Phononic Thermal Transport in Alloyed Nanowires of Gold and Silver

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ABSTRACT

We have performed a computational study of the phononic thermal transport in alloyed nanowires of gold and silver (Au_mAg_n; the indices m, n= 0.25, 0.5, 0.75, represent concentration of Au and Ag atoms, respectively) with main focus on exploring the effect of alloying on thermal transport in the linear and double zigzag topologies. For this purpose, we have calculated the phonon spectra using the force field method as implemented in the Phonons module of ASE python code, followed by the calculation of phononic thermal conductance k_{ph} using the Non-equilibrium Green's Function approach. Parallel to the pristine Au and Ag nanowires, k_{ph} is found to rise sharply with temperature, before saturating to a characteristic constant value. Interestingly, in linear topology the alloying suppresses k_{ph} over the pristine nanowires of both Au and Ag, with the (0.25, 0.75) configuration having its minimum value. In double zigzag topology, there result in total eight configurations for the considered concentration of Au and Ag. All configurations show a reduction in k_{ph} with respect to the pristine Ag nanowire, while an opposite trend is found with regard to the pristine Au nanowire. Reduction in phononic conductance via alloying constitutes an important finding of our study keeping in view application of noble metal nanowires in thermoelectricity.

INTRODUCTION

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From last few decades, the heat transport in noble metals nanostructures has been attracting huge interest due to its importance and expected applications in various fields of modern technology such as electronic chip heat management and thermoelectric energy generation. This interest has triggered a spate of theoretical and experimental studies on magnetic [1], optical [2–5], electrical [6] and thermal properties [7] of nanostructures. Among these nanostructures, ultrathin nanowires (NWs), have gained wide attention, since they show very fascinating properties [8, 9] drastically different from their bulk counterpart, as well as emerged as one of the most promising candidates for next generation interconnectors in nano-electronics [10, 11]. Moreover, these nanowires can be made as a suitable candidate for thermoelectric applications, if one would reduce the thermal conductivity compared to electrical conductivity. The phonon scattering at boundaries is one of the best ways to reduce thermal conductivity. The scope of application has been further explored by synthesizing NW's of alloys of different metals. Theoretically, Kumar et. al. [9, 12] has studied ultrathin nanowires of noble metal atoms by using the density functional theory (DFT). It is found that electronic, optical, magnetic and electrical transport properties of these NW's vary with structure of NW's as well as type of alloying. More recently, Singh *et al.* [13, 14] have presented a comprehensive study on thermoelectric transport properties of pristine and alloyed noble metal atomic wires modeled in different topologies using the NEGF approach. Importantly, it has been shown that the thermoelectric efficiency of atomic wires can be enhanced significantly via topology and alloying. Similar structural dependence on electronic properties of Au-Pt bimetallic NW's also has been reported by Asaduzzaman et. al. [15]. Compared to both theoretical and experimental research concerning the electronic properties, a very much less attention has been paid so for on the thermal properties of pristine NW's of noble metal, while in case of alloyed NW's, it remains untouched. In this paper, we have performed a computational study of the phononic thermal transport in alloyed nanowires of gold and silver (Au_mAg_n; the indices m, n= 0.25, 0.5, 0.75, represent concentration of Au and Ag atoms, respectively) with main focus on exploring the effect of alloying on thermal transport in the linear and double zigzag (DZZ) topologies. We have calculated the phonon transmission spectra using the force field method combined with NEGF approach [16-19], followed by the calculation of phononic thermal conductance k_{ph}.

WIRE MODEL AND COMPUTATIONAL DETAILS

In our study, we have considered linear topology and double zigzag topologies of alloyed nanowires of gold and silver (Au_mAg_n; the indices m, n= 0.25, 0.5, 0.75, represent

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concentration of Au and Ag atoms, respectively). For instance, to model $Au_{0.75}Ag_{0.25}$, unit cells used for geometry optimization in linear topology and double zigzag topologies (DZZ1 and DZZ2) are shown in Fig. 1. Atomic structure is relaxed along with the lattice within a force tolerance of 0.001 eV/Å using LBFGS [20] technique. A super cell of 72 atoms, divided into three basic parts of left electrode, scattering region and right electrode containing 12, 48 and 12 atoms respectively, is constructed using the required repetitions of the unit cells. The wire atoms are placed in x − y plane with its length is taken along the transport direction (x-axis). A vacuum gap of more than 20 Å is chosen between wires to prevent the interactions between periodic images. To get dynamical matrix *K*, we make use of empirical inter-atomic potentials [21-22] within ASE [23-24] code combined with ATOMISTICA [25]. Here, the force constant is determined from the second derivative of potential energy with respect to atomic positions in Cartesian space and thus, we include only the harmonic interactions. From the *K*-matrix, the phonon transmission function $\tau_{ph}(E)$ is then calculated using the NEGF utility PHTrans [26] and SISL [27]. Finally, the phononic thermal conductance k_{ph} is evaluated using the Landauer formula for the phonon heat current as:

$$\kappa_{ph} = \frac{1}{h}\int_0^\infty \tau_{ph}(E)E\left(\frac{\partial f_B(E,T)}{\partial T}\right)dE. \qquad (1)$$

where $f_B(E, T) = \{\exp(E/k_BT) - 1\}^{-1}$, is the distribution of phonons in each electrode.

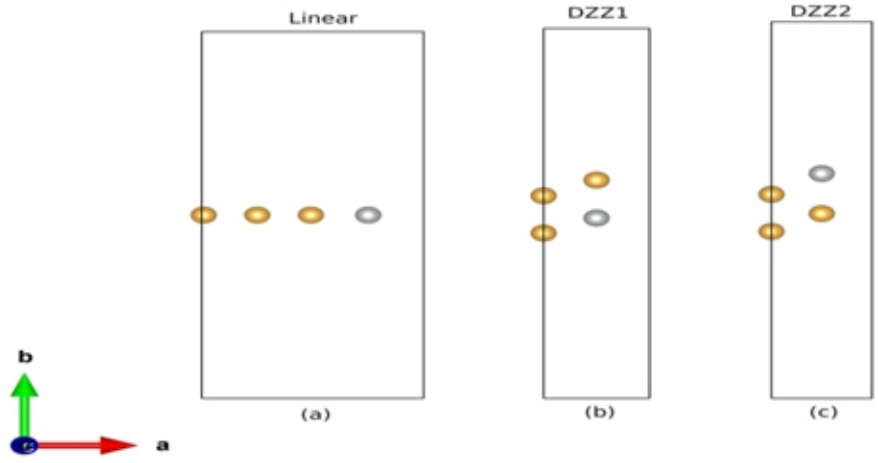

Fig. 1 Unit cells for $Au_{0.75}Ag_{0.25}$ alloyed nanowire in (a) Linear (b) double zigzag-1 and (c) double zigzag-2 topologies with periodicity taken along x-direction.

RESULTS AND DISCUSSION

First of all we calculate the phonon transmission function $\tau_{ph}(E)$ the main ingredient involved in the calculation of k_{ph}. Numerical results of $\tau_{ph}(E)$ for the pristine and alloyed

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NWs having different concentrations of Au and Ag in are shown in Fig. 2. A comparison with the pristine NWs reveals that the Debye energy (the maximum energy a phonon can have) is reduced in the alloyed NWs in all topologies over the pristine Ag, with the DZZ2 topology in both $Ag_{0.25}Au_{0.75}$and $Ag_{75}Au_{25}$ exhibiting the maximum reduction.

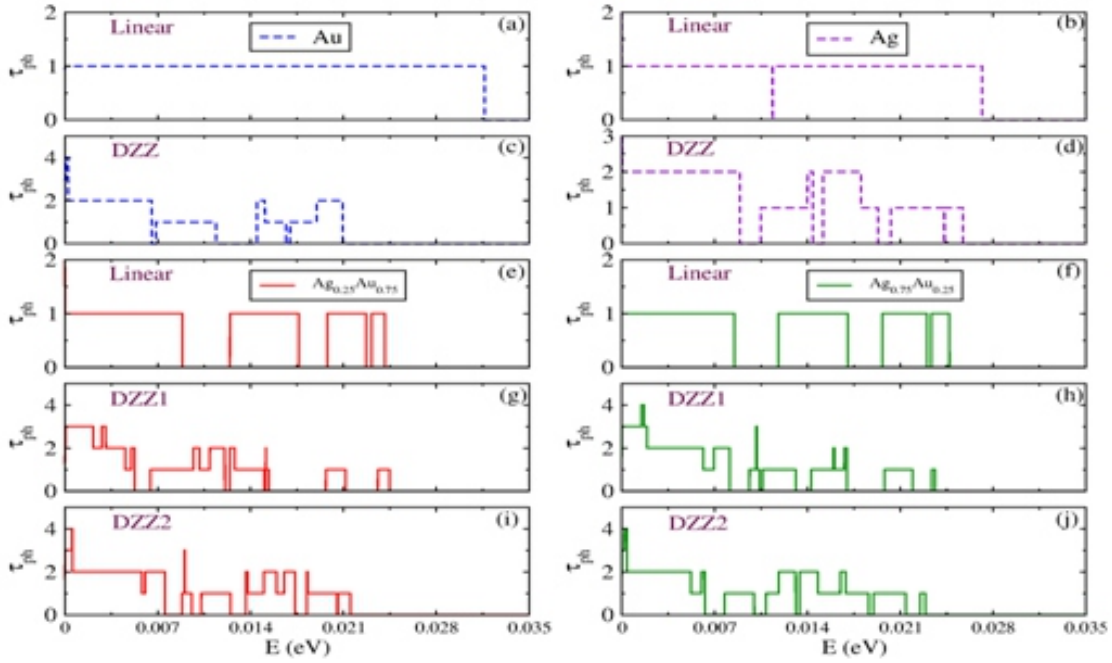

Fig. 2. The phonon transmission coefficient plotted τph as a function of phonon energy *E* for the pristine and alloyed NWs in different topologies.

Zero transmission regions in alloyed NWs, indicate that the number of conduction channels available for phonon transport decrease. Figure 3 depicts the variation of k_{ph} with temperature T for pristine and alloyed NWs. Parallel to the pristine Au and Ag nanowires, k_{ph} in alloyed NWs also found to rise sharply with temperature, before saturating to a characteristic constant value. Saturation is achieved after the characteristic Debye temperature. Interestingly, in linear topology the alloying suppresses room temperature k_{ph} over the pristine nanowires of both Au and Ag, with the (0.25, 0.75) configuration having its minimum value. All configurations show a reduction in k_{ph} with respect to the pristine Ag nanowire, while an opposite trend is found with regard to the pristine Au nanowire. In DZZ topology, $Au_{0.5}Ag_{0.5}$ attains the lowest k_{ph} with DZZ2 configuration (for structure see ref. [14]). Reduction in phononic conductance via alloying constitutes an important finding of our study keeping in view application of noble metal nanowires in thermoelectricity.

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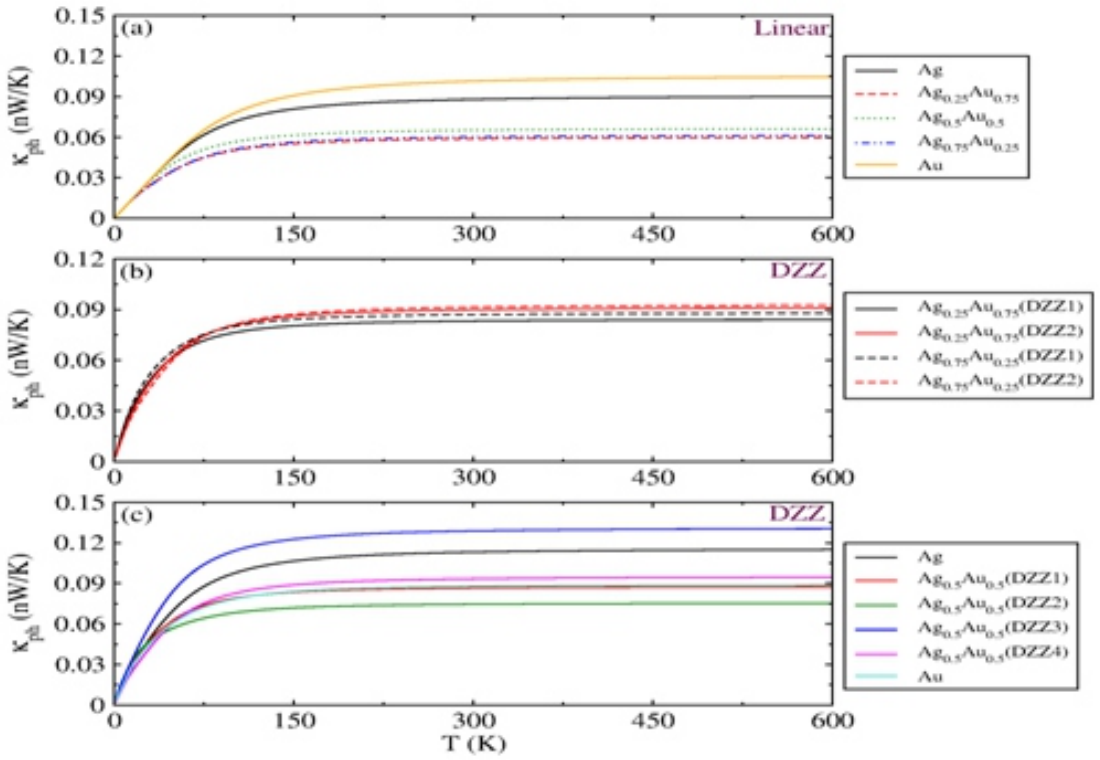

Fig. 3. The phonon thermal conductance k_{ph} plotted as a function of temperature T for the pristine and alloyed NWs.

REFERENCES

1. A. Chen and P. Holt-Hindle, Chemical Reviews 110, 3767–3804 (2010).
2. G. Albrecht, M. Ubl, S. Kaiser, H. Giessen, and M. Hentschel, ACS Photonics 5, 1058–1067 (2018).
3. X. Huang, P. K. Jain, I. H. El-Sayed, and M. A. El-Sayed, Nanomedicine 2, 681–693 (2007).
4. M. Hu, J. Chen, Z.-Y. Li, L. Au, G. V. Hartland, X. Li, M. Marquez, and Y. Xia, Chem. Soc. Rev. 35,
5. 1084–1094 (2006).
6. O. Kvıtek, J. Siegel, V. Hnatowicz, and V.Svorcik, Journal of Nanomaterials 2013 (2013)
7. P. Kapoor, M. Sharma, A. Kumar, and P. K. Ahluwalia, AIP Conference Proceedings 1942, p. 050050 (2018).
8. J. C. Klockner, M. Matt, P. Nielaba, F. Pauly, and J. C. Cuevas, Phys. Rev. B 96, p. 205405Nov (2017).
9. A. Kumar, A. Kumar, and P. Ahluwalia, Physica E: Low-dimensional Systems and

ISBN: 978-81-955611-7-9

Nanostructures 46, 259– 269 (2012).

10. X. Teng, M. Feygenson, Q. Wang, J. He, W. Du, A. I. Frenkel, W. Han, and M. Aronson, Nano Letters 9,3177–3184 (2009).
11. Y. Lu, J. Y. Huang, C. Wang, S. Sun, and J. Lou, Nature Nanotechnology 5, 218 EP –Feb (2010).
12. A. Kumar and P. Ahluwalia, Journal of alloyed and Compounds 615, 194-203 (2014).
13. G. Singh, K. Kumar and R. K. Moudgil, Physica E, 109 114 (2019).
14. G. Singh, K. Kumar and R. K. Moudgil, Phys Chem Chem Phys, 21 20965 (2019).
15. M. Asaduzzaman and M. Springborg, Phys. Rev B 72, 165422 (2005).
16. T. Yamamoto and K. Watanabe, Phys Rev Lett, 96 255503 (2006).
17. N. Mingo, Phys Rev B, 74 125402 (2006).
18. T. Ouyang, Y. Chen, Y. Xie, K Yang, Z. Bao and J. Zhong, Nanotechnology, 21 245701 (2010).
19. T. Ouyang, Y. Chen, Y. Xie, G. M. Stocks and J. Zhong, Appl. Phys Lett, 99 233101 (2011).
20. D. C. Liu, and J. Nocedal, Mathematical Programming, 45 pp. 503-528 (1989).
21. H. W. Sheng, M. J. Kramer, A. Cadien, T. Fujita, M. W. Chen, Phys. Rev. B, 83 p. 134118 (2011).
22. X. W. Zhou, R. A. Johnson, H. N. G. Wadley, Phys. Rev. B, 69 p. 144113 (2004).
23. A. H. Larsen *et al*., Journal of Physics: Condensed Matter, 29 p. 273002 (2017).
24. S. R. Bahn, K. W. Jacobsen, Comput. Sci. Eng. 4 pp. 56-66 (2002).
25. atomistica: v.0.8.0, 2019. URL: https://github.com/Atomistica/atomistica
26. N. Papior, N. Lorente, T. Frederiksen, A. García, M. Brandbyge, Computer Physics Communications, 212 pp. 8-24 (2017).
27. Nick Papior, Jonas L. B., pfebrer, Thomas Frederiksen, & Sofia Sanz Wuhl. (2021). zerothi/sisl: v0.11.0 (v0.11.0).

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