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Origin of one-photon and two-photon absorption peaks in PbSe nanocrystals from atomistic pseudopotential calculations[1] ALBERTO FRANCESCHETTI, JUN-WEI LUO, ALEX ZUNGER, National Renewable Energy Laboratory — PbSe nanocrystals represent the paradigm nanoscale system exhibiting carrier multiplication upon light absorption, yet the origin of the absorption peaks remains elusive. The second peak in the one-photon absorption spectrum has been often attributed to dipole-forbidden P-S and S-P inter-band transitions. Recent two-photon photo-luminescence excitation experiments have shown that the S-P and P-S transitions, which are selectively excited by two-photon absorption, are indeed close in energy to the second one-photon absorption peak. Here we report atomistic pseudopotential calculations of the one-photon and two-photon absorption spectra of PbSe nanocrystals 30.6 Å in radius. We find that the main two-photon absorption peak indeed originates from P-S and S-P transitions, and is blue-shifted by 36 meV with respect to the second one-photon absorption peak, in excellent agreement with experiment. However, our calculations show unequivocally that, contrary to previous interpretations, the second one-photon absorption peak originates from dipole-allowed P-P transitions.

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