Interacting two-level defects as sources of fluctuating high-frequency noise in superconducting circuits

Clemens Müller,1, 2 Jürgen Lisenfeld,3 Alexander Shnirman,4 and Stefano Poletto5, *

1ARC Centre of Excellence for Engineered Quantum Systems, School of Mathematics and Physics, University of Queensland, Brisbane, Queensland 4072, Australia
2Département de Physique, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1
3Physikalisches Institut, Karlsruhe Institute of Technology, Karlsruhe, Germany
4Institut für Theorie der Kondensierten Materie, Karlsruhe Institute of Technology, Karlsruhe, Germany
5IBM T.J. Watson Research Center, Yorktown Heights, New York 10598, USA
(Dated: March 6, 2015)

Since the very first experiments, superconducting circuits have suffered from strong coupling to environmental noise, destroying quantum coherence and degrading performance. In state-of-the-art experiments it is found that the relaxation time of superconducting qubits fluctuates as a function of time. We present measurements of such fluctuations in a 3D-Transmon circuit and develop a qualitative model based on interactions within a bath of background two-level systems (TLS) which emerge from defects in the device material. Assuming both high- and low-frequency TLS are present, their mutual interaction will lead to fluctuations in the noise spectral density acting on the qubit circuit. This model is further supported by direct measurements of energy fluctuations in a single high-frequency TLS.

PACS numbers: 85.25.Cp, 03.67.Lx, 03.65.Yz
Keywords: superconducting circuits, noise, two-level systems

Superconducting qubits [1] are on the best way towards achieving the prerequisites for fault-tolerant quantum computation schemes [2–4]. With the advent of highly coherent superconducting circuits for quantum applications, previously neglected sources of environmental noise become important. One such cause of decoherence are spurious two-level systems (TLS), which are believed to be present in large numbers in the amorphous dielectric oxide layer covering the superconducting films [5, 6]. Ensembles of TLS naturally explain the low-temperature properties of glasses [7, 8] and are used as a universal model for 1/f-type low-frequency noise in electric circuits [9].

Virtually all designs of superconducting qubit circuits tested so far show a pronounced frequency dependence in their relaxation rates [10–13], which is indicative of strongly coloured high-frequency noise acting on the circuits [14]. A natural explanation of these observations relies on weak interactions between the circuit dynamics and spurious environmental TLS. If the coupling strength is much weaker than the individual decoherence rates of qubit and defect, the effect of the TLS on the qubit will be that of a strongly peaked high-frequency noise spectrum. Additionally, strongly coupled coherent TLS are often found to cause avoided level crossings in superconducting circuits which include Josephson junctions [5, 6]. Those TLS are believed to reside in the dielectric forming the tunnelling barrier inside the circuits Josephson junctions, enabling their stronger coupling to the circuit dynamics. Otherwise they are conjectured to be of the same origin as the TLS observed as resonances in the high-frequency noise spectrum. Using the qubit as a probe, it is possible to fully characterise the properties of these strongly coupled defects, for example by measuring their level-structure and coherence times [15, 16].

In this work, we discuss the origin of time-dependent fluctuations in the energy relaxation time $T_1$, which are observed in superconducting 2D-Transmons [17], flux qubits [18] and 3D-Transmons [13], as shown in Fig. 1. Qubit relaxation may occur through its weak coupling to environmental TLS whose characteristic eigener-
gies are comparable in size to the qubit’s energy splitting. The environmental noise spectral density originating from coupling to a single such TLS is strongly peaked around its eigenenergy. A natural approach to explain the fluctuations in the qubit relaxation rate is thus to assume random changes in the energy splitting of individual two-level defects, c.f. Fig. 2. Our model for the origin of the fluctuations is then based on the presence of a large number of interacting TLS at both low and high eigenfrequencies. Due to the interactions between TLS, thermal switching of the state of low-frequency TLS will then lead to fluctuations in the energy splitting of high-frequency TLS, providing a qualitative description of the observed data. This model is further underpinned by our direct observation of fluctuations in a high-frequency TLS’ energy splitting, which occurs on time scales comparable with the qubit’s $T_1$ relaxation. In the following, we will indicate TLS with eigenenergies much larger than temperature as TS (tunnelling systems), while those at energies much lower than temperature will be named TF (thermal fluctuators).

![Figure 2](image_url)

**Figure 2.** (Color online) Illustration of the mechanism behind the fluctuations in the relaxation rate of a superconducting qubit. We plot the noise spectral density $C(\omega)$, Eq. (2), of a single high-frequency TS as a function of frequency $\omega$. The qubit level splitting is indicated as $\omega_{10}$ and the fluctuating TLS energy as $E$. Fluctuations in $E$, as indicated by the arrow and the dashed contours, will cause strong changes in the noise spectral density at the qubit frequency, leading to significant changes in the qubit relaxation rate $\Gamma_1 \propto C(\omega_{10})$. The inset shows an illustration of the interaction between a central high-frequency TS (red, centre) with a surrounding bath of low-frequency TF (black), where the interaction is limited to a small spatial range, indicated by the grey shaded region.

Our model provides a qualitative description of the origin of fluctuations in the electrical susceptibility of mesoscopic circuits, an area which has recently started to attract attention from both experiment and theory [19–21]. We also note that interactions between TLS have recently been observed directly in two strongly coupled defects [22] and that such a coupling has been invoked as a model of noise before, e.g. to explain the line-width broadening and spectral diffusion of ultrasonic excitations of TLS ensembles in glasses [23, 24] as well as spectral blinking in quantum dots [25, 26]. Moreover, Refs. 27 and 28 make a connection between slow fluctuations in the resonance frequency of superconducting resonators causing phase noise, and ensembles of interacting TLS leading to fluctuations in the energy splitting of high-frequency TS, much along the same lines as we describe here. While in those works the real part of the susceptibility was considered, leading to fluctuations in the level splitting of a resonator, here we are concerned with its imaginary part that is responsible for energy dissipation.

The fluctuations of the $T_1$-time reported here (Fig. 1) were measured in a superconducting qubit in the 3D-Transmon design [13], with an average relaxation time $T_1$ of $\sim 80 \mu$s. In our 3D-Transmon circuit, the qubit energy, i.e. the level splitting of its two lowest levels, is fixed at $\omega_0 = 3.58$ GHz and not tuneable as in other designs [29–33]. Each datapoint results from a series of individual measurements, each time resonantly exciting the qubit and detecting the qubit population after waiting for some time $t$. The resulting traces where fitted to an exponential decay curve $\propto e^{-t/T_1}$. The observed strong fluctuations of the qubit’s relaxation rate $\Gamma_1$ in time do not show any apparent structure. The largest experimentally resolvable fluctuation rate is given by the inverse of the time it takes to obtain a single value of $T_1$, here $\sim 1$ min. Additional data, including for experiments performed at different sample temperatures is shown in appendix B.

In a second experiment, we use a superconducting phase qubit to directly monitor the properties of a single TLS. Fig. 3 (a) shows the energy splitting of an individual high-frequency TS as a function of time. Here, the TS’ resonance frequency $E$ was repeatedly measured by varying the frequency of a long microwave pulse applied to the qubit circuit with a pulse amplitude that was large enough to allow for its direct excitation. During the microwave pulse, the qubit was kept far detuned from the TS. After the pulse, qubit and TS were brought into resonance in order to swap the TS excitation into the qubit, whose population was then measured. Details of this technique can be found in Ref. 16 as well as appendix A. We observe that the TS level splitting is also fluctuating as a function of time, with the observable timescales again restricted by the measurement duration.

In the following, we describe our model explaining the observed fluctuations in the relaxation rate $\Gamma_1 = 1/T_1$ of superconducting circuits. We note that in a master equation description of dissipative quantum dynamics the relaxation rate of a qubit is proportional to the unsymmetrized spectrum of its environment at the frequency of the qubit’s level-splitting, $\Gamma_1 \propto C(\omega_{10})$ [34]. Here we assume effectively zero temperature, $k_B T \ll \omega_{10}$, so that thermal excitations can be neglected. It is then our
Fourier transform of the two-time correlation function of superconducting circuits via their electric dipole moment are believed to be charged entities interacting with the level-splitting double well potential, and $\sigma$ describes the position of a particle on either side of a well. Diagonalizing Eq. (1) yields $\hat{H}_{\text{TLS}} = -\frac{1}{2} \epsilon \sigma_z + \frac{1}{2} \Delta \sigma_x$, where $\epsilon$ is the asymmetry energy between the two wells and $\Delta$ is the tunnel splitting. The Pauli-matrix $\sigma_z$ here describes the position of a particle on either side of a double well potential, and $\sigma_z$ induces transitions between the wells. Diagonalizing Eq. (1) yields $\hat{H}_{\text{TLS}} = \frac{1}{2} E \hat{\sigma}_z$ with the level-splitting $E = \sqrt{\epsilon^2 + \Delta^2}$.

The TS observed in the high-frequency noise spectrum are believed to be charged entities interacting with the superconducting circuits via their electric dipole moment $\propto \sigma_z$ [5]. Assuming the interaction strength to be weak, their effect on the qubit will be that of a strongly coloured noise spectral density, which can be calculated from the Fourier transform of the two-time correlation function of their coupling operator $\sigma_z$ [14]. We obtain

$$C(\omega) = \int dt e^{-i \omega t} \langle \sigma_z(t) \sigma_z(0) \rangle$$

$$= \cos^2 \theta \left[ 1 - \langle \sigma_z \rangle^2 \right] \frac{2 \gamma_1}{\gamma_1^2 + \omega^2}$$

$$+ \sin^2 \theta \left[ \frac{1 + \langle \sigma_z \rangle}{2} \right] \frac{2 \gamma_2}{\gamma_2^2 + (\omega - E)^2}$$

$$+ \sin^2 \theta \left[ 1 - \langle \sigma_z \rangle \right] \frac{2 \gamma_2}{\gamma_2^2 + (\omega + E)^2}$$  \hspace{1cm} (2)

with the TLS’ equilibrium steady-state population $\langle \sigma_z \rangle = \tanh (E/2k_B T)$, the intrinsic TLS relaxation rate $\gamma_1$ and $\gamma_2 = \frac{1}{2} \gamma_1 + \gamma_\phi$, where $\gamma_\phi$ is the pure dephasing rate of the TLS. Here, $\gamma_\phi = \Delta/\epsilon$ defines the mixing angle of the TLS. Eq. (2) describes the noise spectrum which acts on the qubit circuit from a single TLS’s electric dipole moment. Depending on circuit design, this interaction may be described by different qubit operators [35], for the Transmon design it generally leads to flips of the qubit state. Eq. (2) is decomposed into three parts, each of which becomes relevant for TLS in different parameter regimes. The first line describes low-frequency noise due to the random switching of the TLS between its two states and is most pertinent for low-frequency TF with $E \ll k_B T$. The second term is a high-frequency contribution which is sharply peaked around the TLS energy and is most pronounced for TS with $E \gg k_B T$. Since the TS are mostly resting in their ground state, they are able to absorb energy from the qubit. It is this contribution that gives rise to the observed resonances in the noise spectrum [10–12] and in which we are mostly interested. The final term contributes at negative frequencies and describes the ability of the TLS to excite the qubit by transferring an excitation to it. For both high-frequency TS in thermal equilibrium as well as low-frequency TF this term will not contribute measurably.

For simplicity, we assume the environmental noise at frequencies close to the qubit level splitting $\omega_{10}$ is dominated by a single, weakly coupled high-frequency TS at energy $E \sim \omega_{10}$. We further assume this TS is interacting with a large number of other TLS which are located in its close spatial vicinity. This is the situation illustrated in Fig. 2 and the one most relevant to experiment [10–12]. If the distribution of TS at high frequencies is dense [14, 27], our results still hold but have to be averaged additionally over the high-frequency distribution. We model the interaction between all TLS in the sample by a Hamiltonian of the form

$$\hat{H} = \frac{1}{2} \sum_j g_j \hat{\sigma}_z \hat{\sigma}_{z,j}$$ \hspace{1cm} (3)

where $g_j$ is the coupling strength between the high-frequency TS and all other TLS, indicated by the index $j$. Coupling of the type Eq. (3) can be caused e.g., by...
dipolar or strain-like interaction, where the asymmetry-energy of either TLS depends on the relative position of the other TLS in its respective double-well potentials [22]. With such an interaction, the energy splitting of any TLS depends on the instantaneous state of all TLS in a certain range around it, determined by the microscopic origin of their interaction, c.f. inset to Fig. 2 and appendix C.

In the calculations one has to carefully separate the different timescales of the problem. The measurement protocol fixes three distinct scales, which have to be compared to the fluctuation rates of individual low-frequency TF to determine the nature of their contribution to the fluctuations in the qubit’s relaxation rate $\Gamma_1$. First, there is the time it takes to do a single measurement of the qubit population, $t_{\text{meas}}$, where many such measurements are averaged to obtain each point in a complete relaxation curve. Fluctuating TF that are faster than $1/t_{\text{meas}}$ will act as an effective broadening of the high-frequency TS resonance, increasing its line-width $\gamma_2$. Second, there is the time to measure a single point of a curve, $t_{\text{point}}$. Fluctuations that are slower than $1/t_{\text{point}}$, but faster than $1/t_{\text{meas}}$ will lead to jitter in the energy relaxation curve, contributing additional noise in the fit of $\Gamma_1$. The final timescale is given by the duration of the measurement of a complete $\Gamma_1$ curve, $t_{\text{T}_1}$. Slow TF that fluctuate at frequencies that are smaller than $1/t_{\text{T}_1}$ will be the ones responsible for the low-frequency fluctuations visible in the $\Gamma_1$ data, as shown in Fig. 1. Note that the microscopic origin of these small TF switching rates is so far unclear [27, 36].

We first calculate the average $\langle \Gamma_1 \rangle$ in the case described above. To this end we write the TS energy as $E = E - \sum_j g_j \sigma_{z,j}$. We then expand the TS induced $\Gamma_1$ for small inter-TLS coupling $g_j \ll \gamma_2 \ll E$ and average the resulting expression over TLS parameter distributions, see appendix C for details. For the ensemble averaged qubit relaxation rate we find

$$\langle \Gamma_1 \rangle \propto \frac{2\gamma_2}{\gamma_2^2 + \delta\omega^2} \propto \left\{ \begin{array}{ll} T^{-(\alpha+1)} & , \delta\omega \lesssim \gamma_2 \\ T^{\alpha+1} & , \delta\omega \gtrsim \gamma_2 \end{array} \right. ,$$

where $\delta\omega = \omega_{10} - E$ is the detuning between qubit and TS and we assumed a TLS parameter distribution of the form $P(\epsilon, \Delta) \propto e^{\alpha} / \Delta$ [14]. For non-interacting TLS, the distribution is usually assumed to be flat, $\alpha = 0$ [7, 8], but might be different from zero in the more realistic case of interacting TLS [14, 27]. The temperature dependence here is due to the behaviour of the TF induced TS dephasing rate $\gamma_2 \propto T^{\alpha+1}$ [14, 27]. Additionally, we find the temperature and frequency dependence of the $\Gamma_1$ fluctuation-spectrum as

$$\langle \Gamma_1(0) \Gamma_1(t) \rangle_\omega \propto \omega^{-1} \left\{ \begin{array}{ll} T^{-5(\alpha+1)} & , \delta\omega \ll \gamma_2 \iota \\ T^{-3(\alpha+1)} & , \delta\omega \sim \gamma_2 \iota \\ T^{2(\alpha+1)} & , \delta\omega \gg \gamma_2 \iota \end{array} \right. ,$$

Here, the $1/\omega$ behaviour stems from the canonical form of the switching rate distribution $P(\gamma) \propto 1/\gamma$ and the fact that we necessarily observe the fluctuations on timescales that are longer than the inverse of the inverse maximum flipping rate. As stated, these results hold for small inter-TLS coupling $g_j \ll \gamma_2$. The opposite case $g_j \gg \gamma_2$, corresponds to on-off switching and is excluded by the experimental data. In the intermediate regime, $g_j \sim \gamma_2$, the overall temperature dependence will be given as an average over our results. More details on the derivation can be found in appendix C, where we additionally discuss the case when the TF switching is solely due to interaction with phonons.

Our model can be directly tested by measuring the relaxation rate at different qubit level splittings and inferring the time and frequency dependence of the noise spectrum acting on the qubit. By using a frequency-tunable qubit, the fluctuations in the noise spectral density might be directly resolvable in time and frequency, depending on the timescale of a single measurement of the relaxation time $T_{\text{T}_1}$. Even for non-tuneable qubits it is possible to probe the noise spectral density in close vicinity of the qubit frequency by measuring the decay of Rabi oscillations of the qubit, c.f. appendix D and Ref. 37. Another possibility is to apply external driving to saturate the TF responsible for the fluctuations in TS energy. If an electric field is applied resonantly with the low-frequency TF, it will lead to oscillations with the Rabi frequency depending on the detuning between drive tone and TF energies, the TF dipole moments and the electric field strength at their position. Assuming the resulting Rabi frequency is fast compared to the duration of a single $T_{\text{T}_1}$ measurement, the effect would be to lower the average $\langle \Gamma_1 \rangle$ while at the same time reducing the amplitude of its fluctuations. This is because the resonant driving of initially very slow TF will alter their contribution towards a simple line-width broadening of the high-frequency TS. In experiments with 3D-Transmon qubits this could be achieved by careful engineering of the cavity modes, such that there exists a suitable low-frequency mode exhibiting strong electric field components spatially close to the qubit. In other qubit architectures, e.g. with phase qubits, this might be possible within existing experimental setups [16]. In our Transmon qubit sample, this experiment proved unfeasible due to design restrictions in the employed cavity. Additional verification could be achieved by a systematic characterisation of the fluctuations of $T_{\text{T}_1}$ at a variety of experimental temperatures $T$. An additional challenge arises from the fact that the exact temperature dependence is connected sensitively to the qubit-TS detuning $\delta\omega$, c.f. Eq. (5), which also has to be determined in this case.

Possible alternative models for the fluctuating noise spectrum include fluctuations of the quasiparticle density in the superconductor. Quasiparticle tunnelling across
the circuit’s Josephson junctions can induce relaxation and dephasing [38], and explains well the temperature dependence of qubit relaxation rates for elevated sample temperatures. In contrast to our model of interacting TLS as sources of the fluctuations, which depends on a structured noise spectrum as background, the quasiparticle induced noise is flat at high-frequencies. Following the theory of Ref. 38 we calculate the fluctuations in quasiparticle density required to effect the observed variance in the relaxation time of the transmons. For the parameters of our sample, we find the fluctuation in the quasiparticle volume density required to change the relaxation rate by 1 kHz as \( \delta n_{qp} \approx 0.5/\mu \text{m}^3 \), see appendix E for details. It follows that this change would require the number of quasiparticles present on either one of the qubit islands to fluctuate by \( \delta N_{qp} \approx 1.5 \times 10^4 \). We are not aware of any mechanism leading to fluctuations in the quasiparticle number of this magnitude.

Another possible model is that in the 3D-Transmon sample we used to obtain Fig. 1, the qubit level splitting was fluctuating as a function of time, e.g. due to changes in the critical current of the circuits Josephson junction [39]. Together with the observed strong structure in the noise spectrum [10–12] this would also explain the fluctuations in the qubit relaxation. This mechanism can however be ruled out since in our measurements the qubit is always resonantly excited, as evident from the fits to the raw data, c.f. appendix B.

In conclusion, we present a simple model of interacting TLS which offers a qualitative understanding of the observed fluctuations in relaxation times \( T_1 \) in superconducting circuits. The model is grounded in our experimental observations, grants a clear route towards further confirmation, and provides a way to verify and refine the existing microscopic TLS models. Moreover, our model clearly indicates that parasitic TLS are a limiting factor in today’s best performing superconducting circuits. A better understanding of this decoherence source is thus vital for further improving the fidelity of superconducting quantum circuits.

We thank G.A. Keefe and M.B. Rothwell for fabricating the 3D-Transmon sample used in this work, and A. Blais, J. Clarke, J.H. Cole, M. Marthaler, T.M. Stace and M. Steffen for valuable comments and discussions. This work was partially supported by IARPA through contract W911NF-10-1-0324. CM acknowledges the support of the RMIT Foundation through an International Research Exchange Fellowship.

* present address: QuTech Advanced Research Center and Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands

[1] J. Clarke and F. K. Wilhelm, Nature 453, 1031 (2008).
[2] R. Barends, J. Kelly, A. Megrant, A. Veitia, D. Sank, E. Jeffrey, T. C. White, J. Y. Mutus, A. G. Fowler, B. Campbell, Y. Chen, Z. Chen, B. Chiaro, A. Dunsworth, C. Neill, P. O’Malley, P. Roushan, A. Vainsencher, J. Wenner, A. N. Korotkov, A. N. Cleland, and J. M. Martinis, Nature 508, 500 (2014).
[3] J. M. Chow, J. M. Gambetta, E. Magesan, D. W. Abraham, A. W. Cross, B. R. Johnson, N. A. Masheskl, C. A. Ryan, J. Smolin, S. J. Srinivasan, and M. Steffen, Nature Communications 5, 4015 (2014).
[4] O. P. Saira, J. P. Groen, J. Cramer, M. Meretska, G. de Lange, and L. DiCarlo, Phys. Rev. Lett. 112, 070502 (2014).
[5] R. W. Simmonds, K. M. Lang, D. A. Hite, D. P. Pappas, and J. M. Martinis, Phys. Rev. Lett. 93, 077003 (2004).
[6] Y. Shalibo, Y. Rofe, D. Shwa, P. Zeides, M. Neeley, J. M. Martinis, and N. Katz, Phys. Rev. Lett. 105, 177001 (2010).
[7] W. A. Phillips, Journal of Low Temperature Physics 7, 351 (1972).
[8] P. W. Anderson, B. I. Halperin, and C. Varma, Philosophical Magazine 25, 1 (1972).
[9] P. Dutta and P. M. Horn, Reviews of Modern Physics 53, 497 (1981).
[10] O. V. Astafiev, Y. A. Pashkin, Y. Nakamura, T. Yamamoto, and J.-S. Tsai, Phys. Rev. Lett. 93, 267007 (2004).
[11] G. Ihler, E. Collin, P. Joyce, P. J. Meeson, D. Vion, D. Esteve, F. Chiarello, A. Shinirman, Y. Makhlin, J. Schriefl, and G. Schönh, Phys. Rev. B. 72, 134519 (2005).
[12] R. Barends, J. Kelly, A. Megrant, D. Sank, E. Jeffrey, Y. Chen, Y. Yin, B. Chiaro, J. Y. Mutus, C. Neill, P. O’Malley, P. Roushan, J. Wenner, T. C. White, A. N. Cleland, and J. M. Martinis, Phys. Rev. Lett. 111, 080502 (2013).
[13] H. Paik, D. I. Schuster, L. S. Bishop, G. Kirchmair, G. Catelani, A. P. Sears, B. R. Johnson, M. Reagor, L. Frunzio, L. I. Glazman, S. M. Girvin, M. H. Devoret, and R. J. Schoelkopf, Phys. Rev. Lett. 107, 240501 (2011).
[14] A. Shirnirman, G. Schönh, I. Martin, and Y. Makhlin, Phys. Rev. Lett. 94, 127002 (2005).
[15] P. A. Bushev, C. Müller, J. Lisenfeld, J. H. Cole, A. Lukashenko, A. Shinirman, and A. V. Ustinov, Phys. Rev. B. 82, 134530 (2010).
[16] J. Lisenfeld, C. Müller, J. H. Cole, P. A. Bushev, A. Lukashenko, A. Shinirman, and A. V. Ustinov, Phys. Rev. Lett. 105, 230504 (2010).
[17] P. Macha, (2014), private communication.
[18] J. Clarke, (2014), private communication.
[19] S. Sendelbach, D. Hover, M. Mück, and R. Mcdermott, Phys. Rev. Lett. 103 (2009).
[20] C. Neill, A. Megrant, R. Barends, Y. Chen, B. Chiaro, J. Kelly, J. Y. Mutus, P. O’Malley, D. Sank, J. Wenner, T. C. White, Y. Yin, A. N. Cleland, and J. M. Martinis, Applied Physics Letters 103, 072601 (2013).
[21] P. Schad, B. N. Narozhny, G. Schönh, and A. Shirnirman, Phys. Rev. B. 90, 205419 (2014).
[22] J. Lisenfeld, G. J. Grabovskij, C. Müller, J. H. Cole, G. Weiss, and A. V. Ustinov, Nature Communications 6, 6182 (2015).
[23] W. Arnold and S. Hunklinger, Solid State Commun. 17, 883 (1972).
[24] J. Black and B. I. Halperin, Phys. Rev. B. 16, 2879.
The three dimensional cavity resonator used in this work was machined from bulk aluminium 6061. The cavity has a nominal size of $(18.6 \times 15.5 \times 4.2) \text{ mm}^3$ engineered to give a resonant frequency of approximately 12 GHz. Two bulk head SMA connectors are used as input and output ports. The loaded quality factor of the waveguide cavity is $3 \times 10^4$, with the output connector stronger coupled than the input connector in order to guarantee a high signal-to-noise ratio. The sample is shielded with a cryoperm can that is thermally anchored to the mixing chamber of the dilution refrigerator.

The qubit manipulation and readout pulses are delivered to the cavity via a single coax line filtered by a 10 dB attenuator at each temperature stage of the refrigerator. Input and output ports are directly connected to SMA filters made of eccosorb in order to block infrared radiation and thermalize the central conductor of the coupling connectors.

The output readout signal is amplified by a chain of cryogenic and room temperature amplifiers for a total gain of 60 dB. A low pass filter and two cryogenic circulators are used between the sample and the cryogenic amplifier. The qubit state is readout via the dispersive shift of the waveguide cavity [40].

The qubit is fabricated on sapphire substrate via aluminium double angle evaporation. Two rectangular pads of $(350 \times 700) \mu \text{m}^2$, separated by 50 $\mu \text{m}$, are connected by a 1 $\mu \text{m}$ wide aluminium strip with a Josephson junction of size $(0.1 \times 0.1) \mu \text{m}^2$. The chip has a total size of $(3.0 \times 6.7) \mu \text{m}^2$ and is kept in place in the waveguide cavity by small pieces of indium. The qubit is placed at the maximum of the electric field of the first cavity mode.

The qubit used in this work has an energy gap $\omega_0$ of $3.5825$ GHz, and anharmonicity ($\approx E_C$) of $171$ MHz. The qubit is designed to work in the transmon regime, with $E_J/E_C \sim 61$ [41].

### Observation of $T_1$-fluctuations of a 3D-Transmon

Our theoretical model to explain time-dependent fluctuations in the energy relaxation time of superconducting qubits is based on their near-resonant coupling to high-frequency TLS, with the additional assumption that those TLS themselves experience resonance frequency variations due to their interaction with thermally fluctuating defects at low frequencies. In this work, we include first experimental evidence that individual high-frequency TLS may indeed show resonance frequency fluctuations in time as shown in Fig. 3.
In order to access TLS individually, we exploit their strong coupling to the state of a superconducting phase qubit when they are residing in the amorphous tunnel barrier of the qubit’s Josephson junction. We were using a phase qubit sample that has been developed in the group of Prof. J. Martinis at University of California, Santa Barbara, USA, with sample parameters as described in Ref. 42.

We recorded the Lorentzian resonance curve of the TLS by varying the frequency of a long microwave pulse applied to the circuit while the qubit was kept far detuned from the TLS. As described in Ref. 16, this allows one to resonantly drive TLS while they remain effectively decoupled from the qubit dynamics. To read out the TLS quantum state, the qubit is first prepared in its ground state and then tuned into the TLS resonance. This realises an iSWAP operation that maps the TLS state onto the qubit, where it can be measured.

Some of the TLS that were investigated with this method showed time-dependent fluctuations of their resonance frequency that were large enough to be resolved spectroscopically. Often, we observe telegraph-signal like switching of TLS resonance frequencies between two similar values, indicating coupling to one dominating thermally activated TLS at low frequency.

To characterise the internal TLS parameters, tunnelling energy $\Delta$ and asymmetry energy $\varepsilon$ were measured by recording the strain dependence [43] of its resonance frequency and performing a hyperbolic fit to the equation $E = \sqrt{\Delta^2 + \varepsilon^2}$. Figure 3 was obtained on a TLS that had $\Delta/h = 7.056$ GHz and whose asymmetry energy was tuned to $\varepsilon/h = 918$ MHz. At this asymmetry, this TLS had an energy relaxation time of $T_1 \approx 590$ ns and a dephasing time of about $T_2 \approx 500$ ns. The sample temperature was kept at $33$ mK.

**APPENDIX B: EXPERIMENTAL DATA**

Figure S1 shows two examples of measured relaxation curves of the 3D-Transmon qubit and the fits to the data. We fit the measurements to decay curves of the form $Ae^{-\Gamma_1 t} + B$ with the free parameters $A$, $B$ and $\Gamma_1$. We show one trace where the fit converged with a very small standard error (a) and another where the convergence was worse (b). The second trace might be better fit by assuming a double exponential decay where at some time the decay rate changed spontaneously due to a change in the environmental noise spectrum (not shown).

Figs. S2-S4 show the full datasets of the fluctuations in the relaxation rate $\Gamma_1$ measured in our 3D-Transmon at three different experimental temperatures. We also show the histograms for the probability of occurrence of a particular value of $\Gamma_1$ for all three temperatures as well as the fluctuations in the fit amplitude $A$ and background $B$. The later two show some fluctuations, but are relatively flat on the scale of the changes observed in $\Gamma_1$. Amplitude fluctuations might be explained if the qubit’s level splitting varies in time, which, together with a strongly coloured high-frequency noise spectrum provides an alternate model for the fluctuations in the qubit’s relaxation rate (c.f. main text). From the data in Figs. S2-S4, we conclude that this mechanism might be present but is weak and not the main contribution. Additionally, we show the two-time correlation function of the relaxation rate as well as its Fourier transform. We fit the $T_1$-fluctuation spectrum to two different functions and show the results in the plots. The red dashed lines are from the best fit to the function $A/\omega^\alpha$, corresponding to a $1/f$-type frequency distribution as is expected from a dense distribution of low-frequency TLS [9, 14] The blue dashed lines are results from a fit to a zero-frequency Lorentzian $\sim A\gamma/(\gamma^2 + \omega^2)$, as would result from a single dominant low-frequency TLS, c.f. Eq. (2). For our data presented here, the temperature dependence of the fluctuation amplitude is inconclusive and does not give any
Supplementary Figure S2. Experimental data on $T_1$ fluctuations in the 3D transmon sample at a temperature of 30 mK. (a) shows the relaxation rates $\Gamma_1$ from fits of the experiments to an exponential decay curve, $P(|1\rangle) = A e^{-\Gamma_1 t} + B$, with error bars corresponding to the 95% confidence interval of the fits. The black dashed lines are a moving average over 10 points and the red dotted lines are the mean values over the full dataset. The inset shows a histogram of the probabilities of values for the relaxation rate $\Gamma_1$. (b) shows the time evolution of excitation amplitudes $A$ and background $B$ from the same fits, including error bars and moving averages in black. (c) depicts the absolute value of the Fourier transform of the two-time correlation function of the relaxation rates $\langle \Gamma_1(t)\Gamma_1(0) \rangle$, with the inset showing the correlation function itself. The red (blue) dashed curve is the result of a fit of the data to a $A/\omega^\alpha$-spectrum (Lorentzian spectrum $A/\gamma^2/(\gamma^2+\omega^2)$) with fit parameters $A = 0.097$ and $\alpha = 0.58$ ($A = 0.18$ and $\gamma = 0.34$ mHz), for details see text.

Supplementary Figure S3. Same as Fig. S2, data taken from experiments performed at a temperature of 50mK. Fit parameters in (c) are $A = 0.079$ and $\alpha = 0.79$ ($A = 0.087$ and $\gamma = 0.052$ mHz) for red (blue) dashed line.

APPENDIX C: CALCULATIONS

Here we give details on the calculations of the mean value and spectrum of the $T_1$-fluctuations in a superconducting circuit due to interactions within a bath of spurious background TLS. We assume a sparse distribution of relatively coherent high-frequency TS, $E \gg k_B T, \gamma_2$, which are weakly coupled to the qubit, leading to sharp indication if our model is accurate. On the other hand, the frequency dependence of the correlations seems to follow roughly a $1/\omega$ dependence, which could be explained in the terms of our model.
features in the noise spectral density [10–12]. Further we conjecture that the overall distribution of TLS energies is strongly peaked towards low frequencies [14]. Then any superconducting circuit will interact with a large total number of TLS, the majority of which is weakly coupled to its dynamics. For the 3D-Transmon measured in this work, and assuming a 5 nm thick oxide layer over the whole area of our sample, we expect the energy density of TLS interacting with our circuit to be $\sim 10^5$ GHz. Here we take the TLS energy/volume density as $10^2$ ($\mu$m$^3$ GHz) [6, 12].

We conceptually divide the distribution of TLS into three parts, (i) high-frequency TS ($E \gg k_B T$) responsible for qubit relaxation, (ii) low-frequency TF ($E \ll k_B T$) responsible for qubit dephasing and (iii) slowly fluctuating TF. As of now, there is no clear consensus in the community as to the origin of the slow fluctuations observed in experiments. Here we simply state that there is a mechanism that leads to random switching of the state of individual TLS on the observed timescales $\sim \text{min}$.

In order to arrive at time-dependent $T_1$ fluctuations, we additionally need a mechanism which randomly changes the energy of high-frequency TS on the timescales seen in experiment. We conjecture that this mechanism is an interaction with TFs at low to intermediate energy scales, whose fluctuations show the timescales corresponding to the experiments.

As explained in the main text, we assume an interaction between individual TLS in the ensemble of the form

$$\hat{H} = \sum_{<ij>} g_{ij} \hat{\sigma}_z,i \hat{\sigma}_z,j,$$  

where the sum includes all pairs of TLS which are coupled with the coupling strength $g_{ij}$. Comparing with Eq. (3), we here write the expression for the coupling between all TLS present, while we earlier restricted ourselves to coupling between a single high-frequency TS and its surrounding defects. This form of the coupling is motivated by recent experiments, where it was used to explain experiments on two TLS that were strongly coupled to a phase qubit and interacting also with each other. Such coupling will originate from electromagnetic interaction between the two TLS dipoles, or related to this a coupling mitigated via deformation of the surrounding atomic potentials through strain [22].

We are looking at fluctuations in the qubit relaxation rate due to slow fluctuations in the TLS energies $E$. In order to calculate expectation values and statistics, we write the level splitting of an individual TLS as an operator

$$\hat{E}_i = E_{i,0} - \sum_{<ij>} g_{ij} \hat{\sigma}_z,j,$$  

now depending on the state of all other TLS via the mutual interaction $g_{ij}$ from Eq. (S1). Here we focus on high-frequency TS with $E_{i,0} \gg k_B T$, $\gamma_2,i$, such that $\langle \sigma_{z,i} \rangle = -1$ and the resulting spectral density is strongly peaked around the TS eigenenergy $E_i$, c.f. Eq (2). We defined the undisturbed TLS level splitting as $E_{i,0} = \sqrt{\epsilon_i^2 + \Delta_i^2}$ with the parameters $\epsilon$ and $\Delta$ from Eq. (1).

We can further write the qubit relaxation rate due to its coupling to TLS as

$$\Gamma_1 = |\langle 1 | \hat{\sigma} | 0 \rangle|^2 \sum_i \tilde{\gamma}_{q,i},$$  

where the operator $\hat{\sigma}$ is the qubit operator that couples the qubit dynamics to the TLS position operator $\sigma_{z,i}$, and $|0\rangle$, $|1\rangle$ are the qubit ground- and excited states. Here, the individual rates induced by a single TLS are
given by the high-frequency components of its spectral density, c.f. Eq. (2), as
\[ \hat{\gamma}_{q,i} = \cos^2 \theta_i \frac{2 \gamma_{2,i}}{\gamma_{2,i}^2 + (\omega_{10} - E_{i,0})^2}. \] (S4)

Assuming the interaction between individual TLS to be weak, \( g_{ij} \ll \gamma_{2,i} \), we can expand this to first order as
\[ \hat{\gamma}_{q,i} = \gamma_{q,i}^{(0)} + \gamma_{q,i}^{(1)} \sum_{\langle ij \rangle} g_{ij} \hat{\sigma}_z + O(g^2), \] (S5)
with the expansion coefficients
\[ \gamma_{q,i}^{(0)} = \cos^2 \theta_i \frac{2 \gamma_{2,i}}{\gamma_{2,i}^2 + (\omega_{10} - E_{i,0})^2}, \] (S6)
\[ \gamma_{q,i}^{(1)} = \frac{\partial \gamma_{q,i}}{\partial E_i} \bigg|_{E_i = E_{i,0}} = \cos^2 \theta_i \frac{4 \gamma_{2,i}(\omega_{10} - E_{i,0})}{(\gamma_{2,i}^2 + (\omega_{10} - E_{i,0})^2)^2}. \] (S7)

These equations will be the basis for further calculations.

### Distribution of parameters

For tunneling TLS one usually assumes flat distributions for both the asymmetry energy \( \epsilon \) as well as the tunneling barrier height \( [7, 8] \). Since the tunneling energy \( \Delta \) depends exponentially on the barrier, the resulting distribution in TLS parameters is \( P(\epsilon, \Delta) \sim 1/\Delta \).

The TLS relaxation rates are then also distributed log-uniformly, \( P(\gamma_1) \sim 1/\gamma_1 \), since the tunnelling strength depends mainly on the size of the tunneling barrier. In Ref. 14 it was found that a linear or super-linear distribution in TLS parameters is as stemming from the same ensemble of TLS. For the sake of generality, we will therefore assume a distribution of TLS parameters of
\[ P(\epsilon, \Delta) = \frac{\alpha^\epsilon}{\Delta} d\epsilon d\Delta, \] (S8)
with \( \alpha \geq 0 \) and the constant \( A \) needed for normalization. Without loss of generality we restrict the integration to the positive real axis. Rewriting Eq. (S8) in terms of the TLS level-splitting \( E \) and the mixing angle \( \theta = \arctan \Delta/\epsilon \), we find
\[ P(E, \theta) = A E^{\cos \theta} \frac{\cos \theta}{\sin \theta} dE d\theta. \] (S9)

When describing the full distribution of TLS for all energies, we integrate the tunnel splitting \( \Delta \) between \( \Delta_{\text{Min}} \geq 0 \) and \( \Delta_{\text{Max}} \) and the asymmetry energy \( \epsilon \) between \( \epsilon_{\text{Min}} = 0 \) and \( \epsilon_{\text{Max}} \). We find for the integration bounds in the new variables: \( \theta_{\text{Min}} = \arctan \Delta_{\text{Min}}/\epsilon_{\text{Max}} \geq 0, \theta_{\text{Max}} = \arctan \Delta_{\text{Max}}/\epsilon_{\text{Min}} = \pi/2 \) and \( E_{\text{Min}} = \sqrt{\Delta_{\text{Min}}^2 + \epsilon_{\text{Min}}^2} = \Delta_{\text{Min}}, E_{\text{Max}} = \sqrt{\Delta_{\text{Max}}^2 + \epsilon_{\text{Max}}^2} \). Here, \( \Delta_{\text{Min}} \) is defined by the minimum tunneling barrier below which the description as a two-level system breaks down and \( E_{\text{Max}} \) provides an upper bound on the TLS level-splitting.

The distribution of inter-TLS coupling strengths \( g_{ij} \) depends strongly on the physical model of their interaction. For example, for dipolar interaction with \( |g| \sim 1/r^3 \), one finds
\[ P(g) dg = P(r) \frac{\partial r}{\partial g} dg = \rho_0 |g|^{-\frac{4}{3}} dg, \] (S10)
where we assumed a constant TLS density in space \( \rho_0 \). It is important to note that the coupling strength \( g \) in most models can be both positive or negative, meaning the coupling between the TLS can either raise or lower the energy of the respective partners. For the dipole coupling model this reflects the fact that the relative orientation of the dipoles can be both parallel as well as antiparallel.

### Performing the average

We now turn to calculating the values for the average and variance of the qubit’s relaxation rate using the distributions motivated above. We concentrate here on fluctuations originating from the low frequency contributions from TLS with small level splitting, \( E \lesssim k_B T \), since those are the ones directly observable in experiment.

Noting that \( \langle \hat{\sigma}_z \rangle = \cos \theta \langle \sigma_z \rangle = \cos \theta \tan \theta \langle E/2k_B T \rangle \), we can directly write down the mean value of the qubit relaxation rate due to the high-frequency TLS to lowest order in the inter-TLS coupling strength \( g \) as
\[ \langle \hat{\gamma}_{q,i} \rangle = \gamma_{q,i}^{(0)} + \gamma_{q,i}^{(1)} \sum_j g_j \cos \theta_j \tan \theta \frac{E_j}{2T} \]
\[ = \gamma_{q,i}^{(0)} + \gamma_{q,i}^{(1)} \int dg \sin \theta dE \frac{P(g, \theta, E) g \cos \theta \tan \theta \frac{E}{2T}}{E}, \] (S11)
where the sum includes all other two-level defects that our high-frequency TLS is interacting with. In the calculation of the average rate \( \langle \gamma_{q,i} \rangle \), we immediately notice that
\[ \int dg g P(g) = 0, \] (S12)
since we integrate an odd function over an even range. Therefore we simply find
\[ \langle \hat{\gamma}_{q,i} \rangle = \gamma_{q,i}^{(0)}, \] (S13)
i.e., the average relaxation rate due to a single TLS is given by its spectrum centred around its undisturbed level-splitting \( E_{i,0} \).

The spectrum of fluctuations of the relaxation rate is then given by the Fourier transform of the rate correlation function as
\[
\langle \hat{\gamma}_{q,i}(0)\hat{\gamma}_{q,i}(t) \rangle_\omega = \int dt \; e^{-i\omega t} \langle \hat{\gamma}_{q,i}(0)\hat{\gamma}_{q,i}(t) \rangle
\]
\[
= \left( \gamma_{q,i}^{(1)} \right)^2 \sum_{j,l} g_j g_l \langle \hat{\sigma}_{z,j}(t)\hat{\sigma}_{z,l}(0) \rangle_\omega
\]
\[
= \left( \gamma_{q,i}^{(1)} \right)^2 \sum_j g_j^2 \cos^2 \theta_j \left[ 1 - \tanh^2 \left( \frac{E_j}{2T} \right) \right] \frac{2\gamma_{1,j}}{\gamma_{1,j}^2 + \omega^2}
\]
\[
= \left( \gamma_{q,i}^{(1)} \right)^2 \int \int \; dg \; d\theta \; dE \; d\gamma_{1} \; P(g,\theta,E,\gamma_{1}) g_j^2 \cos^2 \theta \left[ 1 - \tanh^2 \left( \frac{E}{2T} \right) \right] \frac{2\gamma_{1}}{\gamma_{1}^2 + \omega^2}, \tag{S14}
\]

where, in evaluating the correlator, we restrict ourselves to only the low frequency contribution of the TLS autocorrelation function Eq. (2), i.e. we focus on TFs with \( E \ll k_B T \). Additionally we have assumed that different TLS are uncorrelated, \( \langle \sigma_{z,j}\sigma_{z,l} \rangle = 0 \). We are also only interested in the bare fluctuations of the rate, so we have already subtracted the mean value above.

For the average over the coupling strength, one finds
\[
\int dg \; g^2 P(g) \propto \text{const}, \tag{S15}
\]
where the constant is mainly determined by the maximum possible coupling strength and thus by the minimal distance between TLS and the microscopic origin of their interaction. Performing the average over the mixing angle \( \theta \) also contributes a constant, with the exact value again depending on details of the microscopic TLS model. The average over TF energy can be written as
\[
\int dE \; P(E) \left( 1 - \tanh^2 \left( \frac{E}{2T} \right) \right) \approx \int_0^T dEE^\alpha = T^{\alpha+1}, \tag{S16}
\]
contributing to the temperature dependence of the final result.

We now have to take care of the separation of timescales in our problem. We assume a large ensemble of low-frequency TLS in our model. The majority of these TLS will be switching fast on the timescale of a single \( T_1 \) measurement, and thus does not contribute directly to the observed \( T_1 \) fluctuations. They will however lead to fluctuations in the energy splitting of the high-frequency TLS close to the qubit frequency, and so contribute to its line width \( \gamma_{2,i} \), which appears in the prefactor \( \left( \gamma_{q,i}^{(1)} \right)^2 \). Following Refs. 14 and 27 we find the temperature dependence of the dephasing rate due to a bath of low-frequency TFs as \( \gamma_{2} \propto T^{\alpha+1} \). Still assuming small interaction strength between TLS, \( g \ll \gamma_{2,i} \), we can now distinguish three regimes related to the initial detuning between our qubit and the high-frequency TS, \( \delta\omega = \omega_{10} - E_{i,0} \). For qubit and high-frequency TLS near resonance, \( \delta\omega \ll \gamma_{2,i} \), we find that \( \gamma_{q,i}^{(1)} \propto \delta\omega/\gamma_{2,i}^3 \), and thus the prefactor behaves as \( \left( \gamma_{q,i}^{(1)} \right)^2 \propto 1/T^{6(\alpha+1)} \).

In the regime of intermediate detuning, \( \delta\omega \approx \gamma_{2,i} \), one finds \( \gamma_{q,i}^{(1)} \propto 1/\gamma_{2,i}^2 \) and therefore \( \left( \gamma_{q,i}^{(1)} \right)^2 \propto 1/T^{4(\alpha+1)} \).

In the far detuned regime, \( \delta\omega \gg \gamma_{2,i} \), we finally have \( \gamma_{q,i}^{(1)} \propto \gamma_{2,i}/\delta\omega^3 \) leading to \( \left( \gamma_{q,i}^{(1)} \right)^2 \propto T^{2(\alpha+1)} \).

\[
\langle \hat{\gamma}_{q,i}(0)\hat{\gamma}_{q,i}(t) \rangle_\omega \propto \begin{cases} T^{-5(\alpha+1)} & , \; \delta\omega \ll \gamma_{2,i} \\ T^{-3(\alpha+1)} & , \; \delta\omega \sim \gamma_{2,i} \\ T^{2(\alpha+1)} & , \; \delta\omega \gg \gamma_{2,i} \end{cases} \tag{S17}
\]

Finally, adopting the standard assumption for tunnelling TLS, \( P(\gamma_{1}) \sim 1/\gamma_{1} \), the frequency dependence of the fluctuation spectrum will be given by
\[
\int_0^{\gamma_{\text{Max}}} d\gamma_{1} P(\gamma_{1}) \frac{2\gamma_{1}}{\gamma_{1}^2 + \omega^2} = \frac{2 \arctan \left( \frac{\gamma_{\text{Max}}}{\omega} \right)}{\omega} \propto \begin{cases} 1 & , \; \omega < \gamma_{\text{Max}} \\ \frac{\gamma_{\text{Max}}}{\omega^2} & , \; \omega > \gamma_{\text{Max}} \end{cases} \tag{S18}
\]

Here the maximum relevant switching rate \( \gamma_{\text{Max}} \) is given by the time of a single \( T_1 \)-measurement. All faster fluctuations will be averaged out in the observations, leading to the behaviour \( \sim 1/\omega \) for \( \omega < \gamma_{\text{Max}} \). In the opposite case \( \omega \gg \gamma_{\text{Max}} \), i.e. when we observe the fluctuations on timescales that are short compared to \( 1/\gamma_{\text{Max}} \), the spectrum will show a \( 1/\omega^2 \) dependence.
The smallness of the switching rate $\gamma_1$ here is a simple fact deduced from the observations without any a priori model. For the very slow fluctuations observed in experiments, on timescales $\sim \min$, to the best of our knowledge no microscopic model exists. A possible candidate might be collective behaviour of large ensembles of TLS that form clusters [36, 44], but so far clear experimental confirmation of this effect is missing.

Alternatively one can assume a microscopic model for the TLS relaxation rate $\gamma_1$. For example for coupling to phonons, and omitting irrelevant prefactors, one arrives at \cite{7}

$$\gamma_1 \propto \Delta^2 E \coth \left( \frac{E}{2T} \right) \propto 2TE^2 \sin^2 \theta,$$  

(S19)

where in the second step we already assumed that the relevant energies of the switching TF are smaller than temperature, $E \ll k_B T$. Since the relaxation rate in this expression depends mainly on the TF mixing angle $\theta$, the restriction on small switching rates will be realised by confining $\theta$ to small values around zero, effectively restricting the value of the coupling strength between the relevant low-frequency TF and their phonon bath. Physically, Eq. (S19) implies that phonons do not induce the frequency dependence of the relaxation rate correlator $\mathcal{C}_\perp$ by confining $\theta$ to small values around zero, effectively setting $\Delta$ out of the expression. The frequency dependence of the switching TF \cite{5, 14}.

Effective inter-TLS interaction range

Here we give a rough calculation of the maximum inter-TLS distance which still allows noticeable interactions between them. We assume the TLS to be realised as microscopic electrical dipoles of uniform dipole size $d_i = e \times 10^{-10}$ m, where $e$ is the charge of a single electron \cite{5, 35}. Then, assuming parallel orientation of the two TLS and using the relation between dipole magnitude and coupling strength

$$\frac{g}{2} = \frac{1}{4\pi \epsilon_0 e_r} \left( d_{1,\perp} d_{2,\perp} - 3d_{1,\parallel} d_{2,\parallel} \right),$$  

(S23)

we can estimate the maximum distance to effect a minimum coupling strength of $g_{\text{Min}} = 1$ MHz (c.f. Fig. 3) as $r_{\max} \approx 110 \times 10^{-9}$ m. The volume in which TLS are interacting strongly enough is thus $V_{\text{TLS}} \sim 5.6 \times 10^{-21}$ m$^3$. Assuming an overall TLS density of $10^2/\mu\text{m}^3/\text{GHz}$ \cite{6, 12}, this leads to the effective frequency density of TLS in the interaction region of a single TLS of $\rho \sim 10^{-1}/\text{GHz}$. We note that the density obtained in Refs. 6 and 12 refers only to high-frequency TS, and a much higher density is expected for low-frequency TF \cite{5, 14}.

APPENDIX D: RABI-SPECTROSCOPY

When using non-frequency-tuneable qubits like single junction transmons, it is still possible to probe the form of the noise spectrum in close spectral vicinity of the qubit transition frequency. To this end one can make use of the fact that for a driven system, the frequencies of the noise spectrum relevant for decoherence will be shifted by the applied driving frequency. This effect can be thought of as a result of interaction of the dressing of the system states with drive photons, or similarly in the context of sideband transitions. The following derivation is based on the work in Ref. \cite{37}, more details can be found there.

For a two-level system driven with Rabi driving strength $\Omega_0$ at frequency $\omega_d$ we write the Hamiltonian

$$\hat{H} = \frac{1}{2} \omega_0 \sigma_z + \Omega_0 \cos \omega_d \sigma_x + \hat{H}_{\text{Sys-B}} + \hat{H}_B,$$  

(S24)

with the qubit level-splitting $\omega_0$, the bare Rabi frequency $\Omega_0$ and driving frequency $\omega_d$. For the system-bath coupling term, we take

$$\hat{H}_{\text{Sys-B}} = \frac{1}{2} b_{\perp} \sigma_z \hat{X}_\perp + \frac{1}{2} b_{\parallel} \sigma_x \hat{X}_\perp,$$  

(S25)

where the qubit level splitting is coupled to the bath variable $\hat{X}_\perp$ with coupling strength $b_{\parallel}$ and additionally the bath variable $\hat{X}_\perp$ might induce transitions between the qubit states due to its coupling with strength $b_{\perp}$. Here the bath coupling constants $b$ are assumed to be small.
with respect to the other energies in the problem, such that we can use perturbation theory in the strength of the system-bath coupling term $\hat{H}_{\text{Sys-B}}$. We will not specify the exact form of the bath Hamiltonian $\hat{H}_B$ but simply assume that is of a suitable form to induce Markovian decoherence, i.e. it possesses a very large number of degrees of freedom and equilibrates on a timescale that is much shorter than all system timescales. Moving into a rotating frame at the drive frequency, we then find the decoherence rates as

$$
\Gamma_{\varphi} = \sin^2 \beta \gamma_{\varphi} + \frac{1}{2} \cos^2 \beta \gamma_1 \frac{S_{X_L}(\omega_d)}{S_{X_L}(\omega_{10})},
$$

$$
\Gamma_{1} = \frac{1}{2} \cos^2 \beta \gamma_1 + \frac{1}{4} (1 - \sin^2 \beta) \gamma_1 \frac{S_{X_L}(\omega_d + \Omega)}{S_{X_L}(\omega_{10})},
$$

$$
\Gamma_{1} = \frac{1}{2} \cos^2 \beta \gamma_1 + \frac{1}{4} (1 + \sin^2 \beta) \gamma_1 \frac{S_{X_L}(\omega_d - \Omega)}{S_{X_L}(\omega_{10})},
$$

where we defined the rates

$$
\gamma_{\varphi} = \frac{1}{2} b^2 S_{X_L}(0), \quad \gamma_1 = \frac{1}{2} b^2 S_{X_L}(\omega_{10}),
$$

and we used the Rabi-frequency $\Omega = \sqrt{\Omega_0^2 + (\omega_{10} - \omega_d)^2}$. Here, we introduced the symmetrized correlation functions for the bath variables $\hat{X}$, defined as

$$
S_X(\omega) = \frac{1}{2} (C_X(\omega) + C_X(-\omega)).
$$

where $C_X(\omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t} \langle X(\tau) X(0) \rangle_{\text{th}}$ and the average $\langle \ldots \rangle_{\text{th}}$ is over the steady state of the bath. For an environment in thermal equilibrium, the unsymmetrized noise spectrum will follow a detailed balance relation, $C_X(\omega) = e^{-\beta \omega} C_X(\omega)$, with the inverse temperature $\beta = 1/k_B T$.

The two rates $\gamma_{\varphi}$ and $\gamma_1$ can be determined in independent experiments, measuring relaxation from decay of the qubit excited state and decay of Ramsey fringes. $\gamma_1$ on the other hand can potentially be estimated using $\gamma_{\varphi}$ and assuming a $1/f$-type dependence of the low-frequency noise spectrum. The angle $\beta$ in these expressions defines the relationship between drive strength $\Omega_0$ and detuning between drive frequency and qubit splitting and is defined as $\tan \beta = \Omega_0/(\omega_{10} - \omega_d)$.

In a Rabi experiment, the decay of the oscillations will be proportional to $e^{-t^2}$ with $\Gamma_2 = \Gamma_{\varphi} + \frac{1}{2} (\Gamma_1 + \Gamma_1)$ and thus measurements of the Rabi oscillations at different drive strengths can be used to infer the noise spectrum in the vicinity of the qubit transition frequency $\omega_{10}$.

### APPENDIX E: CALCULATIONS OF QUASIPARTICLE DENSITY

Experimentally it was found that the temperature dependence of the relaxation rates of superconducting qubits could be well explained when assuming interacting with thermally excited quasiparticles [38]. In this theory, the low temperature limit of the relaxation time $T_1$ stems from assuming a remaining density of non-equilibrium quasiparticles, the origin of which is not yet understood. Following the ideas developed in Ref. [38], we conjecture that a fluctuating quasiparticle density, i.e. due to recombination events or tunnelling to an outside reservoir, might lead to the observed fluctuations in relaxation time $T_1$. We calculate the required fractional changes in density as well as in terms of absolute number of quasiparticles for a given qubit design.

The following calculations follow closely the theory of Ref. [38], and we here only repeat their main steps for clarity. To derive the effects of the interaction between quasiparticles and superconducting circuits, we start with a low-energy Hamiltonian describing tunnelling of quasiparticles across a Josephson junction at phase difference $\varphi$

$$
\hat{H}_T = i t \sum_{n,m,\sigma} \sin \frac{\varphi}{2} a^R_{n,\sigma} a^L_{n,\sigma} + \text{h.c.}
$$

where $t$ is the tunnelling amplitude and the operators $a^L/R_{n,\sigma}$ destroy a quasiparticle in state $n$ with spin $\sigma$ in the left/right lead. Eq. (S29) is valid as long as the qubit energy $\omega$ as well as the characteristic energy $\delta E$ of the quasiparticles is much smaller than the superconducting gap $\Delta_{\text{sc}}$, a condition which is well satisfied in experiments. Starting from this equation, the authors in Ref. [38] derive the quasiparticle linear response function and thus the complex admittance of the Josephson junction due to quasiparticle tunnelling.

Using the golden rule, we write the transition rates between qubit states due to quasiparticle tunnelling as

$$
\Gamma_{i \rightarrow f} = \left| \langle i | \sin \frac{\varphi}{2} | f \rangle \right|^2 S_{qp}(\omega_{if})
$$

where $\omega_{if} = \omega_i - \omega_f$ is the energy splitting between qubit states $|i\rangle$ and $|f\rangle$ and $S_{qp}(\omega)$ is the quasiparticle spectral density, which can be calculated from the complex admittance via the fluctuation-dissipation theorem. For low temperature, $T \ll \Delta_{\text{sc}}$ and high frequencies $\omega_{if} \gg \delta E$, one finds

$$
S_{qp}(\omega) \approx x_{qp} \frac{8 E_j}{\pi} \sqrt{\frac{2\Delta_{\text{sc}}}{\omega}}
$$

with the junction’s Josephson energy $E_j$ and the fractional quasiparticle density normalized to the density of Cooper pairs $x_{qp} \approx n_{qp}/2\nu_0\Delta_{\text{sc}}$. Here $\nu_0$ is the density of states of electrons in the leads, which we assume to be the same on both sides.

For the case relevant to experiments, where a single junction 3D-transmon was used, the relaxation rate due to quasiparticles can then be calculated as

$$
\Gamma_{1 \rightarrow 0} = \frac{\omega_{10}^2 x_{qp}}{\omega_{10}^2 \pi} \sqrt{\frac{2\Delta_{\text{sc}}}{\omega_{10}}}
$$

(S32)
with the junction’s plasma frequency \( \omega_p = \sqrt{8E_J E_C} \) and its charging energy \( E_C \). Eq. (S32) directly relates a qubit’s relaxation rate to the density of quasiparticles.

From Eq. (S32) we can extract the fractional quasiparticle density \( x_{qp} \), with the value for the superconducting gap of thin-film aluminium [13]:

\[
\Delta \approx 200 \mu eV \approx 50 \text{ GHz} \approx 3.2 \times 10^{-21} \text{ J}.
\]  

(S33)

Then, for a relaxation time of \( T_1 = 100 \text{ ms} \), corresponding to \( \Gamma_1 = 10 \text{ kHz} \) (c.f. Fig. 2 in the main text) we find the canonical value of \( x_{qp} \approx 5 \times 10^{-7} \).

We want to use the relative quasiparticle density determined above to calculate the actual number of quasiparticles interacting with the qubit sample. For this we need the electron density of states at the Fermi edge for aluminum, which we take from literature as \( \nu_0 = 4.65 \times 10^{47} \text{ m}^{-3} \text{ J}^{-1} \) [45]. We thus find the quasiparticle volume density for the above used relaxation rate \( \Gamma_1 = 10 \text{ kHz} \) as

\[
n_{qp} = 2\nu_0 \Delta sc x_{qp} \approx 5 \times 10^{18} \text{ m}^{-3}.
\]  

(S34)

The 3D-transmon used in the experiments consists of two paddles of dimensions \( 350 \times 10^{-6} \text{ m} \times 700 \times 10^{-6} \text{ m} \times 120 \times 10^{-9} \text{ m}^3 \), with a total volume of \( V_{Al} \sim 3 \times 10^{-14} \text{ m}^3 \). We then find

\[
\delta N_{qp} = V_{Al} n_{qp} / 10 \approx 1.5 \times 10^4
\]  

(S35)

as the number of quasiparticles that, for the sample used, leads to a change in the relaxation rate of \( \delta \Gamma_1 = 1 \text{ kHz} \).