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Calculation of Lattice Thermal Conductivity of $UO_{2+0.25}$ by Solving Phonon Boltzmann Equation

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Abstract

The lattice thermal conductivity of UO2 and $UO_{2+0.25}$ is investigated by the relaxation time approximation solution of the Boltzmann transport equation for the steady-state phonon distribution function. To do this, we have calculated the 2nd and 3rd IFCs with finite displacement method. Results show that thermal conductivity of $UO_{2+0.25}$ is decreased by adding oxygen defects. The reason behind the decreasing the thermal conductivity is the increasing of scattering rate by defects.

Keywords: Uranium Dioxide, Density Functional Theory, Lattice Thermal Conductivity, Phonon Boltzmann Equation (BTE), Relaxation Time Approximation, Hubbard Correction.

Introduction

Nowadays UO_2 is a common nuclear fuel in nuclear plants. Urania under operating conditions or after fabrication often exists in hyperstoichiometric form, UO_{2+x}. In UO_{2+x}, oxygen ions occupy interstitial or interstitial-like sites in the parent fluorite lattice (CaF2). Because of the important role of nuclear fuels in the reactors, knowing the mechanisms that they act is critical. The thermodynamic and kinetic properties of the excess oxygen ions govern radiation tolerance, fission product accommodation, fission gas release and micro-structural evolution. As experimental studies in this field are very difficult, computational works are of great value.

Experiments have determined the crystal structure of UO_2 as a 3k-order antiferromagnetic (AFM) at $T < 30\,K$, and paramagnetic at higher temperatures [1] . The uranium atoms sit on the sites of an FCC structure with lattice constant of 5.47Å, while the oxygen atoms are positioned at sites with $Pa\bar{3}$ symmetry [2] as shown in figure 1. Uranium-dioxide is electrically an insulator material (the so-called Mott insulator), and because of the localized partially filled f-orbitals on U atoms, it is a strongly correlated electron system.

In this study, we investigate the lattice thermal conductivity of bulk UO_2 and $UO_{2+0.25}$ with considering Hubbard correction and solving the BTE. Two software packages are used in this study. GULP [3] that is designed to perform variety of tasks based on force-fields methods and ALAMODE [4] that is designed for analyzing lattice thermal conductivity.

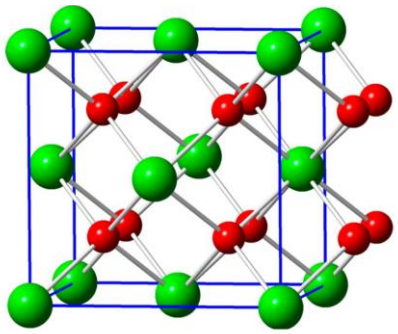

Figure 1. The fluorite structure of UO_2. The green balls represent uranium atoms and red balls represent oxygen atoms.

Computational Detail

The lattice thermal conductivity is calculated by solving the linearized-BTE [5] for the steady-state phonon distribution function f_λ :

$$\nabla T.v_\lambda \frac{\partial f_\lambda}{\partial T} = \left.\frac{\partial f_\lambda}{\partial T}\right|_{scatt.} \qquad (1)$$

Where the left-hand side of the equation corresponds to the phonon diffusion due to temperature gradient, and the term in the right-hand side is the time rate of change of phonon distribution due to all allowed scattering processes. Here, v_λ is the group velocity of phonon in mode λ and $\lambda \equiv (s,q)$ with s and q being the phonon branch index and wave vector in reciprocal space, respectively.

The BTE can be solved using a full iterative algorithm [6], or using relaxation time approximation (as is done here) by employing ALAMODE code package.
Using a 1k-order AFM calculation, we have first fully optimized the geometry of the UO_2 primitive unit cell with space group number 225 and obtained the optimized value of lattice constant for the FCC structure as 5.56 Å.
Using the optimized geometry lattice parameter, the 2nd-IFCs and 3rd-IFCs were calculated employing the finite displacement method.

Results and discussion

In this study, we have taken a supercell for the bulk UO_2 with 96 atoms and for the $UO_{2+0.25}$ with 104 atoms. In each case, the systems were optimized using GULP at first and then we calculated the 2nd and 3rd IFCs from finite displacement method using ALAMODE code package. In both systems, the displacement value for the 2nd order force constant was 0.01 angstrom and all possible displacements are taken into account. For 3rd IFCs calculations, the displacement value was 0.03 angstrom and again all possible displacements are taken into account. In each step, the values was chosen according to the convergency tests. Finally, the thermal conductivity was obtained. Thermal conductivity as a function of temperatures is shown in figure 2. As shown in the figure 2, thermal conductivity of $UO_{2+0.25}$ is reduced compared to the thermal conductivity of UO_2.
As expected, in the presence of defects the scattering rate increase and then the thermal conductivity decrease.

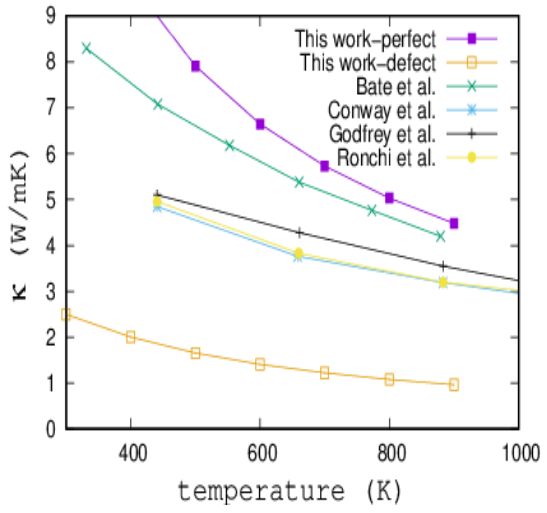

Figure 2. Thermal conductivity of UO_2 (purple squares) and $UO_{2+0.25}$ (orange squares) as functions of temperature compared to experimental results[7-10]. The experimental results are for stoichiometric UO_2.

Conclusions

In this study we investigate the thermal conductivity of UO_2 and $UO_{2+0.25}$. In $UO_{2+0.25}$, the defects cause to increase the scattering rate that leads to decreasing the thermal conductivity.

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