Splitting of Phonon Polariton Dispersion in van der Waals Crystal Heterostructure

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Phonon-polaritons (PhPs), which emerge from the coupling between the atomic lattice vibrations (phonons) and photons, have been observed in many two-dimensional van der Waals (vdW) materials such as alpha-phase molybdenum trioxide (α-MoO_3) and hexagonal boron nitride (hBN).[1] Notably, α-MoO_3 is a biaxial crystal, so that PhPs exhibit in-plane anisotropy. In addition to α-MoO_3, various other two-dimensional van der Waals materials with in-plane anisotropy, such as β-Ga_2O_3, have been discovered.[2] Heterostructures made by stacking and twisting of these materials have demonstrated exotic properties, prompting active research efforts aimed at engineering the polariton dispersion. A representative example is the twisted bilayer α-MoO_3, where twisting the layers relative to each other induces topological transitions and flat bands in the PhP dispersion contours and enables low-loss canalized propagation of PhPs.[3]

At the same time, when hBN is placed on a gold crystal, it can form high-momentum image modes that have higher momentum than those in the same but free-standing hBN layer.[4] Furthermore, by introducing an air gap between the gold and hBN, the momenta of the first-order symmetric and antisymmetric modes can be tuned.[5]

However, no previous study has reported a PhP mode splitting into the bonding-antibonding modes with similar momentum by introducing a gap between the two layers of the same material. While some studies show that the varying gap between gold and hBN can simultaneously increase or decrease the momentum of symmetric and antisymmetric modes, this is limited by the antisymmetric nature of image charges, which inherently restricts the available modes according to their field symmetry.[5] In contrast, using two material slabs instead of image charges enables formation of additional modes. As the gap between the slabs decreases, coupling between the eigenmodes of the two slabs arises, causing the dispersion splitting into a symmetric mode with reduced momentum and an antisymmetric mode with increased momentum, where symmetry refers to the out-of-plane component of the electric field. Importantly, such splitting results in modes with close momenta, in contrast to image modes, akin to the bonding-antibonding energy splitting.

Here, we report for the first time the splitting of PhP modes in a α-MoO_3/hBN/α-MoO_3 heterostructure, modulated by a varying thickness of the hBN spacer. We use near-field imaging to observe the splitting of PhP modes with anomalous dispersion in 975-990 cm^{-1} spectral window and compare the experimental results with theoretical predictions based on transfer matrix method.[6]

References

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