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

Kleiman, Agnolet, and Bishop[1] recently reported very low-temperature anomalies in the acoustic velocity and mechanical dissipation of pure single crystals at frequencies between 500 Hz and 5 kHz. They compared their results for the crystal with those observed in glasses and claimed that the differences required major revisions to tunneling state theory. In fact, an alternative analysis suggests that the results are as expected for impurities in crystals.

As a starting point, we assume that the silicon contains double-well potentials, in the absence of any information on symmetry, with a well-defined tunnel splitting Δ_0 but an asymmetry distribution $f(\Delta)$ produced by local strains and represented by a Gaussian of width Δ_1, to be determined from experiment. A well-defined tunnel splitting implies a very narrow range of relaxation times and so the results of tunneling theory as applied to glasses are no longer valid. It is clear from the fact that Q^{-1} decreases as $1/\omega$ that the relaxation times satisfy the condition $\omega\tau > 1$, and so[2]

$$Q^{-1} = \frac{\gamma^2}{\rho v^2 k_B T} \frac{1}{\omega} \int_0^\infty \frac{\Delta^2}{E^2} \operatorname{sech}^2(E/2k_BT)\tau^{-1} f(\Delta)\, d\Delta , \tag{1}$$

where $E = (\Delta_0^2 + \Delta^2)^{1/2}$ and where τ, the one-phonon relaxation time, is given by

$$\tau^{-1} = AE\Delta_0^2 \coth(E/2k_BT) , \tag{2}$$

with $A = \sum_a \gamma_a^2/2\pi v_a^5 \rho h^4$, where v_a is the sound velocity for polarization a, ρ is the density, and γ_a is the phonon coupling constant. The results, calculated numerically, agree well with Ref. 1 and can be understood by noting that for $k_BT < \Delta_1$ the range of integration is determined by the sech^2 factor to give an effective number of contributing states proportional to T. For each state the $1/T$ prefactor is canceled by the variation, approximately proportional to T, of the inverse relaxation time τ^{-1}, which leads to Q^{-1} proportional to T. At higher temperatures, $k_BT > \Delta_1$, the range of integration and hence Q^{-1} is approximately constant. This change in temperature dependence gives a value of Δ_1 of $10^{-1}k_B$. At even higher temperatures, where $\omega\tau < 1$, Q^{-1} decreases slowly with temperature.

The velocity variation is given by a combination of a resonance contribution at the lowest temperatures and relaxation above 0.2 K, the former giving a negligible contribution to Q^{-1} at these low frequences. The magnitude of the relaxation contribution is determined by Q^{-1}, but the relative importance of the resonance term depends on C [Eq. (2)] and Δ_0. For the calculations $\Delta_0 = 10^{-3}k_B$ and the value of C leads to a coupling constant of approximately 5 eV. In the absence of experimental information at higher temperatures no attempt has been made to optimize the choice of parameters.

With use of these parameters, the density of tunneling states can be calculated from the magnitude of the loss as approximately $10^{43}\,\mathrm{J}^{-1}\,\mathrm{m}^{-3}$, consistent with the value quoted by Kleiman, Agnolet, and Bishop[1] from measurements of the heat capacity. The total number of tunneling states then turns out to be surprisingly low, below $10^{20}\,\mathrm{m}^{-3}$, and significantly smaller than the quoted concentration of impurities. If the width of the asymmetry distribution arises from interactions between tunneling states and other impurities, the required impurity concentration can be estimated[2] from $\Delta E \sim \gamma^2/\rho v^2 R^3$ (where R is the average impurity atom spacing) to be of order $10^{23}\,\mathrm{m}^{-3}$ (a few ppm) for $\gamma = 5$ eV, consistent with the stated values.

One further interesting feature of the experimental results is a strain dependence observed for strains greater than 10^{-6}. This is expected in the present model, where the tunnel splitting is well defined and the asymmetry has a relatively narrow distribution, because for large strains the asymmetry is increased to such an extent that the energy of the states becomes larger than k_BT, thereby preventing them from contributing to the acoustic properties. With use of the parameters Δ_1 and γ the critical strain can be estimated from $\gamma e \sim \Delta_1$ as approximately 10^{-6}, in satisfactory agreement with experiment.

In summary, it appears that a simple tunneling model can explain the main features of these elegant acoustic experiments by use of a distribution function appropriate to a slightly disordered crystal.

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