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Thermal characterization of anisotropic thin dielectric films using harmonic Joule heating

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Abstract

A technique for thermal characterization of anisotropic dielectric films is developed. The technique is applicable to a film which has thickness of the order of 1 μm and is on a substrate with high thermal conductivity. Metal lines with various widths are deposited on the film using standard IC fabrication processes and are subjected to harmonic Joule heating. Monitoring the resulting amplitude of temperature oscillations in the metal lines allows the determination of the in-plane and out-of-plane thermal conductivity. The present work performs thermometry using the electrical method known as the 3ω technique and also the thermoreflectance technique. Measurement results are reported for polyimide films on silicon substrates.

Keywords: Thermal conductivity; Anisotropy

1. Introduction

Many thin films of scientific and technological importance were found to exhibit anisotropy in their thermal conductivity. In addition to the intrinsic mechanisms present in bulk materials, the observed anisotropy is caused by grain [1] or molecular structures [2,3] unique to thin films, and by the size effect associated with boundary or interface scatterings of heat carriers [4]. Anisotropy also exists for a composite thin film structure consisting of layers with different thermal conductivities.

A variety of techniques have been developed to determine the thermal conductivity of thin films, especially that in the out-of-plane direction. Reviews of these techniques are available in the literature [5,6]. In-plane thermal conductivity has received less attention. Previously reported experimental studies required free-standing films or films placed on thermally-insulating substrates [4,7–9]. These approaches have the advantage that temperature gradient can be formed in the direction essentially perpendicular to that of heat flow, allowing independent measurements of the in-plane thermal conductivity. However, they often require demanding processing steps, such as back-etching of a substrate or grafting of a film grown originally on substrates with high thermal conductivity. In addition, for techniques employing a laser as a heat source, interactions of a beam with a film need to be carefully tailored or modeled [10,11]. Non-negligible radiation heat loss for free-standing films also requires special attention [12]. Photothermal methods for the measurement of anisotropic thermal conductivity have also been developed [13], which induce localized and modulated optical heating and map the resulting temperature oscillations to extract the thermal conductivity tensor. For thin films deposited on substrates with much higher thermal conductivity, however, the requirement of high spatial resolution renders the photothermal approach especially difficult to apply. In view of these difficulties, it is desirable to develop techniques that are applicable to films supported by substrates and can complement the previous approaches.

The present manuscript reports a technique for studying the anisotropic thermal conductivity of dielectric films or composite structures, which have thickness of the order of 1 μm and are grown on substrates with high thermal conductivity. The technique employs metal lines with varying widths, which are microfabricated through standard IC processes, and does not require special processing steps. The metal line is subjected to harmonic Joule heating and serves as both a heater and a thermometer. Due to substantial heat conduction through the substrate and the small surface area of the heat affected region, the importance of radiation loss is minimized [14]. As a demonstration, measurements are performed and reported here for polyimide films.

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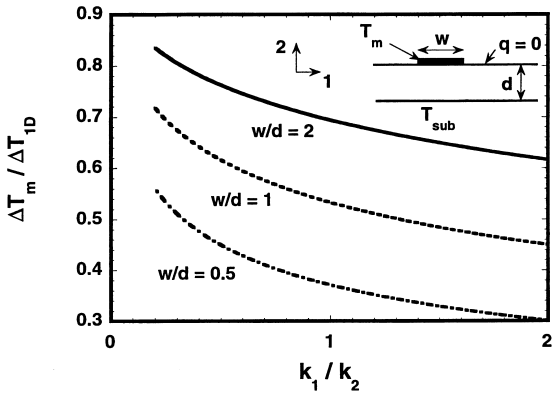

Fig. 1. Steady-state temperature rise ΔT_m calculated as a function of the ratio of the in-plane and out-of-plane thermal conductivities. The boundary conditions used in conjunction with the heat diffusion equation are shown as an inset.

2. Measurement principle

When the surface of a thin film is heated over a finite region, lateral spreading of heat inside the film modifies the temperature field from the one that would exist for a strictly one-dimensional case. The lateral heat spreading is governed by the thickness and thermal conductivity of the film and also by the dimensions of the heat source, which is a microfabricated metal line in the present study. By comparing temperature rises in metal lines with different widths, the anisotropic thermal conductivity can be deduced.

The two-dimensional heat diffusion equation for an anisotropic film can be written as [15]

$$k_{11}\frac{\partial^2 T}{\partial x_1^2} + k_{22}\frac{\partial^2 T}{\partial x_2^2} = C\frac{\partial T}{\partial t} \tag{1}$$

Here the ks are the components of the thermal conductivity tensor and C is the heat capacity per unit volume. The thermal conductivity components are assumed to be constant. The dimension of the heat source in the x_3-direction is assumed to be sufficiently large to neglect any variation of temperature fields along the direction. The contribution from the off-diagonal components, $k_{12} + k_{21}$, is assumed to be absent. This is true when one of the principal axes of the conductivity tensor is perpendicular to the film plane defined by x_1 and x_3 axes. One example is a $YBa_2Cu_3O_7$ film on a MgO substrate, where the c-axis of the film is perpendicular to the film plane. Eq. (1) is also a good approximation when the term $\partial^2 T/\partial x_1 \partial x_2$ is negligible, which is the case, for example, in silicon-on-insulator structures. For films exhibiting the size effect due to the boundary or interface scatterings of heat carriers, the diffusion equation is not strictly valid and should, instead, be interpreted as describing an effective phenomenon.

To provide further physical insight an analytic solution to Eq. (1) is obtained under simplifying assumptions. Consider the case where the conductivity of a substrate is much larger than that of a film, so that the variation in the substrate temperature at the interface between the film and the substrate, T_{sub}, is negligible. Assume similarly that the region covered by the metal line is isothermal at a temperature $T_m = \Delta T_m + T_{sub}$ and the rest of the upper surface of the film is adiabatic (Fig. 1). The validity of these approximations depends on the thickness and thermal conductivity of both the metal line and the film of interest. A previous study [16] derived a series solution to a similar problem by applying the constant surface heat flux boundary condition at the region occupied by the heating line.

Under the steady state condition, a relation between the heat generation rate per unit metal line length Q' and the temperature rise can be obtained by solving Eq. (1) for the thin film with the zero right hand side. Eq.(1) is first transformed through a change of variables according to

$$X_1 = \left(\frac{k}{k_1}\right)^{1/2} x_1 \tag{2}$$

$$X_2 = \left(\frac{k}{k_2}\right)^{1/2} x_2 \tag{3}$$

where $k = (k_1 k_2)^{1/2}$ is chosen to achieve conformal mapping. The resulting heat diffusion equation under the steady-state condition becomes

$$\frac{\partial^2 T}{\partial X_1^2} + \frac{\partial^2 T}{\partial X_2^2} = 0 \tag{4}$$

The solution to the above Laplace equation with the assumed boundary conditions can be found by means of the Schwarz–Christoffel transformation [17]. The temperature rise satisfies the following relationship

$$\frac{\Delta T_m}{\Delta T_{1D}} = \left(\frac{k_2}{k_1}\right)^{1/2} \frac{w}{d}\frac{K}{2K'} \tag{5}$$

where ΔT_{1D} is the temperature rise that would result in the absence of lateral heat spreading and is equal to $Q'd/wk_2$. The argument κ of the complete elliptical integral of the first kind K and its complementary counterpart K' is given as

$$\frac{1}{\kappa} = \cosh\left[\frac{\pi}{4}\frac{w}{d}\left(\frac{k_2}{k_1}\right)^{1/2}\right] \tag{6}$$

Here w and d are the width of the metal line and the thickness of the film, respectively. Note that Eq. (5) depends on $(w/d)(k_2/k_1)^{1/2}$ and not on their individual values. Anisotropy in thermal conductivity has the net effect of modifying the geometric ratio w/d.

Fig. 1 shows the effect of lateral heat spreading on ΔT_m as a function of the ratio between the in-plane and out-of-plane thermal conductivity for three different values of w/d. For a film with isotropic thermal conductivity, the temperature rise is roughly halved for $w/d = 1$. The temperature rise is more sensitive to the anisotropy when the in-plane thermal

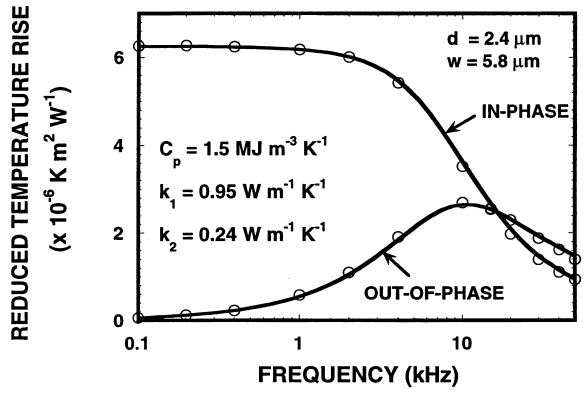

Fig. 2. Predicted and measured frequency dependence of the temperature rise in a 5.8 μm wide metal line deposited on a 2.4 μm thick polyimide film.

conductivity is smaller than its out-of-plane counterpart and also when the heating line is narrower. In the limit of very large *w/d*, the effect of lateral heat spreading becomes negligible, allowing the independent determination of the out-of-plane thermal conductivity of anisotropic films. In addition to a very wide heating line, a mesa structure with appropriate dimensions can accomplish the same goal by minimizing the lateral temperature gradient [18]. Once the out-of-plane thermal conductivity is determined, the in-plane conductivity can be extracted from data on narrower heating lines.

Measurements of the steady-state temperature rise due to a thin film, which is given, for instance, by Eq. (5), often hampered by difficulties associated with establishing appropriate thermal boundary conditions at the bottom of the substrate. Thermal contact resistance between the substrate and the sample holder is hard to control and reproduce, but it often dominates the total thermal resistance. One way to resolve this problem is to employ additional thermometry lines located at two different distances from a narrow heating line and indirectly measure substrate temperature rises [16,20]. This method requires independent measurements of substrate thermal conductivity and rather involved analysis to extract in-plane and out-of-plane thermal conductivity. The present study adopts an alternative method to overcome substrate heating problem. The method induces harmonic heating at frequencies that are high enough to confine the heat affected region within the substrate, but at the same time low enough to be able to treat heat conduction within the film as steady state [21]. The latter condition is fulfilled for measurement frequencies that are much lower than the thermal diffusion frequency of the film. For most dielectric films with thickness of the order of a micrometer on silicon substrates, the required frequency is of the order of a few hundred hertz at room temperature. The total temperature rise at the low frequency limit consists of a frequency-independent component due to the film and a frequency-dependent component due to the substrate heating [14]. Fig. 2 illustrates the amplitudes of the in-phase and out-of-phase components of reduced temperature rises in a metal line, which is deposited on a polyimide film studied here. The reduced temperature rise is defined as temperature rise per unit surface heat flux. Due to the very small thermal conductivity of the polyimide film, the substrate heating is negligible in the particular case.

At higher frequencies, harmonic heating provides additional information on heat conduction within a film. The heat capacity of thin films can be determined from high-frequency data once the thermal conductivity is determined from the low-frequency data. A particularly convenient measurement parameter is an effective heat diffusion frequency, at which the magnitudes of the in-phase and out-of-phase components coincide. Alternatively, this parameter can be used to determine the thermal conductivity of thin films [22], an approach that does not require calibration. The measured frequency-dependence also contains information on thermal boundary resistance at film interfaces, which should be distinguished from the intrinsic film thermal resistance. Thermal boundary resistance at the interface between the metal heater and the dielectric film adds a frequency-independent component to the total temperature rise and thus cannot be distinguished from the film thermal resistance at low frequencies. Further discussion will be provided in Section 3.2.

The amplitude of the temperature oscillations can be calculated using the transient heat conduction equation. A convenient procedure is to substitute $Q = \tilde{Q}e^{i\omega t}$ and $T = \tilde{T}e^{i\omega t}$ into the equation and solve for $\tilde{Q}$ and $\tilde{T}$ [15]. Here $\tilde{Q}$ and $\tilde{T}$ are complex numbers carrying phase as well as amplitude information. Numerical solutions are obtained in the present study to accurately model the influence of the metal line and the substrate.

3. Experiments

3.1. Experimental procedure and sample preparation

Harmonic Joule heating is induced in a metal line deposited on a dielectric film using a waveform generator. The resulting temperature oscillations at twice the current frequency are measured electrically using the method known as the 3ω technique [14] and also optically using the thermoreflectance technique [22,23]. The optical technique has the advantage of high temporal resolution, which permits the measurement at high frequencies. The actual measurement frequencies are limited in the present study by the lock-in amplifier to around 60 kHz for the electrical technique and 100 kHz for the optical technique. Total temperature rise in the Joule heated metal line is kept to be less than 10 K during the measurements. Details of the individual technique can be found in the cited references and will not be repeated here.

The experimental setup is first verified using thin films whose properties are relatively well characterized. The ther-

Table 1
Measured thermal conductivity of the polyimide films in units of W/mK together with experimental uncertainties.

Thickness (μm)	Out-of-plane conductivity	In-plane conductivity	
1.4	0.24 ± 0.01	1.42 ± 0.28	1.42 ± 0.23
2.4	0.24 ± 0.01	0.98 ± 0.17	0.98 ± 0.15

mal conductivity of thermally-grown silicon dioxide and low-temperature CVD silicon-dioxide films are measured using both the temperature rises at low frequencies, where heat conduction through the films can be treated as steady sate, and the effective diffusion frequency. The deduced values are consistent with each other and agree well with those in the literature [24].

The polyimide samples used in the present study are made from commercially available benzophenone tetracarboxylic acid dianhydride (BTDA-ODA-MPD) polyamic acid precursor (DuPont PI 2556). To enhance the adhesion, 0.15 μm-thick silicon nitride layers are deposited on polyimide layers before metal deposition. Processing details are reported elsewhere [22]. The heating lines are made of sputtered aluminum and are 2000 μm long and 0.25 μm thick. The widths of the metal lines used are around 1, 5.5, and 200 μm. Line dimensions vary slightly among different samples. The schematic of the metal lines is similar to what was used in the previous studies [14].

3.2. Results and discussion

The out-of-plane thermal conductivity of the polyimide films is first determined using the 200 μm-wide metal lines based on the 3ω technique for thin films [21]. The thermal conductivity of the silicon nitride layers was characterized independently. Table 1 lists the obtained values for two different thicknesses of the polyimide films. These values agree with the results of previous measurements using the optical thermometry technique and those using micro-mesa structures with polyimide films sandwiched between two metal layers [22].

The reported out-of-plane thermal conductivity can deviate from the intrinsic film thermal conductivity due to the existence of thermal boundary resistance at interfaces. Direct measurements of the thermal boundary resistance between silicon-nitride and polyimide layers, and between polyimide layers and silicon are not available. The present study analyzes data for the 0.5 μm-thick polyimide film with a 0.25 μm-thick metal line, which are not very sensitive to the polyimide film heat capacity [22]. This feature allows the issue of thermal boundary resistance to be addressed largely independent of the uncertainty in the film heat capacity. Measurement of the heat capacity of the polyimide film will be discussed shortly. Predictions are made on the frequency-dependent temperature rises for several different combinations of the thermal boundary resistance and the out-of-plane thermal conductivity. The combinations are selected such that the total thermal resistance extracted from the low-frequency data is reproduced. Comparison of these predictions with the high-frequency data gives an upper bound of 2×10^{-7} $m^2/(K\ W)$ on the thermal boundary resistance. The resulting uncertainty in the out-of-plane thermal conductivity is estimated to be less than 4 percent, which is incorporated in the uncertainty reported in Table 1.

With the out-of-plane thermal conductivity determined above, data for the thicker films obtained at frequencies up to 60 kHz are analyzed to obtain the film heat capacity, 1.5 ± 0.15 MJ/m^3 per K. This is close to the bulk value of the polyimide with identical molecular composition (DuPont data sheet). Predicted frequency-dependent temperature rises based on the measured thermal properties agree with the data over the entire measurement range, whose maximum limit has corresponding heat diffusion length of near 0.5 μm. In extracting the thermal transport properties, the present work assumes that the films are homogeneous.

In-plane thermal conductivity of the polyimide films is deduced from the low-frequency data on samples with 1 and 5.5 μm-wide metal lines and are also listed in Table 1. An example plot of the measured and predicted amplitudes of temperature oscillations per unit heat flux is given in Fig. 2. In the data analysis the present study assumes that the principal axes of the thermal conductivity tensor are aligned with the film axes. This is reasonable considering the fact that preferential alignment of molecular chains parallel to the film plane is conceived to be the main source of anisotropy in polyimide films [25]. Previous studies noted importance of molecular orientation on the anisotropy in thermal and mechanical properties [3,26,27]. The assumption is also justified by the close agreement of the results obtained from the 1 and 5.5 μm-wide metal lines for the 2.4 μm-thick polymer film.

The measured ratio of the in-plane to out-of-plane thermal conductivity is larger than that of a previous study on different types of polyimide films [3]. Large difference in the out-of-plane thermal conductivity between the previous and current study, however, suggests that molecular structures are very different and therefore direct comparison of the two results is not meaningful. The measured in-plane thermal conductivity increases with decreasing film thickness. The molecular in-plane orientation of BPDA-PDA polyimide films was observed to increase as the film thickness decreases [28]. The present results, however, are in contrast to existing data on different kinds of polyimide films, where no systematic thickness-dependence of in-plane thermal conductivity was observed [10]. Polyimide chemistry and molecular weight of the polymer solution were shown to be some of the factors that influence the microstructure of polyimide films and its thickness dependence [29].

The uncertainty in the in-plane conductivity comes mainly from the uncertainty in the measured temperature

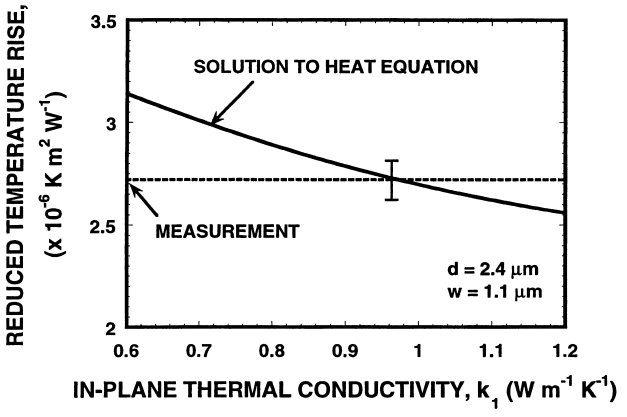

Fig. 3. Steady-state temperature rise per unit heat flux associated with a 2.4 μm thick polyimide film. The vertical bar represents the experimental uncertainty limit in the measured temperature rise.

rises, that in the out-of-plane thermal conductivity, and that in the relevant geometric dimensions. As a part of the sensitivity analysis, the steady-state temperature rise due to the 2.4 μm-thick film is calculated as a function of the in-plane thermal conductivity and is compared with the experimental result in Fig. 3. Similar analysis is performed for the other parameters and the overall uncertainty is determined by combining these analyses using a sum of squares method. Advancement in optical lithography techniques and metrology tools allows the use of sub-micrometer wide metal lines in thermal characterization studies [19] which can improve measurement accuracy.

4. Summary

A technique is developed for thermal characterization of anisotropic thin dielectric films, on substrates with high thermal conductivity. The technique does not require demanding sample preparation processes and consequently avoids potential damage to samples during the substrate removal process, which is necessary for techniques requiring free-standing film structures. Harmonic Joule heating is induced in metal lines with varying widths and their temperature is monitored to extract information on lateral spreading of heat inside the film. Measurements of temperature rises are performed over wide frequency ranges, which can yield information regarding heat capacity and thermal boundary resistance as well as thermal conductivity.

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