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Photoluminescence enhancement of monolayer tungsten disulfide in complicated plasmonic microstructures



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ABSTRACT

Two-dimensional van der Waals materials are interesting for fundamental physics exploration and device applications because of their attractive physical properties. Here, we report a strategy to realize photoluminescence (PL) enhancement of two-dimensional transition-metal dichalcogenides (TMDCs) in the visible range using a plasmonic microstructure with patterned gold nanoantennas and a metal-insula tor-semiconductor-insulator-metal structure. The PL intensity was enhanced by a factor of two under Y-polarization due to the increased radiative decay rate by the surface plasmon radiation channel in the gold nanoantennas and the decreased nonradiative decay rate by suppressing exciton quenching in the SiO₂ isolation layer. The fluorescence lifetime of monolayer tungsten disulfide in this structure was shorter than that of a sample without patterned gold nanoantennas. Tailoring the light-matter interactions between two-dimensional TMDCs and plasmonic nanostructures may provide highly efficient optoelectronic devices such as TMDC-based light emitters.

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1. Introduction

Two-dimensional van der Waals materials are receiving attention for fundamental physics exploration and device applications because of their interesting physical properties [1]. Twodimensional van der Waals materials include gapless graphene and monolayer transition-metal dichalcogenides (TMDCs), which are two-dimensional semiconductors that undergo an indirect-todirect bandgap transition as their dimensions are decreased from bulk to several layers and finally to a monolayer [2,3]. Because of the above features, typical two-dimensional TMDCs, such as monolayer molybdenum disulfide (MoS₂), tungsten disulfide (WS₂), molybdenum diselenide (MoSe₂) and tungsten diselenide (WSe₂), are fluorescent, with the photoluminescence (PL) maxima at 680 nm (1.83 eV), 618 nm (2.01 eV), 792 nm (1.57 eV) and 752 nm (1.65 eV), respectively [2–9]. Although two-dimensional TMDCs are interesting for use in optoelectronic devices [10,11], the interaction between light and the material is weak because of the intrinsic monolayer thickness, resulting in poor light emission and absorption behavior. Therefore, a technique should be used to improve the PL properties of two-dimensional TMDCs [12]. The PL properties of TMDCs can be improved in simple photonic (or plasmonic) nanostructures, such as photonic crystal nanocavities [13], nanoparticles [14], nanocavities [15], nanoshells [16] and nanoantennas [17–21]. However, to date, the PL properties of TMDCs in complicated plasmonic microstructures have received little attention.

2. Materials and methods

2.1. Materials

Here we use a complicated plasmonic microstructure with a configuration of metal-insulator-semiconductor-insulator-metal (MISIM), as shown in Fig. 1(a), to realize PL enhancement of monolayer WS₂. The complicated plasmonic microstructure consists of a two-dimensional periodic array of gold nanoantennas (i.e., gold nanoparticles), an ultrathin SiO₂ isolation layer, a monolayer of WS₂, an ultrathin SiO₂ isolation layer, and a gold layer on top of a SiO₂ substrate, as shown in Fig. 1(a)–(c).

2.2. Gold nanoantennas

The gold nanoantennas increase the local light field in the following ways. When interacting with incident light, each single gold unit produces a local surface plasmon (LSP) mode, which



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Fig. 1. Schematics of (a) the complicated plasmonic microstructure, (b) the gold nanoantenna array and (c) a single gold nanoantenna unit. (d) Calculated local-field magnitude at the position of monolayer WS₂ as a function of wavelength for a single gold nanoantenna unit (black line, magnified by 1000 times) and gold nanoantenna array (red line) under Y-polarized excitation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

induces a strong resonant field in its vicinity [22]. Meanwhile, the array of gold units forms a two-dimensional plasmonic crystal that produces lattice diffraction modes. The coherent coupling between the LSP of gold units and lattice diffraction modes forms collective LSP, i.e., lattice LSP, with narrow linewidth and large quality factor [23,24]. We compared the local-field magnitude in the position of monolayer WS_2 as a function of wavelength for a single gold nanoantenna unit and gold nanoantenna array under Ypolarization incidence by calculation using the finite element method in the finite element solver package COMSOL Multiphysics. In COMSOL Multiphysics, we first drew the geometry of the complicated structure and set the parameters of the refractive index and sizes of each layer of the structure. As for the metamaterial (i.e., gold nanoantennas) and gold layer, the data of permittivity at different wavelength were adopted. We only drew one unit of metamaterial. To calculate the local-field magnitude for a single unit and an array, we set scattered boundary condition and periodic boundary condition around the unit of metamaterial, respectively. The incident direction of illumination beam was set to be perpendicular to the surface of the structure. The polarization of illumination beam was set to be Y direction, which means the electric field vector was along the long side of gold nanoantenna. And the electric field intensity of illumination beam was set to be $E_x = 0$, $E_v = 1 V/m$, $E_z = 0$. The wavelength of illumination beam was swept from 550 nm to 750 nm, which covers the center wavelength of the PL spectrum for monolayer WS_2 . We set a probe at the WS_2 layer to detect the local-field magnitude induced by the structure. The simulation results is presented in Fig. 1(d). The local-field magnitude of the gold nanoantenna arrays is much larger than that of a single gold nanoantenna unit. The peak of the local-field magnitude locates at a wavelength of 615 nm, i.e. the surface plasmon resonant wavelength, which is close to the PL peak of monolayer WS₂ (centered at 618 nm).

2.3. Fabrication procedure

The fabrication procedure (shown in Fig. 2) of the complicated plasmonic microstructure began with depositing a 150-nm-thick gold layer on a SiO_2 substrate to form the bottom reflective layer using an electron-beam evaporation system (E-beam Evaporator, 50B, Cello Tech, China). This was followed by depositing a 6nm-thick SiO₂ isolation layer by using the electron-beam evaporation system. Monolayer WS₂ prepared by chemical vapour deposition was then transferred onto the SiO₂ isolation layer. Another 6-nm-thick SiO₂ isolation layer was deposited on the monolayer WS₂ using the electron-beam evaporation system. Finally, using electron-beam lithography (Raith 150, Raith, Germany) and standard lift-off process, a 2-nm-thick chromium adhesion layer and 30-nm-thick gold nanoantenna array were patterned on the top SiO₂ layer. The lattice constant was 200 nm in the X direction and 650 nm in the Y direction, as illustrated in Figs. 1(c) and 3(a). The area of the gold nanoantenna array was around $120 \times 120 \,\mu\text{m}$ (Fig. 3(b)). We also fabricated a reference sample consisting of a monolayer WS₂ film on an Au/SiO₂ substrate.

2.4. Micro-spectrum measurement system

The PL spectrum of the complicated plasmonic microstructure was measured using a micro-spectrum measurement system. A beam from a Ti:sapphire femtosecond laser system (Chameleon Ultra Vision and Vision-S Diode-Pumped Lasers, Coherent Company, USA) with a wavelength of 405 nm, pulse duration of 140 fs and repetition rate of 4 MHz was used as the excitation light. We excited and collected PL signals at normal angle and analyzed them using a spectrometer, as shown in Fig. 3(c).

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