Cross-material exciton-phonon coupling in van der Waals heterostructures

COLIN CHOW, AARON JONES, XIAODONG XU, University of Washington, Seattle, USA., HONGYI YU, WANG YAO, University of Hong Kong, Hong Kong, China, JIAQIANG YAN, DAVID MANDRUS, Oak Ridge National Laboratory, Oak Ridge, USA., TAKASHI TANIGUCHI, KENJI WATANABE, National Institute for Materials Science, Tsukuba, Japan. — Exciton-phonon interaction plays a major role in the optical response of van der Waals (vdW) heterostructures, which, if appropriately controlled, is a valuable tool in engineering optoelectronic devices based on 2D semiconductors. Here, we report a novel exciton-phonon interaction at the interface between different vdW atomic layers, as well as with the substrates. By interfacing a monolayer WSe\textsubscript{2} with atomically-thin hexagonal boron nitride (hBN), we observe the activation of Raman silent hBN $A_{2u}$ mode stemming from the coupling between WSe\textsubscript{2} exciton and hBN phonons. A comparison between hBN-overlaid and hBN-sandwiched WSe\textsubscript{2} samples shows that the coupling between hBN $A_{2u}$ phonon and WSe\textsubscript{2} exciton is suppressed in the latter, while that between hBN $A_{2u}$, WSe\textsubscript{2} $A_{1}'$ phonons and WSe\textsubscript{2} exciton remains strong. This demonstrates that the interfacial exciton-phonon coupling can be manipulated by symmetry reconstruction. Moreover, we observe enhanced Raman signals originating from surface mode in SiO\textsubscript{2}, as well as $E_g$ mode in sapphire substrates. This highlights the ubiquity of interfacial exciton-phonon coupling in vdW heterostructures. Due to their nanoscaled geometries, they are highly susceptible to vibrational surroundings, especially from supporting substrates, an aspect commonly overlooked.

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