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ArF Excimer Laser Annealing of Polycrystalline Silicon Thin Film

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1. Introduction

Crystallization of amorphous silicon (a-Si) using excimer laser annealing (ELA) has been reported since 1994 by Watanabe group. It is known as the best method to fabricate a good poly-silicon because it can heat the film up to the melting point and, at the same time no thermal damage occur into the glass substrate (Carluccio et al., 1997; Matsumura and Oh, 1999). ELA technique is widely used to increase the grain size and changes the microstructure of polysilicon thin film which is the most important characteristics of excellent built-in polysilicon devices (Palani et al., (2008). The principle advantage of excimer lasers is the strong absorption of ultraviolet (UV) light in silicon beside having larger beam size and high energy density than other laser light sources (Watanabe, et al. 1994). Another major advantage of the excimer lasers is it low-temperature polysilicon annealing. Excimer laser with 308 nm wavelength for example has been reported can transform 50-nm-thin layers of amorphous silicon into high-quality polycrystalline silicon with greatly enhanced electron mobility, for use in flat-panel displays for mobile phones and flat-screen televisions (Delmdahl, 2010). In the low-temperature annealing of polysilicon, excimer lasers with UV output energies of over 1 J per pulse and output powers of 600 W are used to manufacture liquid-crystal and organic LED backplanes at a rate of 100 cm² s⁻¹. A new VYPER/LB750 line beam annealing system enables volume production of low-temperature polysilicon (LTPS) on large generation 6 glass panels. LTPS is the key material for high-resolution liquid crystal displays (LCDs) and organic lightemitting diode (OLED) displays for smartphones, tablet PCs and TVs.

Today, the most prominent applications of excimer lasers are in semiconductor chip manufacturing. This relies on the deep-UV emission wavelength of 193 nm, with semiconductor chip manufacturing capitalizing on the optical resolution of excimer lasers. For example, high repetition rate line-narrowed excimer laser models with 10 mJ per pulse, pulse frequencies of up to 6 kHz and narrowed bandwidths of 0.35 pm are used in advanced photolithography to produce computer chips with feature sizes of 45 nm.

Ongoing miniaturization in microelectronics and the trend towards thin-film technologies demands increasing lateral resolution and selective machining. Functional structures and active layers are often only tens of nanometres thick and has to be annealed, patterned and removed in a selective manner without damaging underlying layers or substrates. Because excimer lasers provide the shortest wavelength of all laser technologies, they will continue

to have a crucial role in many industries over the next decade. In years to come we will see high-power applications of excimer lasers, with output energies reaching 2 J and output power levels of 1,000 W and above (Delmdahl, 2010).

The aim of this chapter is to demonstrate the crystallization of polysilicon thin film by excimer laser annealing. Various aspects of excimer laser annealing process will be covered in this chapter including principle and experimental results. The definition and the history of excimer laser annealing are described in introduction section. Since excimer laser is the key component in this annealing process, the detail regarding excimer laser system is discussed. Mechanism of laser annealing is explained under third section of laser ablation which includes the ablation phenomena and photochemical mechanism. The detail sample preparation using solid phase crystallization technique is described under section 4. Section 5 will give information regarding polycrystalline silicon. The basic annealing process relies on transformation temperature in which three important parameters, comprising critical point, recalescence effect and super lateral growth energy are desired to be identified. The crystallization of silicon thin film is characterized via atomic force microscope analysis. Finally the excimer laser annealing on silicon thin film is summarized in section 8.

2. Excimer laser

2.1 History and application

An excimer laser is a form of ultraviolet laser which is commonly used in the production of microelectronic devices (semiconductor integrated circuits or "chips"), eye surgery, and micromachining. The excimer laser was invented in 1970 by Nikolai Basov, V. A. Danilychev and Yu. M. Popov, at the Lebedev Physical Institute in Moscow, using a xenon dimer (Xe₂) excited by an electron beam to give stimulated emission at 172 nm wavelength. A later improvement, developed by many groups in 1975 was the use of noble gas halides (originally XeBr).

The excimer laser typically uses a combination of a noble gas (argon, krypton, or xenon) and a reactive gas (fluorine or chlorine). Under the appropriate conditions of electrical stimulation and high pressure, a pseudo-molecule called an excimer (or in the case of noble gas halides, exciplex) is created, which can only exist in an energized state and can give rise to laser light in the ultraviolet range.

Laser action in an excimer molecule occurs because it has a bound (associative) excited state, but a repulsive (dissociative) ground state. This is because noble gases such as xenon and krypton are highly inert and do not usually form chemical compounds. However, when in an excited state (induced by an electrical discharge or high-energy electron beams, which produce high energy pulses), they can form temporarily-bound molecules with themselves (dimers) or with halogens (complexes) such as fluorine and chlorine. The excited compound can give up its excess energy by undergoing spontaneous or stimulated emission, resulting in a strongly repulsive ground state molecule which very quickly (on the order of a picosecond) dissociates back into two unbound atoms. This forms a population inversion. Excimer lasers are usually operated with a pulse repetition rate of around 100 Hz and a pulse duration of ~10 ns, although some operate at pulse repetition rates as high as 8 kHz and some have pulsewidth as large as 30 ns.

Excimer lasers are widely used in high-resolution photolithography machines, one of the critical technologies required for microelectronic chip manufacturing. Current state-of-theart lithography tools use deep ultraviolet (DUV) light from the KrF and ArF excimer lasers with wavelengths of 248 and 193 nanometers (the dominant lithography technology today is thus also called "excimer laser lithography), which has enabled transistor feature sizes to shrink below 45 nanometers. Excimer laser lithography has thus played a critical role in the continued advance of the so-called Moore's law for the last 20 years.

The most widespread industrial application of excimer lasers has been in deep-ultraviolet photolithography, a critical technology used in the manufacturing of microelectronic devices (i.e., semiconductor integrated circuits or "chips"). Historically, from the early 1960s through the mid-1980s, mercury-xenon lamps had been used in lithography for their spectral lines at 436, 405 and 365 nm wavelengths. However, with the semiconductor industry's need for both higher resolution (to produce denser and faster chips) and higher throughput (for lower costs), the lamp-based lithography tools were no longer able to meet the industry's requirements. This challenge was overcome when in a pioneering development in 1982, deep-UV excimer laser lithography was proposed and demonstrated at I.B.M. by Kanti Jain. With phenomenal advances made in equipment technology in the last two decades, and today microelectronic devices fabricated using excimer laser lithography totaling \$400 billion in annual production, it is the semiconductor industry view that excimer laser lithography has been a crucial factor in the continued advance of Moore's law, enabling minimum features sizes in chip manufacturing to shrink from 0.5 micrometer in 1990 to 32 nanometers in 2010. This trend is expected to continue into this decade for even denser chips, with minimum features approaching 10 nanometers. From an even broader scientific and technological perspective, since the invention of the laser in 1960, the development of excimer laser lithography has been highlighted as one of the major milestones in the 50-year history of the laser.

For micro- and nano-technologies, laser micromachining is currently used in a large number of R&D and industrial applications. The range of applications to which laser methods are applied is continuously expanding, supported also by the development of novel processing techniques. Over the last decades the excimer laser has obtained the key position among lasers in various sectors of micromachining. Excimer lasers have developed into powerful manufacturing tools mainly because of two reasons: (i) The short wavelengths of the excimer laser offers excellent quality of machining and a great versatility in features which can be produced. (ii) The progress in basic excimer laser technology has made the excimer lasers to reliable machines suitable for the industrial environment.

There are certainly not many types of lasers which have found such broad markets as the excimer laser. Over the last years the main growth results from increasing industrial use followed by medical applications while new sales into R&D applications stay nearly constant. Today the largest known industrial applications of excimer lasers are (i) based on micromaching of different materials as polymers, ceramics and glasses, applied for example in the production of ink jet cartridges by drilling the nozzles, (ii) excimer laser radiation is being used for changing the structure and properties of materials as oxides, silicon or glass in bulk or thin films, as applied for the production of active matrix LCD monitors, fiber Bragg gratings in telecommunication, and high temperature superconducting films, (iii) employing the excimer laser as "short wavelength light bulb" in optical microlithography

for the production of computer chips with critical dimensions below 0.25 μ m (the largest homogeneous market for excimer lasers).

All the widespread applications of excimer lasers in micromachining and medicine are based on the early use of excimer lasers leading to the discovery of the ablation of materials under intense illumination with ultraviolet laser pulses by R. Srinivasan.

2.2 Crystallization source

The source of energy used for crystallization in this work is an argon fluoride excimer laser. The excimer laser generates deep ultraviolet light measuring 193 nm when high voltage energy is discharged into a cavity containing a mixture of a rare gas argon and a halogen (i.e., fluoride) Almost, 90% or more of the mixture contain buffer rare gas (typically helium) that does not take part in the reaction. This gas mixture is pre-ionized by a set of electrodes before a high-voltage current (about 30,000 eV is applied) resulting in formation of highly unstable rare-gas halide molecules, which rapidly dissociate, emitting UV light whose wavelength is determined by the particular gas mixture chosen. Excimer lasers use in this particular project is comprised of a mixture of argon and fluoride gas. The term excimer is derived from the two words "excited" and "dimer" which are used to describe the reaction in which the laser transfer energy through an ultraviolet beam of light (Schneider, 1998). All emitted powerful pulses lasting in 10 nanoseconds at wavelength of ultraviolet region of 193 nm.

2.3 Excimer laser structure

The gain of excimer lasers is extremely high, so the output is superradiant. A single rear mirror is employed and an output coupler of 4 to 8% which transmit in the region of interest (which is in UV region) is used (Figure 1). Divergence of the beam is reduced when a full optical cavity is used, and alignment is easy since the laser operates even when cavity mirrors are completely misaligned. The beam is rectangular profile. Quartz cannot be used as an output coupler for argon fluoride (ArF) Excimer laser since fluorine attacks the material, hence magnesium fluoride are used to replace quartz as it absorbs less UV at most wavelength compare to quartz (Csele, 2004). In discharge excitation, electric current flows through the laser medium, typically ranging from a kilovolt (kV) to over tens of kilovolts deliver energy to the laser gas (Hecht, 1992).



Fig. 1. An electrical discharge exciting a gas laser (Hecht, 1992)

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Excimer laser is excited by passing a short, intense electrical pulse through a mixture of gases containing the desired rare gas and halogen. However, the molecule were found more stable when it is on excited state and become less stable on ground state and this properties can be found on argon fluoride (ArF), krypton fluoride (KrF₂) and xenon chloride (XeCl₂). This phenomenon could be explained in detail by referring on Figure 2.



Fig. 2. Excimer laser potential energy curve.

Figure 2 shows a graph of potential energy curve as a function of atomic distance between the molecules. Generally, the potential energy is minimized at equilibrium molecule distance. However, for ArF, KrF₂ and XeCl₂ material, these properties did not occur. They are found not stable even when they are in their ground state. Thus, excitation process cannot directly be done on ground state. Hence, indirect excitation in discharge system is employed. For example, in ArF excitation process, electron reaction process is as followed,

$$e + F^2 \to F^- + F \tag{1}$$

The negative ion produced will combine with positive ion to produce excited molecule

$$Ar^+ + F^- \rightarrow (ArF)^*$$
 (2)

This reaction can produce an efficient excited dimer molecule. The unstable molecules at ground state leads to a small number of population. Hence, it causes the population inversion to easily happen and laser transition will trigger. The nature of dimer material that is not stable on their ground state causes the atom to easily breaks-up (Shah, 2009). Because of the ground state essentially does not exist, there is a population inversion as long as there are molecules in the excited state. This process is performed again and again; this is how the pulse excimer laser is trigger out.

2.4 External triggering

An argon fluoride ArF excimer laser, manufactured by GAMLaser model EX5/200-110, is employed as a source of ablation. The wavelength of the laser is 193 nm with pulse duration

of 10 ns and repetition rate which can be varied from 20 Hz to 200 Hz. The beam size of the laser output when it hits the sample is 4 mm x 6 mm. The size of the beam also depends on the output energy of the laser beam (GAMLaser Inc., 2003).

The excimer laser is internally operated via the aids of computer using 32 bit Windows software (GAMLaser Inc., 2003). The laser parameters including the frequency, high voltage and number of pulse can be commendable according to the requirement of the experiment. The output energy of the laser can be varied by manipulating the capacitor voltage from 10 kV to 15 kV. The dose number of laser exposure is not limited; however, the minimum number of pulses can be set at 100 pulses. However, to trigger the number of pulse as minimum as 1 pulse, a Sony Tektronix arbitrary function generator model AFG 310 is employed. Figure 3 shows photograph of the excimer laser, which coupled to an arbitrary function generator in the real field. In this particular experiment, only a few pulses of laser exposure are required to obtain the desired optical surface treatment.



Fig. 3. An excimer laser connected to a function generator for external triggering.

The function generator can be used to control the number of pulses lower than 100 pulses. As an external trigger unit, the function generator is set up at '5 Volts', operated at 'pulse' output function, and with 'burst' mode (Sony Tektronix Inc., 2003). The frequency of the function generator is synchronized with the excimer laser system.

2.5 Excimer laser annealing

A GAM LASER Argon Fluoride ArF excimer laser is used to anneal the hydrogenerated silicon thin film. The ArF excimer laser generates ultraviolets laser light at 193 nm with 10 ns pulse duration. The lasers parameters such as number of pulses, repetition rates and pumping voltages can be controlled via a computer program. In this experiment, the laser capacitor voltage is kept constant at 12 kV with repeatition rate of 20 Hz, operating at room temperature. The gas pressure is set at 3200 Torr.

The excimer laser annealing experimental setup is shown in Figure 4. The laser energy (in mJ) for each laser pulses is measured using 13PEM001 Broadband power and energy Meter.

The laser is controlled externally using a AFG310 function generator for obtaining laser pulse below than 100 pulses. In this present works, the number of pulse is varied from one to ten pulses. The operational voltage of the function generator is set at 5 Volts 'pulse' output function, and with 'burst' mode. The laser pulse is generated after 2.5 µs upon application of the trigger pulse rising edge with DC charging is operating in external trigger mode. The frequency of the function generator and the excimer laser system is fixed at 20.0 Hz. The silicon copper thin film placed on *x* and *y* translational stage in and vertical position with respect to the excimer laser beam axis. The number of pulses of excimer laser beam exposed on the thin film target is, controlled by the external trigger unit.

Maximum laser performance can be achieved by aligning the laser mirrors. The mirrors are alligned via the guidance of a visible of helium-neon He-Ne laser. The He-Ne laser is aligned to coaxis with excimer laser beam. The excimer laser is optimized by adjusting the output coupler via the aids of power/energymeter. A bright and rectangular dimension of beam spot is recorded permanently using beam profiler. The images are arranged in order of 3D, 2D and the bright beam spot in the real field such as shown in Figure 5 a, b and c respectively. The dimension of excimer laser beam spot is 4 × 6 mm². The a-Si films is recrystalized at different energy densities. Finally, nanostructure of the crystallized silicon is examined via atomic force microscope AFM.



Fig. 4. The schematic diagram of excimer laser annealing process.



Fig. 5. Beam profile and beam spot of excimer laser. a. 3D beam profile, b. 2D beam profile, and c. The beam spot in the real field.

3. Laser ablation

In laser annealing process the main key for its successful work is the ablation process between optical material and UV light from argon fluoride (ArF) excimer laser. In order to understand the laser-matter interaction, it is better to understand the fundamental mechanism of laser ablation and optical material behavior with respect to the ultraviolet (UV) light illumination. In this section, the photochemical process occurred during ablation process will be discussed.

Ablation is usually described in terms of physical mechanism such as vaporization and shock effects. It is often performed in vacuum or air. Examples of ablation application include polymer ablation, removal of thin metal films from an insulating substrate, and deposition of high-temperature superconducting thin films by ablation of bulk targets (Brannon, 1997).

The laser ablation process requires an intense UV light (λ = 193 nm- 351 nm) produced by an excimer laser. Because of the high absorption of UV light and relatively poor conductivity of many materials, particularly silicon, the energy is deposited in a very thin layer. Whenever the energy density or fluence exceeds the ablation threshold value for the material, chemical bonds are broken, fracturing the material into energetic fragments. The fragments are atoms, groups of atoms, ions and electrons. Because the fragments leave the reaction zone as an energetic gas and solid debris, the ablation process resembles explosive evaporation of the material (Speidell, 1997).

Generally, there are two classes of laser ablation mechanisms: thermal and electronic (nonthermal). Thermal processes rely on an intense laser pulse to very rapidly heat a surface at rates of the order of 10¹⁰ K/sec. These processes can produce expansion and vaporization from solid and melted regions.

Laser ablation of given material can involve both thermal and non-thermal mechanisms. For example, during the interaction of the leading edge of the laser pulse with the solid, nonthermal processes may predominate because significant heating has not yet occurred. As the rest of the pulse is absorbed, the temperature rise rapidly and thermally activated mechanism may commence. It should be noted that laser excitation of a strongly absorbing solid will always result in substantial heating regardless of the ablation mechanism. Although non-thermal process may occur, there is no way to "turnoff "the thermal processes. The dominant process is ultimately determined by the detailed chemistry and physics of the solid.

3.1 Photochemical

In the photochemical processes the photon absorption event break a chemical bonding within a molecule forming molecules with smaller number of atoms. The resulting photoproducts occupy a large volume and create pressure inside the irradiated volume that can then convert to the translational energy of ablation (Sato et al., 2001). During this process, thermal and mechanical damage to the surrounding material is minimized, therefore achieving more precise control over the ablated region.

There has been much un-certainty and debate over the fundamental ablation mechanisms in material. Several photochemicals and photothermals models have been developed to

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explain the ablation mechanisms. In photochemical mechanisms, the photon break the chemical bonds of the material directly where as in photothermal mechanism the material is ablated by heating, melting and vaporizing the material (Chen et al., 1995). It is found that the material removal by laser ablation approximately obeys the Beer lamberts Law at lower laser fluences (Srinivasan, 1982). For photochemical ablation to occur, energy of the photons at that wavelength should overcome the intermolecular bond energies of the target material. The relation between the photon energy of light and laser wavelength is given by (Sauerbrey, 1989),

$$E = 1.245 / \lambda \tag{3}$$

Where λ is wavelength of light (193 nm), E is energy of photon (eV). The photon energy of the lights depends on the wavelength of the light and as the wavelength increases, the energy decreases. An UV laser with a wavelength of 193 nm has photon energy of 6.45 eV. It can be seen that for photochemical ablation to occur in thin film the photon energy of the light should be greater than the bond energy of the material. While excimer laser does not produce as high average powers as longer wavelength infrared lasers such as Nd:YAG and CO₂, their shorter wavelength carried high photon energy. As for example, argon fluoride (ArF) excimer laser with wavelength of 193 nm have photon energy of 6.45 eV, while Nd:YAG with wavelength of 1064 nm have photon energy of 0.18 eV. Hence, only laser with short wavelength can initiate photochemical reaction while the laser with longer wavelength may only lead to heating the material only.

Apparently, the advantage of photochemical reaction, is that the mechanism can change the molecule structure as well as the refractive index of the material, but not involve in external damage unless the energy deliver is too high. As a result the photochemical mechanism is important effect in the nucleation and the growing of grain size or in the crystallization process.

4. Sample preparation

Target used to be annealed is an amorphous silicon. In this works silicon is chosen to be the main material and deposited on the glass substrate as the first layer of thin film. Silicon is widely used in semiconductors industry. It can stand at higher temperatures compared to germanium. Furthermore its native oxide is easily grown in a furnace. Plus it can form a better dielectric interface rather than any other materials.

An ordinary glass substrate is conducted to make it possible for low annealing temperature. The glass substrate used in this research has maximum operation temperature in the range of 350°C to 400°C. The thickness of the glass substrate is in between 1.0 mm to 1.2 mm. The glass substrates are cut into small pieces using diamond cutter. Each of them having dimension of $10 \times 10 \text{ mm}^2$. All glass substrates are cleaned using Ultrasonic Branson 3210 cleaning machine to ensure the glass surface is free from any dust, oil, or contaminations.

4.1 Thin film fabrication

Solid phase crystalization (SPC) is prepared by using a vacuum chamber technology. An Edwards 360 thermal evaporation is a vacuum chamber used to fabricate the silicon thin

film. Silicon is deposited on a glass substrate. Initially, the source material to be deposited or evaporant is loaded into a molybdenum boat. The glass substrate is placed 10 cm above the evaporant. The chamber is set at a vacuum condition until the pressure achieved down to 10⁻⁶ mbar. The current is increased slowly until the evaporant on the molybdenum boat begin to melt. The thickness of film is detected by quartz crystal thickness detector inside the vacuum chamber. The thickness is measured via an Edwards FTM 7 which connected to the vacuum system. To deposit the silicon for about 100 nm thick, 0.3 g of silicon powder is needed. A schematic diagram of the thin film is shown in Figure 6.



Fig. 6. Silicon thin film on glass substrate

4.2 Annealing process

The annealing of silicon thin film is a combination from heat treatment and excimer exposure. Immediately after fabricated the thin film, it is heated via conventional Tube Furnace type F21100. This is important procedure in order to dry out the water inside the thin film so that the hydrogen percentage can be lowered. The hydrogenated thin film is then annealed by using an argon fluoride (ArF) excimer laser.

4.2.1 Heat treatment

The prepared solid phase crystallization of silicon thin film is initially under-went heat treatment. As mention earlier the aim is to extract the water contained in the film during deposition. An encapsulated annealing technique is utilized for this heat treatment in order to avoid vaporization of thin film material. The silicon thin film is placed in a carbon block (both the container and its cover are made from the same element). The capsulated thin film is then placed in the tube furnace. The annealing process is carried out under atmospheric pressure for four hours in air. The annealing is set at a fixed temperature of 350 °C. This furnace has a single set point control which enables the end user to control the furnace up to a preset temperature and remained constant at the set temperature.

4.2.2 Laser annealing

Nearly all optical materials show moderate to intense absorption in the ultraviolet region. This absorption is usually ascribed to electronic transition from a ground singlet to the first excited state. The unique feature in the UV laser ablation of optical material is encountered only in those wavelength region in which such electronic absorption exist (Srinivasan et al., 1988). Material ablation depends on two key conditions, as shown in Figure 7. First, the material must absorb light strongly at the laser's wavelength. Clean, precise ablation usually requires linear absorption coefficients of at least 10⁴ cm⁻¹. Secondly, ablation occurs only after the material has absorbed a minimum energy per unit volume, i.e., the laser intensity must exceed a threshold value (Brannon et al., 1985).

The laser is absorbed in amorphous silicon thin film surface without heating the substrate. A homogenized excimer laser beam spot is exposed on the thin film surface. Within the laser pulse duration the silicon layer is rapidly heated and melted as shown in Figure 7(a, b). As it cools down the crystallization into poly-silicon occurs (Figure 7c, d). In the process of excimer laser annealing, the amorphous silicon thin film is exposed by multiple pulses of laser. The exposed area is partially melted. The period for heating as well as for freezing is the same as the pulse duration of the excimer laser. The heat release in high speed freezing is more compared to the slow rate once. As a result during cooling the heat released by convection force-liquid flow which break the planar symmetry so that the crystal grow develops along the columns or finger such as shown in Figure 7c and d. In Figure 7c, there is partially melted and partially crystallized growth. The totally crystal growth is illustrated in Figure 7d. The crystal grows in the finger formation or poly-crystallization.



Fig. 7. Schematic diagram of excimer laser annealing on amorphous silicon copper thin film.

5. Polycrystalline silicon

Silicon films are fabricated with a microstructure tailored to the application depending on economic and performance requirements. Silicon film is typically divided into three categories: monocrystalline, amorphous, and polycrystalline.

Monocrystalline silicon is a form in which the crystal structure is homogenous throughout the material; the orientation, lattice parameter, and electronic properties are constant throughout the material (Green, 2004). Dopant atoms such as phosphorus and boron are often incorporated into the film to make the silicon n-type or p-type respectively. Monocrystalline silicon is fabricated in the form of silicon wafers, usually by the Czochralski Growth method, and can be quite expensive depending on the radial size of the desired single crystal wafer (Green, 2004). This monocrystalline material is one of the chief expenses where approximately 40% of the final price of the product is attributable to the cost of the starting silicon wafer used in cell fabrication (Campbell, 2001).

Amorphous silicon has no long-range periodic order. The application of amorphous silicon as a standalone material is somewhat limited by its inferior electronic properties (Streetman and Banerjee, 2000). When paired with microcrystalline silicon in tandem and triple-junction solar cells, however, higher efficiency can be attained than with single-junction solar cells (Shah et al., 2003). This tandem assembly of solar cells allows one to obtain a thin-film material with a bandgap of around 1.12 eV (the same as single-crystal silicon) compared to the bandgap of amorphous silicon of 1.7-1.8 eV bandgap (Shah et al., 2003). Tandem solar cells are then attractive since they can be fabricated with a bandgap similar to single-crystal silicon but with the ease of amorphous silicon.

Polycrystalline silicon is composed of many smaller silicon grains of varied crystallographic orientation (Figure 8). This material can be synthesized easily by allowing liquid silicon to cool using a seed crystal of the desired crystal structure. Additionally, other methods for crystallizing amorphous silicon to form polycrystalline exist such as high temperature chemical vapor deposition (CVD). Each grain is crystalline over the width of the grain. The grain boundary separates the grains where the adjoining grain is at a different orientation than its neighbor. The grain boundary separates regions of different crystal structure thus serving as a center for recombination. '*d*' here is a characteristic grain size, which should be maximized for maximum film efficiency. Typical values of *d* are about 1 micrometer.

Presently, polysilicon is commonly used for the conducting gate materials in semiconductor devices such as MOSFETs; however, it has potential for large-scale photovoltaic devices (Streetman and Banerjee, 2000; Ghosh et al., 1980). The abundance, stability, and low toxicity of silicon, combined with the low cost of polysilicon relative to single crystals makes this variety of material attractive for photovoltaic production (Ghosh et al. 1980). Grain size has been shown to have an effect on the efficiency of polycrystalline solar cells. Solar cell efficiency increases with grain size. This effect is due to reduce recombination in the solar cell. Recombination, which is a limiting factor for current in a solar cell, occurs more prevalently at grain boundaries as shown in Figure 8 (Ghosh et al., 1980).



Fig. 8. Grain boundaries of polysilicon

The resistivity, mobility, and free-carrier concentration in monocrystalline silicon vary with doping concentration of the single crystal silicon. Whereas the doping of polycrystalline silicon does have an effect on the resistivity, mobility, and free-carrier concentration, these properties strongly depend on the polycrystalline grain size, which is a physical parameter

that the material scientist can manipulate (Ghosh et al., 1980). Through the methods of crystallization to form polycrystalline silicon, a scientist can control the size of the polycrystalline grains which will vary the physical properties of the material.

5.1 Novel ideas for polycrystalline silicon

The use of polycrystalline silicon in the production of solar cells requires less material and therefore provides for higher profits and increased manufacturing throughput. Polycrystalline silicon does not need to be deposited on a silicon wafer to form a solar cell, rather it can be deposited on other-cheaper materials, thus reducing the cost. Not requiring a silicon wafer alleviates the silicon shortages occasionally faced by the microelectronics industry. An example of not using a silicon wafer is crystalline silicon on glass (CSG) materials (Basore, 2006).

A primary concern in the photovoltaics industry is cell efficiency. However, sufficient cost savings from cell manufacturing can be suitable to offset reduced efficiency in the field, such as the use of larger solar cell arrays compared with more compact or higher efficiency designs. Designs such as CSG are attractive because of a low cost of production even with reduced efficiency (Basore, 2006). Higher efficiency devices yield modules that occupy less space and are more compacts however the 5-10 % efficiency of typical CSG devices still makes them attractive for installation in large central-service stations, such as a power station (Basore, 2006). The issue of efficiency versus cost is a value decision of whether one requires an "energy dense" solar cell or sufficient area is available for the installation of less expensive alternatives. For instance, a solar cell used for power generation in a remote location might require a more highly efficient solar cell than one used for low-power applications, such as solar accent lighting or pocket calculators, or near established power grids.

Thin film silicon photovoltaics are typically produced by chemical vapor deposition processes yielding an amorphous, polycrystalline, or nanocrystalline film. Conventionally, amorphous silicon thin films are most common. Silicon is usually deposited on glass, plastic, or metallic substrates coated with a transparent conducting oxide material. While chalcogenide-based Cadmium-Tellurium (CdTe) and Copper-Indium-Selenium (CIS) polycrystalline thin films cells have been developed in the lab with great success, there is still industry interest in silicon-based thin film cells. Silicon-based devices exhibit fewer problems than their CdTe and CIS counterparts such as toxicity and humidity issues with CdTe cells and low manufacturing yields of CIS due to material complexity. Additionally, due to political resistance to the use non-"green" materials in solar energy production, there is no stigma in the use of standard silicon. Three major silicon-based module designs dominate: amorphous silicon cells, amorphous or microcrystalline tandem cells, and thin-film polycrystalline silicon on glass (Green, 2003).

Amorphous or microcrystalline silicon consists of a mixed phase of small crystalline regions surrounded by amorphous material. This material typically behaves more like crystalline silicon than the amorphous variety. A 3-month field study has shown that hybrid amorphous or microcrystalline cells degrade roughly to the same degree as triple-junction amorphous cells while maintaining higher conversion efficiencies (7.0% versus 5.0% as measured at the conclusion of the study). This result suggests hybrid designs of this type may supplant traditional amorphous-based modules (Green, 2003).

A new attempt to fuse the advantages of bulk silicon with those of thin-film devices is thin film polycrystalline silicon on glass. These modules are produced by depositing an antireflection coating and doped silicon onto textured glass substrates using plasma-enhanced chemical vapor deposition (PECVD). The texture in the glass enhances the efficiency of the cell by approximately 3% by reducing the amount of incident light reflecting from the solar cell and trapping light inside the solar cell. The silicon film is crystallized by an annealing step, temperatures of 400 – 600 °C, resulting in polycrystalline silicon.

These new devices show energy conversion efficiencies of 8% and high manufacturing yields of >90%. Crystalline silicon on glass (CSG), where the polycrystalline silicon is 1-2 micrometers, is noted for its stability and durability; the use of thin film techniques also contributes to a cost savings over bulk photovoltaics. These modules do not require the presence of a transparent conducting oxide layer. This simplifies the production process twofold; not only can this step be skipped, but the absence of this layer makes the process of constructing a contact scheme much simpler. Both of these simplifications further reduce the cost of production. Despite the numerous advantages over alternative design, production cost estimations on a per unit area basis show that these devices are comparable in cost to single-junction amorphous thin film cells (Green, 2003).

5.2 Low temperature induced crystallization of amorphous silicon

Amorphous silicon can be transformed to crystalline silicon using well-understood and widely implemented high-temperature annealing processes. This typical method is the typical method used in industry but requires high-temperature compatible materials, such as special high temperature glass that is expensive to produce. However, there are many applications for which this is an inherently unattractive production method. Flexible solar cells have been a topic of interest for less conspicuous-integrated power generation than solar power farms. These modules may be placed in areas where traditional cells would not be feasible, such as wrapped around a telephone pole or cell phone tower. In this application a photovoltaic material may be applied to a flexible substrate, often a polymer. Such substrates cannot survive the high temperatures experienced during traditional annealing. Instead, novel methods of crystallizing the silicon without disturbing the underlying substrate have been studied extensively. Aluminum-induced crystallization (AIC) and local laser crystallization are common in the literature, however not extensively used in industry. In both of these methods, amorphous silicon (a-Si or a-Si:H) is grown using traditional techniques such as plasma-enhanced chemical vapor deposition (PECVD). The crystallization methods diverge during post-deposition processing.

Another method of achieving the same result is the use of a laser to heat the silicon locally without heating the underlying substrate beyond some upper temperature limit. An excimer laser or, alternatively, green lasers such as a frequency-doubled Nd:YAG laser is used to heat the amorphous silicon, supplying energy necessary to nucleate grain growth. The laser fluence must be carefully controlled in order to induce crystallization without causing widespread melting. Crystallization of the film occurs as a very small portion of the silicon film is melted and allowed to cool. Ideally, the laser should melt the silicon film through its entire thickness, but not damage the substrate. Toward this end, a layer of silicon dioxide is sometimes added to act as a thermal barrier (Yuan et al., 2009). This allows the use of

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substrates that cannot be exposed to the high temperatures of standard annealing, polymers for instance. Polymer-backed solar cells are of interest for seamlessly integrated power production schemes that involve placing photovoltaics on everyday surfaces.

A third method for crystallizing amorphous silicon is the use of thermal plasma jet. This strategy is an attempt to alleviate some of the problems associated with laser processing – namely the small region of crystallization and the high cost of the process on a production scale. The plasma torch is a simple piece of equipment that is used to thermally anneal the amorphous silicon. Compared to the laser method, this technique is simpler and more cost effective (Lee et al., 2009).

Plasma torch annealing is attractive because the process parameters and equipment dimension can be changed easily to yield varying levels of performance. A high level of crystallization (~90%) can be obtained with this method. Disadvantages include difficulty achieving uniformity in the crystallization of the film. While this method is applied frequently to silicon on a glass substrate, processing temperatures may be too high for polymers.

6. Transformation temperature

Laser annealing on the thin film is a high-speed process. It depends on the pulse duration of the excimer laser normally in nanoseconds region and the threshold energy to nucleate and optimum energy to stop crystallization. Such short duration of annealing process is referred as transient crystallization of amorphous silicon film (Viatella and Singh, 1997). Within transient duration, the annealing process still needs to pass through several critical points. In the next section, such phase transformation point will be described in detail.

6.1 Critical points

In order to figure-out the physical change involve in laser annealing it is better to understand the basic concept of phase transformation during heating and cooling of hardening process. There are several important stages in heating and cooling of material. The crucial parameter is temperature at several critical points (Figure 9). The "critical points" are the temperatures at which certain changes in the chemical composition of the material take place, during both heating and cooling. Steel for example at normal temperatures has its carbon (which is its chief hardening element) in a certain form called *pearlite* carbon, and if the steel is heated to a certain temperature, a change occurs and the pearlite becomes martensite or hardening carbon. If the steel is allowed to cool slowly, the hardening carbon changes back to pearlite. The points at which these changes occur are the decalescence and recalescence or critical points, and the effect of these molecular changes is as follows: When a piece of steel is heated to a certain point, it continues to absorb heat without appreciably rising in temperature, although its immediate surroundings may be hotter than the steel. This is the *decalescence* point. Similarly, steel cooling slowly from a high heat will, at a certain temperature, actually increase in temperature, although its surroundings may be colder. This takes place at the recalescence point. The recalescence point is lower than the decalescence point, and the lower of these points does not manifest itself unless the higher one has first been fully passed. These critical points have a direct relation to the hardening of steel. Unless a temperature sufficient to reach the decalescence point is obtained, so that

the pearlite carbon is changed into a hardening carbon, no hardening action can take place; and unless the steel is cooled suddenly before it reaches the recalescence point, thus preventing the changing back again from hardening to pearlite carbon, no hardening can take place. The critical points vary for different kinds of material, and must be determined by tests in each case.

Similar phase transformation also experience by other materials during heating and cooling process. The different may be arisen in the aspect of temperature and the time taken to complete the same process. If the steel is replaced with other material like any amorphous silicon thin film and take into account the duration involves in nanosecond time domain then the whole event is considered as a high-speed phenomenon. This includes the rate of absorption and release of heat which occurs in transient time and enough energy to cause the phase change. In case excimer laser ablation which posses in nanoseconds pulse duration and fluence more than 500 J/cm², having high potential to cause the target thin film experience the same process as hardening steel. This implies excimer laser annealing on thin film should passing through the decalescence and recalescence points to ensure the formation of crystallization.

In general when amorphous silicon thin film is heated and cooling, normal phase change is occurred including the melting and re-solidification transformation. The phenomenon of this phase transformation is similar as illustrated in Figure 9. The red line is indicated the heating process, while the blue line represents the cooling process. In heating process, there are two important points that are decalescence and critical points. Decalescence is a phenomenon that occurs when a material is being heated and there is a sudden slowing in the rate of temperature increase. The slowing rate is due to the change in the internal crystal structure of the material. Meanwhile during cooling down or re-solidification, the phase change is passing through a recalescence point. A sudden spontaneous increase in the temperature of cooling resulting from an exothermic change in crystal structure occurs. This is the critical state in cooling process whereby the formation of crystallization is taken place.

Furthermore, a single pulse of the excimer laser, carried out temperature of 6.42 eV which is higher than threshold to break the bonding of silicon molecules. Once the excimer laser pulse is exposed on the film, it must passing through the decalescence and recalescence points in order to form crystallization. If a lower energy than threshold material or too high energy achieve up to the peak temperature in the transformation process, no crystallization will be formed on silicon film.

In both temperatures, there is latent heat of fusion is liberated from the solid during heating and from liquid during cooling. The latent heat is liberated during both processes to delay either the process of heating or cooling. The coexistent both solid and liquid associated in heating and cooling, resistant the rate of transformation. The advantage of this drawback is given an opportunity to nucleate and grow the crystallization in the silicon.

If the heating is over than optimum energy by exposing with higher number of pulses, the thin film is totally melted. No solid form is left to be as a seed for crystallization. In this case the treatment is achieved up to the peak temperature, passing over the decalescence point. Subsequently re-solidification occurs faster without passing through the recalescence point. Hence no formation of crystallization and end up with homogenous or no change in crystal structure in the excimer laser annealing process.

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Fig. 9. Transformation temperature

6.2 Recalescence effect

There are several researches on the recalescence phenomenon have been reported. Among them are Zhang and Atren, (1992), Armengol et al., (2003) and Mariucci et al., (2003). Zhang and Atren, (1992) have studied the recalescence characteristics in rapid solidification of copper by using a thermokinetic model. They have investigated the effects of the heat transfer coefficient, the melt thickness and the nucleation temperature. Their results had shown that the lower nucleation temperature and thinner melt lead to a longer recalescence effect while larger heat transfer coefficient results in a weaker recalescence effect. A dimensionless number was derived to measure the extent of recalescence.

Mariuci group (2003) had claimed that crystallization less than complete melting point will experience the recalescence effect. Structural properties of thin polycrystalline silicon films, crystallized by single shot excimer laser annealing at different laser energy densities, have been investigated by various researchers (Armengol, et al., 2003,; Mariucci et al., 2003; Razak and Bidin, 2010; Bidin and Razak, 2011a, b). Formation of disk structures has been observed in a wide range of energy densities, from complete melting down to 180 mJ/cm². These structures have been correlated to the lateral growth of grains starting from the small grains present in the central regions of the disks. They had proposed a new crystallization scenario for energy densities below the complete melting. In this framework, the recalescence effect plays an important role while the super lateral growth-regime is no longer a particular crystallization condition but simply represents the upper energy density limit of partial melt crystallization regime.

Similar recalescence effect after bulk solidification in germanium films melted by nanosecond laser pulses had also reported by Armengol, et al., (2003). Rapid solidification

dynamics in amorphous germanium films melted by nanosecond laser pulses has been analyzed by means of single-shot subnanosecond time resolved reflectivity measurements using a streak camera based setup. The results showed that once a minimum melt depth is induced, a bulk solidification process followed by the release of the solidification enthalpy dominates the solidification process. Moreover, the laser-melted material solidifies completely before being remelted as a consequence of the solidification enthalpy release, something only observed, up to date, upon irradiation with picosecond laser pulses (Armengol et al., 2003).

Bossuyt and Greer, (2004) have been investigated the effects of positive feedback on crystallization kinetics and recalescence. They described that in a series of alloys in the Cu-Ni-Ti-Zr system, with compositions close to the bulk glass forming alloy Cu47Ni8Ti34Zr11 (Vit101), the nucleation density has been shown to be spatially inhomogeneous; in an amorphous matrix there are spherical clusters with a high density of nanocrystals. The implied positive feedback in the nucleation rate was analyzed in terms of recalescence instability, where the latent heat released upon crystallization causes the nucleation rate to increase locally, if the nucleation rate is an increasing function of temperature and thermal diffusivity is low enough to avoid distributing the heat evenly over the sample. In deeply under cooled liquids, the first of these requirements is satisfied, but not for the second requirement.

Bossuyt and Greer have used numerical technique to trace the cooling rate with and without the feedback heat in crystallization formation. They identified a critical cooling rate for recalescence to occur. At slightly faster cooling rates, the heat released by crystallization causes the actual cooling rate to drop to nearly zero, but the heat output is never sufficient to actually raise the temperature. When that does occur, there is an abrupt transition from approximately 40% crystallization at cooling rates just above the critical cooling rate, to full crystallization below the critical cooling rate. Furthermore they claimed that disabling the heat of crystallization eliminates the recalescence effect from the temperature traces, and the transition to full crystallization is now much more gradual, without a clearly defined critical cooling rate.

Other feedback mechanisms have effects similar to those of recalescence: there is always a threshold condition where the acceleration due to feedback offsets the quenching effect of decreasing temperature. At cooling rates above this threshold, the crystallization kinetics did not deviate much from those in the absence of feedback. The effect of feedback becomes noticeable over a relatively narrow range of cooling rates just above the threshold, and then dominant below the threshold, abruptly increasing the degree of crystallization (of at least one of the phases). The decalescence and recalescence effect are shown in Figure 9. Further explanation about the critical points will be discussed on section 6.5.

However not many researchers have been realized about the decalescence effect in the recrystallization process. Without determination the decalescence effect during heating process the recalescence point cannot be achieved. Therefore in order to crystallization of polycrystalline silicon film the emphasized is focusing on these both phenomena (decalescence and recalescence effects). In this project, silicon thin film is crystallization by controlling the number of laser pulses.

In most cases, silicon is deposited onto a corning glass substrate and annealed with high energy excimer laser which requires high fabrication cost for industrial purposes. In this attempt, low energy ArF (wavelength of 193 nm) excimer laser was employed together with

conventional annealing method to anneal the silicon thin film. As reported by previous researchers (Carluccio et. al., 1997, Han et. al., 2003) combination annealing techniques between conventional and laser annealing would produce a better poly-si thin film. Figure 10 shows a different in terms of grain size structure between polysilicon thin film PSTF annealed by (a) conventional annealing and (b) combination of conventional and excimer laser annealing. Polycrystalline silicon thin film (PSTF) has been an interest currently due to high potential as electrical interconnections in microelectronics and microsensors.



Fig. 10. FESEM images of poly-Si (a) annealed at 600 °C for 24 h (*a*) and the combination annealing with ELA at 470 mJ cm⁻² (Han et. al., 2003)

6.3 Super lateral growth energy

In order to optimize the annealing process, a super lateral growth SLG energy density needs to be identified. The SLG point is known as the critical energy density or laser fluence desires to generate the maximum crystallization. Greater than this energy normally the crystallization will be reduced and soon eliminated. In other word the SLG energy density is equivalent with the critical point in the temperature transformation curve (Figure 9) during heating process. Beyond this energy, the silicon film will totally melt and cause elimination of spontaneous crystallized formation. This implies that calibration needs to be done in determining this crucial energy.

There are two parameters commonly used to quantify the degree of crystallization that are, grain size and surface roughness. The later parameter is due to the melting effect. Both parameters are utilized for measurement of crystallization. If the boundaries of the grain size is expanding or the surface roughness is increasing after being exposed by a series of excimer laser pulses that means the annealing process is within the decalescence and recalescence region. Once the optimum energy density that is super lateral growth energy is achieved that is at the critical point in the temperature transformation curve, no more number of pulses can be added or no more laser energy density can be increased.

Otherwise, the excess energy exposed that would cause the film totally melted and no solid form leaves at the interface between substrate and film to seed the crystallization.

Hence in excimer laser annealing the most crucial process is to identify the SLG energy. Under this particular SLG energy, the heating and the cooling are associated with the decalescence and recalescence effects. This implies that, the phase change is accompanied with the coexistence of liquid and solid forms. This is illustrated in Figure 11. At the interface between film and substrate, there are some particles remain in solid form whereas the rest in liquid phase. The left over the particles are used to seeding the crystallization. This region is similar as the condition achieved at recalescence point in the transformation temperature curve during quenching or cooling period.



Fig. 11. Nearly complete melt of amorphous silicon layer. The liquid/solid interface seeds the crystallization process

It is well known that laser ablation involves photothermal and photochemical process, depending on the nature of the material used and the experimental conditions, for example laser fluence, wavelength, and pulse duration (Sato et al., 2001; Serafetinides et al., 2001; Hahn et al., 1999). A 193 nm wavelength radiation can cause a neat and clean etch region because at this wavelength the molar extinction coefficient of the material is high ($\sim 10^4$ L mol⁻¹ cm⁻¹) so that 95% of the light is absorbed in the first 300 nm of the substrate. On the other hand, a laser with longer wavelength (~ 500 nm) light melts and damage nearby regions of the sample as a longer wavelength light vibrationally excites the irradiation sites. This process can eventually cause material to desorb from the heated region (Barbara et al., 1984).

7. Atomic force microscope analysis

The crystallization result is quantified based on metallurgical technique. The annealed silicon thin film is examined under the atomic force microscope. The scan area for each sample is fixed to 5000 nm X 5000 nm and zoomed until 1000 nm X 1000 nm to get clearer image of the thin film surface. The typical of the nucleation and the growth of silicon thin

film is shown in Figure 12. The frames of crystallization are arranged in the increasing order of the energy density. Prior annealing, the grain size of the amorphous silicon thin film is measured. The average nanocrystal size, G_{av} of the amorphous silicon film before doping and heating as shown in Figure 12(a) is approximately 17 nm. After experienced four hours heat treament via conventional annealing, a new crystal structure is realized. The crystallization pattern almost uniform with an increment up to 56 nm as depicted in Figure 12(b). Immediately after crystalization with a single shot of excimer laser corresponding to energy density of 65.50 mJcm⁻², another new pattern nanostructure is realized. The crystallization of the silicon film is accelerated further to a grain size of 75 nm as illustrated in Figure 12(c). The crystallization of thin film is continuously increasing after received the sequential number of pulses from argon fluoride ArF excimer laser. The maximum crystallization of silicon film is notified after received five number of pulses as depicted in Figure 12(g). The enlargement achieved approximately to 143 nm corresponding to the super lateral growth energy of 345 mJcm⁻². Beyond the critical energy as shown in Figure 12(h) the crystallization reduced drastically.



Fig. 12. AFM images of silicon thin film with magnification area of 1000 nm x 1000 nm at different energy density.

The stress developed during solidification due to the melting effect of silicon thin film is quantified based on surface roughness measurement. A typical example of the surface roughness of the silicon copper film is shown in Figure 13. Figure 13(a) indicates the roughness before annealing process. The measurement of surface roughness for the amorphous silicon thin film is 0.3 nm. After annealing the intergration of crystalization formation comprised of the agglomeration and coalesence of grain. Simialr results have also reported by He et al., (2007). The surface roughness of the annealed silicon thin film then increased up to 3.0 nm. The profile of the grain sized and surface roughness measurement are presented in Figure 14 and 15 respectively. However the maximum roughness is not overlapping with maximum grain size. The possible reason for such different may be due to the measured surface roughness is not matched with measured area of the grain size.



Fig. 13. Surface roughness of silicon at different number of pulses: a. o pulses, b. 3 pulses.



Fig. 14. Grain size as function of energy density



Fig. 15. Surface roughness of silicon thin film with respect to number of pulses

8. Conclusion

In general, the excimer laser annealing is capable to crystallize the silicon thin film based on the increment of the grain size as well as the surface roughness. In the process of annealing, the film experience transient heating and freezing. The UV light of excimer laser carried out energy of 6.42 eV which is equivalent with 7.5×10^4 K that is more than enough to melt the thin film. Considering the temporal length of pulse duration each of laser pulse is only 10 ns, this means the silicon copper thin film is experience an ultra-speed heating and freezing. The energetic excimer is enough to control the phase transformation with the decalesence point in which the melted is associated with some solid form to nucleate the crystal growth. The decalescence effect is controlled via the number of pulses. In contrast during rapid resolidification the liquid form is associated with recalescence effect which allows the formation of crystallization. The latent heat is released to delay the transformation from liquid to solid. Inherently the recalescence effect responsible to spontaneously crystallize the silicon film. When the energy density greater than the end limit of decalescence point which equivalent with the super lateral growth energy, the heating achieved the peak temperature where the film totally melted. Consequently no recalescence effect occur which result homogenous solidification without crystallization formation during cooling. Thus, in annealing it is crucial to determine the super lateral energy in order to ensure the process is within the decalescence and recalescence points to allow the formation of crystallization.

In addition, laser annealing process of silicon thin film promising well-arranged and large poly-Si grains. As far as we can see, laser annealing has become the most technique used to fabricate a good quality of poly-Si film for ultra-large scale integrated circuit devices, thin film transistors, active matrix liquid crystal displays, solar cells and other optoelectronic devices for industrial purposes. The employment of excimer laser for low temperature poly-Si (LTPS) thin film fabrication makes it possible to replace glass substrate with plastic substrate. This would certainly reduce the cost for large scale productions. Furthermore the high speed crystallization process will shorten the time to anneal as compare to the conventional method. The combination of time for annealing and the low energy consumption will give advantage for the production of high quality silicon thin film.

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