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Nanoscale heat conduction across tunnel junctions

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Nanoscale heat conduction across tunnel junctions prepared through natural oxidation of metal electrodes is experimentally studied. The thermal resistance of AlOx tunnel barriers increases linearly with barrier thickness, which is consistent with the prevailing theory of heat conduction in highly disordered materials. Heat conduction across tunnel junctions is strongly impeded by finite thermal resistance at interfaces between barrier and electrode layers, which can be exploited to create superior thermal barrier coatings. The thermal conductivity of nanolaminates consisting of a series of Ta/TaOx tunnel junctions is determined to be well below the minimum thermal conductivity limit. © 2005 American Institute of Physics. [DOI: 10.1063/1.1931827]

Understanding nanoscale thermal phenomena is important in optimizing or enabling a variety of nanotechnology applications employing tunnel junctions. Reliable operation of a magnetic tunnel junction is compromised by degradation of thin tunnel barriers under simultaneous electrical and thermal stresses.[1] Nanolaminates consisting of a series of tunnel junctions are also promising as thermal barrier coatings and components of thermoelectric devices because heat conduction is strongly impeded by the thermal interface resistance.[2,3] Nanolaminates are attractive as thermal protection coatings for gas turbine blades, machining tool bits, and gun barrels since they do not incorporate pores or microscale defects that compromise mechanical or chemical protection properties. They are also well suited as thermal barrier layers in nanoscale electronic and data storage devices[2] that have stringent roughness requirements. Heat conduction across tunnel junctions is also of great fundamental interest. The type of dominant heat carriers switches between phonons and electrons across the metal–barrier interface. The situation is very different from semiconductor superlattices and quantum wells where phonons are dominant heat carriers throughout the entire thickness.

The present letter reports an experimental study of heat conduction across tunnel junctions and nanolaminates consisting of a series of multiple tunnel junctions. We measure the thermal resistance of aluminum oxide tunnel barriers and TaOx tunnel junctions that are synthesized through repeated deposition and oxidation of thin metal films. Our data show that the thermal interface resistance between a tunnel barrier and electrodes is significant but is smaller than most previously reported values for metal–dielectric interfaces. The effective thermal conductivity of Ta/TaOx nanolaminates as small as 0.35 W/m K is demonstrated at room temperature. This is well below the minimum thermal conductivity limit and among the smallest values obtained for nonporous inorganic solids.

During the past two decades a number of studies reported measurements of heat conduction properties of thin dielectric films and interfaces between metallic and dielectric materials.[4–6] In the present work, we employ electrical techniques that use a lithographically patterned metal stripe as a heater and electrical resistance thermometer.[7,8] We apply either dc or harmonic current to the heater and monitor resulting changes in its electrical resistance, which can be precisely calibrated as a function of temperature.

We prepare tunnel junctions using multitarget magnetron sputtering systems with a base pressure of $\sim 1 \times 10^{-8}$ Torr. One of the systems has been used extensively to synthesize magnetic tunnel junctions for data storage applications. Further discussion of the deposition system and properties of tunnel barriers produced were described elsewhere[9] and will not be repeated here. We prepare two different types of samples, one incorporating AlOx barriers of different thicknesses and the other consisting of multiple Ta/TaOx junctions, on prime quality silicon wafers.

The first set of samples contains 20 nm Ta/AlOx barrier/20 nm Ta. The AlOx barriers are produced by fully oxidizing 1-nm-thick Al films in 1 Torr O_2 for 15 min without breaking vacuum. Transmission electron microscopy (TEM) studies[10] showed that the thickness of naturally oxidized AlOx barriers is approximately 1.3 times that of starting Al layers. The Al deposition and oxidation step are repeated multiple times to obtain thicker barriers.

In the second set of samples alternating steps of Ta layer deposition and oxidation are repeated to yield nanolaminates of Ta/TaOx. The thickness of the Ta layer before oxidation is varied from 2 to 10 nm. Part of the Ta layers is converted into TaOx during oxidation. In calculating the laminate thermal conductivity, we use the measured total thickness of the nanolaminates, which ranges from 12 to 105 nm depending on the initial Ta thickness and the number of repetitions. A SiOx insulation layer and an aluminum layer are subsequently deposited on each nanolaminate sample. The aluminum layer is patterned lithographically to create metal stripe heaters with various widths $(1-40 \ \mu m)$ and lengths $(250-1500 \ \mu m)$.

In studying heat conduction across nanoscale films and interfaces, accurate determination of the substrate and other parasitic thermal resistance in the sample is critical. Analytic

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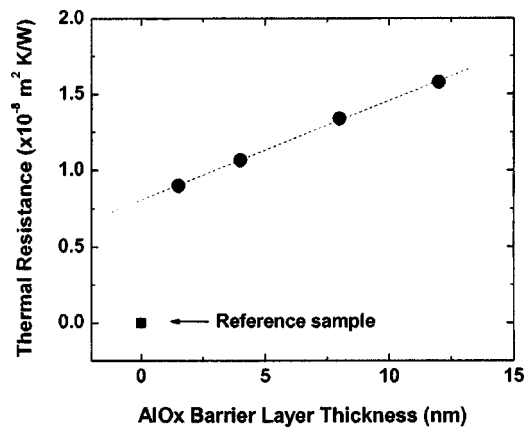a)Author to whom correspondence should be addressed; electronic mail: just@seas.ucla.edu

FIG. 1. Thermal resistance of AlOx tunnel barriers as a function of thickness.
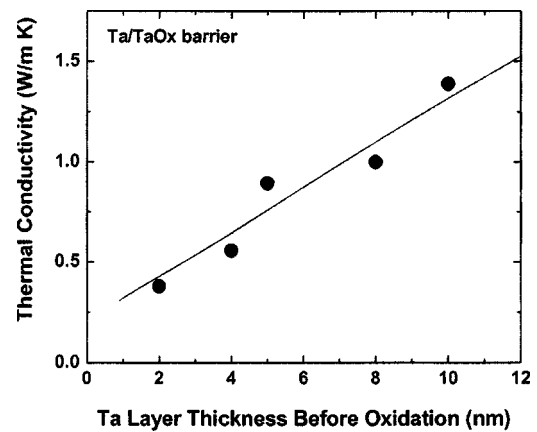

FIG. 2. Effective thermal conductivity of Ta/TaOx tunnel junction nano-laminates as a function of the Ta layer thickness before oxidation. The solid line corresponds to the model prediction that takes into account electron–phonon nonequilibrium in the metal layers as discussed in the main text.

heat conduction models are often used to predict substrate thermal resistance. Although these models yield very useful physical insights, they invoke various approximations whose validity requires careful examination for each situation. We perform measurements on dedicated reference samples to independently determine the substrate and other parasitic thermal resistance. The differential measurement approach was demonstrated for the characterization of sputtered AlOx films.[8] By comparing data obtained from heaters of different dimensions with numerical simulation results, we check the consistency in our data reduction procedure.

Figure 1 shows the thermal resistance of the AlOx tunnel barriers as a function of thickness. The substrate and other parasitic thermal resistances have already been subtracted from the data. The reference sample is identical to the tunnel junction samples except that it does not have an AlOx barrier. The thermal resistance of the AlOx tunnel barriers increases approximately linearly with thickness. This is consistent with the prevailing theory of heat conduction in amorphous materials where strong structural disorder is believed to limit the effective mean free path of heat carriers to the order of interatomic spacing. The thermal conductivity of the AlOx tunnel barriers is estimated to be 1.5 W/m K, which is comparable to that of sputtered AlOx films.[8]

By extrapolating the linear fit to zero barrier thickness, we estimate the average thermal interface resistance between the barrier and Ta electrode layers to be approximately 3.5 $\times 10^{-9}$ m^2 K/W. This is greater than the thermal boundary resistance of TiN films epitaxially grown on crystalline MgO substrates.[11] There is some question, however, about the accuracy of the TiN data since analysis of the thermal resistance of SiOx films reported in the same study yields negative thermal interface resistance. The present value is smaller than most other reported values for metal–dielectric interfaces, which often deviated considerably from predictions of the diffuse mismatch theory.[6] The presence of defects or contamination layers near the interfaces has often been cited as a possible source of discrepancy. Interfaces in the present tunnel barrier samples are formed on freshly deposited metal films in a controlled vacuum environment. The thermal resistance obtained here is indeed similar to that of interfaces found in AlOx–W nanolaminates[3] and ZnS–SiOx nanocomposites synthesized in an ultrahigh vacuum chamber.[12]

Our data, however, should not be considered as direct evidence of the possible impact of interface defects or con-

tamination layers. The thermal interface resistance values obtained for epitaxially grown TiN–MgO and TiN–Al$_2$O$_3$ interfaces were similar, for example, even though the latter is expected to possess significant defects due to large lattice mismatch.[11] We also would like to point out the considerable challenges involved in accurate measurements of thermal interface resistance. Data reduction procedures can introduce significant bias errors.

Figure 2 shows the effective thermal conductivity of Ta–TaOx tunnel junction nanolaminates. The thermal conductivity decreases monotonically with decreasing Ta thickness and hence increasing interface density. This is consistent with the fact that the interface resistance dominates the total thermal resistance. The out-of-plane thermal conductivity of the present nanolaminates is as small as 0.35 W/m K, which is smaller than that of W–AlOx nanolaminates,[3] ZnS:SiO nanocomposite films commonly used in phase change recording media,[12] and ZrO and related films widely used as thermal barrier coatings for gas turbine blades.[13] Controlled oxidation or nitridation of thin metal films is a promising approach to pushing the limits of interface density in nanolaminate-based thermal barriers.

Recently we proposed a model for the thermal resistance of a metal nanostructure surrounded by a dielectric medium.[14] When energy transfer across a metal–insulator interface is mediated only by phonons, it introduces size-dependent extra thermal resistance resulting from nonequilibrium between electrons and phonons in the metal. For a planar metal film sandwiched between two insulating films, the continuum two-fluid heat conduction equations are solved to express the thermal resistance of the metal film R''_{metal} as

$$R''_{\text{metal}} = \frac{L}{k_{\text{ph}} + k_e} + 2\left(\frac{k_e}{k_{\text{ph}}}\right)\left(\frac{\delta}{k_{\text{ph}} + k_e}\right)\left(\frac{e^{L/\delta} - 1}{e^{L/\delta} + 1}\right). \quad (1)$$

Here, L is its thickness, k_{ph} and k_e are the phonon and electron thermal conductivity, respectively, and δ is the electron–phonon coupling distance given as $[k_e k_{\text{ph}}/G(k_e + k_{\text{ph}})]^{1/2}$. The first term of Eq. (1) is the thermal resistance as predicted from the conventional continuum theory. The second term is the extra resistance due to electron–phonon spatial nonequilibrium. The electron–phonon coupling constant G can be determined using ultrashort pulse pump–probe experiments

and typical values for transition metals are of the order of 10^{17}–10^{18} W/m^3 K.[15] The corresponding electron-phonon coupling distance δ is of the order of one nanometer.

The total thermal resistance of the present nanolaminates is the sum of the thermal resistance of the Ta layers as given in Eq. (1), the thermal resistance of the TaOx layers, and the intrinsic thermal interface resistance between the Ta and TaOx layers. The intrinsic thermal interface resistance is expected to be primarily a result of mismatch in atomic vibrational properties as assumed in the diffuse mismatch model. The solid line in Fig. 2 corresponds to the model prediction with the following parameters: $k_e = 10$ W/m K, $k_{\text{ph}} = 1$ W/m K for the Ta and barrier layer, and $\delta = 1$ nm. The electronic thermal conductivity is estimated from the measured electrical resistivity data but the other parameters should be viewed only as first order estimates. The intrinsic thermal interface resistance of 2×10^{-9} m^2 K/W, which is comparable to prediction of the diffuse mismatch theory, yields a good fit to the present data.

In summary, we experimentally study nanoscale heat conduction across tunnel junctions. Substrate heating and other parasitic thermal resistance in the samples are determined independently using a reference sample to ensure accurate measurements of the thermal resistance of nanoscale thin films. The thermal resistance of AlOx tunnel barriers increases linearly with barrier thickness, providing evidence that nanoscale heat conduction in highly disordered materials can be described using the continuum theory. The significant thermal resistance at the barrier and electrode interface suggests a promising approach to realizing superior thermal barriers. We report the effective thermal conductivity of Ta/TaOx nanolaminates as small as 0.35 W/m K at room temperature, which is well below the minimum thermal conductivity limit and among the smallest values obtained for nonporous inorganic solids.

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