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Analysis of wavelength influence on a-Si crystallization processes with nanosecond laser sources

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ABSTRACT

In this work we present a detailed study of the wavelength influence in pulsed laser annealing of amorphous silicon thin films, comparing the results for material modification at different fluence regimes in the three fundamental harmonics of standard DPSS (diode pumped solid state) nanosecond laser sources, UV (355 nm), visible (532 nm) and IR (1064 nm).

The crystalline fraction (% crystalline silicon) profiles resulted from irradiation of amorphous silicon thin film samples are characterized with MicroRaman techniques. A finite element numerical model (FEM) is developed in COMSOL to simulate the process. The crystalline fraction results and the local temperature evolution in the irradiated area are presented and analyzed in order to establish relevant correlation between theoretical and experimental results.

For UV (355 nm) and visible (532 nm) wavelengths, the results of the numerical model are presented together with the experimental results, proving that the process can be easily predicted with an essentially physical model based on heat transport at different wavelengths and fluence regimes. The numerical model helps to establish the optimal operation fluence regime for the annealing process.

1. Introduction

Pulsed laser crystallization has been widely applied to formation of polycrystalline silicon films and has been introduced to fabrication process of polycrystalline silicon thin film transistors (poly-Si TFTs) and solar cell applications [1–3]. Crystallization and grain growth technique of thin film silicon are among the most promising methods for improving efficiency and lowering cost of solar cells.

A major advantage of laser crystallization and annealing over conventional heating methods is its ability to limit rapid heating and cooling to thin surface layers. This is mainly controlled by the pulse duration time and the absorption depth of the laser light used in the material [4].

Laser energy is used to heat the amorphous silicon thin film, melting it and changing the microstructure to polycrystalline silicon (poly-Si) as it cools. Phase change from a-Si to poly-Si depends on the absorbed energy from the incident laser-pulse. Pulsed excimer lasers such as XeCl and KrF have been widely used for annealing of a-Si films [5–9] due mainly to the strong absorption of UV light in Silicon. However, regarding some limitations of these lasers, numerical and experimental analysis with other wavelengths, like CW diode lasers (808 nm) and pulsed Nd:YAG lasers (532 nm, 355 nm and 1064 nm) have been recently studied [10–13].

Regarding the proved advantages of DPSS laser sources in production plants, the authors think that experimental and numerical comparative study on local crystallization using the three fundamental harmonics of these laser sources, UV (355 nm), visible (532 nm) and IR (1064 nm), could give useful information for the process implementation in solar cell industry.

In the present work, we report experimental analysis of a-Si local crystallization by a single laser pulse irradiation in these three wavelengths, using nanosecond time regime. Numerical simulation is used to study the energy absorption, phase change and temperature evolution regarding the nanosecond laser pulse interaction on a-Si. The samples are irradiated with different energy regimes in order to evaluate the effect of the laser fluence on the a-Si crystallization. These experimental results are compared with the data obtained from the numerical simulation. We present a different approach for the analysis of the data, explaining some new correlation between numerical and experimental results which can be used to establish the operation range of the process. And as result of this, it is proposed the use of a simple heat transfer model to predict the operation range for DPSS laser sources in the annealing process.

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2. Theory

The incident energy of the laser pulse is partially absorbed rising the temperature of the silicon layer. The absorbed light simultaneously excites the electronic states of silicon. The energy of the excited states is relaxed to lattice vibration states within the time on the order of 10^{-12} s. In the case of ns-order pulsed laser irradiation, lattice heat is, therefore, the most important interaction [7]. The heat energy generated at the surface region caused by light absorption diffuses into interior regions of the thin film. When the material is melted and cools down then solidifies to poly-Si phase.

The heating process in the a-Si thin film is governed by the general heat transfer equation:

$$\rho(T)C_p(T)\frac{dT}{dt} = \nabla[k(T)\nabla T] + S \quad (1)$$

where T is the temperature, ρ is the material density, C_p the specific heat and k the thermal conductivity, when a heat source S is considered.

The specific heat, density and thermal conductivity of the material ($C_p(T)$, $k(T)$) are assumed temperature dependent. The source term (S) in Eq. (1) refers to the incident laser beam in the model, which is assumed to be Gaussian both for the irradiance distribution and the time evolution. The distribution in depth follows the Beer–Lambert law. So, the whole source term $S(r, z, t)$ can be described by the following equation:

$$S(r, z, t) = P_i(r, t)[(1 - R(T))\alpha(T)\exp(-\alpha(T)|z|)] \quad (2)$$

where $\alpha(T)$ is the material absorption coefficient, $R(T)$ the surface reflectivity and P_i the incident laser power (W/m^2) given by the product of the corresponding exponential r and t dependent functions, as follows:

$$P_i = P_p\left(\frac{2}{\pi r_w^2}\right)\exp\left[-2\left(\frac{r}{r_w}\right)^2\right]\exp\left[-\left(\frac{t - t_0}{\tau/2}\right)^2\right] \quad (3)$$

where P_p is the peak power, r_w is the beam radius, that is, the half-width at $1/e^2$ of the laser intensity along the radial direction, t_0 the time shift and τ is the time-width of the laser pulse.

The Gaussian fluence profile is given by:

$$F(r) = F_p\exp\left[-2\left(\frac{r}{r_w}\right)^2\right] \quad (4)$$

where F_p is the peak fluence, that is, the value of the function F in the center of the laser pulse.

The 2D non-linear heat transfer equation is difficult to solve by analytical approach, so finite element method (FEM) was used to solve it numerically by means of COMSOL multiphysics [14].

2.1. COMSOL multiphysics implementation

According to the mathematical model described before, a heat transfer time-dependent study is applied using *Heat Transfer in Solids* equations with a 2D-axisymmetric model.

Regarding the symmetry and the boundary conditions of the problem, the model is restricted to two dimensions, r and z in cylindrical coordinates system, where r is the distance to the center of the laser pulse and z the depth dimension in the material. The model sample consists of one 2 μm-thick a-Si layer on c-Si substrate. Due to the thickness of the a-Si layer and the maximum laser fluence involved, the substrate is virtually not affected during the process. Therefore the conclusion remains valid when using glass instead of c-Si for the substrate.

The specific heat of a-Si, $C_p(T)$, is replaced by $(C_p(T) + \delta L_m)$ in the heat transfer equation, where L_m is the latent heat of melting and δ is a Gaussian function given in Table 1. The additional term δL_m takes into account the phase change effect within the temperature range of this study [11]. Analogous treatment is made in case of liquid–vapor phase change at the corresponding temperature T_v.

The boundary conditions applied include initial room temperature, thermal insulation for right, left and bottom boundaries, and finally, surface-to-ambient radiation and convective cooling at the top surface. Absorption coefficient $\alpha(T)$ and surface reflectivity $R(T)$ are taken regarding the wavelength of each laser source. Thermal properties and relevant parameters used for the simulation are listed in Table 1 [10,15,16].

The meshing of the model accounts for the laser pulse irradiation area. Therefore a finer meshing is used in the area closest to the center ($r = 0$) and a coarser meshing is used for the substrate.

The simulation time is 0.5 μs with a maximum time-step of 1 ns. PARDISO direct solver algorithm is used to solve the linear system [17]. The numerical simulation gives the temperature evolution in the sample. The material is heated by a single laser pulse resulting in a fast increase of the a-Si surface temperature. When T_m (1420 K) temperature is reached, the surface begins to melt. Then, the temperature of the melting layer will remain constant until the energy needed for phase change is absorbed. The crystallization of a-Si can take place not only via melting, but also via solid phase crystallization (SPC), nevertheless, the SPC mechanism is not considered in our numerical model.

Depending on the laser fluence, the vaporization temperature T_v can be reached at the center of the irradiated area. In these cases ablation effects are expected and the annealing process becomes ineffective. As discussed later, the numerical model help to establish the fluence range needed to assure the melting and crystallization without damage or ablation of the silicon surface.

3. Experimental procedure

Two different PECVD systems were used for depositing a-Si:H thin films simultaneously onto glass substrates and onto crystalline silicon wafers from the decomposition of silane (SiH4). The thickness (2 μm) was selected in order to obtain the same amorphous degree in the a-Si deposited on glass and on c-Si and in order to inhibit the pulsed laser induced epitaxial (PLIE) process. Laser irradiation was performed using different fluence regimes in the three fundamental harmonics of standard DPSS laser sources, UV (355 nm), visible (532 nm) and IR (1064 nm), with Gaussian irradiation profile.

The laser sources used in this study were three solid state DPSS laser systems: Nd:YAG Spectra Physics Navigator (30 ns pulse width at 10 kHz repetition rate) for IR (1064 nm) irradiation, Nd:YVO4 Spectra Physics Explorer and Hippo systems (15 ns, and 12 ns pulse width at 50 kHz repetition rate) with fundamental frequency in IR (1064 nm), doubled and tripled to green (532 nm) and UV (355 nm) respectively.

In order to control the number of pulses, lines of irradiated spots were carried out using a scanner. No overlapping of pulses was produced so as to study the crystallization by a single pulse. The morphology of the irradiated area was characterized by confocal laser scanning microscopy (Leica DCM3D).

The structural properties of the irradiated surface were studied by micro-Raman spectroscopy (Renishaw, in Via Raman microscope). The measurements were performed in the range from 200 cm^{-1} to 850 cm^{-1} with a 100X optical microscope objective, scanning the irradiation area with an incident measurement spot of 1 μm diameter. The excitation optical source consisted of an Ar+ laser tuned at the wavelength of 514 nm.

Non irradiated samples show the usual Raman spectrum from the amorphous silicon with a broad band centered at 480 cm^{-1} corresponding to the TO phonon mode [18]. However, the irradiated