Decoupling between propagating acoustic waves and two-level systems in hydrogenated amorphous silicon

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Specific heat measurements of hydrogenated amorphous silicon prepared by hot-wire chemical vapor deposition show a large density of two-level systems at low temperature. Annealing at 200 °C, well below the growth temperature, does not significantly affect the already-low internal friction or the sound velocity, but irreversibly reduces the non-Debye specific heat by an order of magnitude at 2 K, indicating a large reduction in the density of two-level systems. Comparison of the specific heat with the internal friction suggests that the two-level systems are uncharacteristically decoupled from acoustic waves, both before and after annealing. Analysis yields an anomalously low value of the coupling constant, which increases upon annealing but still remains anomalously low. The results suggest that the coupling constant value is lowered by the presence of hydrogen.

Amorphous solids show anomalous thermal, elastic, and dielectric properties at low temperatures. The observation of anomalous thermal properties, through thermal conductivity and specific heat measurements [1], led in 1972 to development of the standard tunneling model (STM) [2,3], in which atoms or groups of atoms tunnel between nearly degenerate configurations with a distribution of asymmetries Δ that are separated by energy barriers on the order of tens of kelvins with a distribution of tunneling parameter λ, resulting in tunneling-induced states with splittings <1 K. The simplest description of these configurations and the resulting states is as a two-level systems (TLSs). The STM was further developed during the following years, providing an explanation for various anomalous low-temperature properties [4–10]. The specific structures that enable the TLSs are generally not known, but have been proposed for some specific materials, such as silica [11]. Recently, we suggested that TLSs in hydrogenated amorphous silicon (a-Si:H) are primarily due to clustered atomic H in low-density regions [12].

The STM assumes that the asymmetry between states, Δ, is small, and that the tunneling parameter λ is uniformly distributed over a range of values, which allows the TLS distribution function, $P(\Delta, \lambda)$, to be written as $P(\Delta, \lambda) \approx \bar{P}$, where $\bar{P}$ is the TLS density. Under this assumption, the specific heat, $C_P(T)$, of an ensemble of TLSs has a linear term, c_1, associated with the TLS density, n_0, of the TLSs that equilibrate with the phonon bath on the time scale of the measurement by

$$C_P(T) = c_1 T = \frac{\pi^2}{6} \frac{k_B^2 N_A}{n_{at}} n_0 T, \qquad (1)$$

where n_{at} is the total atomic density, k_B is Boltzmann's constant, and N_A is Avogadro's number. The specific-heat-derived TLS density n_0 is proportional to the TLS density $\bar{P}$ through a relationship first proposed by Black and Halperin [6] and Black [7], and later experimentally verified [13,14], which establishes

$$n_0 = \frac{1}{2} \bar{P} \ln\left(\frac{4t}{\tau_{min}}\right), \qquad (2)$$

where t is the measurement time ($\approx$10 ms for the nanocalorimetric system used in this paper at low T) [15] and τ_{min} is the TLS minimum relaxation time. At low temperatures, typically below 10 K, the main TLS relaxation mechanism is the one-phonon process, in which τ_{min} is minimized when the energy difference between two TLS states $E = \sqrt{\Delta^2 + \Delta_0^2} = \Delta_0$, where Δ_0 is the TLS tunnel splitting. This condition implies that the minimum relaxation time is achieved by symmetric TLSs ($\Delta = 0$). In dielectric glasses, the main contribution to specific heat and mechanical loss comes from TLSs with energy splitting $E = k_B T$, and their minimum relaxation time is given by [16–18]

$$\tau_{min} = \left(\frac{\gamma_\ell^2}{v_\ell^5} + 2\frac{\gamma_t^2}{v_t^5}\right)^{-1} \frac{\pi \rho \hbar^4}{k_B^3} T^{-3} = a T^{-3}, \qquad (3)$$

where subscripts ℓ and t denote the longitudinal or transverse wave polarization, respectively, which from now on and as a generalization are denoted by the subscript α. γ_α is the coupling constant between TLS and phonons, v_α is the sound velocity, ρ is the mass density, and T is the system temperature. Typically, $a \approx 10^{-8}$ s K^3 [16,19], and therefore $\tau_{min} \approx 1$ ns at 2 K. Note that because of the logarithmic time dependence in Eq. (2), the ratio $n_0/\bar{P}$ depends only weakly on the measurement time t and on the TLS minimum relaxation time τ_{min}, such that $n_0/\bar{P} \approx 10$ for essentially all experimentally realized measurements [20].

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The interaction between phonons and TLS at low temperature leads to energy loss, which may occur by resonant or relaxation mechanisms. At temperatures above $\sim$10 K, losses arise from thermal activation over the energy barriers separating the states. At the experimental temperatures and excitation frequencies ω used in this paper, relaxation dominates over resonant mechanisms through TLSs with $\omega\tau = 1$. The interaction between a phonon and a TLS is given by the deformation potential, $\gamma_{i,\alpha} = \partial\Delta_i/2\partial u_\alpha$, where u_α is the strain tensor. $\gamma_{i,\alpha}$ thus depends on the specific TLS. In the limit assumed by the STM, a single value of the deformation potential is used for all TLSs, the coupling constant γ_α. In that limit and in the relaxation regime (when $\omega\tau_{\min} \ll 1$), the mechanical loss $Q_\alpha^{-1}(T)$ is temperature independent, typically from 0.1 to 10 K [4], and is given by

$$Q_{0,\alpha}^{-1} = \frac{\pi}{2}\frac{\bar{P}\gamma_\alpha^2}{\rho v_\alpha^2}. \qquad (4)$$

Experimentally, γ_α can be determined combining acoustic dispersion and attenuation measurements [21], or specific heat and acoustic measurements [22]. Mechanical loss, $Q_\alpha^{-1}(T)$, measurements report the imaginary part of the elastic constant, the so-called anelastic energy loss. In amorphous solids, other contributions to $Q_\alpha^{-1}(T)$ are negligible in this temperature range. Surprisingly, $Q_{0,\alpha}^{-1}$ ranges from 10^{-4} to 10^{-3} for most glasses and varies little with differing chemical compositions [23]. This "universal" glassy range is not a consequence of the STM and has been long considered one of the great mysteries of glasses. Relatively recent explanations of this effect center on the concept of self-interacting TLSs, which renormalize the TLS density and yield approximately the observed universal number [24,25].

The relative change with temperature of the sound velocity $\Delta v_\alpha/v_{0,\alpha}$ reports the real part of the elastic constant and is measured from the relative variation of the resonant frequency of the oscillator with temperature, $\Delta f/f_0$, with f_0 being the frequency at a reference temperature T_0. Elastic properties measurements have shown that the thermally activated relaxation rate of the TLS causes a reduction in $\Delta v_\alpha/v_{0,\alpha}$ proportional to temperature,

$$\frac{\Delta v_\alpha}{v_{0,\alpha}} = -\beta_\alpha(T - T_0), \qquad (5)$$

where $v_{0,\alpha}$ is the sound velocity at an arbitrary reference temperature T_0. Equation (5) is not predicted by the STM, but an extension of this model considering thermally activated relaxation processes predicts the linear dependence of $\Delta v_\alpha/v_{0,\alpha}$ with T [26]. Experimentally, β_α has been found to be proportional to $Q_{0,\alpha}^{-1}$ for a large number of systems, including amorphous solids, quasicrystals, and disordered crystals. Specifically, the average proportionality for all the studied systems yields $\beta_t/Q_{0,t}^{-1} = 0.5$ K^{-1} [27]. The results presented in this paper are obtained from transverse modes, so from now on, and unless noted, we always refer to transverse components and omit the wave polarization notation.

It has been shown that vapor-deposited films grown under specific conditions surpass constraints that limit the properties of glasses obtained from the liquid phase, producing ultrastable glasses, i.e., glasses with higher glass transition temperature and atomic density [28–30]. A correlation between atomic density and TLS density, derived from both specific heat and mechanical loss (extending far below the "universal" glassy range), in a-Si films grown under conditions suggesting ultrastability has been shown [31–33]. a-Si:H was the first material for which Q_0^{-1} values below the "universal" glassy range were reported; such values were initially suggested to be due to the presence of hydrogen [34,35] but were subsequently found as well in a-Si (without H) grown at elevated temperature [32,33]. Recently, the TLS density of a-Si:H films grown under different conditions has been studied using specific heat, $C_P(T)$, measurements [12]. The aim of this work is to compare n_0 and Q_0^{-1} measured on equivalent samples of a-Si:H in the as-prepared and annealed states, where it is known that annealing allows hydrogen to redistribute while preserving the silicon network [12], and thus obtain a better understanding of the STM.

We report measurements of specific heat $C_P(T)$, mechanical loss $Q^{-1}(T)$, longitudinal sound velocity v_ℓ, and relative change of the sound velocity $\Delta v/v_0$ on device-quality a-Si:H films prepared by hot-wire chemical vapor deposition (HWCVD), at a growth rate of 2 nm/s, and at a substrate temperature $T_S = 370\,^\circ$C, yielding films with 7% atomic hydrogen (7 at. % H). This preparation yields device-quality material with low dangling bond density [36]. Growth, infrared spectroscopy, and time-domain thermoreflectance characterizations of similar HWCVD films were discussed elsewhere [36,37]. The total atomic density (Si + H atoms) was determined by Rutherford backscattering spectrometry, in combination with a KLA-Tencor Alpha-Step IQ profilometer used to measure the films' thickness ($\sim$85 nm). These data show that the total atomic density depends nonmonotonically on growth temperature and at. % H. v_ℓ was measured at room temperature by a picosecond ultrasonic pump-probe technique [38], yielding 7.7 km/s. Oxygen resonant scattering was used to look for oxygen content, and none was detected ($<$1 at. %). Films were measured in their as-deposited state and then annealed in vacuum at $200\,^\circ$C for 10^4 s and measured again. Hydrogen forward scattering was used to measure H content, and, in particular, it proved that annealing did not remove any hydrogen from the films. Nuclear magnetic resonance (NMR) measurements show that annealing causes H to disperse more uniformly, primarily remaining in isolated Si-H bonds and sometimes forming H_2 molecules [39,40]. Clustering of H_2 can occur and cause bubbles, but this was not observed in the present films under the annealing conditions used.

$C_P(T)$ was measured, both before and after annealing, from 2 to 300 K using the small ΔT relaxation method [15] and a microfabricated thin-film nanocalorimeter [41]. $Q^{-1}(T)$ and $\Delta v/v_0$ measurements were made from 0.3 to 100 K using the antisymmetric torsional mode of a double paddle oscillator (DPO) at $\omega = 2\pi \times 5431$ Hz [42]. We note that the interaction between the TLS and phonons is dominated by relaxation processes, for both $C_P(T)$ and $Q^{-1}(T)$, in these experimental conditions. An extended set of specific heat measurements and their analysis are reported elsewhere [12], showing the effect of growth temperature, annealing, and H content on the total specific heat, atomic density, sound velocity, and TLS density n_0 for a range of HWCVD a-Si:H thin films.

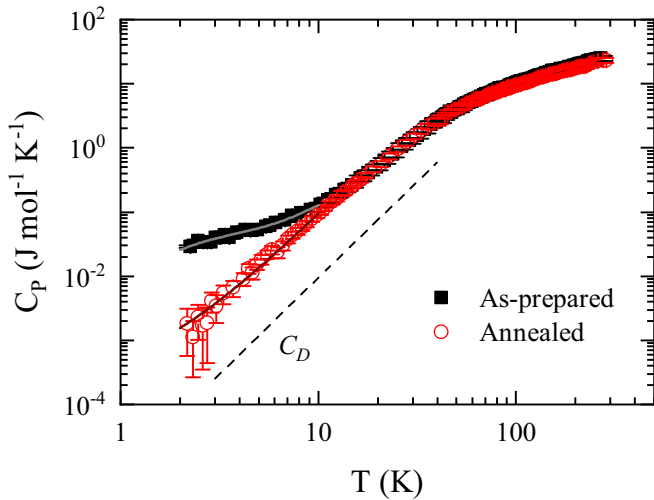Specific heat data on this 7 at. % H sample are shown in Fig. 1. The sound velocity and hence the Debye specific

FIG. 1. Specific heat of a-Si:H film grown at $T_S = 370\,°C$ (7 at. % H) in the as-prepared and annealed states. The Debye specific heat (dashed line), $C_D = c_D T^3$, is calculated from the measured sound velocity v_ℓ. The solid gray and brown lines show the fitting to the as-prepared and annealed states, respectively, using Eq. (6).

heat, C_D, due to the phonon contribution did not change on annealing; C_D is shown with a dashed line. C_D was calculated from the measured atomic density and longitudinal sound velocities as $C_D = (12\pi^4/5)N_A k_B (T/T_D)^3$, where $T_D = (\hbar/k_B)\{6\pi^2 n_{at}[1/(3v_l^3) + 2/(3v_t^3)]^{-1}\}^{1/3}$ is the Debye temperature. Transverse sound velocity was calculated using the relationship $v_t = (0.56 \pm 0.05)v_\ell$, established between longitudinal and transverse modes for amorphous materials [43], including our own work on a-Si [33].

The specific heat is significantly larger than the Debye specific heat, as is commonly found for amorphous materials. Data for the as-prepared and annealed states match above 10 K, making clear that vibrational modes are unchanged and the sample has not been modified by annealing, except for its H distribution and TLS parameters. Below 10 K, the values for the as-prepared sample are significantly larger than those of the annealed sample and show a sublinear temperature dependence, which must represent the high-temperature side of a peak in order to have nondivergent entropy as T → 0. The simplest and most likely model for this peak is a Schottky anomaly [12]. $C_P(T)$ is therefore fit to the following form:

$$C_P(T) = C_{Sch}(T) + c_1 T + c_3 T^3, \qquad (6)$$

where $C_{Sch}(T)$ is the Schottky specific heat given by $C_{Sch}(T) = N k_B \left(\frac{\Delta_0}{k_B T}\right)^2 \frac{\exp(\Delta_0/k_B T)}{[1+\exp(\Delta_0/k_B T)]^2}$, with N being the number of states with energy splitting Δ_0, c_1 is given by Eq. (1), and $c_3 = c_D + c_{ex}$, with c_D being the Debye specific heat coefficient and c_{ex} being an excess cubic coefficient that is not predicted in the STM but has also been reported for a-Si thin films [31], and is discussed at length in Ref. [12], including a discussion of the form of this fit.

As discussed in Ref. [12], after annealing, $C_P(T)$ below 10 K is significantly reduced but remains larger than c_D due to both c_1 and c_{ex}, and, in particular, the Schottky term is gone. We note that the low-T $C_P(T)$ of a-Si:H and a-Si does not depend on magnetic field, where we observed variations <1%

TABLE I. Sample state (as prepared or annealed at $T_A = 200\,°C$); Schottky specific heat C_{Sch} at 2.4 K; TLS density n_0 calculated from Eq. (1); slope of the relative change of the sound velocity with temperature, β, obtained from Eq. (5); and temperature-independent mechanical loss Q_0^{-1} at 1 K.

State	C_{Sch} $\times 10^{-3}$ (J mol^{-1} K^{-1})	n_0 $\times 10^{46}$ (J^{-1} m^{-3})	β $\times 10^{-6}$ (K^{-1})	Q_0^{-1} $\times 10^{-5}$
As-prepared	11.4 ± 3.7	209 ± 26	11.4 ± 0.2	1.07 ± 0.02
Annealed	0	29 ± 3	12.2 ± 0.5	1.15 ± 0.02

in fields up to 8 T (see Fig. 7.7 in Ref. [44]). These results confirm that electronic states are not responsible for the excess $C_P(T)$ at low temperature.

The TLS density n_0 is calculated from the data in Fig. 1 using Eq. (6) and is reported in Table I. The fits for n_0 and c_{ex} are robust to details of the Schottky fit needed to explain the as-deposited data (e.g., multiple energy levels, varying N while fixing Δ_0, degeneracy, etc.) as discussed in Ref. [12], which showed and discussed the specific heat of several samples grown at different temperatures and exhibiting different H content. We note here only that the lowest value of n_0 is obtained from the sample reported in this paper ($T_S = 370\,°C$ and 7 at. % H) both before and after annealing.

Mechanical loss, $Q^{-1}(T)$, of a-Si:H in the as-prepared and annealed states was measured using ring-down measurements of a DPO before and after deposition of the a-Si:H sample and also after annealing [42]. Results of $Q^{-1}(T)$ are shown in Fig. 2, where Q_0^{-1} starts at 1 K. $Q_0^{-1} \sim 1 \times 10^{-5}$ for the as-deposited state and increases ~8% upon annealing. For other HWCVD a-Si:H, Q_0^{-1} has been found to range from 4×10^{-7} to 2×10^{-5} [45], consistent with our data; no explanation for this variation in Q_0^{-1} has yet been suggested. Figure 2 also
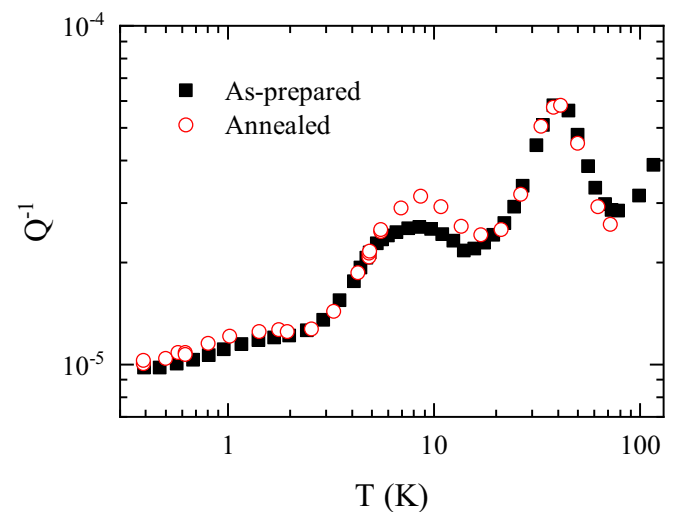

FIG. 2. Mechanical loss Q^{-1} as a function of temperature T for an a-Si:H film grown at $T_S = 370\,°C$ (7 at. % H) in its as-prepared (black solid squares) and annealed (red open circles) states. The loss difference between the bare DPO and the DPO with the film is of almost one order of magnitude, which ensures the accuracy of the measurement.
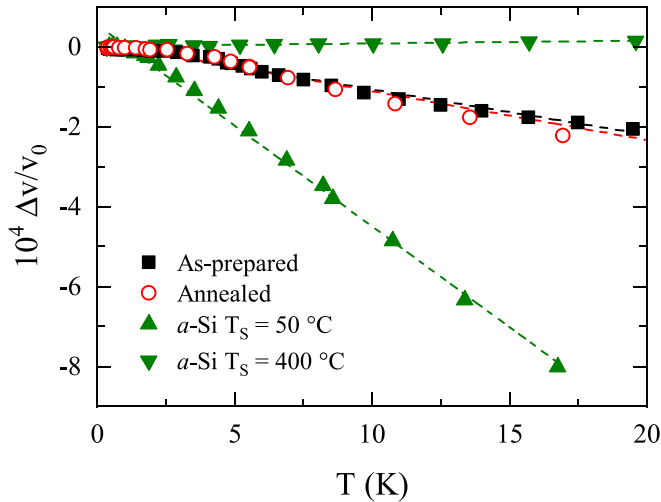
FIG. 3. Relative change of the sound velocity $\Delta v/v_0$ as a function of temperature T of a-Si:H in both as-prepared (black solid squares) and annealed (red open circles) states. For comparison, $\Delta v/v_0$ of a-Si samples grown at 50 °C (green upward-pointing triangles) and 400 °C (green downward-pointing triangles) are also shown from Ref. [32]. Dashed lines are linear fits showing β (see Table I).

shows two peaks: at 6 K (8 K after annealing) and 40 K. The peak near 6 K is likely due to H_2 in a nonbulk environment, caused by the liquid-solid transition of small clusters of H_2 in restricted geometries [46], including two-dimensional H_2 [47], which suppress the transition temperature. The lack of an H_2 triple point signature in both $C_P(T)$ and $Q^{-1}(T)$ suggests the latter possibility, specifically, that H_2 clusters exist but are small enough that they do not behave as bulk H_2. After annealing at 200 °C, the 6-K peak intensity is enhanced by almost 30% and becomes wider, which suggests an increase either in the H_2 content or in the ratio of clustered H_2 to isolated H_2, consistent with literature on the effects of annealing below 200 °C [39,40]. The 40-K peak is not seen in any a-Si:H literature of which we are aware and is unchanged by annealing; it might be associated with contamination of some type.

Data on $\Delta v/v_0$ for the a-Si:H sample are shown in Fig. 3. Again, little change is observed between the as-prepared and annealed states: $\sim 7\%$ increase in β, consistent with the 8% increase seen above in Q_0^{-1}. We note the ratio between dispersion and attenuation properties $\beta/Q_0^{-1} = 1.1$ K^{-1}, which is more than two times the average experimental ratio of 0.5 K^{-1} reported in previous works [27,45]. For comparison, $\Delta v/v_0$ for a-Si is also shown in Fig. 3 for low and high growth temperatures [32], which yields high and low TLS density, respectively, and is quantitatively consistent with the data for a-Si:H.

To go further with the analysis, we calculate $\bar{P}$ of a-Si:H from the specific heat values of n_0 using Eq. (2), along with the experimental value of $t = 10$ ms, and $\tau_{\min} \approx 1.25$ ns, obtained from Eq. (3) with $T = 2$ K, the lowest temperature of the specific heat measurements presented in this paper. This approach yields $n_0/\bar{P} \approx 9$; therefore $\bar{P} \approx 2.4 \times 10^{47}$ J^{-1} m^{-3} before annealing, and $\bar{P} \approx 3.4 \times 10^{46}$ J^{-1}m^{-3} after. We note

again that the logarithm in Eq. (2) makes the choice of $\tau_{\min}$ not important (three orders of magnitude change in $\tau_{\min}$ modifies $n_0/\bar{P}$ less than twofold).

The experimental values of Q_0^{-1} along with the values of $\bar{P}$ calculated above, and combined in Eq. (4), yield $\gamma = 7$ and 19 meV for the as-prepared and annealed states, respectively. These results, however, lead to the nonintuitive conclusion that both $\bar{P}$ and γ must change upon annealing in such a way that Q_0^{-1} barely changes. The usual approach of assuming a value for γ, e.g., 1 eV, leads to a value of $\bar{P}$ from Q_0^{-1} that is orders of magnitude less than that found from C_P measurements, i.e., to a decoupling between propagating acoustic waves and TLSs, thus not explaining why the large $\bar{P}$ found in heat capacity does not cause losses (an equivalent statement to the low γ derived on comparing the two measurements).

Very few studies have been made that estimate γ [22,43,48–51], which typically ranges from 0.1 to 1.5 eV, is considered a material constant, and has not been measured for a-Si:H. While the values of γ reported in this paper are unexpectedly low, there is nothing in the STM that prevents such low values. These low γ values are on the order of those reported in H-Nb systems [49,50], suggesting that H atoms might be responsible for the tunneling states in a-Si:H. The increased value of γ, obtained for the annealed state, and the fact that the sound velocity does not change, imply that upon annealing the minimum relaxation time decreases significantly (sevenfold) since $\tau_{\min} \propto v^5/\gamma^2$ ([see Eq. (3)]). It is remarkable that Q_0^{-1} and β, both proportional to $\bar{P}\gamma^2$, increase very slightly after annealing, whereas n_0 and thus $\bar{P}$ decrease noticeably. This apparent contradiction is mathematically solved if γ increases after annealing. We know that sound velocity does not change upon annealing, which suggests that the silicon structure is not altered. Additionally, the small increase in Q_0^{-1} and $\Delta v/v_0$ may be caused by local structural changes when hydrogen diffuses. However, if the structural rearrangement of hydrogen atoms leads, as measured by $C_P(T)$, to a reduction in TLSs that turn out to be those with the lowest γ_i values, then γ should increase. This hypothesis explains why a reduction in $\bar{P}$ upon annealing would correlate with an increase in γ, or vice versa, in a-Si:H.

Previous work has found that large variations in specific heat are often not associated with changes in thermal conductivity [52,53]. A similar inconsistency is found in $C_P(T)$ and $Q^{-1}(T)$ measurements of chemical-vapor-deposited amorphous silicon nitride, notably a material that contains of the order of 1–2 at. % H [54,55]. An alternative approach to the decoupling between specific heat and acoustic properties results was proposed by Black [7]; in this approach the samples may contain "anomalous" TLSs, besides "standard" TLSs. Black argued that anomalous TLSs relax fast enough to be measured by specific heat techniques but too slowly to be detected by acoustic properties techniques. In this approach, Black considered an additional TLS density for the anomalous states that would only contribute to the specific heat measurements. Despite the addition of anomalous TLSs, which to some extent improved the agreement between the STM and the experimental results, Black could not satisfactorily reconcile theory and results. We suggest that these apparent discrepancies are due to a misunderstanding of the coupling mechanisms between TLSs and propagating waves, i.e., the

deformation potential and, particularly, that it varies from TLS to TLS, and hence to the value of the coupling constant to be used within the STM. We *speculate* that if γ_i is not the same for all TLSs within the system, then the average value of such a distribution, γ, may depend on the relaxation times probed. We note that τ ranges from τ_{min} to ~ 10 ms for the $C_P(T)$ results and $\tau = 1/\omega \sim 30\,\mu s$ for the $Q^{-1}(T)$ results presented in this paper.

In conclusion, the TLS density measured by $C_P(T)$ in HWCVD a-Si:H is significantly larger than that measured by $Q^{-1}(T)$. Furthermore, annealing promotes hydrogen redistribution while the silicon network is preserved, which leads to a reduction by an order of magnitude in the TLS density from $C_P(T)$ while Q_0^{-1} and β slightly increase. The coupling constant of a-Si:H, derived from the STM prediction of $n_0/\bar{P} \approx 10$, yields $\gamma = 7$ meV and increases to $\gamma = 19$ meV after annealing, suggesting not only that γ is not a constant

material quantity but also that its value strongly depends on the microstructure. The low values of the coupling constant suggest that the TLSs that couple to applied elastic fields in a-Si:H are likely related to the presence of hydrogen within the amorphous silicon network. The discrepancy observed between $C_P(T)$ and $Q^{-1}(T)$ measurements shows that propagating acoustic waves are decoupled from TLSs with very low coupling constants.

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