Surface induced phonon decay rates in thin film nano-structures

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Abstract. Nano-scale structure significantly impacts phonon transport and related phonon relaxation rates, with order of magnitude effects on the thermal conductivity of dielectric thin films and quantum wires, and even larger effects on the lifetimes of ultrasonic phonons of micro- (nano-) oscillators. In both cases, efforts to explain the data have been hampered by our lack of knowledge of the effects of confined dimensionality on phonon-phonon scattering rates. Using a phonon Boltzmann equation with appropriate boundary conditions on the free surfaces to take surface roughness into account, we have obtained an expression yielding phonon lifetimes in 2-D dielectric nanostructures (thin films) resulting from phonon-phonon scattering in conjunction with phonon-surface scattering. We present these theoretical results and, in the limit in which surface induced losses dominate, obtain explicit predictions for the phonon lifetimes. The predicted temperature dependence of the ultrasonic loss does not explain the observed saturation of the loss at low temperatures ($\tau(T) \to \text{const}$), but does give results of the order of magnitude of measured ultrasonic lifetimes.

1. Introduction
Determining the effect of nano-scale structure on phonon scattering and transport phenomena has been of great interest recently because of the order of magnitude scale dependence of the thermal conductivity that has been observed in dielectric thin films and nano-wires[1–4]. This empirical observation suggests that it may be possible to engineer the phonon transport properties of dielectric nano-structures through a judicious choice of materials, geometry, and surface preparation, provided we develop a knowledge of the phonon scattering mechanisms.

Recent theoretical approaches to predict the thermal conductivity have been successful[5, 6] in the regime in which discretization effects due to phonon confinement can be neglected. As is typical in phonon transport predictions however, these models use a tunable parameter, in this case the surface specularity $p$, a measure of the phonon-surface scattering, to fit the data and hence lack predictive power. The comparison of theory to experiment does nevertheless indicate that surface scattering is playing a large role in these structures.

Ultrasonic decay rates $Q^{-1}$ of nano-scale structures are thought to be similarly sensitive to phonon-surface scattering[7, 8] based on observations of a strong empirical relationship of $Q^{-1}$ to surface to volume ratios. Ultrasonic measurements could hence potentially be used in conjunction with thermal conductivity measurements to further our understanding of phonon scattering processes in nano-structures. Unfortunately, however, the corresponding theoretical developments in the context of ultrasonic attenuation have not yet been reported, and such an approach cannot be carried out.
We have taken the first steps in this direction. We have extended the Akheiser-like theoretical approach employed by Ehrenreich and Woodruff[9] and Maris[10, 11] for elastic waves in bulk media to elastic waves in nano-scale thin films. In the limit in which internal phonon scattering processes can be neglected, we obtain results showing that the ultrasonic decay rate $Q^{-1}$ indeed depends on the specularity $p$ in a similar fashion as that found for the thermal conductivity. Assuming a temperature independent Gruneisen parameter, we have found the ultrasonic decay rate due to this mechanism to vary as $T^4$ for the low temperature range in which such an approximation would be valid. Extrapolating our results to higher temperatures yields internal friction estimates of the order of magnitude of those observed.

2. Theoretical approach

Consider the vibration of a free standing film of thickness $a$ as shown in Fig. 1. Our aim in this section is to obtain an expression for the attenuation of the ultrasonic modes in the film, the imaginary part of the frequency shift resulting from their interaction with thermal phonons. The surface enters this calculation in the interaction of the thermal phonons with the surface.

Figure 1. Geometry of a free standing film.

Following Maris[11], we write the total potential energy per unit volume of the crystal semiclassically as,

$$\Phi = C + C_{\alpha\beta} \epsilon_{\alpha\beta} + \frac{1}{2} C_{\alpha\beta\gamma\delta} \epsilon_{\alpha\beta} \epsilon_{\gamma\delta} + \frac{1}{V} \sum_{kj} (N(kj) + 1/2) \hbar \omega(kj) + \ldots$$

(1)

where $V$ is the film volume, $\epsilon_{\alpha\beta}(x) = 1/2(\partial_{\alpha} u_{\beta}(x) + \partial_{\beta} u_{\alpha}(x))$ is the Lagrangian strain field, $u_{\alpha}(x)$ is the classical displacement field, $C_{\alpha\beta}(C_{\alpha\beta\gamma\delta})$ are elastic constants, and $\omega(kj)$ and $N(kj; x)$ are the frequency and number of phonons with wavenumber $k$ and polarization $j$ at position $x$. Imposing the condition that the stress $\sigma_{\alpha\beta} = \partial \Phi / \partial \epsilon_{\alpha\beta} = 0$ in the equilibrium, zero strain state at temperature $T$ yields to leading order in the strain

$$\Phi = \frac{1}{2} C_{\alpha\beta\gamma\delta} \epsilon_{\alpha\beta} \epsilon_{\gamma\delta} - \frac{1}{V} \sum_{kj} \Delta N(kj) \hbar \omega_0(kj) \gamma_{\alpha\beta}(kj) \epsilon_{\alpha\beta}.$$  

(2)

Here, $\Delta N(k\lambda) = N(k\lambda) - n(k\lambda)$ is the difference between $N(k\lambda)$ and the Bose distribution $n(k\lambda) = (\exp(\beta \hbar \omega(k\lambda)) - 1)^{-1}$, $\gamma_{\alpha\beta}(kj) = -(\partial \omega(kj) / \partial \epsilon_{\alpha\beta}) / \omega(kj)$ is the Gruneisen tensor, and $\beta = 1/k_b T$. We have now dropped the strain independent contributions to the potential energy; the thermal phonons contribute to the potential energy only through the strain dependence of the phonon frequencies, $\hbar \omega(kj) \approx \hbar \omega_0(kj)(1 - \gamma_{\alpha\beta}(kj) \epsilon_{\alpha\beta})$. We have also ignored the second order term in the expansion of the resonance frequencies because this quantity leads only to shifts in the ultrasonic frequencies as opposed to attenuation, our focus here.

The action for the ultrasonic modes is thus given approximately by

$$S = \int dt dV \left\{ 1/2(\rho \dot{u}^2 + u_i \Phi^0_{ik} u_j) + \Phi^{int} \right\}.$$  

(3)

where the operator $\Phi^0_{ik} = \partial_i c_{ijkl} \partial_l$ is obtained by integrating the first term in the potential energy by parts, and the interaction term is the phonon contribution. The operator $\Phi^0$ is easily
A term, this poses difficulties, but we can make progress if we assume this term to be negligible. The eigenfunctions solve the eigenvalue-eigenfunction equation $\delta S = \Omega_0(k\lambda) A_{k\lambda}(t)$ and satisfy the orthogonality conditions $\int dV \psi_{k\lambda}(x) \cdot \psi_{k'\lambda}(x) = (2\pi)^2 \delta^2(k-k') \delta_{\lambda\lambda'}$.

Using the expansion $u(t) = \sum A_{k\lambda}(t) \psi_{k\lambda}(x)$, and changing variables to $A_{k\lambda}$ and $A_{k\lambda}^*$, we find $S = S_0 + S_{int}$ where

$$S_0 = \int dt \sum_\lambda \int \frac{d^2k}{(2\pi)^2} \frac{1}{2} \rho \left( |\dot{A}_{k\lambda}(t)|^2 - \Omega_0^2(k\lambda) |A_{k\lambda}(t)|^2 \right)$$

(4)

and

$$S_{int} = -\int dt \sum_\lambda \int \frac{d^2q}{(2\pi)^2} \frac{1}{2V} \sum_{k_j} \left\{ \Delta N_{\alpha\beta}(k; q) h\omega(k_j) \gamma_{\alpha\beta}(k_j) A_{q\lambda}^* + c.c. \right\}. \quad (5)$$

The semi-classical equations of motion for the ultrasonic modes are obtained by setting $\delta S = 0$ with respect to variations in $A_{k\lambda}(t)$ and $A_{k\lambda}^*(t)$. To leading order, this yields the equation $\ddot{A}_{k\lambda}(t) + \Omega_0^2(k\lambda) A_{k\lambda}(t) = 0$ which gives the Lamb mode solutions[12] for the ultrasonic waves in the film. It is straightforward to demonstrate to leading order in perturbation theory,

$$\Omega(q\lambda) = \Omega_0(q\lambda) + \frac{1}{2\rho V \Omega_0(q\lambda) A_{k\lambda}(\Omega)} \sum_{k_j} \Delta N_{\alpha\beta}(k; q) \gamma_{\alpha\beta}(k_j) h\omega(k_j). \quad (6)$$

Eq. (6), a generalization of Eq. 54 of Reference [11], is the desired expression.

### 3. Results with no internal phonon scattering

The non-equilibrium phonon number distribution $\Delta N$ must be determined in order to evaluate the attenuation of the ultrasonic waves in the film. We employ the Boltzmann equation (BE) to describe the dynamics of the thermal phonons. Maris gives

$$\frac{\partial N}{\partial t} = -\omega^\alpha_{\alpha\beta} \frac{\partial N}{\partial k_\gamma} - v_\alpha \frac{\partial N}{\partial x_\gamma} + \left( \frac{\partial N}{\partial t} \right)_{coll}$$

(7)

where the term $(\partial N/\partial t)_{coll}$ is the phonon scattering term. The first term on the right hand side is an inhomogeneous source term resulting from the imposed strain field. Prior work[9,11] assumes $\Delta N$ to be proportional to the strain field in the medium. This assumption fails here because the strain field has gradients of the order of the inverse thickness of the film, while the mean free path is typically greater than the thickness of the film. In the presence of the collision term, this poses difficulties, but we can make progress if we assume this term to be negligible.

Taking the collision term to vanish, we Fourier transform the BE in the in-plane directions $xy$ and time to obtain an ordinary differential equation in $z$. We then impose the Ziman boundary conditions $\Delta N(k; q, z = \pm a/2) = p\Delta N(-k; q, z = \pm a/2)$ employed recently by a number of investigators[5,6] in the context of the thermal conductivity predictions. The boundary condition contains the following physics: A fraction $p$ of the incoming thermal phonons are specularly reflected from the surface, while a fraction $(1-p)$ are thermalized by interaction with the surface - an inelastic process which should produce damping of the ultrasonic waves.

For the lowest order Lamb mode, the flexural mode, the strain field is simply proportional to $z$ and the resulting equation for the non-equilibrium phonon distribution is straightforward to
solve. Substituting the expression for $\Delta N$ so obtained into Eq. (6), we obtain the loss factor $Q^{-1} = \text{Im} (\Delta \Omega(q)/\Omega)$ of the flexural mode,

$$Q^{-1} \approx \frac{\pi^3 \ln 2}{30(12)^{1/2} \rho c_j^2} \left(\frac{k_p a}{\bar{h}}\right)^4 \frac{1-p}{1+p} \left\langle \gamma_F^2 \right\rangle$$

(8)

where $k_p^2 = E/\rho(1-\sigma^2)$ is the plate wave speed, $c_j$ is the speed of the thermal phonons (j denoting polarization), $\sigma$ is Poisson’s ratio, and $\gamma_F = \gamma_{11} - \frac{\sigma}{1-\sigma}\gamma_{33}$ is a combination of Gruneisen parameters appropriate to flexural waves. The averaging brackets denote an angular average.

4. Discussion and outlook

Equation (8) gives the loss due to the following decay process: ultrasonic vibration $\rightarrow$ thermal vibration $\rightarrow$ surface absorption. The loss vanishes to leading order for $p = 1$, specular surfaces, but the frequency shift, a result not given here, does not vanish in this limit, and hence the thermal and ultrasonic vibration are not uncoupled, but instead beat against one another. It is interesting to note that the loss depends on the parameters of the film only through the geometrical factor $k_p a$ and is otherwise a universal function of temperature. This leads to a scale independence of the loss, a result which is consistent with our intuitive sense that the loss per cycle depends only on the number of boundary scattering events per cycle and the energy lost per scattering event, quantities which are scale independent in the absence of internal loss.

A detailed evaluation of this expression is difficult to achieve because of unknown elastic constants appearing in the effective Gruneisen parameter $\gamma_F$. Nevertheless, some qualitative observations can be made: 1) The loss due to the surface is proportional to the ratio $(1-p)/(1+p)$, the same dependence on $p$ as is found in the thermal conductivity predictions[5, 6]; 2) The approximation is valid in the low temperature range for which $\Omega \tau \gg 1$; 3) The $T^4$ dependence is inconsistent with the observed saturation($Q^{-1} \to \text{const}$ as $T \to 0$) of the absorption at low temperatures in current measurements; 4) The predictions for $10 < T < 100$, universal predictions as noted above, are in the range $10^{-8} < Q^{-1} < 10^{-4}$, of the order of magnitude of experimental results[13, 14] obtained in this regime. Obtaining predictions at higher temperatures to compare with experiment appears to be a promising avenue of research.

Acknowledgements

This research was supported by the US Office of Naval Research.

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