Transfer of excess charge carriers in an *a*-Si:H/crystalline-silicon heterojunction measured during the growth of the amorphous silicon layer

H. C. Neitzert,* W. Hirsch,† and M. Kunst‡
Department Solare Energetik, Hahn-Meitner-Institut, Glienickerstrasse 100, 1000 Berlin-39, Germany
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The plasma-induced deposition of hydrogenated amorphous silicon on crystalline silicon substrates has been investigated *in situ* by contactless transient photoconductivity measurements. In the initial stage of the deposition process the signals reflect charge-carrier kinetics in the substrate with the surface recombination rate modified by the deposition process. In the final stage the signal is dominated by mobile electrons in the film deposited. Between these extrema an additional contribution of excess charge carriers optically induced in the amorphous silicon film and transported to the substrate is observed. It is shown that this transport is inhibited by a high defect density in the film deposited and by a highly defective interface.

INTRODUCTION

Heterojunctions consisting of amorphous and crystalline silicon layers have recently received some attention because of their application in solar energy conversion devices,[1] where the amorphous silicon has replaced the thin top *n*-type layer usually used in crystalline silicon photovoltaic devices, with the advantage of requiring only low-temperature processing steps. For a similar type of heterojunction, conversion efficiencies up to 18% have been reported.[2] Further applications of this type of heterojunction are hetero-bipolar-transistors,[3] where the amorphous silicon wide-band-gap emitter allows for a thinner and highly doped base, thus increasing the operating speed of the transistors used in the microwave frequency range. For vidicon applications[4] the use of an *a*-Si:H layer decreases the effect of blooming by decreasing lateral currents due to its high resistance.

For the above-mentioned applications the electronic transport in the amorphous silicon layer and the charge-carrier kinetics at the heterointerface are of predominant importance for the device characteristics. Apart from these interesting applications in semiconductor devices, the *a*-Si:H/*c*-Si heterointerface can also serve as a model system for studies of semiconductor interfaces because of the high asymmetry of the electronic properties, e.g., the charge-carrier mobilities, in the adjacent layers. Transient current measurements on highly asymmetric double-layer structures of chalcogenide semiconductors have, for example, already been used to study the transport mechanism in As_2Se_3.[5,6]

Another advantage of the amorphous top layer is the fact that band-gap tailoring is possible by modifying the *a*-Si:H deposition conditions and by alloying the amorphous silicon with other elements such as carbon or germanium.

It is well known that plasma etching introduces electronic defects in the processed layers (see, for example, Ref. 7) and therefore the plasma deposition of hydrogenated amorphous silicon is also likely to influence the defect structure of the crystalline silicon substrate during the formation of the *a*-Si:H/*c*-Si heterojunction. A characterization of such a heterojunction by capacitance spectroscopy showed distinct differences between heterodiodes where the crystalline silicon was exposed directly to the plasma during *a*-Si:H deposition, leading to a high density of interface states, and diodes where the plasma was shielded by an additional electrode in order to minimize the direct impact of the energetic ions.[4]

In a previous work, it has been shown that the time-resolved microwave conductivity (TRMC) method, which is based on the change in microwave reflection after the creation of excess charge carriers in semiconductor layers, is able to monitor charge-carrier kinetics at interfaces between semiconductors[8] and as a contactless technique allows an *in situ* characterization of plasma deposition processes.[9] Measurements of the change of the decay of TRMC transients during the initial stage of *a*-Si:H plasma-enhanced chemical-vapor deposition (PECVD) on crystalline silicon devices showed a highly increased recombination at the *c*-Si surface after ignition of the silane plasma, followed by a passivation of the plasma-induced defects depending strongly on the *a*-Si:H deposition conditions.[10]

Due to much higher charge-carrier mobilities in the crystalline compared to those in the amorphous silicon, for thin *a*-Si:H layers the TRMC transients reflect only the excess charge carriers in the crystalline part of the heterojunction. However, this is no longer true for the thicker *a*-Si:H layer when the exciting light is mostly absorbed in the amorphous silicon film.

In this paper, we will investigate in particular the possibility of a transport of excess charge carriers from the amorphous to the crystalline layers, and its dependence on the state of the *a*-Si:H film and the interface. For this purpose, *a*-Si:H layers with a thickness exceeding 1 μm were deposited on the crystalline silicon. The kinetics of photoinduced excess charge carriers during growth of the heterostructures has been probed *in situ* on a very short time scale (until 150 ns after excitation).

EXPERIMENT

Intrinsic amorphous silicon (*a*-Si:H) layers have been deposited on crystalline silicon substrates by plasma-enhanced chemical-vapor deposition (PECVD) in a capacitively coupled rf-glow discharge system under the following conditions: silane flow, 10 sccm; gas pressure, 35 Pa; rf-power density, 35 mW cm^{-2}; self-biased (U_{bias}: -10 V). The substrate temperature was 120 or 250 °C, as indicated.

In one case an argon plasma treatment has been applied for 120 s prior to the amorphous silicon deposition, using the following parameters: argon flow, 30 sccm; gas pressure, 35 Pa; rf-power density, 35 mW cm^{-2}; a substrate temperature of 250 °C and self-biased (U_{bias}: -14.5 V).

The boron-doped single crystalline silicon (*c*-Si) wafers ($\langle 111 \rangle$ oriented; ρ:10 Ω cm; thickness: 510 μm) were stripped of their natural oxide by a 30-s dip in 40% hydrofluoric acid rinsed with deionized water and dried with nitrogen immediately before being loaded into a plasma deposition chamber. After being transferred into the deposition system, they were annealed in vacuo at 250 °C for 5 min (even if deposition was to take place at 120 °C). This ensured a uniform state of the *c*-Si surface at the start of all experiments.

Time-resolved microwave conductivity (TRMC) measurements using *Ka*-band equipment were performed during the plasma deposition (without interruption of the plasma) in a configuration where the crystalline silicon substrate terminates the open end of a waveguide, separated by a vacuum window. The change of the reflected microwave power after laser-pulse illumination of the sample from the front side was recorded on a transient recorder. Signal averaging was used, and it took about 30 s to record one transient. The illumination was performed with laser pulses [full width at half maximum (FWHM) = 10 ns] from a Nd:YAG (yttrium aluminum garnet) laser at a wavelength (λ) of 532 nm and light excitation densities ranging from 1–1000 μJ cm^{-2}. The excitation densities are changed by filters and in the following the optical densities (OD's) of the used filters will be given, where OD = 0.6 refers to 1000 μJ cm^{-2}. Further details of the experimental setup are given in Ref. 11. The overall time resolution is about 15 ns, determined mainly by the duration of the excitation pulse.

The TRMC signal [the relative change in reflected microwave power upon pulsed illumination, $\Delta P(t)/P$] is proportional to the total number of mobile excess charge carriers at time (t) weighted by their respective mobilities:

$$\Delta P(t)/P \sim \sum_i \Delta n_i(t)\mu_i \ , \tag{1}$$

where $\Delta n_i(t)$ is the number of excess charge carriers of species i, and μ_i is their mobility. For the special case of a crystalline silicon/intrinsic *a*-Si:H heterojunction, Eq. (1) can be simplified, because only excess mobile electrons and holes in the crystalline silicon characterized by their respective mobilities μ_n and μ_p, and excess mobile electrons in *a*-Si:H with mobility μ_d have to be taken into account, yielding

$$\Delta P(t)/P \sim \Delta c(t)(\mu_n + \mu_p) + \Delta a(t)\mu_d \ , \tag{2}$$

where $\Delta c(t)$ is the number of excess charge carriers in the crystalline silicon and $\Delta a(t)$ the number of excess mobile electrons in the *a*-Si:H. The contribution of the holes in the *a*-Si:H layer can be neglected in intrinsic *a*-Si:H due to their lower mobilities.[12]

TRMC signals [$\Delta P(t)/P$] as a function of the time elapsed since triggering of the exciting laser pulse are characterized in this work by the maximum signal height (TRMC amplitude) and the decay behavior. The TRMC amplitude [$A(t_1)$] as it is used here is defined as the maximum of the TRMC signal in a given time interval (t_1) after start of the laser-pulse illumination. In the following, we choose a time interval of 20 ns, if not otherwise indicated. To characterize the shape of the TRMC signal further, we use the ratio of the TRMC signal height at 150 ns divided by $A(t_1)$, which gives a first estimate of the kinetics of the TRMC transients.

RESULTS AND DISCUSSION

TRMC transients on crystalline silicon wafers, which are used as substrates in this work, typically show decay times in the μs range. The decay depends not only on the bulk minority-carrier lifetime but also on the surface recombination rate. In particular, if excess carriers are generated by light with a wavelength of 532 nm, as used in the present study, the decay in the short-time range is due mainly to surface recombination. Because of the high absorption coefficient of *c*-Si at this wavelength (10^4 cm^{-1}), excess carriers are induced in a near-surface region and, consequently, in the short time range only this region is probed by the TRMC technique. From the absence of any appreciable decay in the short-time range for the *c*-Si used in this work before deposition, it can be concluded that bulk decay of excess carriers does not play any role in this time range for the *c*-Si investigated here, and the surface is well passivated. The passivation can be achieved by an oxide coverage of the crystalline silicon, and under some conditions by an *a*-Si:H layer as well. In a former study it has been shown,[8] that the temperature of the substrate during PECVD growth is of crucial importance for the annealing of the interfacial defects created in the initial stage of the plasma deposition. In particular, at 250 °C an approximately complete quenching of the decay in the short-time range due to these defects is observed after growth of a 30-nm-thick *a*-Si:H layer, while at 120 °C, even for thicker *a*-Si:H layers, only a partial quenching has been found. It should also be mentioned that the bulk density of defects in *a*-Si:H is also more than one order of magnitude higher in films grown at a substrate temperature of 120 °C than at 250 °C. In the last case this density is below 10^{16} cm^{-3}.[13]

In order to study the influence of interface defects and *a*-Si:H bulk defects on the charge-carrier kinetics independently, we performed *in situ* TRMC measurements for four different cases of *a*-Si:H/*c*-Si heterostructures, with either high or low bulk defect density in the amorphous silicon and either high or low defect density at the *a*-Si:H/*c*-Si interface.

The cases where both defect densities were low or high, respectively, could be easily achieved, as mentioned above, by the change of the substrate temperature, which was 250 °C in the first case and 120 °C in the second case. The case of low interface and high bulk defect density has been reached by a prior deposition of a 30-nm-thick *a*-Si:H "interfacial" layer at 250 °C before the growth of the "bulk" layer deposited at 120 °C. In a last case, the high interface defect density has been achieved by a 2-min argon plasma treatment of the crystalline silicon surface prior to the growth of the *a*-Si:H layer at 250 °C, leading again to a low bulk defect density. Earlier experiments showed that the surface defects created during this argon plasma treatment cannot be completely annealed even after the deposition of a thick *a*-Si:H film at 250 °C.[14] Reactive ion etching with an argon plasma has been reported to decrease the minority-carrier generation lifetime in silicon by two orders of magnitude.[15]

In Fig. 1, typical TRMC transients measured at different times during the deposition of amorphous silicon at a substrate temperature of 250 °C are shown. The illumination is performed through the growing *a*-Si:H layer, and therefore an increasing fraction of the incident light is absorbed in the *a*-Si:H rather than in the crystalline silicon substrate. This leads to a decreasing amplitude of the TRMC signal with increasing *a*-Si:H thickness, as long as the signal is dominated by the free carriers in the crystalline part of the heterojunction due to the higher charge-carrier mobility in the crystalline silicon.

In order to follow the TRMC transients during the growth of a 2-μm-thick *a*-Si:H layer with an acceptable signal-to-noise ratio, the light intensity has been increased in the course of the experiment. In the present experiments this has been achieved by changing the filters attenuating the excitation pulse at regular intervals, indicated by the optical density of the filter used. The signal in Fig. 1 taken shortly after the plasma start rises within 15 ns to its maximum value, and is flat up to 150 ns. This relatively low interface recombination rate after 115-s deposition means that the plasma-induced surface degradation is appreciably reduced already at this time. A better evaluation of the surface recombination rate can be obtained by measuring the TRMC transients on a longer time scale.[16] The rise time of the transient obtained after 115 s reflects the rise time of the exciting laser pulse. The transients measured at longer deposition times show monotonically increasing rise times with increasing deposition time, and also show no subsequent decay on the measured time scale. The last transient displayed (taken at 12 140 s) additionally exhibits a sharp peak directly after excitation, which decays rapidly within 30 ns followed by the slow rise described above for the other transients. The rise time of the fast peak corresponds again to the duration of the laser pulse.

The change of the amplitude of the TRMC signal with deposition time is shown in Fig. 2. The data for different optical densities have been normalized by setting equal the values of the TRMC amplitude before and after a change of the filter. As mentioned above, the TRMC amplitude descreases in the beginning of the *a*-Si:H growth monotonically with deposition time, because with increasing thickness of the *a*-Si:H film there is a shift from absorption, and so generation of excess charge carriers in the *c*-Si part of the device to that in the *a*-Si:H film. After 9500 s of deposition, this is followed by a roughly constant TRMC amplitude for longer deposition times.

With the definition of the TRMC amplitude used here, this is not necessarily the maximum value of the TRMC signal, because the TRMC signal may still increase after the end of the time window in which the TRMC amplitude is determined. This is, for example, the case for the transients with the slow rise time shown in Fig. 1.

To check the impact of the width of this time window, in Fig. 2 the TRMC amplitude has been plotted for two different windows. The data obtained with the 200-ns window have been taken in a new experimental run, measured in a longer time range but under otherwise identical conditions. While the amplitude determined with the time window of 20 ns decays almost exponentially with deposition time over almost three orders of magnitude, the decay is initially about the same and then more gra-

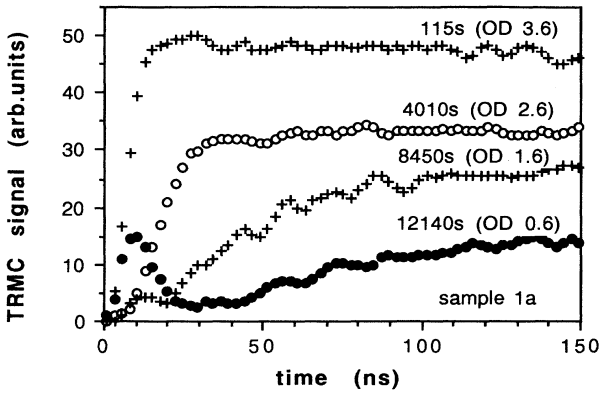

FIG. 1. TRMC transients, measured at different times after start of the deposition (as indicated) during the growth of amorphous silicon at $T_{dep}=250$ °C on *p*-type crystalline silicon.

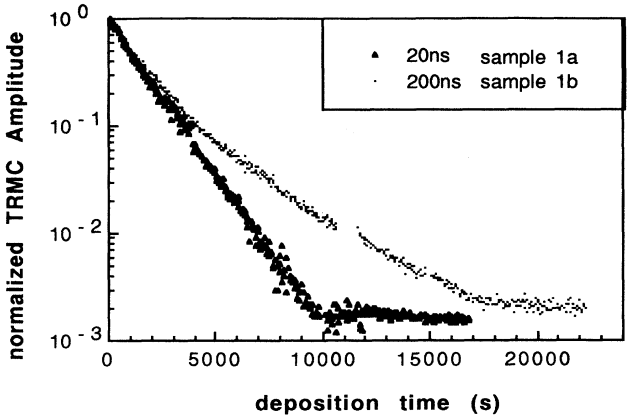

FIG. 2. Normalized TRMC amplitude measured during *a*-Si:H deposition on *p*-type crystalline silicon at $T_{dep}=250$ °C as a function of the deposition time with different windows for the determination of the TRMC amplitude.

dual in the case of the 200-ns window. The saturation level for long deposition times is about the same for both curves.

An explanation for the differences between both curves can be given, assuming that the TRMC signal consists of three contributions. The first is induced by the fraction of the light which is directly absorbed in the crystalline silicon substrate. According to Beer's law, this contribution should decrease exponentially with the deposition time, respectively with the *a*-Si:H thickness. The second part is the contribution of the free electrons in the amorphous silicon layer. Because of the low electron mobility in *a*-Si:H, it should only be observable at high excitation densities and when the density of free carriers in the crystalline silicon is much lower than the one in the amorphous silicon. A third contribution comes from charge carriers generated in the *a*-Si:H layer and subsequently injected into the silicon substrate. They would contribute to the TRMC signal only after their injection into the crystalline silicon. Because the transport and injection processes require time, increasing with increasing thickness of the *a*-Si:H layer, this signal will be delayed with respect to the two other ones, and its delay time will increase with increasing *a*-Si:H film thickness.

This component explains the increase of the rise times of the TRMC signals in Fig. 1 with increasing deposition times. This—on the other hand—leads to the slower than exponential decay of the TRMC amplitude with a window of 200 ns. Also inside this window, a remarkable fraction of charge carriers injected into the silicon from the *a*-Si:H layer contributes to the TRMC signal. If, however, the window is set to about the width of the exciting laser pulse, this contribution is rather small. A more detailed analysis suggests that even with this window for deposition times exceeding about 3000 s, a contribution of injected carriers has to be accounted for.

Knowing the deposition rate, a calculation of the absorption coefficient (α) at 532 nm in *a*-Si:H has been done. In the present case this leads to a value of α of about 1×10^5 cm^{-1}. This is comparable to the value reported recently.[17]

Figure 3 shows some TRMC transients at increasing times exceeding 11 000-s deposition. One can see that the initial peak remains the same in all transients, while the slowly increasing part rises more slowly with increasing deposition time and almost vanishes on this time scale for 16 440 s of deposition. We therefore assign the constant peak to the contribution of mobile electrons in the *a*-Si:H layer, while the slowly rising part reflects the carriers injected into the crystalline silicon. This is confirmed by the dependence of the TRMC amplitude (20-ns window) on the excitation density. After the long deposition time, the linear dependence of the *c*-Si has been replaced by a square-root dependence characteristic of amorphous silicon at high excitation densities.[13]

A simple way to estimate the influence of charge injection on the TRMC signal of *a*-Si:H/*c*-Si heterostructures is to compare the maximum signal of the 20-ns window with the value of the TRMC signal at a suitable later time (in our case at 150 ns). The value of 1 of this ratio up to about 4000 s reflects the fact that the signal is flat and the

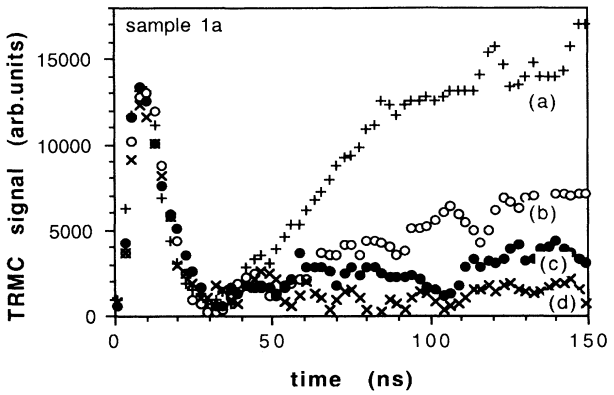

FIG. 3. TRMC transients, measured at different times during the growth of amorphous silicon at $T_{dep}=250$ °C on *p*-type crystalline silicon: (a) 11 880, (b) 13 620, (c) 15 000, and (d) 16 440 s, after start of the deposition.

contribution of charge carriers directly generated in the crystalline silicon dominates. The subsequent increase to a value of 4 shows that, in this regime, charge injection dominates, and the further decrease to a value below 0.2 reflects the more and more dominant contribution of electrons in the *a*-Si:H layer. Because no saturation of this ratio has been found even for these long deposition times, it can be stated that, even for *a*-Si:H thicknesses exceeding 2 μm in low-defect *a*-Si:H, there is still a small contribution of the injected fraction of charge carriers. This is in good agreement with experiments where the influence of interfaces in *a*-Si:H pin diodes on the charge-carrier decay has been studied.[8]

The same type of experiment has been performed during *a*-Si:H deposition at a substrate temperature of 120 °C. The transients in Fig. 4 show that the initially passivated *c*-Si substrate (with a TRMC signal which is not decaying on the displayed time scale) is heavily damaged by the silane plasma leading to a fast decay of the

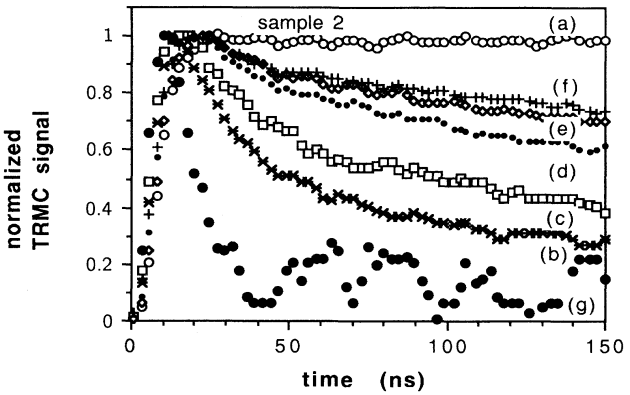

FIG. 4. TRMC transients measured before and during growth of an *a*-Si:H layer deposited at 120 °C on *p*-type crystalline silicon at different times after start of the deposition (as indicated): (a) before plasma start, (b) 120, (c) 500, (d) 2090, (e) 4690, (f) 7000, and (g) 9920 s after the plasma start.

signal even in the nanosecond time range. During the course of the *a*-Si:H deposition, a relatively weak anneal is seen. Again, for very long deposition times a signal which is typical of *a*-Si:H appears. In contrast to the experiment at 250 °C, no change in the slope of the TRMC signal is seen. This suggests that no injection into the crystalline silicon takes place. These findings are supported by consideration of the ratio between the signal value at 150 ns and the TRMC amplitude for deposition at 120 °C. Directly after ignition of the silane plasma, there is a sharp drop in this ratio due to a fast decay of the signal caused by a defective *c*-Si surface, and hence high recombination velocities. The ratio increases slightly, but stays well below the value of 1. Under otherwise identical conditions, but with a substrate temperature of 250 °C, this ratio amounts to 1 and higher. At 120 °C, this ratio decreases for long times down to zero. In the system deposited at 120 °C the electronic transport is expected to be highly suppressed by the high defect density in the bulk of the *a*-Si:H. Excess charge carriers eventually reaching the *a*-Si:H/*c*-Si interface additionally have a very high recombination probability there. Charge-carrier injection is therefore very unlikely in this structure, which is confirmed by the TRMC transients. This can also be concluded from the fact that the signal shape of *a*-Si:H films deposited at 120 °C on glass substrates is identical to that found here at long deposition times (Fig. 4).[13]

Another reason for the better electronic transport in the earlier experiment at 250 °C is the higher charge-carrier mobility in amorphous—in contrast to crystalline—silicon at higher temperatures. From extrapolation of existing data, the electron mobility should roughly be a factor of 2 higher at 250 °C than at 120 °C (compare Ref. 18).

In the next type of heterostructure, a low interface defect density has been achieved by a thin 250 °C layer prior to the deposition of a thick *a*-Si:H layer at 120 °C. The transients measured in Fig. 5 before and during the latter deposition show the typical form of signals on a well-passivated *c*-Si sample and do not change their form during the whole deposition, which is confirmed by the constant ratio between the signal value at 150 ns and the TRMC amplitude. Also in this structure, no change of the TRMC rise time, and hence no contribution of injected carriers, can be seen. The deposition here was too short to see the typical *a*-Si:H signal, which would be expected for measurements after very long deposition times.

In the last example, the surface of a *c*-Si substrate has been heavily damaged by an argon plasma before *a*-Si:H growth at 250 °C. The deposition at 250 °C leads to an *a*-Si:H layer with a low bulk defect density. The decay induced by the plasma damage and the subsequent (partial) quenching is seen in the transients displayed in Fig. 6. The transient taken after 8660 s shows again the typical decay of a low defect *a*-Si:H layer. The rise time for all transients is about the same. Before the argon plasma exposure the ratio between the height of the TRMC signal at 150 ns and the TRMC amplitude is about 1, which is a characteristic value for a passivated surface. After exposure to an argon plasma it drops to 0.2, indicating a heavily damaged surface of the crystalline silicon. During the *a*-Si:H deposition at 250 °C, the ratio increases again, but saturates at a value of 0.8, thus indicating only a partial and slow anneal of the surface, damaged by the argon plasma. At longer deposition times it decreases again and reaches a plateau at about 8000 s at a value of 0.18, a typical value for a TRMC signal of low-defect amorphous silicon at the excitation density used.[13]

It can also be concluded that in this last experiment no charge-carrier injection occurs. In this case the TRMC signal can be described simply as a superposition of the contribution of excess charge carriers in the crystalline silicon with that of excess electrons in amorphous silicon.

Figure 7 is a comparison of the rise times of the TRMC signals during the different *a*-Si:H depositions shown. Only in the case of sample 1 is a distinct increase with increasing *a*-Si:H thickness seen. That means that the excess charge-carrier injection from *a*-Si:H into the crystal-

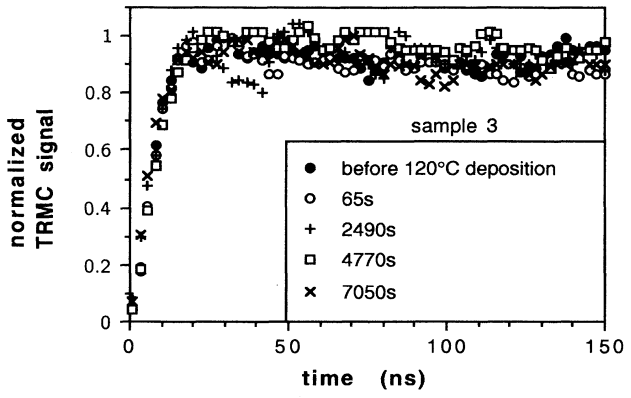

FIG. 5. TRMC transients measured before and at different times after start of the deposition (as indicated) during growth of an *a*-Si:H layer deposited at $T_{dep}=120$ °C on *p*-type crystalline silicon after prior deposition of a thin *a*-Si:H layer (about 30 nm thick) at $T_{dep}=250$ °C.

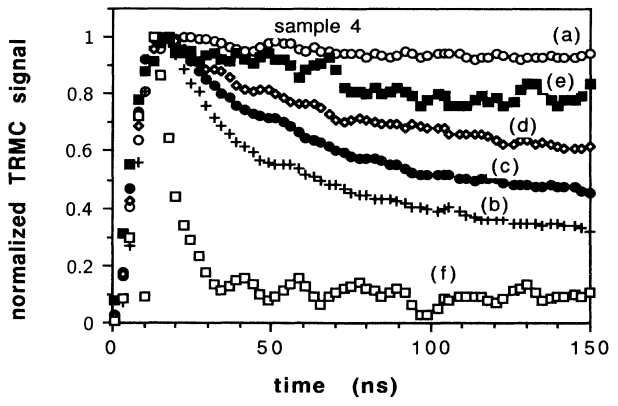

FIG. 6. TRMC transients measured at different times during growth of an *a*-Si:H layer deposited at 250 °C on *p*-type crystalline silicon after a prior 2-min exposure of the crystalline silicon to an argon plasma: (a) before argon plasma, (b) after 2-min argon plasma, (c) 50, (d) 760, (e) 1980, and (f) 8660 s after the start of the silane plasma.

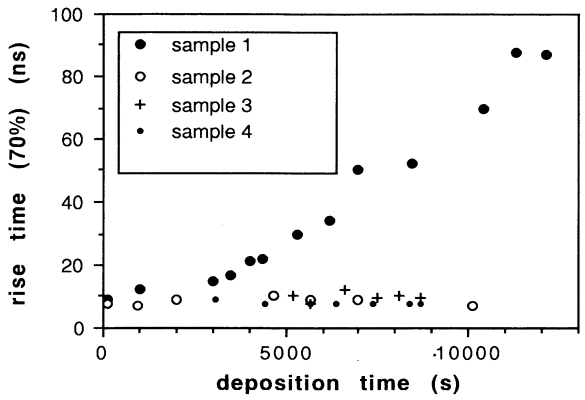

FIG. 7. Rise time (to 70% of the value at 150 ns) of the TRMC signals measured during *a*-Si:H deposition under different deposition conditions.

line silicon can effectively be suppressed either by a high *a*-Si:H bulk defect density or by recombination centers at the interface.

Another feature of these measurements is the saturation of the TRMC amplitude at very long deposition times, being identified as the TRMC amplitude of a pure amorphous silicon layer. Under reproducible experimental conditions such as microwave geometry and excitation density, the ratio between this amplitude and the initial amplitude before deposition gives a relative measure of the effective mobilities of electrons in different *a*-Si:H films, by comparing them to the mobility of a crystalline silicon sample which serves as a reference. A value of about 2×10^{-3} for this ratio has been found for samples 1 and 4, where in all cases the thick *a*-Si:H layer has been deposited at 250 °C and a value of 5×10^{-5} for sample 2, with a layer grown at 120 °C. For a comparison of these values, however, the influence of the different measurement temperatures on the mobilities has to be taken into account. The mobility of *a*-Si:H increases roughly by a factor of 2,[18] and the mobility of *c*-Si decreases by a factor of 2 (Ref. 19) with a temperature change from 120 to 250 °C. Correcting the ratios between the saturation level of the TRMC signal and the initial TRMC amplitude with this factors, there remains still a factor 10 of higher saturation level for the 250 °C film, which is due to a real difference in the electron mobility of both films.

CONCLUSIONS

Transient photoconductivity signals have been measured by a noncontact measurement technique based on the change of microwave reflection from silicon wafers after excess carrier creation during the growth of intrinsic amorphous silicon layers on top of these silicon substrates. For thin *a*-Si:H layers, the measured signals reflect the charge-carrier kinetics in the crystalline silicon only, but the surface recombination rates modified by the PECVD-growth process. In the case of amorphous silicon layers exceeding a thickness of 2 μm, the signal is dominated by the amorphous silicon layer only. For an intermediate thickness, however, both contributions have to be taken into account, but cannot simply be superimposed. This is due to an additional contribution to the TRMC signal originating from excess charge carriers created by light absorption in the *a*-Si:H layer and subsequently transported from the amorphous silicon into the crystalline silicon layer. This transport is inhibited either by a high defect density in the *a*-Si:H or by a high interface recombination rate due to a highly defective interface after exposure either to a silane or to an argon plasma.

*Present address: LPICM, Ecole Polytechnique, Palaiseau, France.
†Present address: PTB, Braunschweig, Germany.
‡To whom correspondence should be addressed.

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