Enabling predictive capabilities for the polaritonic response of the biaxial van der Waals crystal α-MoO_3

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Highly anisotropic crystals have recently attracted considerable attention because of their ability to support polaritons with a variety of unique properties, such as hyperbolic dispersion, negative phase velocity, or extreme confinement. Particularly, the biaxial van der Waals semiconductor α-phase molybdenum trioxide (α-MoO_3) has recently received substantial attention due to its ability to support in-plane hyperbolic phonon polaritons (PhPs) —infrared (IR) light coupled to lattice vibrations in polar materials—, offering an unprecedented platform for controlling the flow of energy at the nanoscale [1,2]. Yet, to accurately predict the IR response of α-MoO_3 and thus to enable predictive capabilities for the extraordinary optical response of this material, it is imperative to develop both an accurate IR dielectric function model for α-MoO_3 and a theoretical study on electromagnetic modes in biaxial crystal slabs.

Here [3], we derive the dispersion relation of electromagnetic modes in biaxial slabs surrounded by semi-infinite isotropic dielectric half-spaces with arbitrary dielectric permittivities. Apart from a general dispersion relation, we provide very simple analytical expressions in typical experiments in nano-optics: the limits of short polaritonic wavelength and/or very thin slabs, allowing for an in-depth analysis of anisotropic polaritons in novel biaxial van der Waals materials.

Moreover, we report the accurate IR dielectric function of α-MoO_3 [4] by modelling polarized IR reflectance spectra acquired on a single thick flake of this material. Unique to our work, the far-field model is refined by contrasting the experimental dispersion and damping of PhPs extracted by near-field polariton interferometry using scattering-type scanning nearfield optical microscopy (s-SNOM) on thin flakes of α-MoO_3, with analytical [3] and transfer-matrix calculations, as well as full-wave simulations (Fig. 1). Through these correlative efforts, exceptional quantitative agreement is attained to both far- and near-field properties for multiple flakes, thus providing strong verification of the accuracy of our model, while offering a novel approach to extracting dielectric functions of nanomaterials, usually too small or inhomogeneous for establishing accurate models only from standard far-field methods. In addition, by employing density functional theory (DFT), we provide insights into the various vibrational states dictating our dielectric function model and the intriguing optical properties of α-MoO_3.

Our findings will enable the interpretation of experimental far- and near-field data, as well as an efficient design of nanostructures supporting such highly anisotropic polaritons supported by α-MoO_3.

REFERENCES

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