Computational modeling of optical properties of host material for laser spectroscopy

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Upconversion (UC) phosphors are able convert incident light into light of a shorter wavelength. The (erbium) Er^{3+}, (ytterbium) Yb^{3+}: $NaYF_4$ system is the most efficient upconversion phosphor known, and yet the quantitative aspects of the mechanism responsible for upconversion, such as the values of microscopic energy transfer rate constants, have to be determined. Ab initio computation of such constants holds promise for further research. We focus on rare-earth-doped hexagon (β) - $NaYF_4$ crystal exhibiting a photoluminescence output as the novel upconversion nanocrystals, where β- $NaYF_4$ with a strong cation disorder [1-2] serving as host material. This material is modeled with atomic models of different size $Na_3Y_3F_{12}$, $Na_6Y_6F_{24}$, $Na_{12}Y_{12}F_{48}$, and $Na_{24}Y_{24}F_{96}$ using periodic boundary conditions. In the project, geometry optimization, thermal modification, and molecule dynamics process are implemented for β- $NaYF_4$ molecular structure based on density functional theory (DFT) with the generalized gradient approximation [3-4]. All structures display very similar properties, i.e. total energy per atom and inter-atomic distances show variation around 1%. The characteristics of β- $NaYF_4$ are investigated including optical absorption spectra, excitation lifetime, orbital's spatial distribution and energies in ground state and excited states, spin density, possibility of electron transfer, and partial charge density. The simulation of absorption spectra is consistent with the literature. [5]

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