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# Optical material anisotropy in high-index transition metal dichalcogenide Mie nanoresonators

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**Resonant optical antennas provide unprecedented opportunities to control light on length scales far below the diffraction limit. Recent studies have demonstrated that nanostructures made of multilayer transition metal dichalcogenides (TMDCs) can exhibit well-defined and intense Mie resonances in the visible to the near-infrared spectral range. These resonances are realizable because the TMDC materials exhibit very high in-plane refractive indices, in fact higher than what is found in typical high-index dielectric materials like Si or GaAs. However, their out-of-plane refractive indices are comparatively low. Here we experimentally and theoretically investigate how this unusually large material anisotropy influences the optical response of individual TMDC nanoresonators made of MoS<sub>2</sub>. We find that anisotropy strongly affects the far-field optical response of the resonators, as well as complex interference effects, such as anapole and resonant Kerker conditions. Moreover, we show that it is possible to utilize the material anisotropy to probe the vectorial nature of the nanoresonator internal near fields. Specifically, we show that Raman spectra originating from individual MoS<sub>2</sub> nanoresonators exhibit mode-specific anisotropic enhancement factors that vary with the nanoresonator size and correlate with specific modes supported at resonance. Our study indicates that exploring material anisotropy in novel high-index dielectrics may lead to new nanophotonic effects and applications.** © 2020 Optical Society of America under the terms of the [OSA Open Access Publishing Agreement](#)

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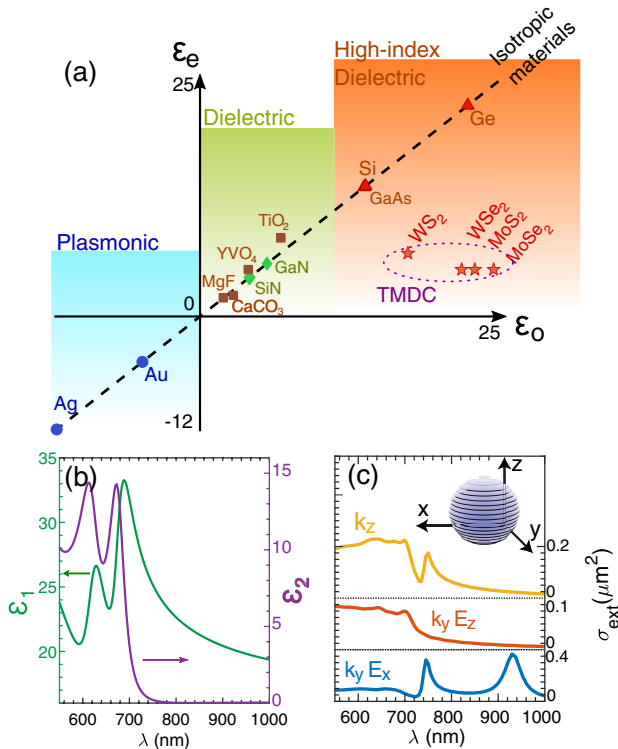
## 1. INTRODUCTION

The ability to fabricate nanostructures of a wide variety of shapes, sizes, and compositions has driven the nanophotonics revolution of the last decades. Nanostructuring enables almost arbitrary control of the optical near fields, which can be explored in the context of enhanced light–matter interaction and unconventional light management. While nanophotonics was for a long time completely dominated by metallic (plasmonic) nanostructures, much recent work has been devoted to high-index dielectrics since the geometrical Mie resonances supported by high-index dielectric nanoantennas exhibit significantly reduced light absorption [1]. High-index nanophotonics is thus a rapidly growing field with potential for applications such as functional metasurfaces [2,3], holograms [4], nonlinear light generation [5,6], light harvesting [7,8], and sensing [9–11]. To date, most dielectric nanoresonators have been fabricated from materials such as Si and GaAs with  $n \approx 3.5$  in the lossless region [12].

Essentially, all bulk materials considered so far in nanophotonics are either isotropic or only weakly birefringent in the visible to near-infrared wavelength range, with ordinary and extraordinary refractive indices differing by not more than  $\sim 10\%$  [Fig. 1(a)]. Researchers working in this wavelength region therefore seldom

had reasons to consider material anisotropy as a useful degree of freedom when designing nanophotonics elements and optical antennas. On the other hand, anisotropy achieved by engineering the effective refractive index through nanostructuring is a well-known inherent property of artificial metamaterials. In this case, the index contrast between different directions makes it possible to create extremely powerful structures that can, for example, tailor the optical wavefront [13], be integrated for optical on-chip communication [14–16], or control polarization beyond what is possible using standard optical elements [17,18]. Optical anisotropy is also prominent in the phonon frequency region of naturally occurring hyperbolic materials such as hexagonal boron nitride, where nanostructuring can be used to generate subwavelength anisotropic localized phonon polariton resonances for infrared wavelengths [19,20].

In contrast to standard materials, group VI semiconductor transition metal dichalcogenides (TMDCs) are layered materials and present an anisotropy ( $\Delta = n_e - n_o$ )  $> 70\%$  and reaching values exceeding 200% at specific wavelengths, with refractive index higher than Si in the plane of the layers and much lower in the out-of-plane direction [see Fig. 1(a)]. Dichalcogenide materials have been the subject of intense research in recent years due to their unique optical and electronic properties [21]. Most recent research



**Fig. 1.** (a) Real ordinary ( $\epsilon_0$ ) and extraordinary ( $\epsilon_e$ ) permittivity of various materials typically used in nanophotonics. Isotropic materials include metals (blue dots,  $\lambda = 533$  nm), dielectric materials (green diamonds,  $\lambda = 800$  nm), and high-index dielectrics (red triangles,  $\lambda = 800$  nm). Birefringent materials (brown squares,  $\lambda = 633$  nm) show differences in refractive index within 10% and only moderately high permittivity. Layered TMDC materials show record-high in-plane permittivity and birefringence larger than 70%. (b) Real (solid line) and imaginary (dashed line) components of the in-plane permittivity of 2H-MoS<sub>2</sub>. (c) Mie resonances of a MoS<sub>2</sub> nanosphere of radius 100 nm in air. Because of material anisotropy, the scattering spectra and the excited modes sensitively depend on the illumination direction and polarization.

has focused on single-layer TMDCs, since these materials exhibit a direct electronic band gap, [22] which makes them interesting for applications like light harvesting, [23–26] catalysis [27–29] and spintronics, [30,31] as well as for fundamental investigations of strong light–matter interactions [32,33]. Recently, we demonstrated an additional and potentially far-reaching functionality of TMDCs, namely, that nanoparticles made of multilayer WS<sub>2</sub> support Mie resonances due to the large in-plane refractive index of the material [34].

In this work, we investigate how material anisotropy influences the optical response of Mie nanoresonators made of TMDC materials. We show that material anisotropy can be used to influence the far-field response of the Mie modes supported by a nanoresonator and that it can be used as a platform for studying new nanophotonic phenomena. Specifically, we used the anisotropic Raman scattering from single MoS<sub>2</sub> nanoparticles to retrieve information on the vectorial nature and relative strength of the nanoresonators' internal near fields. This is possible because TMDCs support distinct Raman-active optical phonons polarized both in the plane of the layers and out of plane according to the symmetry of the specific modes. Thus, the in-plane and out-of-plane phonon modes are selectively enhanced depending on the orientation and strength of the induced local fields. Additionally, the material anisotropy

also influences the far-field response of the complex Mie modes supported by a nanoresonator. These results are, to the best of our knowledge, the first demonstration of how material anisotropy can be used as a new degree of freedom within the context of all-dielectric nanophotonics.

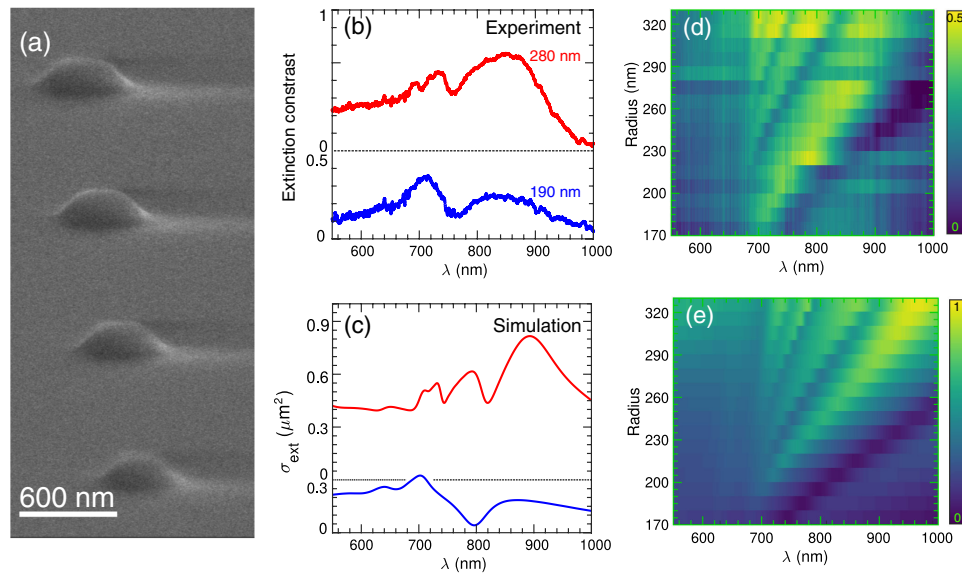
## 2. MATERIAL OPTICAL ANISOTROPY IN MoS<sub>2</sub>

Because of its low loss in the NIR and the very high birefringence [see Fig. 1(a)], we primarily focus on MoS<sub>2</sub> to demonstrate the effects of material anisotropy. The complex in-plane permittivity  $\epsilon_{xy} = \epsilon_{1xy} + i\epsilon_{2xy}$  of bulk MoS<sub>2</sub> is presented in Fig. 1(b) [35]. The material exhibits a very high in-plane refractive index ( $n_{xy} = \text{Re}(\sqrt{\epsilon_{xy}}) > 4$ ) and low absorption ( $\epsilon_{2xy} \approx 0.01$ ) in the NIR region and strong excitonic absorption bands at shorter wavelengths. Despite the interest in TMDC materials and extensive studies during the 1970s, [36] the out-of-plane refractive index is a much less explored quantity [37,38]. In the following, we use the in-plane value from Fig. 1(b) and  $\epsilon_z = 4.9$  reported for 2H-MoS<sub>2</sub> in Ref. [34], while noting that there is considerable uncertainty regarding the out-of-plane value [39].

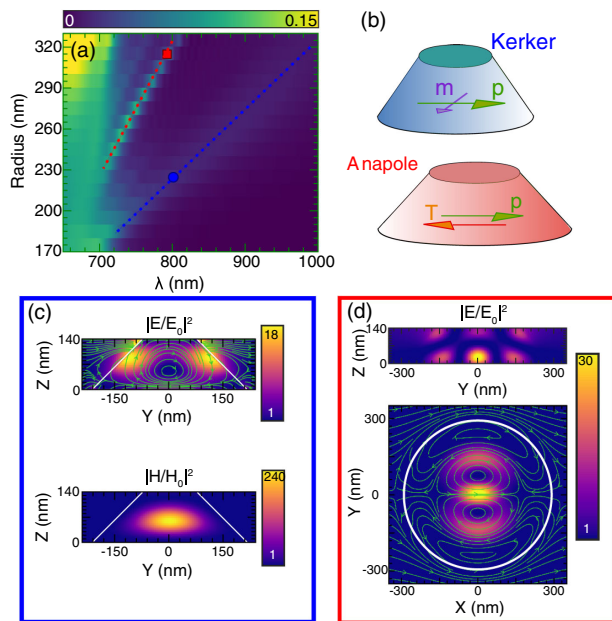
We first investigated how the highly anisotropic permittivity influences the optical response of a spherical nanoresonator, where shape anisotropy (shape birefringence) cannot occur [Fig. 1(c)]. We simulated scattering cross-section spectra for a 100 nm radius nanosphere in air for the three principal incident wavevector-polarization configurations. All spectra exhibit pronounced resonance peaks, but the peak positions, widths, and intensities vary greatly depending on how the nanosphere is excited, despite the spherical geometry. To reveal the nature of the spectral features, we tracked the resonance positions as a function of sphere radius (Fig. S1, Supplement 1). The results show that the resonances disperse linearly with size above  $\sim 700$  nm, where material dispersion  $\epsilon(\lambda)$  and absorption is relatively weak. This is the expected behavior for geometrical Mie resonances in high-index nanostructures. We also performed multipolar decomposition of the induced polarization inside the nanosphere as a function of illumination conditions (see Fig. S2 and Methods, Supplement 1 for details). This analysis indicates that in-plane polarized incidence excites electric-dipole-like and magnetic-dipole-like modes of the sphere, whereas out-of-plane polarized light only excites a magnetic resonance because of the longer effective wavelength perpendicular to the MoS<sub>2</sub> planes.

## 3. SAMPLE FABRICATION AND OPTICAL CHARACTERIZATION

The theoretical results above clearly demonstrate that material anisotropy can profoundly influence the optical properties of a nanoresonator. Unfortunately, spherical TMDC nanoantennas are currently impossible to realize in practice. In the following we therefore focus on in-plane circularly symmetric nanoparticles. The deviation from spherical symmetry implies that any anisotropic response of the structures will in principle be due to a combination of material and shape anisotropy. We start by fabricating particles of varying diameter from transferred MoS<sub>2</sub> flakes by a combination of electron-beam lithography and dry etching (see Supplement 1 for details). In Fig. S3 of Supplement 1, we show that it is possible to apply the same method to fabricate TMDC nanoparticles of MoS<sub>2</sub>, MoSe<sub>2</sub>, WS<sub>2</sub>, and WSe<sub>2</sub>. The resulting



**Fig. 2.** MoS<sub>2</sub> nanostructures support Mie resonances in the visible and near-infrared wavelength range due a high in-plane refractive index. (a) SEM images of the fabricated MoS<sub>2</sub> nanoresonators. (b) Measured extinction contrast spectra and (c) simulated extinction cross-section spectra of MoS<sub>2</sub> nanocones, showing the presence of Mie resonances. (d) Experimental and (e) simulated extinction cross section in  $\mu\text{m}^2$  of MoS<sub>2</sub> nanocones versus base radius and wavelength demonstrating a linear mode dispersion in the near-infrared.



**Fig. 3.** MoS<sub>2</sub> nanoresonators support multiple resonant modes with complex near-field profiles. (a) Simulated absorption cross-section spectra in  $\mu\text{m}^2$  of MoS<sub>2</sub> nanocones as a function of base radius. (b) Sketch of the modes responsible for anapole and Kerker interference effects. (c) Electric and magnetic field distributions for a 230 nm radius nanocone at 785 nm indicating the presence of both electric and magnetic modes. (d) Anapole-like out-of-plane and in-plane electric field distributions and field lines for a 300 nm base nanocone at 785 nm.

particles are single crystalline and can be potentially realized on any substrate of choice.

Scanning electron microscope (SEM) images of representative nanostructures are presented in Fig. 2(a). We note that the dry-etch step yields conical nanodisk shapes. This is likely due to

carbon that deposits on the particle sidewalls during etching and then acts as a mask material. Interestingly, we found that while Mo-based materials yielded nanostructures with sloping sidewalls, i.e., “nanocones,” the W-based samples showed an almost vertical profile (Fig. S3, Supplement 1), confirming a chemical origin of this effect.

Next, we studied the optical properties of individual MoS<sub>2</sub> nanocones as a function of their in-plane dimension [Figs. 2(b)–2(e)]. To this end, we measured transmission  $T$  using a  $1\ \mu\text{m}^2$  confocal probe area centered on the particle and normalized the recorded spectrum with the transmission  $T_0$  through an empty glass slide. The measured effective single particle extinction contrast  $E = 1 - T/T_0$  is shown in Fig. 2(f) as a function of the base radius of the nanocones. The extinction contrast exceeds 0.5 for the largest particles at resonance, indicating a large optical cross section. As expected for high-index dielectric nanostructures, the nanocones exhibit prominent resonant features that disperse linearly with the size in the NIR region, where both material dispersion  $\delta n/\delta\lambda$  and absorption are low. Simulated extinction spectra of MoS<sub>2</sub> nanocones with the same size and tapering angle [Figs. 2(c) and 2(e)] are in good overall agreement with the experimental data despite the complex shape of the nanostructures. The excitonic absorption bands at  $\sim 620$  and  $\sim 680$  nm [Fig. 1(b)] induce strong bending of the Mie resonance dispersion with radius in the visible wavelength range. This effect, which also occurs for WS<sub>2</sub> nanodisks, is caused by hybridization between the geometrical modes and the excitons, resulting in an avoided crossing [26]. However, the coupling is less pronounced in the MoS<sub>2</sub> case, possibly due to a smaller exciton oscillator strength and higher dissipation compared to WS<sub>2</sub> [31].

In the NIR, where the material in-plane absorption is low, the TMDC Mie nanoresonators can be used as a low-loss nanophotonics platform similarly to isotropic all-dielectric nanoresonators. In Fig. 3(a), we show simulated absorption cross-section spectra of MoS<sub>2</sub> nanocones. The data reveals two absorption features

that disperse linearly with size as indicated by the dashed lines. Note that, due to the relatively large in-plane sizes of the MoS<sub>2</sub> nanoparticles, elastic scattering dominates over absorption, which contribute less than  $\sim 10\%$  to the overall extinction. Thus, scattering interferences between the resonances, indicated in Fig. 3(b), play a crucial role in determining the shape and amplitudes of the extinction spectra shown in Fig. 2.

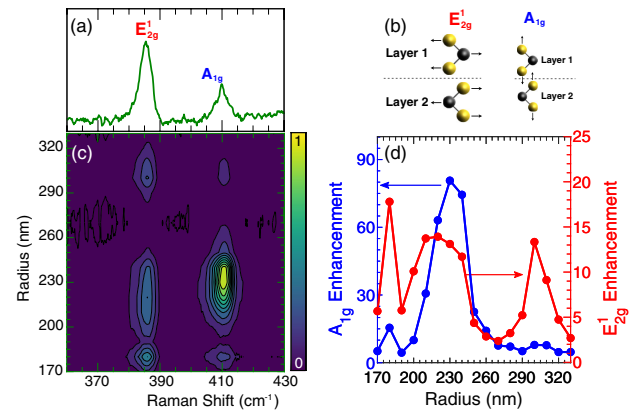
In the following, we analyze the two absorption features at the laser excitation wavelength 785 nm used in the Raman experiments discussed in the next section. The first absorption peak crosses 785 nm at  $\sim 230$  nm radius [blue dot in Fig. 3(a)]. Plots of the corresponding electric and magnetic intensity enhancement factors in a vertical cut in the  $y - z$  plane reveals circulating currents indicative of a magnetic Mie resonance [Fig. 3(c)]. To identify the multipoles contributing to this field pattern, we decompose the induced polarization into Cartesian multipoles (Fig. S4, see Supplement 1). The analysis reveals that the 230 nm radius nanocone exhibits both resonant electric ( $p$ ) and magnetic ( $m$ ) dipole moments with comparable magnitudes and vanishing phase difference. This situation corresponds to the first Kerker condition, characterized by a large forward-to-backward (F/B) scattering ratio [40–42]. In fact, the data indicates that is possible to reach F/B ratios of 50–100 for specific particle radii and wavelengths. Experimental reflection measurements (Fig. S5, Supplement 1) indicate that the MoS<sub>2</sub> nanostructures indeed enter a regime of reduced backscattering, but a detailed experimental quantification of this effect goes beyond the scope of this work.

The second absorption peak crosses 785 nm at a radius of  $\sim 300$  nm [red dot in Fig. 3(a)]. The corresponding near-field distribution [Fig. 3(c)] in the  $x - y$  plane resembles a so-called anapole—an interference effect characterized by enhanced internal fields and suppressed far-field scattering [43]. This phenomenon is due to destructive interference of the far fields generated by electric dipole  $p$  and toroidal electric dipole  $T$  moments in the particle. Indeed, we observe suppression of extinction, which is dominated by scattering, in experiment and simulations around 785 nm, but also an enhancement of internal in-plane near fields.

Similarly to the case of the sphere [Fig. 1(c)], one would like to pinpoint how material anisotropy influences the far-field properties for the fabricated MoS<sub>2</sub> nanoresonators. To this end we performed polarization-dependent dark-field scattering measurements by illuminating the particles with either  $p$ - or  $s$ -polarized light at a high oblique angle (see Fig. S6, Supplement 1). The data show complex polarization-dependent scattering, but a detailed analysis is difficult due to the low symmetry of the nanostructure and the off-axis illumination condition.

#### 4. RAMAN ENHANCEMENT IN MoS<sub>2</sub> NANORESONATORS

We have thus far analyzed the far-field response of Mie resonators made of anisotropic materials. These resonators act as optical cavities that enhance internal electromagnetic fields at specific resonance wavelengths. This allows for enhancement of various optical processes originating from within the particle, such as non-linear frequency conversion [5], fluorescence, [44], and Raman scattering [45]. Here we focus on Raman scattering, since the presence of Raman-active phonon modes with well-defined displacement patterns can be correlated with material anisotropy. A typical Raman spectrum obtained in backscattering geometry from



**Fig. 4.** Raman spectrum of bulk crystalline MoS<sub>2</sub> is enhanced and modified in Mie resonators. (a) Typical unpolarized Raman spectrum of bulk MoS<sub>2</sub> obtained in backscattering geometry for incidence normal to the basal plane and nonresonant excitation at 785 nm. (b) Sketch of the atomic vibration related to the two Raman modes. (c) Single-particle Raman spectra of MoS<sub>2</sub> nanoresonators, demonstrating enhancement of the scattering signal at specific particle sizes. (d) Volume-averaged Raman intensity of the  $A_{1g}$  (blue) and  $E_{2g}^1$  (red) modes measured in single nanocones, relative to the signal obtained from an unpatterned film, as a function of particle bottom radius.

the basal plane of a bulk MoS<sub>2</sub> crystal using nonresonant 785 nm laser excitation is shown in the top panel of Fig. 4(a) (see Methods in Supplement 1). There are two peaks in the frequency range investigated. These peaks, centered at  $385\text{ cm}^{-1}$  and  $410\text{ cm}^{-1}$ , are due to the  $E_{2g}^1$ - and  $A_{1g}$ -symmetry optical phonon modes with vibration displacements within and perpendicular to the MoS<sub>2</sub> planes, respectively [see Fig. 4(b)].

Figure 4(c) shows Raman spectra from single MoS<sub>2</sub> nanoresonators as a function of their base radius. The relative peak intensities, here normalized by the nanoresonator volume, show a nontrivial dependence on the nanodisk size, suggesting that different enhancements originate from the geometrical resonances of the nanoparticles. Remarkably, the relative intensities of the two peaks are inverted at around 230 nm and 300 nm radii. This behavior is also seen in Fig. 4(d), where the volume-averaged Raman intensities of the  $A_{1g}$  and  $E_{2g}^1$  modes instead have been normalized to the intensities obtained from an unpatterned flake of the same thickness as the nanocones and by assuming a diffraction-limited sampling area. The resulting Raman enhancement factor reaches  $\sim 13$  for the  $E_{2g}^1$  and  $\sim 80$  for the  $A_{1g}$  mode, confirming a strong internal field enhancement. Scattering from the  $A_{1g}$  mode is clearly enhanced in resonators with base radius of approximately 230 nm, while the  $E_{2g}^1$  mode exhibits a distinct enhancement maximum at around 300 nm. These nanocone sizes match the maxima observed in the absorption spectra [Fig. 3(a)] at the Raman excitation wavelength 785 nm. As is well known, absorption scales with the electric field intensity inside the nanostructure and the imaginary component of the material's permittivity as a multiplying factor. As the permittivity tensor is isotropic within the basal plane and real in the perpendicular direction, the absorption cross section reads as  $\sigma_{\text{abs}} \propto \epsilon_2(\omega) \int_V (|E_x(\omega_L, r)|^2 + |E_y(\omega_L, r)|^2) d^3r$ , where  $\epsilon_2$  is the imaginary part of the in-plane permittivity from Fig. 1(b). The strong modulation of the Raman intensity with the nanostructure size thus reflects the geometric modes of the nanostructure

going into resonance with the excitation and emission frequencies. However, because of the material anisotropy, mode-specific near-field profiles are probed in the Raman scattering process in different ways compared to standard high-index dielectric materials. We note that the additional peak appearing for  $E_{2g}^1$  at 180 nm does not correspond to a specific mode [cf. the mode dispersion in Fig. 3(a)], and it is likely caused by overestimated noise in the volume-normalized enhancement at small radii.

To analyze and interpret the obtained results, we need to establish a relationship between the electromagnetic field induced in a single-crystalline resonator and the Raman signal recorded in a particular scattering geometry. For a bulk material, the intensity of Raman scattering from a particular phonon mode  $\sigma$  is proportional to

$$I_{\text{Raman}} \propto \left| \sum_{\sigma} \mathbf{e}_{\text{inc}} \overleftrightarrow{\mathbf{R}}_{\sigma} \mathbf{e}_{\text{scat}} \right|^2, \quad (1)$$

where  $\mathbf{e}_{\text{inc}}$  and  $\mathbf{e}_{\text{scat}}$  are unit vectors defining the polarization of the incident and scattered photons, respectively, and  $\overleftrightarrow{\mathbf{R}}_{\sigma}$  is the Raman polarizability tensor of phonon mode  $\sigma$ . The Raman polarizability is determined by the symmetry of the crystal structure, the vibrational displacement pattern of the phonon mode and the electron-phonon coupling strength [46]. MoS<sub>2</sub> has  $D_{6h}$  point symmetry, and the Raman tensors of the  $E_{2g}^1$  and  $A_{1g}$  modes have the structure [47]

$$\overleftrightarrow{\mathbf{R}}_{E_{2g}^1} = \begin{bmatrix} d & 0 & 0 \\ 0 & -d & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \begin{bmatrix} 0 & d & 0 \\ d & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix},$$

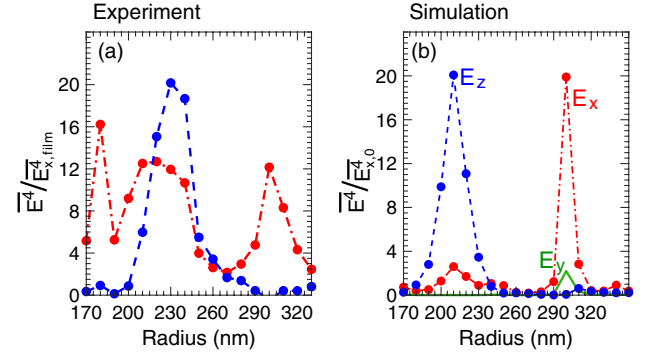
$$\overleftrightarrow{\mathbf{R}}_{A_{1g}} = \begin{bmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{bmatrix}. \quad (2)$$

The relative strengths of the Raman tensor elements  $a$ ,  $b$ , and  $d$  for 785 nm excitation were determined by recording Raman spectra from thick MoS<sub>2</sub> flakes in different scattering geometries A(B,C)D, where A/D indicate the direction of the incident and scattered fields and B/C their polarization. From this analysis (see Supplement 1 and Fig. S7), the relative matrix elements were estimated to  $a \approx 0.46$ ,  $b \approx 1.0$ , and  $d \approx 0.72$ .

In order to describe Raman scattering from MoS<sub>2</sub> nanoresonators, we now employ the classical electromagnetic description of Raman scattering [48]. In this framework, the local excitation field at the laser frequency  $\omega_L$  inside the particle,  $\mathbf{E}_{\text{exc}}(\mathbf{r}, \omega_L)$  determines the local induced Raman polarization  $\mathbf{P}_S$  oscillating at the Stokes-shifted frequencies. The Stokes Raman polarization is then given by

$$\mathbf{P}_S(\mathbf{r}, \omega_L, \omega_S) = \overleftrightarrow{\mathbf{R}}_{\sigma}(\mathbf{r}, \omega_L, \omega_S) \mathbf{E}_{\text{exc}}(\mathbf{r}, \omega_L), \quad (3)$$

where  $\omega_S = \omega_L - \Omega$  and  $\Omega$  is the phonon frequency. The total intensity of the radiated Raman signal can be calculated by integrating the Poynting vector generated by the Raman polarization  $\mathbf{P}_S$  over a surface enclosing the entire particle. Generally, this requires knowledge of the Green tensor of the system and consecutive calculation of the radiated electric and magnetic fields. However, we are only interested in the emission radiated in the backscattering direction. Thus, we utilize the electromagnetic reciprocity theorem and instead calculate enhancement factors for



**Fig. 5.** Extracting the vectorial near-field enhancement from Raman measurements. (a) Variation in volume-averaged  $|\mathbf{E}_x|^4$  and  $|\mathbf{E}_z|^4$  near-field amplitudes with the nanocone bottom radius. The data is extracted from Raman data by inverting Eq. (4) and normalized by the average in-plane field extracted from the Raman signal of a thin film. (b) The corresponding simulated electric field enhancement factors  $|\mathbf{E}_v/\mathbf{E}_0|^4$  ( $v = x, y, z$ ) as functions of the nanocone bottom radius induced at the wavelength of 785 nm.

the  $E_v$  ( $v = x, y, z$ ) field component generated by a normally incident  $x$ -polarized plane wave at the Stokes-shifted wavelength. This quantity is a measure of the Raman enhancement in backscattering of a  $v$ -polarized point electric dipole oscillating at the Raman emission frequency. We also assume a small Stokes shift compared to the excitation frequency for both the  $E_{2g}^1$  and  $A_{1g}$  mode (i.e.,  $\omega_L \sim \omega_S$ ). This treatment is equivalent to the  $|E|^4$  approximation often used in surface-enhanced Raman scattering [49]. Furthermore, we will assume that the Raman polarization is uncorrelated in space; that is, each Raman point dipole inside the particle contributes incoherently to the total Raman intensity. Finally, we will assume that the induced  $y$  component of the electric field inside the particle is negligible for  $x$ -polarized excitation, which is justified by finite-difference time domain (FDTD) simulations in the following. Under these assumptions, we arrive at the following simple expressions for the Raman intensities emitted by the two independent vibrational modes of the crystal (see Supplement 1):

$$I_{E_{2g}^1} \propto |d|^2 \int_V |E_x(r, \omega_L)|^4 d^3r,$$

$$I_{A_{1g}} \propto |a|^2 \int_V |E_x(r, \omega_L)|^4 d^3r + |b|^2 \int_V |E_z(r, \omega_L)|^4 d^3r. \quad (4)$$

The integration is performed over the interior volume of the nanostructure, and the electric fields represent the induced local fields inside the nanostructure.

Equation (4) implies a direct relation between the Raman intensity and the average vectorial electric field intensity inside the nanoparticle. The relative intensity of the two Raman modes can thus be exploited as an experimental probe of the vectorial components of the near fields induced in the nanoresonator. Figure 5(a) shows this effect by inverting Eq. (4) for extracting the volume-averaged magnitudes of the induced near fields  $|\mathbf{E}_v|^4$  in MoS<sub>2</sub> nanoresonators via analysis of the anisotropic Raman modes. The values were normalized by the averaged near field  $|\mathbf{E}_x^{\text{film}}|^4$  extracted from the Raman spectrum of a thin MoS<sub>2</sub> flake of the same thickness, where we assumed a signal originated by

a diffraction-limited illumination area. To verify the obtained electric field magnitudes, we compared them to the electric field enhancement factors obtained from FDTD simulations of the corresponding structures illuminated at the wavelength of 785 nm [Fig. 5(b)]. Despite the approximations made in our derivation, the values extracted from the Raman measurements are in good overall agreement with the simulations. The resonances in the simulated data are shifted to slightly smaller radii compared to the experimental data, but this does not come as a surprise if one considers the differences between the experiment and FDTD simulations already observed in the elastic scattering spectra in Fig. 2. These differences highlight the needs for experimental measurements to estimate the internal fields, particularly for those structures with complicated spectral features.

## 5. CONCLUSION

In summary, we have investigated the effects of material anisotropy in cone-shaped single-crystal MoS<sub>2</sub> transition metal dichalcogenide nanoresonators using extinction measurements, inelastic scattering from Raman active phonons, and extensive electrodynamic simulations. The nanoresonators support geometrical Mie resonances in the visible to near-infrared spectral range due to their large in-plane refractive index and in spite of a much lower out-of-plane index. We showed that is possible to utilize the material anisotropy not only to modify the resonance of the nanostructures but also to probe the polarization and relative magnitudes of the enhanced near fields inside a nanoresonator using Raman scattering analysis.

Anisotropic optical responses in nanophotonics are typically associated with shape anisotropy induced through nanostructuring. TMDC materials offer an additional degree of freedom in the design of nanophotonic structures as the intrinsic material anisotropy also strongly influences far-field spectra, field enhancement, and near-field polarization. Nanostructuring and material anisotropy thus constitute an extremely powerful and flexible means for tailoring the optical response of nanophotonic systems and for exploring new avenues for research and applications.

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**Disclosures.** The authors declare no conflicts of interest.

See Supplement 1 for supporting content.

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