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IN-PLANE THERMAL CONDUCTIVITY OF ULTRA-THIN Al_2O_3 FILMS MEASURED BY MICRO-RAMAN

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In the past decades, heat transport in thin film structures has been extensively studied due to its importance in performance optimization of micro/nano-scale devices. A large amount of heat is generated during the operation of electronic devices and must be dissipated efficiently via dielectric electrical insulating films, thus it is crucial to investigate and engineer the thermal properties of thin film structures. Numerous methods have been developed to measure thin film thermal conductivity, many of which took advantage of electrical heating and temperature sensing such as the 3ω method [1], scanning thermal microscopy (SThM) [2], etc. These methods require the heaters/sensors to directly contact the samples surface, inducing the risk to bias the temperature field; also, the contacting components add a significant complexity to the originally simple 2-D film structure, which increases the difficulty of modeling and data analysis.

Because of the disadvantages of contacting methods, non-contact measurement techniques are desirable to study thin film thermal properties. Recently micro-Raman technique emerged in thermal conductivity research of nanoscale materials [3]. A laser beam is irradiated on the sample, inducing Raman scattering and heating effect. Temperature information is obtained by measuring Raman peak shift due to bond softening at high temperature. In these works, the film thickness needs to be one order larger than the laser spot size, otherwise one has to consider interface and substrate contribution in heat transport, resulting in increasing difficulty in modeling sub-micrometer thin film thermal properties. In this work, we use micro-Raman technique to measure the in-plane thermal conductivity of suspended ultra-thin Al_2O_3 film. Due to the small overall thickness (less than 70 nm) and a large area (50 μm square) of the film stack, heat propagation is in-plane dominated and can be neglected in the cross-plane direction, thus eliminating the influence of film interfaces. Using a simple modeling process we were able to isolate the in-plane thermal conductivity of Al_2O_3 film.

The samples we used are aluminum oxide films deposited by the atomic layer deposition (ALD) method on suspended 20 nm thick, 50 μm × 50 μm silicon dioxide films, and we are interested in measuring the in-plane thermal conductivity of the aluminum oxide film. On top of the aluminum oxide layer, a 20 nm thick bismuth film was coated using vacuum thermal evaporation as Raman temperature sensor. Bismuth has two characteristic Raman peaks at 70 cm^{-1} and 97 cm^{-1}, the latter of which has a clear linear dependence of its peak position and sample temperature due to bond softening. By analyzing the peak position shift, the local temperature at laser focus spot can be simultaneously measured during laser heating process. To calibrate this linear relationship between peak position and sample temperature, we coated a 40 nm bismuth film on bulk glass, put this calibration sample into a heating stage and collected Raman spectra under a series of different temperature settings, which were measured by a platinum resistor.

Calibration result is shown in Figure 1. Raman peak shifts to blue-side with a nearly linear relation of 0.02521 cm^{-1}/K. It is worth noting that the strain induced by Bi-glass or Al_2O_3 interfaces is likely to shift the Raman peak position by a constant number and barely affect the temperature dependence of Raman peak position, as another Bi-Si calibration sample gave almost the same peak shift rate of 0.02528 cm^{-1}/K while the intercept changed by 0.3 cm^{-1}.

During the experiment, a 633 nm HeNe laser was focused by a 50x objective on the center of the sample area to heat the sample. This configuration suggests a radial symmetric temperature distribution as well as a uniform temperature in the z direction since the thickness of the film structure is much less than the radial dimension. The same laser was used as Raman excitation source as well, and the Raman spectrum of bismuth was collected using a HORIBA LabRam HR800 Raman spectroscopy system. By changing the laser power using a variable ND filter, we recorded the change of Raman peak position, which could be converted to the change of local sample temperature at the laser focus spot. This temperature vs. laser power relationship is predicted to be linear by the radial heat transfer model; by fitting the slope, the thermal conductivity of Al_2O_3 film can be extracted.

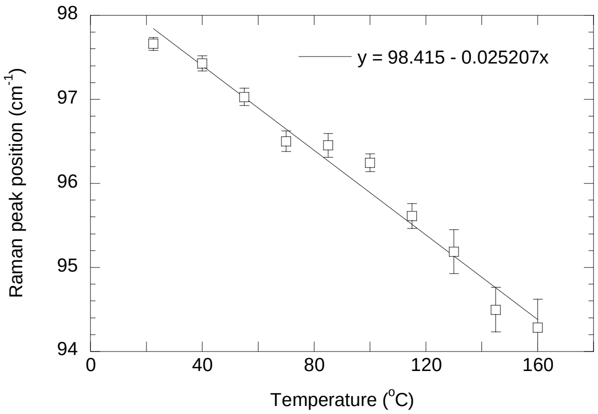

FIGURE 1. Temperature dependence of bismuth Raman peak position. Solid line shows a linear fit.

To solve the radial energy equation for our thin-film laser heating problem

$$\frac{1}{r}\frac{d}{dr}\left(kr\frac{dT}{dr}\right)+\dot{q}=0 \tag{1}$$

one needs to carefully model the heat source distribution and assign proper boundary conditions. The heat source term is attributed to the absorbed laser power in bismuth film, which decays exponentially along z direction and spreads as a Gaussian function along lateral direction. A power meter was placed underneath the sample to measure the optical transmissivity T; at the entrance of the Raman microscope, a beam splitter isolated part of the laser reflected by the sample, by measuring which and using a metallic mirror as reference we obtained the reflectivity R of the sample. Hence, with the total laser power P measured and laser beam profile taken as Gaussian shape, the heat source term can be expressed as product of a Gaussian function of r and an exponential function of z, which is

$$\dot{q}=\alpha(1-R)\frac{2P}{\pi r_0^2}\exp\left(-\frac{2r^2}{r_0^2}\right)\exp(-\alpha z) \tag{2}$$

where α is the optical absorption coefficient of bismuth film determined by transmissivity and reflectivity, P is the total laser incident power and r_0 is the radius of the laser focus spot. Due to the large aspect ratio of sample radius to thickness, heat transfer inside the film can be considered as one-dimensional along radial direction (in-plane direction) only, and the heat source term can be approximated as uniformly distributed along z direction, which gives

$$\dot{q}=\frac{1-R-T}{t}\frac{2P}{\pi r_0^2}\exp\left(-\frac{2r^2}{r_0^2}\right) \tag{3}$$

where t is the total thickness of the sample film. Since surface convective heat transfer is negligible according to our two-dimensional numerical calculation, the radial energy equation becomes

$$\frac{1}{r}\frac{d}{dr}\left(kr\frac{dT}{dr}\right)+\frac{1-R-T}{t}\frac{2P}{\pi r_0^2}\exp\left(-\frac{2r^2}{r_0^2}\right)=0 \tag{4}$$

Here, k stands for equivalent thermal conductivity of the triple-film structure in radial direction

$$k=k_{eq}=\frac{1}{t}\sum_{i=1}^{3}k_i t_i \tag{5}$$

where k_i and t_i is the thermal conductivity and film thickness of each layer respectively. For the boundary conditions, 1) at r=0, heat flux equals zero due to symmetry; 2) T (r = r_b = 50 μm) = T_0 is assumed at the far end of the calculated domain, since the supporting silicon frame has a much higher thermal conductivity than the film stack thus acting like a heat sink that immediately lower the temperature at the film edge down to the ambient level. Apply these boundary conditions to the previous solution and we obtain

$$T(r)=T_0+\frac{(1-R-T)P}{2\pi kt}\left\{\left[\frac{1}{2}Ei\left(-2\frac{r^2}{r_0^2}\right)-\ln\frac{r}{r_0}\right]-\left[\frac{1}{2}Ei\left(-2\frac{r_b^2}{r_0^2}\right)-\ln\frac{r_b^2}{r_0}\right]\right\} \tag{6}$$

where $Ei(x)$ is the exponential integral. The temperature measured by Raman laser beam is the Gaussian-weighted average temperature,

$$T_{Raman}=\frac{\int_0^\infty T(r)\exp\left(-\frac{2r^2}{r_0^2}\right)rdr}{\int_0^\infty \exp\left(-\frac{2r^2}{r_0^2}\right)rdr} \tag{7}$$

It can be shown that the Raman-measured temperature at laser focus spot rises linearly with laser power, and the rate of temperature rise is a function of equivalent thermal conductivity of the sample film stack.

We assumed SiO_2 thermal conductivity to be 1.4 W/mK, Bi thermal conductivity to be 12 W/mK and accordingly fit laser focused spot radius to be 2.68 μm. The Al_2O_3 in-plane thermal conductivity results are shown in Figure 2. The thermal conductivity of Al_2O_3 films are generally below 3 W/mK, consistent with the result from Stark et al [4]. This value is well below that of Al_2O_3 crystals. Kinetic theory suggests that solid bulk thermal conductivity can be expressed as

$$k=\frac{1}{3}C_v vl \tag{8}$$

where C_v is the volumetric specific heat, v is the phonon propagation speed, and l is the phonon mean free path. For a highly disordered dielectric material such as amorphous Al_2O_3, phonons are scattered frequently and the phonon mean free path is equal to the separation distance between atoms [5,6], much smaller than the phonon mean free path in crystalline Al_2O_3, resulting in the “minimum thermal conductivity” for amorphous dielectric materials.

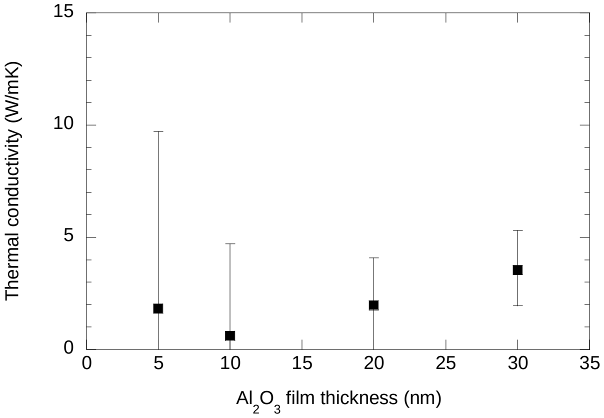

FIGURE 2. Thermal conductivity of Al_2O_3 films of 5 nm, 10 nm, 20 nm and 30 nm thicknesses.

However, due to relatively large error bars, the trend of film thickness dependence of in-plane thermal conductivity due to phonon boundary scattering is not clear. Three factors are considered to be responsible for the large measurement uncertainty. 1) Temperature resolution of Raman thermometry. Generally, for a Raman thermometry system, the temperature measurement deviation can vary between 5 and 20 K [7]. The Raman spectral resolution of our equipment is 0.27 cm^{-1} which gives a peak fitting uncertainty ranging from 0.07~0.26 cm^{-1} and correspond temperature uncertainty of 3~10 K, depending on the Raman spectrum contrast. 2) Radial heat flow route. In our experiments, bismuth was chosen as the Raman sensor film coated on Al_2O_3-SiO_2 films, both of which have much lower thermal conductivity (~1 W/mK) than bismuth (~10 W/mK). As a result, most of the heat flow occurs in the bismuth film rather than Al_2O_3 film, which is unfavorable since it makes heat transfer insensitive to Al_2O_3 thermal conductivity. 3) Laser absorptivity characterization. During the experiments, bismuth film may oxidize and degrade at high temperature. It was noticed that the transmissivity of the sample could increase, for example, from 49% to 52% within 30 seconds for the 10 nm Al_2O_3 sample at 47μW laser power.

In summary, we presented a method to measure in-plane thermal conductivity of dielectric thin films using micro-Raman technique. Al_2O_3 films with different thicknesses were studied, and the thermal conductivity results were consistent with previously reported experiments and theories. Despite the relatively large errors, there are several ways that can be implemented to increase the measurement accuracy. Micro-Raman thermometry, as a non-contact optical method, could be promising in characterizing thermal properties of thin films.

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