

Supplementary Information for

Anisotropic Thermal Transport in Weyl semimetal TaAs: a First-principles Calculation

Tao Ouyang^{a,*}, Huaping Xiao^a, Chao Tang^a, Ming Hu^{b,c,*}, and Jianxin Zhong^a

^a*Hunan Key Laboratory for Micro-Nano Energy Materials and Device and Department of Physics, Xiangtan University, Xiangtan 411105, Hunan, China*

^b*Institute of Mineral Engineering, Division of Materials Science and Engineering, Faculty of Georesources and Materials Engineering, RWTH Aachen University, 52064 Aachen, Germany*

^c*Aachen Institute for Advanced Study in Computational Engineering Science (AICES), RWTH Aachen University, 52062 Aachen, Germany*

1. Workflow for the thermal transport properties calculated in this paper

The macro/micro thermal transport properties such as lattice thermal conductivity, phonon dispersion, group velocities and phonon relaxation time are determined by harmonic (second-order) and anharmonic (third-order) interatomic force constants (IFCs), which can be obtained from *ab-initio* calculations. Such calculations are carried out with the Vienna Ab-initio Simulation Package (VASP). Once the harmonic and anharmonic IFCs are obtained, the thermal transport properties can be calculated by solving the phonon Boltzmann transport equation (BTE)

*E-Mail: ouyangtao@xtu.edu.cn (T.O.)

*E-Mail: hum@ghi.rwth-aachen.de (M.H.)

as implemented in the ShengBTE code¹. The detailed information on the workflow can be found in the Fig. S1.

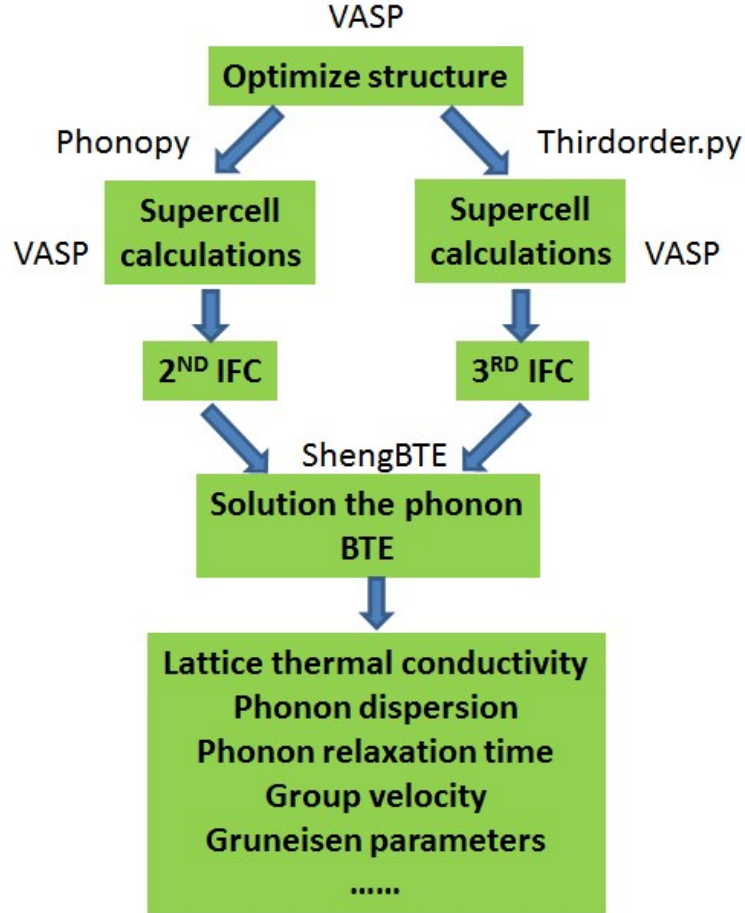


Fig. S1. Workflow for the thermal transport properties calculations using density functional theory combining phonon BTE method.

From the solution of the BTE, the lattice thermal conductivity could be expressed as:

$$\kappa_{\alpha} = \frac{1}{V} \sum_{\lambda} C_{\lambda} \vec{v}_{\lambda, \alpha}^2 \tau_{\lambda} \quad , \quad (S1)$$

where V is the crystal volume, \vec{v} the group velocity vector $\vec{v} = d\omega/d\vec{k}$. The C_λ is the mode specific heat of system:

$$C_\lambda = \frac{\hbar\omega\partial f}{V\partial T} = \frac{k_B x^2 e^x}{V [e^x - 1]^2} \quad (S2)$$

where $x = \hbar\omega/k_B T$. When the phonon-phonon scattering and isotope scattering are considered, the phonon lifetime τ_λ can be obtained from the Matthiessen rule ²:

$$\frac{1}{\tau_\lambda} = \frac{1}{\tau_\lambda^{Ph}} + \frac{1}{\tau_\lambda^{iso}} \quad (S3)$$

The phonon-phonon lifetime for a three-phonon scattering process is computed as the inversion of the intrinsic scattering rate ¹:

$$\tau_\lambda^{Ph} = \frac{1}{\Gamma_\lambda} = N \left(\sum_{\lambda\lambda''} \Gamma_{\lambda\lambda\lambda''}^+ + \frac{1}{2} \sum_{\lambda\lambda''} \Gamma_{\lambda\lambda\lambda''}^- \right)^{-1} \quad (S4)$$

where λ' and λ'' denote the second and third phonon mode that scatter with phonon λ . $\Gamma_{\lambda\lambda\lambda''}^+$ and $\Gamma_{\lambda\lambda\lambda''}^-$ are the intrinsic three-phonon scattering rates for absorption processes $\lambda + \lambda' \rightarrow \lambda''$ and emission processes $\lambda \rightarrow \lambda' + \lambda''$, respectively. For more details about the BTE method we refer the reader to Refs. 1-4.

2. Lagrangian multiplier method used in ShengBTE for enforcing the translational invariance constraint of anharmonic force constants

Owing to the fact that the system energy does not change when the system as whole is displaced, one have the sum rules for third-order IFCs:

$$\sum_k \Phi_{ijk}^{\alpha\beta\gamma} = 0 \quad (\text{S5})$$

However the directly calculated force constants from *ab-initio* package do not exactly satisfy all the sum rules. Therefore, the IFCs have to be enforced by changing the calculated value slightly since the sum rules are crucial to obtain the correct scattering rates especially the low frequencies. These can be done by solving an optimization problem. The idea is to add a compensation d_i to each independent nonzero element ϕ_i , where i ranges from 1 to the total number of independent nonzero elements, such that the sum rule condition can be satisfied. In order to guarantee that the compensation is small, some additional constraints need to be considered. The ShengBTE code minimize the sum of the squares of the compensation for each independent nonzero element, and the enforcement of sum rules turns out to be a minimization of a quadratic polynomial subject to constraints, which can be easily done by using the Lagrangian multiplier method ^{5,6}.

Since all the IFCs can be deduced from the independent elements, the sums can be written in terms of these elements as:

$$\sum_j A_{ij} \Phi_j = B_i \quad (\text{S6})$$

where A_{ij} are integers in the case of a cubic system, and j ranges from 1 to the total number of independent sums. Since the sum rules have to be satisfied, the constraints on the compensation are:

$$g_i \equiv \sum_j A_{ij} \Phi_j + B_i = 0 \quad (S7)$$

The function to be minimized is:

$$f = \frac{1}{2} \sum_j d_j^2 \quad (S8)$$

After introducing the Lagrangian multiplier λ_i , the expression of d_i in terms of λ_i could be obtained from

$$\frac{\partial(f + \sum_i \lambda_i g_i)}{\partial d_j} = 0 \quad (S9)$$

from which it follows:

$$d_j = - \sum_i \lambda_i A_{ij} \quad (S10)$$

Substituting this relation into Eq. (S7), one have

$$\sum_j C_{ij} \lambda_j = B_i \quad (S11)$$

with $C_{ij} = \sum_m A_{im} A_{jm}$, λ_j can be obtained by solving the linear equation arrays, and d_j is further obtained by using Eq. (S10), When d_j is added to the independent IFC elements ϕ_i , the sum rules are completely satisfied and the compensations are minimized.

3. Electronic band structure and density of state (DOS) of TaAs

In order to verify the thermal transport in TaAs is dominated by the phonons. We calculate the electronic band structure and the corresponding density of state (DOS). The results is presented in Fig. S2. It can be seen that the band structure agrees very well with the previous theoretical work ⁷. The DOS around the Fermi level is quite small ⁸ and implies that the electronic contributed thermal conductivity in TaAs is negligible.

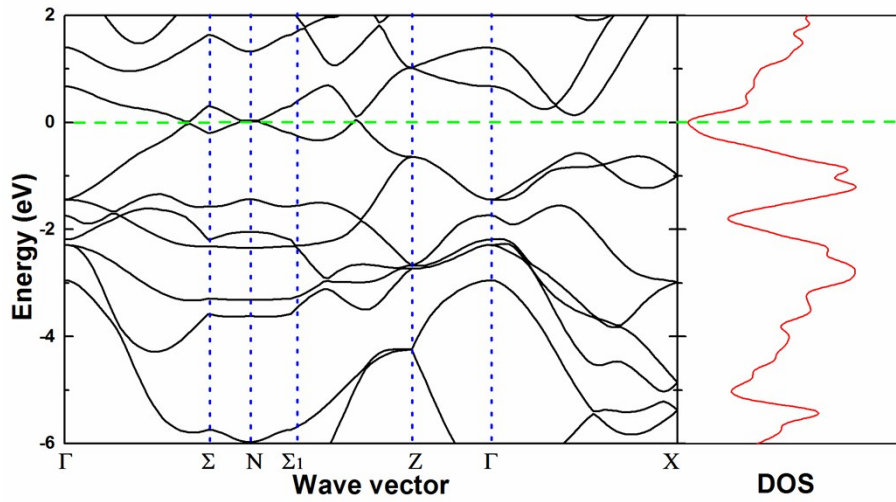


Fig. S2 (color online) The electronic energy band structure of TaAs along several high symmetry directions and the corresponding density of state (DOS).

4. Self-consistent test of the Grüneisen parameters of TaAs

In order to verify the accuracy of the third-order anharmonic IFCs, we calculate the mode specific Grüneisen parameters (γ_λ) which reflect the anharmonicity of a crystal and can be obtained from the anharmonic IFCs. Owing to the lacking of experimental data of TaAs, we perform a self-consistent test via changing the interaction cutoff of third-order force constant as

shown in Fig. S3. It can be seen that the Grüneisen parameters vary obviously for the anharmonic IFCs when the interaction cutoff is short. However, the Grüneisen parameters stay almost unchanged for the interaction cutoff larger than fourth nearest neighbors, corresponding to 4.7 Å. From the mode weighted accumulative Grüneisen parameter (MWGP) defined as $\gamma = \sum_{\lambda} C_{\lambda} \gamma_{\lambda} / \sum_{\lambda} C_{\lambda}$, we can see this behavior more clearly (see the inset of Fig. S3). That is to say, the Grüneisen parameters of TaAs are convergent when the interaction cutoff of IFCs is larger than fourth nearest neighbors, which could qualitatively characterize the accuracy of anharmonic force constants. Taking the computational accuracy and time-consuming of calculation into consideration, the sixth nearest neighbors are chosen as the interaction cutoff for the third-order IFCs in the following works.

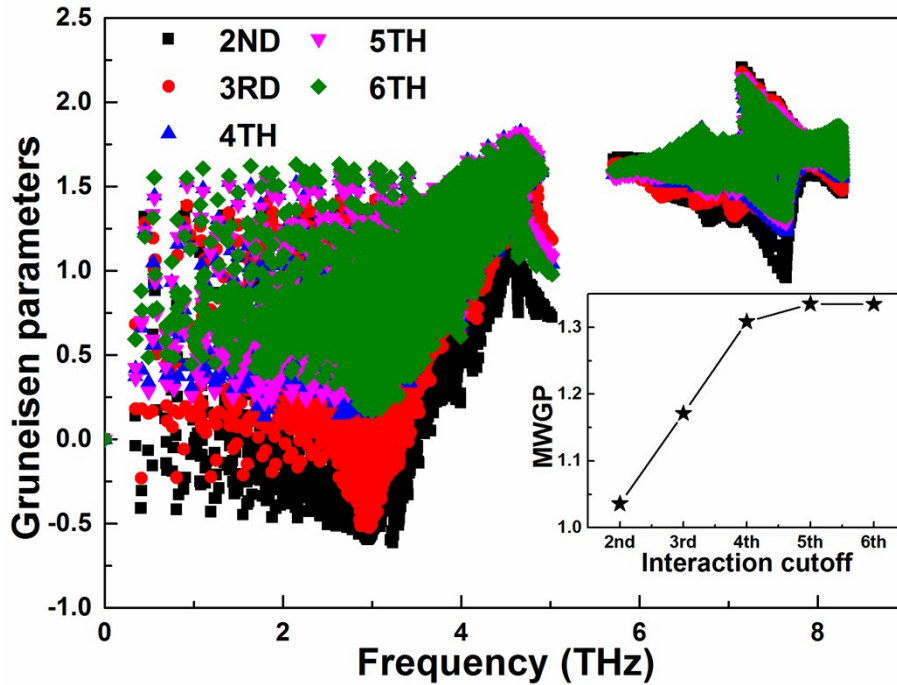


Fig. S3 (color online) Frequency dependent Grüneisen parameter of TaAs with different interaction cutoff of third-order force constants. (Inset) The mode weighted accumulative Grüneisen parameter(MWGP) with respect to the interaction cutoff.

References

1. W. Li, J. Carrete, N. A. Katcho and N. Mingo, *Comput. Phys. Commun.*, 2014, **185**, 1747-1758.
2. T. Feng and X. Ruan, *Journal of Nanomaterials*, 2014, **2014**, 25.
3. M. Omini and A. Sparavigna, *Phys. Rev. B*, 1996, **53**, 9064-9073.
4. J. E. Turney, E. S. Landry, A. J. H. McGaughey and C. H. Amon, *Phys. Rev. B*, 2009, **79**, 064301.
5. W. Li, L. Lindsay, D. A. Broido, D. A. Stewart and N. Mingo, *Phys. Rev. B*, 2012, **86**, 174307.
6. N. Mingo, D. A. Stewart, D. A. Broido and D. Srivastava, *Phys. Rev. B*, 2008, **77**, 033418.
7. S.-Y. Xu, I. Belopolski, N. Alidoust, M. Neupane, G. Bian, C. Zhang, R. Sankar, G. Chang, Z. Yuan, C.-C. Lee, S.-M. Huang, H. Zheng, J. Ma, D. S. Sanchez, B. Wang, A. Bansil, F. Chou, P. P. Shibayev, H. Lin, S. Jia and M. Z. Hasan, *Science*, 2015, **349**, 613-617.
8. C.-C. Lee, S.-Y. Xu, S.-M. Huang, D. S. Sanchez, I. Belopolski, G. Chang, G. Bian, N. Alidoust, H. Zheng, M. Neupane, B. Wang, A. Bansil, M. Z. Hasan and H. Lin, *Phys. Rev. B*, 2015, **92**, 235104.