Photoluminescence from disc-shaped Si₃N₄/WS₂/Al₂O₃ heterojunction

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Abstract. Transition metal dichalcogenides (TMDCs) have a direct band gap and exhibit more peculiar properties than graphene. Two-dimensional TMDCs (2D TMDCs) are widely used in many fields such as catalysis, energy storage, and composite materials due to their unique sandwich structure. Compared to the three-dimensional structure of silicon materials, TMDCs have a two-dimensional layered structure at the nanometer scale, which can be used to manufacture semiconductors or smaller, more energy-efficient electronic chips, which will be widely used in the next generation of nanoelectronic devices.

In this paper, continuous light and femtosecond laser are used to irradiate disc-shaped Si₃N₄/WS₂/Al₂O₃ heterojunction, and the photoluminescence characteristics of the heterojunction are studied by changing the distance between the sample and the light source to change the incident power.

1. Introduction

TMDCs are two-dimensional semiconductors that change from an indirect band gap to a direct band gap [1]. 2D TMDCs have high optical absorption, strong luminescence emission, large exciton binding energy, and relatively high carrier mobility [2]. The rate is a high-quality optical material that creates new advantages for the next generation of ultra-compact photonics and optoelectronics, making it ideal for next-generation optoelectronic devices [3]. Importantly, they are compatible with existing planar technologies and can be easily grown on Si or SiO₂ substrates [4]. Compared with traditional III-V quantum well lasers, 2D TMDCs are expected to have the advantage of quantum well lasers. The high refractive index of 2D TMDCs can significantly increase the limiting factor in the laser active region [5]. The Coulomb interaction can further enhance the optical gain due to the reduced dielectric shielding in these materials. The extremely short radiated and non-radiative carrier lifetimes of these materials indicate the possibility that the two-dimensional material has a light-emitting device that can achieve a high modulation bandwidth [6]. The transition between the valence band and the edge of the conduction band is essentially derived from excitons in this single layer system. Strong exciton properties, including neutral and charged excitons, have been observed and studied in this paper. Strong excitons in two-dimensional TMDCs can be used not only to control the emission characteristics of photons, but also to achieve high optical gain and to provide the long-lived population inversion required for stimulated emission [7].

Therefore, the laser emission of a single-layer WS₂ by embedding a single-layer WS₂ in a ring resonator was studied [5]. In this experiment, the paper mainly studies the stimulated radiation
properties of the two-dimensional material Si₃N₄/WS₂/Al₂O₃ heterojunction, which are shown in Figure 1.

Figure 1. (a) Schematic of disc-shaped Si₃N₄/WS₂/Al₂O₃ heterojunction. (b) Lateral view of the structures. (c) Schematic of the photoluminescence from sample experiment setup.

2. Results and discussion

Microcavity structure has been widely used as an effective way to effectively improve the luminous efficiency of silicon materials. The working principle of the microdisk is that when light is transmitted from a high refractive index material to a low refractive index material, and the incident angle satisfies a certain size, total reflection occurs [8]. Therefore, when transmitting in a silicon nitride-based circular cavity structure, as long as the total reflection condition is satisfied, light is continuously transmitted in the disk, and interference constantly occurs to form a stable mode. A silica disk was placed on a suspended silicon column. Due to the fluorescent nature of the two-dimensional material heterojunction, the two-dimensional material is transferred onto a silicon nitride disk to form a high-gain microdisk. There is air on both sides of the microdisk. The refractive index of silicon nitride (nₙ$_{st}$=2.0) is larger than the refractive index of air (n$_{air}$=1.0), and the light is strongly restricted when it is transmitted in the silicon nitride cavity structure. The light field is mainly concentrated in the core region with a higher refractive index, and the energy loss is small. There are two functions of Al₂O₃ in this structure. One is to avoid direct contact between tungsten disulfide and air, and to reduce the influence of defects; the second is to increase the modal gain.

The silicon nitride disk cavity structure suspended in the paper is prepared by photolithography. First, a single crystal silicon wafer was chosen to grow 300 nm thick silicon nitride on a single crystal silicon wafer. And the photoresist was applied evenly on the silicon nitride and bake the photoresist to evaporate the solvent in the photoresist. This improves adhesion and exposure accuracy. Next, a mask for the pattern required for photolithography was prepared, and the mask was irradiated with ultraviolet light to chemically react the irradiated photoresist, and then etched to transfer the photoresist pattern onto the silicon nitride material. The sensitivity of the photosensitive and non-photosensitive photoresist to the alkaline solution was different, and the pattern on the mask was completely transferred to the surface of the silicon nitride. The exposed photoresist was then immersed in the developer to reveal the latent image in the film by dissolving a portion of the photoresist. Finally, after another baking, the residual solvent content in the gel is further evaporated to a minimum to harden it. Then, the material was placed in a KOH solution, and KOH reacted only with Si, did not react with Si₃N₄, and the periphery of Si was corroded by reaction, and the preparation of suspended Si₃N₄ was completed. The transfer of the two-dimensional material heterojunction to the silica completes the preparation of the suspended disk cavity. The diameter of the silicon nitride microdisk was 5 μm and the thickness was 300 nm, supported by a silicon pillar (the lateral dimension of it was about 1 μm) in the middle of the microdisk, which are shown in Figure 1.
Figure 2. (a) Photoluminescence of the disc-shaped Si$_3$N$_4$/WS$_2$/Al$_2$O$_3$ when the pump light is continuous light with the power is 1400 μW (black) and 2000 μW (blue). (b) Comparison of photoluminescence of femtosecond lasers and continuous light. (c) Photographs of Si$_3$N$_4$ discs after partial etching of silicon with KOH, the central yellow area shows silicon support, and the surrounding green is the edge of the suspended Si$_3$N$_4$ disc. (d) Photograph of disk stimulated radiation. (The middle area is the disc-shaped Si$_3$N$_4$/WS$_2$/Al$_2$O$_3$ heterojunction and the right side is the focusing fiber lens).

Figure 2 (a) shows the photoluminescence of a sample irradiated with continuous light with a wavelength of 405 nm. Figure 2 (b) shows the comparison of photoluminescence of femtosecond lasers and continuous light. Herein, we adjust the output power by changing the distance between the sample and the light source and the center wavelength of the femtosecond laser is 880 nm. We choose two output powers for continuous light: 1400 μW and 2000 μW. For Figure 2(a), the heterojunction emits fluorescence under continuous light of different powers, and as the power increases, the fluorescence intensity increases. Through Figure 2 (b), it is found that the growth rate of peaks near 795 nm, 710 nm, 693 nm and 660 nm is significantly larger than that of background noise, and it can be roughly determined that laser is generated at these wavelengths. Comparing the two figures, the photoluminescence excited by the femtosecond laser is very different, especially at around 650 nm, its emission intensity is higher than that of continuous light excitation. This is because the carrier recombination in WS$_2$ is not in thermal equilibrium. In the ultrafast process, due to the strong linear carrier-carrier scattering process in WS$_2$, hot electrons holes pairs with energy higher are relaxed quickly to lower energy level [9].

At 1400 μW excitation power, the whispering gallery modes (WGMs) superimposed on the WS$_2$ spontaneous emission spectrum are clearly observed. The free spectral range (FSR) is defined as the
wavelength difference or frequency difference between the modes of the optical microdisk. According to the FSR definition formula, the free spectral range of the two modes increases as the wavelength increases. Through calculation, we can get the FSR = 18 nm. As can be seen from the figure, the mode width of the center wavelength of 650 nm is 17 nm. The calculation results are basically consistent with the experiment.

The Zhang Xiang team of the University of Berkeley in the United States measured the two-dimensional WS₂ laser shot in the WS₂ disk laser reported in 2015 [5]. The test was carried out at low temperature (10K) using a 473 nm femtosecond fs laser (190 fs pulse duration, 80 MHz repetition rate) to pump WS₂ with a measured WS₂ laser threshold of approximately 5–8 mw/cm². In our experiments, the Si₃N₄/WS₂/Al₂O₃ laser threshold measured under continuous light irradiation was about 1.1×10⁴ W/cm², which was about two orders of magnitude lower than that of the previous Zhang Xiang group. Our experiment was carried out at room temperature and did not use low temperature conditions. The pump light we use is a continuous light emitting diode, not a femtosecond laser. The structure we have made is greatly reduced compared to the experimental conditions of the Zhang Xiang group.

3. Conclusion
In summary, our study demonstrates photoluminescence of the disc-shaped Si₃N₄/WS₂/Al₂O₃, which are typically linearly excited non-emissive materials. The disc structure greatly reduces heat dissipation. Ultrafast excitation can pump materials to very high electrical temperatures compared to continuous light. Hot carriers induce phonons to play a key role in the disc-shaped Si₃N₄/WS₂/Al₂O₃. Multimode lasing spectra were obtained by photoluminescence, which provides a reference for further research.

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