

An electron-phonon coupling model for studying the effective thermal conductivity of nanocrystalline materials

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Abstract. The thermal conductivity of Nan crystalline materials has become one of the most advanced topics in international research. In this work, the effective thermal conductivity of NC materials based on the two-temperature model of heat conduction was studied. In the frame work of the developed model, we take the Nan crystalline materials as a composite materials composed by grain interior and grain boundary affect zone. The obtained analytical results showed that: (i) the thermal conductivity increases with the increasing of grain size with respect to the electron-phonon coupling length; (ii) the thermal transport behaviours appear not only through interfacial thermal resistance but also by means of the electron-phonon coupling.

1. Introduction

Because of the small size effect, nanocrystalline (NC) materials own unique property in heat transfer compared with their coarser grain counterpart. Generally, it is known that the thermal conductivity of the NC materials decreases pronounced with the decreasing of grain size [1,2]. As the grain size is gradually comparable or smaller than the mean free path and the wavelength of the carriers (phonon and electron), some classical models and theories based on macroscale heat conduction are no longer applicable [3]. Researchers modified moderately these models and theories to analyze the heat transport of NC materials.

Roberts et al. [4] studied the thermal conductivity of NC composites composed of a matrix argon with embedded nanocrystals krypton using molecular dynamics method, their results revealed that the thermal conductivity of the composites with NC particles embedded is reduced by 25%. Dong et al. [5] established a thermal conductivity prediction model, they focused on the influence of grain size and grain boundary on the thermal conductivity of NC diamond. They pointed out that as the grain size increases, the grain boundary effect becomes weaker and weaker. We prepared NC copper and developed the Kapitza thermal resistance theoretical model [6,7]. In our model, it is no need to consider the specific transport process of heat carriers in grain interior and grain boundary, we can obtain the thermal conductivity based on the macro-performance of carriers' scattering.

In addition to the above theories, researches also studied the thermal conduction of NC materials by considering the electron-phonon (e-p) coupling effect. In the 1950s, Kaganov et al. [8] firstly conducted a study on the e-p coupling process of femtosecond laser heating films. In 1993, Qiu et al. [9] proposed a more rigorous TTM, they derived this model from the Boltzmann Transport Equation by evaluating its scattering term using quantum mechanical and statistical considerations. In 2014, Liao et al. [10] has



explored the effect of electron-phonon interaction on the lattice thermal conductivity of silicon. They found that a significant reduction of the lattice thermal conductivity at room temperature as the carrier concentration goes above 10^{19} cm^{-3} (the reduction reaches up to 45% in p-type silicon at around 10^{21} cm^{-3}).

To understand the laws of NC material thermal conductivity, in this work, a modified two-temperature model of heat conduction is used to study the effective thermal conductivity of bulk NC materials. We assume that the NC materials is a composite materials composed by grain interior(GI) and grain boundary affect zone (GBAZ), as shown in Fig. 1. In Fig. 1, GI is envisaged as perfect lattice, the shape of grain is spheres. The basis of this assumptions is that as the grain size down to nano-scale, the grain boundary fraction will become significant, as shown in Fig. 2.

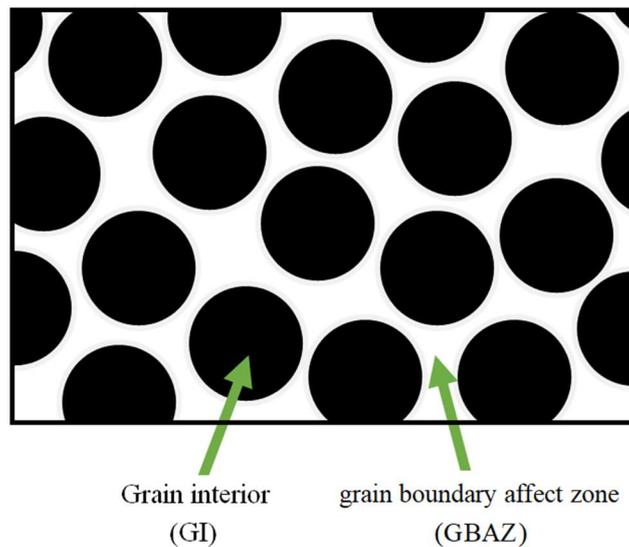


Fig 1. Schematic of the two-phase composite model of NC materials.

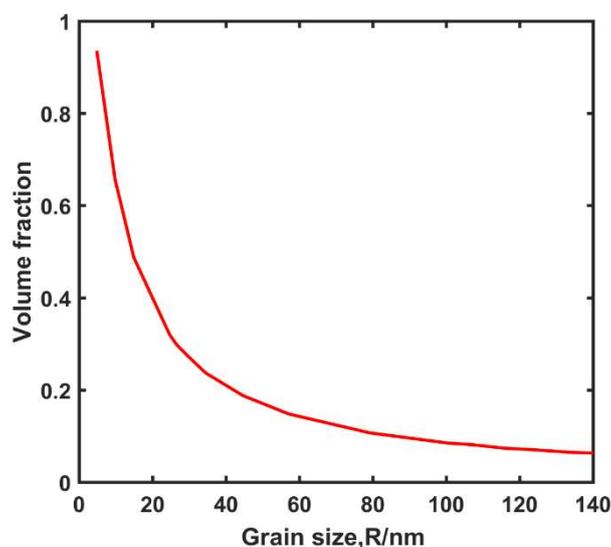


Fig 2. Evolution of volume fraction with grain size corresponding to GBAZ.

2. Theoretical Analysis Model

Under the thermal excitation of an external heat source, the energy transport process in a crystalline material occurs in two steps [11]. At the microscopic level, the electrons and phonons have different thermal energy levels, and therefore, they are not at the same temperature (in general) [12]. As shown by the TTM, the prediction of the electron temperature T_e , and the lattice temperature T_l are given by [13]

$$C_e(T_e) \frac{\partial T_e}{\partial t} = \nabla [k_e(T_e) \nabla T_e] - G(T_e - T_l) + A(z, t) \quad (1)$$

$$C_l(T_l) \frac{\partial T_l}{\partial t} = \nabla [k_l(T_l) \nabla T_l] + G(T_e - T_l) \quad (2)$$

Where C_e is the electron heat capacity, C_l is the lattice heat capacity, k_e is the electron thermal conductivity, k_l is the lattice thermal conductivity, G is the electron-phonon coupling coefficient, A is the laser source term.

Considering the nonequilibrium between the electron and phonon, Eqs. (1) And (2) are generally expressed as

$$k_e \nabla^2 T_e - G(T_e - T_p) = 0 \quad (3)$$

$$k_p \nabla^2 T_p + G(T_e - T_p) = 0 \quad (4)$$

Where T_p is the lattice or phonon temperature. Typical values of the coupling coefficient for variety of materials at room temperature are not vary much [14]. In the present work, for simplicity, we assume that the e-p coupling coefficient is an average constant, this assumption will not affect the calculating results [15].

By combining Eqs. (3) And (4), we can get

$$\nabla^2 \theta - \frac{\theta}{d^2} = 0 \quad (5)$$

$$\frac{1}{d^2} = \frac{1}{d_e^2} + \frac{1}{d_p^2} \quad (6)$$

$$d_e^2 = \frac{k_e}{G} \quad (7)$$

$$d_p^2 = \frac{k_p}{G} \quad (8)$$

Where θ is the electron and phonon temperature difference, d is the electron-phonon coupling length.

we can use the spheroidal coordinates to simplify the solution of the problem, given the azimuthal symmetry of the problem and based on the method of separation of variables, the difference of the electron and phonon temperature can be written as $\theta(\eta, \xi) = S(\eta)R(\xi)$.

We firstly consider that the uniform heat flux \vec{q}_0 is applied in the z-axis, then based on the Fourier law, the temperature $T_0 = -\frac{q_0}{k_2} z = -\frac{q_0}{2k_2} \eta \zeta$. The temperature of GBAZ T_2 can be written as

$$T_2(\eta, \xi) = AT_0(\eta, \xi)[1 + BF(\xi)] \quad (9)$$

Where $F(\xi) = iF(i\xi)$, $i_1(\)$ is the modified spherical Bessel function of the first kind and order one, and A, B are constants.

After writing out the expression for temperature distributions, Eq. (12), we now need to develop interface and boundary conditions to find the specific solutions. The boundary conditions for the temperature and heat flux continuity at the interfaces can be written by

$$k_e \frac{\partial T_e}{\partial \xi} \Big|_{\xi=\xi_1} = 0 \quad (10)$$

$$T_p \Big|_{\xi=\xi_1} = T_2 \Big|_{\xi=\xi_1} \quad (11)$$

$$k_p \frac{\partial T_p}{\partial \xi} \Big|_{\xi=\xi_1} = k_2 \frac{\partial T_2}{\partial \xi} \Big|_{\xi=\xi_1} \quad (12)$$

$$T_2 \Big|_{\xi=\xi_2} = T_0 \Big|_{\xi=\xi_2} \quad (13)$$

Where ξ_1 and ξ_2 are the inner and outer surface of GBAZ, respectively. An enormous amount of algebraic calculations can be saved during the evaluation of these boundary conditions by recognizing that the general form of the temperature profiles is dictated by the form of the external thermal excitation [16].

By substituting Eq. (15) into the three boundary conditions given by Eqs. (13-16), the temperature T_2 is expressed as

$$T_2(\eta, \xi) = T_0(\eta, \xi) \left[1 + \beta_{33} \frac{F(\xi)}{\xi_1 F'(\xi_1)} \right] \quad (14)$$

Where the prime (') indicates derivative of F , and

$$\beta_{33} = \frac{k_{33} - k_2}{k_2 + (k_{33} - k_2)L_{33}} \quad (15)$$

$$k_{33} = k_2 \frac{k_2 + L_{33}(k_1 - k_2)(1 - \nu) + \nu(k_1 - k_2)}{k_2 + L_{33}(k_1 - k_2)(1 - \nu)} \quad (16)$$

$$k_1 = \frac{k_e + k_p}{\chi} \quad (17)$$

$$\chi = 1 + \frac{k_e}{k_p} \frac{d}{R} \frac{i_1(R/d)}{i_1'(R/d)} \quad (18)$$

Where k_1 is the thermal conductivity of GI, χ is a parameter of the ratio between the grain radius R and the coupling length d defined by Eq. (7), which indicates the effect of the e-p coupling. If the size of the GI is compared to the coupling length ($R \leq d$), the coupling term χ tends to the limit $1+k_e/k_p$. If the grain size is much larger than the coupling length, the coupling term reduces to unity, which can be neglected ($d \rightarrow 0$, $G \rightarrow \infty$). ν is the volume fraction, L_{33} is the geometrical factor along the minor z-axis.

Thus, the thermal conductivity of NC materials can be expressed as

$$K = k_2 \frac{(k_e + k_p)(1 + 2\alpha) + 2\chi k_2 + 2\nu[(k_e + k_p)(1 - \alpha) - \chi k_2]}{(k_e + k_p)(1 + 2\alpha) + 2\chi k_2 - \nu[(k_e + k_p)(1 - \alpha) - \chi k_2]} \quad (19)$$

Where α is a dimensionless parameter.

3. Results and Discussion

Based on present theoretical model, we can firstly calculate the coupling parameter χ which is shown in Fig. 3. χ introduces the effect of the electron-phonon coupling. As can be seen from Fig. 3, with the increase of the relative size R/d , the coupling term χ decreases, which is consistent with the variation of thermal resistance of electrons and phonons with the relative size. As the thermal resistance gets smaller, the direct energy coupling between electrons and phonons will become larger. Fig. 3 shows that the e-p coupling term reduces to unit if the dimension of the grain is much larger than the coupling length, it is a special case, where χ can be neglected, the electrons and phonons are in equilibrium. When the relative size becomes smaller, the coupling term will gradually increase. In that condition, the coupling term χ is mainly determined by k_e/k_p . It is clear that the influence of the electron-phonon coupling is not only determined by the relative thickness of the layers, but also by the ratio of electron and phonon thermal conductivities.

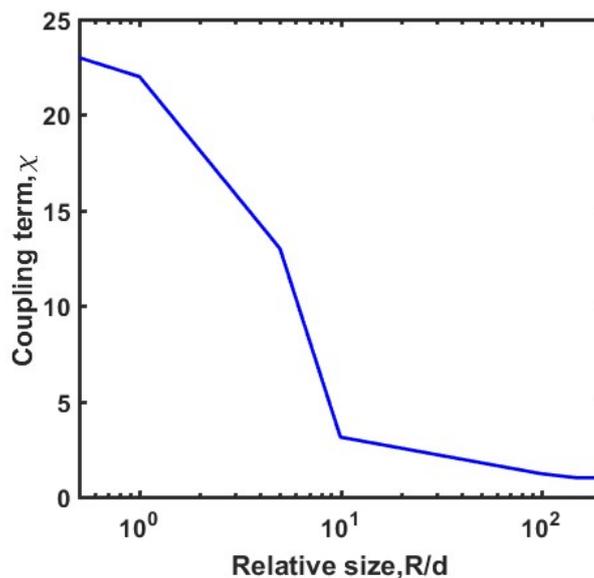


Fig 3. The normalized coupling term as a function of the relative size of grains.

As show in Fig. 4, the calculated thermal conductivity increases with the increased grain sizes, especially when the grain size is in the range of 0-200nm, the thermal conductivity decreases rapidly with the decreasing grain size. This is mainly contributed by the varying volume fraction of GBAZ. It

also can be concluded from Fig. 5 that the thermal conductivity increases with the decrease of coupling length. It is because when the coupling increases, the distance of carriers transport will become shorter and it makes the thermal transport become easier.

In order to study the effect of e-p coupling on the effective thermal conductivity, the normalized thermal conductivity K / K_2 as function of the normalized size of the grain and the e-p coupling length is calculated as shown in Fig 5. By combining the effects of the relative size on coupling term shown in Fig. 4 and normalized thermal conductivity shown in Fig. 5, we can conclude that i) the thermal conductivity is strongly influenced by the size of the grain with respect to the coupling length R/d , and the thermal conductivity increases with the decreased of e-p coupling term χ ; ii) the coupling effects are remarkable within the interval $d/R \leq 10d$ while in the other interval, the coupling effects will become weak.

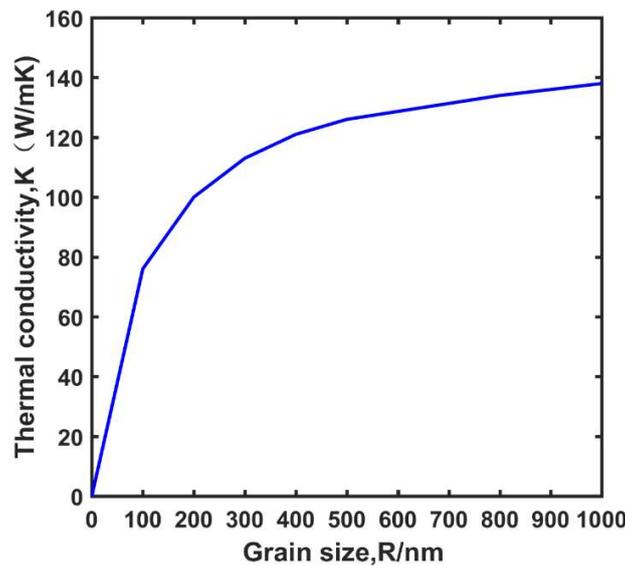


Fig 4. Thermal conductivity versus grain size.

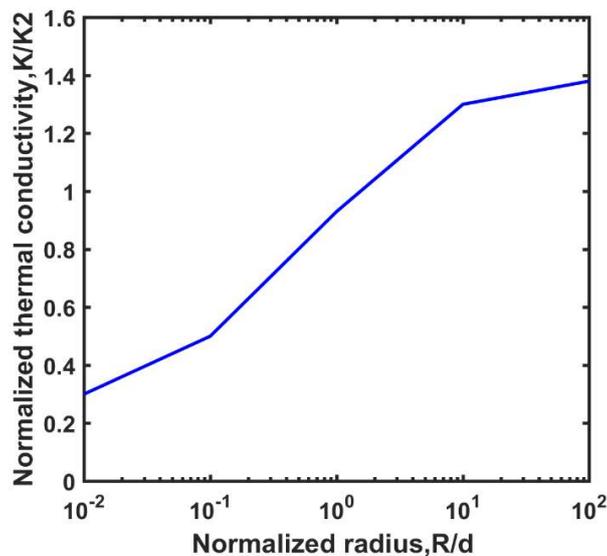


Fig 5. Normalized thermal conductivity of NC materials as a function of the normalized radius of grains.

4. Conclusion

The expression for the thermal conductivity of NC materials takes into account the electron–phonon coupling. Our calculating results indicate that (i) the effect of the e-p interaction has a simple interpretation as thermal resistance, so, the effect of the e-p coupling in NC materials leads to a reduction of thermal conductivity; (ii) the thermal conduction of NC materials is dependent on grain size, it is shows that the thermal conductivity decreases with the decreased grain size; (iii) the boundary scattering effect becomes more and more evident as the grain size decreases to 200 nm.

Acknowledgements

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