Quantum sensors for microscopic tunneling systems
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The anomalous low-temperature properties of glasses arise from intrinsic excitable entities, so-called tunneling Two-Level-Systems (TLS), whose microscopic nature has been baffling solid-state physicists for decades. TLS have become particularly important for micro-fabricated quantum devices such as superconducting qubits, where they are a major source of decoherence. Here, we present a method to characterize individual TLS in virtually arbitrary materials deposited as thin films. The material is used as the dielectric in a capacitor that shunts the Josephson junction of a superconducting qubit. In such a hybrid quantum system the qubit serves as an interface to detect and control individual TLS. We demonstrate spectroscopic measurements of TLS resonances, evaluate their coupling to applied strain and DC-electric fields, and find evidence of strong interaction between coherent TLS in the sample material. Our approach opens avenues for quantum material spectroscopy to investigate the structure of tunneling defects and to develop low-loss dielectrics that are urgently required for the advancement of superconducting quantum computers.

INTRODUCTION
We are still lacking an explanation for the behaviour of amorphous materials at low temperatures <10 K. Why is it that even widely different materials ranging from biatomic glasses to polymers show quantitatively identical properties such as specific heat and thermal conductivity? The Standard Tunneling Model (STM) has been a first attempt to explain these universal anomalies on the basis of two-level systems (TLS) believed to arise from the tunneling of atoms between two energetically similar con...
RESULTS

Sample capacitor design

Single TLS can be detected if their energy exchange rate with the qubit (which equals their coupling strength $g$) is comparable to the energy decay rate $1/T_1$ of the isolated qubit. The criterion $g \approx 1/T_1$ together with the TLS’ above-mentioned coupling strength $hg = |pF|$ define a suitable thickness $d$ of the dielectric layer in overlap capacitors: $d = \frac{p}{T_1 V_{rms}/h}$, where the electric field $|F|$ is substituted by $V_{rms}/d$. Here, $V_{rms} = \sqrt{\hbar \omega_0/2C_{tot}} \approx 4.5 \, \mu V$ is the vacuum voltage fluctuation on the qubit island at the designed plasma oscillation frequency $\omega_0 = 2\pi \cdot 6.2 \, \text{GHz}$ when $C_{tot} \approx 100 \, \text{fF}$ is the sum of all capacitances shunting the qubit. To be able to detect a TLS dipole moment $p_d$ of minimum $0.1 \, e \, \text{Å}^2$, and assuming a rather conservative $T_1 = 1 \, \mu s$, we arrive at a dielectric layer thickness $d \approx 70 \, \text{nm}$. We chose $d = 50 \, \text{nm}$ and a capacitor size of $(0.25 \times 0.3) \, \text{μm}^2$, resulting in $C_s \approx 0.15 \, \text{fF} \ll C_{tot}$ which ensures that the energy that is stored in the lossy sample capacitor remains limited to a small fraction of the qubit’s total energy, and coherence is preserved. A picture of the employed sample capacitor is shown in Fig. 1e while further details on the capacitor design are given in Supplementary Methods 1.

It is furthermore necessary to be able to distinguish TLS in the sample material from those on electrode interfaces and from TLS in Josephson junctions. This is accomplished by probing the TLS’ response to a local electric field generated by voltage-biassing the sample capacitor’s electrode as indicated in Fig. 1a, where the additional capacitor $C_f = 250 \, \text{fF}$ serves as a DC-break. The bias voltage will not induce an electric field in the transmon’s shunt capacitor $C_g$ nor inside the Josephson junctions’ tunnel barrier since the DC-electric potential difference of the transmon island and ground will be compensated by Cooper-pair tunneling, so that only TLS in the sample capacitor responds to the applied voltage $V_g$. In addition, we can tune $V_g$ residing at the perimeter of the qubit capacitor by a globally applied DC-electric field that is generated by an electrode installed above the qubit chip as shown in Fig. 1b. Moreover, all TLS including those residing inside the tunnel barriers of junctions can be tuned via physical strain by bending the chip with a piezo actuator, which is useful to enhance the number of observable TLS. The table in Fig. 2b summarizes how to identify the location of a TLS from its tunability characteristics.

We chose amorphous aluminum oxide AlO$_x$ as the sample material for this work since it is well characterized and of general importance for superconducting quantum circuits where it is ubiquitously used as a reliable tunnel barrier material. The sample capacitor is patterned with electron-beam lithography, where the bottom electrode is deposited and connected to the qubit island in the same step as the qubit’s Josephson junctions, followed by a third lithography step depositing $50 \, \text{nm}$ of AlO$_x$ by eBeam-evaporation of Al in an oxygen atmosphere, and capping it by a top Al electrode. The filter capacitor $C_f$ is formed simultaneously as a wider section in the top electrode. Here, we report results for samples employing small sample capacitors of size $(0.25 \times 0.3) \, \text{μm}^2$ which did not contribute significantly to decoherence. Two tested $C_f$-shunted qubits reached $T_1$-times of $3.3 - 4.2 \, \mu s$, which is comparable with an isolated reference qubit ($T_1 \approx 4.3 \mu s$) on the same chip. In another batch, we also tested larger sample...
capacitors (0.3 × 2.1) μm², which did limit the qubit’s T₁ time. This allowed us to measure the loss tangent of the employed AlOₓ dielectric as tan δ₀ ≈ (1.7 ± 0.2) · 10⁻³, comparable with other reports.²⁴–⁴⁷

TLS spectroscopy
To distinguish whether a TLS is located in a tunnel barrier, at the qubit’s film edges, or in the sample capacitor dielectric, we track its resonance frequency for a range of voltages applied to the global DC electrode (Vg), to the sample dielectric (Vₛ), and to the piezo (Vp). An example of such a measurement is presented in Fig. 2a, showing the frequency dependence of the qubit’s T₁ time estimated by swap spectroscopy,³³,³⁵,⁴³ where dark traces reveal enhanced qubit energy relaxation due to resonant TLS. These segmented hyperbolic traces are fitted to obtain the TLS’ coupling constants γ which determine their bias-dependent asymmetry energy ε = ε₁ + γ₁Vₛ + γ₂Vp up to an intrinsic offset ε₁. The fit also results in the value of the TLS’ tunneling energy Δ₀ if it lies within the tunability range of the qubit’s resonance frequency.

Thanks to the well-specified DC-electric field Vp/d in the sample capacitor, the coupling electric dipole moment p₁ = γ₁d/2 of TLS in the sample material is directly obtained from the identity 2p₁Vp/d = γ₁Vₛ. In contrast, a measurement of the TLS’ coupling strength to a quantum circuit results in the effective dipole moment size p where the matrix element (Δp/E) is often unknown. From measurements on two identical qubits in one cool-down, we characterized in total 138 TLS. Of those, 13 TLS were found inside the sample material, with a spectral density of 4.1 GHz⁻¹ (see calculation details in Supplementary Methods 3), which results in a volume density of the employed AlOₓ (εₓ = 10) of tan δ₀ = nPr₀²(3ε₀εₓ)⁻¹ ≈ 10⁻³, comparable with the number quoted above. The statistics shown in Fig. 2c indicate that the qubits were mostly limited by TLS hosted inside the tunnel barrier of the stray Josephson junctions (light green in Fig. 1d), which are a fabrication artefact that could have been avoided by shorting them in an additional lithography step.

For the 1.5–2 nm thin ⁵¹–⁵³ and 17.17 μm² large tunnel barriers of the two stray junctions shown in Fig. 1d, our measurements indicate a TLS volume density of P₀ wart = 360 to 270 (μm⁻³ · GHz⁻¹) for each junction, in good agreement to previous work.⁴⁰ Notably, this is about six times smaller than the TLS density found in the thicker layer of deposited AlOₓ in the sample capacitor. This is probably due to the minimum detectable TLS dipole moment size, i.e., qubit’s sensitivity, which is smaller for sample-TLS due to stronger oscillating qubit fields (≈90 V/μm) inside the sample capacitor, compared to the field inside the tunnel barrier of the stray junctions (≈15 V/μm). We speculate that this notion might be dressed up with a lot of reasons like a reduced dangling bond density due to facilitated atom diffusibility and self-annealing in the thin tunnel barrier, or enhanced shielding of TLS by the evanescent Cooper-pair condensate, or reasons related to the material’s different growth conditions.

E-field spectroscopy also revealed coherent mutual interactions between TLS in the sample material, which manifest themselves in avoided level crossings as shown in Fig. 3. The coupling between the TLS is described by the interaction Hamiltonian Ĥ int = ν ⋅ (gₓσₓ + gᵧσᵧ), where σₓ and σᵧ are the Pauli matrices of TLS i. As an advancement over earlier work, the combined control of strain and local E-field allowed us to mutually detune the TLS and shift the avoided level crossing through the symmetry point of the observed TLS as demonstrated by the lower panels of Fig. 3. Since the longitudinal coupling component νₓ changes its sign when TLS 1 is tuned through its symmetry point, its effect can be well distinguished from the transversal component νᵧ. This enabled fitting of both components νₓ = −19 (μs)⁻¹ and νᵧ = 25 (μs)⁻¹. More details on the description of coherently interacting TLS can be found in a previous work and in Supplementary Methods 4.
In conclusion, we demonstrated that superconducting qubits can serve as interfaces for studying quantum properties of individual atomic-size tunneling systems located in arbitrary materials deposited as thin films. Qubit swap spectroscopy in dependence on the applied electric field bias to the sample material enables precise measurement of the TLS' coupling dipole moments and reveals avoided level crossings, which herald coherent interaction between TLS. The possibility to mutually detune interacting TLS by using mechanical strain as a second control parameter allows one to fully characterize the type of the interaction. The demonstrated approach has a large potential to provide further insights into the puzzling physics of amorphous solids. It may serve as a valuable tool in the search for low-loss materials urgently needed to advance nano-fabricated devices and superconducting quantum processors where TLS play a detrimental role.

METHODS
Sample fabrication
The qubit samples were fabricated and characterized at KIT. A microchip contained three independent Xmon qubits of whom two were shunted by a sample capacitor, and a third one served as a reference qubit. The qubit electrode, ground plane and resonators were patterned into a 100-nm thick Al film with an inductively coupled Ar–Cl plasma. After Argon-ion milling of the optically patterned electrodes in a PLASSYS shadow evaporation device, the Josephson junctions were deposited in a subsequent electron-beam lithography step.

Qubit samples with large and small sample capacitors were studied. The bottom electrode of large sample capacitors consisted a narrow extension of the qubit island. The bottom layer of the small sample capacitor (see Fig. 1e) was made simultaneously with the Josephson junctions. Sample dielectric and top electrodes of both capacitor types were formed in the PLASSYS device using an MMA/PMMA copolymer mask patterned in an electron-beam lithography step. After removing the native oxide of the bottom electrode with the Ar milling process, the sample dielectric (here 50 nm AlOx) was formed during a perpendicular deposition of Al at a rate of 0.2 nm s⁻¹ in an oxygen atmosphere (chamber pressure of 3 × 10⁻⁴ mBar, oxygen flow of 5 sccm). The dielectric was in situ covered by perpendicularly deposited 100-nm thick layer of Al that formed the top electrode. Further details are reported in the PhD thesis by AB, Chap. 3.2.344.

Experimental setup
The sample was measured in an Oxford Kelvinox 100 wet dilution refrigerator at a temperature of 30 mK. The qubit chip was installed in a light-tight aluminium housing protected by a cryoperm magnetic shield. The coaxial control lines were heavily attenuated, filtered, and equipped with custom-made infrared filters. The qubit state was detected via the dispersive shift of a notch-type readout resonator, which was capacitively coupled to the qubit, and probed in a standard homodyne microwave detection setup.

The DC-gate for tuning the surface-defects consisted of a copper-foil/Kapton foil stack that was glued to the lid of the sample box. It was connected via a twisted pair equipped with an RC-lowpass filter (cutoff ca 10 kHz) at the 1K-stage, and a custom-made copper powder lowpass filter (1 MHz cutoff) at the 30 mK stage. The top electrode of the sample capacitor was controlled via an attenuated microwave line, as further detailed in the Supplementary Methods 2.

DATA AVAILABILITY
Data are available upon reasonable request.
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AUTHOR CONTRIBUTIONS
The qubit samples were designed and fabricated by A.B. Experiments were devised and performed by J.L. in a setup implemented by A.B. and J.L. S.V. performed calculations for the mutually coupled TLS system, and J.B. simulated the electric-field distribution of nanogap capacitors. The manuscript was written by J.L. and A.B. with contributions from all authors.

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