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Quantum Entangled Photon-Pair Generation From van der Waals NbOI₂

Shaohua Fu¹ | Divyanshu Kumar¹  | Xiaodong Yang²  | Jie Gao¹ 

¹Department of Mechanical Engineering, Stony Brook University, Stony Brook, New York, USA | ²Department of Mechanical and Aerospace Engineering, Missouri University of Science and Technology, Rolla, Missouri, USA

Correspondence: Jie Gao (jie.gao.5@stonybrook.edu)

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ABSTRACT

The realization of quantum light sources is crucial for advancing quantum technologies. Two-dimensional van der Waals (vdW) materials offer a compelling platform for ultracompact entangled photon-pair sources due to their strong optical nonlinearities and relaxed phase-matching constraints. Here, the generation of entangled photon pairs from vdW NbOI₂ flakes on Au film via spontaneous parametric down-conversion (SPDC) process is demonstrated. The observed inverse relationship between second-order correlation function and pump power and the linear relationship between coincidence rate and pump power confirm the characteristic features of SPDC process. Furthermore, the measured polarization-dependent coincidence rate shows the anisotropic entangled photon-pair emission in NbOI₂ crystal. The dependence of coincidence rate on flake thickness is characterized by considering both optical interference and the phase-matching effects. The demonstrated results open new avenues for the development of on-chip integrated quantum photonic devices for applications in quantum state engineering, quantum communication, and quantum information processing.

1 | Introduction

The generation of quantum entangled photon pairs is crucial to the development of quantum information science and technology. The entangled photon pairs have become valuable resources for many applications, such as quantum teleportation, [1, 2] quantum cryptography [3], quantum computation [4, 5], quantum metrology [6], and secure quantum key distribution [7, 8]. To meet those demands, a wide variety of entangled photon-pair sources capable of generating high-fidelity entangled states have been developed, most of which rely on the spontaneous parametric down-conversion (SPDC) in second-order nonlinear crystals [9–11]. Conventionally, bulk crystals such as beta barium borate (BBO) and potassium titanyl phosphate (KTP) have been widely used as SPDC sources. However, these bulk crystals generally exhibit relatively low down-conversion efficiency because of their modest second-order nonlinear susceptibility. To improve

conversion efficiency, quasiphasematched nonlinear crystals such as periodically poled lithium niobate (PPLN) and PPKTP have shown great promise as high-efficiency SPDC sources within a restricted phase-matching range. In addition, these crystals typically require precise temperature tuning and stabilization to maintain the desired phase-matching condition. Such constraints greatly limit the development of ultracompact and efficient entangled photon sources for on-chip quantum photonics integration.

Compared with nonlinear bulk crystals, 2D vdW materials with strong second-order nonlinear optical properties have attracted great attention in recent years as a promising platform for building functional nonlinear photonic devices due to their ultrathin layer thickness and the relaxed phase-matching conditions [12–15]. Among them, the layered niobium oxide dihalide NbOX₂ (X = Cl, Br, or I) is an emerging group of 2D materials with strong second-order nonlinear response attributed to their

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ferroelectric crystal structures, where the spontaneous polarization is generated along the crystal's polar axis. It has been reported that NbOX_2 crystals exhibit giant second-harmonic generation (SHG) responses, and the SHG emission can be tuned with a variety of external stimuli [16–20]. It has also been shown that entangled photon pairs can be generated in NbOCl_2 thin flakes through SPDC process, and the polarization entanglement has been demonstrated from orthogonally stacked crystals [21, 22]. Recently, large SHG enhancement of 2D material flake on planar metallic film has been demonstrated, which is attributed to both the excitation field enhancement and the reflection enhancement from the surface and bulk SHG contributions of nonlinear 2D materials [23].

In this work, we demonstrate the generation of entangled photon pairs through SPDC process in the NbOI_2 thin flakes on Au film. It is shown that both the second-order correlation function at zero time delay $g^{(2)}(0)$ and the integrated coincidence rate from the NbOI_2 flake on Au substrate exhibit large enhancement compared to those from the flake on SiO_2/Si substrate. The inverse relationship of $g^{(2)}(0)$ with pump power is observed with a large $g^{(2)}(0)$ value of more than 1500 at a low pump power of $30\ \mu\text{W}$. Meanwhile, the linear relationship of coincidence rate with pump power is revealed, and a high coincidence rate above 30 Hz is achieved at a pump power of $300\ \mu\text{W}$. Furthermore, it is shown that the polarization-dependent photon-pair coincidence rate exhibits an anisotropic two-lobed pattern with the maximum coincidence rate occurring when the pump polarization is along the crystallographic b -axis due to the in-plane anisotropic lattice structure in NbOI_2 crystal. In addition, the dependence of coincidence rate as a function of NbOI_2 flake thickness is characterized, and the observed oscillation peaks in coincidence rate are modeled by considering both the interference effect and phase-matching effect. The presented results pave the way for the realization of ultracompact and efficient entangled photon sources in 2D materials and the development of on-chip integrated quantum photonic devices for many potential applications

in quantum state engineering, quantum communication, and quantum information processing.

2 | Results and Discussion

Figure 1a shows the side view of the NbOI_2 lattice in the ac -plane and the top view in the bc -plane. The NbOI_2 crystal is monoclinic and belongs to the $C2$ space group. NbOI_2 layers are stacked along the crystallographic a -axis, and each layer consists of $[\text{NbO}_2\text{I}_4]$ octahedra, which are connected by sharing opposite iodine corners along the c -axis and opposite oxygen corners along the b -axis [24, 25]. Owing to the 1D Peierls distortion, the NbOI_2 lattice exhibits unequal distances between two Nb atoms (L_1 and L_2) along the c -axis, while there are unequal Nb–O bond lengths (D_1 and D_2) alternating along the b -axis, which gives rise to the spontaneous polarization in b -axis direction. The optical microscope image of a representative NbOI_2 flake mechanically exfoliated from a bulk crystal on 200 nm Au film is shown in Figure 1b. Figure 1c shows the atomic force microscopy (AFM) image of the flake in the uniform region marked by the white box in Figure 1b. The thickness of the NbOI_2 flake is measured to be approximately 450 nm, as shown in the AFM height profile. Figure 1d shows the measured Raman spectrum of a NbOI_2 flake exhibiting five major characteristic peaks. The peaks located at 102, 205, 269, and $606\ \text{cm}^{-1}$ are assigned to A_g -like modes, while the peak at $510\ \text{cm}^{-1}$ is attributed to a B_g -like mode [25].

SPDC is a second-order nonlinear optical process involving the $\chi^{(2)}$ susceptibility, where a single high-energy pump photon is converted into two lower-energy correlated signal and idler photons, as shown in Figure 2a. Entangled photon pairs with frequency ω are generated within the NbOI_2 flake via SPDC process when the flake is excited by pump photon with frequency 2ω . As illustrated in Figure 2b, the generated signal and idler photon pairs from the NbOI_2 flake are collected in a back-reflection geometry and characterized using a Hanbury

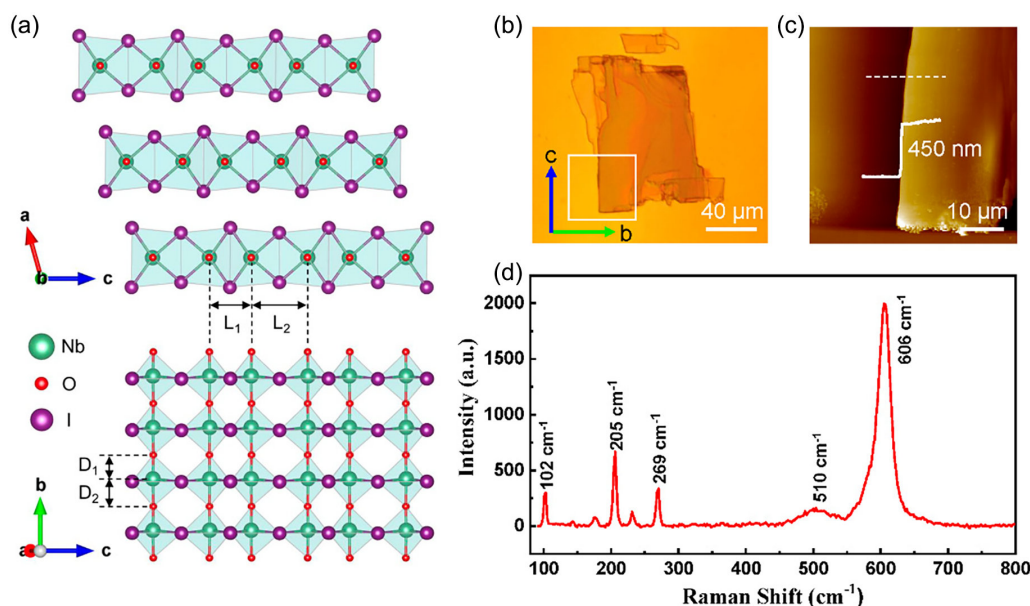


FIGURE 1 | (a) Crystal structure of NbOI_2 . (b) Optical microscope image of a NbOI_2 flake on Au film. (c) AFM image of the flake in the region marked by the white box in (b). Inset: AFM height profile along the white dashed line. (d) Raman spectrum of a NbOI_2 flake on quartz substrate.

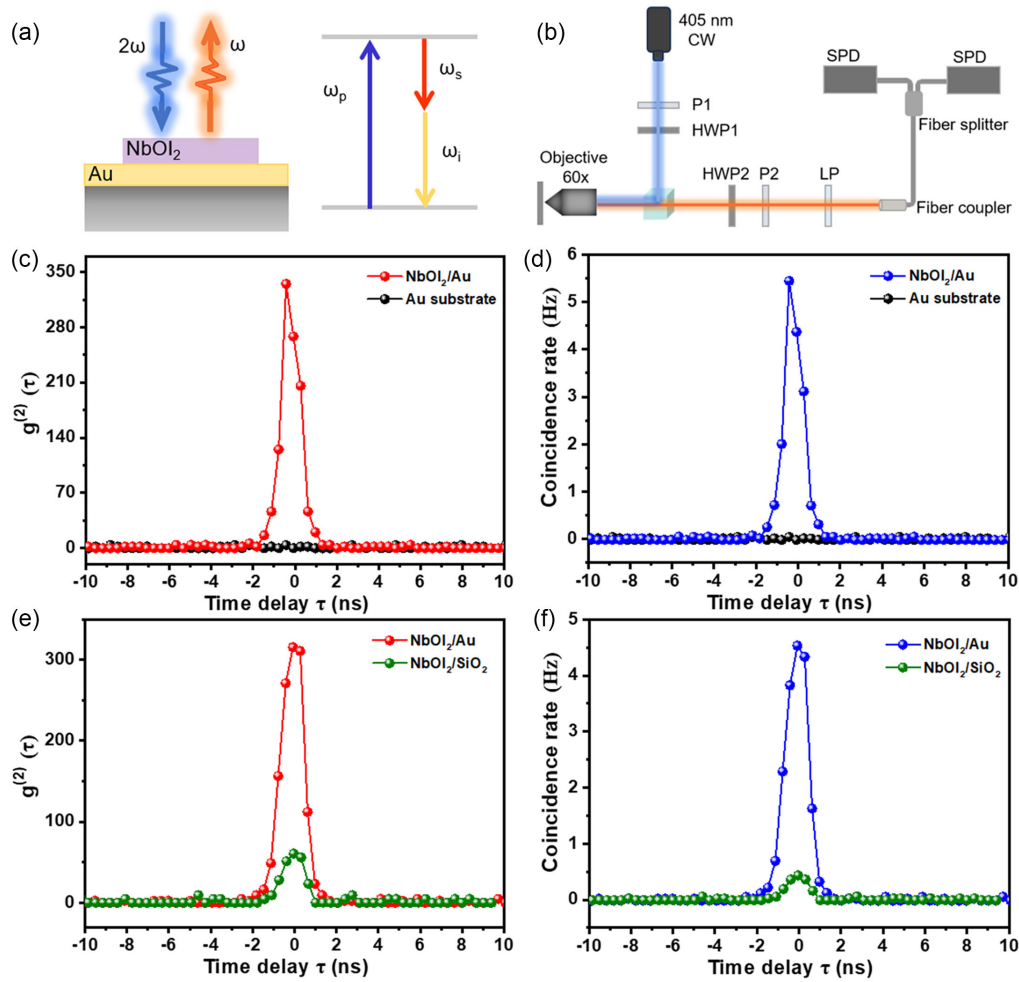


FIGURE 2 | (a) Schematic illustration of the SPDC process from NbOI₂ flake on Au film. (b) Optical setup for the SPDC measurements. (c) Normalized second-order correlation $g^{(2)}(\tau)$ as a function of time delay from the NbOI₂ flake on Au film (red) and from the bare Au film (black). (d) Coincidence rate measured from the NbOI₂ flake on Au film (blue) and from the bare Au film (black). (e) Normalized second-order correlation $g^{(2)}(\tau)$ as a function of time delay from 320 nm-thick NbOI₂ flakes on Au film (red) and SiO₂/Si substrate (green). (f) Coincidence rate from 320 nm-thick NbOI₂ flakes on Au film (blue) and SiO₂/Si substrate (green).

Brown–Twiss setup for photon coincidence measurements. The pump polarization and detection polarization are both set along the b -axis of the flake. Figure 2c shows the normalized second-order correlation function, $g^{(2)}(\tau)$, measured from the 450 nm-thick NbOI₂ flake on 200 nm Au film under a pump power of 150 μ W with a time bin width of 0.35 ns and a collection time of 30 s. A pronounced bunching peak at zero time delay with $g^{(2)}(0)$ value of 335 indicates the generation of correlated photon pairs with very high coincidence-to-accidental ratio $\text{CAR} = g^{(2)}(0) - 1$. In contrast, no such peak is observed from the bare Au film. This confirms that the correlated photon pairs originate from the SPDC process in the NbOI₂ flake and rules out contributions from other effects such as thermal bunching and Au substrate itself. Figure 2d plots the coincidence rate as a function of time delay from the NbOI₂ flake on Au film and the bare Au film. Coincidence rate of more than 5 Hz is achieved at the zero delay time for the NbOI₂ flake on Au film, indicating the bright SPDC emission. Furthermore, the SPDC emission from a 320 nm-thick NbOI₂ flake on 200 nm Au substrate and a 320 nm-thick NbOI₂ flake on 300 nm SiO₂/Si substrate is compared under the same pump power of 150 μ W. The same thickness of 320 nm for the two flakes is confirmed with the AFM measurements. As shown

in Figure 2e, the NbOI₂ flake on Au substrate exhibits a $g^{(2)}(0)$ peak value of 315, which is much higher than the value of 60 on SiO₂/Si substrate. In Figure 2f, the integrated coincidence rate is 18.1 Hz for the NbOI₂ flake on Au substrate, showing an approximately 10-fold enhancement compared to 1.7 Hz for the NbOI₂ flake on SiO₂/Si substrate. It is worth noting that the observed brightness of 11.3 Hz/mW for the NbOI₂ flake on SiO₂/Si substrate is comparable to the rBN crystal with brightness of 9.15 [12] and 8 Hz/mW [26] and the NbOCl₂ crystal with brightness of 5 Hz/mW [27] in a similar back-reflection measurement configuration (see more comprehensive comparison in Table S1). For the NbOI₂ SPDC source on Au substrate, brightness of 121 Hz/mW is achieved, which is one order of magnitude higher than that of the flake on SiO₂/Si substrate. The presence of the 2D material-metal interface enhances the SPDC coincidence counts, similar to the observed SHG enhancement of 2D material flakes on metallic film [23]. Such enhancement is attributed to the enhancement of the excitation field and the enhancement of the reflected SPDC signals consisting of the surface and bulk contributions from the 2D material layer. On the other hand, the physical mechanism underlying the enhanced photon-pair generation for the NbOI₂ flake on Au substrate with respect to the NbOI₂ flake on

SiO₂/Si substrate can also be understood from the enhancement of both dipolar and quadrupolar contributions to the nonlinear polarization, where the dipolar term couples only to the electric field amplitude, while the quadrupolar term couples to the electric field amplitude and its gradient [28]. Due to the larger reflectivity of the NbOI₂/Au interface compared to that of the NbOI₂/SiO₂ interface, the electric field amplitude is increased to provide a stronger dipolar contribution to the nonlinear polarization. Meanwhile, the electric field gradient also gets larger near the NbOI₂/Au interface. Together with the increased electric field amplitude, this further amplifies the quadrupolar contribution to the nonlinear polarization. As a result, the SPDC coincidence rate is significantly enhanced for the NbOI₂ flake on Au substrate.

Next, the power dependence of entangled photon-pair generation from the NbOI₂ flake on Au film is examined to characterize the SPDC process. Figure 3a shows the normalized second-order correlation function $g^{(2)}(\tau)$ under different pump powers. With the pump power increasing, the peak value of $g^{(2)}(\tau)$ gradually decreases. As shown in Figure 3b, the normalized second-order correlation function at zero time delay $g^{(2)}(0)$ exhibits an inverse dependence on pump power, which is attributed to the increase in accidental counts at higher pump power. At a low laser power of 30 μ W, a $g^{(2)}(0)$ value of more than 1500 is obtained. Figure 3c shows that the coincidence rate gradually increases with the pump power, and Figure 3d plots the integrated coincidence rate within the central ± 10 ns time delay window. The coincidence rate

scales linearly with the pump power, showing the characteristic behavior of the SPDC process. Notably, a coincidence rate above 30 Hz is achieved at a pump power of 300 μ W. These results demonstrate that a higher pump power will lead to an increased coincidence rate of entangled photon-pair generation in NbOI₂ flakes, but the associated accidental coincidences reduce the $g^{(2)}(0)$ value.

Furthermore, the polarization-dependent photon-pair generation is investigated to confirm the Type-0 SPDC process and reveal the anisotropic response in the NbOI₂ flake. Figure 4a shows the measured normalized second-order correlation function $g^{(2)}(\tau)$ when the signal and idler photons are copolarized and cross-polarized to the pump photon, while the pump polarization is set along the *b*-axis of NbOI₂ crystal. The result indicates that most of the SPDC photon pairs are produced in the copolarized configuration instead of the cross-polarized configuration. This observation confirms that the entangled photon-pair generation in the NbOI₂ crystal follows the expected Type-0 SPDC process, where the produced photon pairs exhibit the same polarization as that of the pump beam. Figure 4b presents $g^{(2)}(\tau)$ under different polarization angles in the copolarized configuration, where the polarization angle θ is defined as the angle between the pump polarization and the crystallographic *c*-axis. As the pump polarization rotates from the *c*-axis ($\theta=0^\circ$) to the *b*-axis ($\theta=90^\circ$), the $g^{(2)}(0)$ value increases significantly and reaches its maximum at $\theta=90^\circ$. Figure 4c shows the corresponding coincidence rate at

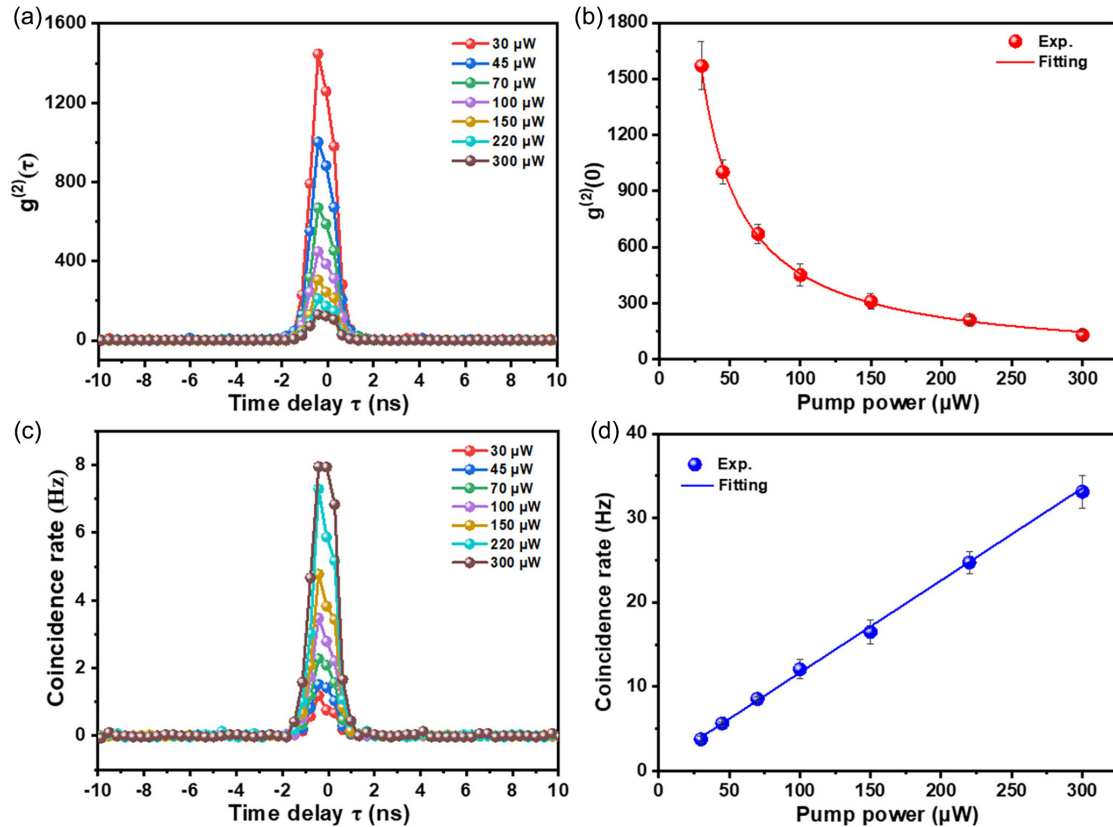


FIGURE 3 | (a) Normalized second-order correlation $g^{(2)}(\tau)$ as a function of time delay for the NbOI₂ flake on Au film at different pump powers. (b) Pump-power dependence of $g^{(2)}(0)$ with the measured data (red dots) and the fitting (red line). (c) Coincidence rate measured from the NbOI₂ flake on Au film at different pump powers. (d) Pump-power dependence of the integrated coincidence rate with the measured data (blue dots) and the fitting (blue line).

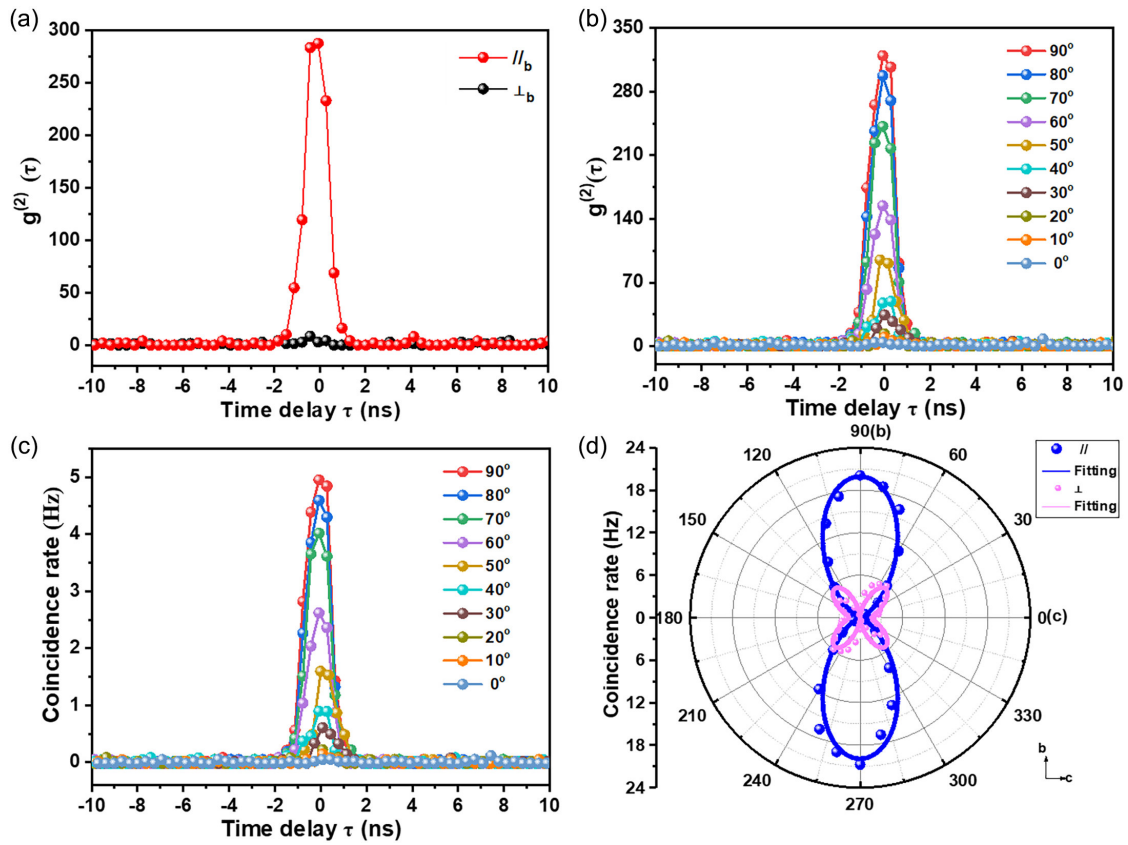


FIGURE 4 | (a) Normalized second-order correlation function $g^{(2)}(\tau)$ when the signal and idler photons are copolarized (red) and cross-polarized (black) to the pump photon, with the pump polarization along the b -axis. (b) $g^{(2)}(\tau)$ and (c) coincidence rate under different pump polarization angles in the copolarized configuration. (d) Polar plots of the measured copolarized (blue dots) and cross-polarized (magenta dots) polarization-dependent coincidence rate with the corresponding theoretical fitting.

different pump polarization angles, exhibiting the same angular dependence as that of $g^{(2)}(\tau)$. The observation indicates that the pump polarization angle affects the efficiency of photon-pair generation due to the in-plane anisotropic lattice structure in NbOI₂ crystals. Figure 4d presents the polar plots of the measured polarization-dependent photon-pair coincidence rate in the copolarized and cross-polarized configurations. Since the SPDC process can be considered as the reverse process for SHG with the same second-order nonlinear susceptibility tensor $\chi^{(2)}$, the polarization-dependent photon-pair coincidence rate via SPDC process can be expressed as $r_{\text{SPDC}}^{\parallel} \propto |3\chi_{\text{bcc}}^{(2)} \cos^2 \theta \sin \theta + \chi_{\text{bbb}}^{(2)} \sin^3 \theta|^2$ and $r_{\text{SPDC}}^{\perp} \propto |\chi_{\text{bcc}}^{(2)} \cos^3 \theta - 2\chi_{\text{bcc}}^{(2)} \sin^2 \theta \cos \theta + \chi_{\text{bbb}}^{(2)} \sin^2 \theta \cos \theta|^2$ where $\chi_{\text{bbb}}^{(2)}$ and $\chi_{\text{bcc}}^{(2)}$ represent the two independent second-order nonlinear susceptibility elements [20]. The theoretical fittings of coincidence rate agree well with the experimental data, as shown in Figure 4d. The coincidence rate of SPDC emission exhibits a two-lobed pattern in copolarized configuration while showing a four-lobed pattern with much smaller values in cross-polarized configuration. The largest coincidence rate occurs in copolarized configuration as the pump polarization and detection polarization are both oriented along the crystallographic b -axis, which is consistent with the C2 space group of NbOI₂ crystal.

Moreover, the dependence of SPDC photon-pair generation on flake thickness is explored with pump polarization and detection polarization along the crystallographic b -axis. Figure 5 presents the coincidence rate as a function of NbOI₂ flake thickness, where

multiple oscillation peaks appear as the flake thickness increases. This thickness-dependent behavior can be understood as the combined result of optical interference effect and phase-matching effect [29]. The interference effect of light propagation within the air-NbOI₂-Au three-layer structure in the back-reflection configuration is modeled using the transfer matrix method. The reflectivity of light from the three-layer structure oscillates periodically

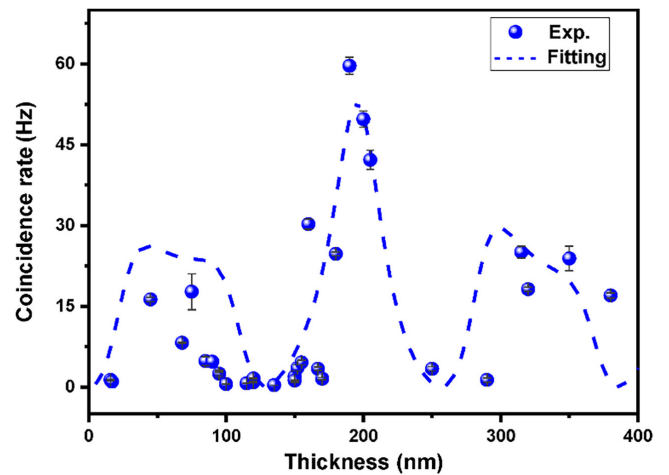


FIGURE 5 | Measured coincidence rate of the photon pairs from the NbOI₂ flake on Au film as a function of the flake thickness shown as the dots, with theoretical modeling shown as the dashed line.

with flake thickness due to the interference effect, [30, 31] which can be expressed as $R(h) = |(r_{12} + r_{23}e^{2i\beta})/(1 + r_{12}r_{23}e^{2i\beta})|^2$, where $r_{ij} = (n_i - n_j)/(n_i + n_j)$ is the reflectivity coefficient from layer i to layer j , $\beta = 2\pi n_2 h/\lambda$ is the phase accumulated across the NbOI₂ layer, and h is the NbOI₂ flake thickness. Here, n_1 , n_2 , and n_3 denote the refractive indices of air, NbOI₂, and Au, respectively. Accordingly, the effective pump light intensity at the flake is $I_p = [1 - R(h)]I_{in}$, with I_{in} as the incident pump intensity which is fixed in the measurement. With the different refractive indices of NbOI₂ for the pump light and the SPDC signal, the phase-matching condition needs to be fulfilled for efficient nonlinear conversion in the NbOI₂ flake. The coincidence rate under phase-mismatched condition can be expressed as $r_{SPDC} \propto |\chi^{(2)}|^2 I_p h^2 \text{sinc}^2(\Delta k h/2)$, where Δk is the wavevector mismatch between the pump light and SPDC signal [32]. The coherence length for the SPDC process is $L_c = \pi/\Delta k$, which is estimated around 45 nm in the back-reflection configuration. Therefore, the thickness-dependent coincidence rate modulated by both the interference effect and phase-matching effect can be approximated as $r_{SPDC} \propto [1 - R(h)]h^2 \text{sinc}^2(\Delta k h/2)$. As shown in Figure 5, the theoretically modeled thickness-dependent coincidence rate exhibits multiple oscillations as the thickness increases, which is in good agreement with the measured data. It is observed that the first peak appears at the thickness around 45 nm, which is consistent with the estimated coherence length. The maximum photon-pair generation rate is obtained at the thickness of around 200 nm, where the interference effect and phase-matching effects interact constructively.

3 | Conclusion

In summary, the generation of entangled photon pairs from van der Waals NbOI₂ flakes on Au film via SPDC process has been demonstrated. Significant enhancement in the second-order correlation and the coincidence rate is achieved for the NbOI₂ flake on Au film, compared to that on standard SiO₂/Si substrate. The observed inverse relationship of $g^{(2)}(0)$ with pump power and the linear relationship of coincidence rate with pump power confirm the characteristic features of the correlated photon-pair generation through SPDC process. Furthermore, the polarization dependence of photon-pair coincidence rate is examined to reveal the anisotropic entangled photon-pair emission in NbOI₂ crystal. Moreover, a distinct thickness-dependent oscillation in the coincidence rate is observed, which is governed by the interplay between optical interference and the phase-matching condition. These findings demonstrate 2D material-metal structures as a compact platform for realizing efficient entangled photon sources and on-chip integrated quantum photonic devices for quantum state engineering, quantum communication, and quantum information processing.

4 | Experimental Section

4.1 | Sample Preparation

The NbOI₂ flakes with different thicknesses are mechanically exfoliated onto polydimethylsiloxane layers using scotch tape from a bulk crystal (HQ graphene) and then transferred

onto a 200 nm Au film coated on glass substrate using dry transfer technique.

4.2 | Optical Setup

The NbOI₂ flake is pumped by a 405 nm continuous-wave laser, and the generated signal and idler photon pairs around 810 nm from SPDC process are collected in a back-reflection geometry and characterized using a Hanbury Brown–Twiss setup. The pump beam passes through a linear polarizer and a half-wave plate for pump polarization control and then is focused on the sample through a dichroic mirror and an objective lens (60×, NA=0.85). The back-reflected SPDC photon pairs are collected through the same objective lens and dichroic mirror, directed through a half-wave plate and a linear polarizer for output polarization control, and passed through two 750 nm longpass filters. Then, the photon pairs are split into the signal and idler paths with a 50/50 fiber beamsplitter and detected with two single photon counting modules (SPCM-AQRH-14-FC), which are connected to a TCSPC (HydraHarp 400) for photon coincidence measurements.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.