Anomalous behavior of the excited state of the A exciton in bulk WS2

Results of optical spectroscopy studies on bulk 2H-WS2 at energies close to its direct band gap are presented. Reflectance and absorption measurements at low temperature show only one dominant feature due to the A exciton of bulk WS2 at similar to 2.02 eV. However, a laser-modulated photoreflectance spectrum looks quite different, revealing a second even stronger feature A* similar to 62 meV above A. The relative intensity of these two features is shown to change significantly in a lateral electroreflectance measurement with electric field applied perpendicular to the c axis of WS2. The experimental results are analyzed by comparison with many-body perturbation theory calculations, including the solutions of the Bethe-Salpeter equation. A* is identified as the first excited state of the A exciton, that is, A(n = 2). The anomalous behavior of A* is explained by its distinct wave function spread along the c axis, the direction of weak van der Waals bonding, which makes it more susceptible to perturbations. Our ab initio calculations suggest that the A exciton in the ground state has a two-dimensional (2D) nature with a large binding energy E-b, in fair agreement with E-b similar to 90 +/- 20 meV estimated from a temperature-dependent reflectance study. The applicability of the 2D hydrogenic Wannier-Mott model for the exciton spectrum of a layered semiconductor like bulk WS2 is discussed.
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