Research Plan

研究计划

Single-Photon Emitters and Charge Density Wave of Two-Dimensional Materials

基于二维材料的单光子源和电荷密度波应用

Qingkai Qian

钱庆凯

Department of Electrical Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802, United States

美国宾夕法尼亚州立大学电子工程系

ABSTRACT

Single-photon emitters (SPEs) are systems that emit one photon at a time, which are important components in photon-based quantum computation and communication. Charge density wave (CDW) is an existing state of some materials with a periodic modulations of charge and lattices. Like superconducting state, CDW is associated with the strongly correlated electrons and electron-phonon interactions. The formation of CDW often induces metal-insulator transition and the conductivity modulations, making CDW have potential applications in memory and storage devices. Both SPEs and CDW have been discovered in two-dimensional (2D) materials. Because of the intriguing properties such as atomic-layer thickness and the dangling-bond-free van der Waals surface, 2D materials possess some special advantages in realizing SPEs and CDW applications. Both SPEs and CDW states can be tuned by defects, and the related studies will also promote the understanding of SPEs and CDW. This research plan will aim to investigate the influences

of defects over SPEs and CDW of 2D materials and the related mechanisms. A better understanding and engineering of SPEs and CDW will be achieved.

摘要

单光子源是一次发射一个光子的系统。单光子源是基于光子的量子计算和量子通信应用中的重要组成部件。电荷密度波是在某些材料的一种稳定存在的相，它表现为材料电荷结构的周期性扰动而实现的稳定状态。和超导体一样，电荷密度波的形成也和电子的交换关联和电声作用有关。电荷密度波的形成导致材料性质的改变，使得它在记忆存储器件方面具有应用。单光子源和电荷密度波都可以在二维材料中实现。由于二维材料具有一些优异的性能，比如具有原子层的厚度同时没有表面悬挂键，它在实现单光子源和电荷密度波的调控方面具有某些独特的优势。例如，基于二维材料的单光子源和电荷密度波可以方便地通过应力和缺陷实现调制，并有利于进一步研究其原理和提高器件性能。本研究计划将集中研究基于二维材料的单光源和电荷密度波，致力于通过应力、缺陷、电场等研究其机制和提高其性能以实现实际应用。

1. BACKGROUND

Single-photon emitters (SPEs) are isolated quantum systems that emit one photon at a time.[1] The signature of an SPE is a characteristic dip in the second-order correlation function $g_2(\tau)$ with value smaller than 0.5.[1] 2D materials can be promising material platforms to realize high-performance solid-state SPEs, which are essential for photon-based quantum information and computing.[1-3] One reason is that by using simple mechanical exfoliation, high-quality samples with unique photonic and optoelectronic properties can be obtained.[3] Another reason is that there is a large library of van der Waals crystals that can be exploited.[4] Besides, the atomically thin 2D materials have the advantage of efficient extraction of the emitted photons without suffering from internal reflections.[2] Moreover, 2D materials have great flexibility in heterogeneous integration and are compatible with established optoelectronic processing technologies.[3] SPEs based on 2D materials have already been demonstrated on WSe_2, WS_2, and hBN.[3,5-8] The mechanism of SPEs in 2D materials is still under debate.[1] One possible mechanism is the quantum dot (QD) effect.[1] The emission originates from a bound exciton that is confined to zero dimension by a potential field generated by local strain and/or a crystallographic defect. The second possible mechanism of SPE is the color center, *i.e.*, an impurity or an isolated crystallographic point defect with electronic states that lie deep within the bandgap and possess the characteristics of an isolated artificial atom.[1]

Of 2D materials, WSe_2 is the most widely studied material for SPEs, which are mainly triggered optically.[3,5,6,8] For exfoliated flakes with naturally occurred SPEs, the emitters are usually located at the flake edges.[3,5,6,8] SPEs in WSe_2 flake needs to be measured at low temperature (<20 K), and the single-photon emission becomes barely visible with temperature >15 K.[3,5,6,8] It is worth to note SPEs based on hBN can operate at room temperature, even though low temperature is better and has smaller peak width.[7] The current studies mostly relied on the random occurrences of natural defects/strain in the 2D material basal planes for quantum emitters. Even though the location of quantum emitters have been deterministically created by strain engineering in 2D materials, the emission wavelength is still widely dispersed out of control.[9-13] The main reason is the lack of understanding of the accurate relationship between quantum emission performance and strain/defect type in 2D materials, and also the engineering challenge of controllably generating strain gradient and defects with a specific type and optimal density. Even though there are studies about the defect level of WSe_2, suggesting that the dissociation of O_2 and the passivation of Se vacancy might be the reason for single-photon emitter, but it is still to be experimentally proved.[14] For the known emitters, quantum optical measurements such as two-photon interference need to be demonstrated. Electrical triggering of SPEs should be pursued to extend the easy deployment advantage of 2D materials in SPE applications. All SPEs based on 2D materials also face the disadvantages of lack of optical spin readout that resembles the nitrogen-vacancy center in diamond. The above gaps have severely hindered the development of engineered 2D materials for desirable quantum photonics.[1]

Charge density wave (CDW) is a periodic modulation of electron densities associated with a periodic displacement of ion positions in some crystals.[15,16] Charge density wave can show varying stages during the formation process. For example, TaS_2 shows an incommensurate CDW phase starting from 543 K, changes to near commensurate phase at 354 K, and becomes commensurate at 183 K.[17] During the whole cooling process, the resistance shows a sharp increase during phase transition. A strong electron correlation often accompanies the CDW, which can induce Mott insulating behavior,[15,18,19] the study of which can also help the understanding of other collective quantum phenomena such as high-temperature superconductivity in doped copper oxides and quantum spin liquids.[20,21] The Mottness becomes important when the Coulomb interaction (U) exceeds the bandwidth (W) in Hubbard-like models.[15] Recent advances in van der Waals 2D material systems have extended the research of CDW and Mott physics to the two-dimensional limit.[15,18,19] One example is 1T-TaS_2, which hosts various CDW phases and metal-insulator transitions with the formation of a star-of-David superstructure. The exact mechanism of forming CDW in TaS_2 remains elusive.[19,22] Fermi surface nesting has already been ruled out, while the phonon instabilities are suggested as an important signature, but the main drive remains uncertain.[19,22,23] Besides, the effects of interlayer stacking on the CDW formation and the metal-insulator transitions are still controversial: interlayer hopping is supposed to increases W while interlayer dielectric screening decreases U, which will suppress the Mott insulating behavior,[15] but a certain stacking order has also been reported resulting in a gap even in the absence of electron correlation.[24-27]

Recent experimental results indicate that the CDW states can be tuned by external and internal control parameters, such as pressure, optical (electrical) excitation, chemical doping and defects.[22,28-32] The substitution doping found that the transition temperature and conductivity of CDW materials can be modulated. Cation doping is found to suppress the transition temperature, and relatively high-level doping will destroy the commensurate CDW phase. The anion disorder at any doping level, on the other hand, doesn't suppress the phase transition toward the commensurate phase.[30] These conductivity switchings may be engineered to realize novel memory devices based on these correlated electrons.[32,33] Domain walls within CDW could also be influenced by the defects and doping, which are shown to play an important role in the metal-insulator transitions of CDW.[15] CDW domain walls are highly conducting channels and are considered as the origin of the metallic property.[15,21] Moreover, the emerging superconductivity out of the Mott-CDW insulating phase has been considered being directly related to the conducting domain walls.[21] The related studies about the atomically thin 1T transition metal dichalcogenides in both theoretical and experimental aspects will offer a great opportunity to differentiate the contributions of electron correlation and interlayer effects during the formation of CDW, which may find engineering applications in memory devices and shed some light on the high-temperature superconductivity.

2. METHODOLOGY AND APPROACH

Single-Photon Emitters. SPEs based on 2D materials such as hBN, WSe_2, and WS_2, have been widely reported.[3,5-8] The single-photon emitters based on 2D materials have some unparalleled advantages, such as the high extraction efficiency of single photons owing to the avoidance of total internal reflections and the easy integration of 2D materials with other electrical and optical components, owing to the small thickness and the dangling-bond-free van der Waals surfaces.[2,3] However, the practical applications of single-photon emitters based on 2D materials still face many challenges. First, the single-photon emission mechanism is still not clear. In the first reports of single-photon emitters of WSe_2, single-photon emissions are found on the edges of the as-exfoliated WSe_2 flakes.[3,5,6,8] The occurrence is not controllable in this way. Later on, researchers show that by creating artificial strain, basically through the tenting of WSe_2 and WS_2 on nanopillars, single-photon emitters can be created with determined positions.[9,10] However, the emission wavelength and emitter number per location are still random or poorly controlled. All these SPEs rely on strain or defects that are randomly occurred to trap excitons for single-photon emissions. For example, even though stain engineering by nanopillar tenting technique has already been able to induce single-photon emission at the designed location with 200 nm accuracy, the emission wavelength is still randomly distributed over a broad range. To realize SPEs with high indistinguishability for practical applications, it is imperative to controllably create defects in 2D materials and gain a comprehensive understanding of the single-photon emission performance. Besides, compared with the state-of-art single-photon emitters based on III-V quantum dots, there still needs more researches to improve the single photon emission purity, indistinguishability. The ability to control the emission time by pulsed laser excitation will also be valuable. The brightness also needs improvements.

Even though it still faces many challenges now, single-photon emitter based on 2D materials possesses great potential applications in quantum photonics. The in-depth exploration of the emission mechanism is vital, which is also meaningful for further improvement over the emission performance and controllability. The related advancement could also provide fundamental building blocks to photon-based quantum information and computation. Currently, the emission locations can be controlled by strain location, but the emission wavelength is not controlled. Whether the emission wavelength is related to the strain strength or the local defect is still under intense debate. The integration of strain technique and controllably created defects could be used to check the role played by defects in single photon emission. The material tested can be WSe_2, WS_2. Strain can be introduced by the usually used nanopillars or even complex structures such as nano-ring to introduce different strain distributions. Defects can be created by plasma treatment or ion/particle bombardment. Depending on the defect creation method, different types of defects will be created, such as substitution, transition metal, or chalcogen vacancies, which may correspond to different emission wavelength and brightness. The related research may resolve the mysterious emission mechanism in 2D materials and greatly promote the advancement of single-photon emission performances.

Charge Density Wave Engineering. Another interesting research area can be the engineering of charge density wave materials. Charge density wave is a periodic disruption of the electron charge density associated with periodic lattice distortion.[16] Charge density wave has been observed a wide range of materials, such as TaS_2, $TaSe_2$, $NbSe_2$.[15,29,34] The formation of charge density wave involves the strong coupling between electron and phonons. Metal-insulator transition can be induced, which can be used as memory devices to store information.[32,33] The Peierls transition of 1D material is often used as an analogy of CDW. However, researches suggest that the related Fermi surface nesting is not the main drive for the formation of charge density wave.[19,22] Instead, phonon instability is a significant signature of charge density wave formation.[19,22] Also, whether Mott insulating caused by the strong electron-electron interaction is an important factor or to what extent it is important to the phase transition is still unknown.[15] Besides, the layer stacking influence is also under debate.[26,27] Some proposed that to form an insulator, the stacking should have an even-layer supercell.[26] But some experiments suggest that the supercell layer changes during the different phase stages from 3 layers to 6 layers depending on the temperature.[17] Some also report that Mott bandgap can be observed using LDA+U calculations with spin-orbit coupling.[27] Even though with all these uncertainties, density functional theory has already been able to correctly predict the charge density wave order with a calculated energy reduction. The formation mechanism of charge density wave in an energy perspective is already practical and worth further investigations.

Experiments have found that doping and defects can change the transition temperature and conductivity of CDW materials.[30,35] Cation doping is found to slightly suppress the onset temperature of CDW, but beyond a doping level of 10%, the commensurate state no longer exists. The anion disorder, on the other hand, does not suppress the commensurate at any doping level.[30] The defect could also cause Anderson localization to influence the conductivity. Besides the defect influence, strain and Fermi level positions are also significant factors that influence the formation of charge density wave order. Since the doping can allow for more degrees of freedom to control the phase transition, the study of which is worth conducting. One way to introduce defects in the experiment is through remote plasma treatment, such as remote N_2 plasma, which can cause anion substitution and has not yet been reported. Since the formation of charge density wave and defects could both influence the supercell structures, the Raman scattering and second harmonic generation can be altered by both factors. As a result, temperature-varied Raman scattering and SHG can be used to characterize the defect level and the influence over the phase transitions. The influence of this anion substitution can be studied by scanning tunneling microscopy and conductivity measurements. In association with these experiments, the superstructure, band, and grain boundaries can be testified and explained by first-principles calculations such as density functional theory.

3. OBJECTIVE

Defect engineering can be important for tailoring the electronic and optical properties of materials. By controllably generating defects and strain within 2D materials, the related studies will promote the understanding of various factors that determine the SPE performance and the formation of CDW. SPE with enhanced performances and CDW with additional engineering freedoms can be achieved, which can become a significant step towards the practical applications of 2D materials in quantum photonics and memory devices. Specifically, the proposed research involving both strain and defects will provide a practical guideline for manufacturing high-performance SPEs in 2D materials. The advantages of 2D materials in SPEs with high photon extraction efficiency and great flexibility in heterogeneous assembly can be fully exploited, and the full potential of 2D materials in solid-state quantum optical light emission and information storage can be realized. The achievement of controllable fabrication of SPEs with high indistinguishability can further facilitate the deployment in scalable quantum communication systems. Similarly, defects used as a knob to tune the CDW phase, in conjunction with the first-principles calculations, can reveal the contributions of electron correlations to the CDW formation and metal-insulator transition, which may shed more light on the strongly correlated system such as high-temperature superconductivity, can provide more degrees of freedom for the engineering of CDW materials.

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