RYDBERG EXCITON STATES IN DIAMOND

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Observation of Rydberg excitons generally requires a high binding energy and thus a large energy gap. Although diamond is an indirect semiconductor with ultrawide bandgap (E_g~5.5 eV) and high binding energy for excitons, the effect of Rydberg excitons remained elusive. Here, I will review recent progress in understanding Rydberg exciton states in diamond.

The Rydberg excitons in diamond were first observed via Lyman transitions from the 1s to *n*p states in the terahertz region [1,2], where *n* is the principal quantum number of the hydrogenic states. The induced absorption from the 1s to ionization continuum of excitons was also found in the mid-infrared absorption spectra [3]. Based on the terahertz time-domain spectroscopy on the Drude response of free carriers and the excitonic Lyman transitions, we evaluated the exciton binding energy to be 93.8 ± 8.2 meV [2]. A more detailed analysis of the fine splitting of 2p exciton states enabled refining the value to $E_b = 93.3 \pm 2.0$ meV and obtaining the Rydberg energy $R_y = 83.5 \pm 2.9$ meV [2], slightly deviating from E_b due to fine structure splitting of the 1s exciton states [4,5].

Meanwhile, we theoretically described the absorption spectrum of diamond [6] by considering the electron-phonon coupling, the 1s exciton with fine structure, and Rydberg states following the relation, $E = E_g - R_y\, n^{-2}$. An additional overall factor of 5.0 was multiplied to the calculation to reproduce the measured absorption coefficients [6,7]. This enhancement factor originates from the increased effective mass of the 1s exciton, compared to the one derived from the spherical bands in the effective-mass approximation [8]. We found that Rydberg states play a crucial role in enhancing the mass, density-of-states, and absorption strengths through mixing with the 1s states.

References

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