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Structural properties and electronic transport in intrinsic microcrystalline silicon deposited by the VHF-GD technique

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Abstract

A series of microcrystalline samples was deposited by the very high frequency glow discharge (VHF-GD) technique, with various input powers while keeping all the other parameters of deposition constant. The goal was to correlate transport and structural properties and avoid as much as possible the problem of a variation of the Fermi level between the samples. The observed decrease of the photoconductivity and of the product mobility-lifetime of hole (as measured by time of flight, TOF) with the increase of the power was surprisingly not connected to the structural properties, which remain approximately unchanged, but with a surface contribution to the transport properties.

Keywords: Microcrystalline; Silicon; Transport properties; Structure; Stability

1. Introduction

In a previous paper [1], we studied the electronic transport parallel and perpendicular to growth direction, in a series of microcrystalline silicon ($\mu\text{c-Si:H}$) samples obtained by various dilutions of silane in hydrogen. It was shown that the transport properties under dark and under illumination conditions increase as the dilution is increased. Furthermore, a correlation between the crystalline fraction in the samples and the transport properties was found. However, due to the uncontrolled incorporation of impurities during growth process, we were not able to find a clear interpretation of the electronic trans-

port, as the amount of impurities changed from one sample to the other.

To solve this problem, we have deposited a series of samples with various input powers (P), while keeping all the other parameters of deposition constant. With this procedure, the amount of impurities incorporated in the film was supposed to be fairly constant (even a priori unknown) with respect to the plasma power. Moreover, we aimed with the increase of P at changing the crystalline fraction of the samples, and thus, to get more information on its role in the transport.

2. Experimental

All the samples were produced by the very high frequency glow discharge (VHF-GD) deposition

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technique [2] at 130 MHz, at an effective deposition temperature of 230°C. The dilution of silane ($[\text{SiH}_4]/[\text{SiH}_4] + [\text{H}_2]$) by hydrogen was kept constant at 5% while the power (HF-power as measured just before the matching network) ranged from 9 to 30 W. The samples were undoped and produced with our gas purifier method [3] to get intrinsic character. Thicknesses of the films ranged between 2.1 and 2.8 μm . For the measurements in the coplanar geometry the films were deposited on glass substrate (AF45; Schott) with aluminium contacts (gap 0.5 mm). All samples were first annealed for 2 h at 180°C in 10 mb of nitrogen, and then cooled slowly down. For time of flight (TOF) measurements, the samples were deposited on glass/semi-transparent chromium substrates, and further fitted with semi-transparent chromium top contacts.

3. Results

3.1. Transport properties

Transport properties in the direction perpendicular to growth direction were as usual monitored by dark conductivity (σ_{dark}), photoconductivity (σ_{photo}) and by the ambipolar diffusion length (L_{amb}). The results

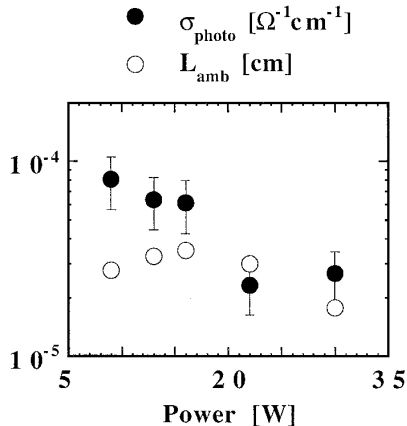


Fig. 1. The σ_{photo} and L_{amb} in function of the power. The measurements were done with an intensity of 20 mW cm^{-2} with red light, which corresponds to a generation of around $1.5 \times 10^{20} \text{ cm}^{-3} \text{ s}^{-1}$ (the error for L_{amb} is estimated around 7%).

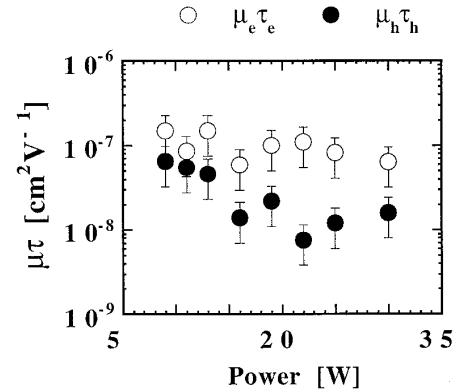


Fig. 2. The $\mu_e \tau_e$ (open circles) and $\mu_h \tau_h$ (closed circles) in function of the power, as measured by TOF.

under illumination are shown in Fig. 1. We observed a continuous decrease in σ_{photo} (factor 3.5) as the power is raised. For L_{amb} , an initial increase is noticed, to a power of around 17 W, followed by a decrease by a factor of 2 (note that for the smallest powers, the evaluation of L_{amb} is difficult, due to a very small photogain [4]). The dark conductivity decreases continuously as the power is increased (see Section 4).

The transport parallel to growth direction was studied by TOF. The product mobility-lifetime for the electrons $\mu_e \tau_e$ remains almost constant for all P with a drift mobility μ_e constant around $2 \text{ cm}^2 \text{V}^{-1} \text{ s}^{-1}$. In contrast, the mobility-lifetime product for the holes $\mu_h \tau_h$ decreases with the increase of P (see Fig. 2). The hole mobility was not measurable, due to the lack of full collection at voltages small enough to avoid RC problems.

3.2. Optical and structural properties

The absorption spectra (determined by the photo-thermal deflection spectroscopy (PDS) and calibrated by transmission/reflection (T&R)) of this power series as well as the X-rays measurements clearly indicate that all the samples are microcrystalline, but all the spectra are similar. Even the crystallite size, as evaluated by the Scherrer formula from X-rays diffraction (between 20 and 22 nm for the $\langle 220 \rangle$ peak), does not change with increase of the deposition power. However, with infrared absorption (IR)

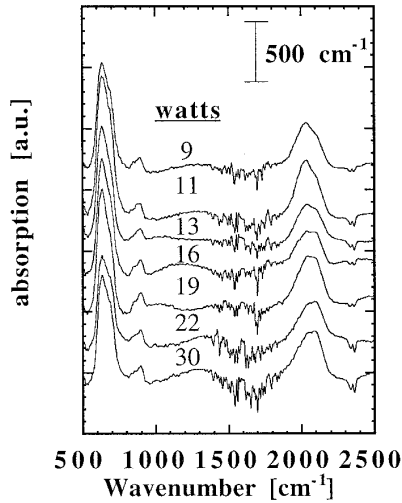


Fig. 3. IR absorption of a series of samples deposited with different powers, as measured after deposition. The spectra are arbitrarily shifted vertically for the sake of clarity.

done after deposition we observed a small change in the modes around 2000 cm^{-1} (see Fig. 3). The band at 2000 cm^{-1} , usually referred as the absorption of the Si-H stretching mode in a-Si:H, slightly shifts to 2100 cm^{-1} , which indicates Si-H bonded at crystallite surfaces [5,6], suggesting a small increase of the crystalline fraction.

4. Discussion

The interpretation of the differences in the electrical transport properties of these samples, structurally similar, is complicated by the observed change with time, when exposed to atmospheric conditions (see Fig. 4). We observed an increase in the dark conductivity when measured approximately 4 months after deposition, in comparison with the initial dark conductivity (measured the day after deposition). Furthermore, this increase becomes more important as P is raised, and is related to the initial value of σ_{dark} : the smaller the initial σ_{dark} , the more important the change.

Secondary ion mass spectroscopy (SIMS) measurements done several weeks after deposition show that there is an in diffusion of impurities (carbon, nitrogen and oxygen) from the surface of the sam-

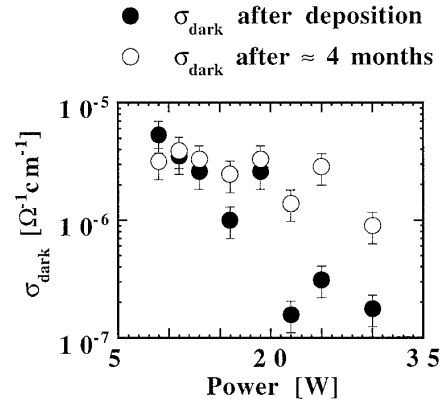


Fig. 4. The σ_{dark} for a series of samples deposited with various powers, as measured 1 day after deposition (closed circles) and around 4 months after deposition (open circles).

ples. These impurities are distributed in the form of a concentration gradient that penetrate deeper in the sample with the increase of the power (see Fig. 5 for the profile of the oxygen concentration). In contrast, the bulk concentration, which is also the concentration immediately after deposition, remains almost constant (within a factor of 2 for the oxygen, at around $3 \times 10^{18}\text{ atoms cm}^{-3}$) with the power.

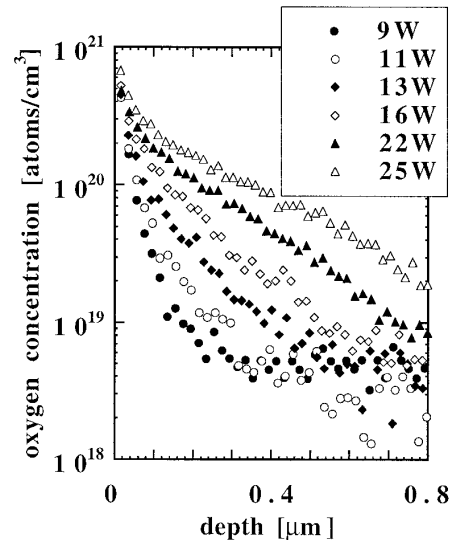


Fig. 5. Oxygen concentration for the series of samples deposited at various powers. For clarity the concentration has been plotted for all samples till $0.8\text{ }\mu\text{m}$ of depth. The accuracy of the values is $\pm 10\%$.

Two facts can be invoked to explain this diffusion of impurities. First of all, X-ray patterns surprisingly do not show any diffraction peak corresponding to the $\langle 111 \rangle$ plane (see Fig. 6). This absence indicates a highly preferential orientation in the direction perpendicular to the substrate (one major intensity due to the diffraction of planes in the direction $\langle 220 \rangle$) compared to the preferential orientation of our standard microcrystalline (two major intensities due to the diffraction of planes in the $\langle 220 \rangle$ and $\langle 111 \rangle$ direction). This very well defined orientation perpendicular to the substrate could favour the formation of voids (eventually even cracks) during growth process. On the other hand, IR spectra also indicate some small changes in the crystallite surface with the change of deposition power. However, these small structural changes seem responsible for the control of the incorporation of impurities in the sample after deposition.

The decrease of the initial σ_{dark} by a factor of 10 with P could hardly be explained by a variation of the oxygen bulk concentration. An increase of the deep defect density with the power is a possible explanation, but since very recently [7], PDS and CPM absorptions, usually used for a-Si:H, do not allow us to extract this density for $\mu\text{c-Si:H}$. The factor of 10 found in the CPM absorption at 0.9 eV between the samples deposited at 13 and 30 W could

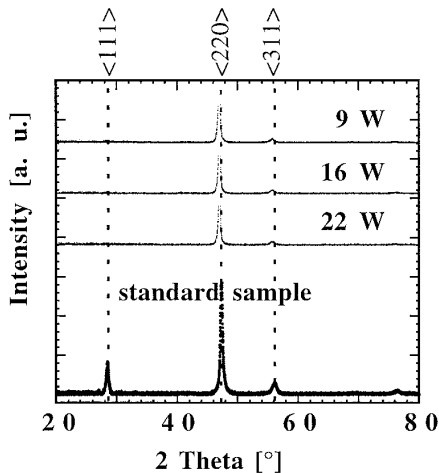


Fig. 6. X-rays diffraction patterns of one of our previous $\mu\text{c-Si:H}$ samples [1] and the diffraction pattern of the samples deposited at powers of 9, 16 and 22 W. The spectra are arbitrarily shifted vertically for clarity.

be due to the increase of the defect density but could also be due to scattering of light. Further measurements are needed with the new method proposed in Ref. [7]. On the other hand, the increase of σ_{dark} with time can be attributed to the post-oxidation, since O, with its n-type dopant character is going to increase σ_{dark} in the O-rich region close to the sample surface.

The post-oxidation could also explain the variation of the product $\mu_h\tau_h$ (measured several weeks after deposition), as TOF measurements are mainly sensitive to the region where the generation of the free carriers occurs. Doping by O is going to induce negatively-charged deep defects (in the amorphous tissue of the grain boundaries) and decreases the apparent magnitude of $\mu_h\tau_h$, without affecting $\mu_e\tau_e$. As far as μ_e is concerned, the highly preferential orientation in the direction perpendicular to the substrate does not bring any benefit; μ_e remains similar to that obtained in standard $\mu\text{c-Si:H}$ samples (which exhibit the $\langle 111 \rangle$ diffraction peak).

For σ_{photo} and for L_{amb} the situation is even more complicated, as illumination moves the quasi-Fermi levels, but measurements as function of time seem to rule out any further effect of the post-oxidation. We do not have, so far, a clear explanation for their change with the power (observed around 4 months after deposition), except the idea of an increase of the defect density with the power. However, even if confirmed, this increase of the defect density leads certainly not straightforward to an interpretation of both σ_{photo} and L_{amb} , when thinking of the situation in a-Si:H [8].

5. Conclusions

The crystalline fraction in our samples depends only weakly on the deposition power and all the samples exhibit similar optical (absorption coefficient) and structural (X-rays patterns) properties, characteristic of microcrystalline material. Only IR absorption shows a structural variation, which may indicate some changes towards more crystalline layers as the power increases. Contamination by impurities (and especially O that is a weak n-type dopant) depends on the deposition power and are responsible

for the variation in the electronic properties in the dark as a function of time. It also renders the interpretation of the transport under illumination problematic.

However, it is important to notice that the deposition power has (at least with deposition parameters used in this study) a strong effect on the initial σ_{dark} and on the post-oxidation, while having very weak influence on structural properties. It also shows that problem of post-oxidation can be controlled by adjusting deposition conditions. On the other hand, its effect in devices could be reduced by the protection offered by the deposition of contacts. As far as we know, $\mu\text{c-Si:H}$ solar cells are stable when exposed to atmospheric conditions and light-soaking (see Ref. [9]).

Finally, this study illustrates very well the problem of studying electronic properties of $\mu\text{c-Si:H}$ layers. Most transport characterisations are performed in coplanar configuration (in opposition with the transport in solar cells, which is in the growth direction), but other problems inherent to the material (such as its non-homogeneity, presence of grain boundaries) and depending of deposition conditions (post-contamination, etc.) render the interpretation rather difficult.

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