Quantum Light Sources Based on van der Waals Materials

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Overview:

Quantum Sensing and Measurement (QSM) techniques encompass a diverse array of technologies and instruments, transcending classical physics limitations. The integration of quantum light sources (QLS) capable of generating entangled photon pairs and indistinguishable photons emerges as indispensable for numerous applications in remote sensing, precise measurement, and optical communications. Within the present technological landscape, there is a preference for solid-state photon sources that offer scalability and seamless integration with existing semiconductor technologies.

Today, the "purest" source of indistinguishable photons comes from optically trapped single atoms/ions. However, they require ultralow operational temperatures, complex equipment, and lack scalability for operations involving thousands of qubits. Conversely, laser pumped spontaneous parametric down conversion (SPDC) offers a proven method for generating entangled photon pairs. Yet, they rely on large scale (mm-size) bulk crystals that are incompatible with on-chip commercial photonic technologies and metal-oxide-semiconductor (CMOS) fabrication. Hence, the development of a singular platform meeting all ideal criteria underscores the imperative for ongoing exploration and development of novel materials conducive to the production of quantum light sources.

For the next generation of QLS, we propose to develop novel sources of single and entangled photons based on a singular platform comprising of van der Waals (*vdW*) ultrathin crystals and mono(few) layers. These sources would have the following advantages:

1) ***vdW* QLS would operate at room temperature and extend the operation wavelength into 1.3-1.5 mm near-IR range for telecommunication windows.**
2) **They would provide highly indistinguishable single photons and entangled photon pairs with orders of magnitude higher generation efficiencies.**
3) **The ultrathin and ultrasmall 2D nature of *vdW* QLS will facilitate ready integration with state-of-the-art, microfabricated photonic integrated circuits (PICs).**

We plan to develop two unique yet different approaches to achieve our goals: First approach will center on entangled photon pair source based on SPDC generated in ultrathin van der Waals crystals such as Group V oxyhalides MOX_2 (M=Nb, Ta, and X=Cl, Br and I) where $NbOCl_2$ has shown features such as vanishing interlayer coupling and monolayer-like excitonic behavior in the bulk form and others remains unexplored. The second approach will concentrate on engineering strain-induced defects in various Indium Selenide (InSe) mono(few) layer flakes acting as deterministic single photon sources (SPS) in the near-IR telecommunication windows. Both approaches would provide for the chip-scale, solid-state QLS that will be a key enabler for QSM applications with unique transition opportunities for remote sensing, imaging, precision measurement and quantum computing/communications.

Technology background:

a) As QSM techniques advance, there is a growing necessity to integrate quantum sources into existing semiconductor processes and chip designs. Parametric downconversion offers a proven method for generating entangled photon pairs. SPDC operates as a second-order nonlinear optical (NLO) process, where one photon (γ_p) splits into a pair of correlated and entangled photons known

as the "signal (γ_s)" and "idler (γ_i)," adhering to energy and momentum conservation principles. Present SPDC-based quantum light sources rely on commercial bulk crystals (mm size) of barium borate and lithium niobate. However, these sources face intrinsic limitations for photonics applications on platforms compatible with CMOS fabrication due to their size and 3D covalent bonding nature.

In contrast, two-dimensional (2D) layered materials composed of van der Waals monolayers have attracted significant attention due to unique optoelectronic properties. These materials exhibit reduced dielectric screening, strong electron-hole interactions, and possess localized defects capable of emitting single photons at elevated temperatures. Moreover, they demonstrate relaxed phase-matching conditions, resulting in enhanced NLO effects at the 2D limit. Due to their intrinsic suitability for bond-free integration without lattice and process limitations at the ultrasmall limit, 2D layered materials are increasingly recognized for their potential in integrated photonics and as on-chip circuit elements. They can be grown by chemical vapor deposition (CVD) methods at low (<450 ^oC) temperatures to allow for direct on-chip integration. However, NLO conversion efficiency of current traditional transition metal dichalcogenides (TMDs, such as MoS_2 or WSe_2) monolayer (ML) is predictably small due to vanishing light-matter interaction as typical thickness of a TMD monolayer is < 1 nm. Increasing thickness (*i.e*. number of layers) significantly modifies the electronic structure due to strong interlayer electronic coupling and dielectric screening that leads to decreased nonlinearity in addition to self-absorbing effects. Therefore, some new types of *vdW* crystal with different structure and scalable second order NLO response will provide revolutionary solutions for future QSM technologies.

b) On the other hand, availability of TMD-based, deterministic single photon sources in the telecom window (*i.e*., near infrared, NIR: 1320-1550 nm) have become highly desirable. SPSs based on spontaneously formed defects in TMD monolayers such as 2D hexagonal boron nitride (*h*BN), $MoSe_2$, WS_2, and WSe_2 have been demonstrated. However, the luminescent spectral range is limited from the edge of near-IR to visible and the origin of the SPSs in these systems are not fully understood. While strain engineering is a promising route for creating SPSs in monolayers, it is still challenging to control the distribution and atomic structures of the defects in 2D crystals deterministically by synthesis and post-growth processes in the majority of 2D systems. So far, the only publication that shows extension of SPS emission outside of the visible range has been the work of Htoon's lab at Los Alamos (LANL) on strain-controlled defects in monolayer $MoTe_2$, with emission in 1100 nm range.[1] Finally, it is highly desirable to have electrically addressed SPS for seamless integration into optoelectronics platforms.

Opportunity and Research Objectives:

1. Recently, a new type of *vdW* crystals such as niobium oxychloride ($NbOCl_2$) featuring vanishing interlayer electronic coupling and monolayer-like excitonic properties in the bulk form, has been discovered[1]. **Our approach will result in further development and engineering this new family of oxyhalide *vdW* crystals with objectives to obtain strong and scalable second-order optical nonlinearity that is <u>3 orders higher than in a monolayer TMD</u> and will present a unique opportunity to integrate compact sources of entangled photon pairs for the rapidly developing hybrid integrated photonic platforms**. **Figure 1a** shows a schematic illustration of the niobium oxyhalide *vdW* crystals which have demonstrated scalable second-order non-linear optical (NLO) response and SPDC photon pair emission at the ultrathin limit, **Figure 1b**.[2] Work by **Lv's** group (PI) has already demonstrated vapor phase growth of high quality $NbOCl_2$ crystals that could be exfoliated into ultrathin flakes, **Figures 1(c,d)**. Using this home-grown $NbOCl_2$ thin

crystal, co-PI **Malko's** lab has demonstrated strong NLO second harmonic generation and large second-order correlation function ($g^2(\tau)$), indicative of photon pair generation, **Figure 1e**. We expect to explore stronger signals from other new group V oxyhalides (M=Nb, Ta, and X=Cl, Br and I) we have grown already. Being unique 2D layered materials, the *vdW* crystals enable bond-free integration without lattice matching and processing limitations. Furthermore, we will focus on incorporating these *vdW* crystals into dielectric tunable cavities that will provide strong enhancement of the NLO and enhance the production rate of the entangled photon pairs. These layered growth techniques are also amenable to direct integration in microfabricated Si structures including means to directly couple photon pair output into micropatterned planar waveguides embedded in the Photonic-Integrated Circuit (PIC) design, *e.g.* the open-access microcavity structure (**Figure 1f)** and multiplexed entangled photon source illustrated in **Figure 1g.**

2. Among many candidates, indium selenide emerges as a rapidly expanding family of layered compounds garnering increasing attention due to its advantageous mechanical, electrical, and

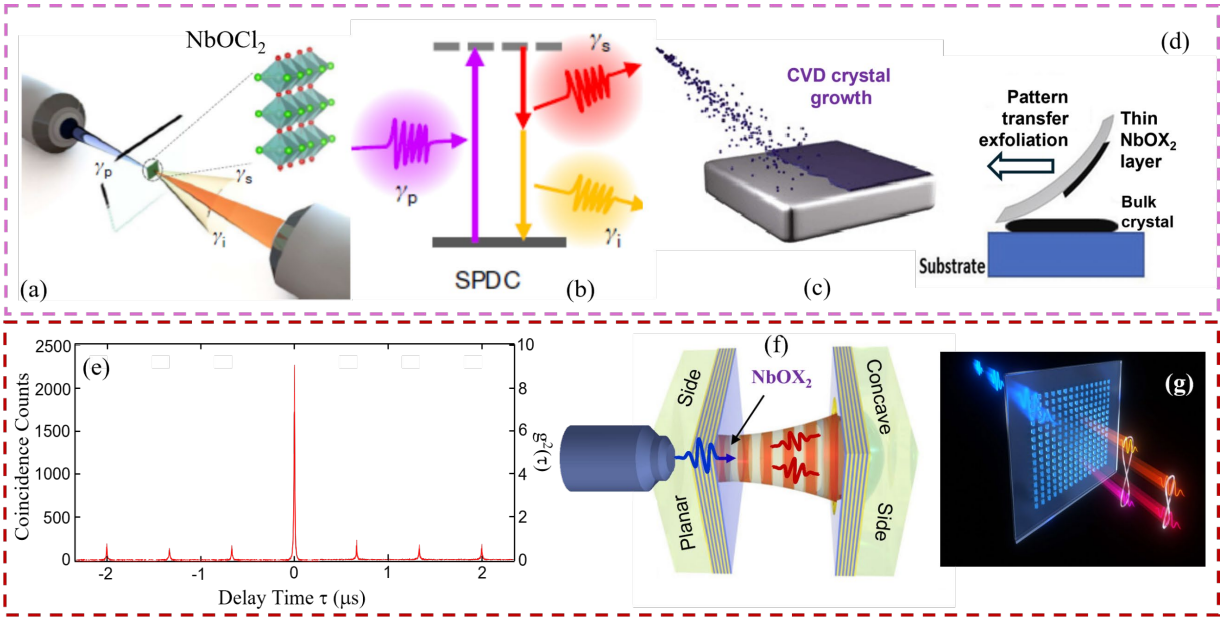

Figure 1. (a) Schematic illustration of the SPDC process and generation of entangled photon pairs using an ultrathin $NbOCl_2$ *vdW* crystal, Ref [1] (b) Energy schematics diagram for pump (*p*), signal (*s*) and idler (*i*) photons. (c) Schematic illustration of the $NbOX_2$ bulk samples *via* CVD growth, (d) preparation of thin crystals by exfoliation, (e) Our recent measurements using home-grown $NbOCl_2$ thin sample that shows normalized two-photon correlation function indicating photon pair emission. (f) schematic of incorporation into open access microcavity and (g) artistic render of entangled photon pairs from multiple arrays.

optical properties. Present in various stoichiometric compositions such as InSe, In_2Se_3, In_3Se_4, etc., and characterized by distinctive phases and crystal structures, indium selenide offers ample opportunities for manipulation of bandgap and defect geometries.[3] In contrast to TMDs, the indirect bandgap of indium selenide monolayers undergoes a transition to a direct bandgap with increasing thickness. Few-layer InSe exhibits promising characteristics beyond traditional TMDs, including strong photoresponsivity, emissive excitons with anisotropic properties, tunable spin polarization, and notably, very high carrier mobility exceeding 10^3 cm^2 V^{-1} s^{-1}, the highest between TMDs. We expect InSe to show a large propensity to bandgap and defect manipulation using strain engineering. Thus, **we propose to develop a range of NIR materials for SPS generation based**

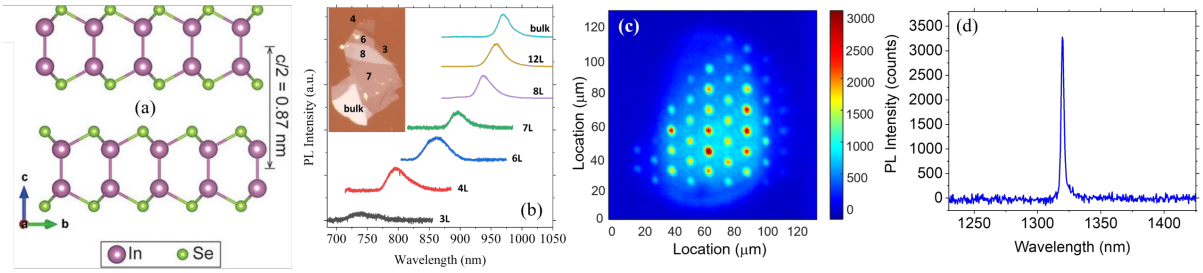

Figure 2. (a) Side view of the InSe crystal structure. (b) PL emission spectra of the γ-InSe samples with different monolayer thickness. Inset: optical microscope image of the few layer flake regions (c, d) PL image and PL spectra of strain-induced emissive defects in InSe few-layer flake extending in the NIR region.

on strain-engineered defects in Indium Selenide in various crystalline forms. In collaboration with Dr. Han Htoon at LANL we worked on several types of InSe materials. **Figure 2** shows crystalline structure and emission spectra of γ-InSe. Using exfoliation method, we produced InSe flakes of different thickness, down to a few monolayers, thus manipulating their PL emission properties. Quite uniquely, transitioning from bulk to few ML thickness, the bandgap is changed by nearly 0.5 eV, **Figure 2b**. Using nanopillar array structures, periodic strain was induced in the flakes placed on top of the pillars, **Figure 2c**. The emission from the strain regions indicates the emergence of well-isolated defects with sharp spectral lines positioned in the telecom emission window as dependent on the amount of strain, **Figure 2d**. Again, the PL emission of the defect line is shifted toward near-IR by nearly 0.6 eV, the lowest energy observed among *vdW* materials. ***Finally, these nanosized materials will be incorporated into tunable micro/nanocavities to enhance emission rate.*** Cavity would play two significant roles: (1) providing Purcell enhancement for fast radiative decay and (2) providing directional emission for highly efficient photon collection and coupling. Guiding photons will lead to much higher brightness of the source.

Team and Budget:

Our team will further leverage existing collaborations between Prof. B. Lv and Prof. A. Malko with complimentary expertise on bulk crystal growth, thin film deposition and exfoliation (**Lv**) and optical characterizations in particularly on NLO and SPDC measurement and strain related optical studies (**Malko**) to build up comprehensive understanding on these two classes of materials. We will continue collaboration with Dr. Htoon who will complement our measurement efforts in the near-IR spectral regions. We would like to budget for $200,000/year for the three-year project period (10/2024 –09/2027), including support for 2 PhD students/year, $10,000/year materials and supplies, $2,000/year for publications, and $3,000/year for travels.

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[1] Zhao, H.; Pettes, M. T.; Zheng, Y.; Htoon, H. *Site-Controlled Telecom Wavelength Single Photon Emitters in Atomically Thin $MoTe_2$*, *Nat. Comm*. **2021**, *12*, 6753

[2] Guo, Q.; Qi, X.-Z.; Zhang, L. et al., *Ultrathin quantum light source with van der Waals $NbOCl_2$ crystal*, *Nature* **2023**, *613*, 53.

[3] Song, C.; Fan, F.; Xuan, N.; et al., *Largely Tunable Band Structures of Few-Layer InSe by Uniaxial Strain*, *ACS Appl. Mater. Interfaces* **2018**, *10*, 3994-4000