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Optical transitions and the nature of Stokes shift in spherical CdS quantum dots[1] DENIS DEMCHENKO, LIN-WANG WANG, Lawrence Berkeley National Laboratory, Berkeley, California 94720 — Resonant Stokes shift observed in CdS quantum dots (QDs) has been previously studied theoretically using **k·p** approach. The large values of measured Stokes shift along with the structure of the excitonic levels obtained by the **k·p** calculations have suggested an optically forbidden P envelope valence state, thus forming a spatial symmetry induced "dark exciton" in CdS QDs, in contrast with the spin-forbidden exchange interaction induced "dark exciton" found in CdSe QDs. Since the **k·p** method has been known to incorrectly predict the energy levels in other QDs, here we apply *ab initio* accuracy methods to study this problem. Using the LDA-based charge patching method to generate the Hamiltonian, combined with the folded spectrum method to solve the single particle states of thousand-atom nanostructures, we find that the top of the valence band state is S-like, thus optically bright, in contrast with all the previous **k·p** calculations. Our results also indicate the range of applicability of the **k·p** method. The calculated electron-hole exchange splitting suggests that the spin-forbidden valence state may explain the nature of the "dark exciton" in CdS quantum dots.

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