Hydrogen-Free Amorphous Silicon with No Tunneling States

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The ubiquitous low-energy excitations, known as two-level tunneling systems (TLSs), are one of the universal phenomena of amorphous solids. Low temperature elastic measurements show that e-beam amorphous silicon (a-Si) contains a variable density of TLSs which diminishes as the growth temperature reaches 400 °C. Structural analyses show that these a-Si films become denser and more structurally ordered. We conclude that the enhanced surface energetics at a high growth temperature improved the amorphous structural network of e-beam a-Si and removed TLSs. This work obviates the role hydrogen was previously thought to play in removing TLSs in the hydrogenated form of a-Si and suggests it is possible to prepare "perfect" amorphous solids with "crystal-like" properties for applications.

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Amorphous solids are commonplace in daily life and important for a wide range of technological applications. Yet, our understanding of these materials is limited when compared with topologically ordered solids where lattice periodicity uniquely determines the relationship between microscopic structures and macroscopic properties. For amorphous solids, there are few established techniques capable of identifying nonperiodic configurations. The lack of long-range order may also be a source of phenomena not generally seen in crystalline solids [1]. One such example is the low-energy excitations (LEEs) [2] found in amorphous solids at low T, where only the long wavelength propagating phonons that give rise to the T^3 Debye specific heat are expected. While, in crystals, defects usually have characteristic energies, these LEEs have a broad and almost energy independent distribution. They are the source of anomalous thermal, elastic, and dielectric properties of amorphous solids [3], such as a linear T dependent specific heat, T^2 thermal conductivity below 1 K, and an almost T independent plateau in internal friction at a few degrees K. These properties are thought to be universal due to the quantitative similarities for a wide variety of amorphous solids. The model of two-level tunneling systems (TLSs) provides a phenomenological description for the LEEs by assuming that tunneling can occur between the minima of neighboring double-well potentials in the amorphous energy landscape [4,5], but the model does not address the microscopic origin of the tunneling entities and the cause of their universal behavior. The nature of the LEEs remains one of the unsolved mysteries in condensed matter physics.

The consensus is that the LEEs, as well as the other universal phenomena of amorphous solids such as the stretched-exponential relaxation at the glass transition [6] and relaxation at the rigidity percolation threshold [7], all

have their origins in the topology of the potential energy landscape. Yet, a microscopic understanding is missing for each of them. The difficulty lies in finding a common mechanism that would support these universal phenomena among a wide variety of amorphous solids, which differ in every aspect, from microstructure to chemical composition. In this regard, a more relevant question is whether these phenomena are intrinsic to the amorphous state. In this work, we show this is not true.

The first amorphous solids without TLSs were found 15 years ago in a type of hydrogenated amorphous silicon (a-Si:H) thin film containing 1 at. % H prepared by hotwire chemical-vapor deposition [8]. Subsequent research was interpreted as showing that the meticulous incorporation of a small amount of H eliminated TLSs by passivating dangling bonds [9]. Now, we show that a-Si thin films prepared by electron-beam (e-beam) evaporation without H at a growth temperature (T_{sub}) of 400 °C, comparable to that of a-Si:H with 1 at. % H, also contain few TLSs, despite still containing significant numbers of dangling bonds. This result points to the structural order as the cause for the disappearance of TLSs, suggesting it is possible to prepare "perfect" amorphous solids without TLSs. This is encouraging, as TLSs in dielectric amorphous thin films have become a bottleneck in an array of cutting edge technologies; they cause both elastic and dielectric losses. TLSs limit the elastic quality factor in both nanomechanical and quantum resonators [10,11] and are the dominant decoherence source in superconducting quantum bits [12,13].

The a-Si studied in this work were grown by e-beam evaporation at $T_{\rm sub}$ between 45 °C and 400 °C [14]. All films were approximately 300 nm thick. Companion a-Si films deposited at the same time or in identical conditions

were examined by Raman spectroscopy, electron and x-ray diffractions, and high resolution cross section transmission electron microscopy (HR XTEM); all films are found to be fully amorphous. The Raman spectra of e-beam a-Si films deposited at $T_{\text{sub}} = 45 \,^{\circ}\text{C}$, 200 $^{\circ}\text{C}$, and 400 $^{\circ}\text{C}$ are given in the Supplemental Material of Ref. [14], which shows that the peak caused by the transverse-optical vibrational modes at 480 cm^{-1} narrows and sharpens with increasing T_{sub} , indicating a progressive reduction of the rms distribution of bond angles of a-Si [15]. The films with $T_{\text{sub}} = 45 \,^{\circ}\text{C}$ and 400 °C were further examined with HR XTEM, and the images are also given in the Supplemental Material of Ref. [14]. No lattice fringes are found in the HR XTEM of either film, confirming the amorphicity of the films. Low resolution XTEM reveals that both films have a columnar structure oriented along the growth direction, as is commonly seen in evaporated amorphous films. For the 400 °C film, the average column size is around 14 ± 4 nm, with clear intercolumnar boundaries. The microstructure of the 45 °C film is harder to quantify, as no clear boundaries are discernible but the oriented growth is visible; the average column size in this film is less than 5 nm, and the film lacks well defined intercolumnar regions. Analysis of Rutherford backscattering shows mass density $\rho = 2.02$, 2.14, and 2.18 g/cm^3 for films with $T_{\text{sub}} = 45 \,^{\circ}\text{C}$, $200 \,^{\circ}\text{C}$, and 350 °C, respectively, and $\rho = 2.17$ and 2.22 g/cm³ for the two films with $T_{\rm sub} = 400\,^{\circ}\text{C}$.

We measured the complex elastic properties, including the real (shear modulus $G_{\rm film}$ and relative change of speed of sound $\Delta v/v_0$) and the imaginary (internal friction $Q_{\rm film}^{-1}$) parts of e-beam a-Si films deposited at different $T_{\rm sub}$ by using the double-paddle oscillator (DPO) technique [16]. We calculate the average shear modulus by subtracting resonant frequency with films from their backgrounds at $T < 30~{\rm K}$ where the resonant frequency is relatively temperature independent. These low temperature values are about 5% higher than room temperature ones. The low temperature shear moduli are shown in the inset of Fig. 1(a); $G_{\rm film}$ increases with $T_{\rm sub}$ but still below that of polycrystalline Si [17].

The relaxational scattering of elastic waves by TLS contributes to the temperature independent plateau in internal friction Q_0^{-1} , which is described by the tunneling strength C in the TLS model, as [18]

$$Q_0^{-1} = \frac{\pi}{2}C,\tag{1}$$

where C is defined as

$$C = \frac{\bar{P}\gamma^2}{\rho v^2},\tag{2}$$

and \bar{P} is the spectral density of the tunneling states, γ the energy with which they are coupled to phonons, ρ the mass density, and v the speed of sound. With the exception of

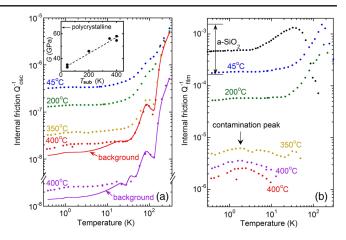


FIG. 1 (color online). (a) $Q_{\rm osc}^{-1}$ of DPOs carrying e-beam a-Si. The two films with $T_{\rm sub}=400\,^{\circ}{\rm C}$ with their respective backgrounds (solid lines) are shown in a y-axis shifted view. The inset shows $G_{\rm film}$ vs $T_{\rm sub}$. The dashed line is a guide to the eye. (b) $Q_{\rm film}^{-1}$ of the e-beam a-Si, and a 107 nm thick dry thermal oxide for comparison. The label "contamination peak" denotes that a large portion of $Q_{\rm film}^{-1}$ for films with $T_{\rm sub}=350\,^{\circ}{\rm C}$ and $400\,^{\circ}{\rm C}$ includes contamination induced loss in the DPO substrate, not related to the a-Si films. The double arrow denotes the "glassy range." See the text for details.

 $a ext{-Si}(:H)$ [8], Q_0^{-1} varies within a range of about 1 order of magnitude for most amorphous solids, often called the "glassy range." The universal magnitude of Q_0^{-1} is surprising, given that the individual parameters that constitute C, like mass density (ρ) or elastic constants (ρv^2) , may vary by many orders of magnitude [18]. Thus, a measurement of Q_0^{-1} directly reveals the universality of the LEEs as well as the spectral density of TLSs.

The internal friction of DPOs $Q_{\rm osc}^{-1}$ carrying *e*-beam *a*-Si films deposited at $T_{\rm sub} = 45\,^{\circ}\mathrm{C}$, 200 °C, 350 °C, and 400 °C are shown in Fig. 1(a). To calculate the internal friction of the films $Q_{\rm film}^{-1}$, the background internal friction of the DPOs $Q_{\rm sub}^{-1}$ was separately measured. We only show $Q_{\rm osc}^{-1}$ with their respective Q_{sub}^{-1} for the two 400 °C films in Fig. 1(a) with the y axis shifted for clarity. Thermoelastic loss dominates Q_{sub}^{-1} above 50 K [19] while external losses, such as attachment loss, are thought to dominate below 50 K. The Q_{film}^{-1} data are shown in Fig. 1(b). For a comparison with prototypical amorphous solids, Fig. 1(b) also shows the Q_{film}^{-1} of a 107 nm thick dry thermal oxide (a-SiO₂) film grown directly on a DPO at 1100 °C. The double arrow shows the glassy range which is the range of Q_0^{-1} found in most other amorphous solids. The Q_{film}^{-1} decreases with increasing T_{sub} . The Q_{film}^{-1} with $T_{\text{sub}} = 45\,^{\circ}\text{C}$ agrees with earlier results [9], as it sits right at the bottom of the glassy range. However, the Q_{film}^{-1} with $T_{\text{sub}} = 200 \,^{\circ}\text{C}$ is smaller than that of an e-beam a-Si film annealed at 350 °C for 5 h after deposition at room temperature [9], indicating that elevated T_{sub} , or surface energetics, is more effective than annealing to remove TLSs (a similar effect was seen in work on a-Tb-Fe [20]). As $T_{\rm sub}$ continues to rise, $Q_{\rm film}^{-1}$ drops rapidly, reaching an almost undetectable level above 10 K for the 400 °C films. Note that the sample with higher ρ has lower $Q_{\rm film}^{-1}$.

It is clear from both Figs. 1(a) and 1(b) that for the films with $T_{\rm sub}=350\,^{\circ}{\rm C}$ and $400\,^{\circ}{\rm C},~Q_{\rm film}^{-1}$ below 20 K is not only very low but is also not flat in temperature, as one would expect from the TLS contribution. This is caused by contamination of DPOs upon annealing. It is known that DPOs are extremely sensitive to contamination by surface dopants at elevated temperature [21]. These dopants have energy-split ground states that cause phonon scattering and elastic dissipation [22]. The broad contamination peak, also seen in the DPOs carrying a-Si:H films [8,9], is determined by the species of contaminants and has been observed before at 0.06 K [22] and 0.2 K [21]. Because of this substrate effect, Q_{film}^{-1} for films with $T_{\text{sub}} = 350 \,^{\circ}\text{C}$ and 400 °C shown in Fig. 1(b) no longer represents the internal friction of these e-beam a-Si. The important observation is that the internal friction plateau characteristic of TLSs diminishes as $T_{\rm sub}$ increases. Since $Q_{\rm osc}^{-1}$ for both 400 °C films is within the experimental uncertainty of their respective backgrounds above 20 K, we presume a detection limit of $\Delta Q_{\min}^{-1} = 5 \times 10^{-10}$ and calculate an upper bound of $Q_{\text{film}}^{-1} \leq 2 \times 10^{-7}$. This is comparable to that of a-Si:H with 1 at. % H [18] and is 3 orders of magnitude smaller than the glassy range shown in Fig. 1(b).

Another way to confirm that the e-beam a-Si films lose their glassy behavior with increasing $T_{\rm sub}$ is to measure $\Delta v/v_0$. The characteristic features of $\Delta v/v_0$ by resonant and relaxational scattering by TLSs occur at lower temperatures than we have used in this work. However, $\Delta v/v_0$ is known to vary linearly with temperature in amorphous solids from a few K up to a few tens of K [23]: $\Delta v/v_0 =$ $-\beta(T-T_0)$, where β is proportional to C and Q_0^{-1} for all amorphous solids studied [24]. This dependence at higher T is understood as the thermally activated relaxation rate of the TLS dominating the quantum tunneling rate [25]. Data of $\Delta v/v_0$ of the same e-beam a-Si and the thermal oxide films shown in Fig. 1(b) are shown in Fig. 2. Similarly to the $Q_{\rm film}^{-1}$ shown in Fig. 1(b), β diminishes to an undetectable level with increasing $T_{\mathrm{sub}}.$ The large β of the thermal oxide film is consistent with its high Q_{film}^{-1} .

Since elastic measurements always probe $\bar{P}\gamma^2$ [Eq. (2)], it is theoretically possible that the decrease of Q^{-1} comes from a diminishing phonon coupling γ of the LEEs. This is, however, ruled out by the results of specific heat from 2–300 K on similarly prepared e-beam a-Si films [14], where a similar reduction of TLS density with increasing $T_{\rm sub}$ was reported [14]. While, above 2 K, C_P is less sensitive to TLSs than Q^{-1} , these results together convincingly confirm a significant reduction of the density of TLSs in a-Si. At the same time, the results of Q^{-1} rule out

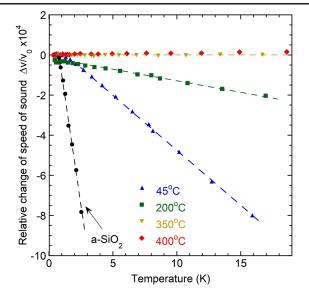


FIG. 2 (color online). $\Delta v/v_0$ of the *e*-beam *a*-Si films, and a 107 nm thick dry thermal oxide film for comparison. The straight lines indicate the linear temperature dependence explained in the text.

anomalous excitations other than TLSs as the source of non-Debye specific heat at low temperatures.

One obvious conclusion of this work is that H is not a necessary ingredient for the absence of TLSs in a-Si. Earlier studies show a strong dependence of the magnitude of Q_0^{-1} on H content [9]. The present work shows that dangling bonds and their passivation by H have no effect on TLS density, as ESR measurements show that the dangling bond density is $\sim 10^{18}$ cm⁻³ in the *e*-beam *a*-Si with $T_{\rm sub} = 400$ °C [14] vs $\sim 10^{16}$ cm⁻³ in *a*-Si:H with 1 at. % H despite similarly low TLSs [8]. As H content is mostly controlled by $T_{\rm sub}$, we suggest that the a-Si:H results should be reinterpreted as a $T_{\rm sub}$ dependence due to surface energetics. High thermodynamic and kinetic stability and high density have been demonstrated in amorphous benzene films deposited at $T_{\rm sub} \approx 0.85 T_q$ [26]. It is an unlikely coincidence that the disappearance of TLSs occurs at comparable $T_{\rm sub}$ for both. In fact, ruling out H as a necessary contributor points to the structure of a-Si as the cause for the disappearance of TLSs, namely, the formation of a dense tetrahedrally bonded network. This supports the suggestion made by Phillips 40 years ago, in which he speculated that TLSs may originate from an "open structure" with "low coordination" [5]. He argued that if every atom is linked to more than two neighbors (like in a-Si and a-Ge), the structure is overconstrained and double-well potentials are unlikely. Previous work has shown empirically that adding H reduced the dangling bond density and the TLS. In this work, we demonstrate that these effects can and should be considered separately. A densely packed and near perfect tetrahedrally bonded amorphous system can be physically constructed without requiring H by growing a-Si at a higher T, which we suggest was the dominant reason for removing the TLSs in both a-Si and a-Si:H.

In light of this work, we propose two possible scenarios for the origin of TLSs: rigidity and heterogeneity. The rigidity scenario is based on Phillips's suggestion. Although TLSs have been found in all amorphous systems, significant reduction or removal (by orders of magnitude) has only been found in overconstrained ones [8,27]. We may therefore divide TLSs into two types: The first occur in fundamentally underconstrained amorphous structures where most atoms have extra degrees of freedom due to low coordination, with some possessing double-well potentials which enable tunneling at low temperatures. These are called intrinsic and constitute the "universal" phenomena of amorphous solids. The other originate from imperfections in an overconstrained amorphous network that is rigidly bonded, like in a-Si, where, according to Phillips, TLSs should not exist. They are called extrinsic. Like defects in crystals, the density of the extrinsic TLS is variable, sensitive to methods of preparation and postdeposition treatments. Elevated T_{sub} may be just one way to experimentally achieve a near perfect high density state without the extrinsic TLS. More work is needed to understand the nature of the extrinsic TLS in a-Si; the present work shows that they are not related to dangling bonds [14]. We note that other overconstrained amorphous systems, such as a-Si₃N₄ [27], a-Ge, and a-C [28], also have Q_0^{-1} that are variable and lower than normal, suggesting that the TLSs in these amorphous systems may also be extrinsic. As the average coordination number is important to other universal phenomena of glasses, it may be that the same potential energy landscape that causes the LEEs is also responsible for glass-forming ability, glass transition temperature, stretched-exponential relaxation, and elastic properties [29].

The heterogeneity scenario comes from the correlation between mass density and the density of TLSs observed here and in the specific heat study [14]. Similar mass density dependence of TLSs has also been found in a-As_xSe_{1-x} [30] and in compacted a-SiO₂ [31,32]. It is known that a-Si cannot be made more dense than c-Si, and a larger mass density deficit ($\sim 10\%$) has been linked to the formation of voidlike structures [33]. As amorphous solids are generally less dense than their crystalline counterparts, it may be that TLSs originate from the low density regions of an amorphous network. In the heterogeneity scenario, TLSs may form as a result of free volume available on the atomic scale [34]. Here, rigidity is less important and all TLSs are intrinsically connected to low density regions, common in all amorphous materials. Even what we called above the extrinsic TLSs may have their origin in heterogeneity. Contrary to rigidity, we may significantly reduce TLS density in any amorphous solid, including those with low atomic coordination, by removing low density regions that cause tunneling.

This is reminiscent of the free volume model [34] where TLSs are formed inside microscopic voids. Recent

scanning tunneling microscope measurements have shown two states' dynamics on surfaces of a-Si involving a cluster of 4–6 atoms [35]. It is possible that similar behavior involving tunneling could happen at low T within the voidlike structures. The local structure of a-Si at the 1–2 nm length scale, where TLSs are believed to originate, has been an area of active research. Fluctuation electron microscopy shows evidence of a heterogeneous structure in a-Si that includes paracrystallites, but this interpretation has been debated [36,37]. Similar signs of such a structure in metallic glass Zr₅₀Cu₄₅Al₅ have been reported as well [38]. We note that as the needle shaped low density regions visible in the low resolution XTEM occur in films both with and without TLSs, it is unlikely that these regions are directly related to TLSs. We believe that the origin of TLSs lies instead in a nanoscale heterogeneous microstructure, not directly visible by XTEM. Recent theoretical work suggests that amorphous solids consist of local regions of enhanced bond angle regularity embedded in a more disordered matrix with TLSs forming at the interfaces between those regions [39]. We propose that further structural investigations include comparison of a-Si with low and high mass density. To distinguish between these two scenarios, both overconstrained (e.g., a-Si and a-Ge) and underconstrained (e.g., a-SiO₂) amorphous systems need to be studied by preparing each under conditions that lead to different atomic densities. The outcome (rigidity vs heterogeneity) will depend on whether TLSs can be removed only in overconstrained (rigidity) or in both the overconstrained and underconstrained systems (heterogeneity).

To conclude, this work demonstrates that TLSs can be removed from hydrogen-free a-Si by proper optimization of deposition conditions, thus revealing a clear dependence of TLSs on macroscopic parameters and microstructure. This opens up an avenue to further elucidate the microscopic origin of the universal LEEs as well as the other universal phenomena of amorphous solids. Our result supports the notion that a-Si can be made a "perfect glass" [1] with "crystal-like" properties, thus offering an encouraging opportunity to use it as an easy-to-prepare alternative to crystalline materials in applications, such as in modern quantum devices where TLSs are the source of dissipation, decoherence, and 1/f noise.

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