Infrared permittivity of the biaxial van der Waals semiconductor α-MoO_3 from near- and far-field correlative studies

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The biaxial van der Waals semiconductor α-phase molybdenum trioxide (α-MoO_3) has recently received significant attention due to its ability to support highly anisotropic 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, beyond what has been achieved with other materials supporting PhPs. However, to fully exploit the extraordinary IR response of this material, an accurate dielectric function is required. Here [1], we report the accurate IR dielectric function of α-MoO_3 by modelling far-field 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, revealed by polariton interferometry using scattering-type scanning near-field optical microscopy (s-SNOM) on thin flakes of α-MoO_3, with analytical and transfer-matrix calculations, as well as full-wave simulations. Through these correlative efforts, exceptional quantitative agreement is attained to both far- and near-field properties for multiple flakes, thereby strongly verifying the accuracy and robustness 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.

References

[1] G. Álvarez Pérez *et al.*, Advanced Materials (accepted).

Figures

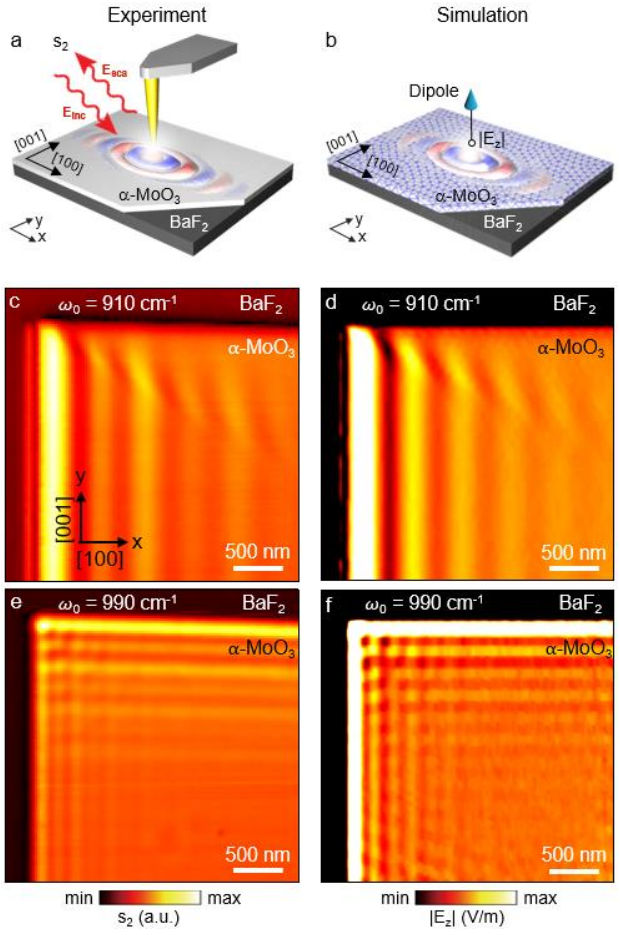

Figure 1: Near-field simulated and experimental images of in-plane anisotropic PhPs on an α-MoO_3 thin flake.