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

Gonzalo Álvarez-Pérez[1,2]
Thomas G. Folland,[3] Ion Errea,[4,5] Javier Taboada-Gutiérrez,[1,2] Jiahua Duan,[1,2] Javier Martín-Sánchez,[1,2] Ana I. F. Tresguerres-Mata,[1] Joseph R. Matson,[3] Andrei Bylinkin,[6,7] Mingze He,[3] Weiliang Ma,[8] Qiaoliang Bao,[8,9] José Ignacio Martín,[1,2] Joshua D. Caldwell,[3] Alexey Y. Nikitin[5,10] and Pablo Alonso-González.[1,2]
[1]*Department of Physics, University of Oviedo, Oviedo 33006, Spain.*
[2]*Center of Research on Nanomaterials and Nanotechnology, CINN (CSIC--Universidad de Oviedo), El Entrego 33940, Spain.*
[3]*Department of Mechanical Engineering, Vanderbilt University, Nashville 37235, TN, USA.*
[4]*Fisika Aplikatua 1 Saila, University of the Basque Country (UPV/EHU), Donostia/San Sebastián 20018, Spain. / Centro de Física de Materiales (CSIC-UPV/EHU), Donostia/San Sebastián 20018, Spain.*
[5]*Donostia International Physics Center (DIPC), Donostia/San Sebastián 20018, Spain.*
[6]*CIC nanoGUNE, Donostia/San Sebastián 20018, Spain.*
[7]*Moscow Institute of Physics and Technology, Dolgoprudny 141700, Russia.*
[8]*Department of Materials Science and Engineering and ARC Centre of Excellence in Future Low-Energy Electronics Technologies (FLEET), Monash University, Clayton, Victoria 3800, Australia*
[9]*Institute of Functional Nano and Soft Materials (FUNSOM), Jiangsu Key Laboratory for Carbon-based Functional Materials and Devices Soochow University, Suzhou, Jiangsu 215123, China*
[10]*IKERBASQUE, Basque Foundation for Science, Bilbao 48013, Spain.*
gonzaloalvarez@uniovi.es

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 [1]. However, to fully exploit the extraordinary IR response of this material, an accurate dielectric function is required. Here [2], 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 [3] 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, 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.

References

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