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Thickness-dependent thermal properties of amorphous insulating thin films measured by photoreflectance microscopy

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ABSTRACT

In this work, we report on the measurement of the thermal conductivity of thin insulating films of SiO_2 obtained by thermal oxidation, and Al_2O_3 grown by atomic layer deposition (ALD), both on Si wafers. We used photoreflectance microscopy to determine the thermal properties of the films as a function of thickness in the 2 nm to 1000 nm range. The effective thermal conductivity of the Al_2O_3 layer is shown to decrease with thickness down to 70% for the thinnest layers. The data were analyzed upon considering that the change in the effective thermal conductivity corresponds to an intrinsic thermal conductivity associated to an additional interfacial thermal resistance. The intrinsic conductivity and interfacial thermal resistance of SiO_2 were found to be equal to 0.95 W/m·K and 5.1×10^{-9} m^{2}K/W respectively; those of Al_2O_3 were found to be 1.56 W/m·K and 4.3×10^{-9} m^{2}K/W.

1. Introduction

Thermal and electronic conductivities are strongly correlated in most materials. However, many applications demand the maximization of one of these properties while minimizing the other. In microelectronics for instance, good electrical insulation is essential (capacitors, interconnects), but low-k dielectrics usually come with poor thermal conductivity, hampering heat dissipation. Conversely, high electrical conductivity and thermal insulation are crucial for thermoelectric conversion, in order to avoid Joule heating while preserving the temperature gradient [1,2]. Nanostructured materials offer a new way to act on these antagonistic requirements, since nanoscale thermal properties can significantly differ from bulk values [3,4]. A lot of attention has been focused recently on understanding the underlying physics, like phonon scattering [5] and heat transport phenomena [6,7].

In this paper, we investigate the thermal properties of two electrical insulators, SiO_2 and Al_2O_3 thin films. SiO_2 is essential to microelectronics and other industrial applications. It has therefore received a lot of attention, and its thermal properties are relatively well known. Some research groups have studied the thermal conductivity of Al_2O_3 amorphous thin films [8–11], but the evaluation of their interfacial thermal resistances is still very incomplete [12]. Al_2O_3 amorphous thin films are promising, since they can reduce electronic recombination losses in solar cells by the passivation of silicon surfaces, thus enabling higher efficiency [13]. Moreover, thin amorphous Al_2O_3 films are good thermal insulators as well as excellent moisture barriers [14] that can be fabricated at low temperatures [15,16], making them highly desirable in electronic components [17].

A broad range of experimental methods is available in order to determine the thermal properties of materials. They essentially differ in their heat generation process (optical, Joule, …), in the property which is probed (temperature of the surface, sample or air, acoustic waves, etc.…), and in the probing mechanism (refractive index, thermal emission, interferometry, fluorescence, electrical resistance…). Temporally, various strategies have also been developed: steady state, transient or modulated. Several reviews of thin films characterization techniques have been proposed [18,19]. Among these techniques, modulated photoreflectance microscopy has the advantages of being contactless, non-destructive and, owing to the high spatial resolution of visible light microscopy, allows measurements on relatively small samples (> 10 μm). It is based on the generation of thermal "waves" by intensity-modulated optical excitation. This technique was first

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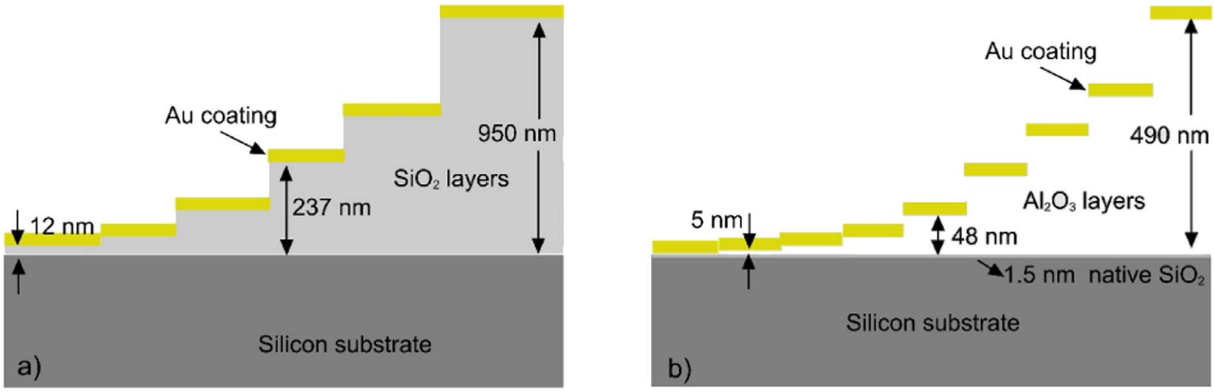

Fig. 1. Schematic of the fabricated samples, a) the SiO_2 layers and b) the Al_2O_3 layers.

proposed by A. Rosencwaig et al. [20], and then widely used to determine the thermal properties of bulk materials [21,22], grains [23], coatings and thin films [24,25]. In this work, the frequency domain photoreflectance method is used to study the effect of thickness on the thermal properties of amorphous SiO_2 and Al_2O_3 thin films. The method requires the deposition of a gold layer to opacify the surface, but is well adapted to this kind of study, where different nanoscale layers have to be distinguished. A 3D heat diffusion model was used to extract the thermal properties of each material independently [22].

2. Experimental

The SiO_2 thin films with different thicknesses were fabricated by Kelvin Nanotechnology Ltd. (KNT), in collaboration with Glasgow University. The starting material is a thick layer of SiO_2 grown by thermal oxidation on a p-type Si wafer. Repeated photolithography steps, followed by timed hydrofluoric acid (HF) etching, were performed to obtain the required thicknesses of 12, 30, 65, 145, 237, 530 and 950 nm, as depicted in Fig. 1a). The layer thickness was measured by white light interferometry [26].

The studied Al_2O_3 samples were fabricated by Picosun using a Picosun™ ALD reactor. ALD is a powerful method to grow fully conformal, pinhole-free layers with atomic accuracy [27]. This is based on the self-terminating nature of gas-solid reactions taking place at the sample surface. The studied thin films were grown on silicon wafer with a 10–14 µm n-type epilayer of resistivity 3–6 Ω·cm and a native SiO_2 layer (thickness of approximately 1.5 nm). The values of the obtained thicknesses (2, 5, 9.5, 24, 48, 98, 152, 196, 490 nm) were measured by ellipsometry [28], as shown in Fig. 1b).

The frequency-domain photoreflectance microscopy is one of the most convenient photothermal techniques to measure the thermal diffusivity of solid materials. It uses an intensity-modulated green laser (λ = 532 nm) focused by an optical microscope onto the surface of the sample [21,29]. The modulated beam excites thermal "waves", and the resulting distribution of the surface temperature modulation is read by a second probe laser (λ = 670 nm) using the temperature dependence of the reflectivity, which is proportional to temperature in a first approximation, and depends on the nature of the reflecting material. The amplitude and the phase of the modulated photoreflectance signals are extracted by lock-in detection and recorded as a function of the distance between the two spots. This method requires a good absorption of the heating laser, for efficient thermal wave generation, and an efficient reflection of the probe laser. In the case of transparent materials, an opaque and reflective transducing surface is therefore always needed in order to create and probe the thermal waves. In our case, the Al_2O_3 and SiO_2 samples were coated with a 100 nm thick gold layer. The measurements were performed at room temperature, with an excitation frequency of 150 kHz. The heat diffusion theoretical model depends on an effective diameter, which is obtained by a convolution of the heating laser spot and the probing laser spot. This effective diameter was found to be 3 µm, a value which can be either determined by convolution of the Airy discs associated to the laser wavelengths and the numerical aperture of the objective, or by fitting measurements obtained on a known sample.

3. Results and discussion

The thermal diffusivity D of an optically and thermally thick, isotropic, bulk material can be straightforwardly extracted from the slope $\frac{dP}{dx} = \sqrt{\frac{\pi f}{D}}$ of the phase lag P of the surface temperature rise with respect to the excitation at a distance x from the excitation, where f is the excitation frequency [30]. For a multilayered sample, there is no such simple relation, since the phase slope.

$\frac{dP}{dx}$ is then a function of the diffusivities and conductivities of the different layers [29]. However, the surface temperature modulation can be calculated using a thermal quadrupole formalism [25]. The thermal diffusivity and conductivity of one layer of known thickness is therefore obtained by using this model and determining the thermal properties which best fit the measured amplitude and phase of the surface temperature modulation. The samples are considered homogenous laterally over the studied region (40 µm), and the anisotropy of the diffusion coefficient, i.e. differences between the in-plane and through-plane diffusion coefficients, $D_{//}$ and $D_{\perp}$ respectively, are neglected. Moreover, the diffusivity and conductivity are not determined independently, but rather upon assuming that the ratio of thermal conductivity k to thermal diffusivity D is constant $\frac{k}{D} = \rho C$=cst, with ρ the density and C the specific heat of the bulk material, taken from the literature. In some cases, e.g. when the thermal contrast and the measurement Signal to Noise Ratio are sufficient, the thermal parameters of up to two media [29] can be obtained simultaneously. However, because the number of variables increases with each additional layer, a reliable determination of the thermal properties of a given layer requires the precise knowledge of the thicknesses and thermal properties of the other layers. Therefore, the thermal properties of the different layers composing the samples were determined in three steps, depicted in Fig. 2:

- Step 1: A gold layer was deposited simultaneously on glass and on the studied samples. Since its thermal properties are well known, and because it is thermally insulating, glass is an appropriate choice to study the properties of the 100 nm gold layer which essentially drive the surface temperature.
- Step 2: An identical gold layer was deposited on a bare Si substrate, to characterize the thermal properties of the substrate.
- Step 3: An identical substrate, supporting the layers to be characterized and the same gold layers was fabricated. Using the properties obtained in steps 1 and 2, the thermal properties of the relevant layer can finally be measured.