Second-Order Optical Nonlinearity in MoS_2 Nanoislands from Density-Matrix Calculations

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Nonlinear optics has many applications in nanophotonics, such as frequency combs and optical modulators. The feasibility and efficiency of these applications are contingent upon nanoscale materials with a sufficiently large optical nonlinearity. As a result, there is ongoing research in the pursuit of nanomaterials and nanostructures with a high nonlinear optical susceptibility. The second-order optical susceptibility vanishes in most materials owing to their centrosymmetric structure. The synthesis of noncentrosymmetric nanostrucutres promises to enhance the optical nonlinearity. Following recent advances in the chemical vapor deposition processes [1], nanoscale equilateral-triangle-shaped islands of transition-metal dichalcogenides, such as MoS_2, WS2, and WSe2, can be accurately synthesized. Owing to the broken centrosymmetry in these nanotriangles, the second-order optical susceptibility is nonzero. Here, we employ the density-matrix approach to calculate the second-order optical susceptibility of MoS_2 nanotriangles at mid-infrared to near-infrared frequencies [2-4]. We show that the second-order optical susceptibility maximizes at plasmon resonances as well as two-photon (intraband) resonances. The two-photon resonances are only dependent on the size of the nanotriangles; however, the plasmon resonances can be tuned via the carrier density. We show that by aligning the plasmon resonances and two-photon resonances, the second-order optical susceptibility can increase up to 10^{-6} mV^{-1}.

[1] S. Xie et al., Science **359**, 1131 (2018).
[2] R. W. Boyd, *Nonlinear optics*, Elsevier (New York) (2008).
[3] F. Karimi, A.H. Davoody, and I. Knezevic, Physical Review B **93**, 205421 (2016).
[4] F. Karimi, A.H. Davoody, and I. Knezevic, Physical Review B **97**, 245403 (2018).

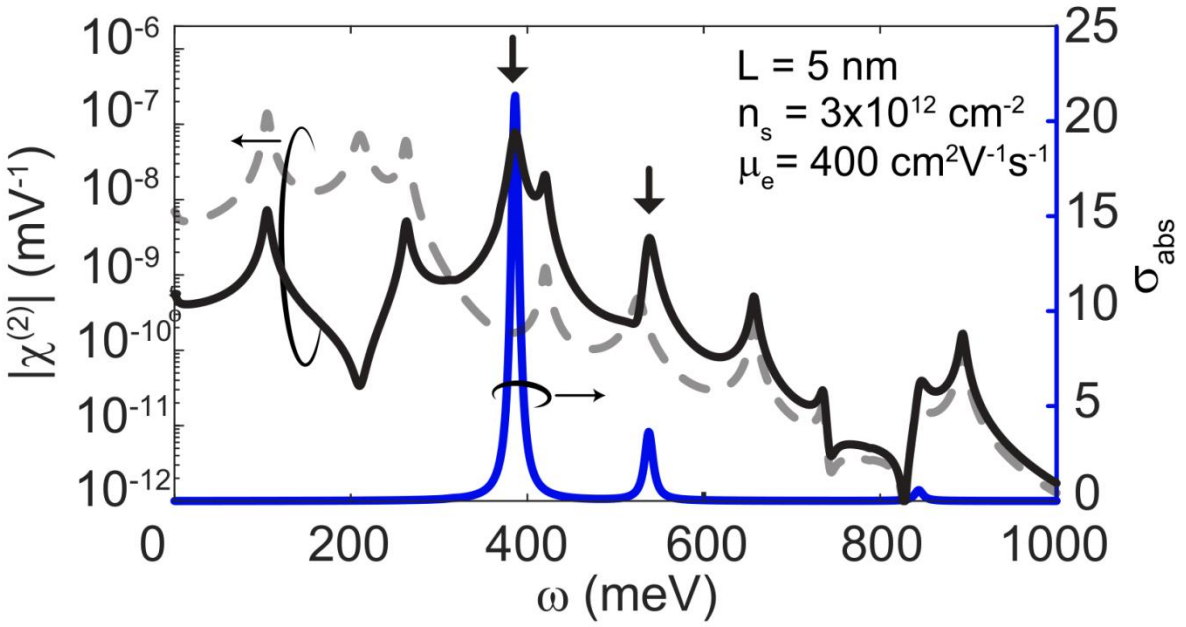

Fig.1: The second-order optical susceptibility with respect to the external field (solid black) and the self-consistent field (dashed black) for a nanotriangle with the side length of 5 nm, electron density of 3x10^{12} cm^{-2}, and electron mobility of 400 cm^{2}V^{-1}s^{-1}. The loss function is shown by the blue curve.

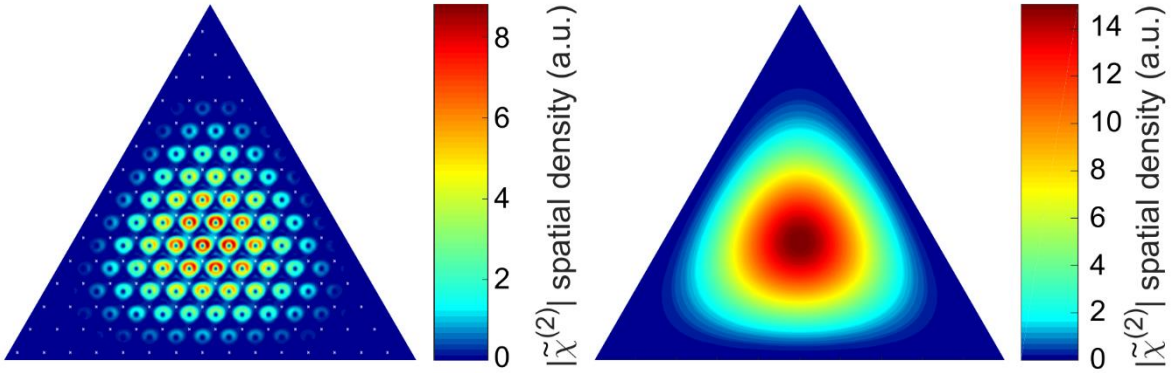

Fig.2: Spatial profile of the second-order optical susceptibility at the plane of Mo atoms and at the plasmon resonance frequency of 380 meV, calculated within the envelope function approximation with (left) and without (right) the Bloch wave function included. The nanotriangle has the side length of 5 nm, electron density of 3x10^{12} cm^{-2}, and electron mobility of 400 cm^{2}V^{-1}s^{-1}. The white crosses in the left figure denote the location of S atoms.