

Enhanced Luminescence of MoS₂, WS₂ and WSe₂, Direct Band Gap Semiconductor Heterostructures

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Abstract: We introduce van der Waals heterostructures to amplify the otherwise negligible cathodoluminescence from monolayer transition metal dichalcogenides, which allowed to visualize the nanoscale variation of strain and photonic modes in the monolayer on engineered surfaces. © 2018 The Author(s)

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We will report on the giant enhancement of the luminescence from monolayers of transition metal dichalcogenides (TMDCs) under free-electron excitation, which makes cathodoluminescence spectroscopy of TMDCs viable at the monolayer level even with the presence of highly luminescing oxide substrates. Such enhancement was enabled by forming a van der Waals (vdW) heterostructure, in which the luminescent TMDC layer is sandwiched between layers of hexagonal boron nitride (hBN) with a wider band gap. The sandwiched TMDC monolayer showed three orders of magnitude stronger cathodoluminescence (CL) compared to that from isolated monolayer (Fig. 1). The high spatial resolution of CL probe enabled the characterization of nanoscale variation of strain and optical density of states in the monolayer on engineered surfaces.

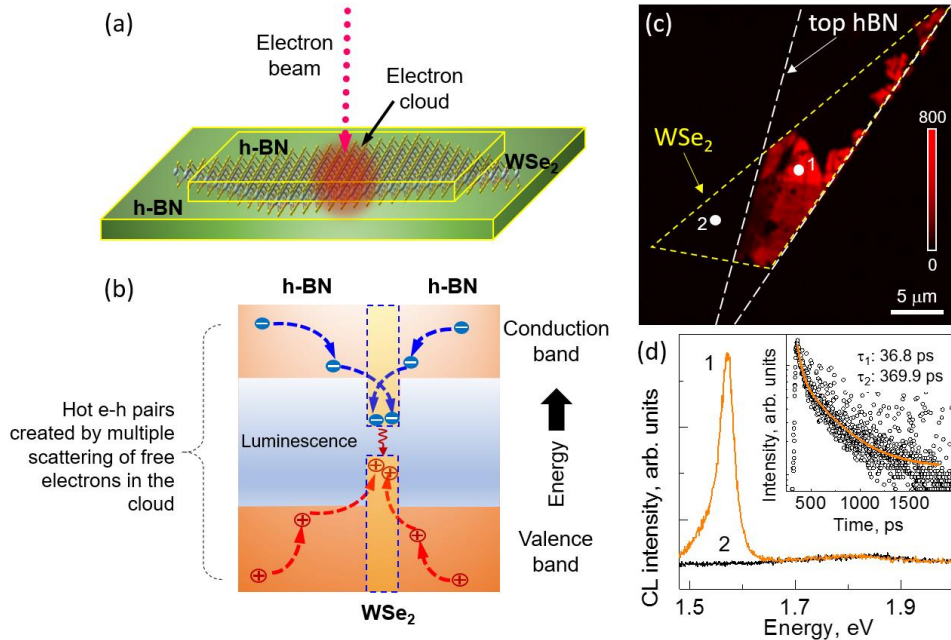


Fig. 1: (a) Illustration of cathodoluminescence in a heterostructure hBN/TMDC/hBN. (b) Process of the generation, diffusion, and recombination of electron-hole (e-h) pairs. The minor number of e-h pairs generated in the TMDC layer is ignored. (c) Monochromatic CL map of the heterostructure at WSe₂ emission energy, 1.66 eV (color coded in red) under 5 keV electron beam irradiation. (d) CL spectra generated by electron beam impact on WSe₂ at position 1 and 2 in (a). Inset shows time-resolved CL decay curve from the sandwiched WSe₂ with a double exponential fit.

Monolayer TMDCs such as MoS₂, WS₂ and WSe₂, are direct band gap semiconductors with large exciton binding energy, which presents them as promising building blocks for opto-electronic applications. It has been

assumed cathodoluminescence spectroscopy is not applicable to these monolayer materials due to the extremely small interaction volume within the monolayer. Here, we show that cathodoluminescence from monolayer TMDCs can be strongly enhanced by sandwiching them between layers of hBN with a wider band gap. The hBN/TMDC/hBN heterostructure can effectively increase the recombination probability of electron-hole (e-h) pairs in the monolayer TMDC in such a way that a good fraction of the electrons and holes generated in the hBN layers diffuse to and then radiatively recombine in the TMDC layer, leading to significant enhancement of the luminescence, comparatively to an isolated layer (see Fig. 1a,b).

Heterostructures of hBN/TMDC/hBN were prepared by stacking individual flakes with a dry transfer technique, where mechanically exfoliated flakes are picked up in sequence by vdW forces. CL measurements of prepared samples were performed in a scanning electron microscope (SEM) equipped with a dedicated CL detection system. Figure 1a shows the CL intensity map of the heterostructure hBN/WSe₂/hBN near 1.66 eV which corresponds to the exciton energy of monolayer WSe₂. Here, WSe₂ monolayer glows only if sandwiched by the two hBN layers, which confirms the quantum well model where the extra e-h pairs created in the adjacent hBN layers diffuse into, trapped and recombine in the monolayer TMDCs. The CL spectra from WSe₂ monolayer on (position 1) and off (position 2) the heterostructure is shown in Fig. 1b, where more than 500-times intensity enhancement is clearly observed. Cooling down the sample to cryogenic temperatures could reveal the trions in the monolayer as well. The enhanced luminescence was sufficient to perform time-resolved measurement as shown in the inset of Fig. 1b. We could demonstrate that CL spectroscopy can be applied to study the strain-induced excitonic peak shift in monolayer TMDCs on patterned substrates.

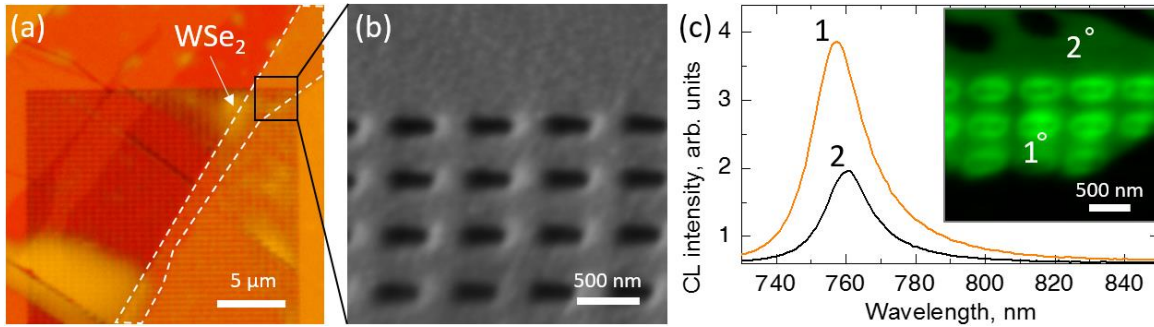


Fig. 2: (a) Optical image of a heterostructure hBN/WSe₂/hBN transferred onto a nano-slit array in 300nm-thick gold film. (b) SEM image of the highlighted area in (a). (c) CL spectra from WSe₂ monolayer on (1: orange) and off the nano-slit array (2: black) Inset shows monochromatic CL map at WSe₂ emission wavelength ($\lambda=756$ nm) acquired concurrently with the SEM image in (b).

CL spectroscopy can be employed to map out subwavelength optical modes in various nanophotonic systems thanks to the broadband excitation capability and high spatial resolution of focused electron beam probes. Here, we employ plasmonic nano-slit arrays to enhance the CL from monolayer TMDCs and clearly visualize the Purcell effect at subwavelength scale. A nano-slit array (slit length = 300 nm) was patterned with a focused ion beam in a 300 nm-thick gold film to have an absorption resonance near the excitonic emission peak of WSe₂. Figure 2a shows an optical image of a heterostructure hBN/WSe₂/hBN after it was transferred onto the nano-slit array with absorption resonance at 750 nm. The introduction of nano-slit arrays made the monolayer glow brighter near the edges of nano-slits where the mode density of the nano-slit array is higher (Fig. 2c inset). Figure 2c shows the CL spectra from WSe₂ monolayer on (position 1) and off (position 2) the patterned area, where the enhancement factor of up to 2.4 was found. Further, we will discuss on the effect of increased mode density on the lifetime of monolayer emission based on time-resolved CL measurement.