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► **To cite this version:**

A. Fefferman, A. Maldonado, E. Collin, X. Liu, T. Metcalf, et al.. Elastic measurements of amorphous silicon films at mK temperatures. *Journal of Low Temperature Physics*, 2017, 187 (5-6), pp.654-660. 10.1007/s10909-016-1686-6 . hal-01588335

HAL Id: hal-01588335

<https://hal.science/hal-01588335>

Submitted on 15 Sep 2017

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Elastic measurements of amorphous silicon films at mK temperatures

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Received: date / Accepted: date

Abstract The low temperature properties of glass are distinct from those of crystals due to the presence of poorly understood low-energy excitations. The tunneling model proposes that these are atoms tunneling between nearby equilibria, forming tunneling two level systems (TLSs). This model is rather successful, but it does not explain the remarkably universal value of the mechanical dissipation Q^{-1} near 1 kelvin. The only known exceptions to this universality are the Q^{-1} of certain thin films of amorphous silicon, carbon and germanium. Recently, it was found that Q^{-1} of amorphous silicon (a-Si) films can be reduced by two orders of magnitude by increasing the temperature of the substrate during deposition. According to the tunneling model, the reduction in Q^{-1} at 1 kelvin implies a reduction in $P_0\gamma^2$, where P_0 is the density of TLSs and γ is their coupling to phonons. In this preliminary report, we demonstrate elastic measurements of a-Si films down to 20 mK. This will allow us, in future work, to determine whether P_0 or γ is responsible for the reduction in Q^{-1} with deposition temperature.

1 Introduction

At temperatures below a few kelvin, the thermal, mechanical and dielectric properties of glass are dominated by low energy excitations (LEEs) that are not present in crystals. The first sign of these LEEs was the discovery that the heat capacity of glass below 1 kelvin is much greater than that of crystals and has a nearly linear temperature dependence, compared with the cubic

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temperature dependence observed in insulating crystals [1]. While the identity of the LEEs remains uncertain [2,3], the tunneling model proposes that they are two level systems (TLSs) formed by atoms tunneling between nearby equilibria in the disordered lattice of the glass [4,5]. The tunneling model successfully describes many aspects of the low temperature behavior of glass, but one major shortcoming is its failure to explain the remarkable universality of the mechanical dissipation Q^{-1} of glass near 1 kelvin. The level of the Q^{-1} plateau near 1 kelvin is universal within a factor of 20 centered at 5×10^{-4} and is insensitive to chemical composition, impurity concentration, preparation of the samples including heat treatment, measurement frequency over 9 orders of magnitude and stiffness of the glass over four orders of magnitude [6]. The only known exceptions to this universality are in four-fold coordinated glasses: certain thin films of amorphous silicon, carbon and germanium have Q^{-1} up to a factor of 100 below the glassy range [6].

The origin of the reduced Q^{-1} in these materials was uncertain, but it was recently found that the Q^{-1} of a-Si films can be tuned by varying the temperature of the substrate during deposition: increasing the substrate temperature during deposition from room temperature to 400 Celsius results in a reduction of Q^{-1} of about two orders of magnitude [7]. The decrease in Q^{-1} was explained in terms of enhanced surface energetics at high growth temperatures [7]. Indeed, the high mobility of atoms within a few nanometers of the glass surface during deposition is known to produce highly stable glasses that are low on their energy landscape [8]. Despite the reduced Q^{-1} of the a-Si films deposited at high temperatures, they remain fully amorphous, as demonstrated by Raman spectroscopy, electron and x-ray diffraction and high resolution cross section transmission electron microscopy [7]. In addition to vapor deposition, the four-fold coordination of the a-Si films may also play a role in suppressing TLSs in this material [5,7].

In this preliminary report, we demonstrate low temperature elastic measurements of an a-Si film deposited at room temperature. This technique will allow us, in future work, to separately determine the density of TLSs and their coupling to phonons. A central assumption of the tunneling model is that the TLSs have a broad distribution of parameters with spectral density $P(\Delta, \Delta_0) = P_0/\Delta_0$, where Δ is the asymmetry in the depth of the two wells of the TLS and Δ_0 is the tunneling amplitude [9]. The energy splitting of a TLS is $E = \sqrt{\Delta^2 + \Delta_0^2}$. Strain ϵ in the glass has little effect on Δ_0 but changes the asymmetry of the TLSs by $\gamma\epsilon$, thus changing E . Consequently, after the glass is deformed, the fraction of TLSs in their excited states no longer satisfies thermal equilibrium and the ensemble of TLSs relaxes at a rate

$$\tau^{-1} = \frac{3\gamma^2}{v^5} \frac{E\Delta_0^2}{2\pi\rho\hbar^4} \coth(E/2k_B T) \quad (1)$$

This relaxation causes softening of the glass as well as dissipation. If all the TLSs had the same energy, a maximum in dissipation would occur when the relaxation rate of the TLS matches the driving frequency, $\omega = \tau^{-1}$. For the distribution of TLS parameters given above, the dissipation due to relaxation

is temperature independent for $\omega \ll \tau^{-1}$ and cubic in temperature for $\omega \gg \tau^{-1}$. Furthermore, a phonon can be absorbed by a subset of the TLSs that have energies matching the frequency of the phonon. This causes significant dissipation when $\hbar\omega \geq k_B T$ (not the case in this work) as well as a contribution to the sound speed that increases logarithmically with temperature.

Liu *et al.* observed a plateau in Q^{-1} of a-Si films between 300 mK and 10 K. The films deposited at room temperature had $Q^{-1} = 2 \times 10^{-4}$ in this temperature range. According to the tunneling model, the plateau occurs because $\omega \ll \tau^{-1}$ for the subset of TLSs that dominate the mechanical response, i.e., those with $E \approx \Delta_0 \approx k_B T$. The value of Q^{-1} at this plateau is $Q_0^{-1} = \pi P_0 \gamma^2 / 2\rho v^2$, where ρ is the mass density of the glass and v is its sound speed [10]. As the temperature decreases, we expect a maximum in the sound speed at the temperature where $\omega \approx \tau^{-1}$. The maximum occurs because the contribution of relaxation to the softening becomes negligible compared with that of resonant phonon/TLS interactions at the lowest temperatures.

2 Experimental Details

The sound speed and Q^{-1} of the a-Si film was measured using a double paddle oscillator (DPO) like the one pictured in the inset of Fig. 1. The DPO has been characterized in detail in previous work [11, 12, 10, 13]. It is etched from a 300 μm thick wafer of crystalline silicon. Of the ten lowest modes of vibration, the AS2 mode around 5.5 kHz has by far the lowest Q^{-1} , which reaches below 10^{-8} at the lowest temperatures. The other modes have Q^{-1} around 10^{-6} at low temperatures. The exceptionally low Q^{-1} of the AS2 mode is due to the low strain amplitude at the clamping position. In this mode, the strain is concentrated in the upper torsion rod, called the neck, where the sample is deposited. A gold or platinum metal film is deposited on the wings, leg and foot of the DPO to facilitate electrostatic drive and detection of the motion and to thermalize it. When the DPO is installed in its electrode structure, each wing is separated from an electrode by a $\approx 100 \mu\text{m}$ gap, forming two of the capacitors represented in the circuit in Fig. 1. The DPO is clamped in an Invar block, whose low thermal expansion coefficient minimizes the risk of cracking the DPO while cooling. A calibrated carbon resistor was screwed to the side of the Invar block for thermometry.

The resonant frequency of the DPO was measured using the ringdown technique with the circuit in Fig. 1. We first applied a driving voltage across the capacitor formed by the drive electrode and the grounded metal film on the DPO. The driving voltage was at half the DPO resonant frequency since the force on the DPO was proportional to the square of the drive voltage. To initiate the ringdown, the driving voltage was reduced by a factor of 250 and its frequency was increased to 10 mHz above the DPO resonance, so that the AS2 mode was effectively undriven. On the detection side, a voltage source was used to bias the detection electrode at voltage V . A blocking capacitor was added to protect the preamplifier from the bias voltage. Due to a 10 M Ω resistor

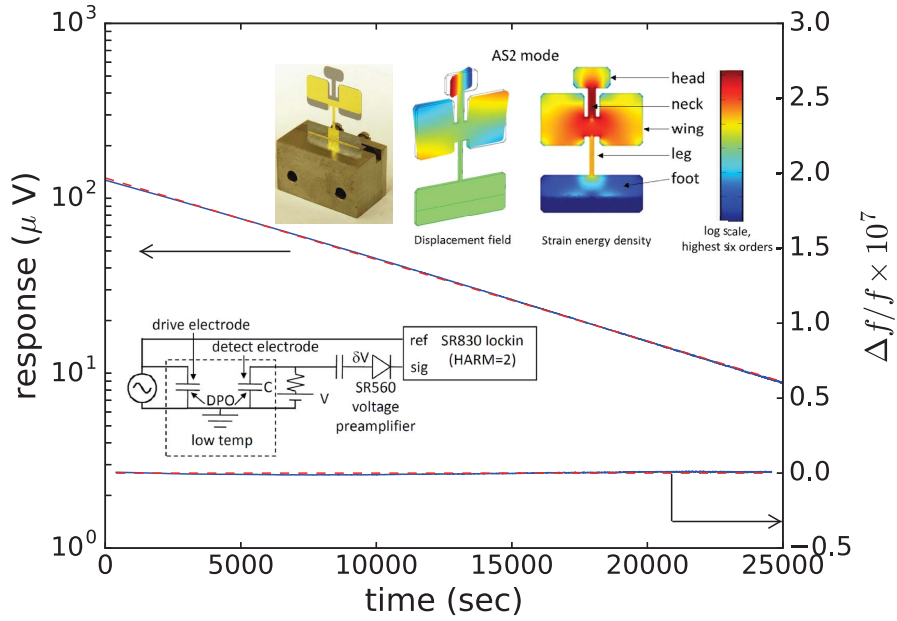


Fig. 1 Inset: Photograph of a silicon double paddle oscillator (DPO) and calculation of its motion. The gold film is used for electrostatic drive and detection of its motion and does not cover the neck, which is the sample region. Also shown is the circuit used for drive and detection. Main Panel: Measurement of a bare DPO (i.e. without a sample) at 10 mK. The free decay of its response (left axis) and the corresponding time evolution of its resonant frequency shift $\Delta f/f$ (right axis) are shown. The horizontal dashed red line is a guide to the eye and the other dashed red line is a linear fit, yielding $Q^{-1} = 6.0 \times 10^{-9}$. The variation of $\Delta f/f$ during the ringdown was negligible compared with the temperature dependence of this quantity (Fig. 2). (Color figure online.)

in series with the voltage source and the $100 \text{ M}\Omega$ input impedance of the preamplifier, the charge on the detection electrode remained nearly constant on the time scale of the DPO oscillations. Consequently, the fluctuations δV of the voltage across the capacitor C formed by the detection electrode and the grounded DPO were proportional to the capacitance fluctuations δC caused by the motion of the DPO: $\delta V/V = -\delta C/C$. The quantity δV was measured using the “Harm=2” setting of an SR830 dual phase lockin amplifier. This setting causes it to generate an internal reference signal at a frequency f_{ref} that is twice the frequency of the signal input to its reference port, which was output by the function generator. The lockin output the phase θ of δV relative to the internal reference signal. We determined the oscillation frequency of the DPO during the ringdown (i.e. its resonant frequency f_{res}) using $f_{\text{res}} = f_{\text{ref}} + (1/2\pi)d\theta/dt$.

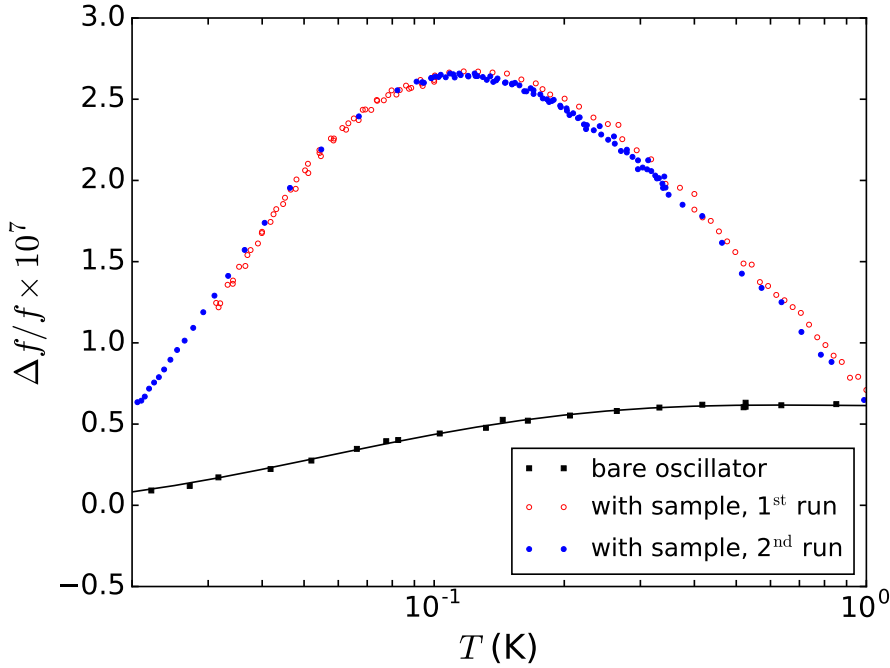


Fig. 2 Temperature dependence of the resonant frequency shift $\Delta f/f = (f - f_0)/f_0$ of a bare oscillator (called DPO1) [black] and the same DPO with a 336.1 nm thick amorphous silicon film deposited on its neck at 22 deg Celsius [blue and red]. The DPO was warmed to room temperature between the acquisition of the blue and the red data. The reference frequency f_0 has three different values for the three data colors. (Color figure online.)

3 Results and Discussion

The amplitude of δV during the ringdown of a bare DPO at 10 mK is plotted as a function of time in Fig. 1. As expected, it has a nearly perfect exponential time dependence, as shown by the dashed red line. The corresponding time dependence of f_{res} is also shown in the figure. The horizontal red dashed line shows that the response of the DPO is linear at these strain magnitudes and that fluctuations in f_{res} are small compared with the temperature dependence of f_{res} , which will be discussed below.

Figure 2 shows the temperature dependence of the resonant frequency of the bare DPO obtained from measurements like those shown in Fig. 1 as well as that of the DPO with an a-Si film. The coincidence of the red and blue data points, which came from two different runs starting from room temperature, demonstrates the reproducibility of the temperature dependence.

In order to determine the temperature dependence of the sound speed in the film, the contribution of the bare paddle was subtracted as follows. The resonant frequency of the AS2 mode scales with the square root of the torsion constant of the DPO neck $\kappa_{\text{neck}} \propto wt^3$, where w and t are respectively

the width and thickness of the neck. Consequently, the frequency shift upon depositing a sample on the DPO neck is [13]

$$\frac{f_{\text{osc}} - f_{\text{sub}}}{f_{\text{sub}}} = \frac{3t_{\text{film}}G_{\text{film}}}{t_{\text{sub}}G_{\text{sub}}} \quad (2)$$

where f_{osc} and f_{sub} are respectively the frequencies of the film-laden and bare DPO, and G_{film} and $G_{\text{sub}} = 62$ GPa are respectively the shear moduli of the film and the substrate. Since $t_{\text{film}} = 336.1$ nm and $t_{\text{sub}} = 300\mu\text{m}$, this equation can be used to calculate G_{film} . The solid black line in Fig. 2 is a polynomial fit to f_{sub} that was used for this purpose. The resulting $\delta v_{\text{film}}/v_{\text{film}} = \delta G_{\text{film}}/2G_{\text{film}}$ is plotted in Fig. 3. The blue and the red data correspond to measurements at two different initial strain levels (each ringdown measurement lasted 500 seconds), and the coincidence of these data demonstrates that the measurements were made in the regime of linear response. The intrinsic non-linear response due to TLSs was analysed theoretically in [14], and it will be studied experimentally in this material in future work. Figure 3 also shows Q^{-1} of the a-Si film studied here as well as Q^{-1} of an a-Si film measured by Liu *et al.* [7], which was deposited at a slightly higher temperature (45 deg C). As expected, the Q^{-1} of the film of Liu *et al.* is slightly less than but almost the same as that of the present film. The superposition of these data demonstrate that our measurements of Q^{-1} are consistent.

It has been proposed that, in a-Si, TLSs can only exist at imperfections in the four-fold coordinated atomic structure [5, 7]. This is in contrast to a-SiO₂, which has a more open structure where most atoms have extra degrees of freedom. Despite these structural differences, the temperature dependence of the sound speed in a-Si below 1 kelvin (Fig. 3) is qualitatively similar to that of a-SiO₂ [15]. Indeed, in a-Si $\delta v_{\text{film}}/v_{\text{film}}$ has a maximum at $T_{\text{co}} = 100$ mK and a logarithmic temperature dependence above and below T_{co} . From this result, we can make a rough estimate of γ , which could not be determined in the higher temperature measurements of [7]. As explained above, the contribution to the mechanical susceptibility from relaxation of TLSs is dominated by symmetric TLSs with energy splittings approximately equal to the temperature of the glass [16]. Setting $\Delta_0 = E = T = T_{\text{co}} = 100$ mK in Eq. 1 and setting $\tau^{-1}(T_{\text{co}}) = \omega$ yields $\gamma \approx 1$ eV, compared with $\gamma = 0.8$ eV in [15]. A more precise determination of γ in a-Si requires a fit of the tunneling model to both $\delta v_{\text{film}}/v_{\text{film}}$ and Q_{film}^{-1} , which will be carried out in future work.

Acknowledgements We acknowledge support from the ERC CoG grant ULT-NEMS No. 647917 and from the US Office of Naval Research.

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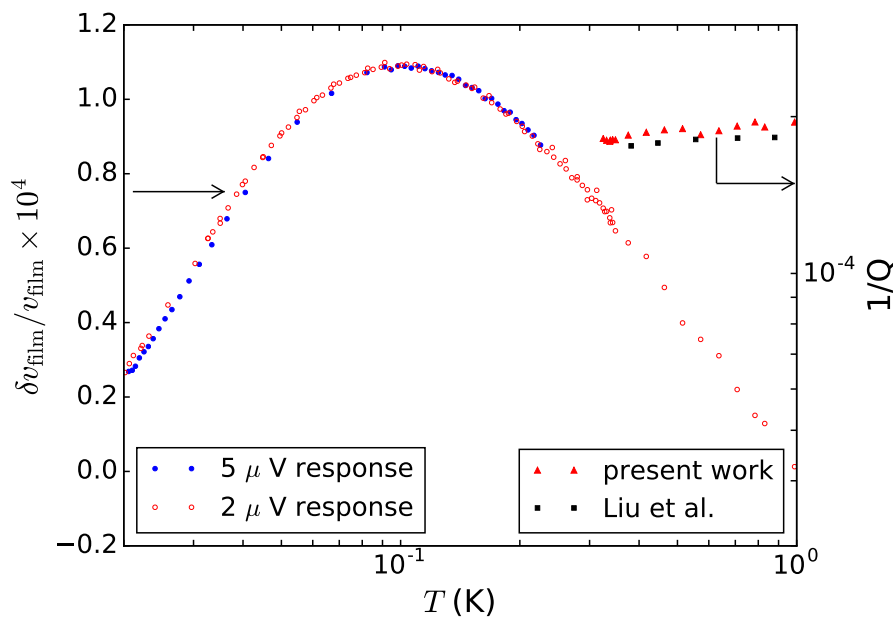


Fig. 3 Temperature dependence of the relative change in sound speed $\delta v_{\text{film}}/v_{\text{film}} = (G_{\text{film}} - G_0)/2G_0$ in the amorphous silicon film, as determined from the measurements in Fig. 2 using Eq 2. We set $G_0 = 37$ GPa. The lack of dependence of the sound speed on the response amplitude demonstrates that the measurements were made in the linear regime. Also shown is the agreement between Q^{-1} of the present film and that measured in [7] (see text). (Color figure online.)

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