

Density-Functional-Theory Analysis of Electron-Phonon Interaction in 2D/3D α -MoO₃

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[Introduction] α -MoO₃, a layered wide-gap semiconductor, has attracted considerable attention for a long time because of its potential in many applications, from sensors to catalysts^[1]. Recently, two-dimensional (2D) α -MoO₃ has also been synthesized, showing high mobility and high permittivity, which is suitable for the application in field effect transistors^[2]. The carrier transport in α -MoO₃ is underpinned by electron-phonon interaction. Therefore, investigation of electron-phonon interaction in this material is high demand in order to improve their transport properties. Here, we focus on the electron-phonon coupling calculation by density functional theory (DFT) to evaluate phonon-limited mobilities of bulk (3D) and 2D α -MoO₃ based on the Boltzmann transport equation.

[Methodology] Our DFT calculations were carried out with Quantum Espresso package. Norm-conserving pseudopotential and the generalized gradient approximation were adopted. 90 Ry was set for the cutoff energy of plane wave basis both in the 3D and 2D α -MoO₃. For the electron (phonon) states of the 2D and 3D cases, the 1st Brillouin zone was sampled by $8 \times 8 \times 1$ ($8 \times 8 \times 2$) and $8 \times 8 \times 2$ ($8 \times 8 \times 2$) uniform meshes, respectively. In the 2D case, a 18.3 Å-thick vacuum layer was added in the z-direction to avoid the adjacent-layer interaction. Phonon dispersions were calculated by density functional perturbation theory.

[Results] The phonon band dispersion of α -MoO₃ shows a large longitudinal optical (LO)/transverse optical (TO) splitting: frequencies of LO phonon depend on the polarization of the phonon wave vector around the Γ point. We found that the strong anisotropy in Born effective charge and dielectric tensor is the origin of the large LO/TO splitting. In order to evaluate the electron-phonon coupling strength, we calculate scattering rate. For the 2D α -MoO₃, we found that the temperature dependence of the scattering rate is small at low temperatures while large at high temperatures [Fig. 1 (a)]. This is because the electron-phonon coupling is dominated by optical phonons and a high temperature is needed to excite them. We also found that the scattering rate has strong wave vector and energy dependence [Fig. 1 (b)]. The rate becomes small at the maximum of valence bands and the minimum of conduction bands, which may be the reason for the high mobility in the 2D α -MoO₃.

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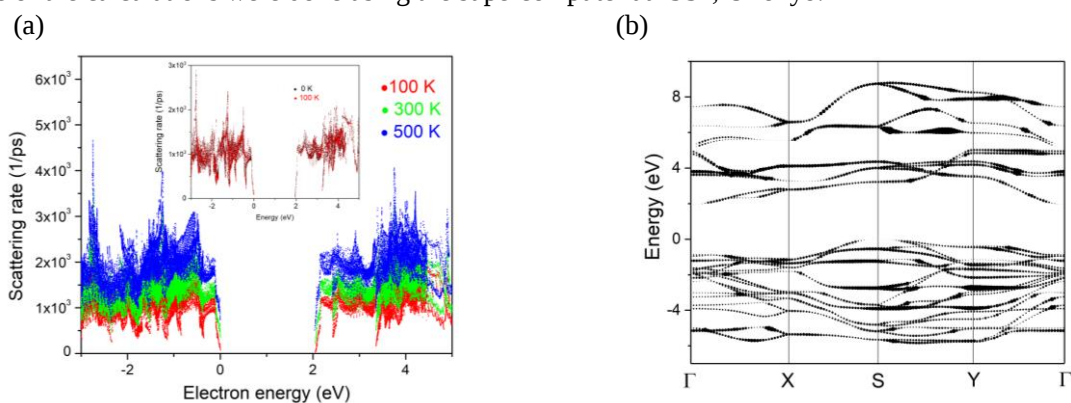


Figure 1 Temperature dependence (a) and wave vector and energy dependence (b) of the scattering rates in the case of the 2D α -MoO₃.

References

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