Pressurized MoS_2 and monolayer WTe_2 as ideal excitonic insulators

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We claim that MoS_2 under pressure [1] and monolayer WTe_2 [2,3] are “excitonic insulators” (EIs). The long-sought EI is a permanent Bose-Einstein condensate of excitons, electron–hole pairs bound by Coulomb interaction, which form in the absence of optical excitation. A surge of experimental claims has recently addressed layered materials, because of reduced Coulomb screening. However, the transition to the putative EI is ubiquitously accompanied by the softening of a phonon inducing a structural change; therefore it remains unclear whether the observed phase is genuinely excitonic or instead stabilized by electron–phonon interaction.

Our calculations from first principles [1] show that MoS_2 for a range of applied pressure is unstable against the spontaneous generation of excitons but stable against lattice distortion. We predict that the EI is an antiferroelectric, electronic density wave. At the onset of the EI phase, those optical phonons that share the exciton momentum provide a unique Raman fingerprint for the EI formation. We identify such fingerprint in a Raman feature that was previously observed experimentally, thus providing direct spectroscopic confirmation of an EI phase in bulk MoS_2 above 30 GPa.

Furthermore, we present theoretical [2,3] and experimental [3] evidence that the two-dimensional bulk of monolayer WTe_2 contains excitons that spontaneously form in thermal equilibrium. On cooling from room temperature to 100 K, the conductivity develops a V-shaped dependence on electrostatic doping, while the chemical potential develops a step at the neutral point. These features are much sharper than is possible in an independent-electron picture, but they can be accounted for if electrons and holes interact strongly and are paired in equilibrium. Our calculations from first principles show that the exciton binding energy is larger than 100 meV and the radius as small as 4 nm, explaining their formation at high temperature and doping levels. Below 100 K, more strongly insulating behaviour is seen, suggesting that a charge-ordered state forms. The observed absence of charge density waves in this state is surprising within an excitonic insulator picture, but we show that it can be explained by the symmetries of the exciton wavefunction. Therefore, in addition to being a topological insulator, monolayer WTe_2 exhibits strong correlations over a wide temperature range.

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