PHONON SCATTERING RATE AND THERMAL CONDUCTIVITY REDUCTION DUE TO DISLOCATION LOOPS IN ALUMINA - D.P. White (Oak Ridge National Laboratory and Merrimack College)

OBJECTIVE

The objective of this work is to calculate the phonon scattering rate (inverse relaxation time) due to dislocation loops in alumina. From this calculation an estimate of the effect of this scattering mechanism on the thermal conductivity is made.

SUMMARY

The phonon scattering rate due to dislocation loops has been calculated. The dislocation loops are modeled as plate-like inclusions in the lattice. Using the calculated value of the phonon scattering relaxation time the reduction in the lattice thermal conductivity is estimated. It is found that for characteristic values of loop size and number density in irradiated alumina dislocation loops will produce a fractional reduction in the thermal conductivity of approximately 33 percent, assuming this is the only scattering mechanism other than intrinsic scattering.

PROGRESS AND STATUS

Introduction

The thermal properties of ceramic materials used in fusion reactors change as a result of being in a radiation environment. Point defects are known to reduce the thermal conductivity and their effect on the thermal conductivity of irradiated ceramics has been calculated [1-3]. Ceramic materials are also known to develop dislocation loops as a result of irradiation. The effect of dislocation loops on the thermal conductivity of ceramics has not been as well developed in the literature as that due to point defects. Estimates of the effect in irradiated ceramics have been presented [4] but a detailed calculation of the phonon relaxation time has not been presented. A calculation of the phonon relaxation time due to dislocation loops is presented below. Using this relaxation time an estimate of the reduction in the thermal conductivity due to dislocation loops is calculated.

Theory

In order to calculate the phonon relaxation time of a dislocation loop the loop will be considered to be a plate-like assembly of atoms within the material. This plate of material will perturb the local phonon velocity and this will give rise to phonon scattering. If this scattering is strong enough then it will lead to a reduction of the thermal conductivity of the material.

First consider a single atom placed into an existing lattice. Assume that this atom occupies a volume, V_0, equal to the average atomic volume of the lattice. (This would not be true for an individual interstitial atom, however we will shortly be considering an assembly of these defects forming the plate-like inclusion and the majority of the atoms in the plate will occupy the average atomic volume). The model to be considered for this inclusion is that of a spherical inclusion in an elastic continuum.

A spherical inclusion at the origin causes a displacement, $\vec{d}(\vec{r}) = a\vec{r} + b\vec{r}/r^3$ [5-6]. This displacement causes a dilation of the lattice. It is this dilation, a part of which will cause a local variation in the phonon velocity, which gives rise to phonon scattering.

By analogy between the strain field and the electromagnetic field the dilation caused by this inclusion can be calculated [5,7,8]. The dilation is given by;

$$\vec{\nabla} \cdot \vec{d}(\vec{r}) = a\vec{\nabla} \cdot \vec{r} + b\vec{\nabla} \cdot \frac{\vec{r}}{r^3}. \tag{1}$$

The second term in the dilation is 0 for r >0 however it can be evaluated at r = 0 using the divergence theorem and Eq. 1 becomes:

$$\vec{\nabla} \cdot \vec{d}(\vec{r}) = 3a + 4\pi b\delta(\vec{r} - \vec{r}_\alpha), \tag{2}$$

which covers the case when the inclusion is at a position $\vec{r}_\alpha$ other than the origin. The first term in Eq. 3 is a uniform dilation of the lattice, this uniform dilation will not lead to a local variation of the phonon velocity and therefore does not give rise to phonon scattering. The second term will lead to phonon scattering.

The uniform dilation of the lattice is given by 3a, so:

$$3a = \frac{V_0}{GV_0} = \frac{1}{G} \tag{3}$$

where G is the number of atoms in the lattice, so a is a very small number. The magnitude of the displacement at the edge of the inclusion is:

$$r_0 = ar_0 + \frac{b}{r_0^{\,2}} \tag{4}$$

but since a is so small $r_0 \approx b / r_0^2$ and so $b \approx r_0^3$. The second term in the dilation now becomes;

$$4\pi r_0^3 \delta(\vec{r} - \vec{r}_\alpha) = 3V_0\delta(\vec{r} - \vec{r}_\alpha) = \theta_2(\vec{r}). \tag{5}$$

The inclusion site, $\vec{r}_\alpha$, is a lattice site, $\vec{x}_i$, and the lattice expression for Eq. 6 is;

$$\theta_2(\vec{x}_i) = 3\delta(\vec{x}, \vec{x}_i), \tag{6}$$

where $\delta(\vec{x}, \vec{x}_i)$ is the Kronecker delta. The change in the local phonon velocity due to this dilation is given by [9,10];

$$\delta v(\vec{x}_i) = \gamma v_0 \theta_2(\vec{x}_i) = 3\gamma v_0 \delta(\vec{x}, \vec{x}_i), \tag{7}$$

where γ is the Gruneisen constant (which typically has a value close to 1), and v_0 is the unperturbed phonon velocity. The scattering of phonons can be described in terms of a perturbation Hamiltonian of the form,

$$H' = \sum_{\vec{q},\vec{q}'} c_2(\vec{q},\vec{q}')a^*(\vec{q}')a(\vec{q}) \quad , \qquad \vec{q} \neq \vec{q}', \tag{8}$$

where $\vec{q}$ and $\vec{q}'$ are the wave vectors of the incident and scattered phonons respectively and a^* and a are phonon creation and annihilation operators respectively. If this perturbation is due to a change in the local phonon velocity the coefficient $c_2(\vec{q},\vec{q}')$ has the following form [10];

$$c_2(\vec{q},\vec{q}') = \frac{2Mv}{G} qq' \sum_{\vec{x}_i} \delta v(\vec{x}_i) e^{i(\vec{q}-\vec{q}')\cdot\vec{x}_i} (\vec{\varepsilon}\cdot\vec{\varepsilon}'), \tag{9}$$

where M is the mass of the average atom in the lattice, and $\vec{\varepsilon}$ and $\vec{\varepsilon}'$ are the phonon polarization unit vectors. The lattice sum in Eq. 10 is a sum over impurity sites. In this model of a dislocation loop the inclusions are considered to be oriented so that they form a plate of radius R and thickness h. It has been shown [11] that the phonon relaxation time for such an orientation of defects is;

$$\frac{1}{\tau(q)} = \frac{4h}{v_0} \frac{R^2 h}{v_{plat}} \left(\frac{\delta v}{v_0}\right)^2 \omega^2 A(q), \tag{10}$$

where,

$$A(q) = 4\pi \int_0^z [J_1(t)]^2 / t(1 - t^2/z^2)^{1/2} dt, \tag{11}$$

where $z = qR$ and $J_1(t)$ is the Bessel function.

Calculation, Results, and Conclusions

The scattering rate given by Eqs. 11 and 12 can now be evaluated. Assuming that the thickness of the platelet is one atomic layer, h = 2.04 X 10^{-10} m for alumina. The term, $1/v_{plat}$, in Eq. 11 is identified as the number of platelets per unit volume, n_{plat}. Irradiated alumina has been observed to have dislocation loops with an average radius of, R = 1 X 10^{-9} m, at a density of, $n_{plat} = 10^{22}$ m^{-3} [12]. The phonon velocity in alumina is, v_0 = 1 X 10^4 m/s [3]. Figure 1 is a plot of the phonon scattering rate (Eq. 11) for these conditions up to the Debye frequency of 1.31 X 10^{14} s^{-1}.

At low frequency the scattering rate is very low, the platelets are transparent to long wavelength phonons. At high frequencies, above approximately 20% of the Debye frequency, the term $A(q)$ approaches a constant value of 2π and the scattering rate is proportional to ω^2. This result is what would be expected for an infinite sheet defect, that is the platelets appear infinite to short wavelength phonons.

This last result may be used in order to estimate the effect of these defects on the thermal conductivity of alumina at high temperatures. At high temperatures Eq. 11 becomes;

$$\frac{1}{\tau(q)} = 2\pi n_{plat} \frac{4h}{v_0} R^2 h \left(\frac{\partial v}{v_0} \right)^2 \omega^2. \tag{12}$$

This expression may be substituted into the thermal conductivity integral in order to calculate the change in thermal conductivity it produces. As mentioned, the expression in Eq. 13 only applies to phonons with frequencies above 20% of the Debye frequency but since equal frequency intervals contribute equally to the thermal conductivity integral this will not produce an error of more than 20% in the calculated thermal conductivity. This approximation will overestimate the effect of platelet (dislocation loop) scattering.

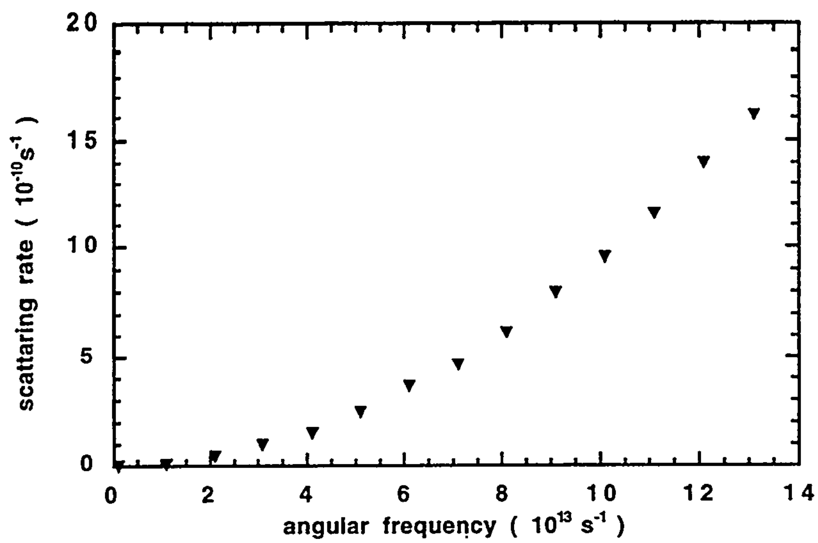

Fig. 1. A plot of the phonon scattering rate due to dislocation loops in alumina. This plot was produced by evaluating Eq. 11 for the case when $h = 2.04 \times 10^{-10}$ m, $R = 1 \times 10^{-9}$ m, and $n_{plat} = 10^{22}$ m^{-3}

The intrinsic phonon scattering rate has the form, $1/\tau_i = B\omega^2$, where $B = (2\gamma^2 K_B T)/(Mv^2\omega_D)$ and where K_B is Boltzmann's constant, M is the average atomic mass, T is the temperature, and ω_D is the Debye frequency. The platelet scattering rate in this approximation has the same form as the intrinsic scattering rate and is $1/\tau_{plat} = A\omega^2$ with A given by Eq. 13. The thermal conductivity without any defects present can be estimated by substituting the intrinsic relaxation time into the thermal conductivity integral,

$$K = \frac{1}{3}\int_0^{\omega_D} S(\omega) v^2 \tau(\omega) d\omega \tag{13}$$

where $S(\omega)$ is the lattice specific heat, it is found that $K_{unirr} = K_B\omega_D / (2\pi^2 vB)$. Similarly the combined intrinsic-loop scattering relaxation time, $1/\tau_c = (A+B)\omega^2$, may be substituted into Eq. 14 to estimate the thermal conductivity with platelet scattering. It is found that the fractional reduction in the thermal conductivity is given by;

$$\frac{K_{unirr} - K_{plat}}{K_{unirr}} = \frac{\delta K_{plat}}{K_{unirr}} \cong 1 - \left(\frac{A}{B} + 1\right)^{-1}. \tag{14}$$

For the platelet size and number density we have been considering in this paper, A = 9.4 X 10^{-18}s and at 300K B = 1.86 X 10^{-17}s. Substituting these values into Eq. 15 it is found that the expected fractional reduction in the thermal conductivity due to dislocation loops of the size and number density given above is about 33%. The actual fractional reduction in thermal conductivity attributable to dislocation loops will be less that the 33% reduction calculated above because this reduction was calculated assuming the only scattering mechanisms were intrinsic and dislocation loop scattering. For example, in the presence of strong point defect scattering, which preferentially scatters high frequency phonons, the frequency range over which dislocation loop scattering is important will be diminished and the actual fractional reduction due to dislocation loops will be similarly diminished.

This calculation shows that dislocation loop scattering can lead to a significant reduction in the thermal conductivity in alumina and must be taken into account in a discussion of the reduction of the thermal conductivity due to irradiation.

FUTURE WORK

This calculation of the effect of dislocation loops on the thermal conductivity of alumina has not considered the effect of the scattering due to the strain field of the dislocation line surrounding the loop. Although is has been suggested that this scattering is less important that that due to the stacking fault [13] it should be estimated and compared to the results of this paper.

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