Evidence for weakly and strongly interacting two-level systems in amorphous silicon

Liuqi Yu,1, 2, ∗ Shlomi Matityahu,3, 4 Yaniv J. Rosen,5 Chih-Chiao Hung,1, 2 Andrii Maksymov,6 Alexander L. Burin,6 Moshe Schechter,3 and Kevin D. Osborn1,7

1Laboratory for Physical Sciences, University of Maryland, College Park, MD 20740, USA
2Department of Physics, University of Maryland, College Park, MD 20742, USA
3Department of Physics, Ben-Gurion University of the Negev, Beer Sheva 84105, Israel
4Institut für Theorie der Kondensierten Materie, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany
5Lawrence Livermore National Laboratory, Livermore, California 94550 USA
6Department of Chemistry, Tulane University, New Orleans, LA 70118, USA
7Joint Quantum Institute, University of Maryland, College Park, MD 20742, USA

Quantum two-level systems (TLSs) intrinsic to glasses induce decoherence in many modern quantum devices, such as superconducting qubits and single-photon astronomical detectors. Although the low-temperature physics of these TLSs is usually well-explained by the phenomenological standard tunneling model (STM) of independent TLSs, the nature of these TLSs, as well as their behavior out of equilibrium and at high energies above 1 K, remain inconclusive. Here we measure the non-equilibrium dielectric loss of TLSs in amorphous silicon using a superconducting resonator, where energies of TLSs are varied in time using a time-dependent electric field. Our results show the existence of two distinct ensembles of TLSs, interacting weakly and strongly with phonons. Analysis within a previously proposed two-TLS model allows insight into the origins and characteristics of the two types of TLSs, contributing to the loss near equilibrium and far from equilibrium, respectively.

The universal properties of amorphous solids at low energies $T \lesssim 1 \text{K}$[1], have been attributed to tunneling two-level systems (TLSs), originating from an atom or a group of atoms that can tunnel between two adjacent structural configurations. They can be generally described by a standard tunneling model (STM)[2, 3], which assumes the existence of independent TLSs with asymmetry energy $\Delta$ and tunneling amplitude $\Delta_0$ that are universally distributed with a distribution function $P(\Delta, \Delta_0) \propto 1/\Delta_0$. It leads to an energy independent density of states (DOS) as a function of the energy splitting $E = \sqrt{\Delta^2 + \Delta_0^2}$ [2–5]. This model accounts well for most of the low-temperature properties observed in a broad range of amorphous systems[1, 6, 7] in terms of weakly interacting TLSs. Recent studies on material density[8–10], stress[11] and TLS-nucleus interactions[12] extend our knowledge of TLS origins. However, an understanding of the nature of TLSs in specific systems and of the origin of low-temperature universality in the phonon attenuation of glasses is still lacking [13, 14].

With the advent of superconducting qubits and other modern quantum devices, this gap in understanding TLSs has turned especially important, since TLSs in the materials and surfaces of these devices serve as a major channel for energy relaxation, decoherence and noise processes[15–32].

To study individual TLSs, some modern probes have been developed to manipulate individual TLSs via adiabatic adjustments in situ of applied strain or electric fields. The external field changes the TLS asymmetry energy $\Delta$, which in turn modifies the TLS energy splitting $E = \sqrt{\Delta^2 + \Delta_0^2}$, while the tunneling energy $\Delta_0$ is fixed. Using static strain and electric fields to tune TLSs in qubits or resonators provides a spectroscopy method to extract energy splittings [33], couplings to phonons [33, 34], dipole moments [35–37], relaxation and decoherence rates [38] of individual TLSs, and recently even their locations [39, 40]. These experiments probe TLSs in resonance with the device in equilibrium conditions at $\sim 5 \text{GHz}$, corresponding to energies of $\sim 0.25 k_B \text{K}$, where $k_B$ is the Boltzmann constant. Consequently, good agreement with the STM predictions is found in the majority of the cases.

A recent technique uses a time-dependent bias electric field to dynamically tune the asymmetry energy of TLSs in superconducting resonators[41]. The TLSs are swept through resonance with the resonator where they may absorb resonator photons and dissipate them after crossing the resonance. With fast bias rates and large bias amplitudes, this technique allows the study of TLSs out of equilibrium and of higher energies, where the applicability of the STM is less clear. When the non-equilibrium loss is measured in silicon nitride[41], the bias rate dependence of the dielectric loss is explained with the STM and the Landau-Zener (LZ) effect[42], which describes the scaling of loss over a range of ac fields and bias rates. The TLS dipole moment is then extracted from the data during scaling. At equilibrium (zero bias field), the TLS microwave absorption is saturated at high driving powers (average photon number $n \gg 1$) and the resonator loss decreases as a function of the driving power, as already shown five decades ago [43]. A time-dependent bias electric field $E_\text{bias}(t)$ sweeps TLSs into the resonance...
FIG. 1. Steady-state and nonequilibrium loss. a Optical image and b schematic of the superconducting resonator device, where the total capacitance, $C$, is made of a bridge of 4 equal capacitances ($C_1 = C_2 = C_3 = C_4 = C$). Bias voltage $V_b$ is applied across $C_1$ and $C_2$ (and also $C_3$ and $C_4$) in series. The inductor $L$ is coupled to a coplanar waveguide for transmission measurements. c Steady-state loss tangent $\tan \delta$ (red squares) plotted as a function of the average photon number $n$ at 20 mK. The loss fits to the standard model of TLS loss: $\tan \delta = \tan \delta_0 / \sqrt{1 + n/n_c}$, where $\tan \delta_0 = 1.6 \times 10^{-4}$ is the intrinsic material loss measured at the single photon regime ($n < 1$) and $n_c = 3.7$, is the quantum-classical crossover photon number. d Calculation of normalized non-equilibrium loss $\tan \delta/\tan \delta_0$ as a function of the dimensionless bias rate $\xi = 2V_b/\sqrt{\epsilon_0 \omega_0 h}$, based on the STM in the regime of strong saturation ($\Gamma_{1,m}/\Omega_{R0} \ll 1$) [42]. Inset: TLS-photon energy-level diagram as a function of the TLS energy bias rate $\nu$. The states $|g, n\rangle$ and $|e, n\rangle$ ($|g\rangle$ and $|e\rangle$ are the TLS ground and excited states, and $|n\rangle$ is a photon number state) are connected by an adiabatic transition (with an avoided crossing gap equal to the TLS Rabi frequency $\Omega_R$) which may lead to single-photon loss; the probability for photon absorption is $1 - e^{-\xi^2}$ (red squares) plotted as a function of the average photon number $n$. e Loss measured at various microwave source powers (-60 dBm to 0 dBm with 5 dBm increment as indicated by the arrow) as a function of $\widehat{E}_b/n$ (proportional to $\xi$). A periodic triangular bias voltage is applied, as indicated by the inset, with a fixed amplitude $E_{b,\text{max}} = 0.44$ V/μm and varying modulation frequency $f_b$, resulting in the bias rate $\dot{E}_b = 4E_{b,\text{max}}\pi f_b$. Data collapse at medium bias rates occurs below the intrinsic loss $\tan \delta_0$ (dashed-dotted black line), which scales according to LZ theory. The scaling deviates at the lowest rates, where TLS relaxation ($T_1$) processes dominate. Excess loss above $\tan \delta_0$ can be seen at $\dot{E}_b/n \gtrsim 10$ V/μm. f Electric field bias rate dependence of $\tan \delta$ at selected microwave source powers. Excess loss above $\tan \delta_0$ can be seen at fast bias rates, which reveals a data collapse at low powers $P_{ac} < -25$ dBm.

field with resonator phonons, with the number of swept TLSs proportional to $\dot{E}_b$. As a result, the dielectric loss increases with $\dot{E}_b$ up to the intrinsic loss tangent $\tan \delta_0$, i.e. the loss in the equilibrium ($\dot{E}_b = 0$) and low-power limit $n \ll 1$, where TLSs are unsaturated. It should be noted that the observations were made previously in silicon nitride[41], but are expected to apply to all amorphous materials where the STM holds. A similar fast-bias technique has been used to demonstrate a defect maser based on TLS population inversion [44] and dynamical decoupling of TLSs from a resonator by multiple coherent resonant transitions [45]. Early work on non-equilibrium measurements of dielectrics revealed small amplitude relaxations [46] which were understood from a small gap in the standard TLS density of states, formed by rare strong interactions [47], however, modern non-equilibrium measurements provide large signal variations due to the direct influence of controlled quantum transitions, providing new opportunities for TLS studies.

Here we employ the fast-bias technique of Ref. [41] but also a much higher maximum bias rate to measure the non-equilibrium loss of amorphous silicon. At low bias
rates the dielectric loss follows the theory of Ref. [42] based on the STM, similar to previous measurements on silicon nitride [41]. However, at high bias rates we observe a striking excess loss, larger than the intrinsic loss, for all applied driving powers. This contradicts the common understanding of the STM and provides strong evidence for another loss mechanism in higher energies and out of equilibrium. Following the two-TLS model proposed in Ref. [48], we analyze the data in terms of two types of TLSs, interacting weakly and strongly with phonons, with their own contributions to the total loss. To our knowledge, our observations provide the first direct experimental evidence for the existence in amorphous solids of two types of TLSs with coupling to phonons that differ by an order of magnitude.

Results

Nonequilibrium excess loss. As shown in Fig. 1a and Fig. 1b, the resonator consists of four equal bridge parallel-plate capacitors with a total capacitance, \( C \) (\( C_1 = C_2 = C_3 = C_4 = C \)). A dc bias field, \( E_b \), is applied across the capacitors. The bridge layout is effective to isolate the ac resonance energy from the bias field input port. Standard transmission measurements of \( S_{21} \) are performed on the resonator as a function of the average photon number, \( n \), or microwave resonator field amplitude \( E_{ac} = \sqrt{2n\hbar\omega_0/(eV)} \), where \( \hbar \) is the reduced Planck constant, \( \omega_0 = 2\pi \times 5.1 \text{ GHz} \) is the resonator resonance frequency, \( e \) is the permittivity, and \( V = 2925 \mu \text{m}^3 \) is the total capacitor volume. The material loss tangent \( \tan \delta \), equal to the inverse resonator quality \( 1/Q_i \), is then extracted. Figure 1c shows the steady state loss tangent, \( \tan \delta_0 \), at zero bias field as a function of \( n \). The resonator photon power dependence arises from the saturation of TLSs, and the loss tangent \( \tan \delta_0 \) is expressed as \( \tan \delta_0 = \tan \delta_1 \sqrt{1+n/n_c} \) [43], where \( \tan \delta_1 \) is the intrinsic material loss measured in the single photon limit \( (n \ll 1) \) and \( n_c \) is the quantum-classical crossover photon number. The fit yields \( n_c = 3.7 \) and \( \tan \delta_0 = 1.6 \times 10^{-4} \), corresponding to \( Q_i = 6200 \).

When the bias field \( E_b \) is varied in time (see inset of Fig. 1e), the asymmetry energy of each TLS is modified as \( \Delta(t) = \Delta(0) - 2pE_b(t)\cos \theta \), where \( p \) is the TLS dipole moment and \( \theta \) is the angle between \( \vec{p} \) and \( \vec{E}_b \). An ensemble of ground-state TLSs is swept through the resonance, described by the condition \( E(t) = \sqrt{\Delta_0^2 + \Delta^2(t)} = \hbar\omega_0 \) for each TLS, thereby leading to an enhanced loss. Close to resonance, the TLS energy changes at a rate of \( \epsilon = |\dot{E}|/\hbar \approx \nu_0\sqrt{1-|\Delta_0/\hbar\omega_0|^2}\cos \theta \), where \( \nu_0 = (2p/h)E_b \) is the maximum bias rate. The dynamics of each resonant passage is of the LZ type, where an adiabatic transition corresponds to the excitation of a TLS by absorption of a single photon (see inset of Fig. 1d). The resulting nonequilibrium loss was analyzed in Ref. [42] and is shown to be a function of the two dimensionless parameters: the standard LZ parameter \( \xi = 2\nu_0/(\pi\Omega_{R0}) \) and the ratio \( \Gamma_{1,m}/\Omega_{R0} \), where \( \Omega_{R0} = pE_{ac}/\hbar \) is the maximum TLS Rabi frequency and \( \Gamma_{1,m} \) is the maximum TLS relaxation rate. As plotted in Fig. 1d, in the regime of strong saturation \( \Gamma_{1,m}/\Omega_{R0} \ll 1 \), the predicted normalized loss \( \delta/\tan \delta_0 \) is a universal function of \( \xi \), approaching 1 in the non-adiabatic limit \( \xi \gg 1 \). It should be noted that the intrinsic loss is recovered at high bias rates provided that the TLS DOS is energy-independent, as assumed by the STM. This is understood as a result of fast LZ passage time compared to \( 2\pi/\Omega_{R0} \), such that the swept TLSs remain unsaturated by the resonator field, as in the single-photon limit.

Fig. 1e shows the measured dielectric loss tangent \( \tan \delta \) as a function of \( E_b/n \), which is proportional to the dimensionless bias rate \( \xi = 2\nu_0V/\pi^{\omega_0p} \cdot (E_b/n) \). The data demonstrate two distinct loss regimes separated by \( \tan \delta_0 \). Below \( \tan \delta_0 \) the curves show a single dependence on \( E_b/n \) for all microwave driving powers, except for small deviations at small \( E_b/n \) (or \( \xi \lesssim \Gamma_{1,m}/\Omega_{R0} \)) due to incoherent LZ transitions [41, 42]. The data collapse of the different curves is in accord with the theory of Ref. [42] and agrees with previous measurements in silicon nitride [41], suggesting that dielectric loss originates from standard TLSs. However, at higher values of \( E_b/n \), \( \tan \delta \) strikingly exceeds the intrinsic loss \( \tan \delta_0 = 1.6 \times 10^{-4} \) and reaches a maximum of \( 5.2 \times 10^{-4} \) except for the high powers, \( P_{ac} > -20 \text{ dBm} \) (see Fig. 1f). The excess loss scales with \( E_b/n \) at high driving powers, \( P_{ac} > -20 \text{ dBm} \) (see Fig. 1e), but scales with \( E_b \) at low driving powers, \( P_{ac} < -30 \text{ dBm} \) (see Fig. 1f). These distinguished saturation behaviors at small and large driving powers imply that the excess loss is due to a second type of TLSs. This saturation occurs at much higher driving powers (\( P_{ac} \sim -30 \text{ dBm} \)) compared to \( -60 \text{ dBm} \) for the saturation of standard TLSs responsible for the loss at low bias rates. It therefore suggests that the second type of TLSs has much higher relaxation rates. In addition, at small powers \( P_{ac} < -25 \text{ dBm} \) where the second type of TLSs is unsaturated, the loss shows a bias rate dependence (Fig. 1f), testifying for a time- and energy-dependent DOS of the second type of TLSs. In comparison, the non-equilibrium loss of unsaturated standard TLSs is equal to the intrinsic value \( \tan \delta_0 \) irrespective of the bias rate.

Loss from two types of TLSs. We now analyze the data in a model which consists of two types of TLSs. First, we use the data at low to intermediate bias rates to calculate the loss resulting from the standard TLSs, \( \tan \delta_1 \). Figure 2a shows the loss of Fig. 1f normalized by \( \tan \delta_0 \). Figure 2b shows a numerical calculation of \( \tan \delta/\tan \delta_0 \) based on LZ theory (see Methods and Refs. [41, 42] for details). The calculated loss at a given bias rate \( E_b \) and photon number \( n \) depends on the dipole moment \( p \) and maximum relaxation rate \( \Gamma_{1,m} \) which serve as fit-
FIG. 2. Loss analysis of the first and the second types of TLSs. a Normalized loss $\tan \delta/\tan \delta_0$ (colored circles), where $\tan \delta_0 = 1.6 \times 10^{-4}$ is the intrinsic loss at low powers and zero bias rate, plotted at each driving power (in dBm). Colored solid curves are calculations based on LZ theory within the STM [42], performed at low bias rates $E_b < 300 \text{V}/\mu\text{m}$. The calculation gives $p_1 = 11D$ ($1D = 0.21$ eÅ, where $e$ is the electron charge) and a maximum relaxation rate $\Gamma_{1, \text{m}} = 5.7 \text{MHz}$ for the standard (first) TLS type. b The excess loss $\tan \delta_2 = \tan \delta - \tan \delta_1$ (colored circles) is attributed to the second type of TLSs. Colored solid curves are theoretical calculations of $\tan \delta_2$ based on an energy-dependent DOS for the second type of TLSs. c Loss data (colored circles) corresponding to Fig. 1f are shown together with the theoretical calculations (colored solid lines), which combine both types of TLS loss.

bias rates. In Fig. 2b we plot the net excess loss $\tan \delta_2$ by subtracting the calculated standard TLS loss from the measured loss, i.e. $\tan \delta_2 = \tan \delta - \tan \delta_1$. The single dependence on $E_b$, for low driving powers $P_{\text{ac}} < -25 \text{dBm}$ then becomes apparent. The independence on driving power implies that the TLSs responsible for the excess loss are unsaturated in this power regime, such that the observed single curve is the equivalent of the intrinsic loss $\tan \delta_0$ of the standard TLSs, and will be denoted as $\tan \delta_{0, b}$. The fact that $\tan \delta_{0, b}$ increases with $E_b$ points to two features of the DOS of the contributing TLSs. First, the DOS is an increasing function of the TLS energy, because the loss is determined by the number of TLSs within the energy range $\hbar \omega_0 < E < \hbar \omega_0 + pE_{b,\text{max}}$ that are swept through resonance. Second, Since $E_b$ is varied by varying the modulation frequency $f_b$ with a fixed amplitude $E_{b,\text{max}}$, the initial energies of the TLSs that are swept through resonance are independent of $E_b$. This means that the large non-equilibrium DOS of these TLSs at the resonance energy $\hbar \omega_0$ tends to restore its small equilibrium value by some mechanism. This mechanism becomes less effective as the sweep time reduces ($E_b$ increases).

Such a scenario arises in a previously proposed two-TLS model, which divides TLSs into two groups, distinguished by their interactions with phonons [48]. As a consequence of their distinct interactions, TLSs that are weakly coupled to phonons are abundant at low energies below $\sim 1 \text{K}$ and form the standard TLSs of the STM with an approximately energy-independent DOS $\rho_1$; TLSs that are strongly coupled to phonons are characterized by an energy-dependent DOS $\rho_2(E)$ exhibiting a soft (power-law) gap at low energies. This soft gap is a result of their mutual interactions with the standard TLSs [49, 50], as dictated by the Efros-Shklovskii mechanism for long-range interacting particles in glassy systems [51, 52]. As a result, strongly interacting (with phonons) TLSs are scarce at low energies, thus rarely observable in conventional measurements performed near equilibrium. However, in our measurement TLSs with maximum energy of $\hbar \omega_0 + pE_{b,\text{max}}$, where their densities are much larger, can be swept into resonance. Out of equilibrium, the interaction between the two types of TLSs acts to reform the equilibrium gap in the DOS of strongly interacting TLSs by rearrangement of the standard TLSs. This sets a typical time scale for the reformulation of the gap, equal to a typical relaxation time of standard TLSs. One observes the excess loss when the bias field modulation frequency $f_b$ exceeds the standard TLS relaxation rate, such that the reconstruction of the gap is incomplete. The non-equilibrium DOS $\rho_2(E, E_b)$ depends on energy due to the energy dependence of the equilibrium DOS $\rho_{2,\text{eq}}(E)$, and also depends on $E_b$ due to the time-dependent reformation of the gap. By extending the Efros-Shklovskii argument...
to our non-equilibrium situation we obtain an approximated expression for $\rho_2(E, \dot{E}_b)$ (see Methods for details). The resulting non-saturated loss tangent $\tan \delta_{2,0}(\dot{E}_b) = \pi p_2^2 \rho_2(\omega_0, \dot{E}_b)/(3\varepsilon)$, where $p_2$ is the dipole moment of the second type of TLSs, reads

$$\tan \delta_{2,0}(\dot{E}_b) = A \exp \left[ B \int_0^\infty dx \exp \left(-\frac{x}{\rho_2(\omega_0, \dot{E}_b)}\right)\right],$$

with the four fitting parameters $A$, $B$, $C$, and $D$. Here $A = \pi p_2^2 \rho_2(\omega_0)/(3\varepsilon)$ is the excess loss at equilibrium ($\dot{E}_b = 0$), and is proportional to the equilibrium DOS $\rho_2(\omega_0)$ at the resonance energy. $B = (8\pi/3)\rho_1^u \ln[1 + E_0/(\hbar\omega_0)]$, where $E_0 = \min\{E_{\text{max}}, \hbar\omega_0 + p_2 E_{b,\text{max}}\}$ with $E_{\text{max}} \sim 10 \text{kHz}$ being the maximum energy of standard TLSs, is proportional to the phonon-mediated interaction strength $u$ between the two types of TLSs. $C = \ln \left(\frac{E_{\text{max}}}{\Delta_{0,\min}}\right)$ is the upper limit of the integral over the normalized tunneling amplitudes of the standard TLSs $x = \ln(\Delta_{0,\min}/\Delta_{0,\min})$, where $\Delta_{0,\min}$ is a minimum cutoff for tunneling amplitudes [4, 5]. Finally, $D = \Gamma_{1,m}^2 E_{\text{max}}^2 \Delta_{0,\min}/E_{\text{max}}^2 E_{b,\text{max}}$, with $D\varepsilon^2 C = \Gamma_{1,m}^2 E_{\text{max}}^2 E_{b,\text{max}}$ being the characteristic bias rate above which standard TLSs cannot change their state in order to equilibrate the DOS of the second type of TLSs. Fitting the collapsed data of Fig. 2(b) (for $P_{ac} < -25\text{dBm}$) yields $A = 1.8 \times 10^{-8}$, $B = 1.27$, $C = 7.8$ and $D = 7.2 \times 10^{-3} \text{V/µm s}$.

At $P_{ac} \geq -15\text{dBm}$ where a saturation effect is observed, we notice that the normalized excess loss $\tan \delta_t/\tan \delta_{2,0}$, which neutralizes the effect of energy-dependent DOS of the second TLS type, resumes a similar scaling with $\dot{E}_b/n$ as the standard TLSs (Fig. 3). This provides a striking evidence for the TLS origin of the excess loss. We therefore repeat the numerical calculation based on LZ theory for the excess loss of Fig. 2b, similarly to the calculation performed for the standard TLSs in Fig. 2a. The colored solid curves shown in Fig. 2b are obtained for $p_2 = 110\text{D}$ and $\Gamma_{1,m} = 800\text{MHz}$ for the second type of TLSs. Combining the theoretical calculations of $\tan \delta_t$ and $\tan \delta_2$ from the standard and second TLS types, respectively, the total loss $\tan \delta = \tan \delta_t + \tan \delta_2$ agrees well with measured loss over the entire range of driving powers and bias rates explored in the experiment, as shown in Fig. 2c.

Comparison to the two-TLS model. From the calculation of $\tan \delta_2$ we can extract information that allows us to examine the predictions of the two-TLS model. First, the ratio $\Gamma_{1,m}^{(2)}/\Gamma_{1,m}^{(1)}$ between the relaxation rates of the two types of TLSs is equal to $(\gamma_2/\gamma_1)^2$, where $\gamma_1$ and $\gamma_2$ are the couplings of the two types of TLSs to phonons. Using the relaxation rates found above, we obtain $\gamma_2/\gamma_1 \approx 12$, which is slightly smaller than expected within the two TLS model[48, 59], but nevertheless describes distinct coupling strengths to phonons differing by an order of magnitude. Second, the ratio between the equilibrium DOS of the two types of TLSs at the resonance energy $\hbar\omega_0 \approx 0.25\text{kHz}$ can be found from $A/\tan \delta_t = p_2^2 \rho_2(\omega_0)/(p_2^2 \rho_1) \approx 10^{-4}$. Together with the dipole moments $p_1 = 11\text{D}$ and $p_2 = 110\text{D}$, we find $\rho_2(\omega_0)/\rho_1 \approx 10^{-6}$. This is consistent with the general success of the STM in describing the low-temperature universality in some acoustic and thermodynamic properties of amorphous solids, since phonon attenuation is dominated by the standard (weakly interacting) TLSs at energies where $\gamma_2^2 \rho_2(\omega_0) < \gamma_1^2 \rho_1$. Lastly, by estimating the largest energy of standard TLSs as $E_{\text{max}} \approx 10\text{kHz}$, from the parameter $B$ one obtains $\rho_1 u \approx 4 \times 10^{-2}$. According to the two-TLS model, the so-called tunneling strength is given by $C_0 = \rho_1 u \cdot (\gamma_1/\gamma_2)$, which for $\gamma_1/\gamma_2 \approx 1/12$ gives $C_0 \approx 3 \times 10^{-3}$. This agrees with the universally small value of $C_0 \approx 10^{-4} - 10^{-3}$ which is experimentally found to hold across a wide range of different amorphous solids [14].

Further comparison with the two-TLS model is achieved by studying the bias rate dependence of the loss at various bias amplitudes. This is shown in Fig. 4a for driving power of $-35\text{dBm}$. It reveals a clear bias amplitude dependence at high bias rates, indicating an energy-dependent DOS for the second TLS type. At the highest bias rates $\dot{E}_b \gg D\varepsilon C$, where $D\varepsilon^2 C = \Gamma_{1,m}^2 E_{\text{max}}^2 E_{b,\text{max}} \sim 5 \times 10^4 \text{V/µm s}$, the gap reconstruction in the non-equilibrium DOS $\rho_2(E, \dot{E}_b)$ is negligible. Thus $\rho_2(E, \dot{E}_b) \approx \Gamma_{1,m}^2 E_{\text{max}}^2 E_{b,\text{max}}$ ap-
FIG. 4. Bias amplitude dependence of the maximum loss. a $\tan \delta$ measured as a function of $\dot{E}_b$ for different bias field amplitudes $E_{b,\text{max}}$. b The measured maximum excess loss $\tan \delta_{\text{max}}$ as a function of $p_2E_{b,\text{max}}$, proportional to the equilibrium DOS of the second type of TLSs at this energy, $\rho_{2,\text{eq}}(p_2E_{b,\text{max}})$. The red line shows a (weak) logarithmic dependence of the measured $\tan \delta_{\text{max}}$, which indicates that we most likely measured in the regime where $p_2E_{b,\text{max}}$ is in the vicinity of or outside the gap edge.

Discussion

Using a fast-swept bias field in addition to microwave fields in a resonator, we have studied the non-equilibrium loss in amorphous silicon. The data suggests the existence of two types of TLSs. At low bias rates the dielectric loss is determined by standard TLSs, which are weakly coupled to phonons. At high bias rates the dielectric loss deviates from the STM predictions, but agree well with the two-TLS model for TLSs strongly interacting with phonons. The analysis of the bias rate dependent loss indicates a gap in the DOS and the fitting yields a large dipole moment for the second TLS type. Technically, the fittings of the data for the entire domain of driving powers and bias rates contain eight parameters (see Fig. 2c). These include the dipole moments $p_1 \approx 11 \text{ D}$, $p_2 \approx 110 \text{ D}$ and relaxation rates $\Gamma^{(1)}_{1,\text{m}} \approx 5.7 \text{ MHz}$, $\Gamma^{(2)}_{1,\text{m}} \approx 800 \text{ MHz}$ for the two types of TLSs, and the four parameters $A$, $B$, $C$ and $D$ used to fit the unsaturated excess loss at low and intermediate powers. The accuracy of these parameters depends on the LZ analysis of the loss at low bias rates, which may lead to some ambiguities in the fitting. However, our main results do not depend on the preciseness of these parameters, since the distinct dipole moments and relaxation rates of the two kinds of TLSs result from the distinct driving power dependence of the two contributions to the loss, below and above the intrinsic loss tangent $\tan \delta_0$. Experimental data consistent with the existence of two types of TLSs, characterized by distinct couplings to phonons, was previously reported [54, 55]. Our results here go beyond previous experiments as data attests directly to the presence of two types of TLSs, the bimodality of their couplings to phonons, and their electric dipole interaction strengths.

Insights of the two types of TLSs gleaned from the experiment are of general interest to understand TLS properties in amorphous solids, and of increasing importance due to their impact in quantum information science. The fast bias technique has been previously used to study the non-equilibrium loss for a couple different materials: silicon nitride[41] and alumina[45]. However, we have not yet seen excess loss in these materials. The two-TLS model is silent with respect to the electric dipole moments of the TLSs, which can vary between materials. Usually, a dipole moment of a few Debye is reported for standard TLSs [23, 35–37], compared to 11 D for amorphous silicon. The dipole moment of 110 D extracted for the second type of TLSs is anomalously large and may be unique to amorphous silicon. The contribution from the second TLS type to the loss at high bias rates may thus not be measurable in other amorphous solids, where the dipole moments are considerably smaller. Interestingly, several works reported a large range of TLS densities in amorphous silicon [7–10] and a recent study suggests that mechanical and dielectric loss in amorphous silicon originates from two different types of TLSs [56]. To check the
generality of the second TLS type in other amorphous solids, one possibility is to repeat our non-equilibrium protocol, but measure both the dielectric and acoustic responses, as large acoustic response out of equilibrium is expected irrespective of the value of the electric dipole moment. Alternatively, one could also measure the thermal conductivity, which is proportional to square of the TLS-phonon coupling, and compare the results with and without a rapidly varying bias field. Such studies would allow detailed characterization of TLSs at low and high energies, and could constitute a significant step towards the long-sought understanding of the origin of TLSs in glasses.

Methods

Experimental setup

The resonators are fabricated with Al/a-Si/Al trilayer films on highly resistive silicon substrates. The bottom aluminum layer (100 nm) and the top aluminum layer (250 nm) are sputtered via DC sputtering. The low-stress amorphous silicon layer (250 nm) is grown by PEVCD at 100°C. The resonator (Fig. 1a) is defined by photolithography and subsequent etching. It consists of four equal bridge parallel-plate capacitors (Figs. 1a and 1b), which is modified from that in ref.31. Assuming the dielectric constant for the amorphous silicon is 11.5, the capacitor has a total capacitance \( C = 1.2 \text{ pF} \) \((C1 = C4 = C)\). The vias are etched by SFE to connect the capacitor to the meander inductor. The resonator is inductively coupled to the coplanar transmission line for standard transmission measurements of \( S21 \). The measurements were performed at the base temperature of the dilution refrigerator \( T_{\text{base}} \approx 20 \text{ mK} \). A dc bias field, \( E_b = V_b/2d \), where \( d = 250 \text{ nm} \) is the thickness of the amorphous silicon dielectric, is applied across the capacitors. The bridge layout is effective to isolate the microwave resonance energy from the bias field input port. A triangular bias voltage is applied. The time dependence of the loss for a resonator when a periodic bias field is applied is studied in Ref.[41]. The maximum bias is \( E_{b,\text{max}} = 0.44 \text{ V/} \mu \text{m} \), and the fastest bias frequency is \( f_b = 4.5 \text{ MHz} \). The resonance frequency \( f_0 = 5.1 \text{ GHz} \). \( S21 \)'s are measured as a function of photon number, \( n \) and bias rate, \( E_b \). The corresponding loss \( \delta \), equal to the inverse resonator quality \( 1/Q_0 \), is found from each \( S21 \) measurement. See Supplementary Note 1 for more wiring and attenuation details in the measurement setup.

Loss due to LZ transitions within the STM

The Hamiltonian of a single TLS driven by the resonator electric field \( E_{\text{res}}(t) = E_{\text{ac}} \cos(\omega_0 t) \) and by a time-dependent bias electric field \( E_b(t) \) is

\[
H = \frac{1}{2} E(t) \sigma_z - \vec{p} \cdot \vec{E}_{\text{ac}} \cos(\omega_0 t) \left( \Delta(t) E(t) \sigma_z - \Delta_0 E(t) \sigma_x \right),
\]

where \( E(t) = \sqrt{\Delta^2(t) + \Delta_0^2} \) is the TLS energy splitting, assumed to be slowly varying on the time scale \( 2\pi/\omega_0 \). A TLS with \( \Delta_0 < \hbar \omega_0 \) can be swept through resonance at time \( t_0 \) for which \( E(t_0) = \hbar \omega_0 \). Near this resonance the energy splitting can be expanded as \( E(t) \approx \hbar \omega_0 + \nu \omega(t - t_0) \) [41, 42], where \( \nu = E(t_0)/\hbar = v_0 \sqrt{1 - (\Delta_0/\hbar \omega_0)^2} \cos \theta \) with \( \theta \) the angle between \( \vec{p} \) and \( \vec{E}_b \), and \( v_0 = (2p/\hbar) E_b(t_0) \) the maximum bias rate.

The TLS absorption in the absence of the bias field can be separated into the so-called relaxation absorption, arising from the longitudinal term \( (\propto \sigma_z) \) in the brackets of Eq. (2), and the resonant absorption resulting from the transverse coupling \( (\propto \sigma_x) \) [4, 5, 57, 58]. In the regime \( \omega_0 \gg k_B T/\hbar \gg \Gamma_1, \Gamma_2 \) considered in this paper (\( \Gamma_1 \) and \( \Gamma_2 \) are the TLS relaxation and decoherence rates, respectively), the resonant mechanism dominates and the longitudinal term in Eq. (2) can be neglected. The TLS dipole moment induced by the resonator field can therefore be written as \( \langle p(t) \rangle = -\nu \cos \theta \langle \sigma_x(t) \rangle = \Re \left[ \langle \sigma_x(t) \rangle \right] \), where \( \chi' = \chi' + i \chi'' \) is the TLS electric susceptibility. The imaginary part of the susceptibility, \( \chi'' \), yields the imaginary part \( \epsilon'' \) of the dielectric constant due to TLSs upon averaging over the ensemble of TLSs \( \chi' \) gives the shift in the real part of the dielectric constant due to TLSs, which is small compared to the material dielectric constant \( \epsilon \). The loss tangent \( \tan \delta = \epsilon''/\epsilon \) is thus proportional to the out of phase \( (\propto \sin(\omega_0 t)) \) component of \( \langle \sigma_x(t) \rangle \). To calculate this expression, we transform to the frame of reference rotating around the z axis with frequency \( \omega_0 \), using the unitary transformation \( U_R = e^{i \omega_0 t \sigma_z/2} \). Since \( U_R \sigma_x U_R^\dagger = \cos(\omega_0 t) \sigma_x - \sin(\omega_0 t) \sigma_y \), in the rotating frame of reference the relation \( \chi'' = \Re \left[ \langle \sigma_x(t) \rangle \right] \) holds [58], where \( \Omega_R = \Omega_{\text{R0}} \cos(\Delta_0/\hbar \omega_0) \) is the TLS Rabi frequency, with its maximum value \( \Omega_{\text{R0}} = p E_{\text{ac}}/\hbar \). Moreover, application of the rotating wave approximation yields the LZ Hamiltonian

\[
H_{\text{LZ}} = \frac{\hbar}{2} \left[ v(t - t_0) \sigma_z + \Omega_R \sigma_x \right],
\]

governing the dynamics of a TLS resonant passage [42]. The probability for photon absorption in a transition is the famous LZ probability for an adiabatic transition from the initial state \( |g, n\rangle \) to the final state \( |e, n-1\rangle \) [Fig. 1(d)]. \( P_{\text{ad}} = 1 - e^{-\eta_g^2/2v^2} \). Finite TLS relaxation and decoherence rates \( \Gamma_1 \) and \( \Gamma_2 \) can be taken into account by means of the Bloch equations within the rotating frame of reference [58]. The Hamiltonian (3) can be written as the Hamiltonian of a spin-1/2 particle in a
magnetic field, \( H_{LZ} = -S \cdot \vec{B} \), where \( \vec{S} = \hbar \vec{\sigma}/2 \) and \( \vec{B} = -(\Omega R, 0, v(t-t_0)/\hbar) \). The corresponding Bloch equations read \( d(\vec{S})/dt = (\vec{S} \times \vec{B}) - (\Gamma_2(S_z), \Gamma_2(S_y), \Gamma_1((S_z - S_{z0})) \), where \( S_{z0} = (\hbar/2) \tanh(\hbar \omega_0/2k_B T) \). In our calculations we neglect pure dephasing of the resonant TLSs, such that \( \Gamma_2 = \Gamma_1/2 \). Assuming TLS relaxation into the phonon bath, one has \( \Gamma_1(\Delta_0) = \Gamma_{1,m}(\Delta_0/h\omega_0)^2 \), where \( \Gamma_{1,m} \) is the maximum relaxation rate for resonant TLSs with energy splitting \( E = \hbar \omega_0 \). If the bias duration is longer than the TLS relaxation time \( T_1 = 1/\Gamma_1 \), the steady state solution depends on time via the detuning from resonance \( \delta E = v(t-t_0) \), and \( \varepsilon'' \) can be obtained by averaging \( \chi'' \) over the ensemble of TLSs with the distribution function \( P(E, \Delta_0) = \rho E/(\Delta_0 \sqrt{E^2 - \Delta_0}) \), where \( \rho \) is the TLS DOS, assumed to be energy-independent within the STM. The integration over energies can then be replaced by integration over time, leading to the expression

\[
\tan \delta \tan \delta_0 = \frac{3}{2} \int_0^1 dyy^2 \int_0^1 \frac{x dx}{\sqrt{1-x^2}} \frac{2v}{\pi \Omega_R} \int_0^\infty \langle \sigma_y(t) \rangle dt,
\]

where \( \tan \delta_0 = \pi \rho p^2/(3c) \) is the intrinsic loss tangent and the integration variables are \( x = \Delta_0/(h\omega_0) \) and \( y = \cos \theta \) (in terms of these variables \( v = v_0 \sqrt{1-x^2} \) and \( \Omega_R = \Omega_{R0}xy \)). This loss is a function of the parameters \( \xi = 2v_0/(\pi \Omega_{R0}^2) \) and \( \Gamma_{1,m}/\Omega_{R0} \), and can be calculated by a numerical integration of the Bloch equations.

In the regime \( \Gamma_{1,m}/\Omega_{R0} \ll 1 \), dissipation can be neglected for \( \xi \gg \Gamma_{1,m}/\Omega_{R0} \), and one obtains the expression for the dielectric loss tangent [42]

\[
\tan \delta \tan \delta_0 = \frac{3}{2} \int_0^1 dyy^2 \int_0^1 \frac{x dx}{\sqrt{1-x^2}} \frac{2v}{\pi \Omega_R^2} \left[ 1 - e^{-\pi \Omega_R^2/(2v)} \right].
\]

By performing numerical integration over \( x \) and \( y \), one obtains the normalized loss \( \tan \delta/\tan \delta_0 \) as a function of the dimensionless bias rate \( \xi \), approaching unity at \( \xi \gg 1 \), as shown in Fig. 1(d).

**Loss due to the second type of TLSs**

Let us derive the expression for the non-equilibrium DOS of the second type of TLSs, \( \rho_2(E, \hat{E}_b) \), under the assumption that their energy-dependent equilibrium DOS results from interactions between the two types of TLSs, as motivated by the Efros-Shklovskii coulomb gap [51, 52] and discussed in Refs. [48, 50]. In equilibrium, the DOS is reduced by interaction with standard TLSs that break the Efros-Shklovskii stability criterion \( E + E' - 2u/R^3 > 0 \), where \( E' \) is the energy of the standard TLS, \( u/R^3 \) is the mutual TLS-TLS interaction and \( R \) is the distance between the TLSs. We note that interactions exist also among the second type of TLSs, but since they are scarce at low energies, the dominating interactions are with the standard TLSs. As discussed in Refs. [48, 50], at higher energies interactions among the second type of TLSs lead to weaker logarithmic energy dependence of the DOS [see Fig. 4]; here we neglect these interactions and consider the gap in the DOS at low energies due to interactions between the two types of TLSs. In the non-equilibrium situation of our experiment, only those standard TLSs that are capable of changing their state during the sweep time \( 1/(2f_b) \) will contribute to the reconstruction of the gap in the DOS. For \( f_b \ll \Gamma_1 \), where \( \Gamma_1 \) is a typical relaxation rate of standard TLSs, the DOS \( \rho_2(E, f_b \ll \Gamma_1) \) should approach the DOS at equilibrium, \( \rho_{2,eq}(E) \), discussed in Ref. [48]. On the other hand, in the limit of instantaneous sweep, \( f_b \gg \Gamma_1 \), the non-equilibrium DOS \( \rho_2(E, f_b) \) should satisfy the condition \( \rho_2(E, f_b \gg \Gamma_1) \approx \rho_{2,eq}(E + p_2 E_{b,\text{max}}) \). Taking into account the fraction \( 1 - e^{-\Gamma_1/(2f_b)} \) of standard TLSs that can flip during time \( 1/(2f_b) \), where \( \Gamma_1 = \Gamma_1(E', \Delta_0) \) is the relaxation time of standard TLSs with energy \( E' \) and tunneling energy \( \Delta_0 \), we obtain the expression

\[
\rho_2(E, f_b) \approx \prod_{d^3 R} \left[ 1 - d^3 R \int_0^{E_0} dE' \int_0^{E_0} d\Delta_0 \frac{d\Delta_0}{\Delta_0} \Theta \left( \frac{2u}{R^3} - E - E' \right) \left( 1 - e^{-\frac{\Gamma_1}{2f_b}} \right) \right] \rho_{2,eq}(E + p_2 E_{b,\text{max}})
\]

\[
= \exp \left[ -\rho_1 \int_0^{E_0} dE' \int_0^{E_0} d\Delta_0 \frac{d\Delta_0}{\Delta_0} \int d^3 R \Theta \left( \frac{2u}{R^3} - E - E' \right) e^{-\frac{\Gamma_1}{2f_b}} \right] \rho_{2,eq}(E),
\]

where \( \Theta(x) \) is the step function and \( E_0 = \min\{E_{\text{max}}, \hbar \omega_0 + p_2 E_{b,\text{max}}\} \) with \( E_{\text{max}} \approx 10 \)K being the maximum energy of standard TLSs. This expression fulfills both limiting conditions discussed above. The exponent in the last line of Eq. (6) gives the enhancement of the equilibrium DOS \( \rho_{2,eq}(E) \) due
to standard TLSs with relaxation rates $\Gamma_1 \lesssim f_b$. Using $\dot{E}_b = 2f_bE_{b,\text{max}}$ and assuming relaxation of standard TLSs to be dominated by interaction with phonons, corresponding to the rate $\Gamma_1(E', \Delta_0) = \Gamma_{1,m}(E') / \Delta_0$ [4], we obtain

$$\rho_2(E, \dot{E}_b) = \rho_{2,\text{eq}}(E) \exp \left[ \frac{8\pi}{3} \rho_1 u \int_0^{E_0} \frac{dE'}{E + E'} \int_0^{\ln(E'/\Delta_{0,\text{min}})} dx e^{-\Gamma_{1,m}(E')(\Delta_{0,\text{min}}/E')^2 (E_{b,\text{max}}/E_b)x^2} \right],$$  

(7)

the last expression by replacing $E'$ by $E_{\text{max}}$ in the last integral and in the upper limit. This results in the expression

$$\rho_2(E, \dot{E}_b) \approx \rho_{2,\text{eq}}(E) \exp \left[ \frac{8\pi}{3} \rho_1 u \ln \left( 1 + \frac{E_0}{E} \right) \int_0^{\ln(E_{\text{max}})} dx e^{-\Gamma_{1,m}(E_{\text{max}})(\Delta_{0,\text{min}}/E_{\text{max}})^2 (E_{b,\text{max}}/E_b)x^2} \right],$$  

(8)

At low power driving, where the second type of TLSs is non-saturated, the excess loss is $\tan \delta_{2,0}(\dot{E}_b) = \pi p^2 \rho_2(\hbar \omega_0, \dot{E}_b)/(3e)$ and thus takes the form of Eq. (1). This loss serves as the intrinsic loss for the second TLS type, and is used for scaling the excess loss in Fig. (3). The LZ analysis described above is then carried out for the normalized excess loss $\tan \delta_2 / \tan \delta_{2,0}$ at all driving powers.

### DATA AVAILABILITY

The data in the main text and Supplementary Materials are available from the corresponding authors upon request.

---

* Correspondence and requests for materials should be addressed to L.Y. (email: liqiu.yu.physics@gmail.com) or K.D.O (email: osborn@lps.umd.edu).

[1] Zeller, C. R. & Pohl, R. O. Thermal Conductivity and Specific Heat of Noncrystalline Solids. *Phys. Rev. B* 4, 2029 (1971).

[2] Phillips, W. A. Tunneling states in amorphous solids. *J. Low Temp. Phys.* 7, 351 (1972).

[3] Anderson, P. W, Halperin, B. I. & Varma, C. M. Anomalous low-temperature thermal properties of glasses and spin glasses. *Philos. Mag. J. Theor. Exp. Appl. Phys.* 25, 1 (1972).

[4] Phillips, A. W. Two-level states in glasses. *Rep. Prog. Phys.* 50, 1657 (1987).

[5] Hunklinger, S. & Raychaudhuri, A. K. Chapter 3: Thermal and Elastic Anomalies in Glasses at Low Temperatures. *Progr. Low Temp. Phys.* 9, 265 (1986).

[6] Yu, C. C. & Leggett, A. J. Low temperature properties of amorphous materials: Through a glass darkly. *Comments Condens. Matter Phys.* 14, 231 (1988).

[7] Liu, X. et al. Amorphous Solid without Low Energy Excitations. *Phys. Rev. Lett.* 78, 4418 (1997).

[8] Queen, D. R. et al. Two-level systems in evaporated amorphous silicon. *J. Non-Cryst. Solids* 426, 19 (2015).

[9] Molina-Ruiz, M. et al. Two-level systems and growth-induced metastability in hydrogenated amorphous silicon. *Mater. Res. Express* 7, 095201 (2020).

[10] Liu, X. et al. Comparing amorphous silicon prepared by electron-beam evaporation and sputtering toward eliminating atomic tunneling states. *J. Alloys Compd.* 855, 157431 (2021).

[11] Southworth, D. R. et al. Stress and Silicon Nitride: A Crack in the Universal Dissipation of Glasses. *Phys. Rev. Lett.* 102, 225503 (2009).

[12] Bartkowiak, M. et al. Nuclear Quadrupole Moments as a Microscopic Probe to Study the Motion of Atomic Tunneling Systems in Amorphous Solids. *Phys. Rev. Lett.* 110, 205502 (2013).

[13] Yu, C. C. & Leggett, A. J. Low temperature properties of amorphous materials: Through a glass darkly. *Comments Condens. Matter Phys.* 14, 231 (1988).

[14] Pohl, R. O., Liu, X. & Thompson, E. J. Low-temperature thermal conductivity and acoustic attenuation in amorphous solids. *Rev. Mod. Phys.* 74, 991 (2002).

[15] Müller, C., Cole, J. H. & Lisenfeld, J. Towards understanding two-level-systems in amorphous solids: insights from quantum circuits. *Rep. Prog. Phys.* 82, 124501 (2019).

[16] Oliver, W. D. & Welander P. B. Materials in superconducting quantum bits. *MRS Bull.* 38, 816 (2013).
[17] Martinis, J. M., Cooper, K. B., McDermott, R., Steffen, M., Ansmann, M., Osborn, K. D., Cicak, K., Oh, S., Pappas, D. P., Simmonds, R. W., & Yu, C. C. Decoherence in Josephson Qubits from Dielectric Loss. Phys. Rev. Lett. 95, 210503 (2005).

[18] Shalibo, Y. et al. Lifetime and Coherence of Two-Level Defects in a Josephson Junction. Phys. Rev. Lett. 105, 177001 (2010).

[19] Zaretskii, V., Suri, B., Novikov, S., Wellstood, F. C. & Palmer, B. S. Spectroscopy of a Cooper-pair box coupled to a two-level system via charge and critical current. Phys. Rev. B 87, 174522 (2013).

[20] Christensen, B. G. et al. Anomalous charge noise in superconducting qubits. Phys. Rev. B 100, 140503 (2019).

[21] Pappas, D. P., Vissers, M. R., Wisbey, D. S., Klime, J. S. & Gao, J. Two Level System Loss in Superconducting Microwave Resonators. IEEE Trans. Appl. Supercond. 21, 871 (2011).

[22] Paik, H. & Osborn, K. D. Reducing quantum-regime dielectric loss of silicon nitride for superconducting quantum circuits. Appl. Phys. Lett. 96, 072505 (2010).

[23] Müller, C., Lisenfeld, J., Shirman, A. & Poletto, S., Phys. Rev. B 92, 035442 (2015).

[24] Klimov, P. V. et al. Fluctuations of Energy-Relaxation Times in Superconducting Qubits. Phys. Rev. Lett. 121, 090502 (2018).

[25] Schlör, S. et al. Correlating Decoherence in Transmon Qubits: Low Frequency Noise by Single Fluoridators. Phys. Rev. Lett. 123, 190502 (2019).

[26] Burnett, J. et al. Decoherence benchmarking of superconducting qubits. npj Quantum Inf. 5, 54 (2019).

[27] de Graaf, S. E. et al. Suppression of low-frequency charge noise in superconducting resonators by surface spin desorption. Nat. Commun. 9, 1143 (2018).

[28] Gao, J. et al. Experimental evidence for a surface distribution of two-level systems in superconducting lithographed microwave resonators. Appl. Phys. Lett. 92, 152505 (2008).

[29] Burnett, J. et al. Evidence for interacting two-level systems from the 1/f noise of a superconducting resonator. Nat. Commun. 5, 4119 (2014).

[30] Faoro, L. & Ioffe, L. B. Interacting tunneling model for two-level systems in amorphous materials and its predictions for their dephasing and noise in superconducting microresonators. Phys. Rev. B 91, 014201 (2015).

[31] Burin, A. L., Matityahu, S. & Schechter, M. Low-temperature 1/f noise in microwave dielectric constant of amorphous dielectrics in Josephson qubits. Phys. Rev. B 92, 174201 (2015).

[32] Connors, E. J., Nelson, J., Qiao, H., Edge, L. F. & Nichol, J. M. Low-frequency charge noise in Si/SiGe quantum dots. Phys. Rev. B 100, 165305 (2019).

[33] Grabovskij, G. J., Peichl, T., Lisenfeld, J., Weiss, G. & Ustinov, A. V. Strain Tuning of Individual Atomic Tunneling Systems Detected by a Superconducting Qubit. Science 338, 232 (2012).

[34] Brehm, J. D., Bilmes, A., Weiss, G., Ustinov, A. V. & Lisenfeld J. Transmission-line resonators for the study of individual two-level tunneling systems. Appl. Phys. Lett. 111, 112601 (2017).

[35] Sarabi, B., Ramanaayaka, A. N., Burin, A. L., Wellstood, F. C. & Osborn, K. D. Projected Dipole Moments of Individual Two-Level Defects Extracted Using Circuit Quantum Electrodynamics. Phys. Rev. Lett. 116, 167002 (2016).

[36] Lisenfeld, J. et al. Electric field spectroscopy of material defects in transmon qubits. npj Quantum Inf. 5, 1 (2019).

[37] Hung, C. C., Yu, L., Foroozani, N., Fritz, S., Gerthsen, D. & Osborn, K. Probing hundreds of individual quantum defects in polycrystalline and amorphous alumina. Preprint at arxiv.org/abs/2107.04131 (2021).

[38] Lisenfeld, J., Bilmes, A., Matityahu, S., Zanker, S., Marthaler, M., Schechter, M., Schön, G., Shirman, A., Weiss, G. & Ustinov, A. V. Decoherence spectroscopy with individual two-level tunneling defects. Sci. Rep. 6, 23786 (2016).

[39] Bilmes, A., Megrant, A., Klimov, P., Weiss, G., Martinis, J. M., Ustinov, A. V. & Lisenfeld J. Resolving the positions of defects in superconducting quantum bits. Sci. Rep. 10, 3090 (2020).

[40] Bilmes, A., Volosheninuk, S., Brehm, J. D., Ustinov, A. V. & Lisenfeld, J. Quantum sensors for microscopic tunneling systems, npj Quantum Inf. 7, 27 (2021).

[41] Khalil, M. S. et al. Landau–Zener population control and dipole measurement of a two-level-system bath. Phys. Rev. B 90, 100201 (2014).

[42] Burin, A. L., Khalil, M. S. & Osborn, K. D. Universal Dielectric Loss in Glass from Simultaneous Bias and Microwave Fields. Phys. Rev. Lett. 110, 157002 (2013).

[43] von Schickfus, M. & Hunklinger, S. Saturation of the dielectric absorption of vitreous silica at low temperatures. Phys. Lett. A 64, 144 (1977).

[44] Rosen, Y. J., Khalil, M. S., Burin, A. L. & Osborn, K. D. Random-Defect Laser: Manipulating Lossy Two-Level Systems to Produce a Circuit with Coherent Gain. Phys. Rev. Lett. 116, 163601 (2016).

[45] Matityahu, S. et al. Dynamical decoupling of quantum two-level systems by coherent multiple Landau-Zener transitions. npj Quantum Inf. 5, 114 (2019).

[46] Natelson, D., Rosenberg, D. & Osheroff, D. D. Evidence for Growth of Collective Excitations in Glasses at Low Temperatures. Phys. Rev. Lett. 80, 4689 (1998).

[47] Burin, A. L. Dipole Gap Effects in Low Energy Excitation Spectrum of Amorphous Solids. Theory for Dielectric Relaxation J. Low Temp. Phys. 100, 300 (1995).

[48] Schechter, M. & Stamp, P. C. E. Inversion symmetric two-level systems and the low-temperature universality in disordered solids. Phys. Rev. B 88, 174202 (2013).

[49] Chirkin, A., Barash, D. & Schechter, M. Nonhomogeneity of the density of states of tunneling two-level systems at low energies. Phys. Rev. B 89, 104202 (2014).

[50] Chirkin, A., Gabdulkin, I., Burin, A. & Schechter, M. The strain gap in a system of weakly and strongly interacting two-level systems. Preprint at http://arxiv.org/abs/1307.0868 (2014).

[51] A. L. Efros, A. L. & and Shklovskii, B. I. Coulomb gap and low temperature conductivity of disordered systems. J. Phys. C 8, L49 (1975).

[52] Baranovskii, S. D., Efros, A. L., Gelmont, B. L. & Shklovskii B. I. Coulomb gap in disordered systems: computer simulation. J. Phys. C 12, 1023 (1979).

[53] Gaita-Ariño, A. & Schechter, M. Identification of Strong and Weak Interacting Two-Level Systems in KBr:CN. Phys. Rev. Lett. 107, 105504 (2011).

[54] Matityahu, S., Shirman, A., Schön, G. & Schechter, M. Decoherence of a quantum two-level system by spectral diffusion. Phys. Rev. B 93, 134208 (2016).
[55] Kirsh, N., Svetitsky, E., Burin, A. L., Schechter, M. & Katz, N. Revealing the nonlinear response of a tunneling two-level system ensemble using coupled modes. *Phys. Rev. Materials* 1, 012601(R) (2017).

[56] M. Molina-Ruiz, M., Rosen, Y. J., Jacks, H. C., Abernathy, M. R., Metcalf, T. H., Liu, X., DuBois, J. L. & Hellman, F. Origin of mechanical and dielectric losses from two-level systems in amorphous silicon. *Phys. Rev. Materials* 5, 035601 (2021).

[57] Jäckle, J., Piché, L., Arnold, W. & Hunklinger, S. Elastic effects of structural relaxation in glasses at low temperatures. *J. Non-Cryst. Solids* 20, 365 (1976).

[58] Carruzzo, H. M., Grannan, E. R. & Yu, C. C. Nonequilibrium dielectric behavior in glasses at low temperatures: Evidence for interacting defects. *Phys. Rev. B* 50, 6685 (1994).

ACKNOWLEDGMENTS

L.Y. would like to thank F. C. Wellstood and S. Dutta for their discussions and support. S.M. acknowledges support by the A. von Humboldt foundation. Y.J.R. acknowledges Lawrence Livermore National Laboratory operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA2. A.L.B. and A.M. acknowledge the support by Carrol Lavin Bernick Foundation Research Grant (2020-2021), NSF CHE-1900568 grant and LINK Program of the NSF and Louisiana Board of Regents. M.S. acknowledges support from the Israel Science Foundation (Grant No. 2300/19).

AUTHOR CONTRIBUTIONS

L.Y., Y.J.R., C.H. and K.D.O. designed and performed the experiments, L.Y., K.D.O. and S.M. analysed the data and wrote the paper. L.Y. and Y.J.R. processed the samples. S.M., M.S. A. M. and A.L.B. developed and carried out the theoretical work and numerical modeling. All authors discussed the results and commented on the manuscript.

COMPETING INTERESTS

The authors declare no competing interests.
Evidence for weakly and strongly interacting two-level systems in amorphous silicon, Yu et al.
SUPPLEMENTARY MATERIAL
Evidence for weakly and strongly interacting two-level systems in amorphous silicon

Liuqi Yu,1,2 Shlomi Matityahu,3,4 Yaniv J. Rosen,5 Chih-Chiao Hung,1,2 Andrii Maksymov,6 Alexander L. Burin,6 Moshe Schechter,3 and Kevin D. Osborn1,7

1Laboratory for Physical Sciences, University of Maryland, College Park, MD 20740, USA
2Department of Physics, University of Maryland, College Park, MD 20742, USA
3Department of Physics, Ben-Gurion University of the Negev, Beer Sheva 84105, Israel
4Institut für Theorie der Kondensierten Materie, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany
5Lawrence Livermore National Laboratory, Livermore, California 94550 USA
6Department of Chemistry, Tulane University, New Orleans, LA 70118, USA
7Joint Quantum Institute, University of Maryland, College Park, MD 20742, USA

CONTENTS

I. Additional details of the experimental setup 2
II. Input microwave power calibration 3
III. Extraction of loss from $S_{21}$ 4

References 5
I. ADDITIONAL DETAILS OF THE EXPERIMENTAL SETUP

The measurements are performed in a dilution refrigerator at a temperature of less than 20 mK. Supplementary Figure 1 shows the schematic of the detailed wirings of the measurement setup. A vector network analyzer (VNA) is used to carry out the $S_{21}$ measurements. The input line is heavily filtered with multiple attenuators at different stages. An additional 12 GHz K&L low-pass filter is added. The output signal is routed through two low-noise circulators (4 – 12 GHz) which are thermally anchored at mixing chamber plate. The output signal is amplified by a high electron mobility transistor (HEMT) at 4 K. For the bias line, a $-10$ dB attenuator is added at room temperature. A 12 GHz K&L low-pass filter and a RC filter are placed at mixing chamber stage, which gives a cut-off frequency of 10 MHz. A time-varying bias voltage as a triangular waveform is applied. The maximum bias is $E_b = 0.44$ V/µm, and the fastest applied bias frequency is $f_b = 4.5$ MHz. In previous related measurements, the time-dependent loss, particularly when the bias rate changes sign, was studied\(^1\). It is known that the change to the measured average loss is negligible. The base temperature of the dilution refrigerator remains below 20 mK throughout all measurements, such that no significant heating from bias leakage current is observed.

1. Schematic of the experimental wiring for measuring the resonator.
II. INPUT MICROWAVE POWER CALIBRATION

To determine the input power on chip due to attenuation in the input line, throughput $S_{21}$'s (bypassing the mounted device) at various source powers have been measured at room temperature. This is particularly important to accurately determine the average photon number in the resonator. The attenuation depends on the signal frequency and the source power from the VNA. Four relatively high source power (10 dBm, 0 dBm, $-10$ dBm and $-20$ dBm) are used to obtain sufficient signal to noise ratio. As shown in Supplementary Fig. 2a, linear fits are conducted on all four semilog $S_{21}$ plots. The frequency dependence is obtained by averaging the four slopes at different applied source powers. As shown in Supplementary Fig. 2b, the source power dependence of the input power at zero frequency is determined by plotting the intercepts as a function of the source powers. The obtained slope is one, which suggests any nonlinearities in the system associated with the power is negligible. Therefore, the eventual input power at mixing chamber can be expressed as

\begin{equation}
  P = -1.87 \times 10^{-9}f + P_{ac} - 70.2
\end{equation}

This expression is used to calibrate the photon number on the measured resonator throughout the main manuscript.
III. EXTRACTION OF LOSS FROM $S_{21}$

3. a An example of magnitude of $|S_{21}|$ vs frequency measured at $P_{ac} = -29$ dBm. b Raw $S_{21}$ data (blue dots) are plotted in IQ plane in the lower left. The red curve is a fitting of Supplementary Eq. 2. The black line represents the scaling of amplitude and the rotation from $(1, 0)$. The yellow line represents a $\phi$ rotation from a small impedance mismatch. We plot the data with adjusted amplitude and electric delays on the right.

The quality factor extraction is following the fitting function

$$S_{21} = Amp \times e^{i\theta} \times \frac{1 - Q|\hat{Q}_{e}^{-1}|e^{i\phi}}{1 + 2iQ\frac{\omega - \omega_0}{\omega_0}}.$$  \hspace{1cm} (2)

The $Amp$ and $\theta$ are the adjusted parameters for the raw data due to the attenuations, amplifications, and electric delays in both input and output line, and $Q$ is the total quality factor. Because of the impedance mismatch at two ports, $\hat{Q}_{e}$ is the external quality factor and also a complex number, and $\phi$ is the angle of $\hat{Q}_{e}^{-1}$. Fig. 3a shows an exemplary $S_{21}$ data (magnitude vs frequency) measured at $P_{ac} = -29$ dBm. The corresponding IQ plot of the data are shown in Fig. 3b: The lower left curve (blue dots) and the right curve (blue dots) are data before and after the adjusted $Amp$ and $\theta$. From the fitting, we extract $Q_{e} = 1/Re\{\hat{Q}_{e}^{-1}\} \approx 7300$, $Q_{i} \approx 9000$, and $\phi = 5.7^\circ$ when the input photon number $n = 1422$. 
Supplementary References

1 Khalil, M. S. et al. Landau-Zener population control and dipole measurement of a two-level-system bath. *Phys. Rev. B* **90**, 100201 (2014).

2 Khalil, M. S., Stoutimore, M. J. A., Wellstood, F. C. & Osborn, K. D. An analysis method for asymmetric resonator transmission applied to superconducting devices. *J. Appl. Phys.* **111**, 054510 (2012).