SURFACE TEMPERATURE MEASUREMENT OF DIELECTRIC MATERIALS HEATED BY PULSED LASER RADIATION

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The temperature of a dielectric surface irradiated by a laser pulse can be measured by recording the time-resolved resistance change of a thin platinum stripe vapor-deposited on this surface. Results are reported for quartz and glass surfaces exposed to a pulsed CO_2 laser, showing heating rates in excess of 10^8 K/s.

1. Introduction

Laser-induced thermal desorption (LITD) techniques have been extensively exploited to analyze surface adsorbates, to clean surfaces and to study processes and reactions on surfaces (for reviews, see ref. [1]). In LITD, a laser pulse irradiates an adsorbate-covered surface which absorbs a portion of the laser energy, depending on the optical properties of the surface. Within picoseconds, the absorbed energy is converted into heat [2]. This causes a temperature jump and induces thermal desorption of adsorbates.

We report here a new method to measure the transient surface temperature induced by pulsed laser irradiation. The method is simple and applicable to almost any insulator surface. It uses an optically thin platinum stripe as a resistivity thermometer for measuring transient surface temperatures with nanosecond time resolution.

There have been a number of methods to measure surface temperatures generated by pulsed laser heating, including photothermal displacement spectroscopy [3], blackbody radiation [4], Raman scattering [5], metal film resistance thermometry [6] and pyroelectric calorimetry [7]. However, these methods have intrinsic limitations: The photothermal displacement method presently cannot be calibrated to measure temperatures. The blackbody radiation method has only been applied successfully to measure very high temperatures induced by cw laser irradiation. Raman scattering works best on well-defined surfaces and even then, has a significant uncertainty (≈ 100 K). Metal film thermometers have never been applied to measure transient temperatures of a surface heated by pulsed laser radiation. Pyroelectric calorimeters operated as thermometers are limited to selected materials which can be deposited as thin films on the pyroelectric transducer.

There have also been theoretical approaches to model surface heating following pulsed laser irradiation. Ready described laser-induced surface heating with a one-dimensional model [8]. For the purpose of calculating the surface temperature changes, the thermal properties of the solid are often assumed to be independent of temperature, which is a reasonable approximation for metals and relatively small temperature changes. Recently, further simplifications have been employed, using a temperature-independent absorption coefficient and Gaussian [9], square and triangular [10] laser pulse shapes. We show that these simplifications may result in large deviations from the actual temperature of a dielectric surface, because in general the optical and thermal properties of many dielectric solids are strongly dependent on temperature, and the temporal and spatial profiles of a laser pulse can be very complex.

2. Experimental

Thin film resistance sensors are fabricated by vapor deposition of platinum on optically flat glass and

quartz surfaces, using an electron beam evaporator and physical masks. The sensor geometry is shown in fig. 1. The actual sensor element is only the 1×0.5 mm stripe with a thickness of 100 Å or less. It has a resistance of several hundred ohms, depending on the sensor thickness. The resistivity of thin metal films can be more than 10 times higher than the bulk value because of the size effect and meander-like film structure [11]. However, ohmic behavior has been found for platinum films less than 10 Å thick [12]. The contacts (see fig. 1) are 2000 Å thick, have a resistivity close to the bulk value, and hence contribute negligibly to the sensor resistance.

The sensor resistance is determined by feeding 0.4 mA from a fast constant current source (10 ns rise/fall time) through the platinum stripe and measuring the voltage drop with a 125 MHz oscilloscope. Because both the resistance and the temperature coefficient of resistance vary with the sensor geometry and thickness, each sensor is individually cali-
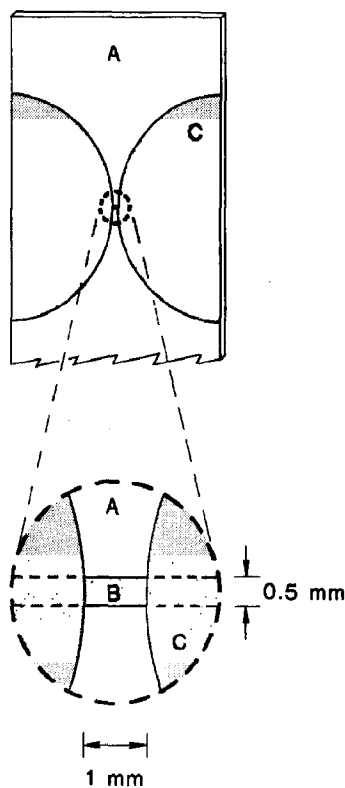
brated whenever a temperature is to be measured. This is done by monitoring the sensor resistance while cooling it slowly after annealing it for 1–2 days at 850 K. Calibration curves are obtained by plotting the output voltage of the readout electronics versus the oven temperature. Calibration curves for the same sensor are reproducible within 10 K. They show two linear regimes with a transition around 650 K, having almost the same slope; this behavior agrees very well with standard tables for platinum resistance thermometers [13].

The heating laser used is a pulsed CO_2 laser (Pulse Systems, LP-30) with a nominal pulse width of 10 μs. Incident laser fluences are controlled by adjusting the distance between the sensor surface and a focusing ZnSe lens. In all experiments, the beam spot is over 1 mm in diameter, so that it covers the entire sensor. Laser pulse energies are measured with a 372 Scientech energy meter. An Eltech 420-2 pyroelectric laser detector serves to determine the temporal laser pulse profiles with 2 ns resolution. For transient measurements, both the sensor and the readout electronics must be shielded in a metal box to avoid rf interference from the CO_2 laser.

The materials studied are fused quartz (Quartz International) with well-known physical properties [#1] and soda-lime glass (Airic Scientific) with rather ill-defined properties [#2].

3. Results and discussion

Fig. 2 shows typical oscilloscope traces obtained from heating a glass surface with the CO_2 laser whose temporal pulse profile is displayed in fig. 2d. The surface temperature evolution is characterized by a

Fig. 1. Sensor geometry: (A) Substrate surface; (B) vapor-deposited Pt stripe, 100 Å thick or less; (C) vapor-deposited contacts, 2000 Å thick.

[#1] Physical constants used for quartz (data from the manufacturer): density $p = 2200$ kg/m^3, thermal conductivity $\lambda = 1.4$ W/m K (at 20°C), specific heat $c = 670$ J/kg K (at 20°C). Optical properties [14]: Complex index of refraction $n' = n + i\kappa$ at 10.6 μm: $n = 2.22$, $\kappa = 8 \times 10^{-2}$. From these values the skin depth ($d = 10$ μm) and the reflectivity ($R = 12\%$) at 10.6 μm could be calculated.

[#2] From the manufacturer, only the density ($p = 2402$ kg/m^3), the softening point (997 K) and a value for the thermal expansion coefficient were available. Tables gave information about the chemical composition and a value for the mean specific heat ($\bar{c} = 1.168$ J/kg K, between 313 and 1073 K, see ref. [13, p.917]).
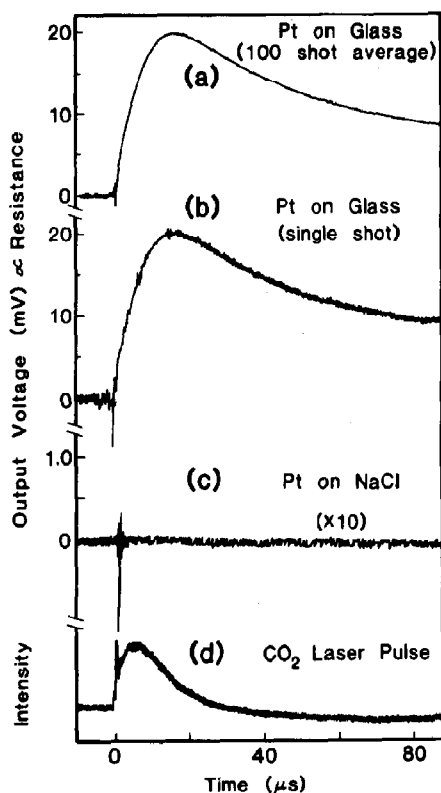
Fig. 2. Typical response from (a) 72 Å Pt sensor on glass, 100-shot average; (b) 72 Å Pt sensor on glass, single-shot measurement; (c) 100 Å Pt sensor on NaCl (IR transparent); (d) CO_2 laser temporal profile.

sharp rise coincident with the initial spike of the laser profile, a slower rise thereafter, a peak about 15 μs after the onset of the laser pulse and a long tail that extends over several hundred microseconds (see fig. 2a). 100 μs after the onset of the laser pulse, the temperature is still high; it has fallen by only about 50% of the peak temperature difference. The slow decay of the signal at later times reflects the poor heat conductivity of the substrate materials used. We do not offer a quantitative analysis of this behavior because lateral heat flow as well as heat sink effects caused by the contacts are believed to play a role at such long times.

In contrast to a number of other methods, single-shot measurements are possible with our approach and agree very well with averaged measurements (see fig. 2b). In fact, one must rely on single-shot measurements for the resolution of very sharp features, since they would be blurred by averaging procedures

because of the inherent trigger instabilities of our CO_2 laser.

A number of doubts and concerns about the meaning of our measurements are examined:

Are we really measuring the surface temperature? The skin depth (defined as $1/e$ decay of the incident laser intensity in the solid) for quartz is 10 μm (see footnote 1), which is over 1000 times larger than the sensor thickness. Furthermore, the heat diffusion time τ, estimated by $\tau = d^2/4\kappa$, through a $d = 100$ Å thick platinum layer (thermal diffusivity $\kappa = 0.25$ cm^2/s) is on the order of 1 ps. In other words, "local thermodynamic equilibrium" of the sensor with the substrate is established rapidly compared with the time scale of the transient heating. Heat diffusion in the quartz substrate occurs with a time constant of 25 ps over a distance of 100 Å and with a time constant of 2.5 ns over a distance of 1000 Å (see footnote 1). This means that within a few nanoseconds, the original temperature profile induced by laser heating of the solid (exponential decay from the surface into the solid) will not be perturbed by the presence of the sensor.

What is the time resolution? Because the sensor responds to the actual surface temperature on a time scale of 1 ns, the temporal resolution in our experiments is completely determined by the present electronics, which is on the order of 20 ns. We note that for heating on a time scale faster than a nanosecond, a 100 Å sensor could not follow the surface temperature accurately.

Do absorptions by the platinum film, or reflections on its surface cause systematic errors? Compared with the wavelength of the laser (10.6 μm), the sensor is 1000 times thinner and should therefore be nearly transparent to the infrared radiation, i.e. should not absorb or reflect it. In contrast, bulk platinum is a very good IR reflector (93% at 10.6 μm). By depositing the same sensor structure on a NaCl substrate which is IR transparent, we are able to assess the contribution from direct absorption of the CO_2 laser by the platinum film. The result is shown in fig. 2c: Except for some electronic noise, no signal is detected. This result demonstrates negligible heating of the platinum stripe by the laser pulse. Similarly, IR reflectivities of the surfaces are independent of the presence or absence of a thin Pt film within experimental error.

Surface temperature profiles for different laser fluences are obtained by focusing or defocusing the laser beam with respect to the surface plane. The results are shown in fig. 3. The curves peak at the same time (15 µs) and show exactly the same behavior, except that the magnitude of the temperature jumps (20, 400, and 710 K) depends on the incident laser fluence.

It is interesting to compare these results with calculated surface temperature jumps. Analytical solutions for this problem can be obtained by using a simple model in which the laser pulse shape is assumed to be triangular [10], and thermal and optical properties of quartz are assumed to be constant, fixed at their room temperature values (see footnote 1). Using this model, maximum temperature differences of 30, 490, and 1730 K are obtained 15 µs after the onset of the laser pulse, for the same fluences as those used in the experiment (fig. 3). Although the shapes of the calculated curves appear similar to the measured ones, the calculated temperature jumps are substantially larger than our experimental results. This discrepancy is worse for higher fluences. For example, the calculated peak temperature for 1.4×10^5 W/cm^2 laser fluence is above the melting point for quartz. However, no surface damage is observed visually for this fluence.

Heating rates are also measured. They depend critically on individual laser pulse shapes and therefore must be determined in single-shot mode. For incident laser fluences ranging from 4×10^4 to 9×10^4 W/cm^2, we find heating rates between 4×10^8 and 7×10^8 K/s caused by the initial spike of the laser pulse. An example is shown in fig. 4. Such high heating rates last only for about 200 ns and raise the temperature by 70 to 100 K; afterwards, the heating continues at about an order of magnitude slower rate until the peak temperature is reached.

Heating rates can also be estimated using the simple model cited above. Values between 5×10^7 and 3×10^8 W/cm^2 are calculated for the range of fluences used, about a factor of 2–5 lower than the measured heating rates corresponding to the spike of the CO_2 laser pulse.

We conclude that this simplified treatment does not adequately describe our experimental findings. To achieve a better fit with the experiment, one must (1) include the exact pulse shape of the heating laser and (2) take into account the temperature dependence of the heat conductivity, the heat diffusivity and the reflectivity of the irradiated solid. This is a complex numerical task which was not attempted.

One problem that became apparent during the course of this work is the spatial inhomogeneity of the CO_2 laser beam. In addition to concentrical diffraction rings originating from the output coupler, there is a central "hot spot" whose influence is particularly significant for fluences below $\approx 1 \times 10^4$ W/cm^2, obtained by defocusing the laser beam with respect to the surface. In this case, the diameter of the hot spot approaches the length of the sensor element
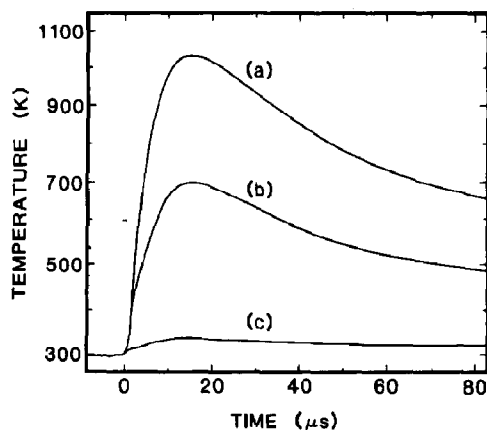

Fig. 3. Temperature deduced from sensor resistance (76 Å Pt on quartz) as a function of time at three different laser fluences: (a) 1.4×10^5 W/cm^2; (b) 4×10^4 W/cm^2; and (c) 2.5×10^3 W/cm^2.
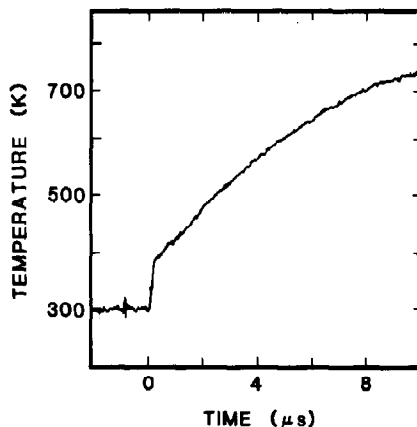

Fig. 4. Temperature change measured in single-shot mode (76 Å Pt sensor on quartz, average laser fluence=4×10^4 W/cm^2). The temperature rises by 80 K within 160 ns, resulting in a heating rate of 5×10^8 K/s.
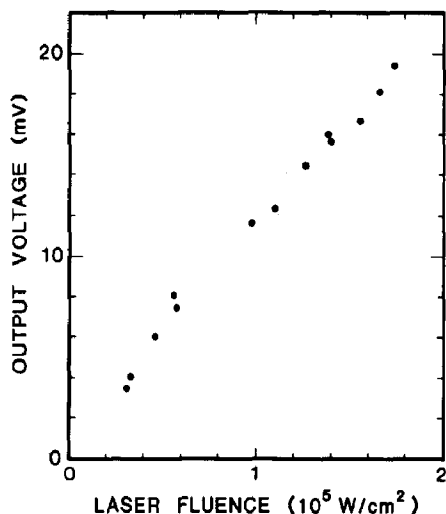
Fig. 5. Dependence of the output voltage (proportional to the peak temperature) on the incident laser fluence, measured in single-shot mode (99 Å Pt sensor on glass).

and temperature jumps can be up to twice as large as expected from measurements at tighter focal conditions. Our method allows us to detect these inhomogeneities. We suggest that a series of miniaturized sensors would permit one to meausure the actual spatial profile of a laser pulse.

In order to measure how the temperature jumps scale with laser fluence, an alternative way of varying the laser power is employed. By taking single-shot measurements and using the natural intensity fluctuations of our CO_2 laser, we can get a variation of fluences within a factor of 2, without changing the focal conditions and the sensor position relative to the beam. Even lower laser fluences can be obtained by attenuating the beam with a 2 mm thick Ge window. Fig. 5 shows the results obtained from a 99 Å sensor on a glass surface. Within the limited range of fluences used, we find a linear relationship between the peak temperature and the fluence.

4. Conclusions

We have demonstrated the successful application of a vapor-deposited resistance thermometer to measure transient temperatures of laser-heated surfaces. This is an intrusive measurement but has the advantage that it is quite generally applicable to many

insulator surfaces where other experimental approaches and simplified theoretical models fail. The method is simple and accurate (± 10 K), allows single-shot measurements to be carried out and provides a nanosecond time resolution.

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