Tunable Trion-Polaritons in Hybrid WS2-Plasmonic System

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***Abstract*—The profound interplay between Two-Dimensional (2D) Transition Metal Dichalcogenides (TMDs) and cavity photons has facilitated a deeper understanding of the fundamental physical properties of many-body complexes and their response to light interaction. The intrinsic properties of trions in TMD semiconductors, together with their interaction with light, remain a scientific puzzle warranting experimental investigation. Here, by integrating WS_2 flakes on a plasmonic metasurface, we demonstrate dynamically tunable weak, strong and ultra-strong couplings of trions to plasmons under ambient conditions. We demonstrate dynamically tunable couplings of trions to plasmons, demonstrating the potential to manipulate these interactions even at room temperature. In addition, the investigation extends to the effects of ultrastrong coupling on the ground state energy, providing insights into the tunable nature of trion-plasmon coupling. The results pave the way for advances in optoelectronic devices and quantum technologies that exploit the tunability of trion polaritons in hybrid WS_2-plasmonic systems.**

***Keywords*—Transition Metal Dichalcogenides (TMDs), trion, tunable strong coupling**

I. INTRODUCTION

The intricate interplay between Two-Dimensional (2D) Transition Metal Dichalcogenides (TMDs) and cavity photons has deepened our understanding of the fundamental physical characteristics of many-body complexes and their interactions with light. Transition Metal Dichalcogenides (TMDs) hold appeal for trion-polaritons owing to their substantial trion binding energy [1], large oscillator strength [2, 3], and robust interaction with light [4, 5]. Unlike excitons, trion states offer electrical controllability, facilitating electromagnetic wave propagation and minimizing losses [6]. Nevertheless, experimental evidence of pure trion-polaritons in the strong coupling regime (excluding exciton involvement) in TMDs [7–9] remains elusive. Trions are widely recognized as three-body fermionic bound states, with recent theoretical and experimental endeavors focusing on elucidating trion-polariton properties in doped Two-Dimensional (2D) semiconductor materials [6, 10] to unveil the physical essence of trions. A recent theoretical model posits a four-body trion state comprising a conduction band electron-hole pair bound to an exciton, highlighting the necessity for reliable experimental data to clarify trion and trion-polariton characteristics in TMDs.

In this study, we investigate the dynamics of trions and trion-polaritons in WS_2 flakes coupled to a flexible plasmonic metasurface. We reveal dynamically tunable weak, strong and ultra-strong couplings of trions to plasmons under ambient conditions. The use of a flexible substrate allows us to immobilise the metallic metasurface during fabrication, regulate the plasmonic resonance and enhance the local field enhancement via uniaxial strain, together with electron doping of the WS_2 flake to enhance the trion oscillator strength and stability, thereby facilitating the formation of trion polaritons. Remarkably, the study achieves a normalised coupling strength greater than 10% in the ultrastrong coupling regime, where trions and plasmons give rise to mixed polariton states with significant variations in ground state properties. This groundbreaking achievement highlights the potential for mechanically tunable interactions spanning weak, strong and ultrastrong trion-plasmon interactions in atomically thin semiconductors, even at room temperature. Such advances hold promise for the realisation of controllable low-threshold lasers within plasmonic systems, heralding a new era of optoelectronic innovation.

II. TRION-POLARITON IN WS_2-PLASMON SYSTEM

To delve deeper into the intricate realm of trion polaritons, we embed the WS_2 flake on a flexible thin gold metasurface, as shown in Fig. 1(a). The flexible substrate, illustrated in Figs. 1(b) and (c), serves multiple purposes throughout the fabrication process. Not only does it serve to secure the metallic metasurface, but it also plays a crucial role in modulating the plasmonic resonance and enhancing the local field via precisely controlled uniaxial strains. In addition, the flexible substrate enables the electron doping of the WS_2 flake, a process that is crucial to enhance the strength of the trion oscillator and ensur its stability. This substrate-induced doping mechanism eliminates the nerf for additional procedures such as electrostatic gating or chemical modification of the WS_2 flake to induce the trion state, as elucidated in previous studies [11]. The gold metasurface is fabricated using a cold-drawing technique to provide densely packed nanometre-sized plasmonic gaps for strong and robust local field enhancement. During our measurement, the typical plasmonic metasurfaces used in our experimental setups have gold layer thicknesses ranging between 16 and 35 nm, thereby fostering plasmonic resonances that seamlessly coincide with the trion transition. In addition, our investigation includes bright-field optical images showcasing the bilayer WS_2 exfoliated on a PDMS tape and meticulously transferred onto the plasmonic metasurface, as delineated in Fig. 1(a).

To unravel the intricacies of trion polaritons, we meticulously analyze dark-field scattering spectra to investigate both uncoupled and coupled plasmonic systems, as exemplified in Figs. 1(b) and (c). This careful analysis reveals the interplay between exciton, trion, and plasmonic modes, with the emergence of three distinct polariton peaks. These peaks, characterized by their distinct Rabi splitting,

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serve as invaluable tools for deciphering the nuanced physical properties inherent in trion entities, as confirmed by previous scientific work [6, 10].

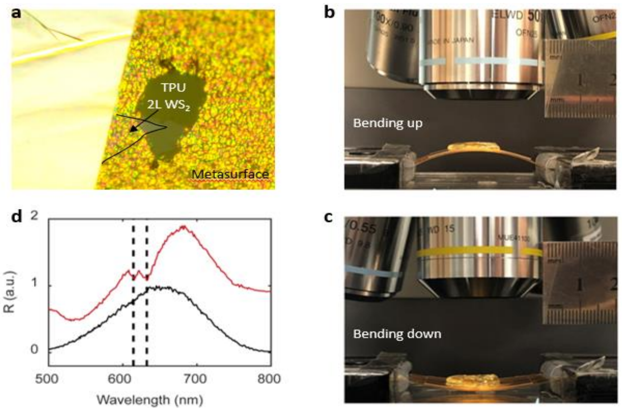

Fig. 1. Plasmonic system and associated trion-polaritons. (a) Bright-field optical microscope image of the WS_2 flake meticulously exfoliated on a PDMS tape and subsequently transferred onto the gold metasurface. 2L refers to bilayer WS_2. The black line refers to the boundary of the bilayer WS_2. Schematic of controlling the coupling between excitons, trions and plamsons by bending; (b) upward and (c) downward the flexible substrate; (d) Dark-field scattering spectra of uncoupled (black lines) and coupled (red lines) plasmonic systems integrated bilayer WS_2. The Rabi splitting is clearly visible in the strong coupling scattering spectrum.

III. Tunable Trion-Polaritons

A. *Tunable Trion-Polaritons in Bilayer WS_2*

Uniaxial strain is applied to tune the coupling strength in the hybrid plasmonic system. In particular, the coupling strength among excitons, trions and plamons decreases (smaller Rabi splitting in Fig. 2(a)) at strains from −1% to 1.5%, which is mainly induced by the redshift of the plasmonic resonance at tensile strains. The fits with the eigenfrequencies obtained from the diagonalization of the JC Hamiltonian analysis consistently provide perfect agreement with the experimentally extracted coupling strength at different strains (Fig. 2(b)). For strains from −1% to 1.5%, the extracted coupling strength varies from $g_x = 82$ meV< $(\gamma_{pl}+\gamma_t)/4$ and $g_t = 63$ meV< $(\gamma_{pl}+\gamma_t)/4$ to $g_x = 127$ meV> $(\gamma_{pl}+\gamma_x)/4$ and $g_t = 89$ meV< $(\gamma_{pl}+\gamma_x)/4$, and the trion-plasmon remains in the weak coupling regime over the applied strain range.

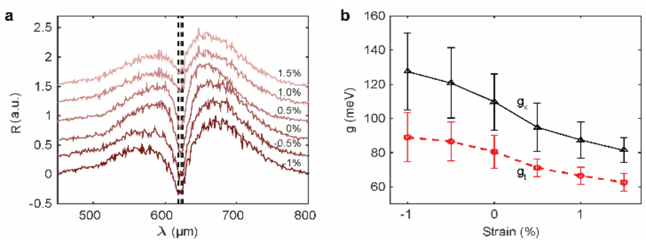

Fig. 2. Tunable exciton- and trion-polaritons within the bilayer WS_2 configuration. (a) Dark-field scattering spectra of the bilayer WS_2 integrated into the plasmonic system with strain varied from −1% to 1.5%. Black dashed lines serve to delineate the wavelengths corresponding to exciton and trion entities, providing valuable insight into their spectral behaviour under varying strain conditions; (b) Coupling strength as a function of the strain for exciton (g_x) and trion (g_t).

B. *Tunable Trion-Polaritons in Four-Layer WS_2*

The comprehensive analysis of the scattering spectra of the four-layer WS_2-based plasmonic system, plotted against laser wavelength and applied strain, is meticulously depicted in Fig. 3(a). Here, the vertical dashed line symbolizes the pivotal trion energy, which serves as a reference point for understanding the observed spectral features. Moving on to Fig. 3(a), (b) detailed investigation follows, where the extracted polariton dispersions of the interconnected plasmonic system are rigorously fitted to the theoretical framework of Hopfield Hamiltonian transition energies. In particular, the influence of compressive strain on the induction of Rabi splitting between trions and plasmons and, conversely, the mitigating effect of tensile strain on this phenomenon, similar to the trends observed in Fig. 2, are elucidated. It is imperative to acknowledge that the observed enhancement of light-matter coupling under compressive strain is mainly due to the enhanced local plasmonic field enhancement, which is attributed to the concomitant reduction of the plasmonic gap size. Furthermore, a comprehensive analysis of the normalised coupling strength as a function of uniaxial strain is meticulously presented in Fig. 3(c) (left panel). Remarkably, when the compressive strain is attenuated to −2%, the normalised coupling strength g_t/ω_t increases to an impressive 11.3%, highlighting the dynamic nature of the coupling mechanism. Conversely, as the tensile strain increases to 2%, there is a notable decrease in the normalised coupling strength to approximately 9.8%, further highlighting the strain dependent nature of the observed phenomena.

The experimental data also allow us to calculate how ultrastrong coupling changes the ground state energy, since the standard approximation breaks down in the ultrastrong coupling regime. The number of excitations in a conserved system leads to a ground state containing virtual excitations. The ground state modification is calculated from the obtained polariton ($\omega_\pm$), trion (ω_t) and bare plasmon (ω_p) energies as $\delta E_g = \hbar(\omega_+ + \omega_- - \omega_p - \omega_t)/2$ which is proportional to the square if the coupling strength (g_t^2) [12]. The obtained ground-state energy modification, ranging from 0.38% to 0.55% (Fig. 3(c), right panel), is a characteristic signature of tunable ultrastrong trion-plasmon coupling. Through this multifaceted analysis, a comprehensive understanding of the intricate interplay between structural dynamics and optical response within the four-layer WS_2 system is achieved, laying the groundwork for further exploration and optimisation of trion-polariton phenomena.

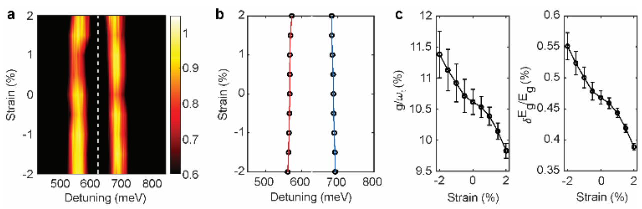

Fig. 3. Tunable trion-polaritons in four-layer WS_2. a, Dark-field scattering spectra plotted against laser wavelength and applied strain. The white dashed line marks the trion wavelength. b, Fit of the measured trion-polariton dispersion of the coupled plasmonic system to the Hopfield Hamiltonian transition energies. Circles denote polariton energies meticulously extracted from experimental scattering peaks, while lines delineate calculated polariton dispersions, offering a comprehensive comparison between theory and observation. c, Normalized coupling strength and normalized ground state modification as a function of uniaxial strain.

IV. Conclusion

We study the dynamics of trions and trion-polaritons in WS_2 flakes coupled to a flexible plasmonic metasurface. By incorporating WS_2 flakes onto a plasmonic metasurface, we demonstrate dynamically tunable weak, strong and ultra-strong couplings of trions to plasmons under ambient conditions. This approach offers a promising advancement towards the realisation of controllable low-threshold lasers in plasmonic systems. In addition, the flexible substrate used in this setup allows immobilisation of the metallic metasurface during fabrication, regulation of the plasmonic resonance and local field enhancement by uniaxial strain, and electron doping of the WS_2 flake to enhance the trion oscillator strength and stability. Dark-field scattering spectra are used to study uncoupled and coupled plasmonic systems, revealing three polariton peaks that facilitate the study of trion physical properties.

We also investigate how the ultrastrong coupling affects the ground state energy, with the obtained ground state energy modification ranging from 0.38% to 0.55%. This characteristic signature of tunable ultrastrong trion-plasmon coupling highlights the potential of the proposed system for various applications. In addition, we present the tunability of trion polaritons in both two-layer and four-layer WS_2 systems under applied strain. Compressive strain is found to promote Rabi splitting between trions and plasmons, while tensile strain reduces Rabi splitting. These results provide valuable insights into the manipulation of trion-polariton properties in hybrid WS_2-plasmonic systems, paving the way for advances in optoelectronic devices and quantum technologies.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

T. Wu and Y. Luo conceived the idea. T. Wu performed the experimental measurements. All authors had approved the final version.

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