

Dual-resonant biphoton generation in 3R-MoS₂ van der Waals membrane metasurfaces

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We propose a general and practical strategy for designing membrane van der Waals (vdW) metasurfaces composed of two distinct hole arrays and supporting a pair of high-order multipole modes at the pump wavelength and spontaneous parametric down-conversion wavelength at once. The resonance excitation of nonradiating components of electric octupole at the pump wavelength allows one to directly concentrate near-field energy within the nonlinear 3R-MoS₂ metasurface. Tuning of the second membrane metasurface resonance for concentration of near field around the holes entails the enhancement of vacuum fluctuations and spontaneous emission of photons from them. This dual-resonance architecture initiates the formation of nonclassical correlations and violation of the Cauchy-Schwarz inequality on reduced times relative to both an unpatterned flake and an off-resonant membrane configuration. The presented approach predicts a biphoton generation rate of 61 Hz/mW and a frequency bandwidth of 11.5 THz, while retaining the transferability and materials flexibility of the vdW membrane platform and offering a scalable pathway toward faster quantum encryption and computing technologies.

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Recently, van der Waals materials have attracted close attention from researchers and have been considered as a new material basis for modern and future nanophotonics [1,2]. The main expectations from these layered materials are related to the record values of refractive indices, optical anisotropy, and high quantum yield [3–6]. Quite recently, it has been discovered that their nonlinear characteristics may also be record-breaking for a certain type of stacking of monolayers. So, 3R polytypes of transition metal dichalcogenides (TMDs) with the chemical formula MX₂ (M represents a transition metal atom and X denotes a chalcogen atom) possess the unique symmetry properties, which cause the high second-order nonlinearity for their thin stacks [7–10]. Unlike the more common 2H polytype, which features an ABA stacking sequence that restores spatial inversion symmetry for any even number of layers (yielding a strictly zero macroscopic $\chi^{(2)}$ response), the 3R polytype features an ABC stacking sequence. Consequently, the 3R phase intrinsically lacks an inversion center regardless of the layer number. This unique crystallographic property enables the in-plane nonlinear dipoles to remain parallel, allowing the nonlinear response to scale constructively with the thickness of the stack. Moreover, their

second-order nonlinear susceptibility tensor provides the possibility of direct generation of maximally entangled signal and idler photons [11–15] in the process of spontaneous parametric down-conversion (SPDC) [see Fig. 1(a)]. The possibility of biphoton generation from 3R-MoS₂ flake placed on quartz substrate with simple polarization control was demonstrated for the first time in Ref. [16]. At the same time, the low rate of biphoton generation and the experimentally observed width of the spectrum limit the possibilities of practical use of the achieved results [17]. The solution of this problem can be the realization of quasi-phase-matching in a stack of flakes for which the symmetry axes are arranged with the periodically changed rotation angle [16], which was implemented in Ref. [18]. Another way to increase the brightness of a source is the integration of nonlinear material with resonators in order to increase the length of interaction with the nonlinear medium [19,20]. In this case, the use of a double-resonant distributed Bragg reflector cavity with resonances at both the pump wavelength and the signal wavelength [21] can allow to increase the spectral brightness of the source. The application of the dual-resonant biphoton generation strategy with conventional periodically poled Brillouin zone folding metasurfaces allowed one to obtain the calculated biphoton rate of about 100 MHz/mW with a frequency bandwidth of 20.5 THz [22].

The problem of obtaining a broadband source of biphotons can be solved by using distributed resonator systems in the form of photonic crystal fibers [23]. Finally, it has been recently shown [24] that metasurfaces composed of conventional nonlinear materials and supporting bound states in the continuum can simultaneously solve the problems of

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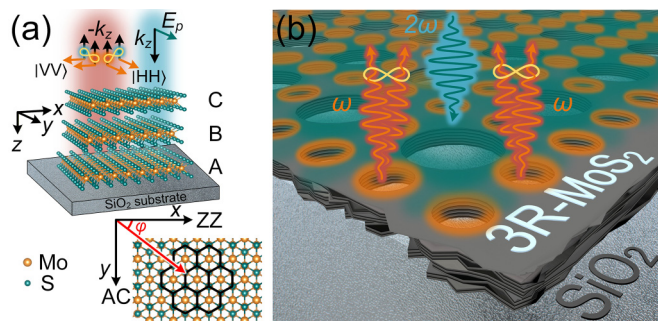


FIG. 1. (a) Artistic picture illustrating the generation of entangled photons in 3R-MoS₂ stack. Top sketch: Three-layer 3R-MoS₂ flake with ABC-stacking generating polarization-entangled signal and idler photons $|HH\rangle$ and $|VV\rangle$ through the SPDC in the regime of reflection from a perfect reflector. Bottom sketch: Atomic structure of 3R-MoS₂ stack with depicted lattice cells and zigzag (ZZ) and armchair (AC) directions. (b) The model of double membrane metasurface in the form of 3R-MoS₂ film perforated with a periodic array of through holes with two different diameters and placed on quartz substrate.

increasing the brightness of photonic states and controlling their spectrum.

At the same time, along with outstanding nonlinear properties, the layered TMDs have high refractive index values for the in-plane component and optical anisotropy in conjunction with almost zero losses in the infrared region. This fact allows the creation of both individual highly efficient Mie resonators [7] and metasurfaces [25] directly based on 3R-MX₂ TMDs. The arrangement of MX₂ material layers in the metasurface plane additionally localizes the quasitrapped modes (QTMs) and increases their quality factor when choosing the appropriate settings [26,27] with an extremely high concentration of electromagnetic fields in the near-field region. The combination of these resonant near fields with the high nonlinearity of the 3R-MX₂ can provide unprecedented capabilities for increasing the efficiency and control of biphoton generation.

In this Letter, unlike previous approaches, to improve the efficiency of the nonlinear response, we propose using membrane metasurfaces, which represent a periodic structure of holes in a thin layer of the 3R-polytype van der Waals material. In this context, we present a general practical strategy for implementing efficient generation of entangled photons using the nonlinear properties of such metasurfaces. The proposed membrane metasurfaces are obtained by perforating the 3R-MX₂ flake with holes of two different radii [see Fig. 1(b)]. The key idea is to find such eigenmodes of the system that correspond to nonradiating or weakly radiating distributions of displacement currents causing the high concentration of near-field energy in the plane of membrane metasurface. The excitation of one mode at the pump wavelength allows the significant increase in its efficiency simultaneously changing the effects of reflection (scattering) for the structure and concentrating the near-field energy within the nonlinear material. Tuning of the second eigenmode to the wavelength being two times greater than original is intended to intensify spontaneous photon emission by increasing the local density of states and enhancing the vacuum fluctuations; see algorithm

for tuning of the system in Sec. S1.1 of the Supplemental Material [28]. The near-field behavior of such modes can be related with contributions of nonradiating high-order multipole moments excited due to multipole coupling in a lattice [26]. Therefore, to control and tune the corresponding modes of the membrane metasurface, we will extensively use the multipole analysis [29–31]. In this method, the transmission and reflection properties of a metasurface are characterized by a superposition of fields generated by the multipole moments of its unit cell, so metasurface's resonant response can be analyzed in terms of the resonant contribution of certain multipole moments [32,33] (see Secs. S1.2 and S1.3 in the Supplemental Material [28]). The optimal choice of unit cell and the conditions for application of the multipole approach (in the case of membrane metasurfaces) are described in Ref. [34]. Although membrane metasurfaces can operate in a homogeneous air environment, in our case, from the beginning, we will assume that the metasurface is located on a SiO₂ substrate. This approach is justified by the fact that, from a practical point of view, the fabrication of the metasurfaces discussed below involves the initial stage of exfoliation of a thin plate from a nonlinear 3R-MoS₂ crystal and its transfer to a solid substrate, similar to other van der Waals materials [35,36].

The proposed strategy can be divided into two main stages: The first involves pretuning to determine the periods of the metasurfaces, and the second provides fine-tuning to the required resonant wavelengths.

The first stage (pretuning). We start by numerical tuning the optical response of a pair of independent membrane metasurfaces in the form of 3R-MoS₂ films of equal thicknesses $H = 100$ nm perforated with circular holes. The criterion for our search is the resonant excitation of nonradiating (from the metasurface plane) component O_{xy} of the electric octupole, as in Ref. [26], at different wavelengths λ_1 and λ_2 in the corresponding metasurfaces. An important condition is the multiplicity of these wavelengths, namely, $\lambda_2 = 2\lambda_1$. Since the excitation of eigenmodes leads to modulation of the reflectivity, each of the membrane metasurfaces must maintain resonant reflection characteristics at the selected wavelengths while simultaneously enhancing the near fields in the membranes. An additional requirement for these two metasurfaces is that their periods also should be multiples of each other, for example, $P_2 = 2P_1$, where $P_{1(2)}$ is the period of the first (second) metasurface. This condition will help us in the second stage to obtain a metasurface that has two sets of holes and supports two resonances at multiple wavelengths.

To accomplish our goal, we fixed the periods $P_1 = 645$ nm and $P_2 = 2P_1 = 1290$ nm for a pair of independent 3R-MoS₂ metasurfaces placed on quartz substrates and varied the hole radii R_1 and R_2 in order to satisfy the condition $\lambda_2 = 2\lambda_1$. To speed up the search, we calculated the contour plots for reflection coefficient R depending on the wavelength λ and radius R_d (R_1 or R_2) [see Figs. 2(a) and 2(b)]. The spectral dependences for the refractive index and extinction coefficient for the 3R-MoS₂ material are taken from Refs. [8,37]. The region of sharp change/modulation of the reflection coefficient appearing in Figs. 2(a) and 2(b) can be associated with the excitation of the metasurface's eigenmodes. The remarkable peculiarity of the emerging feature is that it arises at the wavelength $\lambda_1 = 1012$ nm for the radius $R_1 = 182$ nm, whereas

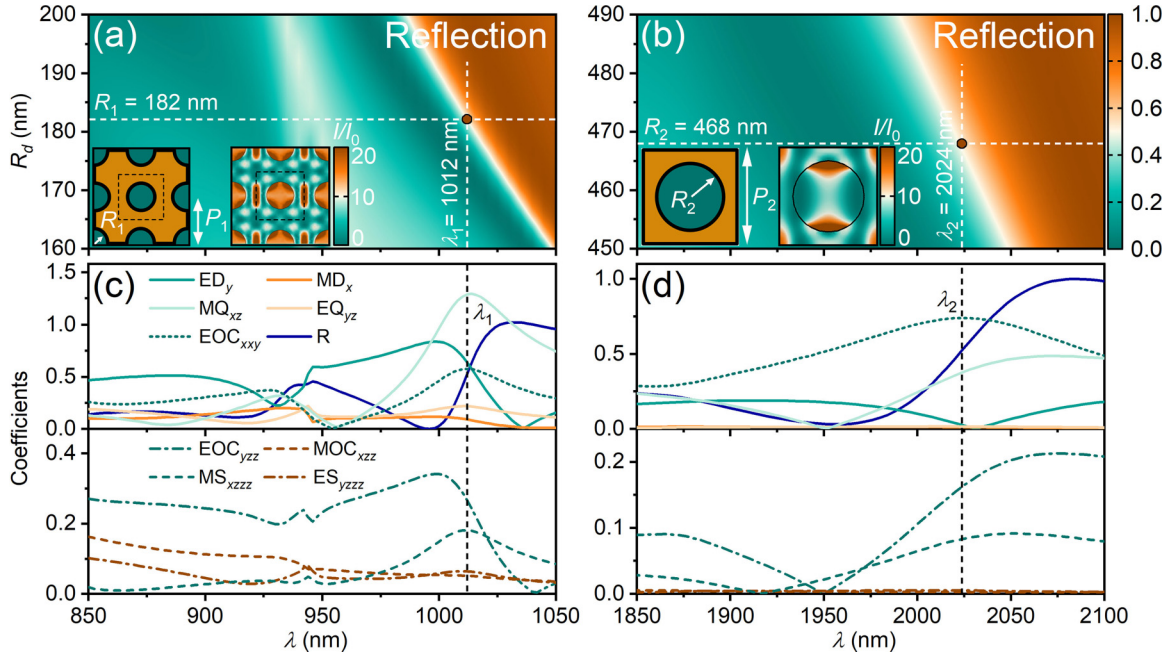


FIG. 2. (a), (b) Contour plot for the reflection coefficient R on the parametric plane (λ ; R_d) for a pair of independent membrane metasurfaces, i.e., 3R-MoS₂ films of thickness $H = 100$ nm placed on a quartz substrate and perforated with hole arrays with periods (a) $P_1 = 645$ nm and (b) $P_2 = 2P_1 = 1290$ nm. (c), (d) Coefficients for the contributions of various multipole moments to the reflection, as well as the normalized nonradiating component EOC_{xxy} , calculated for (c) $R_d = R_1 = 182$ nm and (d) $R_d = R_2 = 468$ nm for which the spectral features are realized at the wavelengths $\lambda_1 = 1012$ nm and $\lambda_2 = 2024$ nm, respectively. The metasurfaces are excited by the electromagnetic wave $E_y(k_z)$. Unit cell for the integration of fields coincides with the left inset in panel (b) and indicated by dashed square in the insets in panel (a). The right insets in panels (a) and (b) show the intensity distribution of electric field.

the choosing of radius $R_2 = 468$ nm shifts this feature to the wavelength $\lambda_2 = 2024$ nm being integer multiple of λ_1 .

To analyze the multipole composition of the considered modes, we choose the unit cells, shown in the insets in Figs. 2(a) and 2(b), as integration regions, centers of which coincide with the centers of holes in membrane, and their height is equal to the film thickness. The lateral size of the unit cells is equal to the period. Approximate expression for the reflection coefficient of a metasurface can be written in the following form [34]:

$$R \cong \left| K \left(p_y + \frac{m_x}{v_d} + \frac{ik_d}{6} Q_{yz} + \frac{ik_d}{2v_d} M_{xz} - \frac{k_d^2}{6} O_{yz} - \frac{k_d^2}{6v_d} MO_{xzz} - \frac{ik_d^3}{24} S_{yz}^{(e)} - \frac{ik_d^3}{24v_d} S_{xzz}^{(m)} \right) \right|^2 \quad (1)$$

corresponding to the square of the linear superposition of multipole contributions with absolute values $ED_y = |Kp_y|$, $MD_x = |Km_x/v_d|$, $EQ_{yz} = |KQ_{yz}ik_d/6|$, $MQ_{xz} = |KM_{xz}ik_d/(2v_d)|$, $EOC_{yz} = |KO_{yz}k_d^2/6|$, $MOC_{xzz} = |KMO_{xzz}k_d^2/(6v_d)|$, $ES_{yz} = |KS_{yz}^{(e)}ik_d^3/24|$, and $MS_{xzz} = |KS_{xzz}^{(m)}ik_d^3/(24v_d)|$ expressed through the corresponding components of electric p_y and magnetic m_x dipoles, electric Q_{yz} and magnetic M_{xz} quadrupole moments, electric O_{yz} and magnetic MO_{xzz} octupole moments, and electric $S_{yz}^{(e)}$ and magnetic $S_{xzz}^{(m)}$ 16-pole moments of the unit cell. Here, $K = ik_d/(E_y^0 2S_L \epsilon_0 \epsilon_d)$, $S_L = P^2$ is the area of a lattice unit cell, P is the lattice period, $k_d = k_0 \sqrt{\epsilon_d}$ and $v_d = 1/\sqrt{\epsilon_0 \mu_0 \epsilon_d}$ are the wave number and speed of light in the surrounding

medium with relative permittivity ϵ_d , respectively, k_0 is the wave number in the vacuum, E_y^0 is the electric field of the incident wave in the metasurface plane, ϵ_0 is the vacuum permittivity, and μ_0 is the vacuum permeability. Integral definitions of multipole moments are given in Sec. S1.2 of the Supplemental Material [28]. It should be noted that Eq. (1) is written for a homogeneous membrane environment. In our case, we use it in the effective medium approximation, where the effective permittivity of the medium outside the membrane can be written as follows: $\epsilon_d = [(1 + n_{\text{sub}})/2]^2$, where n_{sub} is the spectral dependence of the refractive index of the SiO₂ material. The applicability of this approach is discussed in Sec. S1.4 of the Supplemental Material [28]. We also note that high-order multipoles, including 16-poles, significantly contribute to the expression (1) for membrane metasurfaces, i.e., the multipole sum (1) is in poor agreement with the numerically calculated curve for R without taking them into account (see Fig. S1.2 in the Supplemental Material [28]).

Note that Eq. (1) includes only those multipole components that directly emit electromagnetic waves from the metasurface plane. However, due to electromagnetic coupling between unit cells, the multipole decomposition of displacement currents in a single unit cell can include multipole components that do not emit electromagnetic waves from the metasurface plane. If these components belong to a resonant mode, the energy corresponding to these components accumulates in the metasurface as strong near fields. In particular, as follows from the multipole decomposition shown in Fig. 2 and Eq. (1), such a nonradiating multipole is an electric octupole, which is

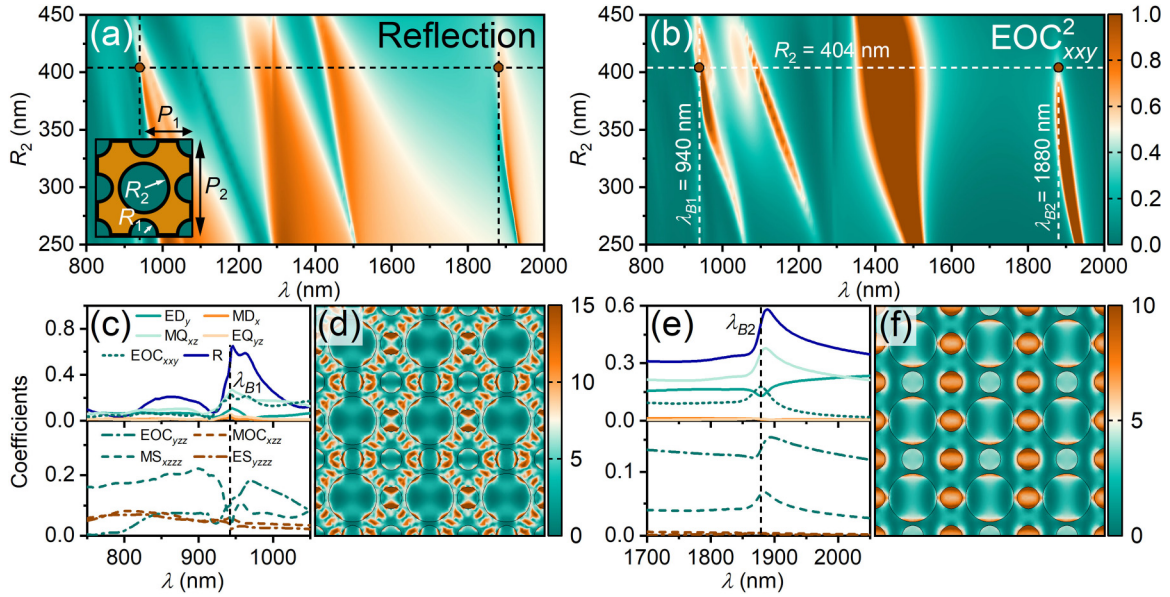


FIG. 3. (a), (b) Contour plot for (a) reflection coefficient R and (b) multipole coefficient EOC_{xxy}^2 on parametric plane (λ ; R_2) for double metasurface, i.e., 3R-MoS₂ film of thickness $H = 100$ nm placed on a quartz substrate and perforated with hole arrays possessing hole radii $R_1 = 182$ nm and R_2 and fixed periods $P_1 = 645$ nm and $P_2 = 2P_1 = 1290$ nm. (c), (e) Coefficients for the contributions of various multipole moments to the reflection coefficient R , as well as the normalized value EOC_{xxy} of nonradiating component, calculated for the optimized radius $R_2 = 404$ nm for which QTMMs are formed at the wavelengths $\lambda_{B1} = 940$ nm and $\lambda_{B2} = 1880$ nm, respectively. (d), (f) Distributions of intensity for the electric component of near field in a metasurface at the wavelengths (d) $\lambda_{B1} = 940$ nm and (f) $\lambda_{B2} = 1880$ nm calculated inside metasurface at the distance $H/2$ from its top surface. The metasurface is excited by the electromagnetic wave $E_y(k_z)$. Unit cell for the integration of fields and the calculation of the multipole moments coincides with the inset in panel (a).

formally consistent with the case of a quasitrapped octupole mode of a conventional metasurface composed of cylindrical resonators [26,38] (see Sec. S1.3 in the Supplemental Material [28]). Indeed, from Figs. 2(c) and 2(d) it is evident that the nonradiating component $\text{EOC}_{xxy} \sim O_{xxy}$ of the electric octupole moment is resonantly excited at $\lambda_1 = 1012$ nm for the first metasurface with $P_1 = 645$ nm, $R_1 = 182$ nm and at $\lambda_2 = 2024$ nm for the second metasurface with $P_2 = 1290$ nm, $R_2 = 468$ nm. As a result, for these parameters we obtain a strong concentration of near fields in metasurfaces [see insets in Figs. 2(a) and 2(b)], which can be identified as the excitation of high-order quasitrapped modes of the membrane metasurface (QTMMs). This completes the realization of the first stage.

The second stage (fine-tuning). Now, we combined two independent arrays of holes obtained in the first stage into single membrane metasurface as shown in Fig. 1(b). Such a membrane metasurface is not an exact superimposition of two metasurfaces in Fig. 2, but corresponds to the replacement of some small holes in Fig. 2(a) by large ones in Fig. 2(b) while retaining the periodicities. As a result, we obtained a hybrid metasurface supporting resonant modes at the wavelengths close to λ_1 and λ_2 . After that, we performed the additional fine-tuning of hole sizes in order to obtain the exact condition $\lambda_{B2} = 2\lambda_{B1}$ for the wavelength of these QTMMs in such a hybrid membrane metasurface. For this, we calculated contour plots for the reflection coefficient and coefficient $\text{EOC}_{xxy} = |KO_{xxy}k_d^2/6|$, corresponding to the nonradiating component O_{xxy} of electric octupole normalized in a similar way to the contributions of radiating components to the reflection coefficient (1) [see Figs. 3(a) and 3(b)], depending on the

wavelength and radius R_2 of large holes for a fixed radius $R_1 = 182$ nm of small holes. The value of R_2 in this case is considered as a fine-tuning parameter. The red dots in Figs. 3(a) and 3(b) corresponds to the excitation of a resonance pair at the wavelengths $\lambda_{B1} = 940$ nm and $\lambda_{B2} = 1880$ nm, being integer multiples of each other, for the hybrid metasurface with $R_2 = 404$ nm. The change in the values of the resonant wavelengths in the hybrid metasurface compared to the case of two independent metasurfaces from the first stage is associated with a change in the configuration of the unit cell of this tuned metasurface [see inset in Fig. 3(a)]. It should be noted that the used approach with sequential optimization, i.e., optimization of ordinary metasurfaces (see Fig. 2), followed by optimization of a double membrane metasurface combined from them, allowed us to save a significant amount of computational resources when obtaining the required parameters for the regime in Fig. 3.

Resonant excitation of a nonradiating component O_{xxy} of electric octupole at the wavelengths λ_{B1} and λ_{B2} in Figs. 3(c) and 3(e), respectively, indicates the retention of the same type of modes as in Fig. 2, whose multipole compositions comprise the components p_y and M_{xz} (see also Sec. S1.5 and Figs. S1.3–S1.5 in the Supplemental Material [28]). However, their relative contributions are changed compared to the cases shown in Figs. 2(c) and 2(d). Moreover, the pattern of near-field energy distribution in the double membrane metasurface in Figs. 3(d) and 3(f) qualitatively changes compared to the case of two separate metasurfaces in Fig. 2. In particular, the maximum enhancement of near field at the wavelength λ_{B1} is realized directly inside the material [see Fig. 3(d)], fulfilling the whole potential of near-field pumping at this

wavelength. On the contrary, the significant enhancement of the near field near and inside the small holes at the wavelength λ_{B2} in Fig. 3(f) should result in the increase in the local density of states and enhancement of spontaneous emission through these holes. The advantage of this geometry is the possibility of nondissipative emission of biphotons not only from the near-surface layers of the flat material [24], but also from each layer of van der Waals stack perforated with a hole. The quality factor of the corresponding resonances for the reflection R is 21 at the wavelength λ_{B1} and 30 at the wavelength λ_{B2} . Thus, the use of pumping at the wavelength λ_{B1} should lead to the high-rate generation of biphotons with a required spectral distribution near the wavelength λ_{B2} .

We also note that there is a fundamental difference between optical membrane metasurfaces considered here and well-known Mie voids [39]. In the latter, the localization of the near field mainly occurs inside holes being cut on the surface of the optically thick material. It is almost impossible to obtain field localization inside material due to the presence of actually open boundary leading to the leakage. The membrane metasurfaces offer the opportunity for strong localization of near fields both inside the holes, which is similar to Mie voids, and directly within the film material. The fabrication of such membrane metasurfaces directly from the functional materials paves the way for their use in lasing, second harmonic generation, SPDC, and other applications.

Biphoton generation. The entangled photon pairs in the SPDC process are usually generated in two different continuous modes, for which the polarizations of signal and idler photons can be either collinear or orthogonal [12,40,41]. The monolayer of the considered MoS₂ material belongs to the D_{3h} symmetry group, which determines the following relationships for the nonzero elements of the nonlinear tensor $\chi_{yyy}^{(2)} = -\chi_{yxx}^{(2)} = -\chi_{xyx}^{(2)} = -\chi_{xyx}^{(2)}$ when choosing the x axis along zigzag direction and y axis along armchair direction in the lattice [16,42] [see Fig. 1(a)]. Here, we focus on a specific case where the pump wave $E_y^{(p)}(k_z)$ results in generation of maximally entangled (Bell) state $|\Phi^-\rangle = 1/\sqrt{2}(|HH\rangle - |VV\rangle)$ for a pair of correlated photons with collinear polarizations $|HH\rangle$ or $|VV\rangle$ [see Fig. 1(a)]. Moving on to the multilayer MoS₂ stack, it should be noted that its 3R polytype not only exhibits strong second-order nonlinearity but also advantageously differs from other phases in terms of nonclassical light state generation. The ABC-ABC stacking order that is inherent to this polytype [43] without inversion center is intrinsic with respect to the generation of entangled photons along the z axis due to the action of in-plane components of nonlinear tensor [16]. The out-of-plane nonlinear tensor components for such a thick flake can also lead to the generation of entangled states, but their contribution is not so considerable, and the emission pattern has a large angular divergence [16].

Despite the ideal entanglement of photons in the case of unstructured 3R-MoS₂ flake [16], the reason for our in-depth analysis is to find a strategy for control of the generation efficiency and spectra of such states using metasurfaces as shown in Fig. 1(b) (see Sec. S2 in the Supplemental Material [28]). For this purpose, we use the second-order autocorrelation $g_{i(s)}^{(2)}(t, \tau)$ and cross-correlation $G_{si}^{(2)}(t, \tau)$ functions [44–47]

depending on interaction time t and time delay τ between counts, as well as the parameter of nonclassical light C that is the left side of Cauchy-Schwarz inequality (see Sec. S3 in the Supplemental Material [28]):

$$C \equiv \frac{[G_{si}^{(2)}(t, \tau)]^2}{g_s^{(2)}(t, \tau)g_i^{(2)}(t, \tau)} \leq 1. \quad (2)$$

Essentially, the photons generated in the stochastic SPDC process have thermal statistics; i.e., each mode satisfies the relation $g_{i(s)}^{(2)}(t, 0) \simeq 2$. At the same time, the combination of near-field intensity and interaction time in a nonlinear medium can lead to the changes in values of $g_{i(s)}^{(2)}(t, \tau)$, $G_{si}^{(2)}(t, \tau)$, and C [48]. In this case, the key, albeit indirect, indication of the strong nonclassical correlations for photon pairs is the violation of the Cauchy-Schwarz inequality, i.e., the satisfaction of the condition $C > 1$ [44,45,49,50].

Describing and comparing various regimes of light-matter interaction (see Sec. S4 and Figs. S4.1, S4.7, and S4.13 in the Supplemental Material [28]), we observe that an increase in the near-field intensity up to maximal value under resonant conditions for the double metasurface shown in Fig. 3 allows the increase in the photon generation rate compared to the cases of both nonperforated flake (compare with the case in Ref. [16]) and considered metasurface but under nonresonant conditions; see the comparative dynamics of the growth of the average number of photons $n_s = \langle b_{s(i)}^+(t)b_{s(i)}(t) \rangle$ in Fig. 4(a), where $b_{s(i)}^+(t)$ and $b_{s(i)}(t)$ are the creation and annihilation photon operators (see Secs. S2 and S3 in the Supplemental Material [28]), respectively. At the same time, the decrease in the function $g_{i(s)}^{(2)}(t, \tau)$ below the unit value at $\tau = 0$ s in Fig. 4(b) corresponds to the single-photon regime in the time interval $t \in (33; 116)$ ps. Finally, the achievement of the maximum for the parameter C in Fig. 4(a) indicates the optimal conditions [51] for the generation of nonclassical biphoton pairs at the time $t_{\max} = 60$ ps. Shift in the wavelength of detected idler and signal photons leads to some increase in this optimal interaction time, but without destroying the regime in Fig. 4(a). It should be noted that the maximum value for parameter C is resulted not so much from the cross-correlation function, which takes the value $G_{si}^{(2)}(t_{\max}, 0) = 2.98$ (see Fig. S4.7 in the Supplemental Material [28]), but is due to a significant decrease in the autocorrelation functions to the values $g_{s(i)}^{(2)}(t_{\max}, 0) = 0.27$ in the regime “Meta ($\lambda_p = 940$ nm)” (see Fig. S4.8 in the Supplemental Material [28]). Actually, the considerable violation of the Cauchy-Schwarz inequality in the considered case is associated with the strong photon antibunching in each of the signal (s) and idler (i) channels, which might be of independent interest for the quantum computing and cryptography protocols. The estimations of specific biphoton generation rate considerably depend on the evaluation method. In particular, the use of closed-form unitary transformations in the Heisenberg representation results in the value of specific biphoton generation rate $R_{\text{SPDC}} = 251$ MHz/mW at the time moment $t_{\max}$ upon choosing the pump power $P_{\text{pump}} = 17.1$ mW and under condition $\tau = 0$ s; see Sec. S6.1 in the Supplemental Material [28] and compare with the value 100 MHz/mW for conventional metasurfaces [22]. At the same time, based on the quantum-classical correspondence approach developed in Refs. [52,53] and employing the

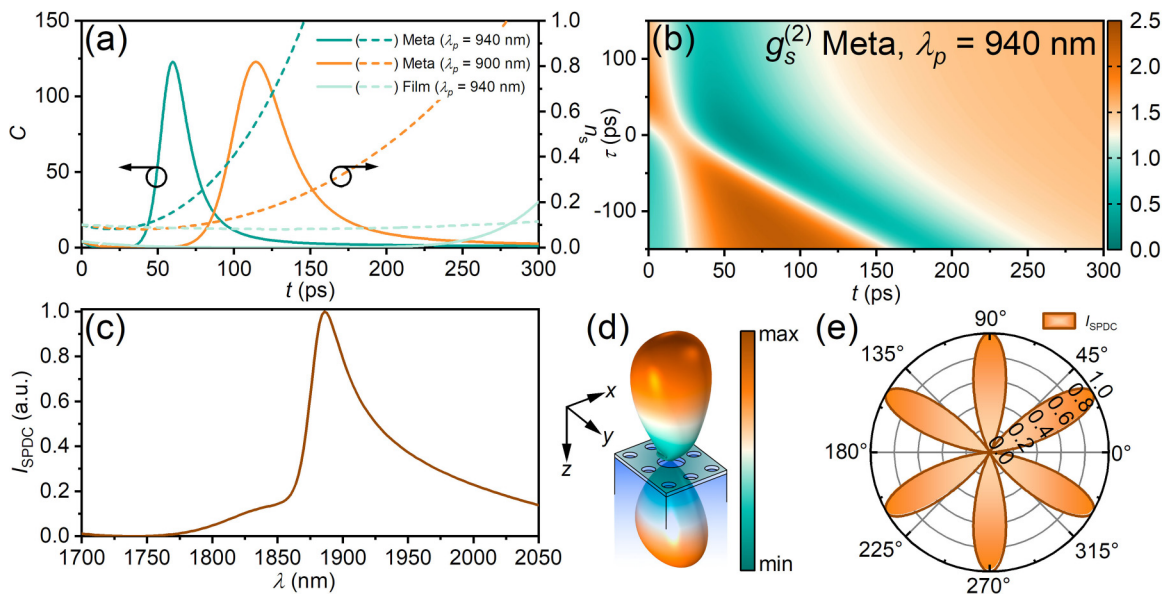


FIG. 4. (a) Quantum dynamics of the average photon numbers n_s (dashed curves) and parameter C (solid curves) for a flat 3R-MoS₂ flake with thickness 100 nm, as well as the resonant at $\lambda_p = 940$ nm and nonresonant at $\lambda_p = 900$ nm interaction regimes for the double metasurface from Fig. 3 and choosing $\tau = 0$ s. (b) Contour plot for $g_s^{(2)}(t, \tau)$ function on time plane ($t; \tau$) for double 3R-MoS₂ metasurface. (c) Generation spectrum, (d) two-photon far-field emission pattern, and (e) dependence of photon-pair rate on pump polarization angle φ measured relative to the x axis (zigzag direction) for synchronously rotation of polarizer and analyzer polarization planes. The geometry of interaction corresponds to Fig. 1.

direct numerical simulation of the second harmonic generation, one can obtain the value $R_{\text{SPDC}} = 61.1$ Hz/mW for the same double membrane 3R-MoS₂ metasurface (see Sec. S6.2 in the Supplemental Material [28]). A significant decrease in the generation rate is associated with accounting for the absorption losses and actual physical geometry of the sample placed on the substrate. This value for membrane metasurface is an order of magnitude greater than numerically calculated one $R_{\text{SPDC}} = 3.5$ Hz/mW obtained in Ref. [54] for a lithium niobate film covered by a SiO₂ metagrating. Moreover, in the latter work, the generation rate enhancement reaches almost 350 times compared to the unpatterned LiNbO₃ film for the similar calculation conditions. In our case, the increase in the SPDC rate for membrane metasurface is only about 30 times, but high absolute value is achieved due to the substantial biphoton generation rate for the used material (1980 Hz/W for the 3R-MoS₂ pristine film versus 10.2 Hz/W for the LiNbO₃ film). This is caused by the large value of the second-order nonlinear susceptibility for 3R-MoS₂ material, which is about 100 pm/V at the considered wavelength versus 20 pm/V for LiNbO₃ [55,56]. In addition, the concept of membrane metasurfaces allows their transfer to optical surfaces, and the simplified methods for their fabrication should be advantageous over the conventional metasurfaces. It should be noted that the values of SPDC rate obtained in real experiments are much lower than the estimated ones for all samples (see Sec. S6.3 in the Supplemental Material [28]). This is due to the experimental features of detection of single-photon states. In practice, tuning of the system to the considered optimal parameters can be achieved by additional controlling the Fabry-Pérot resonances [10] upon depositing the additional reflective coatings, including metallization of the metasurface [25]. A detailed comparative analysis for the

regimes of biphoton pair generation using resonant and non-resonant conditions for a double metasurface and flat flake is presented in Sec. S4 in the Supplemental Material [28].

Note that all the dependences shown in Fig. 4 correspond to the case when the far-field pumping at the wavelength $\lambda_p = 940$ nm forms a near-field source inside the metasurface at the wavelength $\lambda_{\text{SPDC}} = 2\lambda_p = 1880$ nm through nonlinear energy transfer during the SPDC process. The spectral characteristics of such a source are calculated numerically [27] and are shown in Fig. 4(c). The pronounced extremum at the wavelength λ_{SPDC} is complemented by a broad band of biphoton generation with a frequency bandwidth 11.5 THz (1840–1980 nm).

In addition, we show the two-photon far-field emission pattern in Fig. 4(d) exhibiting the characteristic directivity along the z axis that is normally oriented to the metasurface plane. Sixfold polarization pattern in Fig. 4(e) for the case of synchronously rotation of polarizer and analyzer polarization planes is calculated according to the dependence $I_{\text{SPDC}} \propto \sin^2(3\varphi)$ for the used type of SPDC generation and exactly matches the symmetry properties of 3R-MoS₂ structure.

Thus, the key result of the work is the proposal of a general strategy to design strongly nonlinear 3R-MX₂ membrane metasurfaces supporting a pair of high-order multipole modes at different wavelengths at once, which are able to efficiently generate trains of entangled photons for the purposes of quantum cryptography and quantum computing. The membrane nature of metasurfaces allows for their postfabrication transfer to optical elements and their integration with chips, which is impossible for conventional metasurfaces composed of rigid building blocks [24]. This paves the way for the creation of nonlinear sticker-type converters and sticker-type sources of

nonclassical photons in particular, and can also be the starting point for the development of concept of a transferable quantum metaphotonics in general.

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