An Attempt to Observe Phonon Dimensionality Crossover Effects in the Inelastic Scattering Rate of Thin Free-Standing Aluminum Films

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We compare the inelastic scattering rate in 220-Å-thick free-standing films with nearly identical films on substrates. The scattering rate was determined by fitting the magnetoresistance to theories of quantum transport. Although the films are sufficiently thin so as to modify the three-dimensional spectrum of thermal phonons, we find no significant difference between the scattering rates in free-standing and supported films. While experiments on other materials down to lower temperatures are necessary, we conclude that the cubic temperature dependence of the inelastic scattering rate in these thin aluminum films is not strongly affected by the phonon dimensionality.

1. INTRODUCTION

The past decade has seen a great deal of activity on low-dimensional electronic systems.[1,2] There are at least two reasons why the analogous problems for phonons have not been explored thoroughly or systematically. First, a suitable sample must be constructed to isolate the microstructure under study from phonons of the adjoining substrate while still permitting the sample to be cooled. Second, a suitable parameter of the sample sensitive to the dimensionality of the phonons must be identified and measured.

We have developed a technique[3] to fabricate free-standing, or self-supported, metallic films of thickness >200 Å together with a contiguous, simultaneously evaporated structure on a substrate. A typical sample consists of two identical films: one on a bulk silicon wafer substrate, and the other on a nearby thinned region of the same wafer. The thinned region is subsequently

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completely removed by exposing the backside of the wafer to a reactive plasma. This process does not appreciably degrade the electrical properties of the free-standing film with respect to the supported film.[3] Consequently, the properties of the free-standing film can be compared directly with an otherwise identical control (the supported film). A related technique[4] has been used by others to prepare free-standing structures. However, their technique causes far more damage to the film, and the data are difficult to interpret.

Traditional probes of the phonon properties include the specific heat and thermal conductivity in high quality insulators at low temperatures.[5] These measurements are difficult to carry out in sub-μm-scale structures since the heat capacity and thermal conductivity are small. The electron-phonon scattering rate, τ_{ep}^{-1}, should be the dominant temperature dependent contribution to the inelastic scattering rate, τ_{in}^{-1}, in aluminum thin films at liquid helium temperatures. The temperature dependence of τ_{ep}^{-1} should reflect the modification of the phonon spectrum due to finite size effects. Consequently, measurements of τ_{in}^{-1} should provide a viable technique to probe phonon properties. Quantum transport measurements, in particular the low-field magnetoresistance, is a proven technique[1,2] for determining the inelastic scattering rate, τ_{in}^{-1}. The technique does not require highly disordered metal films to adequately resolve the temperature dependence of τ_{in}^{-1}. Also, low measuring currents are used, and the carriers are not significantly perturbed from their equilibrium thermal distribution.

We have chosen aluminum in this study since it is easy to deposit, pattern, and render free-standing. Also, its low-temperature transport properties have been extensively studied by others.[6,7] For temperatures well above the superconducting transition temperature, these experiments reveal that the inelastic rate, τ_{in}^{-1}, varies as $AT + BT^3$. The linear term was associated with the electron-electron scattering rate and the cubic term with the electron-phonon rate. These studies spanned a range of sample dimensions and disorder but consistently yielded a T^p term for the electron-phonon component of the inelastic rate with $p = 3$, the anticipated value for clean-limit *bulk* systems. This value of p was attributed to the strong coupling between phonons in the film with those emanating from the supporting (bulk) substrate. Studies on much more disordered and typically thinner films of other materials yield p around two, which has been tentatively attributed to a reduced phonon dimensionality.[8,9]

In this paper, we restrict our attention to aluminum thin films which are relatively clean. Given the well documented T^3 power-law component of τ_{in}^{-1} for these films on substrates, we initially thought that a comparison of τ_{in}^{-1} in free-standing and supported films would provide a simple and

appealing approach to revealing phonon dimensionality effects. In particular, the T^3 dependence of τ_{in}^{-1} at higher temperatures was expected to crossover to a T^2 dependence at lower temperatures for the free-standing samples. However, as we shall show, the anticipated dimensionality crossover signature is not observed. This suggests that the T^3 power law for τ_{in}^{-1} in aluminum has a physical origin different from three-dimensional electron-phonon scattering. This conclusion is different from that found in the literature.[6,7] We are confident of our results because we have examined τ_{in}^{-1} in identical supported and free-standing films whose thickness was small enough to ensure that the thermal phonon excitation spectrum is modified from that of the bulk.

2. DESCRIPTION OF THE EXPERIMENT

The speed of sound, c_s, in aluminum ranges from 3040 m/sec (transverse mode) to 6420 m/sec (longitudinal mode).[10] This implies that the quantization of the phonon wavevector in the direction perpendicular to the film's plane will have an energy (in Kelvin) given by $\hbar c_s \pi / k_B d$, where d is the film thickness. For the case of our film thickness of 220 Å, this leads to an energy splitting of 7 K for longitudinal phonons and 3.3 K for transverse phonons. Thus, one would expect that the phonon spectrum should be largely two-dimensional (2D) below ~3 K. These estimates are rather crude, but are consistent with a more detailed numerical calculation to be discussed later.

For the present experimental study, the nominal film thickness was measured to be 220 Å. Operationally, this is close to the limit that we can reliably fabricate free-standing films. A lift-off process was used to define a lateral film dimension of 5 μm. The film on the thinned region of the substrate was then made free-standing by reactive-ion etching the backside of the substrate.[3] The resulting sample is illustrated in Fig. 1, which shows both the free-standing and the contiguous supported structures. A meandering current path is used to provide a higher resistance sample to increase the accuracy of the measurements. The finger-like projections provide mechanical support for the free-standing structures. They are replicated on the supported structures for consistency and serve no other function there. Unless otherwise indicated, a measuring current of 1.0 μA was used, a level which produces no significant self-heating. Thermometry was achieved by monitoring the resistance of a calibrated germanium sensor. Temperature stability is typically better than 0.5 mK.

The etching process is non-invasive,[3] resulting in similar electrical properties for the supported and free-standing structures. The sheet resistance, R_s, at 4 K is 1.61 Ω (1.98 Ω); the residual resistance ratio is 1.85 (1.78); the
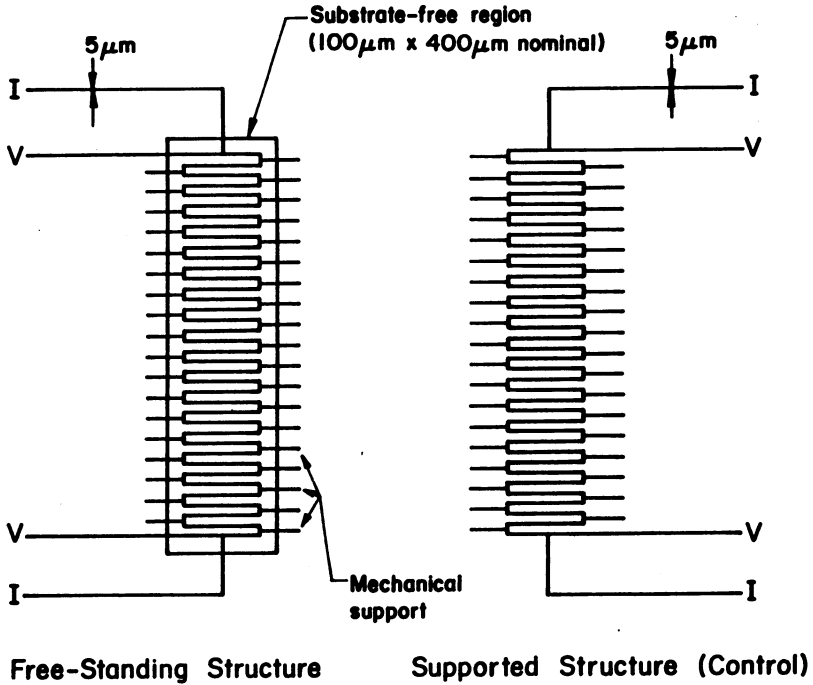
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Fig. 1. Schematic of a typical sample, showing the free-standing film together with the contiguous supported film (the control). Film thickness is nominally 200 Å.

diffusion constant, D, as determined[11] from the temperature dependence of the upper critical field near the transition temperature T_c, is 29.2 cm^2/s (24.0 cm^2/s); and the T_c is 1.462 K (1.364 K). Here, the values in the parentheses are those for the free-standing structure.

3. EXPERIMENTAL RESULTS

The low-field magnetoresistance (MR) of both films were measured for 1.4 K $< T <$ 6.5 K and for -100 Oe $< H <$ 400 Oe. Typical data, normalized to the classical Drude resistance, R_0, of the film, are shown in Fig. 2 for $T = $ 5.3 K. The MR, $(R(H) - R_0)/R_0$, of a 2D thin film at low temperature is due to three quantum corrections:[1,2] weak localization, superconducting fluctuations (both Aslamazov–Larkin and Maki–Thompson terms), and the electron-electron interaction in the particle-particle channel. The last can be ignored at fields smaller than $\pi c k_B T/2eD$, which in the case of our films is approximately 550 Oe at 1 K.[7] In our measurements, the applied field is $<$400 Oe throughout the entire temperature range studied, and we ignore
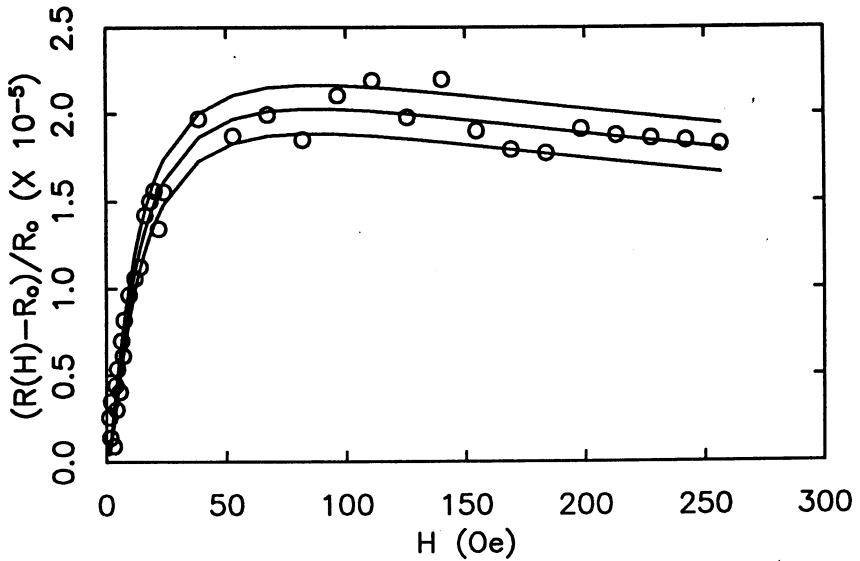
Fig. 2. The magnetoresistance (MR) versus applied magnetic field H at $T = 5.2$ K. Data are symmetric about $H = 0$. The raw data are normalized by the Drude resistance R_0 and fitted to theory (solid lines). The upper and lower lines correspond to $\pm 9\%$ of the value of τ_{in}^{-1} obtained from the middle line (the least-square fit).

this contribution. The determination of $\tau_{in}^{-1}(T)$ by fitting MR to theoretical models is a well-established procedure. Following previous workers,[1,6,7] we have used a two-parameter fit at higher temperatures to obtain the spin-orbit scattering rate, τ_{so}^{-1}. This rate is temperature insensitive and is used in a one-parameter fit to obtain τ_{in}^{-1} at all other temperatures. We found $\tau_{so}^{-1} = 1 \times 10^9$ s^{-1} to be appropriate for both free-standing and supported samples. (Varying the choice of τ_{so}^{-1} would, in general, alter the magnitude of τ_{in}^{-1} but *not* its temperature dependence.) The result of this analysis for both the supported in free-standing films is shown in Fig. 3. Other samples measured show similar behavior.

It is immediately evident that the rates for both films have essentially the *same* temperature dependence in the entire temperature range investigated other than the narrow region near T_c. The noticeable deviation at the lowest temperatures is a consequence of the different T_c's of the two films, resulting in different contributions from superconducting fluctuations. For T somewhat above T_c, the electron-phonon scattering is the dominant contribution to τ_{in}^{-1}. For $T > T_c$, we find that τ_{in}^{-1} follows the relation $AT + BT^3$, where $A = 2 \times 10^7$ s^{-1} K^{-1} and $B = 1.4 \times 10^7$ s^{-1} K^{-3}. Following earlier workers, we associate these contributions with 2D electron-electron and 3D electron-phonon scattering processes respectively.
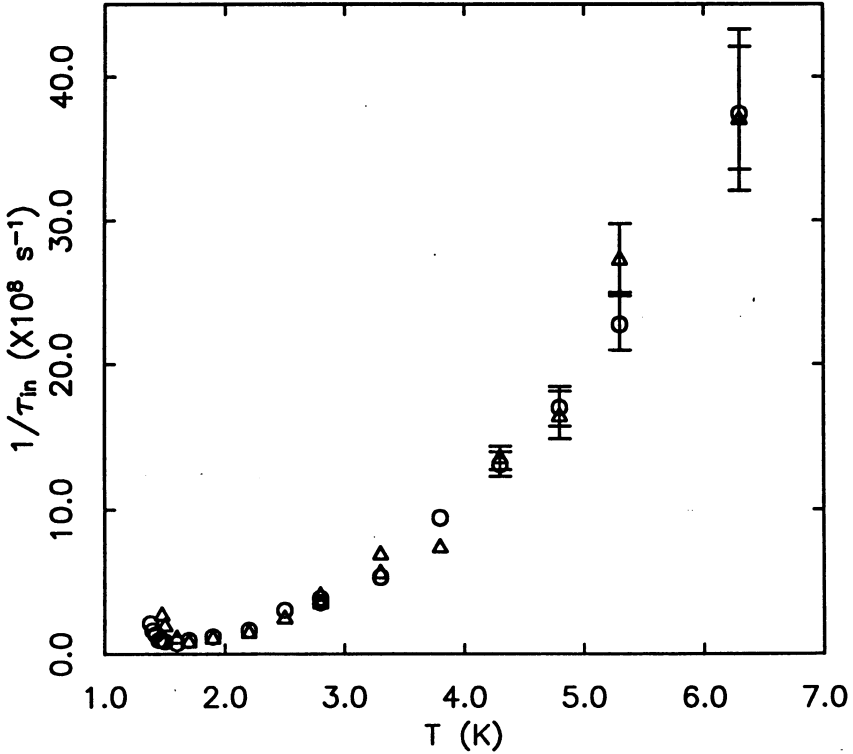
Fig. 3. The inelastic rate τ_{in}^{-1} versus T for the free-standing film (O) and the supported film ($\triangle$).

The power-law behavior can be more clearly seen in Fig. 4 where we plot τ_{in}^{-1}/T against T^2. The slope of the straight line in Fig. 4 yields the parameter B, which is ~0.9 of the theoretical prediction.[12] Our value of A is on the order (~0.2) of the theoretical prediction for electron-electron interaction in the 2D dirty limit.[2] For temperatures corresponding to the minimum τ_{in}^{-1} where electron-electron scattering and fluctuation effects dominate, the inelastic length $l_{in} = (D\tau_{in})^{1/2}$ approaches the film width of 5 μm. This may have rendered the 2D theory less accurate. However, the contribution from electron-electron scattering to τ_{in}^{-1} is relatively small in the temperature range of interest and should not affect our conclusion regarding the electron-phonon rate. Within experimental scatter, τ_{in}^{-1} for both the supported and free-standing films can be fitted to the same values of A and B. Therefore, provided the T^3 power law is a manifestation of bulk phonon dimensionality in the electron-phonon scattering rate, the expected crossover in the phonon dimensionality at ~3 K is not observed in these films.
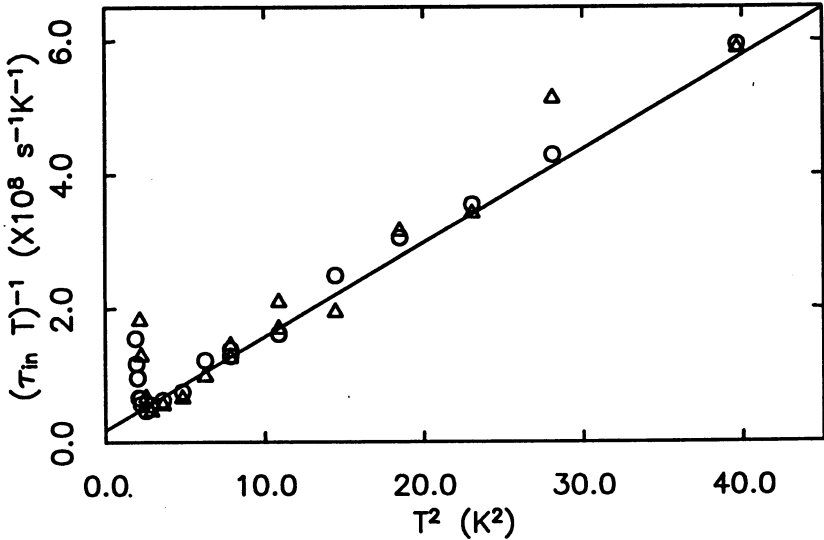
Fig. 4. τ_{in}^{-1}/T versus T^2, clearly showing the same T^3 contribution to τ_{in}^{-1} for both the free-standing film (O) and the supported film ($\triangle$). The y intercept and the slope of the solid line yields the parameters A and B respectively (see text). The low temperature increase in τ_{in}^{-1}/T is due to fluctuation effects.

4. CALCULATION OF FINITE SIZE EFFECTS IN THE PHONON SPECTRUM

In order to carry out an accurate comparison between data and theory, we have to consider details of the phonons' thermal excitation spectrum in addition to the simple dimensional estimate described in Sec. 2. Suppose the free-standing thin film has thickness d_z and lateral dimensions d_x and d_y such that $d_z \ll d_x, d_y$. Provided that the film thickness is comparable to the wavelength of the dominant thermal phonons, λ_T, where $\lambda_T = hc_s/k_BT$, the planes of the reciprocal lattice in the k_z direction are separated by energies significant when compared to the thermal energy. The reciprocal lattice of the thin film is illustrated schematically in Fig. 5. Due to the small film thickness, the wavevector k_z perpendicular to the film takes on discrete values k_{zn}, where $k_{zn} = n(\pi/d_z)$; $n = 0, 1, 2, \ldots$. For a plane with $k_z = k_{zn}$, the density of states, $F(k_p)$, is linear in k_p, where k_p is the projection of the wavevector (k_x, k_y, k_{zn}) onto the k_z-plane. (Thus, $k = [k_p^2 + k_{zn}^2]^{1/2}$, with $k_p = [k_x^2 + k_y^2]^{1/2}$.)

The electron-phonon scattering rate due to a *single plane* and for a particular mode (transverse or longitudinal) is given by

$$\tau_{ep}^{-1} \propto \int_0^\infty \frac{\alpha^2 F(k_p) \, dk_p}{\sinh(\beta \hbar c_s k)} \tag{1}$$
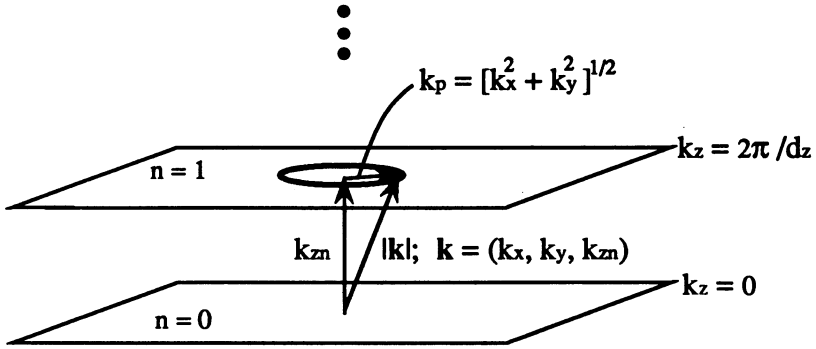
Fig. 5. Schematic representation of the reciprocal lattice of a defect-free thin film. Note that only discrete values of k_z are allowed due to quantization. Only the first two planes in the k_z direction are depicted.

where $\beta = 1/k_B T$. The $(\sinh)^{-1}$ factor accounts for the number of available phonon and electron states which participate in electron-phonon scattering events. The product $\alpha^2 F$ is the Eliashberg function which, in 2D systems, is proportional to k.[13] Following Allen,[14] we can qualitatively understand this dependence by decomposing the spectral function, $\alpha^2 F$, into the density of phonon states, F, and the "coupling" part α^2, which characterizes the strength of the electron-phonon coupling. Providing the function α is not strongly k dependent, we can rewrite $\alpha^2 F \propto k$ since the phonon density of states is linear in k. For the $n=0$ plane, $k_{zn}=0$ and $k=k_p$; eq. (1) is the same as that derived by Belitz and Das Sarma.[9]

Thus, we can rewrite eq. (1) for each plane

$$\tau_{ep}^{-1} \propto \int_0^\infty \frac{k_p \, dk_p}{\sinh(\beta \hbar c_s k)} = \int_{k_{zn}}^\infty \frac{k \, dk}{\sinh(\beta \hbar c_s k)} \tag{2}$$

Changing the variable of integration to ε, where $\varepsilon = \hbar \omega = \hbar c_s k$, we can express the contribution from all planes as a single summation:

$$\tau_{ep}^{-1} \propto \sum_n \int_{\hbar c_s k_{zn}}^\infty \frac{\varepsilon \, d\varepsilon}{\sinh(\varepsilon/k_B T)}$$

This expression can be rewritten in a more convenient form for the purposes of numerical evaluation:

$$\tau_{ep}^{-1}(T) \propto \sum_n \int_0^\infty H\left(\varepsilon - n \frac{\pi \hbar c_s}{d_z}\right) \frac{\varepsilon \, d\varepsilon}{\sinh(\varepsilon/k_B T)} \tag{3}$$

where H is the Heaviside step-function. This is the expression with which we evaluate the temperature dependence of τ_{ep}^{-1}.
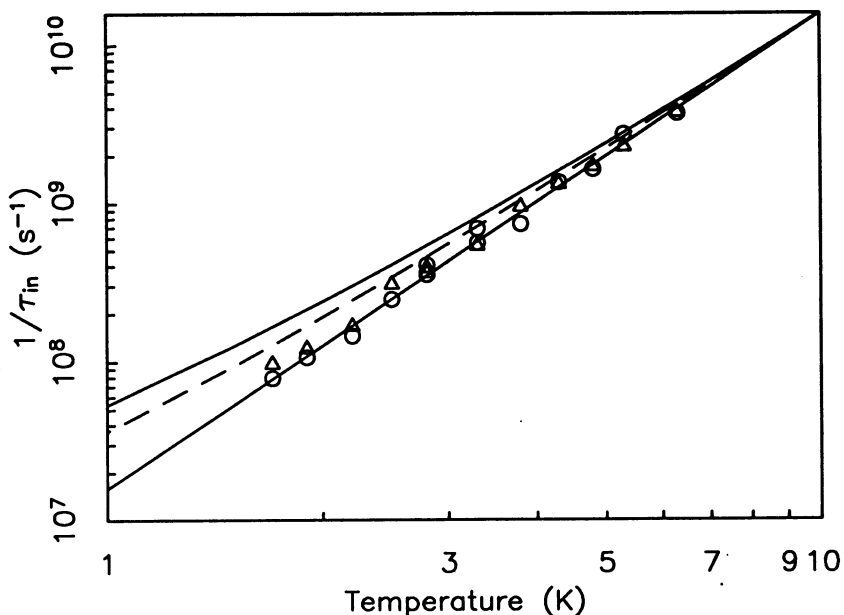
Fig. 6. We plot the scattering rates measured for supported ($\triangle$) and free-standing ($\bigcirc$) films, together with the results of the numerical calculation. We omit results below 1.7 K (which are dominated by fluctuations) for clarity. The upper solid line is the result for τ_{in}^{-1} for longitudinal scattering alone, while the result considering both longitudinal and transverse contributions is shown as the dashed line. Both numerical results are set equal to the T^3 fit to the data of Fig. 4 (lower solid line) at 10 K for purposes of comparison. Both sets of data closely follow the T^3 dependence.

In Fig. 6, we compare the results of the calculation for both longitudinal phonons alone as well as the case for a combination of longitudinal and transverse phonons (with the latter weighted for two modes). The results indicate a clear deviation from the T^3-dependence of τ_{in}^{-1} at a temperature below ~5 K. Given experimental scatter, the differences between the measured results and the numerical calculation become significant below 3 K. The T^2 dependence does not develop fully until temperatures below 1.5 K.

5. DISCUSSION

A critical examination of the results illustrated in Fig. 6 reveals that while neither sets of data deviate from the T^3 dependence, the disagreement between the numerical calculation and the experiment is statistically significant below ~3 K. Unfortunately, the superconductivity of the aluminum films and the effects of fluctuations preclude us from making measurements

at temperatures lower than ~1.7 K in this system. Nevertheless, the results should exhibit a significant deviation from the bulk-like T^3 behavior in the experimentally accessible temperature range.

The apparent insensitivity of $\tau_{in}^{-1}(T)$ to the absence of the substrate raises a number of questions. It is unlikely that the dimensionality of the phonon density of states, $F(\varepsilon)$, is not reduced in the free-standing films. The explanation for the behavior of our free-standing and supported films must therefore lie elsewhere.

A factor of possible importance is the relatively large grain size of thermally evaporated aluminum films. Transmission electron microscopy indicates that our films have typical grain size of ~100 Å. This is smaller than, but comparable to, the estimated wavelength of thermal phonons in the film at 5 K. It is unclear what effect, if any, this may have on phonon dimensionality or, more generally, on $\alpha^2 F(\varepsilon)$. In general, the role of disorder in thin films is to increase the power law of the inelastic scattering rate to T.[4,15] However, there is clearly no evidence for this behavior in our data.

Throughout this work, we have attributed the T^3 contribution in the τ_{in}^{-1} of our films to 3D electron-phonon scattering. This is an interpretation adopted by previous investigators of aluminum thin films on substrates. In doing the same, we are forced to conclude that the electron-phonon scattering rate appears to be much less sensitive to the dimensionality of the phonon density of states than expected. Experiments on other thin film systems (typically much more disordered than the films discussed in this paper) do *not*, as a rule, yield a T^3 electron-phonon term in the scattering rates. The typical, sub-T^3 dependences in diverse systems[8] have frequently been attributed to reduced phonon dimensionality or alternatively to $\alpha^2 F$ being linear in ε. We believe that such a view may be problematic.

Removal of the substrate, together with the ability to compare nearly identical samples, allows us to assert that the phonon spectrum must be modified. We note that the coefficient for the T^3 term, $1.4 \times 10^7 \text{ s}^{-1} \text{ K}^{-3}$, is nearly identical to that determined by Santhanam *et al.*[7] In view of our results which demonstrate that the magnetoresistance shows no significant differences between the free-standing and supported films, there is strong evidence that the modification of the thermal phonon excitation spectrum due to the finite size of the metallic samples does not influence the T^3 coefficient or power law in the inelastic scattering rate.

It is widely believed that the cubic temperature dependence of τ_{in}^{-1} in supported aluminum films is a consequence of the scattering of a 2D electron gas by 3D phonons emanating from the substrate. This view is now adopted by theorists and experimentalists in order to explain the body of quantum transport data. Since the phonon spectrum must be modified by finite size effects, an alternate explanation of our data requires that the T^3 rate is *not*

a manifestation of a 3D phonon spectrum in aluminum films. This would imply that the power law behavior of $\tau_{in}^{-1}(T)$ is a property intrinsic to the films and depends only weakly (if at all) on the phonon dimensionality. This conclusion is consistent with the results of a recent experiment (of very different design) on copper films.[16] This related work reveals no difference in the electron-phonon scattering rate between free-standing and supported films, of thickness ranging from 100 Å to 1000 Å, over a temperature range between 0.5 K and 10 K. In view of this data, it would be instructive to examine free-standing films of other materials over a wider temperature range. This could be done in equilibrium (as in this work), or by measuring the hot-electron properties as explored by DiTusa *et al.*[16]

6. CONCLUSIONS

We have performed a systematic investigation of the inelastic scattering rates in free-standing and nearly identical supported structures. This investigation was carried out in the regime for which a phonon dimensionality crossover signature was expected. The absence of such a signature is both surprising and intriguing. The absence of the effect of dimensionality of the thermal phonons on τ_{in}^{-1}, particularly in the free-standing films, may indicate that the T^3 component is associated with processes intrinsic to the film and not to bulk-like electron-phonon scattering. Satisfactory resolution of this problem will necessarily require a more detailed theoretical understanding on electron-phonon processes, as well as experimental work on other free-standing systems. The role of disorder must also be further explored.

ACKNOWLEDGMENTS

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