2P25

Excited state study on yttrium compounds using theoretical methods.

○Agalya Govindasamy[1)], Chen Lv[1)], Hideyuki Tsuboi[1)], Michihisa Koyama[1)], Momoji Kubo[1,2)], Ewa Broclawik[3)], Akira Miyamoto[1,3)]

[1]Grad. Sch. Eng., Tohoku Univ. (6-6-07 Aoba, Aramaki, Aoba-ku, Sendai 980-8579)
[2]JST-PRESTO (4-1-8 Honcho, Kawaguchi, Saitama 332-0012)
[3]NICHe, Tohoku Univ. (6-6-10 Aoba, Aramaki, Aoba-ku, Sendai 980-8579)

【Introduction】

Rare earth materials have been widely used in optoelectronic devices, because their photoluminescence exhibits high quantum yields and very sharp spectral bands. Recently many oxides, such as TiO_2, ZrO_2, Al_2O_3 and Y_2O_3 films have been studied in order to use them as wave-guide materials in optoelectronic integrated circuits. Among these oxides, Y_2O_3 film has received particular attention because of its large band gap (5.8eV). Furthermore, Eu doping techniques can control the optoelectronic properties of yttrium oxide. The main applications of Y_2O_3: Eu as commercial phosphor are in lighting and cathode ray tube, display material, in tricolor fluorescent lamps, field emission displays and laser devices. Because of promising applications the further investigations of Y_2O_3: Eu to improve its properties is desirable. Consequently study of the excited state of europium-doped yttrium oxide is of major significance. In the present study we report studies performed on the different possible spin multiplicities of europium doped yttrium oxide as well as yttrium oxy-sulphide using density functional theory method to unveil the influence of europium ion in yttrium compounds.

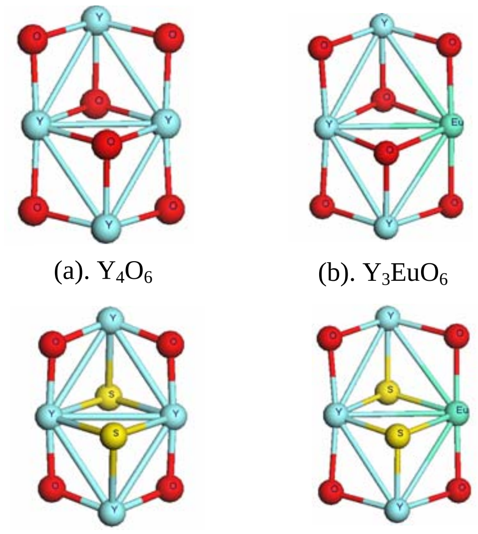
【Method】

We used the program package $DMol^3$ that is based on DFT. The geometries were optimized at Generalized Gradient Approximation level with Becke exchange and Lee-Yang-Parr 1988 correlation correction. DNP (Double Numerical and Polarization) Basis set and Effective core potential were used. Cluster models of Y_4O_6, Y_3EuO_6, $Y_4O_4S_2$, $Y_3EuO_4S_2$ were investigated

【Results and Discussion】

A small cluster model with four yttrium and six oxygen atoms was taken from the crystal structure of Y_2O_3 with BCC lattice and is shown in Fig. 1(a). Cluster (a) was then doped with one Eu atom and is shown in Fig. 1(b). Yttrium oxy-sulphide cluster was obtained by replacing two oxygen atoms by sulphur in cluster (a) and the structure is shown is Fig. 1(c). Yttrium oxy-sulphide was then doped with one Eu atom and is shown in Fig. 1(d). The geometries of yttrium oxide and yttrium oxy-sulphide clusters and its Eu doped analogues were optimized for different spin multiplicities based on density functional theory and the energies are given in Table.1.

The results show that the septet states of Y_4O_6 and $Y_4O_4S_2$ are the most stable and the quintet states are the least stable. This indicates that the ground states of Y_4O_6 and $Y_4O_4S_2$ have septet multiplicities. In the case of their Eu doped analogues, septet states are found to lie close to singlets and are stable. This can be explained in terms of all the six outer electrons being paired or unpaired. So the ground state of the Eu doped analogues might be singlets or septets. Moreover the excited states have triplet or quintet multiplicities. The triplet states of Eu doped analogues are the least stable. These results indicate that in absorption/emission spectroscopy the transition from septet state to quintet state should be observed, which aggress with experiment.



(a). Y_4O_6 (b). Y_3EuO_6 (c). $Y_4O_4S_2$ (d). $Y_3EuO_4S_2$

Fig. 1. Cluster models of Yttrium oxide compounds studied.

Table.1. Energies for different spin multiplicities using DFT.

Multiplicity	Energy (eV)			
	Y_4O_6	Y_3EuO_6	$Y_4O_4S_2$	$Y_3EuO_4S_2$
Singlet	-58.46	-54.53	-53.58	-49.90
Triplet	-55.96	-51.31	-51.08	-46.61
Quintet	-52.47	-52.05	-48.03	-47.63
Septet	-59.95	-54.52	-54.49	-49.90