

RESEARCH ARTICLE

Mirror-Induced Electromagnetic Asymmetry Enables Greatly Enhanced Second-Harmonic Generation From 2H-Phase WS₂ Nanoparticles

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ABSTRACT

Second-harmonic generation (SHG) in centrosymmetric materials is forbidden within the electric-dipole (ED) approximation, posing a fundamental challenge for nonlinear nanophotonics. Here, we demonstrate an effective approach to activate and enhance SHG from centrosymmetric WS₂ nanoparticles using a nanoparticle-on-mirror architecture. By placing hexagonal WS₂ nanoparticles onto a metallic film, mirror-induced field asymmetry and strong nanoscale field confinement create an inversion-asymmetric local electromagnetic environment, enabling pronounced SHG emission from individual nanoparticles. Excitation of the ED-dominated hybrid nanoparticle-mirror resonance leads to a maximum SHG enhancement factor of approximately 2000 compared with bulk WS₂ under identical excitation conditions. Polarization-, power-, and wavelength-dependent measurements confirm the second-order nonlinear nature of the emission and reveal a clear correlation between linear optical resonances and SHG enhancement. Numerical simulations further reveal that the metallic mirror induces strong asymmetric electric-field localization at the WS₂-substrate interface, providing a physical origin for the observed nonlinear response. These results establish mirror-induced electromagnetic asymmetry as an effective strategy for enabling nonlinear optical processes in centrosymmetric two-dimensional materials and provide a versatile platform for designing efficient nonlinear nanophotonic devices through substrate engineering and resonant mode control.

1 | Introduction

Second-harmonic generation (SHG) is a fundamental nonlinear optical phenomenon that serves as the basis for numerous applications, including frequency conversion, bioimaging, and

on-chip photonic signal processing [1–3]. At the nanoscale, the realization of efficient SHG is particularly critical for the advancement of compact and integrable nonlinear optical devices [4, 5]. Nevertheless, within the framework of the electric-dipole (ED) approximation, SHG is forbidden in centrosymmetric materials,

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thereby substantially restricting the selection of materials suitable for nonlinear nanophotonic applications [6, 7]. Consequently, overcoming this symmetry-imposed limitation remains a pivotal challenge in the development of nanoscale nonlinear optical platforms.

Two-dimensional transition metal dichalcogenides (TMDs) have garnered significant interest as promising materials for nanophotonics due to their pronounced light–matter interactions and compatibility with nanophotonic architectures [8–14]. Monolayer TMDs, which lack inversion symmetry, exhibit strong SHG [15–17]. However, their atomic-scale thickness constrains the overall nonlinear conversion efficiency [18, 19]. Conversely, multilayer and bulk TMDs with the thermodynamically stable 2H stacking configuration are centrosymmetric, resulting in the suppression of ED-allowed SHG and posing challenges for achieving efficient nonlinear emission [20–22]. Although noncentrosymmetric 3R-stacked TMDs intrinsically support SHG even in multilayer forms [23–25], the 2H phase remains the most prevalent and stable crystal structure. This circumstance motivates the investigation of alternative strategies to activate nonlinear optical responses in centrosymmetric TMD systems [26–29].

An alternative and comparatively unexplored route of enabling SHG in centrosymmetric nanostructures is to engineer symmetry breaking through the electromagnetic environment rather than through the material itself [6, 30, 31]. In particular, nanoparticle-on-mirror (NPoM) structures can produce hybridized gap modes with ultrasmall mode volumes and intense, spatially asymmetric fields through mirror-image interactions [32–34]. The confinement and modal symmetry of these modes depend strongly on the particle morphology and nanogap geometry, enabling strong light–matter coupling and enhanced nonlinear optical processes [35–40]. Previous NPoM-based SHG studies have exploited resonance matching in intrinsically noncentrosymmetric monolayer TMDs [41, 42] and light-induced symmetry breaking of metallic-surface nonlinearities [35, 43]. Despite these advances, experimental demonstrations of mirror-induced activation of SHG in centrosymmetric TMD nanostructures remain scarce.

In this work, we demonstrate mirror-induced activation and enhancement of SHG from centrosymmetric 2H-phase WS₂ nanoparticles using an NPoM architecture. Unlike previous metallic NPoM systems or NPoM cavities incorporating intrinsically noncentrosymmetric monolayer WS₂, the hexagonal WS₂ nanoparticle here serves as both the semiconductor resonator and nonlinear medium and is integrated with a SnO₂/Ag/SiO₂ substrate, where the mirror creates strong vertical field asymmetry and renders the WS₂/air and WS₂/SnO₂ interfaces electromagnetically inequivalent, preventing complete cancellation of their interface nonlinear polarizations, while strong z-directed field gradients may activate nonlocal bulk-like contributions. Combining optical measurements with numerical simulations, we show that excitation of an ED-dominated hybrid nanoparticle–mirror resonance produces pronounced SHG emission, with the detected SHG intensity enhanced by a factor of approximately 2000 relative to the thick bulk WS₂ reference under identical excitation and detection conditions. These results establish mirror-induced electromagnetic asymmetry as an effective strategy for activating nonlinear optical processes in centrosymmetric two-dimensional materials and provide a versatile platform for engineering

nonlinear nanophotonic functionalities through substrate and resonance design.

2 | Results and Discussion

2.1 | Electromagnetic-Asymmetry-Enabled SHG From Centrosymmetric WS₂ Nanoparticles

It is well established that SHG requires a nonvanishing second-order nonlinear susceptibility $\chi^{(2)}$, which is forbidden in centrosymmetric media under the ED approximation. Nevertheless, in nanoscale structures coupled to a metallic mirror, the local electromagnetic environment can become inversion asymmetric because of finite-size effects and mirror-induced field redistribution [43]. This electromagnetic asymmetry facilitates unconventional SHG even in centrosymmetric materials. Figure 1a schematically depicts this phenomenon using a hexagonal WS₂ nanoparticle situated on a metallic thin film. In this dielectric NPoM configuration, the metallic film enhances and spatially redistributes the local electric field at the lower WS₂/SnO₂ interface, thereby creating an inversion-asymmetric electromagnetic environment that enables SHG in centrosymmetric WS₂. It should be emphasized that the metallic mirror does not modify the intrinsic centrosymmetric lattice of 2H-phase WS₂. Instead, the NPoM geometry creates an inversion-asymmetric electromagnetic environment by making the upper WS₂/air and lower WS₂/SnO₂ interfaces inequivalent. The image-charge-mediated interaction with the Ag mirror strongly localizes the fundamental field near the lower interface. Consequently, the surface nonlinear polarizations generated at the two interfaces possess substantially different amplitudes and cannot completely cancel, giving rise to a finite net interface ED contribution. In contrast, thick bulk WS₂ flakes remain intrinsically centrosymmetric, and their lower WS₂/SnO₂ interface is optically remote from the upper surface from which most of the detected SHG is collected; consequently, their weak SHG signal primarily arises from surface- and edge-related contributions [21, 28].

To experimentally validate this phenomenon, WS₂ nanoparticles were synthesized on a SnO₂/Ag/SiO₂ substrate using a solution-based method, as described in our previously reported procedure [44, 45]. The resulting hexagonal WS₂ nanoparticles, with representative long-axis lengths ranging from approximately 748 to 1145 nm, were supported by a 5 nm SnO₂ spacer layer atop a 50 nm Ag film (Figure S1). The Ag layer was chosen due to its favorable plasmonic response in the near-infrared spectral region, while the SnO₂ spacer serves to prevent Ag oxidation and reduce quenching of the nonlinear emission from the WS₂ nanoparticles [46]. Consistent with our prior findings, WS₂ nanoparticles synthesized from the powder precursor predominantly adopt the thermodynamically stable 2H phase [45], which is centrosymmetric in the multilayer limit and thus intrinsically suppresses ED-allowed SHG. Therefore, efficient SHG emission from these nanoparticles requires an inversion-asymmetric electromagnetic environment and local-field enhancement, which can be provided by the NPoM configuration [43].

Figure 1b presents the optical characterization of an individual WS₂ nanoparticle, with the structure shown in the inset scanning electron microscopy (SEM) image. The investigated nanoparticle

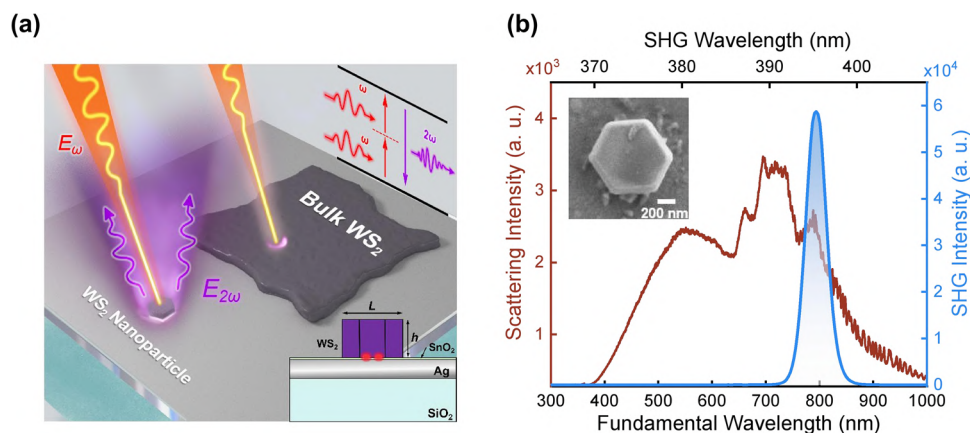


FIGURE 1 | Mirror-induced SHG from centrosymmetric WS_2 nanoparticles on a metallic substrate. (a) Schematic illustration of enhanced SHG from a WS_2 nanoparticle supported on a $\text{SnO}_2/\text{Ag}/\text{SiO}_2$ substrate. The SnO_2 spacer (~ 5 nm) separates the nanoparticle from the Ag film (~ 50 nm thick). The inset shows the side-view geometry with nanoparticle long-axis length L and height h . The upper-right panel illustrates the SHG process under excitation at frequency ω , generating emission at 2ω . (b) Experimental dark-field scattering spectrum (red curve) and the corresponding SHG spectrum (blue shaded curve) measured from an individual WS_2 nanoparticle on the $\text{SnO}_2/\text{Ag}/\text{SiO}_2$ substrate. The inset shows the SEM image of the measured WS_2 nanoparticle.

has a long-axis length of approximately 1115 nm and a height of approximately 135 nm. The measured scattering spectrum reveals a distinct resonance near 800 nm within the low-loss spectral window of WS_2 . Simulations further show that the corresponding hybrid resonance progressively redshifts as the effective nanoparticle size increases (see Note S2 for more details), in agreement with previous studies [44, 45, 47]. The nanoparticle analyzed was selected based on the corresponding dark-field microscopy image (Figure S3). At wavelengths below approximately 700 nm, the scattering response is strongly affected by the intrinsic excitonic and interband absorption of WS_2 and is therefore excluded from the subsequent analysis of the low-loss optical resonance [44, 45, 48]. Under resonant excitation at approximately 790 nm, a pronounced SHG signal centered near 395 nm is observed, confirming resonance-assisted frequency doubling in the WS_2 nanoparticle.

2.2 | Experimental Observation and Enhancement of SHG

Having established resonance-enhanced SHG from WS_2 nanoparticles deposited on the $\text{SnO}_2/\text{Ag}/\text{SiO}_2$ substrate, we proceeded to investigate the spatial distribution and polarization properties of the nonlinear emission utilizing the experimental configuration depicted in Figure 2a. Due to the solution-based fabrication method, WS_2 nanoparticles were randomly dispersed across the substrate surface. Consequently, a two-photon laser scanning confocal microscope was employed to efficiently identify nanoparticles exhibiting strong SHG signals. A representative scanning image is presented in Figure 2b, revealing a ring-shaped SHG intensity distribution that corresponds to the spatially localized nonlinear emission from individual nanoparticles. Subsequently, polarization-resolved SHG measurements were conducted on a selected WS_2 nanoparticle under resonant excitation at 790 nm (Figure 2c). Under a fixed polarization of the incident fundamental beam, the collected SHG intensity exhibits a pronounced two-lobed dependence on the analyzer angle, indicating

predominantly linearly polarized SHG emission along a preferred in-plane polarization direction. Similar polarization-resolved SHG responses have been observed in TMD nanostructures with anisotropic geometries or optical resonances, although their geometrical and crystallographic symmetries differ from those of the present 2H-phase WS_2 nanoparticle [21, 44, 45, 49]. This polarization behavior reflects the combined effects of nanoparticle geometry and substrate-induced electromagnetic asymmetry. In all subsequent measurements, the excitation polarization was fixed to ensure consistent experimental conditions.

Figure 2d presents a comparison of SHG spectra obtained from WS_2 nanoparticles deposited on various substrates under identical excitation power (~ 1 mW). Two configurations involving Ag-based substrates are examined: WS_2 nanoparticles exhibiting an optical resonance near 790 nm (“Ag res.”) and those off-resonant at this wavelength (“Ag off-res.”). For comparison, WS_2 nanoparticles supported on indium tin oxide (ITO)/ SiO_2 substrates, with and without a corresponding optical resonance (see Figure S4 for spatial SHG mapping), as well as a bulk WS_2 flake on $\text{SnO}_2/\text{Ag}/\text{SiO}_2$ (see Figure S5), are also included. All nanoparticle configurations demonstrate stronger SHG signals than the bulk WS_2 flake, underscoring the significance of nanoscale geometry. Notably, the WS_2 nanoparticle on the Ag thin film that supports an optical resonance yields the most intense SHG among all samples. Under otherwise identical conditions, the SHG intensity of the resonant WS_2 nanoparticle is approximately 1.8 times that of the off-resonant WS_2 nanoparticle on the Ag thin film. Furthermore, both Ag-supported configurations exhibit stronger SHG than the samples under the other substrate conditions. These comparisons indicate that the mirror-induced interfacial asymmetry is primarily responsible for enabling the effective second-order response, whereas spectral overlap with the hybrid nanoparticle–mirror resonance further enhances and localizes the fundamental field near the lower WS_2/SnO_2 interface, thereby amplifying the generated SHG (see Note S6 for further details). Consistent with this interpretation, multilayer WS_2 flakes on the $\text{SnO}_2/\text{Ag}/\text{SiO}_2$ substrate exhibit a more pronounced SHG signal

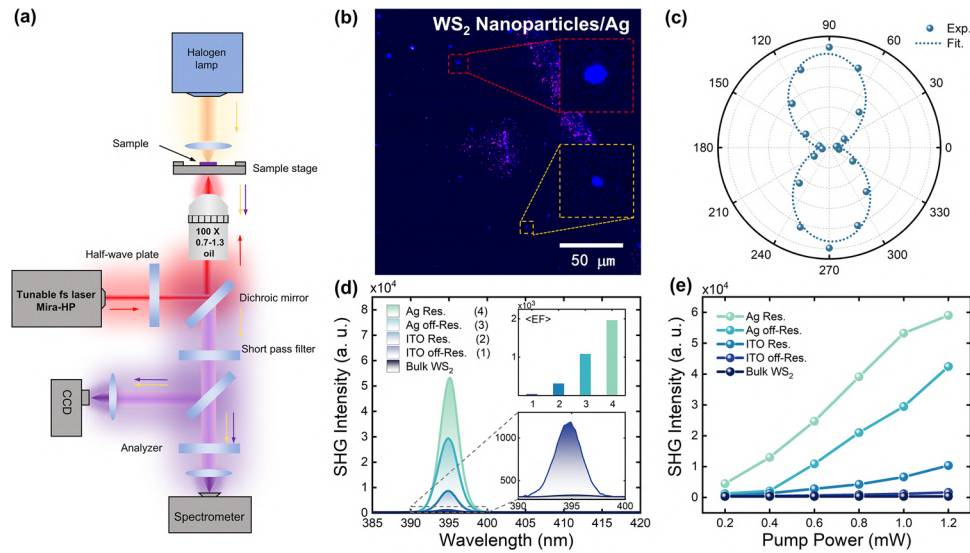


FIGURE 2 | Optical resonance enabled the enhancement of SHG from WS₂ nanoparticles. (a) Schematic of the experimental setup for excitation and collection of SHG in a reflection geometry using a wavelength-tunable femtosecond laser. (b) Laser-scanning confocal microscopy image of SHG emission from WS₂ nanoparticles on the SnO₂/Ag/SiO₂ substrate under an excitation power of 1 mW. Enlarged views of representative nanoparticles highlight the spatial localization of the SHG signal. (c) Analyzer-angle-dependent SHG intensity of an individual WS₂ nanoparticle on the SnO₂/Ag/SiO₂ substrate under resonant excitation at 790 nm with fixed incident polarization. Circles denote experimental data, while the dotted curve represents the corresponding theoretical fit. (d) Comparison of SHG spectra from WS₂ nanoparticles on different substrates under resonant and off-resonant excitation at the same incident power (~1 mW). The SHG spectrum from a bulk WS₂ flake is included for reference. The inset shows the enhancement factor, defined as the ratio of the peak SHG intensity of the nanoparticle to that of bulk WS₂ under identical excitation conditions. An enlarged view of the low-intensity region is included to clearly display the weak SHG spectra from the ITO off-resonant nanoparticle and the thick bulk WS₂ reference. (e) Excitation power dependence of the SHG peak intensity for WS₂ nanoparticles on different substrates.

than bulk WS₂ flakes on the same substrate (Figure S7), indicating that reducing the WS₂ thickness allows the mirror-induced interfacial symmetry breaking to contribute more effectively to the measured nonlinear response. Nevertheless, the SHG intensities of both flake samples remain substantially lower than that of the resonant WS₂ nanoparticle, further underscoring the additional enhancement provided by the resonance-induced local field.

To quantitatively assess the nonlinear enhancement, an enhancement factor $\langle EF \rangle$ is defined as the ratio of the peak SHG intensity of the nanoparticle to that of the bulk WS₂ flake, measured under identical excitation conditions (see inset of Figure 2d). Although absolute SHG conversion efficiencies and effective nonlinear coefficients provide useful quantitative benchmarks for calibrated nonlinear nanoantenna systems [50], their determination requires calibration of the generated SHG power and the overall collection and detection response. In the present setup, however, the wavelength-dependent collection and detection efficiencies were not independently calibrated, preventing a reliable conversion of the measured spectrometer counts into absolute SHG power. Accordingly, $\langle EF \rangle$ is used here as a relative measure of the detected SHG enhancement under identical excitation and detection conditions. The resonant WS₂ nanoparticle on the Ag film exhibits $\langle EF \rangle \sim 2000$, which substantially surpasses that observed for WS₂ nanoparticles on ITO/SiO₂ substrates. Notably, even in the absence of a pronounced optical resonance, WS₂ nanoparticles on the Ag film produce SHG signals nearly three times stronger than those from resonant nanoparticles on ITO/SiO₂. This finding suggests that the metallic mirror itself plays a crucial role in establishing an inversion-asymmetric elec-

tromagnetic environment through mirror-induced image fields and interface-localized electromagnetic enhancement.

Figure 2e illustrates the dependence of SHG intensity on excitation power under various conditions. For WS₂ nanoparticles deposited on the SnO₂/Ag/SiO₂ substrate, the SHG signal exhibits a rapid increase with excitation power, irrespective of the presence of optical resonance. Conversely, WS₂ nanoparticles on ITO/SiO₂ substrates show a more gradual increase in SHG intensity, while bulk WS₂ flakes display negligible SHG even at higher excitation powers. According to prior studies, the SHG enhancement factor for WS₂ nanoparticles exhibiting resonance on ITO substrates is approximately 30 relative to the monolayer WS₂ [45]. In comparison, the enhancement factor for WS₂ nanoparticles on the Ag film supporting optical resonances is estimated to be approximately 150 under comparable excitation conditions. These findings indicate that the NPom configuration constitutes an effective approach for achieving strong nonlinear emission from centrosymmetric WS₂.

2.3 | Linear Optical Resonances and Mirror-Induced Electromagnetic Asymmetry

To reveal the physical origin of the enhanced SHG, we conducted numerical simulations of the linear optical response of WS₂ nanoparticle/Ag film heterostructures. The nanoparticles were modeled using an idealized hexagonal geometry based on the SEM-measured dimensions. Figure 3a displays the simulated scattering spectrum of a WS₂ nanoparticle on a SnO₂/Ag/SiO₂

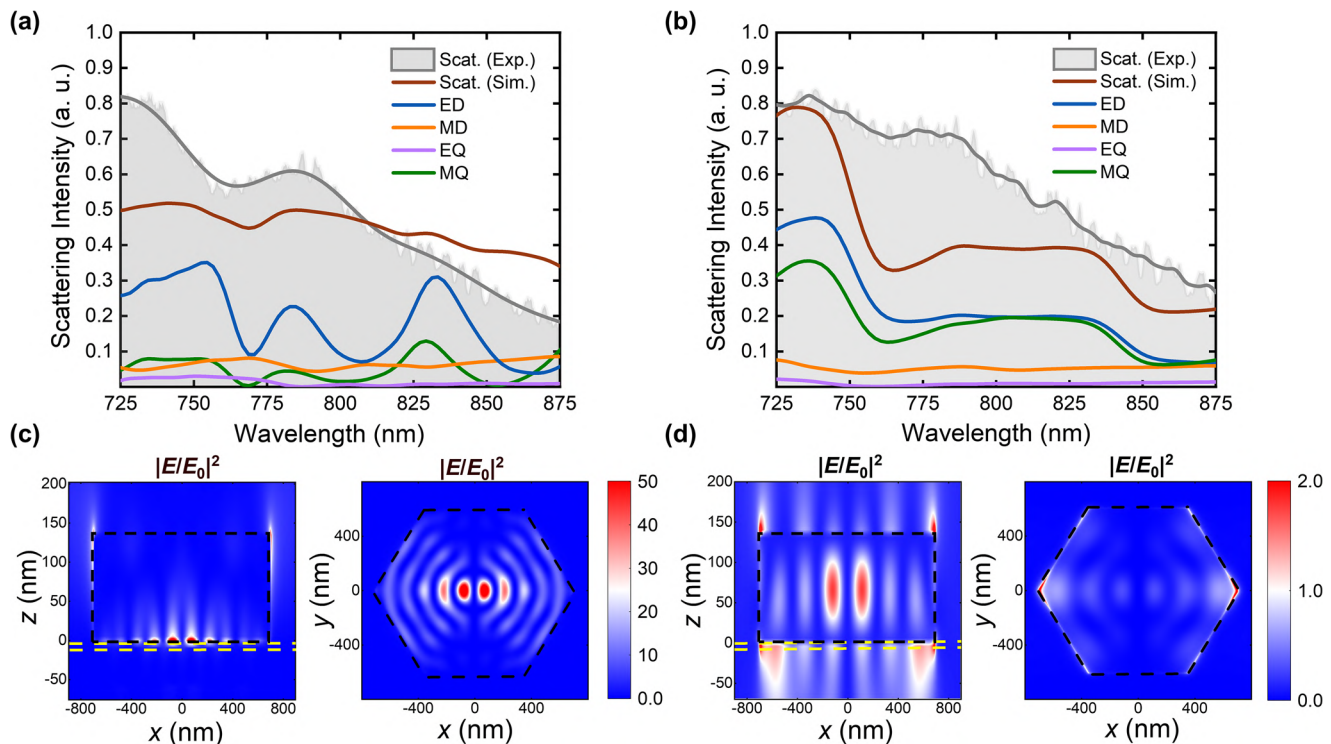


FIGURE 3 | Linear optical response of WS₂ nanoparticles on different substrates. (a,b) Scattering spectra (simulation and experiment) and the corresponding multipole decomposition of WS₂ nanoparticles supported on SnO₂/Ag/SiO₂ (a) and ITO/SiO₂ (b) substrates. (c) Simulated electric-field intensity distributions ($|E/E_0|^2$) of a WS₂ nanoparticle on the SnO₂/Ag/SiO₂ substrate at the resonant wavelength of 790 nm, shown in the x-z (side view) and x-y (top view) planes. (d) Corresponding electric-field intensity distributions of a WS₂ nanoparticle on the ITO/SiO₂ substrate at the same wavelength as in (c) for comparison. The same WS₂ nanoparticle geometry, with identical lateral dimensions and a height of 135 nm, was used in both simulations.

substrate, accompanied by a multipole decomposition analysis [51]. For visual comparison, the normalized experimental scattering spectrum, simulated total scattering spectrum, and multipole-decomposition results were overlaid in Figure 3a using different scaling factors (see more details in Figure S8). The multipole decomposition indicates that the far-field scattering near 790 nm is predominantly associated with the ED channel, while the magnetic dipole (MD), electric quadrupole (EQ), magnetic quadrupole (MQ), and higher-order contributions remain comparatively weaker. Nevertheless, the spatial field distribution is more complex than that of an isolated ED because the nanoparticle mode is hybridized with its electromagnetic image in the Ag mirror and strongly confined near the WS₂/SnO₂ interface (see more details in Figure S9). We therefore identify this resonance as an ED-dominated hybrid nanoparticle-mirror resonance rather than a purely ED Mie mode. It should be noted that although additional ED-dominated resonances appear at shorter and longer wavelengths in the simulated spectrum, their exact spectral positions and strengths are sensitive to the WS₂ refractive index and nanoparticle geometry. No comparably distinct resonances are resolved experimentally at the corresponding spectral positions, which may reflect differences between the actual nanoparticle and the idealized numerical model, including possible deviations of the actual WS₂ refractive index from the literature-derived values used in the simulations [48], as well as variations in lateral dimensions, particle height, and edge morphology. These additional simulated resonances are therefore not discussed further here. Experimental scattering spectra are

superimposed for comparison, demonstrating good agreement with the simulated resonance position and thereby validating the numerical results.

Figure 3c illustrates the electric-field intensity distributions at the resonant wavelength of 790 nm. A significant field enhancement is observed near the lower WS₂/SnO₂ interface above the Ag film, with a maximum electric-field intensity enhancement factor of approximately 50. Notably, the confined field distribution exhibits a pronounced asymmetry across the nanoparticle, and the highly inhomogeneous optical field, particularly the strong z-directed field gradient near the WS₂/SnO₂ interface, may activate nonlocal bulk-like nonlinear polarizations associated with electric-quadrupole and field-gradient terms, providing a possible nonlocal contribution to the effective second-order response (see Figure S6). Additionally, the field distribution in the x-y plane supports the assignment of the resonance as an ED-dominated hybrid mode with a complex near-field distribution, and the strong field localization and pronounced top-bottom field asymmetry further indicate that the lower WS₂ interface plays an important role in the mirror-enabled nonlinear response. For comparison, the linear optical response of a WS₂ nanoparticle on an ITO/SiO₂ substrate is presented in Figure 3b,d. Although ED and magnetic quadrupole contributions are evident in the scattering spectrum, the electric field is predominantly confined within the nanoparticle, resulting in an almost two-fold enhancement. The significantly weaker field confinement and diminished asymmetry account for the substantially lower detected SHG

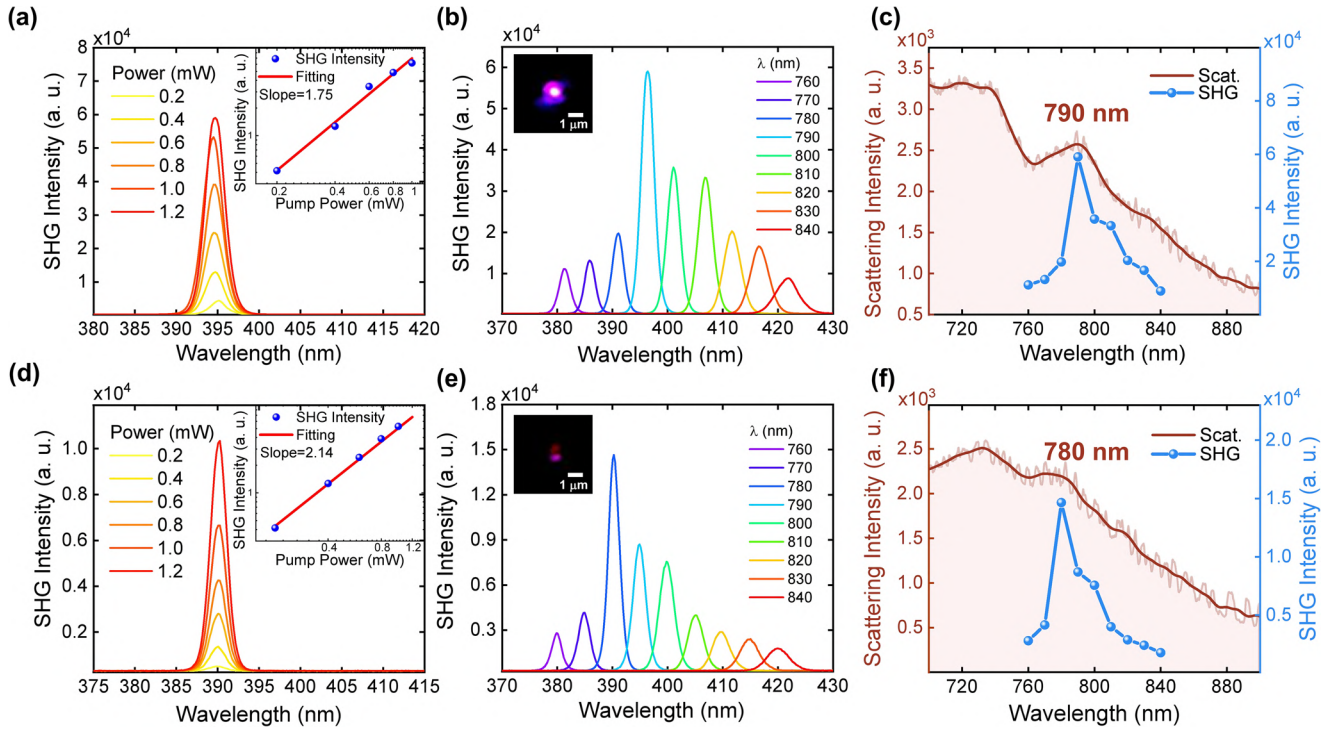


FIGURE 4 | Excitation wavelength-dependent SHG response of WS₂ nanoparticles. (a,d) SHG spectra of WS₂ nanoparticles on the SnO₂/Ag/SiO₂ (a) and ITO/SiO₂ (d) substrates, measured at different excitation powers. The inset shows the SHG peak intensities extracted from the spectra in the main panel as a function of incident power, together with the corresponding power-law fit. (b,e) Excitation wavelength-dependent SHG spectra of WS₂ nanoparticles on SnO₂/Ag/SiO₂ (b) and ITO/SiO₂ (e) substrates measured at a fixed excitation power of 1 mW. Insets show the SHG emission images recorded by a charge-coupled device (CCD). (c,f) Comparison between the linear scattering spectra and the excitation wavelength dependence of the peak SHG intensity for WS₂ nanoparticles on SnO₂/Ag/SiO₂ (c) and ITO/SiO₂ (f) substrates.

intensity of WS₂ nanoparticles on ITO/SiO₂ substrates. Taken together, these results indicate that the measured SHG is consistent with the combined contributions of the net interface ED response generated at the two inequivalent interfaces and possible nonlocal bulk-like nonlinear effects.

2.4 | Excitation-Condition Dependence and Resonance Correlation of SHG

To further validate the resonance-mediated mechanism underlying the enhancement of SHG, we conducted excitation power-dependent measurements. Figure 4a,d illustrate the relationship between excitation power and SHG intensity for WS₂ nanoparticles deposited on SnO₂/Ag/SiO₂ and ITO/SiO₂ substrates, respectively. The SHG peak intensities extracted from the power-dependent spectra in the main panel are summarized in the inset and fitted to obtain the power-law exponent. The extracted power-law exponents are approximately 1.75 and 2.14, consistent with the near-quadratic dependence expected for a second-order nonlinear optical process. An additional measurement on an independent WS₂ nanoparticle under the same conditions yields comparable power-law behavior (Figure S10), supporting the reproducibility of the SHG response across independently measured nanoparticles. The modest deviations from the ideal exponent of 2 may arise from small variations in the incident power, focal positioning, and nanoparticle alignment, uncertainty in SHG signal extraction, and particle-to-particle differences in resonance-mediated local-field enhancement. Figure 4b

illustrates the dependence of SHG on the excitation wavelength for a WS₂ nanoparticle situated on a SnO₂/Ag/SiO₂ substrate. The SHG intensity reaches its maximum near 790 nm, coinciding with the pronounced linear scattering resonance of the complete nanoparticle-mirror system. Multipole decomposition indicates that the far-field radiation of this resonance is dominated by the ED channel. Although additional spectral features appear in the ED contribution at other wavelengths, they do not correspond to comparably pronounced resonances in the experimentally measured scattering spectrum and do not produce resolvable maxima in the wavelength-dependent SHG response. This correlation is further evident in Figure 4c, where the excitation-wavelength dependence of the peak SHG intensity closely follows the corresponding scattering resonance, supporting resonant field confinement to the enhanced nonlinear response. To examine the particle-to-particle reproducibility of this spectral correlation, we performed the same measurements on two additional WS₂ nanoparticles supported on the SnO₂/Ag/SiO₂ substrate (Figure S11). Although their optical resonances occur at different wavelengths because of particle-to-particle geometrical variations, the maximum SHG response of each nanoparticle remains spectrally correlated with its corresponding linear scattering resonance. This consistent resonance-SHG correspondence across independently measured nanoparticles further supports the resonance-mediated origin of the observed SHG enhancement.

To demonstrate the general applicability of this mechanism, wavelength-dependent SHG measurements were conducted

on WS₂ nanoparticles supported on ITO/SiO₂ substrates (Figure 4e,f). In this context, the SHG enhancement similarly corresponds to the optical resonance of the nanoparticle–substrate system, although with a markedly reduced magnitude. These findings suggest that resonance-enhanced nonlinear responses are an intrinsic characteristic of WS₂ nanoparticles, while field confinement induced by metallic mirrors offers an additional, highly effective approach to augmenting SHG intensity. The integration of nanoscale geometry, substrate engineering, and low-loss optical resonances thus provides a versatile platform for enhancing nonlinear optical processes in centrosymmetric TMD nanostructures. It is important to note that the observed resonance-assisted SHG intensity enhancement in individual nanoparticles remains limited by the relatively low quality (*Q*) factor of the hybrid nanoparticle–mirror resonance. Further improvements may be realized by employing higher-*Q* optical resonances, such as bound states in the continuum (BICs) [52–55], guided-mode resonances (GMRs) [56, 57], or hybrid Mie–plasmonic resonance [58, 59], achievable through the design of advanced TMD nanostructures fabricated via advanced nanofabrication techniques [60–62].

3 | Conclusions

In conclusion, we have demonstrated an effective strategy for realizing strong SHG from centrosymmetric WS₂ nanoparticles by employing an NPoM architecture. While bulk WS₂ inherently lacks ED-allowed SHG due to its inversion symmetry, the combination of nanoscale geometry and a metallic mirror creates an inversion-asymmetric electromagnetic environment and intense electromagnetic field confinement at the WS₂ nanoparticle–mirror interface, thereby producing pronounced SHG emission from individual nanoparticles. Comparative analyses between metallic and dielectric substrates indicate that mirror-induced electromagnetic asymmetry enables a finite effective second-order response, whereas optical resonance provides substantial additional enhancement through local-field confinement. Resonant excitation of an ED-dominated hybrid nanoparticle–mirror resonance results in a maximum SHG enhancement factor of approximately 2000 relative to bulk WS₂, consistent with numerical simulations that demonstrate strong asymmetric field localization. Power- and wavelength-dependent measurements further validate the second-order nature of the emission and its direct association with linear optical resonances. These findings establish mirror-induced electromagnetic asymmetry as a viable strategy for activating nonlinear optical processes in centrosymmetric two-dimensional materials and offer a versatile platform for the design of nonlinear nanophotonic devices through substrate engineering and resonant mode manipulation.

4 | Methods

4.1 | Sample Preparation and Characterization

WS₂ nanoparticles were synthesized from commercially available WS₂ powder (Sixcarbon Technology, Shenzhen, China). The synthesis procedure followed the same protocol reported in our previous studies. The resulting WS₂ nanoparticles were dispersed and subsequently deposited onto prefabricated SnO₂/Ag/SiO₂ substrates. The morphology and geometry of

individual WS₂ nanoparticles were characterized using scanning electron microscopy (SEM, Zeiss), which provided the dimensional parameters used for optical measurements and numerical simulations.

4.2 | Optical Characterization

The optical properties of individual WS₂ nanoparticles supported by Ag-based and dielectric substrates were measured using an inverted optical microscope (Observer A1, Zeiss) equipped with both broadband white-light and femtosecond laser excitation sources. A wavelength-tunable femtosecond laser (Mira 900S, Coherent) with a pulse duration of 130 fs and a repetition rate of 76 MHz was used to excite the WS₂ nanoparticles at different excitation wavelengths. The fundamental excitation beam was linearly polarized along the fixed laboratory *y*-axis and maintained at this polarization unless otherwise specified. The fundamental laser beam was focused onto an individual WS₂ nanoparticle using a 100× objective with a numerical aperture of 0.7. The full width at half maximum of the focal intensity profile at the sample plane was approximately 1 μm, as characterized previously using the same optical configuration [63]. The average incident power was measured at the sample plane. Taking the measured focal-spot width as the effective diameter of a circular illumination area, an average power of 2 mW corresponds to a nominal average incident intensity of approximately 2.55×10^5 W/cm². The emitted SHG signals were collected by the same objective lens in a backscattering configuration and directed either to a spectrometer (SR-500i-B1, Andor) for spectral analysis or to a CCD camera (DU970N, Andor) for SHG radiation imaging. For polarization-resolved measurements, the incident polarization was kept fixed while an analyzer in the SHG collection path was rotated. For spectral measurements, the collected nonlinear emission was passed through a short-pass filter to suppress residual fundamental-frequency light before entering the spectrometer. The SHG intensity was obtained by integrating the isolated SHG peak after background subtraction and was used for relative comparison under identical measurement conditions. For spatial mapping and rapid identification of SHG-active nanoparticles, a two-photon laser scanning confocal microscope (A1MP, Nikon) was employed to excite and detect SHG emissions from individual WS₂ nanoparticles.

4.3 | Numerical Simulation

The linear optical response and electric-field enhancement of WS₂ nanoparticles supported on SnO₂/Ag/SiO₂ substrates were simulated using the finite-difference time-domain (FDTD) method. In the simulations, a hexagonal WS₂ nanoparticle with a thickness of 135 nm and a long-axis length of 1200 nm was modeled based on dimensions extracted from SEM images. The refractive index of WS₂ was adopted from literature values [48]. The Ag film thickness was set to 50 nm, with its dielectric function taken from reported experimental data [64]. The SnO₂ spacer layer was modeled with a thickness of 5 nm and a refractive index of 1.9. A minimum mesh size of 1 nm was applied in regions containing the WS₂ nanoparticle and the Ag film to accurately resolve near-field distributions, while self-adaptive meshing was used elsewhere to optimize computational efficiency.

Author Contributions

Sheng Lan: supervision, funding acquisition, writing – review and editing, resources. **Shulei Li:** funding acquisition, formal analysis, data curation. **Jiancheng Xu:** methodology, software. **Jingdong Chen:** funding acquisition, formal analysis, software. **Fu Deng:** writing – review and editing, writing – original draft, funding acquisition, project administration, conceptualization, supervision. **Yuheng Mao:** methodology, software, formal analysis. **Chaomian Wu:** investigation, data curation, visualization, validation, writing – original draft, formal analysis, software. **Lujun Huang:** writing – review and editing, funding acquisition, conceptualization. **Yeshun Guo:** methodology, data curation, visualization, validation, writing – original draft, investigation, conceptualization, software, project administration.

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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.

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