POLARITONS

Graphene unlocks dispersion of topological polaritons

Heterostructure of graphene and biaxial van der Waals crystal supports a species of plasmon-phonon-polaritons whose isofrequency dispersion contour can be manipulated while experiencing a topological transition.

Sergey G. Menabde and Min Seok Jang

The past decade has witnessed explosive growth in research on van der Waals crystals supporting propagating polaritons — quasiparticles of light coupled to the collective oscillations of material charges[1]. Because of the inherently 2D nature of van der Waals crystals, their polaritons naturally possess an extremely compressed electromagnetic field relative to the light wavelength, providing strong light-matter interaction and offering ways to manipulate light at the nanoscale. Two types of mid-infrared polaritons are particularly important owing to their unusual properties: collective electron oscillations known as plasmons, such as those occurring in highly doped graphene; and phonon-polaritons associated with ionic motion in van der Waals crystals such as hexagonal boron nitride[2] (hBN), orthorhombic vanadium pentoxide[3] (α-V_2O_5) or orthorhombic molybdenum trioxide[4] (α-MoO_3). The dispersion of graphene plasmons can be tuned by doping the material (for example, by electrostatic gating or chemical adsorption), while phonon-polaritons possess intrinsic anisotropy. Now, Hu et al. have demonstrated[5] that the van der Waals heterostructure formed by graphene deposited on a film of α-MoO_3 supports hybrid plasmon-phonon-polaritons (Fig. 1a) that combine unique properties of both modes: the tunability of graphene plasmons and the anisotropic dispersion of phonon-polaritons in α-MoO_3. As a consequence, by tuning the doping level of graphene, the topology of the isofrequency dispersion contours of the hybrid modes can be actively changed[6,7] from a hyperbolic (open) to an elliptical (closed) shape (Fig. 1b). In the process, the researchers observe intermediate flat isofrequency contours, which they use to achieve polariton canalization along desired directions.

Hu et al. take advantage of widely tunable optical conductivity of graphene that can be controlled via doping. As a consequence, the dispersion of graphene plasmons can be manipulated via doping as well. However, the isotropy of the graphene conductivity prohibits topological transitions in the

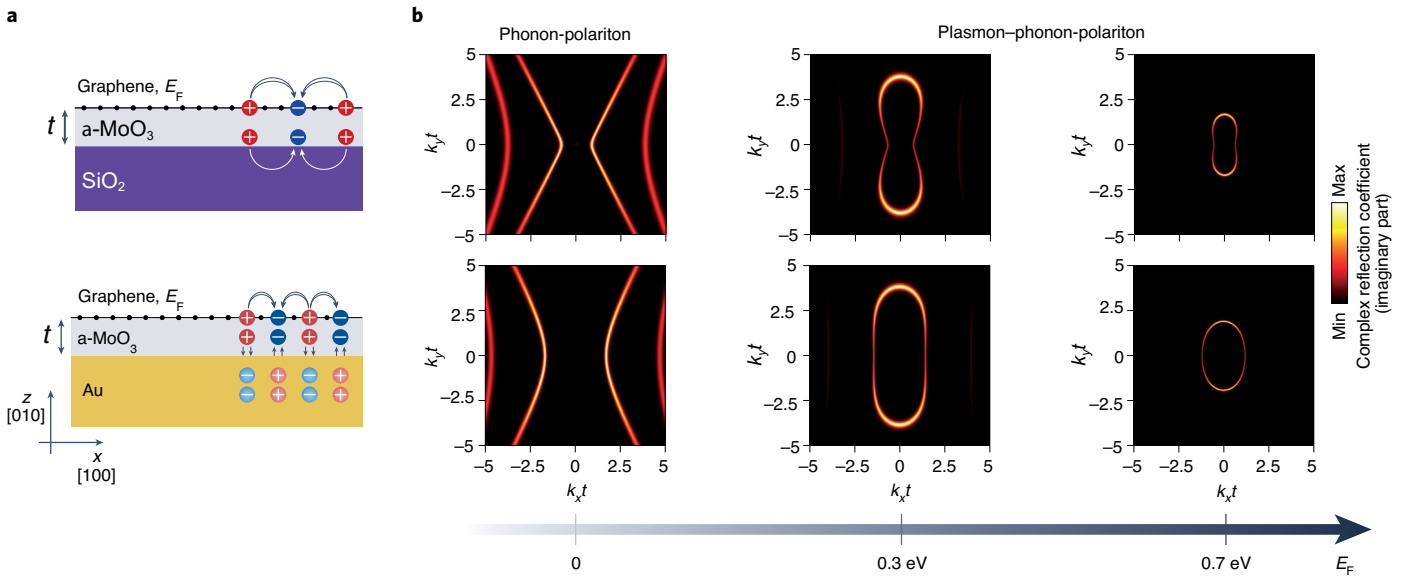

Fig. 1 | Schematics and isofrequency contours of the hybrid plasmon-phonon-polaritons. a, Top: schematic representation of the plasmon-phonon-polariton formed by hybridization between the isotropic graphene plasmon and anisotropic phonon-polariton in α-MoO_3 on a dielectric substrate. Bottom: image charges in the metal substrate result in a more compressed image polariton mode. **b**, Varying Fermi level in graphene leads to the transitioning of the hybrid polariton's isofrequency dispersion contour from the open shape at zero doping (that of the hyperbolic phonon-polariton in α-MoO_3) to a closed shape at higher Fermi levels. The higher-momentum image mode (bottom row) exhibits flattened isofrequency contour at Fermi level of ~0.3 eV, providing a diffractionless canalization of the in-plane mode energy. Isofrequency contours are revealed by the imaginary part of the complex reflection coefficient of the heterostructure calculated at 910 cm^{-1} assuming negligible ohmic loss in graphene. E_F, Fermi level in graphene; k, polariton momentum; t, thickness.

plasmon momentum space. In contrast to graphene, multilayer polaritonic van der Waals materials are typically anisotropic and support phonon-polaritons with hyperbolic shapes of their associated isofrequency contours — the hyperbolic phonon-polaritons. In particular, the mid-infrared polaritons supported by α-MoO_3 within its second Reststrahlen band (from 816 cm^{-1} to 976 cm^{-1}) possess a strong in-plane anisotropy, stemming from the real part of the permittivity being positive along the [001] and negative along the [100] crystallographic directions. Although the properties of α-MoO_3 cannot be externally tuned, mode hybridization with graphene plasmons unlocks the dispersion of hyperbolic plasmon-phonon-polaritons, allowing the manipulation of the isofrequency contour in momentum space.

Importantly, a high Fermi level in graphene is not required to switch the dispersion topology of the hybrid polaritons. By resorting to chemical doping, Hu et al. demonstrate that a topological transition from the purely hyperbolic mode at zero doping occurs already at a Fermi level of 0.2–0.3 eV (Fig. 1b). Further increasing the doping up to 0.7 eV leads to an elliptic isofrequency contour and enables polariton propagation along the otherwise forbidden [001] direction.

In addition to the graphene Fermi level, the substrate material supporting the heterostructure may play an important role in tailoring the dispersion. Hu et al. show how a gold screen under the α-MoO_3 layer (bottom panel in Fig. 1a) increases the polariton momentum and flattens the isofrequency curve (bottom panels in Fig. 1b). This is due to the manifestation of a more compressed image mode[8], stemming from polariton coupling with its mirror image in a highly conductive metallic substrate (such as gold at mid-infrared frequencies). The gold substrate thus leads to a rather flat isofrequency contour at intermediate stages along the transition between hyperbolic and elliptic morphologies. The researchers make use of this to achieve a diffractionless beam-like canalization of the hybrid mode when its propagation is allowed only in a certain direction.

Another intriguing phenomenon spotlighted by Hu et al. is the different curvature of the dispersion contour of the hybrid mode depending on the substrate. At high doping levels of graphene, the isofrequency contour in [100] direction is convex on a gold substrate and concave on SiO_2 (rightmost panels in Fig. 1b). Therefore, by altering the permittivity of the environment, they can further manipulate the propagation of hybrid polaritons. Using the in-plane heterostructure of gold–SiO_2–gold as a substrate, the researchers demonstrate the effect of partial focusing of the propagating mode using a 1.5-μm-wide SiO_2 patch, achieving a 4.5-fold intensity enhancement at the focal spot compared with the same area without SiO_2.

Recently, the manipulation of polariton dispersion topology has been demonstrated in a system of two α-MoO_3 layers twisted relative to each other[9]. This approach is based on the hybridization of phonon-polaritons with different dispersion along the selected propagation direction according to the crystallographic orientation of each crystal. However, physical stacking of thin α-MoO_3 crystals is not amenable to dynamic tuning of the hybridized polaritons. In contrast, graphene doping can be modified through electrical gating, with the gold substrate playing the role of a back gate. Such versatile control over the highly compressed hyperbolic polaritons could lead to exciting on-chip applications such as nanoimaging, optical sensing, and manipulation of energy transfer at the nanoscale. ❐

Sergey G. Menabde and Min Seok Jang ✉

School of Electrical Engineering, Korea Advanced Institute of Science and Technology, Daejeon, Korea.
✉*e-mail: jang.minseok@kaist.ac.kr*

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Competing interests

The authors declare no competing interests.