Electronic structure of oxygen deficient nonstoichiometric tantalum oxide

T.V. Perevalov[1,2], V.A. Gritsenko[1,2], V.N. Kruchinin[1], V.Sh. Aliev[1], V.A. Pustovarov[3], I.A. Prosvirin[4]

[1]Rzhanov Institute of Semiconductor Physics SB RAS, 13 Lavrentiev Ave, 630090, Novosibirsk, Russia
[2]Novosibirsk National Research University, 2 Pirogov Str., 630090, Novosibirsk, Russia
[3]Ural State Technical University, 19 Mira Str., 620002, Ekaterinburg, Russia
[4]Boreskov Institute of Catalysis SB RAS, 5 Lavrentiev ave., 630090, Novosibirsk, Russia

Tantalum oxide (Ta_2O_5) is a perspective high-κ dielectric for a wide range of applications in microelectronics. Ta_2O_5 films have promising for use as a recording medium in flash memory devices [1]. Ta_2O_5 is one of the promising materials for ReRAM [2]. As a storage medium in ReRAM is used non-stoichiometric metal enriched tantalum oxide (TaO_x) with a high concentration of oxygen vacancies [3].

Ta_2O_5 films were deposited using sputtering of a Ta target with a reactive ion beam in oxygen. Stoichiometry was changed by reducing oxygen pressure in grow chamber. XPS spectra were obtained using a SPECS's machine with monochromatic Al*Kα* radiation (1486.74 eV). Quantum-chemical simulations were performed for the λ-Ta_2O_5 (as the closest to the amorphous [4]) in terms of the density functional theory using the hybrid B3LYP functional in the Quantum-ESPRESSO code [5]. The refractive index n(ω) and extinction coefficient k(ω) of TaO_x films were determined by means of spectroscopic ellipsometry. Photoluminescence (PL) spectra obtained using a 400W deuterium discharge lamp and the primary double-prism monochromator DMR-4. PL excitation spectra were normalized to the equal number of photons incident on the sample using a yellow lumogen.

TaO_x films have broadening of Hf*4f* level to lower binding energy and a formation of defect states at the 2.2 eV above the valence band top caused by oxygen depletion. The superposition of calculated valence band XPS for crystalline Ta_2O_5 with oxygen vacancies and the experimental one for TaO_x show good agreement in a defect level position and intensity. The [O]/[Ta] atomic ratio for TaO_x estimated from the XPS peak area close to atomic ratio estimated from the theoretical XPS. The optical absorption spectrum Ta_2O_5 with oxygen vacancies shows a peak at 4.55 eV. The PL spectra of in tantalum oxide film with different oxygen vacancies' concentrations show the PL band centered near a 2.7 eV peak. There is a broad band at 4.5 eV in the PLE spectrum for 2.7 eV emission. Thus, the 2.7 eV PL emission is excited in the absorption/PLE band near 4.5 eV in Ta_2O_5. The thermal activation trap energy of oxygen vacancy determined as half of Stokes shift, is equal to 0.9 eV. It was concluded that the 2.7 eV luminescence excited near 4.5 eV in a Ta_2O_5 film is associated with the oxygen vacancy.

1. C. H. Cheng et al., Appl. Phys. Lett. **91**, 192903 (2007).
2. S.Z. Rahaman et al., Nanoscale Research Letters **7**, 345 (2012).
3. S.U. Sharath et al., Appl. Phys. Lett. **109**, 173503 (2016).
4. S.-H. Lee, Phys. Rev. Lett. **110**, 235502 (2013).
5. P. Giannozzi et al., J. Phys.: Condens. Matter **21**, 395502 (2009).