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Internal loss of superconducting resonators induced by interacting two level systems

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In a number of recent experiments with microwave high quality superconducting coplanar waveguide (CPW) resonators an anomalously weak power dependence of the quality factor has been observed. We argue that this observation implies that the monochromatic radiation does not saturate the Two Level Systems (TLS) located at the interface oxide surfaces of the resonator and suggests the importance of their interactions. We estimate the microwave loss due to *interacting* TLS and show that the interactions between TLS lead to a drift of their energies that result in a much slower, logarithmic dependence of their absorption on the radiation power in agreement with the data.

High quality superconducting CPW resonators are used in a number of diverse fields, ranging from astronomical photon detection [1, 2] to circuit quantum electrodynamics [3–6]. In these applications, the CPW resonator is operated in a regime of low temperature ($\sim$ 10mk) and low excitation power (single photon). The performance of these devices is directly related to the resonator quality factor, Q, defined by the photon decay rate, γ_{ph}, as $Q = \omega_0/\gamma_{ph}$, where ω_0 is the resonance frequency. γ_{ph} is given by the sum of the escape rate from the resonator and the rate of the intrinsic decay; the latter sets the limit on the performance. At low temperature and single photon regime, the intrinsic decay is usually attributed to the excitations of TLS located in the dielectrics (bulk and surfaces) surrounding the resonator [7]. This belief is strongly supported by the observation of temperature-dependent resonance frequency shift that closely agrees with the one predicted by the conventional theory of microwave absorption of TLS in glasses [8, 9]. According to this theory, one expects also that, as the power of the radiation applied to resonator is increased, TLS in the dielectrics get saturated, thereby limiting the maximal power that can be dissipated by photons. This results in a strong power dependence of the quality factor: $Q \propto \sqrt{P}$ above a critical power P_c. This power dependence is indeed observed in many resonators characterized by intrinsic loss tangent $\sim 10^{-3}$ at very low powers [10, 11]. However resonators characterized by lower intrinsic loss at low powers typically show much weaker power dependence [12–16].

In fact, careful fitting of the loss versus power to the empirical equation $Q \propto (1 + P/P_c)^{\varphi}$ gives $\varphi \sim 0.03-0.16$ for the resonators made of Nb and Al on sapphire and of Nb on undoped silicon [13]; similarly high-Q single-layer resonators of different geometries made of Al on sapphire substrate show $\varphi \sim 0.1 - 0.2$ [15]. Corrections of the conventional theory of TLS which include the geometric dependence of the applied electric field failed to reproduce the weak power dependence observed experimentally [12, 14–16].

The failure of the conventional theory of TLS to predict the power dependence of the quality factor for the high quality resonators is an indication of a serious gap in our understanding of TLS in amorphous insulators. In this Letter we argue that in high-Q superconducting CPW resonators the TLS located in the interface oxide surfaces are subject to stronger interactions than the TLS located in the bulk dielectrics. These TLS interactions lead to a drift of the TLS energies that results in a logarithmic dependence of their absorption on the radiation power, P, in agreement with the data. We begin by sketching the most important assumptions that lead to the $Q \propto \sqrt{P}$ prediction of the conventional theory of microwave absorption of TLS; then we discuss the implications of the much weaker power dependence reported experimentally in high-Q superconducting CPW resonators.

In the conventional theory, TLS are described by pseudo-spin operators, S, and are characterized by the uniform distribution of the energy difference, ε, between their ground and excited state. In the basis of the eigenstates the Hamiltonian has a simple form $H = \varepsilon S^z$. The ground and the first excited state of the TLS correspond to the quantum superposition of the states characterized by different atomic configurations. Each TLS is characterized by a dipole moment $\hat{\mathbf{p}} = \mathbf{p}(\sin\theta S^x + \cos\theta S^z)$, which is an operator with both diagonal and off-diagonal components. $\mathbf{p}$ denotes the difference between the dipole moments in the two different atomic configurations, its magnitude $p_0 = |\mathbf{p}|$ sets the scale of the dipole moment. θ describes the fact that the eigenstates of the dipole correspond to the superposition of its states in real space. Because many dipoles have exponentially small amplitude to tunnel between different positions in real space, the parameters θ and ε are assumed to have distribution $\mathcal{P}(\varepsilon, \theta) d\varepsilon d\theta \sim \nu/\theta d\varepsilon d\theta$ for small θ, where $\nu = 10^{20}/cm^3 eV$ is the typical density of states of TLS [17]. The interaction between different TLS is essentially of a dipole-dipole nature with an effective strength given by the dimensionless parameter $\lambda = \nu p_0^2/\epsilon$, here ϵ is the dielectric constant of the medium, it can also be viewed as coming from TLS coupling to elastic strain [18]. Straightforward analysis shows that the same parameter also controls the phonon mean free path at low temperatures [19]. The direct measurement gives values of $\lambda \approx 10^{-3}$ in bulk materials, so the interaction between TLS is usually assumed to be irrelevant.

The intrinsic microwave loss is due to the coupling of the electric dipole moment $\hat{\mathbf{p}}$ of the TLS to the applied electric field $\mathbf{E}\cos\omega t$ of the resonator. The resonator quality factor is related to the imaginary part of the dielectric function, $\epsilon(\omega)$, i.e. $Q^{-1} \propto |\Im\epsilon|$. At microwave frequencies and low temperatures, only the resonant contribution to the electric susceptibility tensor is relevant. One can compute the change in the dielectric function due to the resonant response of an ensemble of TLS by assuming that TLS are relaxed by phonons and using the Bloch equations that neglect the interaction between TLS [18]. In fact, one can get the power dependence of quality factor Q by following a more general, qualitative argument. In the steady state driven by sinusoidal electric field, the power dissipation density is given by the time average

$$\langle P_d \rangle = \left\langle \mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial \mathrm{t}} \right\rangle = \frac{\omega}{2} |\mathbf{E}|^2 |\Im\epsilon| \qquad (1)$$

where $\mathbf{D} = \epsilon\mathbf{E}$ is the displacement vector. In the absence of TLS-TLS interactions, each TLS can decay from its excited state only by emitting a phonon; the rate of this emission gives the decay rate, $\Gamma(\varepsilon)$, of its excited state. The maximal power that a given TLS can absorb from the photons in the resonator is $\varepsilon\Gamma$. At small applied powers, the TLS that absorb photons have energies close to the frequency, ω_0, of the photon field, i.e. $|\varepsilon - \omega_0| \lesssim \Gamma$. It is convenient to characterize the effect of the electric field on TLS by their Rabi frequency $\Omega = \frac{1}{2}\mathbf{p}\cdot\mathbf{E}\sin\theta$; for small $\Omega < \Gamma$ TLS at resonance are excited with probability $(\Omega/\Gamma)^2$. Since their density is $\nu\Gamma$, the total power dissipation density reads $\langle P_d \rangle = \nu\varepsilon\Omega^2/\Gamma$. Different is the situation at larger applied powers. There, even the TLS with energies further away from the frequency of the resonator, ω_0, get excited. For TLS exactly at resonance, the electric field $\mathbf{E}\cos\omega_0 t$ results in oscillations with frequency Ω. When $\Omega > \Gamma$, TLS with energies $|\varepsilon - \omega_0| \lesssim \Omega$ are excited. Since their density is $\nu\Omega$, the total dissipated power density reads $\langle P_d \rangle = \nu\varepsilon\Gamma\Omega$. By substituting it into Eq. 1, one recovers immediately the quality factor power dependence $Q \propto \sqrt{\mathrm{P}}$.

The generality of these qualitative arguments implies that, in the resonators where a much weaker power dependence of Q has been observed, the dipoles that absorb the radiation do not get saturated as the applied power P is increased. This is possible if the interactions between dipoles are not negligible and lead to energy diffusion.

The bulk of evidence indicates that the loss in high-Q superconducting CPW resonators is due to TLS located on the interface oxide surfaces (metal-air, metal-substrate or substrate-air) [10, 12–14, 16], even though it remains unclear which of the surfaces is more relevant[20]. The estimates show [21] that the concentration of TLS in these thin surfaces is higher than in the bulk and consequently the average interaction between TLS is larger, implying that one has to develop the theory of microwave absorption in *interacting* TLS in these resonators. In contrast, the resonators that show $Q \propto \sqrt{\mathrm{P}}$ and low intrinsic loss in single photon regime are made of Nb on SiO_2/Si [10] and AlO_x coated Nb on Si substrate [11] and thus contain a significant amount of bulk amorphous dielectrics (SiO_2 or AlO_x). It is natural to assume that the large intrinsic loss in these resonators is due to TLS located in the dielectric bulk which is described well by the conventional theory of independent TLS.

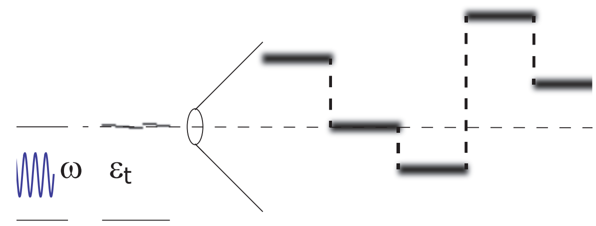

Figure 1: Drift of the energy of a given TLS in the field of a few fluctuators.

Developing the full theory of interacting TLS is a very difficult problem that was first discussed by Yu et al. [22] and still remains controversial [23–26]. Fortunately, as we shall see below, one does not need to solve the full theory of interacting TLS in order to estimate the internal loss of superconducting resonators due to *interacting* TLS. In our approach, we assume that the effective degrees of freedom that remain active in the amorphous oxides surrounding the superconductors are described by fluctuating dipoles: some of these dipoles are characterized by fast transitions between their states ($\varepsilon\sin\theta \sim \omega$) and relatively small decoherence rates, i.e. $\Gamma \ll \omega$, and are effectively coherent TLS; other dipoles are instead slow and characterized by decoherence times shorter than the typical time between the transitions, we shall call them fluctuators. Due to the interaction between fluctuators and TLS, the frequency of the TLS jumps when the fluctuators in its vicinity change its state. This translates into the fact that instead of staying constant the energy of a given TLS drifts with time (see Fig. 1). Notice that this phenomenological model is in a full agreement with experiments on charge noise performed in superconducting SET and qubits [27].

We now use this phenomenological model to estimate the internal loss of the CPW resonators. We begin with qualitative arguments. Due to the interaction with fluctuators, the TLS with energy level ε in resonance with the applied electric field stays effectively in resonance only for a short dwell time $\tau = \gamma^{-1}$, where γ is the combined relaxation rate of the fluctuators that affect it (see Fig.1). If the time τ is short, i.e. $\tau\Omega \ll 1$, the TLS gets into the excited state with probability $\mathcal{P}_e \sim (\Omega\tau)^2$ and then dissipates the energy away from the resonance. The average dissipation rate of this process is $\bar{\Gamma} = \gamma\mathcal{P}_e \propto \Omega^2/\gamma$. Only TLS with energy level located within energy γ from the resonator frequency ω contribute to this process: their density is $\nu\gamma$ and the total dissipated power density reads $\langle P_d\rangle = \nu\varepsilon\Omega^2 \int_{\Omega}^{\gamma_{max}} \mathcal{P}(\gamma)d\gamma$, where $\mathcal{P}(\gamma) = P_0/\gamma$ is the probability distribution of the fluctuator relaxation rates [28]. After integration, we obtain that $\langle P_d\rangle = \nu P_0 \varepsilon\Omega^2 \ln(\gamma_{max}/\Omega)$ and by inserting it into Eq.1 we find that in this case $Q \propto \ln(\Omega/\gamma_{max}) \propto \ln P$, i.e. there is only a slow, logarithmic dependence of the quality factor with the applied power P.

These qualitative arguments can be confirmed by more quantitative analytical computations. Before proceeding with the analytic derivation, we stress that the essential ingredient of the phenomenological model is the large value of the jumps in the TLS frequency, $\delta\varepsilon_t \gg \Gamma$ caused by fluctuators. This implies that the interaction between the high frequency dipole and the fluctuators in its vicinity is large: $V(r) = \frac{p_0^2}{\epsilon r^3} \gg \Gamma$. Because the typical distance between thermally activated TLS is $\bar{r} = \rho^{-1/3}$, with $\rho = \nu T$, the typical interaction is $V(\bar{r}) = \lambda T$. Comparing it with the relaxation rates of $\Gamma \sim 10^6 s^{-1}$ observed experimentally at frequencies $\omega \sim 10$ GHz (and expected theoretically for phonon emissions) we see that in typical experimental conditions $\delta\varepsilon \sim \Gamma$ for $\lambda \sim 10^{-3}$. The condition $\delta\varepsilon \gg \Gamma$, however, can be satisfied for lower frequencies or in materials with larger λ. The estimates of the dissipation per unit volume of surface oxides show that there their density of TLS is at least 10 times larger than in the bulk amorphous materials [21]. This translates into 10 times larger value of the parameter λ and a completely different physics of microwave dissipation described above.

We now give the details for the analytical derivation of the $Q \propto \sqrt{P}$ and its generalization for the model that takes into account energy drifts of TLS caused by fluctuators. The collection of TLS is characterized by the Hamiltonian $H_{int} = \sum_i \varepsilon_i S_i^z + \frac{1}{2}\sum_{i,j} V_{ab}(r_i - r_j)\mathbf{p}_a\mathbf{p}_b$. Here $V_{ab}(r)$ is the interaction between TLS that is due to their dipole moments or virtual phonon exchange. In either case, in the static limit, the interaction scales as $V(r) \sim 1/r^3$. At zero temperatures and for a small value of the parameter $\lambda \ll 1$, the Hamiltonian H_{int} gives a coherent dynamics of the individual TLS. In this limit, the levels at high frequencies are broadened by phonon emission [9] that leads to the relaxation rate $\Gamma_1 \propto \varepsilon^3$. At non-zero temperatures the levels are further broadened by dephasing caused by thermal phonons and other TLS [29, 30]. The combined effect of all these processes on a single TLS can be described by the Bloch equations: $\frac{d}{dt}\langle\mathbf{S}(t)\rangle = [\langle\mathbf{S}(t)\rangle \times \mathbf{B}(t)] - \mathbf{R}(t)$, where $\mathbf{R}(t) = (\Gamma_2\langle S_x\rangle, \Gamma_2\langle S_y\rangle, \Gamma_1(\langle S_z\rangle - m))$ is the relaxation matrix, $\mathbf{B}(t)$ is the total field acting on the TLS and $m = \dfrac{1}{2}\tanh[\varepsilon/2T]$ denotes the equilibrium population of the TLS levels and T is the temperature. In the conventional theory of TLS, this field has two components: $\mathbf{B} = \mathbf{B}^0 + \mathbf{B}^1$, with $\mathbf{B}^0 = (0, 0, \varepsilon)$ and $\mathbf{B}^1 = (\Omega, 0, \Omega')\cos\omega t$, where $\Omega' = \frac{1}{2}\mathbf{pE}\cos\theta$. The presence of the fluctuators results in the additional time dependence of the energy levels, $\varepsilon_t = \varepsilon + \xi(t)$. Here $\xi(t)$ denotes a multi-level telegraph noise signal characterized by switching rate γ.

The nonlinear Bloch equations are dramatically simplified in realistic conditions, because the feasible electric field acting on the TLS has very little effect on them away from the resonance. Formally, in the stationary solution of the Bloch equations driven with $\mathbf{E}\cos\omega t$, all spin components oscillate with frequencies $n\omega$: $\mathbf{S} = \sum_n \mathbf{S}_n \exp(-in\omega t)$ with $\mathbf{S}_n = \mathbf{S}^*_{-n}$. Relaxation to the stationary solution is determined by the decay rate $\Gamma_1 \ll \Gamma_2 \ll \omega$ which sets the longest time scale in the problem. It can be described by allowing slow time dependence of $\mathbf{S}_n$ components. Finally, introducing the operators $S^\pm = S^x \pm iS^y$ and leaving only the leading resonant terms the Bloch equations reduce to:

$$i\frac{dS_1^+}{dt} = \Omega S_0^z - (\omega - \varepsilon_t + i\Gamma_2)S_1^+ \tag{2}$$

$$\frac{dS_0^z}{dt} = \Omega\Im S_1^+ - \Gamma_1(S_z^0 - m) \tag{3}$$

The macroscopic response is given by the average polarization, $\mathbf{P}_\omega$ of TLS at frequency ω:

$$\mathbf{P}_\omega = \frac{1}{2}\left\langle \mathbf{p}\sin\theta S_1^+\right\rangle \tag{4}$$

where the average is taken over the distribution of TLS and their dipole moments. The quality factor is directly related to $\mathbf{P}_\omega$: $Q^{-1} \propto |\Im\mathbf{P}_\omega|/\mathbf{E}$.

If the jumps $\xi(t)$ induced by the fluctuators are small, i.e. $\xi < \Gamma_2$, the stationary solution of Eqs.(2,3) gives the spin component:

$$S_1^+ = \frac{\Omega(\omega - \varepsilon - i\Gamma_2)m}{(\omega - \varepsilon)^2 + \Gamma_2^2 + \Omega^2\Gamma_2\Gamma_1^{-1}}. \tag{5}$$

At small fields, one can neglect the last term in the denominator of Eq.(5). By substituting Eq.(5) into Eq.(4) and then averaging over the distribution of TLS, one gets:

$$|\Im\mathbf{P}_\omega| = \frac{\pi m}{6}\left\langle \sin^2\theta\right\rangle \lambda\mathbf{E} \tag{6}$$

and consequently a Q factor that does not depend on the field strength $\mathbf{E}$.

At large fields ($\Omega^2 > \Gamma_1\Gamma_2$), the third term in denominator of Eq.(5) dominates the second and this results in a rapid decrease of the response. By averaging over the distribution of TLS, one gets:

$$|\Im \mathbf{P}_\omega| = \frac{\pi m}{6}\left\langle \sin^2\theta \frac{\sqrt{\Gamma_1\Gamma_2}}{\Omega}\right\rangle \lambda \mathbf{E} \quad (7)$$

which translates into the usual $Q \sim \sqrt{P}$ dependence.

In the opposite case of large jumps $\xi \gg \Gamma_2$ one should solve the full dynamical Eqs.(2,3) with noise $\xi(t)$. The details of the solution depend on the relation between the relaxation rates Γ_1, Γ_2 and the jump rate γ. We discuss first the simplest case of large γ in which a given TLS spends most of its time away from the resonance, so that when it moves back into the resonance its magnetization S_0^z and S_1^+ have their equilibrium values: $S_0^z = m$ and $S_1^+ = 0$. In this case the only quantity that controls the response is the rate, γ, of incoming and outgoing jumps into the resonance. The stochastic nature of this process implies that, in order to find the average value of S_1^+ that determines the response to electric field, we have to solve the equation for the probability, $\varrho(\mathbf{s})$, to find the resonant TLS characterized by the three dimensional vector $\mathbf{s} = (x, y, z)$, where x and y are the real and the imaginary parts of S_1^+ and $z = S_0^z$. This probability obeys the evolution equation:

$$\frac{\partial \varrho}{\partial t} + \frac{d}{d\mathbf{s}}\left(\frac{d\mathbf{s}}{dt}\varrho\right) = \gamma\left[\delta(z-m)\delta(x)\delta(y) - \varrho\right]$$

where $d\mathbf{s}/dt$ is given by Eqs.(2,3) with constant ε. The equation is further simplified in the limit of large $\gamma \gtrsim \Omega \gg \Gamma_1, \Gamma_2$ where the analytic solution gives (see Supplementary material), after averaging over the distributions of ε and γ, $\mathcal{P}(\varepsilon, \gamma) = \nu P_0/\gamma$:

$$|\Im \mathbf{P}_\omega| = \frac{\pi m}{6}\left\langle \sin^2\theta\right\rangle \lambda P_0 \ln\left(\frac{\gamma_{max}}{\Omega}\right) \mathbf{E} \quad (8)$$

which results in a weak, logarithmic dependence of the quality factor Q on the strength of the electric field.

A similar logarithmic dependence occurs in the cases when the dephasing rate is larger than the jump rates as well. In this case, one can neglect the time derivative terms in Eq. (2) and express S_1^+ through S_0^z and get the general evolution equation:

$$\frac{\partial \varrho_k}{\partial t} + \frac{\partial}{\partial z}\left[\left(\Gamma_1(z-m) - \Xi_k z\right)\varrho_k\right] = \gamma_{kn}\varrho_n \quad (9)$$

where $\varrho_k(z,t)$ is the probability for a given TLS to have value $S_0^z = z$ while subjected to the effective driving field $\Xi_k = \Omega^2\Gamma_2/[(\omega-\varepsilon_k)^2 + \Gamma_2^2]$ in the state k of the fluctuators. The matrix γ_{kl} is the matrix of the transition rates between the states of the fluctuators. Similarly to the case of the small dephasing rate the saturation of TLS does not happen if they stay mostly away from the resonance, so that the probability to find a *given* TLS in resonance $n < \Gamma_1/\gamma$ (see Supplementary material). Because the number of states of classical fluctuators grows exponentially with their number this condition is satisfied even for a moderate number of fluctuators that affect a given TLS. In this situation the dissipation is dominated by TLS with $\gamma > \Xi \sim \Omega^2/\Gamma_2$:

$$|\Im \mathbf{P}_\omega| = \frac{\pi m}{6}\left\langle \sin^2\theta\right\rangle \lambda P_0 \ln\left(\frac{\gamma_{max}\Gamma_2}{\Omega^2}\right) \mathbf{E} \quad (10)$$

Both Eqs(8,10) lead to the weak, logarithmic dependence of Q at large electric fields ($\Omega^2 > \Gamma_2\Gamma_1$), while at smaller fields, the imaginary part of the average polarization is given by Eq.(6) and Q is constant.

As a final remark let us notice that in this model the dimensionless coupling between TLS remains small, $\lambda \ll 1$. This implies that thermodynamics properties and the real part of the dielectric constant are not affected by the interaction. Therefore we expect the same temperature-dependent resonance frequency shift as predicted by the conventional theory of TLS in agreement with experiments [8, 9].

In conclusion, we have shown that the resonance absorption from TLS is strongly affected by their interaction with classical fluctuators; this does not change the absorption at low powers but changes the square root dependence of the absorption into the logarithmic one at high powers. The important condition is that each fluctuator should move the TLS frequency more than its width due to the relaxation rate. This translates into a higher (~ 10 times larger) concentration of TLS than the typical density in amorphous bulk materials.. We interpret the results of recent experiments displaying slow power dependence of the quality factor in high-Q superconducting CPW resonators as the evidence for a large concentration of TLS located in the interface oxide surfaces of the resonator. Very likely it implies that these TLS have a different nature, e.g. localized electron states at the superconductor oxide boundary.

Note added: recent experiment directly observed the energy drift of TLS which is the basis of our model [31]

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