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Transient Optical Transmission Measurement in Excimer-Laser Irradiation of Amorphous Silicon Films

The transient temperature field development during heating of an amorphous silicon (a-Si) film, deposited on a fused quartz substrate, by pulsed excimer laser irradiation is studied. Static reflectivity and transmissivity measurements are used to obtain the thin film optical properties at elevated temperatures. Experimental in-situ, transient, optical transmission data are compared with heat transfer modeling results. The variation with temperature of the material complex refractive index across the thin film thickness is taken into account. The effects of the film thickness and thermal diffusivity, as well as of the laser pulse shape, are discussed.

I Introduction

Pulsed laser irradiation is employed over a wide spectrum of materials processing applications, including surface hardening, alloying, curing, synthesis of compounds, and deposition of thin films. In semiconductor systems (Wood et al., 1984), it is used to anneal ion-implantation surface damage, recrystallize amorphous and polycrystalline films, and enhance dopant diffusion. Recent studies (Zapka et al., 1991) have shown that one of the most effective ways of removing submicron-sized particles from solid surfaces is by deposition of a liquid film on a substrate surface and application of a UV excimer laser pulse on the surface. One of the main issues in improving such processes is the control of the induced transient temperature field. Time-resolved optical transmission and reflection measurements have been reported for the investigation of the irradiation of c-Si silicon on sapphire structures at picosecond (Lompre et al., 1983) and nanosecond (Lowndes, 1982; Lowndes and Jellison, 1984) time scales.

The complex melting and solidification behavior observed in pulsed laser irradiated materials, including nonequilibrium effects, has been studied numerically by Wood and Geist (1986a, 1986b). Time-resolved reflectivity measurements on bulk silicon and germanium using a pulsed excimer KrF laser heating beam were combined with melting-model calculations to explain the mechanism of inhomogeneous melting nucleation (Jellison et al., 1986a). The initial formation and subsequent explosive propagation of buried molten layers in amorphous silicon irradiated by excimer lasers was studied by time-resolved visible and infrared reflectivity and transmissivity measurements (Lowndes et al., 1987). A variety of experimental techniques, including time-resolved transmission, x-ray diffraction, and electrical conductivity measurements, have been used in conjunction with numerical simulations to show that the pulsed laser irradiation of semiconductors is a thermal phenomenon (Jellison et al., 1986b).

The optical transient reflectivity and transmissivity measurements are based on the variation of the material complex refractive index with temperature. The temperature variation of the normal incidence surface reflectivity for bulk samples is generally small (for crystalline silicon (c-Si) $d\mathcal{R}/dT \sim 10^{-5}$ K^{-1}). Interference effects significantly enhance the variation of the thin film reflectivity with temperature. This effect was demonstrated by Grigoropoulos et al. (1991) in their measurements of the thin polysilicon film reflectivity during continuous wave (CW) laser annealing. The thin film reflectivity is expected to be a strong function of the film thickness for semitransparent films. This work presents an optical transmission probing technique for the transient, in-situ monitoring of the temperature field in pulsed excimer laser irradiation of thin amorphous silicon (a-Si) films. A numerical conductive heat transfer model for the transient temperature field in the thin film structure and the substrate is applied. Static measurements of the thin film complex refractive index at elevated temperatures are used to obtain the transient thin film transmissivity from the temperature profile across the thin film. The predicted and experimental transmissivities are in excellent agreement, thus validating the use of this technique for nonintrusive experimental temperature measurement.

II.1 Static Measurements. Amorphous silicon films of 0.2 and 1.0 μm thickness were deposited by e-beam evaporation of crystalline silicon in vacuum onto 250-μm-thick fused quartz substrates. The substrate temperature was kept to 140°C and the deposition rate at 10 Å/s. The uniformity and accuracy of the thickness of the a-Si layer were confirmed using a Tencor Alpha-Step 200 surface profilometer. The samples were then annealed at a temperature of 380°C, in a nitrogen chamber for about 5 hours until optical measurements (reflectivity and transmissivity) were reproducible. The annealing process does

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not cause nitridation, for which much higher temperatures (above 1200°C) would be needed (Nakamura et al., 1983).

It has been reported (Siregar et al., 1979) that the optical properties of submicron-thick, a-Si films do not vary significantly with temperature at the Nd:YAG, $\lambda = 1.064$ μm laser light wavelength. Recent studies (Do et al., 1992) have revealed a significant variation of the optical properties of amorphous silicon films with temperature at the $\lambda = 0.752$ μm diode laser light wavelength.

The standard static reflectivity, $\mathcal{R}$, and transmissivity, $\mathcal{T}$, measurement procedure was used to determine the temperature dependence of the components of the thin film complex refractive index. An infrared diode laser beam ($\lambda = 0.752$ μm) was directed onto the sample at normal incidence. The sample was mounted on a heated aluminum block. The heater-thermocouple system varied the sample temperature from room temperature to about 400°C. The reflectivity and transmissivity signals are detected by photodiodes, which are connected to an oscilloscope. The components of the complex refractive index, $n(T)$, and $k(T)$, are then obtained from these measurements iteratively, by applying thin film optics, Fresnel-type relations. A thermal expansion rate of 3×10^{-6}°C^{-1} is assumed for the a-Si film. For the 0.2-μm-thick a-Si films used in this work, at the $\lambda = 0.752$ μm wavelength, and in the temperature range of 293–650 K, the following linear expressions are obtained:

$$n = n(T) = 4.0 + 1.30 \times 10^{-4} \quad (T - 293) \qquad (1a)$$

$$k = k(T) = 0.055 + 2.30 \times 10^{-4} \quad (T - 293) \qquad (1b)$$

Figures 1 (a, b) compare these expressions to measured complex refractive index components of 1.0-μm-thick amorphous silicon films, and to high-temperature optical data of bulk crystalline silicon (c-Si) samples (Jellison and Burke, 1986). Figure 2 shows the computed thin film reflectivity, $\mathcal{R}$, transmissivity, $\mathcal{T}$, and absorptivity, $\mathcal{A} = 1 - \mathcal{R} - \mathcal{T}$, for the 0.2-$\mu$m-thick film, as functions of temperature, at the $\lambda = 0.752$ μm wavelength. The results reproduce the steady-state measurements well. The estimated fractional uncertainty in the measurement of the components of the complex refractive index is ±2 percent for n and ±3 percent for k.

II.2 Transient Measurements. The layer is illuminated by a KrF ($\lambda = 0.248$ μm) excimer laser beam (Fig. 3). The probing diode laser beam ($\lambda = 0.752$ μm) is approximately normal onto the sample surface. The transmitted signal is captured by a fast photodiode and a digitizing oscilloscope. The photodiode has a rise time of 1 ns, and the oscilloscope sampling speed is 1 Gsample/s. Care is taken in the experiment to (a) minimize electrical noise interference by shielding the connecting cables to the oscilloscope and (b) collect all the transmitted light to the detector by using appropriate focusing lenses before the detector. The laser beam fluence, F, is determined by measuring the pulse energy using an energy meter. The estimated fractional uncertainty in the measurement of the laser pulse fluence is ±5 percent. The laser beam spot area on the sample surface, is measured to be about 0.5 cm^2.

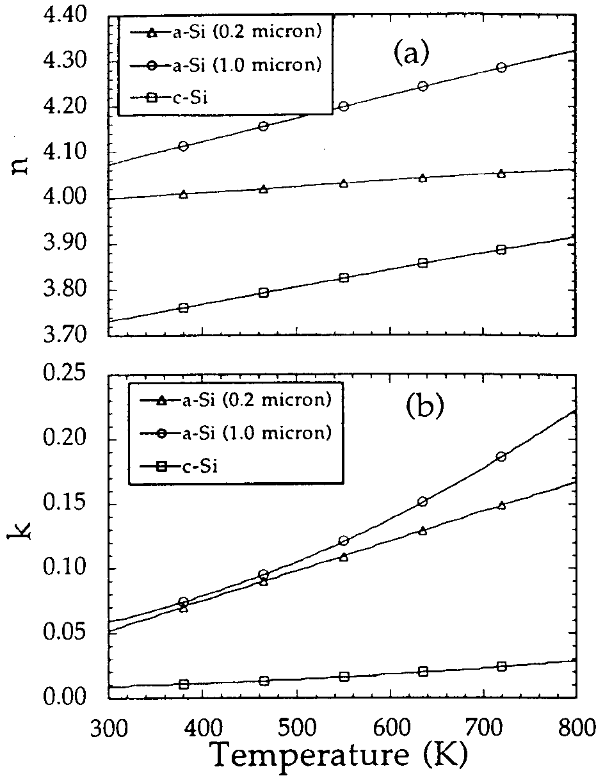

Fig. 1 Temperature dependence of the complex refractive index of 0.2 and 1.0-μm-thick amorphous silicon (a-Si) films and bulk crystalline silicon (c-Si) at $\lambda = 0.752$ μm: (a) real part; (b) imaginary part

II.3 Heat Transfer Modeling. The temperature profile penetration in the glass substrate is of the order of 1 μm. Thus, it may be assumed that the heat transfer at the center of the laser beam is essentially one dimensional. For the nanosecond time scales considered in this work, nonequilibrium and non-Fourier, thermal wave effects are negligible. For temperatures below the melting temperature, the conductive heat transfer in the solid silicon layer is given by:

$$\rho C_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(K \frac{\partial T}{\partial x}\right) + Q_{ab}(x, t) \qquad (2)$$

Nomenclature

$\mathcal{A}$ = absorptivity
C_p = specific heat
d = layer thickness
d_p = absorption penetration depth
d_{si} = semiconductor layer thickness
d_s = substrate thickness
E_g = optical energy gap
F = laser beam fluence
h = Planck's constant
i = imaginary unit
I = incident laser light intensity
K = thermal conductivity
k = extinction coefficient
$\mathcal{M}$ = transmission matrix
n = real part of refractive index
$\hat{n}$ = complex refractive index
N = number of layers in the thin film
Q_{ab} = power absorbed by the thin silicon layer
$\mathcal{R}$ = reflectivity
r = Fresnel reflection coefficient
t = time
t_l = length of laser pulse
t_p = time of occurrence of peak laser intensity
t_r = Fresnel transmission coefficient
T = temperature
$\mathcal{T}$ = transmissivity
T_∞ = ambient temperature
x = coordinate in the direction normal to the sample surface
α = thermal diffusivity
γ = absorption coefficient
λ = laser light wavelength
ν = light frequency
ρ = density

Subscripts

si = silicon
s = substrate

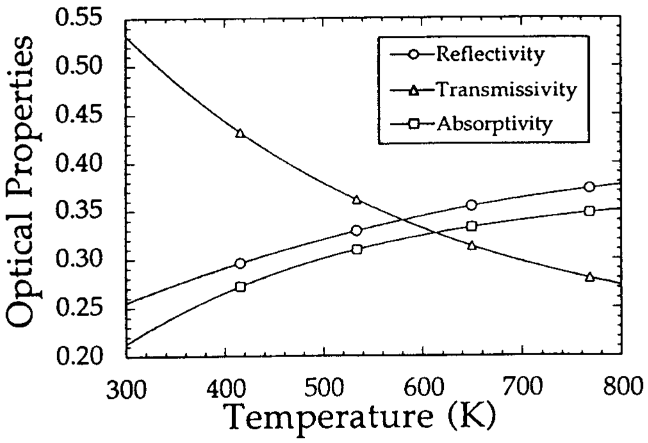

Fig. 2 Temperature dependence of the reflectivity, transmissivity, and absorptivity for the 0.2-μm-thick amorphous silicon film, at a wavelength λ = 0.752 μm

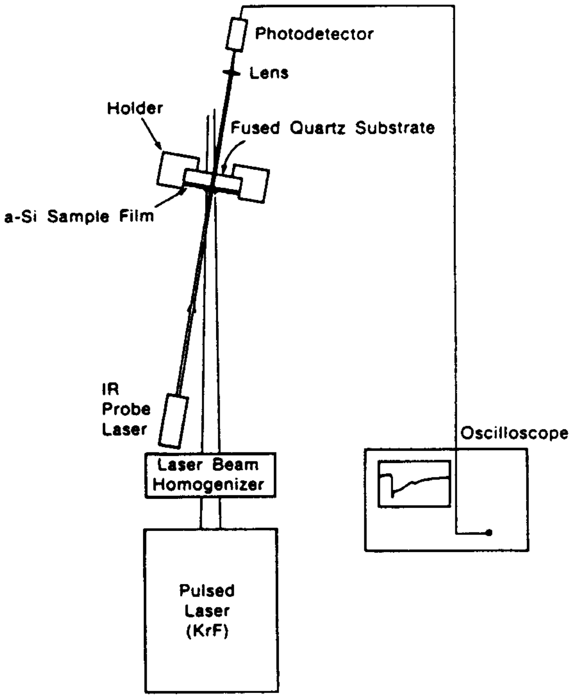

Fig. 3 Schematic of the experimental apparatus

In the above equation, x is the coordinate normal to the sample surface ($x = 0$), ρ is the density, T is the temperature, C_p is the specific heat for constant pressure, K is the thermal conductivity. The variation of the material thermal properties (Ong et al., 1986) is considered. The amorphous silicon complex refractive index at the KrF excimer laser light wavelength ($\lambda = 0.248$ μm) is taken as $\hat{n} = n - ik = 1.69 - i\,2.76$ (Palik, 1985). These optical constants yield an absorption coefficient, $\gamma = 1.40 \times 10^6$ cm^{-1}. The corresponding absorption penetration depth in the thin film, $d_p = 1/\gamma$, is of the order of seven nanometers, thus prohibiting any radiation absorption in the quartz substrate. The radiation photon energy for the KrF excimer laser light, $h\nu$, is much larger than the semiconductor optical energy gap, E_g, and the absorption coefficient, γ, is so high, that the temperature dependence of the material optical properties at $\lambda = 248$ nm is expected to be small (Wood and Jellison, 1984). The optical property data for crystalline silicon (c-Si), at elevated temperatures (Jellison and Modine, 1982) show little dependence of the optical properties on temperature in the UV range, but to the authors' knowledge, no detailed data for a-Si exist. It is thus reasonable to assume that no interference and internal refraction-reflection effects modify the local energy absorption in the thin film. Such effects have been shown to be important in pulsed ruby ($\lambda = 0.694$ μm), and frequency doubled Nd:YAG ($\lambda = 0.532$ μm) laser irradiation of silicon layers (Grigoropoulos et al., 1992), and also in absorption detection of defects in hydrogenated, amorphous silicon, (a-Si:H) films (Asano and Stutzmann, 1991). The laser energy is transferred to the lattice in times of the order of 10^{-12}–10^{-11} s, for both nanosecond and picosecond laser pulses. It is not expected that carrier diffusion and recombination effects play a significant role in nanosecond laser processing of semiconductor materials (Wood and Jellison, 1984). The energy absorption $Q_{ab}(x, t)$, follows an exponential decay in the material:

$$Q_{ab}(x,t) = (1-\mathcal{R})I(t)\gamma e^{-\gamma x} \tag{3}$$

In the substrate there is no energy absorption, and Eq. (2) is valid, with $Q_{ab} = 0$. Measurements of the laser pulse temporal profile, $I(t)$, have shown that the pulse fluence, F, is distributed in a triangular shape, with the pulse length, $t_l = 26$ ns, and the peak intensity occurring at $t_p = 6$ ns.

$$\begin{aligned} I(t) &= \frac{2Ft}{t_l t_p} && 0<t<t_p \\ I(t) &= \frac{2F(t_l-t)}{t_l(t_l-t_p)} && t_p<t<t_l \\ I(t) &= 0 && t_l<t \end{aligned} \tag{4}$$

Convection and radiation losses are negligible, due to the high incident laser intensities (of the order of 10^{11} W/m^2) and the short time scales considered in this problem. The temperature penetration in the structure is small, so that the bottom substrate surface remains at the ambient temperature, T_∞.

$$\left.\frac{\partial T}{\partial x}\right|_{x=0} = 0 \tag{5a}$$

$$T(x=d_{si}+d_s,\, t) = T_\infty \tag{5b}$$

Initially the structure is isothermal, at the ambient temperature:

$$T(x,0) = T_\infty \tag{6}$$

Continuity of both temperature and heat flux is applied at the film/substrate interface.

The heat conduction was solved numerically by an implicit finite difference algorithm. A depth of $x = 5$ μm is sufficient to contain the temperature field penetration. The temperature field in the semiconductor film induces changes in the a-Si film complex refractive index at the $\lambda = 0.752$ μm wavelength. These changes were accounted for in the picosecond irradiation of thin c-Si films (Lompre et al., 1983) by assuming an average temperature for the fitting of the measured optical properties. In this study, the semiconductor film is treated as a stratified multilayer structure (Jacobsson, 1965; Born and Wolf, 1980; Knittl, 1976) composed of N layers of varying complex refractive index. Wave optics effects are also considered in the substrate, which is represented by the $m = N + 1$ layer, and is transparent at the probing laser light wavelength, $\lambda = 0.752$ μm. Utilizing the formalism of the characteristic transmission matrix, the lumped structure reflectivity and transmissivity can be obtained. The mth layer of thickness d_m, which may be absorbing, having a complex refractive index $\hat{n}_m = n_m - ik_m$, is represented by the 2×2 matrix $\mathfrak{M}_m$, whose elements are complex:

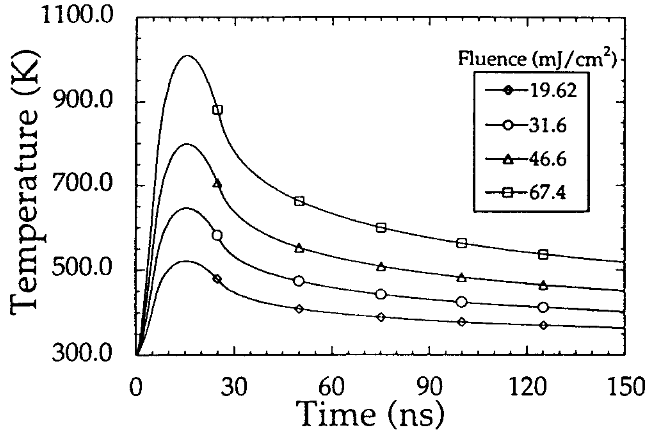

Fig. 4 Surface temperature histories for a 0.2-μm-thick amorphous silicon layer, irradiated with a KrF excimer laser (λ = 0.248 μm) for laser fluences F = 19.62, 31.6, 46.6 mJ/cm², 67.4 mJ/cm²; the laser pulse length t_l = 26 ns

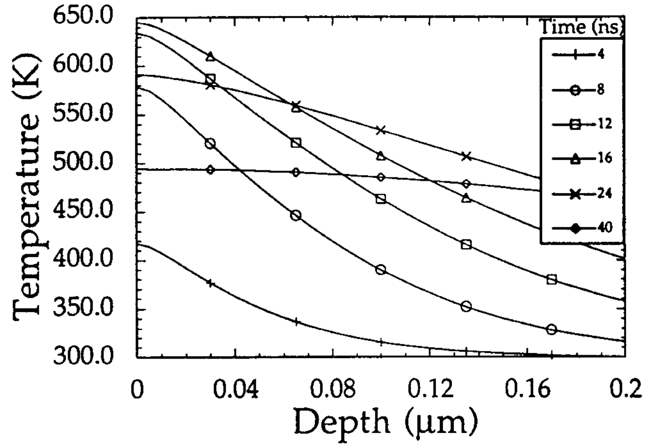

Fig. 5 Computed temperature profiles in a 0.2-μm-thick amorphous silicon layer, irradiated with a KrF excimer laser; the laser fluence F = 31.6 mJ/cm² and the pulse length t_l = 26 ns

$$\mathcal{M}_m = \begin{pmatrix} \cos\left(\frac{2\pi}{\lambda}\,\hat{n}_m d_m\right) & \frac{i}{\hat{n}_m}\sin\left(\frac{2\pi}{\lambda}\,\hat{n}_m d_m\right) \\ i\,\hat{n}_m \sin\left(\frac{2\pi}{\lambda}\,\hat{n}_m d_m\right) & \cos\left(\frac{2\pi}{\lambda}\,\hat{n}_m d_m\right) \end{pmatrix} \qquad (7)$$

The multilayer transmission matrix, $\mathcal{M}$, is:

$$\mathcal{M} = \prod_{m=1}^{N+1} \mathcal{M}_m \qquad (8)$$

The reflection and transmission Fresnel coefficients, r and t_r, are:

$$r = \frac{(\mathcal{M}(1,1)+\mathcal{M}(1,2)) - (\mathcal{M}(2,1)+\mathcal{M}(2,2))}{(\mathcal{M}(1,1)+\mathcal{M}(1,2)) + (\mathcal{M}(2,1)+\mathcal{M}(2,2))} \qquad (9a)$$

$$t_r = \frac{2}{(\mathcal{M}(1,1)+\mathcal{M}(1,2)) + (\mathcal{M}(2,1)+\mathcal{M}(2,2))} \qquad (9b)$$

The structure reflectivity and transmissivity in terms of r and t_r follow

$$\mathcal{R} = |r|^2 \qquad (10a)$$

$$\mathcal{T} = |t_r|^2 \qquad (10b)$$

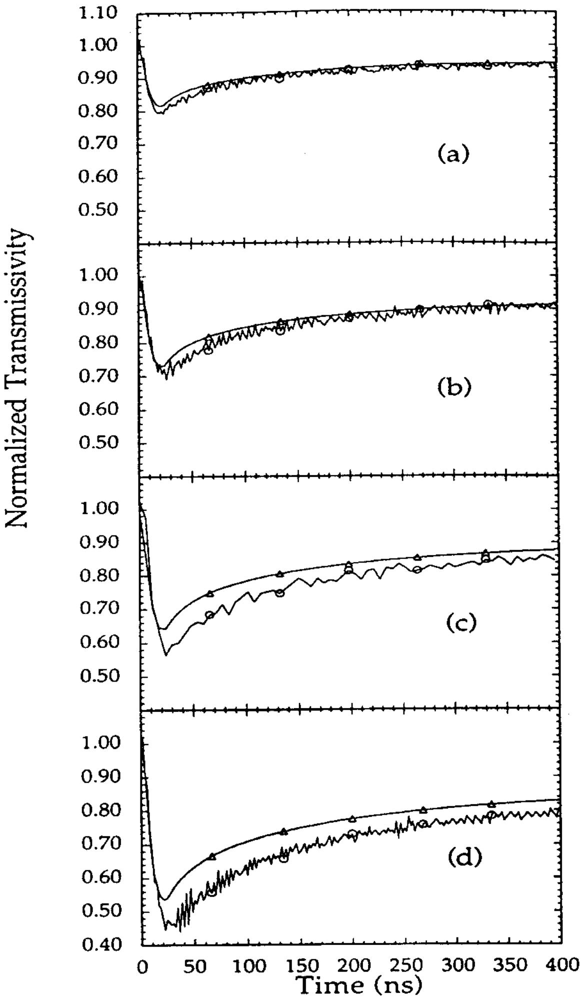

Fig. 6 Comparison between the numerical predictions (smooth solid line) and the experimental transient transmissivity measurement (noisy signal) for a 0.2-μm-thick amorphous silicon layer, irradiated with a KrF excimer laser (λ = 0.248 μm); results are shown as laser beam fluences, F: (*a*) 19.62 mJ/cm²; (*b*) 31.6 mJ/cm²; (*c*) 46.6 mJ/cm²; (*d*) 67.4 mJ/cm²; the pulse length, t_l = 26 ns

III Results

The amorphous silicon layer was irradiated by laser pulse fluences, F = 19.62, 31.6, and 46.6, and 67.4 mJ/cm². Some results obtained during these experiments have been reported by Park et al. (1992). Figure 4 shows predicted surface temperature histories for these fluences. The peak temperature occurs approximately at a time of 15 ns. The calculated temperature profiles across the thickness of the silicon layer are shown in Fig. 5 for a laser fluence, F = 31.6 mJ/cm². The experimental transmissivity signal was normalized by the steady-state value before heating. The predicted transmissivity was also normalized by the transmissivity at a temperature T_∞ = 300 K. This normalization is consistent with the measurement of the complex refractive index of the layer from reflectivity and transmissivity data. The comparison between experiment and model for the laser beam fluences, F = 19.62 and 31.6 mJ/cm², is shown in Figs. 6(*a*) and 6(*b*). It can be stated that the model accurately captures the experimental trend. The calculated peak temperature for the fluence, F = 31.6 mJ/cm², is approximately 650 K (Fig. 4). It is recalled that the thin film optical properties have been measured up to this temperature range. At higher fluences, the agreement is not as close (Figs. 6*c*, *d*), though both the time occurrence of the peak temperature and the cooling trend are predicted quite well.

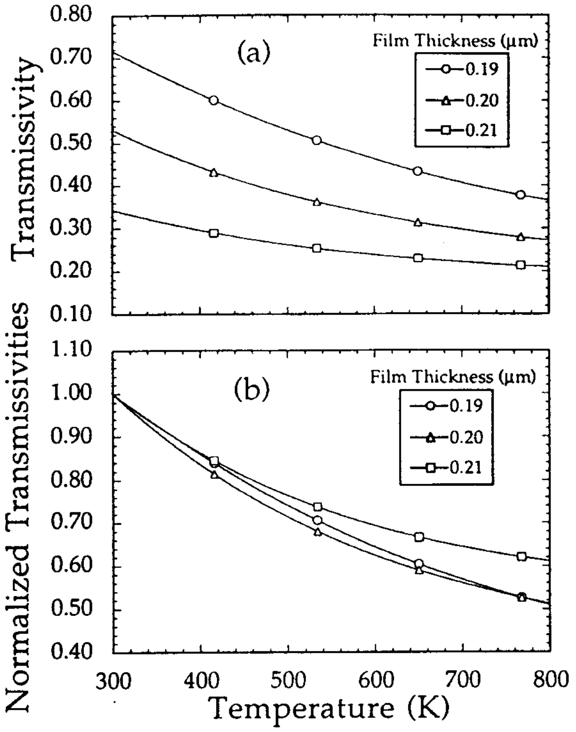

Fig. 7 Thickness dependence of the thin film transmissivity: (*a*) absolute values; (*b*) normalized values

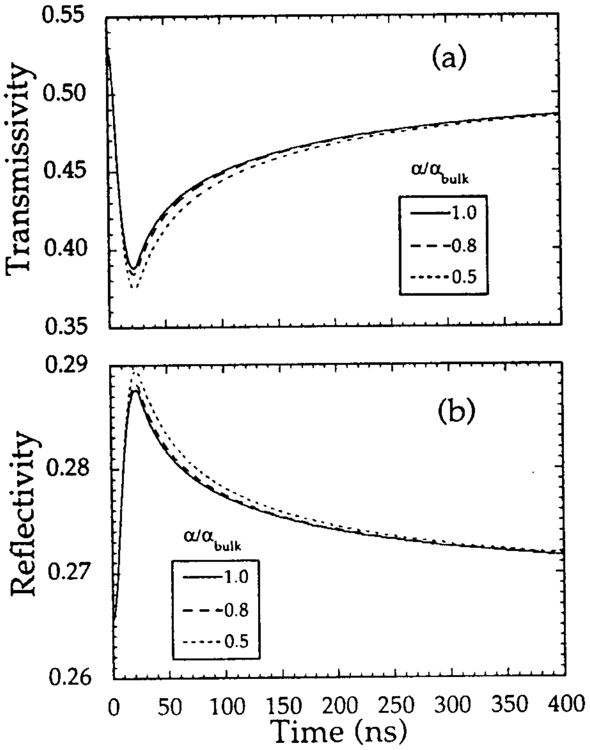

Fig. 8 Effect of variations of the thin film thermal diffusivity on the predicted: (*a*) reflectivity; (*b*) transmissivity; the excimer laser beam (λ = 0.248 μm) fluence, F = 31.6 mJ/cm^2, the pulse length, t_l = 26 ns

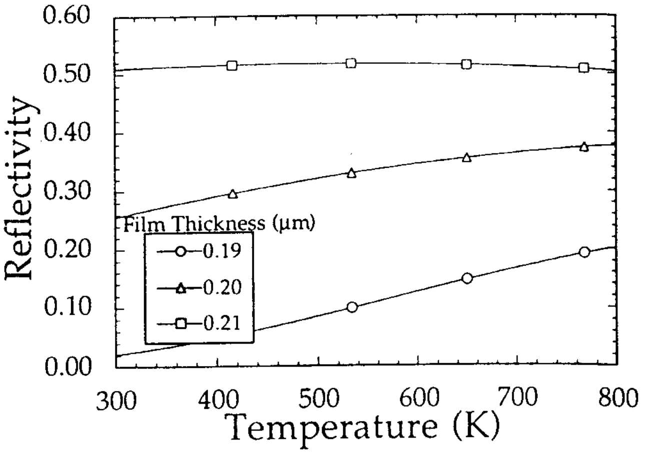

Fig. 9 Thickness dependence of the film reflectivity for the λ = 0.752 μm wavelength

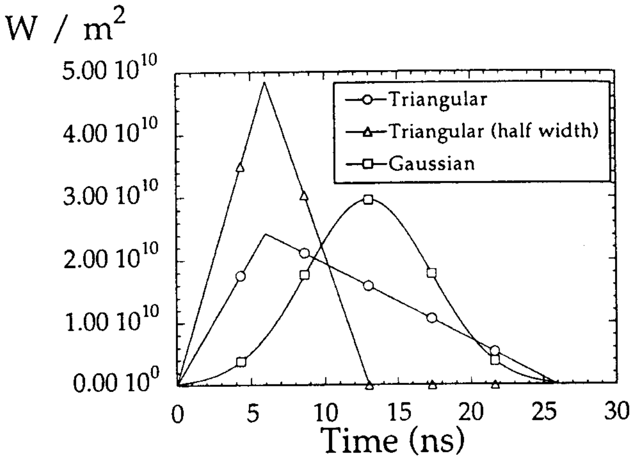

Fig. 10 Different laser pulse intensity distributions for a total fluence, F = 31.6 mJ/cm^2; the laser wavelength λ = 0.248 μm

Variations of the thin film thickness by ±0.01 μm, cause absolute transmissivity departures of about 40 percent from the values that correspond to the nominal 0.2 μm amorphous silicon layer thickness used in this work (Fig. 7*a*). The use of the normalized transmissivity measurements significantly reduces this deviation to about 10 percent at a temperature T = 650 K (Fig. 7*b*). The uncertainty in the normalized transmissivity measurement is estimated to be ±0.04. Numerical computations have shown that the magnitudes of the transient reflectivity and transmissivity are not very sensitive to reduction of the thin film thermal diffusivity by 50 percent (Figs. 8*a*, *b*). On the contrary, the thin film steady-state reflectivity measurement is quite sensitive to film thickness variations. Figure 9 shows that the reflectivity for a 0.21 μm film thickness slowly decays with temperature, whereas it increases almost linearly for the 0.20 μm and 0.19 μm film thicknesses. On the contrary, the decay of the transmissivity signal with temperature is monotonically behaved, with decreasing values for increasing thicknesses. The long-term temperature field depends mainly on the substrate thermal properties. The pulse intensity temporal profile used in these calculations (Eq. (4)) was fitted to experimentally acquired data. This profile may vary from pulse to pulse. Numerical experiments with different laser intensity temporal profiles (Fig. 10) corresponding to a laser beam fluence of 31.6 J/cm^2 were conducted to estimate the effect of the related experimental uncertainty. Whereas much shorter pulses (t_l = 13 ns) raise the temperature faster, intensity distributions over the measured pulse length, t_l = 26 ns, produce little change to the calculated reflectivity and transmissivity signals (Figs. 11*a*, *b*).

IV Conclusions

A method for monitoring the transient temperature field in

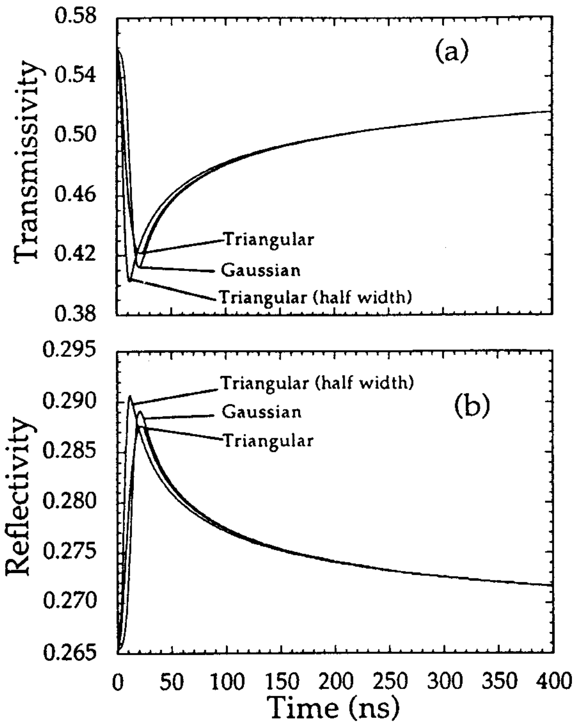

Fig. 11 Calculated probing laser (λ = 0.752 μm) (*a*) transmissivity and (*b*) reflectivity signals for the laser beam intensity profiles given in Fig. 10

pulsed excimer laser irradiation of thin a-Si films has been presented. An infrared (λ = 0.752 μm) diode laser has been used as a probe for in-situ optical transmission measurements. The temperature dependence of the components of the complex refractive index of the a-Si films has been determined at this wavelength in the temperature range from room temperature to about 400°C using static reflectivity and transmissivity measurements. The heat transfer in the excimer laser irradiated a-Si film has been calculated numerically. The transient transmisssivity for the probe laser light has been calculated by assuming a variation of the a-Si material complex refractive index according to the statically measured optical property data. The predicted and experimental transmissivity signals were in excellent agreement for the excimer laser fluences that produce temperatures within the range of the static optical property measurement. The optical transmission measurements presented in this work accurately capture the transient temperature field in excimer laser irradiated amorphous silicon films.

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