

Two-Level Systems Observed in the Mechanical Properties of Single-Crystal Silicon at Low Temperatures

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(Received 8 June 1987)

Using the high- Q mechanical-oscillator technique we have measured the sound velocity and mechanical dissipation of high-purity single-crystal silicon as functions of temperature (0.005–4.2 K), frequency (0.6–6.0 kHz), and strain amplitude (10^{-5} – 10^{-8}). In the mechanical properties we find a surprisingly strong temperature dependence with the same qualitative behavior for silicon as for vitreous silica. This implies a density of two-level systems only 2 orders of magnitude lower for silicon than amorphous silica. In silicon we find evidence for a new dissipation mechanism at low temperatures and report the first observation of a saturation with strain of the resonant contribution to the sound velocity.

PACS numbers: 62.65.+k, 62.20.Dc

The acoustical properties of crystalline materials at very low temperatures have not been extensively studied and the dominant dissipation mechanisms are not well understood.¹ This is despite the fact that such systems have obvious importance to such areas as gravity-wave detection and high-precision measurement.² Indeed it is not at all clear in most experiments on high-quality single crystals whether the observed mechanical dissipation is intrinsic to the material or due to extrinsic experimental difficulties.

In this Letter we report careful measurements of the low-temperature (0.005–4.2 K) mechanical properties of high-quality single-crystal silicon. We have been able to show conclusively that in this temperature range the dominant contribution to the mechanical dissipation arises from the presence of two-level systems (TLS). These occur in silicon with a density equal to roughly 1% of the density of TLS found in pure glass. This low density keeps the overall dissipation in our system small unlike pure glasses and allows us to follow its behavior to very low temperatures. Because of this feature we are able to make quantitative comparisons over a wide range in temperature with the theory developed for describing the acoustical properties of TLS. We have found several new results. In contradiction to current theory we find that at low temperatures the dissipation varies linearly with temperature as opposed to the T^3 dependence expected for phonons, and we have for the first time observed a saturation with strain of the resonant sound velocity. The similarity of the features shown here to those seen and not previously understood in other high- Q systems suggests that we now know what limits the intrinsic mechanical Q in a wide variety of systems at low temperatures.

The method we have used is the low-frequency, high- Q mechanical-oscillator technique described in more detail in Ref. 2. Using photolithographic patterning and anisotropic etching techniques we have fabricated double torsional pendulums from thin wafers of single-crystal silicon (see inset in Fig. 1). A double torsional pendulum

design was chosen in order to assess the significance of clamping losses. In the design shown, clamping losses are diminished by a factor of ≈ 1000 in the high-frequency torsional mode as compared to the low-frequency torsional mode.² We have found that the two modes behave in the same qualitative manner, allowing us to rule out the significance of clamping losses. In fact our oscillator as shown has seven clearly identifiable high- Q resonances in the range 0.6–6.0 kHz, all of which show the same qualitative features. This oscillator design has been useful in the study of the frequency dependence of the effects to be described and is analogous to a Birnboim multiple resonator.³ In addition, measurements were performed as a function of wafer thickness and served to rule out the importance of sur-

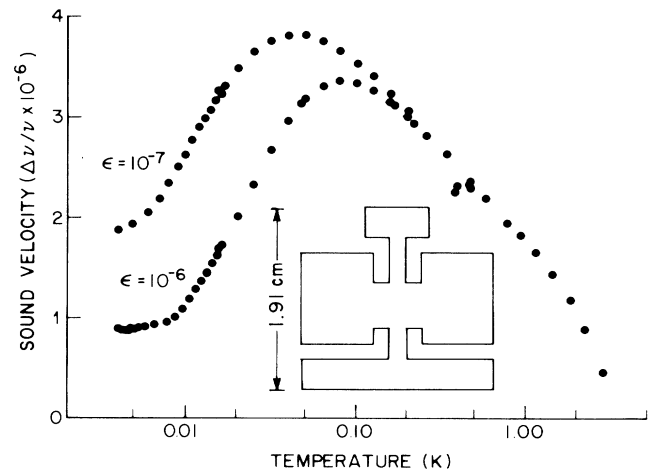


FIG. 1. The sound velocity $\Delta v/v$ vs T for two oscillation amplitudes (10^{-7} and 10^{-6}). $\Delta v/v$ is highly nonlinear in the region $T < T_{co}$. Data are from a 2164-Hz resonant mode. While the reference point for the overall velocity changes is arbitrary, no offset has been made between the two sets of data. Inset: Oscillator design and dimensions. The thickness of the sample used is ≈ 0.033 cm.

face effects and show that what we see are bulk, intrinsic properties of the silicon.

The oscillators are driven and their motion detected electrostatically in a phase-locked loop. The resonant frequency and amplitude give us the dissipation (Q^{-1}) and sound velocity ($\Delta v/v$) directly with a negligible correction for linear expansion. To address strain-dependent effects, in the data shown here, the oscillator was driven in a constant-amplitude mode.

The silicon used for the data presented here was float-zone ultrapure single-crystal silicon from Hughes Aircraft. It had a resistivity of $30 \text{ k}\Omega \cdot \text{cm}$ with approximately 4 ppma oxygen, 0.2 ppma carbon, and 0.6 parts per 10^9 (atomic) electronically active impurities as determined from a measure of the minority-carrier lifetime.

Shown in Fig. 2 are the changes in sound velocity (top) and dissipation (bottom) for our best silicon sample as a function of temperature (note that the tempera-

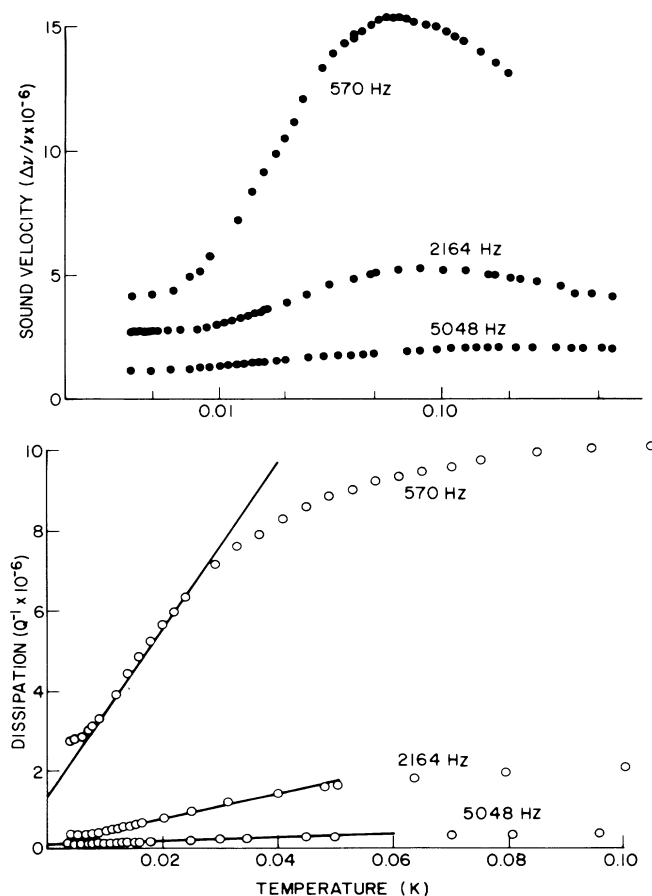


FIG. 2. The sound velocity, $\Delta v/v$ (top), and mechanical dissipation, Q^{-1} (bottom), vs temperature for three frequencies (570, 2164, and 5048 Hz). All data are from the same sample. Straight lines are drawn through the low-temperature dissipation data. Note the different temperature scales.

ture scales are not the same).⁴ At high temperatures the dissipation is roughly constant and the sound velocity is increasing slowly. However, below a crossover temperature $T_{co} \approx 0.1 \text{ K}$ the dissipation decreases markedly. The sound velocity passes through a maximum at $\approx T_{co}$. These results are surprising for a number of reasons. First, it is unusual to see the sound velocity in a pure, single crystal soften at low temperatures. Secondly, one would intuitively expect that as the dissipation decreases the sound velocity would *increase*.

Our data can be understood qualitatively as indicating the presence of a large number of two-level systems in our samples. Indeed the behavior shown in Fig. 2 is just what is seen for the acoustical properties of vitreous silica⁵ except for an overall scale change of ≈ 100 . What we will show below is that this low density of defects in our system as compared to a glass allows us to measure the behavior over a wide temperature range and quantitatively to compare it to the standard theories of the acoustical properties of glasses. We find that despite qualitative agreement the theories are incapable of explaining our results in several fundamental ways.

The interaction of ultrasonic phonons and two-level systems is composed of two different kinds of interactions, resonant and relaxation absorption.⁶⁻⁹ They are not coupled and the effects simply add and the magnitudes are proportional to the density of TLS. The contributions from the two mechanisms give rise to changes in the sound velocity and dissipation. The effects predicted by theory are summarized in Table I.

In the resonant case, ultrasonic phonons, of energy $\hbar\omega$, are absorbed by TLS of comparable energy, and reemitted after a time τ_1 , leading to dissipation and changes in the sound velocity. When the dimensionless strain, ϵ , is large compared to some critical strain, ϵ_c , all energy levels with $E \approx \hbar\omega$ are saturated and the absorption vanishes. Since $\hbar\omega \ll kT$ in our type of experiment, the resonant absorption is insignificant as compared to the relaxation absorption, except at extremely low temperatures which are not relevant to our experiment. At ultrasonic frequencies both effects are observable.

In the case of relaxation absorption, the ultrasonic phonons modulate the tunneling parameters disturbing the equilibrium of the thermal phonons. The absorption has a different temperature dependence, according to whether the modulations in the strain field at frequency ω occur faster or slower than the fastest relaxation time for a given energy splitting τ_m . When $\omega \ll \tau_m^{-1}$, at high temperatures, the phonons relax quickly enough to follow the modulation due to the strain field, giving rise to a constant dissipation and a logarithmic temperature dependence. When $\omega \gg \tau_m^{-1}$, at low temperatures, there is a changing phase lag between the TLS and the strain field, giving rise to a stronger temperature dependence for both the dissipation and sound velocity. The relaxation occurs via the dominant elementary excitation. In a

TABLE I. Predictions for the temperature and frequency dependence of the sound velocity, $\Delta v/v$, and mechanical dissipation, Q^{-1} , arising from the interaction of phonons with TLS. Here we have defined $C_D = n_0 D^2 / \rho v^2$, $C_M = n_0 M^2 / \rho v^2$; $\langle M^2 / v^5 \rangle_{av} = M_l^2 / v_l^5 + 2M_t^2 / v_t^5$, where l and t denote longitudinal and transverse phonon polarization; and $I = \int_0^\infty x^6 dx / (e^x - e^{-x})^2 \approx 5.72$. In the case of the resonant velocity, T_0 is an arbitrary reference temperature, but for the high-temperature relaxation velocity, T_0 corresponds to the crossover temperature, T_{co} . K is a dimensionless coupling constant describing the interaction between the TLS and electrons (typically $K \approx 0.0015$). The remainder of the terms are defined in the text.

Interaction	Effect	$\omega \gg \tau_m^{-1} (T < T_{co})$	$\omega \ll \tau_m^{-1} (T > T_{co})$
Resonant	Q^{-1}	$C_M \tanh(\hbar \omega / 2kT) (1 + \epsilon^2 / \epsilon_c^2)^{-1/2}$	
Resonant	$\Delta v/v$	$C_M \ln(T/T_0)$	
Relaxation	Q^{-1}	Phonons, $\frac{\pi^3 C_D}{24} \frac{\langle M^2 / v^5 \rangle_{av}}{\rho \hbar^3} \frac{k^3 T^3}{\hbar \omega}$	$\frac{1}{2} \pi C_D$
		Electrons, $\frac{\pi^3 C_D}{24} K^2 \frac{kT}{\hbar \omega}$	
Relaxation	$\Delta v/v$	$\frac{16IC_D}{\pi^2 \rho \hbar^3} \frac{\langle M^2 / v^5 \rangle_{av}}{\rho \hbar^3} \frac{k^6 T^6}{\hbar^2 \omega^2}$	$-\frac{3}{2} C_D \ln(T/T_0)$

dielectric material, for phonons, the dependence of Q^{-1} goes as T^3 ; in a metal, for electrons,¹⁰ the dependence goes as T . The crossover temperature between the high- and low-temperature regimes is most easily defined by our equating the two dependences. For the dissipation with phonons this predicts $T_{co} \sim \omega^{1/3}$.

The details of the dependences rely only on a uniform density of states and the correct elementary excitation. There are essentially no parameters which are sensitive to the details of the material. Thus any data taken for a dielectric glass at a given frequency should look virtually the same. The extent to which theory and experiment agree then tells us about violations of any assumptions, such as an energy dependence to the density of states.

Because of the low frequencies (kilohertz) of our experiment throughout the entire temperature range, we will only get a contribution to the dissipation from relaxation processes. At high temperatures above T_{co} we see a broad plateau region up to 4.2 K as expected by theory which predicts a T^0 dependence in this regime. Below T_{co} we find $Q^{-1} \sim T$. The theory predicts $Q^{-1} \sim T^3$ if the dominant relaxation is due to phonons and $Q^{-1} \sim T$ through electrons. At ultrasonic frequencies $Q^{-1} \sim T^3$ has been observed for dielectric crystals. Our crystals are completely insulating and the observed $Q^{-1} \sim T$ behavior suggests a breakdown of the theory at low frequencies. The predicted $T^0 \omega^0$ dependence at high temperatures and T/ω at low temperatures suggests that $T_{co} \sim \omega$. The data show this dependence reasonably well. As can be seen in Fig. 2 the plateau value of the dissipation, $\frac{1}{2} \pi C_D$, also has a strong dependence on frequency. We consistently find $C_D \sim \omega^{-1}$. The theory predicts no frequency dependence to C_D and none is seen in the ultrasound work. Thus we again attribute these deviations from theory to a breakdown at low frequencies.

The changes in sound velocity are due to contributions from *both* resonant absorption *and* relaxation absorption. At low temperatures we find a logarithmic temperature dependence with a positive slope characteristic of resonant absorption. The leveling off at low temperatures is due to sample heating and disappears at low amplitudes of oscillation. As in the relaxation absorption we find $C_M \sim \omega^{-1}$. In the high-temperature regime ($T > T_{co}$) we find that the temperature dependence is logarithmic with a negative slope. This is from the combined effects of resonant *and* relaxation absorptions. Table I predicts a logarithmic slope of C_M at low temperatures and $C_M - \frac{1}{2} C_D$ (or $\sim -\frac{1}{2} C_M$) at high temperatures which is what is seen in our experiment.

As shown above, the changes in sound velocity at low temperatures are due exclusively to resonant absorption. We have observed for the first time a saturation of the resonant sound velocity. Shown in Fig. 1 are the changes in sound velocity versus temperature for two oscillation amplitudes. For $T > T_{co}$ there is no amplitude dependence but for $T < T_{co}$ there is a pronounced decrease in the sound velocity for increasing amplitude. Note that the effect goes the wrong way for simple sample heating which we do see, however, at much lower temperatures. The nonlinear changes in sound velocity are not predicted by theory, nor have they been previously observed. We would expect that the resonant contribution to the sound velocity should be essentially unsaturable since only those energy levels with $E \approx \hbar \omega$ are saturated whereas the sound-velocity changes are due to the unsaturated $E \approx kT$ splittings.⁶ Also it is surprising that the saturation is evident only for $T < T_{co}$ whereas there is a resonant contribution to the sound velocity for both $T > T_{co}$ *and* $T < T_{co}$. Because $T_{co} \sim \omega$ this effect would be quite difficult to observe in ultrasound experiments and hence the failure of previous workers to see it.

At this point a discussion of the density of TLS is in order. All of the effects in Table I are proportional to C_D ($\sim n_0 D^2$) and C_M ($\sim n_0 M^2$), where n_0 is the density of states for the TLS and D and M are the appropriate deformation potentials (~ 1 eV). It is not possible by use of only acoustics to measure independently n_0 or D or M . However, it is useful to compare our results with similar measurements on vitreous silica. Assuming $D_{\text{silicon}} \approx D_{\text{vitreous silica}}$ we find⁹ $n_0(\text{Si})/n_0(\text{silica}) \sim 0.01$. This surprising result suggests that pure glass has a density of states of TLS only 100 times higher than the best single-crystal silicon available! Previous measurements⁹ on vitreous silicon find $D \approx M \approx 0.4$ eV which allows us to estimate $n_0 \approx 5 \times 10^{30} \text{ erg}^{-1} \text{ cm}^{-3}$. Independent measurements of the linear term in the specific heat for a different sample¹¹ find that $n_0 \sim 8 \times 10^{30} \text{ erg}^{-1} \text{ cm}^{-3}$, in reasonable agreement with our measurement. For silicon this implies $n_0 = 2 \times 10^{-4}$ states/(Si atom)·eV. With a guess of ≈ 1000 K for E_{max} this suggests that the total number of states is $\approx 10^{-6}$ states/(Si atom). This is roughly the concentration of oxygen and carbon impurities in our silicon. It should also be noted that similar measurements on a wide variety of high- Q metallic oscillators with such materials as BeCu and CuAg have seen qualitatively the same behavior¹² as is presented here for silicon. This suggests that for a wide variety of high- Q materials that the intrinsic damping mechanism at low temperatures is due to two-level systems and that very high Q 's can be achieved by cooling to below 10 mK. This work also suggests that mechanical Q measurements are a very good way to measure the concentration of electrically inactive impurities which are impossible to measure in any other way at these concentrations. Simple estimates suggest that our technique should be sensitive to electrically inactive impurities to the part-in- 10^9 level which is 3 orders of magnitude better than present techniques.

In conclusion, we have shown that in high-purity single-crystal silicon there are surprisingly large anomalies in the internal friction and sound velocity for temperatures much less than 1 K. These effects are due to the presence of two-level systems at an unexpectedly high density given the purity of the crystals. Because silicon acts as a "diluted" glass we are able to use this system to make detailed quantitative comparisons with theory over a wide temperature range. We find evidence that a new relaxation mechanism dominates the low-

temperature dissipation. We also observe for the first time the saturation of the resonant sound velocity. Both results are contrary to expectations and suggest that general modifications in TLS theory are required to explain the low-frequency, low-temperature regime. This work clearly identifies for the first time the dominant mechanical dissipation process in a single crystal at low temperatures and has technological implications for high-precision measurement with use of single-crystal silicon as well as other high- Q systems. It also suggests a useful system for the performance of detailed quantitative comparisons with the theory of two-level systems.

The authors would like to thank Brage Golding for many helpful discussions, T. Gmitter and E. Yablono- vitch for the measurement of minority carrier lifetimes, G. Kaminsky and J. D. Reppy for early contributions to this work, and D. D. Osheroff for the use of one of his dilution refrigerators as well as many helpful suggestions.

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