Contribution ID: **18** Type: **Contributed talk**

Raman spectra of a-Ta_2O_5: a first-principles analysis

Monday, 30 September 2024 15:20 (20 minutes)

We present a first-principles study of the vibrational spectra, i.e. vibrational density of states (v-DOS), infrared (IR), and Raman spectra, in amorphous tantala (a-Ta_2O_5). Model structures of upto three-hundreds atoms have been generated by means of classical and *ab-initio* molecular dynamics. An unprecedented series of *ab-initio* test calculations of the v-DOS and the IR dielectric function, with different kinds of exchange-correlation functionals and pseudopotentials, have been performed by using the Quantum Espresso package. Comparisons with available experimental data have then been carried out to select the optimal DFT setup for further investigations. Our study confirms that the vibrational modes above $\sim$550 cm^{-1} consist mainly of Ta-O stretching motion of oxygen (O) atoms, meanwhile Ta motion gives a major contribution to the v-DOS only below $\sim$200 cm^{-1}. In particular, the vibrational modes underlying the main Raman peak at 670 cm^{-1} are related to Ta–O bond stretching motions, and a major contribution to the peak appears to come from twofold O atoms. Finally, by means of projectional analysis we infer that vibrational modes with frequencies above 550 cm^{-1}, arise mostly from the asymmetric and symmetric stretching modes of TaO_n ($n = 5, 6, 7$) polyhedra. The latter analysis, in particular, allows to explain the origin of the experimental Raman doublet at $\sim$830-840 and $\sim$935-945 cm^{-1}.

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Session Classification: Coating and Materials

Track Classification: Coatings and Materials