Enhanced second-harmonic generation from two-dimensional $MoSe_2$ by waveguide integration

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Abstract: We demonstrate enhanced second-harmonic generation from a monolayer $MoSe_2$ through Si waveguide integration. This is achieved by exciting the monolayer through the guided mode, which dramatically increases the interaction length and allows for phase-matching.

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Two-dimensional (2D) transition-metal dichalcogenides (TMDCs) with intrinsically-broken crystal inversion symmetry and large second order nonlinear response have shown great promise for future nonlinear light sources [1, 2] . However, the sub-nanometer thickness of such active materials limits their overall nonlinear conversion efficiency. In this work, we demonstrate, for the first time to our knowledge, enhanced second-harmonic generation (SHG) from 2D $MoSe_2$ through integrating it with Si waveguide, as schematically illustrated in Figure 1a.

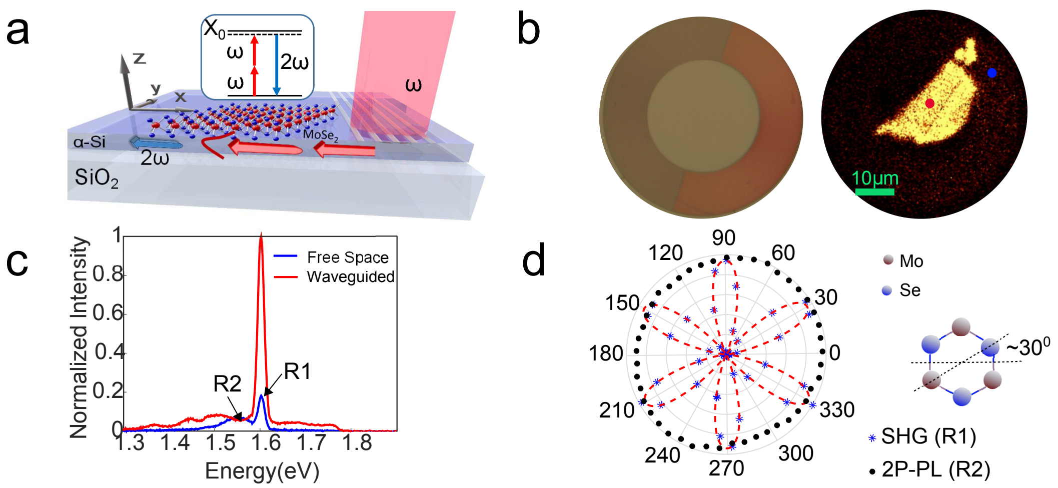

Fig. 1. (a) Schematic design of the hybrid integration of $MoSe_2$ onto a Si-waveguide. (b) Optical image (left) and 2D photoluminescence mapping (right) of the sample. (c) Emission spectrum when excited from grating (red) and free space (blue). (d) Co-polarized component of the signal varying with pumping polarization.

Light from free space is coupled into the waveguide by inscribed grating, and the evanescent field of the guided mode at the fundamental frequency (FF) of $\sim 1550\,$nm overlaps with the $MoSe_2$ material on top to generate second harmonic (SH). The generated SH can be guided and extracted out of the waveguide into free space. In practice, grating coupler of circular geometry is used in order to enable multiple angles of excitation and optimal coupling, the optical image (left) and photoluminescence mapping (right) of the sample are shown in Figure 1b, the bright piece inside the grating is a monolayer $MoSe_2$.

Under the excitation by the focused FF beam at around 0.8 eV, we observed approximately 5 times enhancement of the SH signal at 1.6 eV, when exciting the monolayer by the evanescent waveguide mode, as compared to pumping the $MoSe_2$ piece directly from free space, as seen in Figure 1c. We also measured how the co-polarized components of the emission depends on the polarization of the pump laser as shown in Figure 1d. The observed six-fold pattern for the main peak (red line) reflects the three-fold rotational symmetry of the crystal and identify it as SH signal [1], while the other weaker peaks are two-photo luminescence. Besides, we measured dependence of both the total and co-polarized signal of SH on pumping laser polarization from different positions: P1 and P2 on the grating, as shown in Figure 2a, P2 is on the armchair axis of $MoSe_2$ and P1 is at an angle of around 30° to it. The results displayed in Figure 2b show that total SH intensity for P1 and P2 have similar distribution patterns, while the distribution profiles for co-polarized component are quite different. Besides, we took snapshots of the SH emission as shown in Figure 2c. We could clearly see from that the SH signal is excited by guided mode and propogates in the waveguide, being subsequently coupled out from the grating on the left.

To further understand the experiments, we also perform theoretical calculation. Our calculation reveals that the SH enhancement is due to that the nonlinear interaction length is amplified by the waveguide geometry. The different distribution and intensity profiles shown in Figure 2b and c are because different guided modes of the SH are excited dominantly, when pumping from various positions relative to the armchair axis of the $MoSe_2$ crystal orientation, calculation results match well with experimental observation. In addition, the calculation shows we could achieve the exact phase-matching point between the waveguide mode of pumping and the SH signal by fine tuning the waveguide thickness, such phase-matching could provide further enhancement of the SH signal..

In conclusion, we have demonstrated the integration of TMDC monolayer on a silicon waveguide achieving quadratic nonlinear optics on a silicon photonics platform. We show 5 times enhancement of SHG from atomically thin monolayer $MoSe_2$, which proves that the light-matter interaction length of 2D TMDCs limited by monolayer thickness could be overcome by integration with waveguide. This paves the way for many other nonlinear applications for 2D materials in optical domain, including parametric amplification as well as generation of entangled photos.

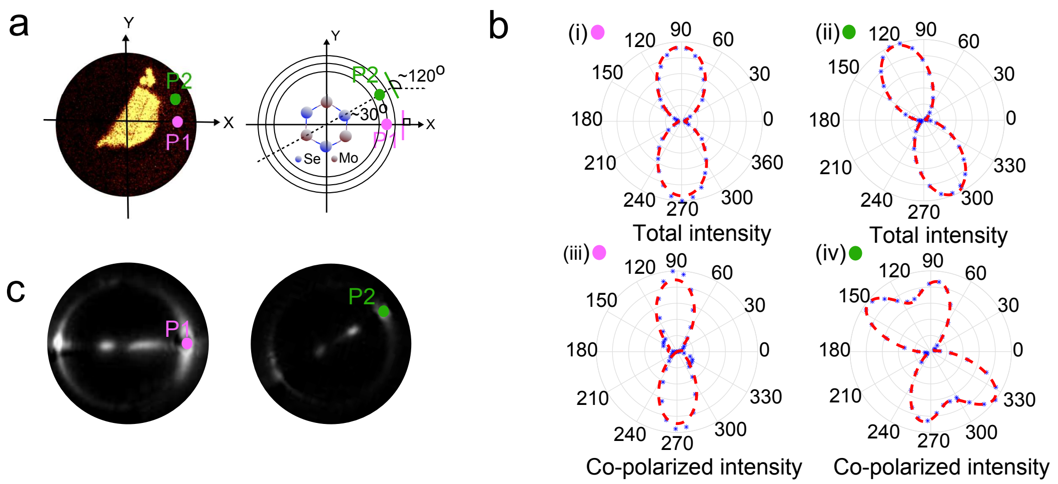

Fig. 2. (a) Map of the sample orientation relative to the polar coordinate system for both real sample (left) and schematic view (right). (b) Total and co-polarized SH emission intensity dependence on pumping laser polarization when focusing on position P1 and P2 respectively, the blue asterisks are the measured data and the dashed red lines are fitting lines. The angle refers to the polarization direction of the excitation laser retative to the X axis. (c) Snapshot images of the SH emission when pumping from P1 and P2 respectively.

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References

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2. C. Janisch, *et al.*, ”Extraordinary Second Harmonic Generation in tungsten disulfide monolayers”, Sci. Rep. **4**(2014).