



# Unveiling room temperature upconversion photoluminescence in monolayer WSe<sub>2</sub>

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**Abstract:** Upconversion photoluminescence (UPL) is a phenomenon describing an anti-Stokes process where the emitted photons have higher energy than the absorbed incident photons. Transition metal dichalcogenides (TMDCs) with strong photon-exciton interactions represent a fascinating platform for studying the anti-Stokes UPL process down to the monolayer thickness limit. Herein, we demonstrate room-temperature UPL emission in monolayer WSe<sub>2</sub> with broadband near-infrared excitation. The measured excitation power dependence of UPL intensity at various upconversion energy gains unveils two distinguished upconversion mechanisms, including the one-photon involved multiphonon-assisted UPL process and the two-photon absorption (TPA) induced UPL process. In the phonon-assisted UPL regime, the observed exponential decay of UPL intensity with the increased energy gain is attributed to the decreased phonon population. Furthermore, valley polarization properties of UPL emission with circular polarization excitation is investigated. The demonstrated results will advance future photon upconversion applications based on monolayer TMDCs such as night vision, semiconductor laser cooling, and bioimaging.

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## 1. Introduction

Photoluminescence (PL) is a light-matter interaction process in which low energy photons are emitted after the absorption of high energy photons, which normally obeys Stokes' law of luminescence [1]. Contrary to PL, upconversion photoluminescence (UPL) is an anti-Stokes process where the emitted photons have higher energy than the absorbed incident photons, due to additional energy gain triggered by series of other events like phonon participation [2] or multiphoton absorption [3,4]. During the UPL process, excitation photon ( $\hbar\omega_{pump}$ ) couples to a real or virtual intermediate state and eventually results in photon emission at higher energy ( $\hbar\omega_{emission}$ ) after gaining additional energy  $\Delta E$  with energy conservation [5,6]. In solid-state systems, phonon-participated UPL process is a one-photon involved linear process, which is distinguished from other nonlinear optical processes such as two-photon absorption (TPA) induced PL and second-harmonic generation (SHG) emission [7]. UPL phenomena have been reported to occur efficiently in certain materials like organic dyes [4,6], rare-earth-doped materials [3], quantum wells [8] and quantum dots [9]. In recent years, UPL has attracted research interests due to its potential usefulness in varieties of applications such as photoluminescence bioimaging [10,11], photovoltaic energy conversion [12], lasers [13], displays [14] and optical refrigeration of solids [15].

Study of nanomaterials in the field of material science particularly two-dimensional (2D) materials has blossomed over the past years. Successful exfoliation of graphene with exceptional electrical [16], optical [17], thermal [18] and mechanical properties [19] flickered a trend on the exploration of 2D materials. 2D transition metal dichalcogenides (TMDCs) with chemical formula MX<sub>2</sub> (M = Mo, W, Re and X = S, Se, Te), where transition metal M is sandwiched