Abstract Submitted
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Optical spectra and exchange-correlation effects in molecular crystals[1] NA SAI, University of Texas at Austin, MURILO L. TIAGO, Oak Ridge National Laboratory, JAMES R. CHELIKOWSKY, University of Texas at Austin, FERNANDO A. REBOREDO, Oak Ridge National Laboratory — We report first-principles GW-Bethe Salpeter Equation and Quantum Monte Carlo calculations of the optical and electronic properties of molecular and crystalline rubrene ($C_{42}H_{28}$). We predict the formation of intermolecular, charge-transfer spin-singlet excitons with energies in close agreement with the observed yellow-green photoluminescence in rubrene microcrystals. In contrast, spin-triplet excitons are localized and intramolecular with a predicted phosphorescence at the red end of the optical spectrum. We find that the exchange energy plays a fundamental role in raising the energy of intramolecular spin-singlet excitons above the intermolecular ones. Exciton binding energies are predicted to bearound 0.5 eV (spin singlet) to 1 eV (spin triplet). The calculated electronic gap is 2.8 eV. The theoretical absorption spectrum agrees very well with recent ellipsometry data.

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