

### 3 - 15 First-principle Investigation of LiTaO<sub>3</sub>

He Wenhao, Gao Xing and Wang Zhiguang

As an important multi-functional material applied in surface acoustic wave devices, optical communications, laser and optoelectronics due to its good photoelectric and piezoelectric properties, lithium tantalate (LiTaO<sub>3</sub>) has drawn extensive interests<sup>[1]</sup>. Numerous physical properties of LiTaO<sub>3</sub>, such as acoustical, electronic structures, optical properties and effective mass, thermodynamic properties, have been reported in several papers.

Point defects will affect the electronic structure which should be responsible for many physical properties, especially optical properties. At present, it has been reported that Oxygen vacancies numerous form in Oxygen-poor environment in LiTaO<sub>3</sub>. There are also massive experiments showing the optical properties change of LiTaO<sub>3</sub> crystal irradiated by various particles. This change in irradiated crystal can be attributed to abundant point defects induced by irradiation, including interstitials and vacancies. For instance, the decline of transmittance of LiTaO<sub>3</sub> crystal irradiated by Ar ions with the energy of 217 keV was observed, and the author confirmed that the result was related to the oxygen vacancies formed in the irradiated LiTaO<sub>3</sub> crystal. In LiTaO<sub>3</sub> crystal irradiated by low-energy Ar ions, it has been found that the different type of defect introduced difference of absorption position in visible light region.

However, the stable vacancies in each different chemical environments are still unclear and the mechanism for which vacancy affects the optical property is also unclear. In order to explain and predict its optical property change in different service environments, we need to study the effect of vacancy on optical properties. As the first step, we have calculated the neutral and charged vacancies formation energy in LiTaO<sub>3</sub> with first-principles, and stable vacancy type and reactions have been revealed in this study.

First-principle total energy calculations were carried out with the Vienna Ab initio Simulation Package (VASP) based on the density function theory (DFT). The projected augmented wave (PAW) pseudopotentials were used within the generalized gradient approximation (GGA) with Perdew-Burke-Ernzerhof (PBE) functional for the exchange and correlation energies in the calculations. A cutoff energy of 500 eV was used for the plane-wave expansion. The criterion for self-consistency in the electronic structure determination was that two consecutive total energies differ by less than  $1 \times 10^{-8}$  eV. The internal structure relaxations were considered to finish when residual forces on each atom was less than 0.01 eV/Å. A  $\gamma$  centered  $4 \times 4 \times 4$  Monkhorst-Pack k-mesh was used for all the calculations of supercells.

We can obtain the atomic chemical potential by using the theorem that atomic chemical potential of each atom type in all the phases is equals at equilibrium points. The Gibbs ternary diagram of Ta-Li-O is shown in Fig. 1. The formation energy of each type vacancies in all the chemical environments is shown in Fig. 2. Compared to others, the formation energy of  $V_{\text{O}}^{2+}$  is lowest in equilibrium point D. The  $V_{\text{O}}^{2+}$  formed easily in equilibrium points D. We can define that points A and B are the oxygen-rich chemical environment and point D is the oxygen-poor chemical environment. As stated above, formation  $V_{\text{O}}^{2+}$  is not easiest in oxygen-rich environment. In contrast, formation  $V_{\text{O}}^{2+}$  is easiest in oxygen-poor environment. This result is satisfied with the phenomenon observed by L. A. Kappers<sup>[2]</sup>.

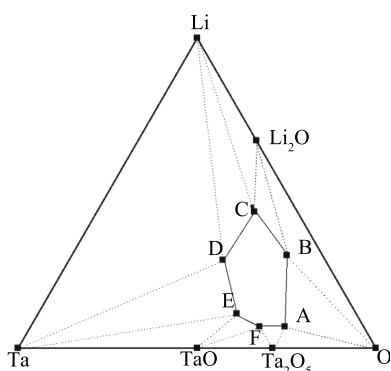


Fig. 1 The Gibbs ternary diagram of Ta-Li-O.

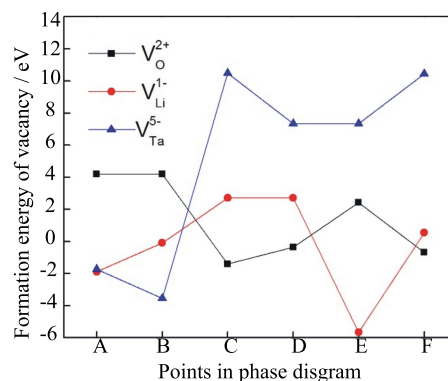


Fig. 2 (color online) The formation energy of each type vacancies in all the chemical environments.

#### References

- [1] D. A. Scrymgeour, V. Gopalan, A. Itagi, et al., Phys. Rev. B, 71(2005)184110.
- [2] L. A. Kappers, K. L. Sweeney, L. E. Halliburton, J. H. W. Liaw, Phys. Rev. B, 31(1985)6792.