spacing is 1.1 nm. Using the approximate expression of the wavelength separation for a ring cavity: $\Delta\lambda = \lambda^2/Ln_g$, where $L = 47 \mu\text{m}$ is the length of the loop, we obtained a group index n_g of 5.4 for the green mode, consistent with the number reported in refs 36 and 37.

One unique property of our looped laser design is the common output position for two-color lasing. Unlike RGB lasers^{1,38} and some other multicolor lasers,^{17,20} our dual-color laser does not require external guiding to mix the multicolor lasing beams. More importantly, this unique feature allows us to achieve color-tunable output via pumping the straight and looped segments separately, as indicated in Figure 1C. A beam splitter was used to split the excitation laser into two beams for pumping the CdSe-rich straight part and CdS-rich loop separately. The neutral density filters are applied after the

beam splitter to precisely control the incident power of two excitation beams. Figure 4A shows the dark-field image of another looped nanowire. By adjusting the intensities of two pump beams, the output color at the junction was tuned from green (B1) to yellow-green (B2) to yellow (B3) to orange (B4). C1–C4 in Figure 4 show the lasing spectra corresponding to B1–B4. In C1 and C4, only one of the two excitation beams was used for excitation while the other one is blocked. Therefore only one lasing color is observed at either 530 nm (green) or 588 nm (orange). In C2 and C3, the relative intensity of the green and orange lasing is controlled at 6:4 and 3:7, respectively. The mixed colors are represented as the intermediate colors between green and orange, which are close to yellowish green (B2) and yellow (B3). In Figure 4D, the chromaticity of C1–C4 spectra are marked on CIE 1931 color space by ★, ▲, ●, and ■, respectively. The monowavelength lasing of C1 and C4 are located at the curved edge of the color space. C2 and C3 can be considered as the linear combination of two equivalent wavelengths, and the corresponding chromaticity are marked on the dashed line in Figure 4D. The calculated colors in Figure 4D match perfectly with the colors in real color images shown in Figure 4B1–B4, further validating the monochromatic property and color tunability of this looped nanowire laser. Additionally, any color on the dashed line can be obtained from this nanowire by precise control of the relative power of the pumping beams. Such continuously controllable colors are critically important for many applications such as color-by-design for lighting, color display, or other technological interfaces.

In summary, we demonstrated a novel design of the dual-color laser based on CdSSe alloy nanowires. By looping the CdS-rich section of the nanowire, we achieved simultaneous green and red color lasing in a single alloy nanowire. The 107 nm wavelength separation of the two lasing colors is much larger than the typical gain bandwidth of a semiconductor.

More importantly, our looped-end design allows for dynamic control or tunability of the combined colors in a wide range, so that any intermediate color between the two lasing wavelengths is achievable by adjusting the relative excitation power. Our design and demonstration show several interesting advantages including a single device with small size, flexible lasing colors and active color tunability in an extremely wide range compared to the gain bandwidth of a single semiconductor. As a result, we believe that this extremely wide-range controllability of laser colors have a great potential in the photonic integrated circuit, small size white color laser and on-chip laser display. Such a capability would be extremely powerful if extended to structures with three colors and especially under electrical injection, so that any desired color or color combinations can be readily achieved with a simple change of relative injection levels. We believe that such a widely tunable full-color source would offer a revolutionary solution to many fields such as lighting, displays, and any other situation where dynamical color control is needed.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information includes more details on material synthesis method, experimental techniques, mode simulation, and supporting figures. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Author Contributions

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Notes

The authors declare no competing financial interest.

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