Low temperature dissipation scenarios in palladium nano-mechanical resonators

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We study dissipation in Pd nano-mechanical resonators at low temperatures in the linear response regime. Metallic resonators have shown characteristic features of dissipation due to tunneling two level systems (TLS). This system offers a unique tunability of the dissipation scenario by adsorbing hydrogen ($H_2$) which induces a compressive stress. The intrinsic stress is expected to alter TLS behaviour. We find a sub-linear power law $\sim T^{0.4}$ in dissipation. As seen in TLS dissipation scenarios we find a logarthmic increase of frequency characteristic from the lowest temperatures till a characteristic temperature $T_{co}$ is reached. In samples without $H_2$ the $T_{co} \sim 1K$ whereas with $H_2$ it is clearly reduced to $\sim 700mK$. Based on standard TLS phenomena we attribute this to enhanced phonon-TLS coupling in samples with compressive strain. We also find with $H_2$ there is a saturation in low temperature dissipation which may possibly be due to super-radiant interaction between TLS and phonons. We discuss the data in the scope of TLS phenomena and similar data for other systems.

Nano-electromechanical systems (NEMS) are not only sensitive transducers but also form an excellent platform to explore basic physical phenomena. The spectrum of phenomena include potential macroscopic quantum states[1], electron-phonon coupling[2] and mechnano-spintronic phenomena[3]. Scaling of NEMS to smaller sizes (or higher frequencies) results in higher dissipation[4]. Typically GHz frequency devices that can satisfy the rudimentary quantum condition $\hbar\omega > k_BT$ at dilution fridge temperatures are limited by intrinsic losses despite geometric aspects like clamping loss at boundaries are overcome by free-free resonators designs[5] or small phonon bottle neck devices[6]. Photon pressure in microwave cavities can also squeeze MHz frequency NEMS resulting in occupation numbers close to the ground state of a quantized harmonic oscillator[7]. So far only one dilation-mode resonator has shown evidence for macroscopic quantum behaviour in mechanical systems[8]. Intrinsic loss mechanisms in NEMS are not yet fully understood and are crucial to understand decoherence of quantum phenomena[9]. At temperatures below $4.2K$ almost all materials freeze except helium which forms a quantum liquid due to overlap of nuclear wave-functions[10]. One may naively expect most solids will show uninteresting behaviour in mechanical response at these temperatures. On the contrary the mechanical responses of solids do vary vibrantly even at temperatures down to even below $T \leq 3mK$[11]. Tunneling two level systems (TLS) have been used to model low temperature mechanical dissipation in bulk solids successfully[10][13]. Some of the key parameters of a phenomenological TLS is shown as a schematic in Fig[1]. Apart from the parameters for an isolated TLS as shown in Fig[1] the overall energy landscape of TLS like the distribution function for TLS energies $P(E, \Delta_0)$, distribution of typical relaxation times $\tau_s$ for tunneling (both $P(E, \Delta_0)$ and $\tau_s$ are functionally related) and how the TLS interact with each other and interaction with quasi-particles like phonons gives rise to unique behaviour for various classes of systems. These models have been successful in explaining many experimentally accessible properties of bulk solids both amorphous and crystalline. The experimentally studied properties usually have a Kramers-Kroning type dispersion for a susceptibility $\chi = \chi_r + i\chi_im$ containing a real (dissipative) and imaginary (dispersive) response, the dissipation $Q^{-1}$ and relative frequency shift $df/f_0$ for measurements in this work. In systems like amorphous silicon films hydrogenation resulted in lower dissipation possibly due to alteration of

FIG. 1. (a) Schematic of a TLS. The key parameters are barrier height $V$ the barrier asymmetry $\Delta$ the relaxation time $\tau$ and the TLS phonon coupling parameter $\gamma$. The tunnel amplitude $\Delta_0 \sim h\Omega e^{-^\lambda}$ where $\lambda = d\sqrt{2m\Omega}/\hbar^2$ m is particle mass like term. TLS splitting energy $E^2 = \Delta^2 + \Delta_0^2$. Possible changes in TLS parameters (b) increase of barrier height (c) enhanced phonon coupling.
the co-ordination of the amorphous network. Recent studies have also shown density dependent voids to scale with TLS density. TLS models for amorphous glasses are general enough to be mapped to several crystalline and polycrystalline systems. In polycrystalline solids one may map the variables to several potential TLS candidates like grain boundary angles, kinks and dislocations but with different energy scales for parameters like phonon TLS coupling, density of TLS etc. The analogy is like crystalline materials showing spin-glass states. Hence it is not hard to conceive of pseudo-spin glass states for TLS scenarios. TLS models predict a range of values for quantities like mechanical dissipation e.g. $Q^{-1} \sim 10^{-3} - 10^{-5}$ for amorphous dielectrics with some exceptions in stressed systems like silicon nitride. The possibility of universality in semiconducting NEMS was suggested in ref. The motivation for studying $Pd$ in normal and superconducting states. Stand alone metallic nano-mechanical systems are indeed simpler systems to study dissipation. It was demonstrated that tensile stress in these systems increases the quality factor (Q-factor) in gold nano-mechanical resonators were studied showing evidence for TLS mechanisms. TLS mechanisms possibly due to quasi 1-D phonon mediated dissipation was seen in aluminum in normal and superconducting states.

In this work we report our studies on mesoscopic $Pd$ beams. The motivation for studying $Pd$ is to probe a system where dissipation scenarios may be modified significantly intrinsically without external dissipation dilution. Palladium’s affinity to adsorb $H_2$ is well known. In ref. nano-scale $Au - Pd$ beams have been used as hydrogen sensors by probing frequency shifts due to adsorbed $H_2$. The $H_2$ not only covers the surface but also forms $H^+$ ions that occupy interstitial sites in the $Pd$ resulting in a compressive strain on the $Pd$ lattice structure. Compressive or tensile stress can affect the barrier height $V$ as in Fig. or TLS phonon coupling constant $\gamma$ as in Fig. Metallic beams at cryogenic temperatures have intrinsic tensile stress due to differential thermal contraction with respect to the substrates. Our goal is to tune this tensile stress with exposure to $H_2$ thereby modifying TLS scenarios. Intrinsic tensile stress is known to drastically alter the TLS scenario in systems like silicon nitride.

The experiments were carried out in a cryo-free dilution fridge. A separate brass vacuum can with homemade RF feed-through were used to introduce an exchange gas of $H_2$. Typical samples had a length ($l$) of $4 - 5 \mu m$, thickness ($t$) around $80 \text{nm}$ and a width ($w$) of $450 - 470 \text{nm}$ were fabricated by e-beam lithography on $Si/\text{SiO}_2$ wafers and undercutting the $\text{SiO}_2$ in buffered oxide etch. The samples were bonded by mechanically pressing indium coated gold wires on to the chip and to micro-strip tracks with RF launchers. We used a standard magneto-motive technique to probe the resonant response. RF current from a vector network analyzer was driven through the sample with a magnetic field parallel to the wafer plane to excite and detect out of plane motion of the beam due to the Lorentz force.

In initial trials we found samples did not survive thermal cycling to room temperature. Hence a set of two samples $Pd4B1L$ ($\sim 4.35 \mu m \times 390 nm \times w$) and $Pd4B1R$ ($\sim 4.35 \mu m \times 366 nm \times w$) forming a RF bridge were cooled in $H_2$ exchange gas of $10^{-3} \text{torr}$ and again with $10^{-2} \text{torr}$ and subsequently pumped below $10^{-4} \text{torr}$ when the mixing chamber temperature was below $160 \text{K}$. In the second round of exposure to $H_2$, $Pd4B1R$ was heated with a $0.5 \mu A$ low frequency current from room temperature down to $160 \text{K}$. This was unstable in frequency with time-scale several hours to few days possibly due to excessive adsorption of $H_2$ causing additional diffusion induced dissipation and the data is not discussed here. The frequencies of the samples were estimated as in ref. accounting for tension due to differential thermal contraction of the substrate and sample at $4.2K$. In both cases the resonant frequency was less than the estimated $24 \text{MHz}$.

A second set of two samples $Pd2C3L$ ($4.5 \mu m \times 430 nm \times w$) and $Pd2C3R$ ($4.5 \mu m \times 470 nm \times w$) were studied in the absence of $H_2$ separately after pumping the system to $\sim 4 \times 10^{-5} \text{torr}$ over one day and cooled down while pumped continuously with a turbo pump. The samples had resonant frequencies of $19 \text{MHz}$ and $29 \text{MHz}$. Although sample dimensions were comparable, $Pd2C3R$ trapped some indium in the etched region below it thereby reducing its effective length to $\sim 3.5 \mu m$ and the predicted frequency matched for this length. An electron microscope image after measurement along with an EDS scan confirmed presence of Indium. The first sample showed some buckling that possibly explains a reduced frequency from the estimated $23 \text{MHz}$.

A Lorentzian fit to the real and imaginary part of the response was used to extract the loaded Q-factor $Q_l$ and resonant frequency $f_l$. As expected in standard magneto-motive technique the eddy current damping showed a linear dependence for $B^2$ vs $Q_l^{-1}$ in all cases satisfying the relation $Q_l^{-1} = Q_0^{-1} (1 + \alpha B^2) = Q_0^{-1} \left[ 1 + \frac{B_0^2}{2\pi f_c} \right]$ where $R_m = \frac{d^2 B_0^2 Q_0}{2\pi f_c m}$ is the mechanical equivalent of re-
sistance depending on resonator parameters (length, frequency and mass and intrinsic Q-factor $Q_0$). The frequency squared also showed a quadratic dependence in field due to presence reactive components like a bias-tee that protected the samples from static charge. The field dependence was used to estimate the intrinsic $Q_0$ and frequency $f_0$ from loaded values measured at 4 T. There was not a significant change in the magneto-motive damping parameter $\alpha$ when $H_2$ was added with all samples showing $\alpha$ was $1.33 - 4 \times 10^{-6}$ $T^2$ and non monotonic with $H_2$. The optimal power to measure in the linear response regime was $\sim -90$ dBm (when pre-cooled with $\sim 2 \times 10^{-3}$ torr of $H_2$, hereafter referred as low $H_2$) and $\sim -110$ to $-105$ dBm (when pre-cooled with $\sim 10^{-2}$ torr $H_2$, hereafter referred to as high $H_2$) and $\sim -80$ dBm when cooled without $H_2$. In all cases the $H_2$ exchange gas was reduced to a pressure of $\sim 10^{-4}$ torr when the system reached a temperature $\sim 160 \text{K}$. This change in linear response regime clearly indicates softening of the beams with $H_2$ due to additional compressive stress.

The data in the absence of $H_2$ for the 19 & 29 MHz samples are presented in Fig2. The frequency shift shows a logarithmic increase with raise of temperature till a characteristic temperature $T_{co} \sim 1 \text{K}$ is reached in both samples and the slope is negative beyond this point. This is similar to ac susceptibility of spin glasses or rather a pseudo spin glass in our case where one may identify a characteristic relaxation time $\omega_0 \tau \sim 1$ at this temperature where $\omega_0$ is the device resonant frequency. In the absence of electrons in dielectric glasses this may be interpreted as a crossover from a domination of resonant TLS interactions in the low temperature $\omega_0 \tau \geq 1$ to a relaxation dominant regime $\omega_0 \tau \leq 1$. In dielectric glasses the slopes on either side are expected to have a ratio 1.5 whereas this is not valid in systems like metallic glasses33. In well studied metallic glasses 33 like PdSiCu there is relaxation due to phonons and possibly conduction electrons as well. The cross-over temperature is much higher $\sim 1.5 \text{K}$ which is large compared to dielectric glasses where it is $\sim 0.2 \text{K}$. In ref 31 the $T_{co}$ is not a simple cross over from resonant to relaxation as it does not scale significantly with frequency for a device of 1000 Hz and 1 GHz whereas dielectric glasses do satisfy a frequency scaling, $\omega T_{co}$, being a constant. We do not have several decades of scaling as in 31, but we do not see a $\Delta T_{co} \sim 75 \text{mK}$ between a 19 MHz and 29 MHz beams. A similar cross over $\sim 1 \text{K}$ was

FIG. 3. (a)$Q^{-1}$ of two samples exposed to $\sim 10^{-3}$ torr $H_2$ during cool-down. A sub-linear fit with $\sim T^{0.37}$ for the 19 MHz and $\sim T^{0.43}$ for the 20 MHz sample is shown for reference. (b) $\frac{\omega}{f_0}$ for both these samples are shown with a logarithmic fit of 1.9 $\times$ $10^{-5}$ and 1.6 $\times$ $10^{-5}$. The last three points show a small deviation from the log fit are corrected by using fit from higher temperatures shown by * symbols. The $Q^{-1}$ in (a) retains the low temperature saturation despite the corrections.

FIG. 4. (a)The dissipation of the stable 20 MHz samples exposed to $\sim 10^{-2}$ torr $H_2$ during cool-down. A sub-linear fit with $\sim T^{0.43}$ is shown for reference. (b) $\frac{\omega}{f_0}$ with a log fit of 1.54 $\times$ $10^{-5}$

FIG. 2. (a) Damping $Q^{-1}$ as a function of temperature on the top graph. A power law of $\sim T^{0.39}$ for the 19 MHz and $\sim T^{0.41}$ for the 29 MHz sample is shown as a guide. (b) $\frac{\omega}{f_0}$ in these samples with a reference frequency $f_0$ at an arbitrary temperature $T_0$. The logarithmic slope quantifying the parameter below $T_{co} \sim 1 \text{K}$ is $C$ is $\sim 1.85$ $\times$ $10^{-5}$ for the 19 MHz and $\sim 3.5$ $\times$ $10^{-5}$ for the 29 MHz samples.
seen for Al resonators from 40 to 350 MHz \[24\]. However the slope on the right side of the crossover is not linear as seen in metallic glasses\[31\]. The ratio of slopes at the cross over is \(\sim 2\) and 2.5 as opposed to 1.5 for dielectric glasses. In poly-crystalline metals generally and also Pd films one finds \(T_{co} \sim 60\) mK as reported in \[32\]. The same work reported no dependence of the TLS phonon coupling constant \(\gamma\) on electronic mean free path and \(\gamma\) was generally 1 to 3 eV for metals. An increase of \(T_{co}\) by 50 mK for annealed Pt films was interpreted as a decrease in the TLS-phonon coupling constant \(\gamma\). Annealed films have tensile stress. Since our beams are under tensile stress, we may conclude that \(\gamma\) is reduced in comparison to bulk films. The dissipation shows a sublinear power law in both cases below the \(T_{co}\). The power law shows a weak change above \(T_{co}\). The deviation from the power law and the onset of a characteristic saturation is obvious at higher temperatures. Overall the behaviour shows features of glass like TLS models but does not fit either a dielectric or metallic glass.

The scenario with low \(H_2\) concentration Fig(3) and high \(H_2\) concentration Fig(4) is qualitatively similar with some key differences. In cases of both low and high \(H_2\) concentrations the samples showed a clear lowering of the \(T_{co}\) to around 700 mK. It is important that for devices close to \(\sim 20\) MHz in both of these samples we see a lower cross over temperature as compared to the \(H_2\) free case.

The dissipation shows a sub-linear power-law around 0.37–0.43. It is easy to adjust the power law by choosing or neglecting a few points near the \(T_{co}\) in frequency due to scattering from onset of a weaker power or plateau like feature near \(T_{co}\). But this dependence is not systematic with or without \(H_2\), hence we quote a mean of \(\sim T^{0.4}\) to describe the behavior. There were no Debye peaks\[30\] in the dissipation indicating that \(H_2\) diffusion induced mechanisms are not prominent at these temperatures which are also well below freezing point of \(H_2\). The overall order of magnitude of dissipation is not significantly lower despite of the softening seen by lower power for probing in the linear response regime. If we take the dissipation in the high temperature plateau or the highest temperature, where the data show a change to weaker dependence the constant \(C\) from TLS theory turns out to be similar order

\[
C = \begin{cases} 
\frac{Q}{\tau} Q^{-1} \sim \frac{P_{b}}{K} \sim 0.1 \times 5 \times 10^{-5} & \text{if } T > T_{co} \\
\gamma_{\nu}^{2} \frac{\nu}{|\epsilon_o|} \sim 1.5 \times 4 \times 10^{-5} & \text{if } T < T_{co}
\end{cases}
\]

in the \(H_2\) free and the low \(H_2\) scenario. This similarity is surprising considering the effective Young's modulus \(E\) has to be lower in the case of the \(H_2\) scenario as reckoned by lower drive power to keep the response linear. The only plausible explanation is either the TLS density \(P\) or phonon coupling \(\gamma\) is enhanced. The standard TLS models reckon \(\omega \sim \gamma^2 T_{co}^3\). The lowering of \(T_{co}\) in our case implies enhancement of \(\gamma\) due to additional compressive stress caused by \(H_2\) adsorption. This is reminiscent of materials like PMMA (softer glass) and silica (harder glass) showing similar low temperature dissipation due to commensurate TLS parameters for \(C\)\[26\]. Surprisingly the ratio of frequency shifts above and below \(T_{co}\) is \(\sim 1.43\) and \(\sim 1.5\) for the low \(H_2\) samples in Fig(3) similar to amorphous dielectrics.

In the low \(H_2\) scenario Fig(3) at temperatures below 200 mK one sees a saturation in dissipation. Such a feature has been predicted \[33\] as a possible super-radiant phonon emission. In some older experiments\[23\] where the temperature was corrected using the high temperature logarithmic frequency shift as a thermometer the feature merged into the power law indicative of thermal de-coupling and these features were at much lower temperatures \[23\] \[25\]. Such a correction also did not remove the feature as seen in Fig(3a). The sample with higher \(H_2\) Fig(4) shows the same feature starting earlier at \(\sim 300\) mK down to \(\sim 250\) mK where we could get the data without any need for corrections to the frequency shift. In ref\[33\] two characteristic loss mechanisms are added \(Q_{total}^{-1} = Q_{0}^{-1} + Q_{pump}^{-1}\) where \(Q_{0}^{-1}\) is the TLS mechanism and \(Q_{pump}^{-1}\) a co-operative phonon emission by a small population of TLS enclosed within phonon wavelength. The effect is a weak renormalization of TLS relaxation time \(\tau\) by \(\tau/N\) where \(N\) is the number of TLS that are cooperatively excited by a phonon. The effect is expected only at very low temperatures due to crossover to non-linear TLS-phonon coupling. The high temperature plateau are different where \(\omega\tau \sim 1\) signifies onset of inelastic processes exciting a spectrum of TLS relaxation rates. When already in the limit \(\omega\tau > 1\) we expect coherent excitation. One of the criterion for the pumping\[34\] is the asymmetry energy \(\Delta_0 \leq \gamma\epsilon_o\) where \(\gamma\) is the TLS phonon coupling and \(\epsilon_o\) some in built strain. We cannot comment on the static strain status of our devices at cryogenic temperatures but devices exposed to \(H_2\) showed some buckling when warmed up and the power to drive them linearly was significantly lower. We also inferred \(\gamma\) to be enhanced from drop in \(T_{co}\) with \(H_2\) exposure. The condition to probe more TLS by a phonon is also enhanced on a lattice with compressive strain. Other possible scenarios like modification of TLS distribution or interactions are expected at ultra low temperatures, typically below 10 mK\[11\]. In our data the small frequency shift corrections in Fig(3a) or none in Fig(4) for higher \(H_2\) sample points towards a super-radiant phonon loss as the most plausible scenario.

In conclusion we have studied a system where dissipation scenarios are tunable. There is prospect for further studies at KHz frequencies to several hundred MHz and other experimental probes like thermal transport. Further experiments in these extreme regimes have the potential to throw light on TLS like mechanisms in submicron metallic structures.

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