DFT STUDY OF POINT DEFECTS IN YTTRIUM ALUMINUM GARNET DOPED WITH CARBON

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Point defects in the YAG crystal are studied using the density functional approach as implemented in the open source SIESTA code. The formation energies of native point defects and C substitution and interstitial defects are computed. The concentrations of charged point defects are found from the overall charge neutrality condition. The defect energy levels in the band gap are obtained.

Keywords: density functional theory, laser crystals, impurity defects

The point defects in pure YAG (Yttrium aluminium garnet, $Y_3Al_5O_{12}$) and YAG doped with C are studied. To calculate defect formation energies we use the Kohn-Sham density functional theory (DFT) method in the generalized gradient approximation with the Perdew-Burke-Ernzerhof parametrization for the exchange-correlation functional and double-zeta basis with polarization orbitals [1]. The pseudopotentials were generated with the improved Troullier-Martins scheme. Lattice vectors were allowed to relax until the maximum residual stress component converged to less than 0.05 GPa. Atomic positions were optimized until the residual forces had been less than 0.005 eV/A. A real-space grid with the plane-wave cutoff energy E_c = 950 Ry was used to calculate the total energy of the system.

The cell of YAG consists of 160 atoms: 24 atoms of yttrium, 40 atoms of aluminium and 96 atoms of oxygen. To perform the numerical calculation one can take the cell of YAG and set the coordinates of all atoms. To make a defect one can remove some atom from the cell (this is a vacancy), or can replace an atom of some sort with an atom of another sort (this is a replacement). When an atom is removed from the cell or is replaced with another one, there can be extra electrons, or there can be lack of electrons. It means, that the cell with defects can become charged.

Let's consider the case when one atom of sort *i* is removed from the cell and/or atom of sort *k* is added to the cell. Let's suppose that the charge of a defective cell is equal to *q* (in units of *e*, the elementary charge). In this case the energy of formation of a defect is given by the equation [2]

$$E_i = E_{def.cell} - E_{perf.cell} + \mu_i - \mu_k + \mu_e q + E_M \qquad (1)$$

where $E_{\text{def.cell}}$ – is the total energy of the cell with defects, $E_{\text{perf.cell}}$ – is the total energy of the perfect cell (without defects), μ_i – is the chemical potential of an atom of sort i, μ_k – is the chemical potential of an atom of sort k, μ_e – is the chemical potential of electrons, E_M is the Madelung energy. For the calculation of μ_k and E_M the approach described in [3] is used.

The concentrations of point defects are given by equation $c_i=(1/\Omega_0)\cdot n_i\cdot\exp(-E/[k_\beta\cdot T]$, where Ω_0 is the unit cell volume, n_i is the number of ways to place a given defect specie in the unit cell, and T is the temperature of sintering or annealing. The chemical potential of electrons is calculated from the charge neutrality equation [2]:

$$\sum_i q_i c_i + c_h - c_e = 0 \tag{2}$$

The concentrations of free carriers (electrons and holes) are evaluated as

$$c_e = \exp(\left[E_{CBM} - \mu_e\right]/\left[k_B T\right]) \int_0^\infty dE\, D(E_{CBM} + E) \exp(-E\left[k_B T\right]) \tag{3}$$

$$c_h = \exp(\left[\mu_e - E_{VBM}\right]/\left[k_B T\right]) \int_0^\infty dE\, D(E_{VBM} - E) \exp(-E\left[k_B T\right]) \tag{4}$$

Where E_{CBM} is the conducting band minimum, E_{VBM} is the valence band maximum, and $D(E)$ is the electron density of states.

In our study we consider the oxygen vacancies, cation vacancies, the C_{Al} and C_o substitutional defects and C interstitial defects in different charge states. For all considered point defects we calculate the band structure and obtain the positions of the defect levels in the band gap.

This work was performed using computational facilities of the Joint computational cluster of State Scientific Institution "Institute for Single Crystals" and Institute for Scintillation Materials of National Academy of Sciences of Ukraine incorporated into Ukrainian National Grid.

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