Enhanced Second-Harmonic Generation in Monolayer $\mathrm{MoS_2}$ Driven by a BIC-based Nonlinear Metasurface

J.T. Wang[1], **J.W. You**[1,2] **and Nicolae C. Panoiu**[1]

[1]University College London, Department of Electronic and Electrical Engineering, Torrington Place, WC1E 7JE, London, United Kingdom
[2]Southeast University, State Key Laboratory of Millimeter Waves, No. 2 Southeast University Road, Nanjing, 211189, China
jitong.wang@ucl.ac.uk

Abstract **– Dielectric metasurfaces have opened novel routes for nonlinear optics in recent years. In this work, we integrate a nonlinear metasurface with monolayer molybdenum disulfide ($\mathrm{MoS_2}$) to enhance second-harmonic generation (SHG) from atomically thin $\mathrm{MoS_2}$. By utilizing bound states in the continuum, we achieve about 600$\times$ of SHG enhancement from monolayer $\mathrm{MoS_2}$ on a resonant metasurface relative to suspended monolayer $\mathrm{MoS_2}$. Moreover, an eigenmode expansion approach is exploited to express second-harmonic power and the corresponding analytical results agree well with the rigorous calculations.**

I. Introduction

Optical metasurfaces have been studied widely in the context of nonlinear optics due to their ability to generate large local field enhancement. Recently, symmetry-breaking dielectric metasurfaces governed by bound states in the continuum (BICs) have opened up a new way for engineering optical resonances with large quality-factors (Q-factors) [1]. This promising approach makes metasurfaces a compelling platform to enhance light-matter interactions in atomically thin transition metal dichalcogenides (TMDs), which possess large second-order optical nonlinearity arising from their non-centrosymmetric structure [2].

In this work, we propose a nonlinear optical system for giant SHG from monolayer $\mathrm{MoS_2}$ on a silicon metasurface. The optical system that supports a symmetry-protected BIC is engineered to be asymmetric so that high-Q resonances can be achieved. In addition, we utilize a recently proposed theoretical approach based on an eigenmode expansion method to express the second-harmonic (SH) power generated from monolayer $\mathrm{MoS_2}$. The results derived from the analytical model agree with the conclusions based on rigorous numerical calculations.

II. Results and Discussion

A. Geometrical Configuration

As described in Fig. 1, our optical system consists of a cruciform silicon metasurface placed on a quartz substrate and a $\mathrm{MoS_2}$ monolayer placed on top of the silicon crosses. The period of the metasurface along both x and y directions is $\Lambda = 506\,\mathrm{nm}$. For each arm of the cruciform nanoresonator, the long and short sides are $316\,\mathrm{nm}$ and $126\,\mathrm{nm}$, respectively, and the depth is $122\,\mathrm{nm}$. The monolayer $\mathrm{MoS_2}$ has the same pattern with cruciform silicon and its thickness is $0.615\,\mathrm{nm}$. In the symmetric case, the centers of the two crossed arms coincide. To employ the BIC-inspired mechanism for enhancing the nonlinear optical response of the metasurface, one arm is shifted so as to break the in-plane inversion symmetry. Then, we can define an asymmetry parameter $s = \Delta a/a$ to quantify the asymmetry of the metasurface, where $a = 95\,\mathrm{nm}$ is the maximum shift distance. Impinging perpendicularly upon this system is an x-polarized plane wave, which acts as external excitation at the fundamental frequency (FF).

B. Linear Optical Response of a BIC-based Metasurface

We first investigate the angle-dependent transmission map and corresponding band diagram of the silicon metasurface under symmetric condition ($s = 0$). By comparing the two panels in Fig. 2a, the band structure of transverse

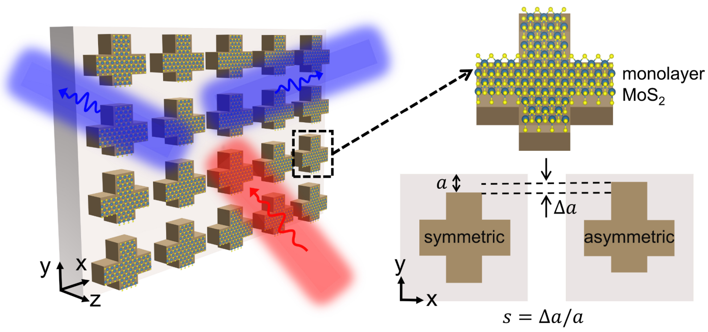

Fig. 1: Schematics of a nonlinear optical system for enhanced harmonic generation (left), design of a unit cell (upper right) and definition of a asymmetry parameter *s* (lower right).

electric (TE)-like modes shows consistent behavior with the transmission map. A BIC is clearly observed under TE-polarized wave at the Γ point with frequency of $345.8\,\text{THz}$. The resonance line-width of the BIC vanishes at the Γ point because of the symmetry incompatibility and increases gradually away from the Γ point.

The existence of a symmetry-protected BIC provides us with a simple approach to obtain high-Q resonances in the form of quasi-BIC. To show the dependence of the BIC on *s*, we sweep *s* from 0 to 1 under *x*-polarized plane-wave excitation. The results are given in Fig. 2b and the sharp resonances are characterized by the *Q* factor presented in Fig. 2c. The transmission spectra are fitted with Fano formula: $T_{\text{Fano}} = |a_1 + ia_2 + \frac{b}{\omega-\omega_0+i\gamma}|$, where a_1, a_2, and *B* are constant real parameters, ω_0 is resonant frequency, and γ represents the leakage rate of the resonance. Then, the *Q* factor can be calculated as $Q = \omega_0/2\gamma$ [3].

To gain deeper insights into the physical properties of the TE-like modes, we present the electric and magnetic field profiles determined at points A and B in Fig. 2d from eigenmode analysis. At point A, the electric and magnetic field patterns of the BIC state possess in-plane inversion symmetry, indicating the complete decoupling of the BIC from the normally incident plane wave. In contrast, the quasi-BIC (point B) reveals asymmetric field distributions and therefore manifests itself in transmission spectra as sharp resonances with high Q factor.

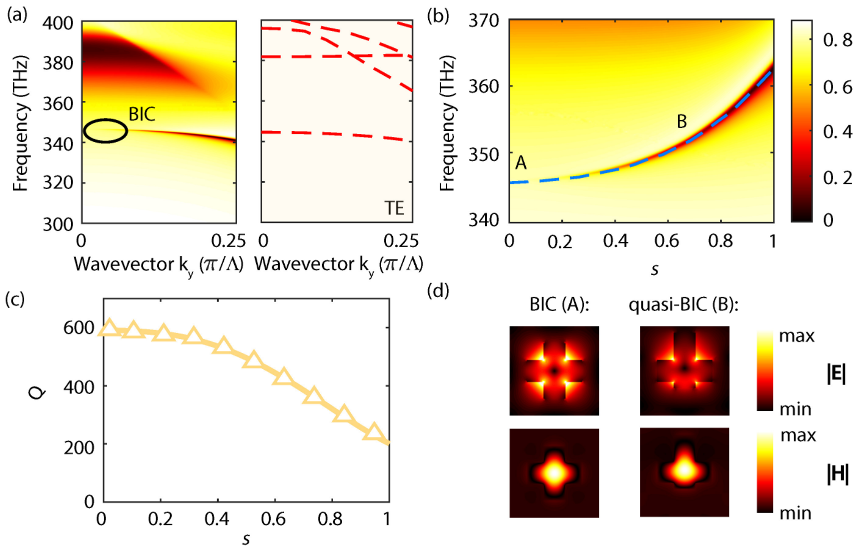

Fig. 2: (a) Angle dependence of transmission map (left) and corresponding eigenmode analysis (right). (b) Transmission spectra vs. s. The blue dashed line indicates the resonant frequency, and letters A and B indicate BIC ($s = 0$) and quasi-BIC ($s = 0.63$) states, respectively. (c) Evolution of Q factor with s. (d) In-plane (*xy* plane) electric and magnetic filed patterns of TE-like eigenmodes computed at the two s points A and B.

C. Giant Second-Harmonic Generation from Monolayer $\mathrm{MoS_2}$

The monolayer $\mathrm{MoS_2}$ generates enhanced second-order nonlinear optical response due to the BIC mechanism. In Fig. 3a, we notice that SHG from monolayer $\mathrm{MoS_2}$ on the resonant metasurface is about $600\times$ larger than that from suspended monolayer $\mathrm{MoS_2}$, appearing at $s = 0.63$ with frequency of 699 THz. Moreover, it is of particular interest to explore the underlying relation between s and SH intensity. Thus, we use a recently proposed eigenmode expansion method to study the nonlinear SHG process. The main idea is to find the induced field using Green's functions that are expanded in eigenfunctions of an optical resonance system. At the FF, linear optical response is supposed to be governed by a resonant state, E_1, while near the SH a single resonant state E_2 dominates the field generated by nonlinear polarization sources. Then, the SH power $P_{SH}(2\omega)$ is determined as explained in [4]:

$$P_{SH}(2\omega) = [P(\omega)\kappa_1(\omega)Q_1 L_1(\omega)]^2 \kappa_{12} Q_2 L_2(2\omega)\kappa_2\alpha(2\omega). \tag{1}$$

Here, $P(\omega)$ is total incident power, $\kappa_1(\omega)$ is the coupling between excitation and E_1, κ_2 is the out-coupling coefficient, κ_{12} is the overlap factor between the modes E_1 and E_2, Q_j and L_j, $j = 1, 2$, are Q factor and spectral mismatch, respectively, and $\alpha(2\omega)$ is smooth envelope factor.

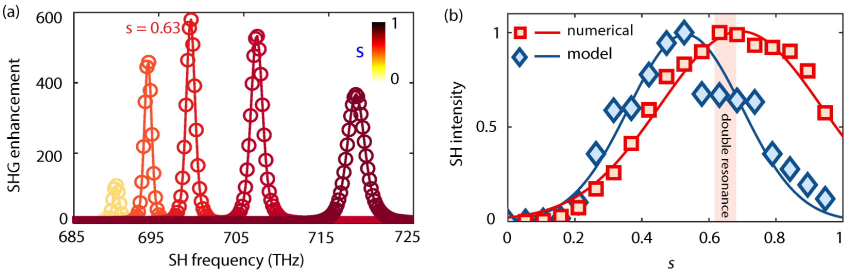

Fig. 3: Harmonic generation from monolayer $\mathrm{MoS_2}$. (a) s-dependent SHG enhancement with maximal enhancement at $s = 0.63$. (b) Comparison of normalized SH intensity determined using numerical and analytical methods.

The results of analytical model from (1) are compared with those of rigorous numerical calculations, as shown in Fig. 3b. The two methods show a maximum up-conversion efficiency at $s = 0.63$ and $s = 0.53$, respectively. The difference in s is explained by the assumption of single-mode field expansion in the analytical model. A double-resonance phenomenon at $s = 0.64$ is also observed, which further enhances SHG.

III. Conclusion

In this contribution, a nearly 600 times enhancement of SH intensity is shown from monolayer $\mathrm{MoS_2}$ in a BIC-based metasurface relative to suspended monolayer $\mathrm{MoS_2}$. In our quantitative analysis of this phenomenon, we employ an eigenmode expansion method to obtain the SH power. Our design suggests a promising way for integrated TMD-based nanodevices in photonic applications.

Acknowledgement

This work was supported by the European Research Council (ERC) (ERC-2014-CoG-648328).

References

[1] K. Koshelev, S. Lepeshov, M. Liu, A. Bogdanov, and Y. Kivshar, "Asymmetric Metasurfaces with High-Q Resonances Governed by Bound States in the Continuum," *Physical Review Letters,* vol. 121, art. no. 193903, 2018.

[2] M. Weismann and N. C. Panoiu, "Theoretical and computational analysis of second- and third-harmonic generation in periodically patterned graphene and transition-metal dichalcogenide monolayers," *Physical Review B,* vol. 94, art. no. 035435, 2016.

[3] Y. Yang, I. I. Kravchenko, D. P. Briggs, and J. Valentine, "All-dielectric metasurface analogue of electromagnetically induced transparency," *Nature Communications,* vol. 5, p. 5753, 2014.

[4] K. Koshelev, S. Kruk, E. Melik-Gaykazyan, J.-H. Choi, A. Bogdanov, H.-G. Park, and Y. Kivshar, "Subwavelength dielectric resonators for nonlinear nanophotonics," *Science,* vol. 367, p. 288, 2020.