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Queen, DR

Liu, X

Karel, J

et al.

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Light-induced metastability in pure and hydrogenated amorphous silicon

D. R. QUEEN¹, X. LIU², J. KAREL³, Q. WANG⁴, R. S. CRANDALL⁴, T. H. METCALF² and F. HELLMAN¹

¹ Department of Physics, University of California - Berkeley, CA 94720, USA

² Naval Research Laboratory - Washington D.C., 20375, USA

³ Department of Materials Science and Engineering, University of California - Berkeley, CA 94720, USA

⁴ National Renewable Energy Laboratory - Golden, CO 80401, USA

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Abstract – Light soaking is found to increase the specific heat C and internal friction Q^{-1} of pure (a -Si) and hydrogenated (a -Si:H) amorphous silicon. At the lowest temperatures, the increases in C and Q^{-1} are consistent with an increased density of two-level systems (TLS). The light-induced increase in C persists to room temperature. Neither the sound velocity nor shear modulus change with light soaking indicating that the Debye specific heat is unchanged which suggests that light soaking creates localized vibrational modes in addition to TLS. The increase can be reversibly added and removed by light soaking and annealing, respectively, suggesting that it is related to the Staebler-Wronski effect (SWE), even in a -Si without H, and involves a reversible nanoscale structural rearrangement that is facilitated by, but does not require, H to occur.

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Amorphous semiconductors are known to be metastable, undergoing structural rearrangements and electronic changes after exposure to light, such as, volumetric expansion and dangling bond (DB) generation in hydrogenated amorphous silicon (a -Si:H) and photo-darkening, bleaching, and structural relaxation in the chalcogenide glasses [1–5]. The light-induced increase of DBs in a -Si:H is problematic for solar cells as the DBs are recombination centers for electron-hole pairs. This is known as the Staebler-Wronski effect (SWE) [3]. It is generally thought that weak Si-Si bonds break when a photon is absorbed and that hydrogen then passivates one of the bonds leaving behind a DB [6,7]. DBs are removed by annealing above 150 °C, the temperature where hydrogen becomes mobile [6].

In addition to generating DBs, light soaking also increases the low-temperature acoustic loss in a -Si:H [8] which is caused by low-energy excitations described by the two-level systems (TLS) model [9,10]. TLS dominate the low-temperature acoustic, dielectric, and thermal properties of amorphous solids [11,12]. TLS are thought to result from the tunneling of single atoms or groups of atoms with energetically similar configurations but the

true microscopic origin of TLS is still unknown. Amorphous silicon is one of the few systems where the TLS density can be reduced below the universal value found in other amorphous solids [13,14]. Until recently, it was thought that including hydrogen was key to removing TLS in amorphous silicon much like it was key to eliminating the deleterious effects of DBs. However, we have recently shown that the same reduction of TLS seen in a -Si:H can be achieved in hydrogen-free a -Si by depositing the films at moderately elevated growth temperature [13–17]. The light-induced increase in both the TLS and DBs in a -Si:H suggests that both phenomena are related [8]. Exploring the relationship between light soaking and TLS may therefore elucidate the underlying structural origin of each. Understanding the origin of the TLS and how to controllably remove them has gained new urgency as decoherence caused by TLS is a major roadblock for quantum devices [18,19].

In this letter, we show that light soaking results in a persistent increase in the specific heat C and internal friction Q^{-1} for both a -Si and a -Si:H. This increase is qualitatively consistent with light-induced TLS but quantitatively the increases in C and Q^{-1} do not scale as expected from the

TLS model. The light-induced increases in C and Q^{-1} can be reversibly removed by annealing at 200 °C suggesting the metastable structures responsible for the TLS are related to the SWE. However, our results suggest that the light-induced TLS do not originate from DBs directly but are rather a consequence of light-induced nanoscale structural rearrangement. This is the first demonstration of light-induced metastability in non-hydrogenated a -Si and suggests that hydrogen is not required for the formation of the light-induced metastable states.

TLS densities can be measured from thermodynamic and acoustic measurements. The low- T specific heat of an amorphous, dielectric material has the form [20]

$$C(T) = c_1 T + c_3 T^3. \quad (1)$$

c_1 is calculated from the TLS model as

$$c_1 = \frac{\pi^2}{6} k_B^2 n_0 \frac{N_A}{n}, \quad (2)$$

where k_B is Boltzmann's constant, n_0 is the TLS density ($\approx 10^{45} \text{ J}^{-1} \text{ m}^{-3}$ for most glasses), N_A Avogadro's number, and n is the atomic number density [20,21]. The T^3 specific heat has two terms

$$c_3 = c_D + c_{ex}, \quad (3)$$

where c_D is the Debye specific heat due to phonons calculated from the sound velocity [22]. c_{ex} is an additional T^3 term attributed (qualitatively —there is no model) to non-propagating vibrational modes with a distribution of frequencies [20]. While c_{ex} is not predicted in the TLS model, we have found a strong correlation between n_0 and c_{ex} in previous work on a -Si suggesting that the same structures are responsible for both [15,17].

Internal friction Q^{-1} measures the scattering of acoustic waves from TLS and is a sensitive probe of the TLS density. The dissipation due to TLS produces a temperature-independent plateau Q_0^{-1} at $T \approx 1 \text{ K}$ [23] with magnitude,

$$Q_0^{-1} = \frac{\pi \bar{P} \gamma_i^2}{2 \rho v_i^2}, \quad (4)$$

where i indicates the polarization of the wave (longitudinal or transverse), $\bar{P}$ is the spectral TLS density, γ_i the coupling parameter between TLS and acoustic waves, ρ the density, and v_i the sound velocity. $Q_0^{-1} \approx 10^{-4}$ ($\bar{P} \approx 10^{44} \text{ J}^{-1} \text{ m}^{-3}$) for a wide range of glasses for both wave polarizations [12]. It is experimentally known that $n_0 > \bar{P}$ which is attributed to C measurements being sensitive to all TLS that equilibrate with the phonon bath during the time scale of the measurement whereas Q_0^{-1} measures those TLS where the relaxation time is close to the inverse measurement frequency ($\omega\tau \sim 1$).

a -Si films were prepared by e -beam evaporation at $\sim 1 \times 10^{-8}$ torr and a growth rate of 0.05–0.1 nm/s. Growth temperature T_S was varied from 45° to

400 °C [15]. a -Si:H films were prepared by hot-wire chemical vapor deposition (HWCVD) where the hydrogen content varied from 10 to 2 at. % by changing T_S from 300 to 470 °C [24]. Annealing the a -Si:H films at 200 °C after growth yielded a reproducible initial state. Films were grown on membrane-based nanocalorimeters and single-crystal double-paddle oscillators (DPO). Film thicknesses were measured with a profilometer with an error of 1%–4%. Film densities were determined from Rutherford backscattering (RBS) and hydrogen content by hydrogen forward scattering (HFS) [25].

Heat capacity was measured from 2 to 300 K using a microfabricated, thin-film nanocalorimeter [15,26–28]. Q^{-1} was measured from 0.3 to 300 K using the anti-symmetric torsional mode of the DPO at 5500 Hz [16,29]. The shift in the resonant frequency of the DPO after deposition of the film yielded the shear modulus $G = \rho v_t^2$ which was used to calculate v_t for a -Si. For a -Si:H, v_t was determined from surface acoustic wave measurements. v_l was measured using an ultrasonic pump/probe technique [30].

After initial C , Q^{-1} , and v_l measurements, films were light soaked using a PV Measurements Inc. small area AM1.5G solar simulator for 7 days which was sufficient to saturate the DB density (n_{DB}) [8,31]. For the specific heat samples, films were exposed through the 50 nm nitride membrane which was measured to be 60% transmissive. Temperature was monitored during light soaking and remained below 30 °C. After light soaking, samples were cooled to 77 K within 1 day and to 2 K by the second day. After measurement of a light soaked sample, the sample was annealed for 10^4 seconds at $T_A = 200$ °C, a temperature at which the SWE effect is known to be gone, and H to be mobile [6]. For C measurements, annealed samples were either slowly cooled or quenched to 4 K. All other samples were slowly cooled. No cooling rate dependence was observed.

Figure 1(a) shows the initial, light-soaked, and annealed states of an a -Si:H film deposited at $T_S = 430$ °C, corresponding to 4 at. % H, and an a -Si film deposited at $T_S = 45$ °C with no H. Crystalline silicon (c -Si) is shown for comparison and only has a T^3 Debye term at low T [32]. For both a -Si:H and a -Si, C at low T is significantly larger than the corresponding phonon Debye specific heat c_D which was calculated from the measured sound velocity [15]. v_l for the 45 °C a -Si film was measured in the initial, light soaked, and annealed states and did not change ($< 3\%$). Similarly, light soaking and annealing did not shift the DPO resonant frequency (< 2 ppm) for a -Si or a -Si:H films.

To further illustrate the light-soaking effect, fig. 1(b) shows the light-induced change in specific heat ΔC for the two films shown in fig. 1(a) along with an a -Si:H grown at 370 °C and an a -Si grown at 200 °C. ΔC persists to room temperature and for a -Si:H is of order ten times larger than both a -Si films at all T but with similar T dependence. We emphasize again that the increase in C up to room temperature is *not* due to a change

Fig. 1: (Colour on-line) (a) Specific heat C of a -Si:H ($T_S = 430^\circ\text{C}$) and a -Si ($T_S = 45^\circ\text{C}$). The legend indicates the order in which the films were light soaked (LS) and annealed (AN). C_D is shown for each film (dashed lines). The Debye temperature $\theta_D = 476\text{ K}$ for the $T_S = 45^\circ\text{C}$ a -Si film and $\theta_D = 590\text{ K}$ for the a -Si:H film. Crystalline silicon (c -Si) is shown for reference [32]. The Dulong-Petit limit is $24.9\text{ J mol}^{-1}\text{K}^{-1}$. (b) Increase in the specific heat ΔC of a -Si and a -Si:H after light soaking. Growth temperatures are given in parentheses. T and T^3 temperature dependences are shown for comparison (solid lines).

in phonons, hence C_D , as the sound velocity did not change ($< 3\%$) after light soaking. Additionally, C did not change in 4 T or 8 T magnetic fields at any T , ruling out the electronic states of DBs as the source of ΔC [33–35].

As is commonly observed in electrical measurements of the SWE, annealing reversibly removes the light-induced changes. We note that annealing the 45°C a -Si film at $T_A > T_S$ produces a nearly 30% reduction in C below 10 K, likely due to relaxation of the a -Si structure. Annealing changed the sound velocity by $< 3\%$, and thus the relaxation in the 45°C sample is occurring in those structures responsible for c_1 and c_{ex} .

The initial, light-soaked, and annealed states of the a -Si and a -Si:H films can be described at low T by eq. (1). The increase in c_1 corresponds to an increase in n_0 and the increase in c_3 corresponds to an increase in c_{ex} only since c_D is unchanged. Figure 2 shows n_0 plotted against c_{ex} for the initial state of a -Si and a -Si:H from fig. 1(a) along with several additional films [15,34]. Shown on the same scale are the changes in each term, Δn_0 and Δc_{ex} , between the initial and light-soaked states. We have previously shown in a -Si that n_0 and c_{ex} are correlated, depend strongly on growth conditions, and are independent of the

Fig. 2: (Colour on-line) n_0 vs. c_{ex} for as-prepared and annealed a -Si (red triangles) and annealed a -Si:H (black circles) from fits of C to eq. (1). Also shown on the same scales are the light-soaked changes Δn_0 vs. Δc_{ex} for a -Si (green open triangles) and a -Si:H (blue open circles).

sound velocity [15,16]. We then suggested that TLS and non-propagating vibrational modes that lead to c_{ex} have a common origin. Figure 2 shows that a -Si:H follows the same general trend observed in a -Si but with less sensitivity to growth temperature (and thus H content). **More importantly though, both Δn_0 and Δc_{ex} show the same trend as n_0 and c_{ex} in a -Si which suggests that light soaking creates additional TLS and non-propagating modes that are indistinguishable from those resulting from the growth process.**

Figure 3(a) shows Q^{-1} for several a -Si and a -Si:H films in the as-prepared, light-soaked, and annealed states. Figure 3(b) shows the light-soaked increase ΔQ^{-1} . The low- T plateau is typical of TLS. The peaks in a -Si:H at 8 K and 40 K are attributed to Si-H bonds [36]. Q^{-1} for the a -Si films shows the same T_S dependence as C [17]. A low value of Q^{-1} has previously been reported for a -Si:H film [13,36] but this value is inconsistent with the large low-temperature specific heat; we attribute this discrepancy to the low-energy excitations that contribute to C but couple very weakly to the acoustic waves, possibly due to H [34]. **ΔQ^{-1} is smaller than expected for all films given the size of ΔC . $\Delta Q^{-1}/Q^{-1}$ for the 45°C and 200°C a -Si films is 5% and 3%, respectively, whereas $\Delta n_0/n_0$ is 200% and 70% and $\Delta c_{ex}/c_{ex}$ is 56% and 53%, respectively. For the a -Si:H film, the magnitudes of both Q^{-1} and $\Delta Q^{-1} \leq 10^{-5}$ are similarly smaller than expected given the large values of C and ΔC but are comparable to previous results [8].** Thus the low-energy excitations seen in C are more sensitive to light soaking than those seen in Q^{-1} .

While it is known that light-soaking increases, and annealing decreases, n_{DB} , the underlying mechanism of the SWE is still not well understood. We begin by eliminating DBs, either intrinsic or light-induced, as the source of TLS in amorphous silicon. a -Si:H has $n_{DB} \approx 10^{16}\text{ cm}^{-3}$ and a -Si has $n_{DB} \approx 10^{18}\text{ cm}^{-3}$ however, at low T , C and ΔC of a -Si:H are ten times larger than a -Si. If DBs were the source of the TLS (either intrinsic or light induced) we would expect the opposite. Furthermore, we have observed no magnetic-field dependence for C , again

Fig. 3: (Colour on-line) (a) Internal friction Q^{-1} of *a*-Si and *a*-Si:H in the as-prepared (solid blue), light-soaked (open blue), and annealed (solid red) states. The plateau between 1 and 10 K is from two-level systems. Peaks at 8 K and 40 K in *a*-Si:H are due to Si-H bonds. (b) The increase in internal friction ΔQ^{-1} after light-soaking as-prepared films.

inconsistent with a DB as the source of TLS. In the HWCVD material, hydrogen is primarily bonded as monohydride (Si-H) and the number of 3-fold coordinated silicon ($\approx 10^{20} \text{ cm}^{-3}$) atoms is larger in *a*-Si:H than *a*-Si where the number of 3-fold coordinated atoms is proportional to $n_{DB} \approx 10^{18} \text{ cm}^{-3}$. We suggest that the TLS are associated with the local environment around these low coordination sites which are topologically distinct from the fully bonded silicon backbone network.

Several structural models for the SWE have been proposed based on weak Si-Si bond breaking. H is thought to stabilize the broken bond by forming a new Si-H bond and a dangling Si bond [6,7]. Given that C increases upon light soaking for both *a*-Si and *a*-Si:H, we suggest that light soaking breaks weak Si-Si bonds even in the absence of H. The larger ΔC for *a*-Si:H suggests that H increases the probability of this process by lowering the energy barrier for the same underlying bond breaking process that occurs in both structures [37,38]. It is notable that ΔC is indistinguishable for the two *a*-Si:H samples, and for the H-free *a*-Si ΔC is slightly larger for the 45°C than the 200°C sample. We draw two conclusions from these results. First, high surface mobility during growth (higher T_s) determines the disorder in the Si backbone, and therefore the sound velocity, whereas the number of weak Si-Si bonds depends on the formation of nanometer size imperfections, which H stabilizes in *a*-Si:H, that are buried during the growth process [15,39,40]. Second, due to the fact that hydrogen tends to cluster in *a*-Si:H, the number

of weak Si-Si bonds may be saturated in these higher hydrogen content regions and thus light-soaking effects are less sensitive to total H content.

Light soaking induces a volume expansion $\Delta V/V \sim 10^{-6}$ – 10^{-4} in *a*-Si:H indicating that low-density regions, such as voids, are formed in the material [1,2]. A heterogeneous density model was proposed for *a*-Si:H to explain this observation. In this model, the boundary between the high- and low-density regions is unstable and diffusion of hydrogen expands the low-density regions [41]. This picture is compatible with our heterogeneous model for hydrogen-free *a*-Si where the TLS are confined to the low-density regions and not present in the fully coordinated network [15,17]. The structures responsible for the low-energy excitations in *a*-Si:H are likely related to those in *a*-Si with the additional hydrogen lowering the energy barrier for TLS formation. A similar heterogeneous model was proposed to connect TLS to light sensitive structures in the chalcogenide glasses [42]. We suggest that Δn_0 and Δc_{ex} , created by light soaking, are also localized in low-density regions and that the non-propagating vibrational modes lead to the increase in C at higher T . $\Delta V/V$ and n_{DB} show the same light intensity and soaking time dependence [1] suggesting that they are correlated, however, the volume increase per DB generated is ~ 20 times larger than the Si-Si bond length which should be comparable in size to the DB/Si-H bond complex [7]. It is likely that the DBs are physically associated with the low-density regions and that DB creation is part of a larger-scale structural relaxation involving tens of atoms.

Recent atomistic, first-principles calculations of *a*-Si [43] and *a*-Si:H [44] provide some insight into the microscopic mechanism responsible for the low-energy excitations. These studies showed that local strains play a key role in the light-induced metastability and that Si-Si bond flipping resulted in a lower-energy final state as compared to the generation of a DB. In *a*-Si:H, the presence of H increased the probability of forming localized strain and that these strained regions form a connected network. This microscopic picture is supported by our results. The strained regions are natural places for non-propagating vibrational modes to occur and it has been proposed that TLS occur at the frustrated interfaces between regions of less frustrated material [45].

It is also notable for *a*-Si that although the light-induced changes, ΔC and ΔQ^{-1} are qualitatively consistent, they are not quantitatively consistent as one would expect from the TLS model which we have recently shown holds for a wide range of *a*-Si samples [17]. Even more striking is the lack of agreement between C and Q^{-1} for *a*-Si:H. In the framework of the TLS model, this can be accounted for by a reduction of the TLS-phonon coupling parameter γ (possibly even $\gamma = 0$) in eq. (4). Weak coupling between the TLS and phonons may result from the structural separation between the TLS in the low-density regions and the phonons which propagate on the backbone network. These are the first results that compare both C and Q^{-1}

from the same materials where the TLS density is not “universal”. Direct measurement of γ in these films and dielectric loss measurements will provide further insight into the assumptions of the TLS model.

In conclusion, we have demonstrated that the excess specific heat over a very wide temperature range in both a -Si and a -Si:H is reversibly created and removed by light soaking and annealing, respectively, in a manner consistent with the SWE. At low temperatures, this light-induced effect is typical of the excess specific heat that is characteristic of amorphous materials, representing TLS and an excess T^3 term. At high temperatures, the light-induced increase in C suggests that additional localized modes are created. The increase in the T^3 term and the high temperature C are not due to an increase in the Debye specific heat as the sound velocity is unaffected by light soaking. Since the states also occur in the non-hydrogenated material, hydrogen is not required for this process but increases the probability of it occurring by lowering the energy of the final state or the energy barrier for the formation of the low-energy excitations. It is likely that the DBs directly associated with the SWE are therefore part of a structural rearrangement involving tens of atoms.

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