Measurement of solid–liquid interface temperature during pulsed excimer laser melting of polycrystalline silicon films

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A nanosecond time resolution pyrometer has been developed for measuring the transient temperature of thin polycrystalline silicon (p-Si) films irradiated by a pulsed excimer laser. The sample design structure and material optical properties allow direct measurement of the temperature at the solid–liquid phase change interface. © *1994 American Institute of Physics.*

Surface melting of semiconductor materials by pulsed excimer lasers has been studied extensively in the literature. In most cases, rapid melting and resolidification induced by the pulsed excimer laser irradiation is understood as a one-dimensional process. The excimer laser irradiation offers a well controlled experimental technique for the study of the interface response when the local equilibrium conditions are disturbed. The interface response function is often described by the interface kinetic theory[1] where it is shown that the solid–liquid interface superheating is approximately proportional to the velocity of the interface: $\Delta T = CV_{\mathrm{int}}$, where C is a material constant. This interface kinetic relation is widely adopted in the literature for numerical simulation of rapid melt propagation. To verify the kinetic relation and quantify the material constant C, both the interface velocity and the interface temperature need to be determined. The interface velocity has been studied by the transient conductance measurements,[2] while various transient temperature measurement techniques have been developed.[3–5] Measurement of interface temperature has also been reported.[6,7] However, rather than being able to measure the temperature right at the phase change interface, these methods measure the temperature at a certain distance away from the interface,[6] or the response due to the temperature-dependent material properties integrated over a certain depth.[7] The accuracy of these methods relies largely on accurate knowledge of material properties. Here we report the measurement of the interface temperature based on the transient thermal emission measurement at the solid–liquid interface with a nanosecond time resolution (notice that the fastest pyrometer reported is of 2 μs time resolution[8]).

The sample structure is a 0.5 μm thick p-Si film deposited on top of a 0.5 mm thick fused-quartz substrate, by low-pressure chemical-vapor deposition. The p-Si film is heated by a pulsed KrF excimer laser with a pulse duration of 26 ns and a wavelength of 248 nm (Fig. 1). A beam homogenizer is used to ensure spatial uniformity in the laser beam. The laser intensity uniformity on the sample surface is measured to be within 10% over the central 90% portion of the laser beam spot. The laser spot size on the sample surface is about 6 mm^2. A germanium diode is used to detect the emission signals. As shown in Fig. 1, thermal emission from the sample is measured from the *back* side of the sample, in the wavelength range between 1.1 and 1.7 μm. In this wavelength range, both the solid silicon and the quartz substrate are transparent, so that the emissivity of these materials is zero according to Kirchhoff's Law. In contrast, the liquid silicon has an emissivity about 0.28. Thus the entire measured thermal emission signal can be ascribed to liquid silicon. At near-IR wavelengths, the liquid silicon has a radiation absorption depth less than 18 nm. Therefore the measured thermal emission comes from the liquid in the immediate vicinity of the solid–liquid interface. The effect of the movement of the interface on the energy collection (depth of field effect) is negligible. This is because the maximum melting depth achieved in this experiment is less than 0.4 μm, which is five orders of magnitude smaller than the focal lengths of the lenses (65 mm).

The germanium diode senses the thermal emission from an area of 1 mm^2 at the center of the heated spot. It is reversely biased to achieve a rise/fall time of 1 ns. The electric signal from the germanium diode is recorded on a digitizing oscilloscope with a 1 GHz sampling rate. Bandpass filters with center wavelengths at 1.2, 1.4, 1.5, and 1.6 μm and bandwidths of approximately 0.08 μm are used to acquire the spectral thermal emission signals. The emissivity of the sample is obtained from a transient reflectivity measurement.

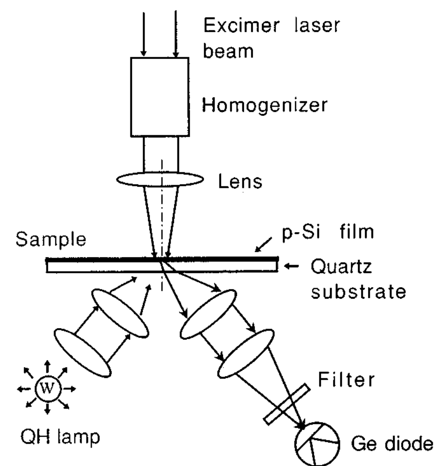

FIG. 1. Experimental setup for transient thermal emission and emissivity measurements during pulsed excimer laser melting of p-Si films.

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A quartz halogen (QH) lamp is used as the light source. The light of the QH lamp (Fig. 1) is focused onto the sample surface with a beam spot size of about 1 mm^2. The reflectivity of this beam is refocused by lenses onto the germanium diode and recorded by the oscilloscope. Bandpass filters (the same as those used in emission measurement) are used to measure the reflectivity at different wavelengths. Two measurements are taken at each wavelength and laser fluence, one for the thermal emission measurement (with the QH lamp off) and one for the reflectivity measurement which also includes the emission signal. The reflectivity is obtained by subtracting the thermal emission signal from the total signal. To eliminate the effect of the laser energy instability, the fluence of each laser shot is measured. The experimental data are accepted when the measured laser fluence has the desired value. To eliminate the effect of the laser energy instability. The surface of the sample shows little change from shot to shot for the range of laser fluences used in this experiment.

Planck's distribution of blackbody radiation intensity law is used to derive the temperature from the measured thermal emissions:

$$e_{\lambda b}=\frac{2\pi C_1}{\lambda^5\ \exp(C_2/\lambda T)-1}, \tag{1}$$

where $e_{\lambda b}$ is the blackbody emissive power, and C_1 and C_2 are blackbody radiation constants. The detector collects thermal emission within a solid angle (θ_1 to θ_2, ϕ_1 to ϕ_2) and a wavelength bandwidth (λ_1 to λ_2). The voltage signal recorded on the oscilloscope, V, is expressed as

$$V=W/\pi\int_{\lambda_1}^{\lambda_2}\int_{\theta_1}^{\theta_2}\int_{\phi_1}^{\phi_2}\epsilon'(\lambda,\theta,\phi,T)\tau(\lambda)D(\lambda)\times e_{\lambda b}\, d\phi\, d\theta\, d\lambda\, dA. \tag{2}$$

In the above equation, W is the impedance of the oscilloscope (50 Ω), $\tau(\lambda)$ the spectral transmission of the lenses and filters, $D(\lambda)$ the responsivity of the germanium diode (in units of A/W), $\epsilon'(\lambda,\theta,\phi,T)$ the directional spectral emissivity, and dA the area on the sample where the thermal emission is sensed. Invoking Kirchhoff's law, the directional spectral emissivity is equal to the directional spectral absorptivity:

$$\epsilon'(\lambda,\theta,\phi,T)=\alpha'(\lambda,\theta,\phi,T). \tag{3}$$

For an opaque material (such as liquid silicon), the spectral directional absorptivity can be expressed as

$$\alpha'(\lambda,\theta,\phi,T)=1-R_s(\lambda,-\theta,\phi,T)-R_d. \tag{4}$$

$R_s(\lambda,-\theta,\phi,\mathrm{T})$ is the specular reflectivity while R_d is the diffuse reflectivity. It is assumed that the melt propagation front is planar, and the diffuse reflectivity at the melt front can be neglected. Thus the emissivity can be obtained from the specular reflectivity measurement:

$$\epsilon'(\lambda,\theta,\phi,T)=1-R_s(\lambda,-\theta,\phi,T). \tag{5}$$

The temperature is obtained by solving Eq. (2) from the thermal emissions measured at four different wavelengths. The temperature measurement is calibrated at steady state with a quartz halogen lamp, whose temperature is measured

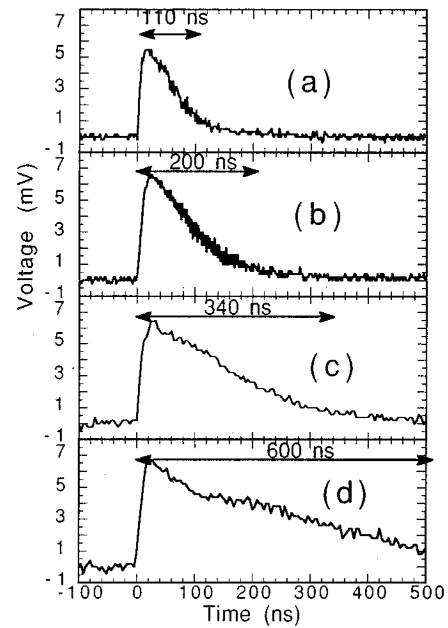

FIG. 2. Transient thermal emission signal at $\lambda=1.5$ μm. (a) $F=0.55$ J/cm^2, (b) $F=0.65$ J/cm^2, (c) $F=0.75$ J/cm^2, and (d) $F=0.95$ J/cm^2.

by a NIST calibrated pyrometer. The confidence level in determining the *absolute* temperature value is estimated to be ±50 K. The error in determining the *relative* temperature (temperature difference at different time or at different fluence), which is determined by the resolution of the digitizing oscilloscope, is ±15 K.

Figure 2 shows thermal emission signals at the wavelength of 1.5 μm. The thermal emission measurement also yields the melting duration (indicated by arrows), since only liquid silicon emits light in the wavelength range between 1.1 and 1.7 μm. Comparing the thermal emission signal at $F=0.55$ and 0.65 J/cm^2, it can be seen that the maximum interface temperature increases with the laser fluence. However, when the laser fluence is higher than 0.65 J/cm^2, the maximum interface temperature does not increase with the laser fluence. The effect of a temperature gradient at the solid–liquid interface is considered in calculating the interface temperature. From numerical simulation,[9] it is found that the temperature gradient at the interface (about 3.3 K/nm at a laser fluence of 0.95 J/cm^2) causes the measured temperature to be approximately 40 K higher than the actual interface temperature. The maximum solid–liquid interface temperatures at different laser fluences are shown in Fig. 3(a).

In our previous work,[9] the transient melting front position was measured using the transient conductance method. The transient melting depth was calculated using a heat conduction model which incorporates a melting front tracking algorithm and the interface response function. The optical refractive index of the p-Si film at the excimer laser wavelength was measured using a variable angle spectral ellipsometer, which is $\tilde{n}_{\mathrm{exc}}=1.2+2.8i$. Thermal and optical properties of bulk silicon[10,11] and quartz[10] were used in the calculation. The measurement matched well with the numerical simulation for quantities such as melt depth and melt

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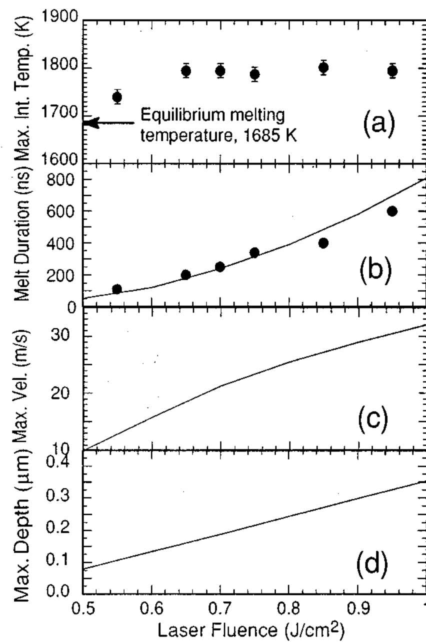

FIG. 3. (a) measured maximum interface temperature. (b) Comparison between the measured and calculated melting duration. (c) Calculated maximum melting velocity and (d) calculated maximum melting depth at different laser fluences.

duration. Here we compare the melt duration obtained from the thermal emission measurement with numerical simulations [Fig. 3(b)]. Good agreement has been achieved between the measured melt duration and the calculations. The possibility that the *high-temperature* solid silicon can contribute to the thermal emission measured from the back side of the sample is examined by measuring the surface reflectance. The surface reflectivity measurement also yields the melting duration due to a large reflectance increase of silicon upon melting. The melting durations measured by surface reflectance are in close agreement with those obtained from thermal emission measurement. Considering that any possible thermal emission from the high-temperature solid silicon can last much longer than the melting duration, we can dismiss the possibility that thermal radiation is emitted from high-temperature solid silicon.

The calculated maximum melt front velocities at different laser fluences are shown in Fig. 3(c). A comparison between the interface velocity and the interface temperature [Figs. 3(a) and 3(c)] allows us to determine the coefficient C in the interface response function. Assuming that there is a linear relation between the interface superheating temperature and the interface velocity at fluences lower than 0.65 J/cm^2, the response function coefficient C is determined to be around 6 K/(m/s). When the laser fluence is higher than 0.65 J/cm^2, the interface superheating temperature is "saturated" at about 110 K. In some cases, the maximum melt depth is used instead of incident energy as an indication of the actual energy coupled to the material. Calculation of maximum melt depth versus laser fluence is presented in Fig. 3(d).

Thermal emission from the top of the sample is also measured to verify that the surface temperature does not reach the boiling temperature (2628 K). In the case of surface evaporation, the solid–liquid interface velocity increases only slightly with fluence; the excess laser energy is consumed by the latent heat of vaporization. The experimental results show that the maximum surface temperature at the laser fluence of 0.95 J/cm^2 is about 2100 K, well below the boiling temperature of liquid silicon.

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