Theory of nonlinear microwave absorption by interacting two-level systems

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The microwave absorption and noise caused by quantum two-level systems (TLSs) dramatically suppress the coherence in Josephson junction qubits that are promising candidates for quantum information applications. It is a challenge to understand microwave absorption by TLSs because of the spectral diffusion resulting from fluctuations in their resonant frequencies induced by their long-range interactions. Here we treat the spectral diffusion explicitly using the generalized master equation formalism. The proposed theory predicts that the linear absorption regime holds while a TLS Rabi frequency is smaller than their phase decoherence rate. At higher external fields, a nonlinear absorption regime is found with the loss tangent inversely proportional to the intensity of the field. The theory can be generalized to acoustic absorption and lower dimensions realized in superconducting qubits.

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I. INTRODUCTION

Quantum two-level systems (TLSs), commonly represented by atoms or groups of atoms tunneling between two states (see Fig. 1, Refs. [1,2]), are ubiquitous in amorphous solids. TLSs restrict the performance of modern nanodevices, including superconducting qubits [3] and quantum dots [4] for quantum computing, kinetic inductance photon detectors for astronomy [5], and nanomechanical resonators [6]. TLSs are commonly found in Josephson junction barriers, wiring crossovers [7–9], and even on the surfaces of the resonators with coplanar superconducting electrodes on crystalline substrates [5,10]. TLSs reduce the coherence in qubits absorbing microwaves in the frequency domain of their oscillations [3,9] and producing a noise in qubit resonant energies [10–15].

Both the microwave absorption and the noise induced by TLSs are dramatically sensitive to their interactions [10,13–18]. While the $1/f$ noise is reasonably interpreted within the interacting TLS model [13,14], the problem of nonlinear microwave absorption has not been resolved yet, in spite of numerous efforts [16,17,19]. Here we propose a solution to this long-standing problem integrating the earlier developed rate equation model for TLS density matrix time evolution [20,21] briefly introduced in Sec. II with the master equation formalism to account for the spectral diffusion [22–24] introduced in Sec. III B. The rigorous solution of the master equation is obtained in Sec. IV for the low-temperature case where the thermal energy is smaller than the microwave energy quantum

$$k_B T < \hbar\omega. \tag{1}$$

Based on this solution, the analytical expression for the loss tangent is derived in Sec. IV B. The opposite high-temperature regime is discussed qualitatively in Sec. IV C. The obtained solution results in the novel behaviors of nonlinear absorption in qualitative agreement with earlier expectations [23,25]. The predictions of theory are partially consistent with the experimental observations in amorphous solids [16], and they deviate from the observed weakening of intensity dependence in Josephson junction resonators and qubits. The possible reasons for deviations are discussed in Sec. IV D. The paper ends with a brief conclusion in Sec. V, including the summary of loss tangent nonlinear behaviors at different temperatures given in Table I.

II. RATE EQUATION FORMALISM AND TLS LOSS TANGENT

The nonlinear absorption of acoustic and electromagnetic waves by TLSs in amorphous solids was discovered experimentally almost half a century ago [16,26–28]. The results have been interpreted using the rate equation formalism applied to the TLS density matrix [16,21] in the Bloch vector representation [24] defined as $\sigma^x = \rho_{ge} + \rho_{eg}, \sigma^y = (\rho_{ge} - \rho_{eg})/i$, $\sigma^z = \rho_{gg} - \rho_{ee}$, where indices g and e stand for the TLS ground and excited states, respectively. The time evolution of the density matrix is determined by TLS frequency detuning from resonance $D = E/\hbar - \omega$, Rabi frequency $\Omega_R = (\Delta_0/E)pF_{\rm AC}\cos(\theta)$ (see Fig. 1), and relaxation and decoherence times T_1 and T_2. In the rotating frame approximation, relevant in the regime of interest, Eq. (1), corresponding to the resonant absorption [20,21,29,30], one has

$$\frac{d\sigma^x}{dt} = D\sigma^y - \frac{\sigma^x}{T_2}, \quad \frac{d\sigma^y}{dt} = -D\sigma^x - \frac{\sigma^y}{T_2} + \Omega_R\sigma^z,$$
$$\frac{d\sigma^z}{dt} = -\Omega_R\sigma^y - \frac{\sigma^z - \sigma^z_{\rm eq}}{T_1}, \quad \sigma^z_{\rm eq} = \tanh\left(\frac{E}{2k_BT}\right). \tag{2}$$

The equilibrium population difference $\sigma^z_{\rm eq}$ for resonant TLSs ($E \approx \hbar\omega$) can be set equal to unity in the case of interest, $k_BT \ll \hbar\omega$. The rotating frames approximation is applicable at sufficiently small external field, $\Omega_R \ll \omega$, which is satisfied in the vast majority of measurements of TLS resonant absorption at microwave frequencies.

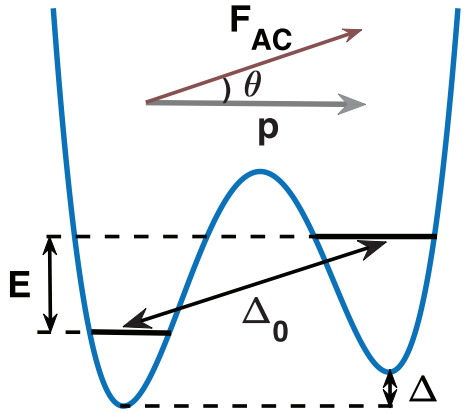

FIG. 1. A tunneling two-level system having an energy splitting E and a dipole moment $\mathbf{p}$ interacting with an external field $\mathbf{F}_{\rm AC}\cos(\omega t)$. The energy $E=\sqrt{\Delta^2+\Delta_0^2}$ is defined by an asymmetry Δ and a tunneling amplitude Δ_0 [1,2].

The TLS relaxation and decoherence times originated from the TLS interaction with phonons are defined as [14,20,31,32]

$$\frac{1}{T_1}=\alpha\frac{\Delta_0^2 E}{k_B^3}\coth\left(\frac{E}{2k_BT}\right)\approx\frac{x^2}{T_{10}},\quad T_2=2T_1, \tag{3}$$

where T_{10} stands for the minimum relaxation time and we introduced the dimensionless parameter $x=\Delta_0/E$ to describe TLS tunneling coupling. TLSs obey the universal distribution $P(E,x)=P_0/(x\sqrt{1-x^2})$ (see Fig. 1 and Refs. [1,2,20,33]). The minimum decoherence time is given by $T_{20}=2T_{10}$ and the constant $\alpha\sim 3\times 10^7\ \mathrm{s^{-1}\,K^{-3}}$ [31] is determined by the TLS-phonon interaction.

The microwave absorption is usually characterized by the TLS loss tangent defined in terms of the integrated reactive response $\Omega_R\sigma^y(D)$. This response is determined by the stationary solution of Eq. (2). For the sake of simplicity, we assume all TLS dipole moments have identical absolute values, p, as argued in Refs. [3,31]. The Rabi frequency can then be expressed in terms of the maximum Rabi frequency $\Omega_{R0}=pF_{\rm AC}/\hbar$ as $\Omega_R=xy\Omega_{R0}$, where $y=\cos(\theta)$ and it is uniformly distributed between -1 and 1. The loss tangent is defined as [21,31]

$$\tan(\delta)=\frac{4\pi P_0\hbar^2}{2\epsilon' F_{\rm AC}^2}\int_{-1}^{1}dy\int_0^1 dx\frac{xy\Omega_{R0}f^y(0,x,y)}{x\sqrt{1-x^2}},$$
$$f^a(q,x,y)=\int_{-\infty}^{\infty}\sigma^a(D,x,y)e^{iqD}dD,\quad a=x,y,z, \tag{4}$$

where ϵ' is a static dielectric constant of the material. The Fourier transforms $f^a(q,x,y)$ will be used in future analysis.

If the TLS Bloch vector obeys Eq. (2), then the Fourier transform of its y component contributing to the loss tangent can be evaluated as (cf. Refs. [16,21])

$$f^y(0)=\frac{\pi\Omega_R\tanh\left(\frac{\hbar\omega}{2k_BT}\right)}{\sqrt{1+\Omega_R^2T_1T_2}}. \tag{5}$$

This steady-state solution of Eq. (2) substituted into Eq. (4) determines a TLS loss tangent in the approximate form [3,5,16,31,34]

$$\tan(\delta)\approx\frac{\tan(\delta_0)}{\sqrt{1+\frac{32}{9\pi^2}\Omega_{R0}^2T_{20}^2}},$$
$$\tan(\delta_0)=\frac{4\pi^2P_0p^2\tanh\left(\frac{\hbar\omega}{2k_BT}\right)}{3\epsilon'}, \tag{6}$$

where $\tan(\delta_0)$ expresses the loss tangent in the linear-response regime. In the analysis below, we set the hypertangent factor $\tanh(\frac{\hbar\omega}{2k_BT})$ to unity since the consideration is limited to low temperatures Eq. (8).

III. MASTER EQUATION FORMALISM FOR SPECTRAL DIFFUSION

A. Spectral diffusion

The *spectral diffusion* that is the target of the present work strongly affects microwave absorption by TLSs leading to a TLS phase decoherence. It is induced by TLS interactions with neighboring "thermal" TLSs having energies and tunneling amplitudes comparable to the thermal energy. They switch back and forth between their ground and excited states with the quasiperiod $T_{1T}=1/(\alpha T^3)$, Eq. (3), and modify the energy and detuning of the given TLS. One can describe the spectral diffusion in terms of the distribution function $W(D,t|D',t')$ for possible detunings D at the time t provided that $D(t')=D'$. In

TABLE I. Summary of intensity dependencies of the TLS loss tangent at different temperatures below 1 K where the TLS model is applicable. The intermediate intensity regime of significant spectral diffusion is not available at low temperatures where the relaxation is faster than the phase decoherence as indicated by "n.a." in a corresponding cell of the table. TLS parameters used within the table are introduced in Eqs. (3) and (7) while the results are given in Sec. IV B. The restrictions for the maximum Rabi frequency and the temperature are introduced to avoid the emergence of relaxational absorption (see the end of Sec. IV C) and keep the rotating frame approximation valid.

			$\frac{\tan(\delta)}{\tan(\delta_0)}$		
	Ω_{c1}	Ω_{c2}	$\Omega_{R0}<\Omega_{c1}$	$\Omega_{c1}<\Omega_{R0}<\Omega_{c2}$	$\Omega_{c2}<\Omega_{R0}<\omega\min(1,(\frac{\hbar\omega}{k_BT})^3)$
$T<(\frac{12\hbar^7\alpha\omega^6}{\pi^6\chi k_B^7})^{1/4}$	$\frac{1.67}{T_{20}}$	$\frac{1.67}{T_{20}}$	1	n.a.	$\frac{1.67}{\Omega_{R0}T_{20}}$
$(\frac{12\hbar^7\alpha\omega^6}{\pi^6\chi k_B^7})^{1/4}<T<\frac{\hbar\omega}{k_B}$	$\frac{3.3}{T_{\rm sd}\ln(\frac{T_{20}}{T_{\rm sd}})}$	$\frac{0.6T_{20}}{T_{\rm sd}^2}\ln[3\ln(\frac{T_{20}}{T_{\rm sd}})]$	1	$\frac{\ln[1+3\ln(\Omega_{R0}T_{\rm sd})]}{\Omega_{R0}^2T_{\rm sd}^2\ln(\frac{T_{20}}{\Omega_{R0}T_{\rm sd}})}$	$\frac{1.67}{\Omega_{R0}T_{20}}$
$\frac{\hbar\omega}{k_B}<T<\min(1\ \mathrm{K},10\hbar\omega)$	$\frac{1}{T_{\rm sd}}\sqrt{\frac{T_{1T}}{T_{10}}}$	$\frac{T_{1T}}{T_{\rm sd}^2}$	1	$\frac{T_{1T}^2}{\Omega_{R0}^2T_{\rm sd}^4}$	$\frac{1.67}{\Omega_{R0}T_{20}}$

a three-dimensional system with TLS-TLS interaction decreasing with the distance as $1/R^3$, this function has a Lorentzian shape [23,35,36]. In a short-time limit $|t-t'| < T_{10} \ll T_{1T}$ [see Eqs. (1) and (3)], the width w of the Lorentzian distribution, $\Phi(D-D',t-t') = W(D,t|D',t')$, for the change in detuning, $D-D'$, can be expressed as [14,35,36]

$$w = k_2^2|t-t'|, \quad k_2^2 = \frac{\sqrt{1-x^2}}{T_{\rm sd}^2}, \quad \frac{1}{T_{\rm sd}^2} = \frac{\pi^6}{24}\frac{\chi k_B T}{\hbar T_{1T}}, \quad (7)$$

where the universal dimensionless parameter $\chi \sim 10^{-3}$ is the product of TLS density P_0 and the average absolute value of TLS-TLS interaction constant U_0, $\langle|U(r)|\rangle = U_0/r^3$ [14,32,36,37]. The rate $1/T_{\rm sd}$ characterizes the TLS phase decoherence observable in echo experiments. Spectral diffusion affects the nonlinear absorption if this rate exceeds the TLS relaxation rate $1/T_{10}$ defined in Eq. (3). For a typical microwave field frequency $\omega/(2\pi) \approx 5$ GHz, the decoherence stimulated by the spectral diffusion goes faster than that due to the relaxation at reasonably high temperatures $T > T_l$, where the temperature T_l is given by

$$T_l \approx \left(\frac{12\hbar^7\alpha\omega^6}{\pi^6\chi k_B^7}\right)^{1/4} \approx 30\ \text{mK}. \quad (8)$$

Upper [Eq. (1)] and lower ($T_l < T$) constraints on the temperature can be satisfied simultaneously at reasonably small resonant frequencies $\hbar\omega/k_B < 10$ K. Our consideration is limited to sufficiently small frequencies and temperatures $T, \hbar\omega/k_B < 1$ K where the tunneling model [1,2] is valid, which justifies the relevance of the consideration of the temperature domain restricted by Eqs. (1) and (8).

To describe the loss tangent behavior within the temperature domain $T > T_l$, it is natural to attempt to use Eq. (2) with the modified decoherence rate

$$1/T_{2*} = 1/T_{20} + 1/T_{\rm sd}. \quad (9)$$

This modifies Eq. (6) for the TLS loss tangent as [16,21,29]

$$\tan(\delta) \approx \frac{\tan(\delta_0)}{\sqrt{1+\frac{16}{9\pi^2}\Omega_{R0}^2 T_{2*}T_{10}}}. \quad (10)$$

The accurate treatment of the spectral diffusion described below within the framework of the master equation formalism predicts that the loss tangent behavior is qualitatively different from Eq. (10).

B. Master equations for TLS Bloch vector

The time evolution of the Bloch vector components $d\sigma^a = \sigma^a(D,t+dt) - \sigma^a(D,t)$ can be separated into two parts. The first part described by Eq. (2) includes the quantum-mechanical evolution and relaxation [16,20,21]. The second part accounts for the change in detuning due to spectral diffusion that can be treated classically [17,23,36]. The change of detuning during the infinitesimal time dt is determined by the conventional Lorentzian probability function $\Phi(D-D',dt)$ introduced above. The evolution of the TLS density matrix in the course of the spectral diffusion can be described by the master equation as suggested by Laikhtman [23] (see also the textbook [22]),

$$d\boldsymbol{\sigma}_{\rm sd}(D,t) = \int_{-\infty}^{\infty} dD'\Phi(D-D',dt)[\boldsymbol{\sigma}(D',t) - \boldsymbol{\sigma}(D,t)]. \quad (11)$$

This equation is valid if there is no correlation between the TLS evolution before and after the time t [22]. Phonon stimulated transitions of resonant TLSs to the ground state occur with the quasiperiod T_{10} and each transition erases memory of the previous TLS dynamic. Since the time between two transitions of thermal TLS $T_{1T} \approx (\alpha T^3)^{-1}$ is much longer than this quasiperiod [see Eqs. (1) and (3)], each neighboring thermal TLS contributes no more than once to the shift of the resonant TLS energy during the time T_{10}. Consequently, there is no correlation of energy shifts during the equilibration time T_{10}, which justifies Eq. (11).

Using the Fourier transform representation of the TLS density matrix, Eq. (4), and the width of the Lorentzian distribution defined in Eq. (7), one can express the time evolution in Eq. (11) as

$$d\mathbf{f}_{\rm sd} = (e^{-k_2^2 dt|q|} - 1)\mathbf{f} = -k_2^2|q|dt\mathbf{f}. \quad (12)$$

The complete set of evolution equations for the density matrix Fourier transforms [see Eqs. (6)] can be obtained adding Eq. (12) and the Fourier transforms of Eq. (2). Then we get

$$\begin{aligned}
\frac{df^x}{dt} &= -|q|k_2^2 f^x - i\frac{df^y}{dq} - \frac{f^x}{2T_1} = 0,\\
\frac{df^y}{dt} &= -|q|k_2^2 f^y + i\frac{df^x}{dq} - \frac{f^y}{2T_1} + \Omega_R f^z = 0,\\
\frac{df^z}{dt} &= -|q|k_2^2 f^z - \Omega_R f^y - \frac{f^z}{T_1} + \frac{2\pi\delta(q)}{T_1} = 0,
\end{aligned} \quad (13)$$

where time derivatives should be set to zero since we are interested in the stationary regime [20]. All Fourier transforms $f^a(q)$ $(a = x,y,z)$ should approach 0 for $q \to \pm\infty$, which defines the boundary conditions to Eq. (13). To evaluate the loss tangent, we need to find the Fourier transform $f^y(0)$ for $q = 0$ in accordance with Eq. (6).

Here and in earlier work [23], the rotating frames approximation assumes the instantaneous adiabatic basis for the Bloch vector following the spectral diffusion of the detuning. This assumption seems to be well justified since a single TLS resonant frequency ω_0 is much larger compared to a characteristic rate of the spectral diffusion, however the collective TLS transitions can be stimulated by their interactions in the course of spectral diffusion [30,38]. This collective dynamics becomes important at very low temperature $T \sim 10$ mK [32], which is outside of the scope of the present work.

IV. NONLINEAR ABSORPTION IN THE PRESENCE OF SPECTRAL DIFFUSION: RESULTS AND DISCUSSION

A. Solution of the master equation for individual TLSs

Further analysis of Eq. (13) is straightforward but tedious, and its details can be found in the Supplemental Material, Sec. I [39]. Using the first and third equations, one can express f^x, f^z, and $\frac{df^x}{dq}$ in terms of f^y and obtain the second-order differential equation for $f^y(q)$ containing the δ-function term $\delta(q)$ as a

nonhomogeneous part. It is convenient to introduce the new variable $v = (A + k_2|q|)^2$, where $A = 1/(k_2 T_2)$. Using the substitution $f^y = e^{-\frac{v-A^2}{2}} F(v)$, we get the equation for $F(v)$ in a form similar to the hypergeometric differential equation (see Refs. [40,41]; remember that $v > A^2$)

$$v\frac{d^2F}{dv^2} - v\frac{dF}{dv} - F\left[\frac{\Omega_R^2\eta}{4k_2^2}\right] = 0, \quad \eta(v) = \frac{1}{1+\frac{A}{\sqrt{v}}}. \quad (14)$$

To account for the δ-function term at $v = A^2$ [$q = 0$, see Eq. (13)] we introduce the boundary conditions at $v = A^2$ for the first derivative of the function F in the form [39]

$$\frac{dF(A^2)}{dv} - \frac{1}{2}F(A^2) = -\frac{\pi\Omega_R}{2}, \quad (15)$$

while the second boundary condition is $F(\infty) = 0$. The parameter of interest $f^y(0)$, that determines the loss tangent in Eq. (4), is equal to $F(A^2)$.

Equations (14) and (15) represent the most significant results of the present work. They have been solved numerically for this problem and can be extended to other problems of interest. The approximate analytical solution for the TLS loss tangent is derived below in the case of a significant spectral diffusion, $A \ll 1$, and compared to the numerical solution. This solution is a subject for comparison to the experimental data and it helps to understand the nonlinear absorption qualitatively.

If the parameter η in Eq. (14) can be set to constant, then the solution satisfying the zero boundary condition at infinity is given by the confluent hypergeometric function of the second kind, $F(v) = cU(\eta(\Omega_R/k_2)^2/4,0,v)$ [41,42], with the constant c defined by the boundary condition in Eq. (15). Comparing the first term in Eq. (14) with the second and third terms, one can see that the solution of Eq. (14) should change remarkably compared to its maximum $F(A^2)$ ($q = 0$) at $v \sim v_* = \min(1, k_2^2/\Omega_R^2)$. Assuming that v_* determines the typical value of the parameter η, one can expect that $\eta \approx 1$ for $v_* \gg A^2$, while in the opposite case $\eta \approx 1/2$. This is confirmed by the comparison of analytical and numerical solutions [39].

In either case, one can evaluate the parameter of interest $f^y(0) = F(A^2)$ [see Eq. (4)] using the boundary condition Eq. (15) and the identity $dU(a,b,v)/dv = -aU(a+1,b+1,v)$ [41] as

$$f^y(0) = \frac{\pi\Omega_R}{1+\frac{\eta B^2}{2}\frac{U(\eta B^2/4,0,A^2)}{U(1+\eta B^2/4,1,A^2)}}, \quad B = \frac{\Omega_R}{k_2}. \quad (16)$$

Consider the case $v_* > A^2$ ($\Omega_R < k_2^2 T_1$, and we set $\eta \approx 1$). In this case, one can use the asymptotic behaviors of confluent hypergeometric functions $U(a,0,v) \approx \Gamma(1+a)^{-1}$, $U(1+a,1,v) \approx [\ln(1/v) - 2\gamma - \psi(1+a)]\Gamma(1+a)^{-1}$, and approximate the digamma function as $\psi(1+a) = \psi(1+B^2/4) \approx 2\ln(B/2 + e^{-\gamma/2})$, where $\gamma \approx 0.5772$ is the Euler constant. Then Eq. (16) can be represented as

$$f^y(0) = \frac{\pi\Omega_R}{1+\frac{\Omega_R^2}{k_2^2}\ln\left(\frac{2e^{-\gamma}k_2^2T_2}{2e^{-\frac{\gamma}{2}}k_2+\Omega_R}\right)}. \quad (17)$$

This solution is in excellent agreement with the exact numerical solution if the argument of the logarithm exceeds 2 [39].

In the opposite limit of a very large field $B > 1/A$ ($\Omega_R > k_2^2 T_1$, and we set $\eta \approx 1/2$), one can approximate Eq. (16) as [39]

$$f^y(0) \approx \frac{\pi\Omega_R}{\sqrt{1+\frac{B^2}{2A^2}}}. \quad (18)$$

This result is identical to the stationary solution of Eq. (2) or Eq. (13) at $k_2 = 0$ as given by Eq. (5). It does not depend on the spectral diffusion. The spectral diffusion is not significant in this regime because the typical detunings of resonant TLSs contributing to absorption, $D \sim \Omega_R$ [13], exceed a TLS frequency shift $D_{\rm sd} \sim k_2^2 T_1$ due to the spectral diffusion occurring during the time T_1 separating two resonant TLS relaxation events [see Eq. (7)]. Substituting Eq. (18) into the loss tangent definition, Eq. (4), and performing the integration, one obtains the earlier established behavior of the loss tangent Eq. (6) [20,33].

The solution of time-dependent master equations has been obtained in Ref. [23] ignoring relaxation terms (i.e., setting $T_1 = T_2 = \infty$) for a finite pulse duration. Although it has a certain similarity with the present work, it leads to a full suppression of absorption in the infinite duration time limit that is a consequence of the lack of dissipation needed for the correct description of absorption in a steady-state regime.

B. Evaluation of the loss tangent

The loss tangent is defined by the integral in Eq. (4) over different tunneling amplitudes and dipole moment orientations of contributing two-level systems. It has been evaluated using the exact numerical solution of Eq. (14) as shown in Fig. 2 by solid lines for several ratios of spectral diffusion and relaxation rates ($T_{20}/T_{\rm sd}$). The predicted behaviors differ from that for the loss tangent in the absence of spectral diffusion, Eq. (6), as shown in Fig. 2 by dashed lines and from the "corrected" Eq. (6) with the modified decoherence time T_{2*} ($1/T_{2*} = 1/T_{20} + 1/T_{\rm sd}$) as shown in Fig. 2 by the dotted line.

Below, we derive the analytical interpolation for the loss tangent suitable for the analysis of experimental data in the case of a significant spectral diffusion $T_{\rm sd} < T_{10}$; otherwise, the loss tangent is always determined by Eq. (6). In the case of an intermediate Rabi frequency, $1/T_{\rm sd} < \Omega_{R0} < T_{10}/T_{\rm sd}^2$, one can use the approximate solution, Eq. (17), substituted into the integral in Eq. (4). Then, one can evaluate this integral with logarithmic accuracy as $\tan(\delta_0)\ln(1+3l)/(\Omega_{R0}T_{\rm sd})^2$ with the logarithmic factor $l = \ln(\Omega_{R0}T_{\rm sd})/\ln(D_{\rm sd}/\Omega_{R0})$ [39]. In the opposite case of large Rabi frequency, $T_{10}/T_{\rm sd}^2 < \Omega_{R0}$, the loss tangent is determined by Eq. (6). One can combine these two asymptotic behaviors within the single interpolation formula

$$\tan(\delta_{\rm sd}) = \frac{\tan(\delta_0)}{1+\frac{\Omega_{R0}^2T_{\rm sd}^2}{\frac{3\pi\Omega_{R0}T_{\rm sd}^2}{4\sqrt{2}T_{20}}+\ln(1+3l_*)}} \quad (19)$$

with the logarithmic factor l_* defined as

$$l_* = \frac{\ln(d_1 + c_1\Omega_{R0}T_{\rm sd})}{\ln\left(d_2 + \frac{c_2T_{20}}{\Omega_{R0}T_{\rm sd}^2}\right)}. \quad (20)$$

The numerical constants $c_1, d_1, c_2, d_2 \sim 1$ in the definition of l_* were introduced to describe the crossover between two

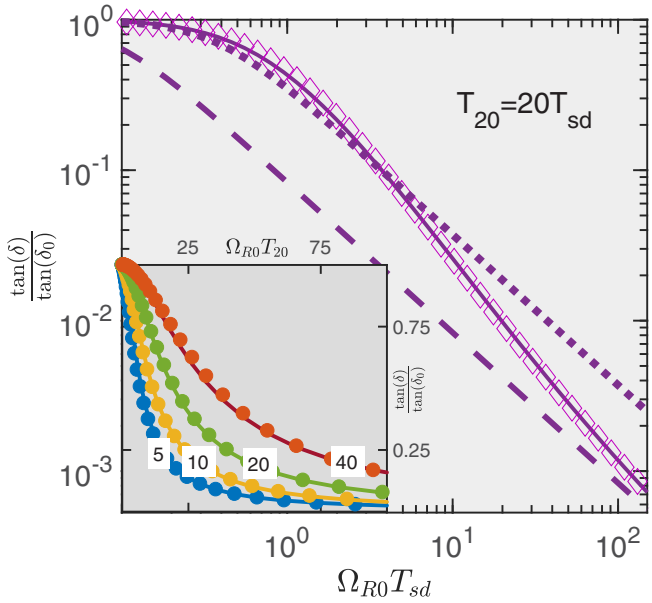

FIG. 2. The loss tangent vs Rabi frequency in the presence of the fast spectral diffusion $T_{20} \gg T_{\rm sd}$. The main graph shows the numerical solution (solid line), analytical interpolation of Eq. (19) (diamonds), the solution, Eq. (6), ignoring the spectral diffusion (dashed line), and the solution Eq. (10) with the modified decoherence rate (dotted line) in the case $T_{20} = 2T_{10} = 20T_{\rm sd}$. In the inset, the numerical (solid lines) and analytical [Eq. (19), circles] solutions are compared for different relative rates of spectral diffusion $T_{20}/T_{\rm sd}$ (indicated at each line).

regimes. They were estimated as 0.5,3,0.55,1.2, respectively, by fitting the exact numerical solution with Eq. (19) [39]. As shown in Fig. 2, this fit is perfectly consistent with the numerical solution and, therefore, it can be used to analyze experimental data. In the asymptotic regime of a large external field, $T_{10}/T_{\rm sd}^2 < \Omega_{R0}$, one can ignore the factor $\ln(1+3l_*)$ in the denominator in Eq. (19), which leads to Eq. (6), while in the opposite regime, the logarithmic term $l_* \approx l$ dominates in the denominator.

Using Eq. (20), one can estimate the crossover Rabi frequencies separating linear and nonlinear regimes (Ω_{c1}) and regimes of significant and negligible spectral diffusion (Ω_{c2}) characterized by the loss tangent behaviors $\tan(\delta) \propto \Omega_{R0}^{-2}$ and $\tan(\delta) \propto \Omega_{R0}^{-1}$, respectively. We assume that the spectral diffusion is much faster than the relaxation, i.e., $T_{\rm sd} \ll T_{20}$. For the first crossover representing the nonlinear threshold, one can estimate $\ln(1+3l_*) \approx 3\ln(d_1)/\ln(T_{20}/T_{\rm sd})$ within the logarithmic accuracy. The threshold Rabi frequency where the loss tangent gets smaller than its linear-response theory value by a factor of 2 can be expressed as

$$\Omega_{c1} \sim \frac{3.3}{T_{\rm sd}\ln\left(\frac{T_{20}}{T_{\rm sd}}\right)}. \tag{21}$$

This crossover frequency is proportional to the squared temperature. Consequently, the nonlinear threshold intensity increases as the fourth power of the temperature.

The second crossover frequency can be estimated setting two factors in the denominator of Eq. (17) responsible for the spectral diffusion $[\ln(1+3l_*)]$ and relaxation $[3\pi\Omega_{R0}T_{\rm sd}^2/(4\sqrt{2}T_{20})]$ regimes equal to each other. Then within logarithmic accuracy one gets

$$\Omega_{c2} \sim \frac{0.6T_{20}}{T_{\rm sd}^2}\ln\left[3\ln\left(\frac{T_{20}}{T_{\rm sd}}\right)\right]. \tag{22}$$

The estimates for the threshold Rabi frequency separating linear and nonlinear regimes has been obtained in Ref. [23] using qualitative arguments and in Ref. [25] using the rigorous perturbation theory analysis. Both estimates are consistent with the present work; moreover, the estimate of Ref. [25] differs only by a factor of $\pi/3.3 \approx 0.95$, which is an excellent agreement. The loss tangent intensity dependence predicted by Eq. (19) is also consistent with the qualitative estimates of Refs. [23,25], yet here it is obtained in a rigorous form. The weakening of that dependence at higher intensities to the inverse square-root behavior to our knowledge was not considered before the present work.

C. Loss tangent behavior in regimes where the present theory is not applicable

The analytical interpolation for the loss tangent given by Eq. (20) is applicable only if the spectral diffusion is faster than the TLS relaxation ($T_{\rm sd} < T_{10}$) that takes place at sufficiently large temperature $T > T_l$, Eq. (8), and the resonant TLS relaxation is faster than that of thermal TLSs responsible for the spectral diffusion suggesting $k_BT < \hbar\omega$, Eq. (1). For the typical microwave experimental frequency of 5 GHz, this limits the theory applicability to temperatures $30 < T < 200$ mK [31]. Below we consider the loss tangent outside of this temperature domain.

At low temperatures $T_{\rm sd} < T_{10}$, Eq. (13) is still applicable. In this regime, its numerical solution shown in the Supplemental Material [39] is almost identical to Eq. (4) so that the nonlinear absorption can be very well described by Eq. (6) ignoring the spectral diffusion. It is noticeable that in the crossover regime of $T_{\rm sd} = T_{10}$ the numerical solution, analytical interpolation of Eq. (20), and standard model described by Eq. (6) predict almost identical behaviors. At higher temperatures ($T_{\rm sd} < T_{10}$) one can use Eq. (20) until $k_BT < \hbar\omega$.

At high temperature, $k_BT > \hbar\omega$, the master equation formalism is no longer applicable and we cannot obtain the accurate quantitative solution for the loss tangent. Below, we suggest the qualitative arguments to predict the nonlinear absorption behavior in this regime ignoring possible logarithmic dependencies.

The crossover Rabi frequency Ω_c separating linear and nonlinear regimes can be estimated considering the probability of absorption during the resonant TLS relaxation time $T_{10} > T_{1T}$ [remember that $T_{1T} = 1/(\alpha T^3)$ estimates the relaxation time of thermal TLSs responsible for the spectral diffusion]. During that time, the TLS energy passes through the resonance T_{10}/T_{1T} times and the probability of absorption during each passage is given by $\Omega_R^2T_{\rm sd}^2$, so the total absorption probability during the time T_{10} can be estimated as $P_{\rm abs} \approx \Omega_R^2T_{\rm sd}^2T_{10}/T_{1T}$, provided that $P_{\rm abs} < 1$. The saturation in absorption is expected to take place at $P_{\rm abs} \sim 1$, suggesting the crossover Rabi

frequency

$$\Omega_{c1} \sim \frac{1}{T_{\rm sd}}\sqrt{\frac{T_{1T}}{T_{10}}}. \quad (23)$$

The more accurate estimate of the nonlinear threshold for $\hbar\omega_0 < k_B T$ including numerical and logarithmic factors can be found in Ref. [25]. Using definitions of Eqs. (3) and (7), one can estimate the temperature dependence of the threshold Rabi frequency as $\Omega_{c1} \propto T$, while the threshold intensity behaves as $I_c \propto \Omega_{c1}^2 \propto T^2$.

At small Rabi frequency, $\Omega_R < \Omega_c$, the loss tangent can be described by the linear-response theory expression given by Eq. (6), while at higher Rabi frequencies the inverse intensity dependence $\tan(\delta) = \tan(\delta_0)(\Omega_c/\Omega_R)^2$ is expected similarly to the intermediate temperature regime and in agreement with Ref. [25]. This dependence holds until the Rabi frequency is smaller than the typical maximum spectral diffusion range ($\Omega_R < \Omega_{c2} \sim T_{1T}/T_{\rm sd}^2$) while at larger intensities the nonlinear absorption is no longer sensitive to the spectral diffusion and it can be described by Eq. (6). It is straightforward to check that different asymptotic behaviors are consistent with each other at the crossovers.

In the case of thermal energy exceeding the field quantization energy, there exists the emergence of relaxational absorption that contributes to the loss tangent as $\tan(\delta_{\rm rel}) \approx \tan(\delta_0)\alpha T^3/\omega$ for $\hbar\Omega_{R0} < k_B T$ [20]. One should notice that the relaxational absorption is suppressed exponentially in the opposite limit of low temperatures given by Eq. (1) so it can be neglected there. Here it limits the maximum temperature to $10\hbar\omega/k_B$ to keep the crossover estimates valid and limits the maximum Rabi frequency to $\omega[\hbar\omega/(k_B T)]^3$ to keep the resonant absorption dominating. We assume both constraints are satisfied, as indicated in Table I.

D. Discussion of experiment

The theoretical predictions for the nonlinear absorption in the presence of spectral diffusion was made in Ref. [16] using Eq. (10) with a modified decoherence rate as described in Eq. (9). These predictions do not have any specific domain of relevance, yet they do not deviate dramatically from the predictions of the present theory [see Eq. (10) and the dotted line in Fig. 2 calculated at a spectral diffusion rate exceeding the TLS relaxation rate by a factor of 10, similarly to Ref. [16]]. Particularly, this approximate agreement could be the reason for the conclusion of Ref. [16] about the relevance of that approach to the experiments.

One should notice that the temperature dependence of the threshold intensity, which separates linear and nonlinear regimes, is experimentally observed as $I \propto T^4$, which conflicts with the theoretical model of Ref. [16] predicting $I \propto T^2$. However, it is consistent with the present theory determining the crossover Rabi frequency as the inverse TLS decoherence rate, $1/T_{\rm sd} \propto T^2$, Eq. (7). The critical intensity is determined by the squared Rabi frequency, leading to the T^4 dependence in agreement with the experiment [16] (cf. Ref. [34]).

We hope that the present work provides a solid background for future experiments that can verify the predictions of the theory. The analytical interpolation of Eq. (17) should serve as a guideline for the data analysis since it covers not only asymptotic regimes but crossovers between them. It is important that the novel behavior takes place in the domain of Rabi frequencies (intensities) restricted from both lower and upper sides [see Eqs. (22) and (22)], and therefore the ratio of relaxation and decoherence times $T_{10}/T_{\rm sd}$ determining the size of this domain should be chosen sufficiently large. The other significant problem of a finite pulse duration can also affect the experimental data [23,34]. This duration should exceed the TLS relaxation time to make the theory applicable [23].

Numerous measurements of the nonlinear loss tangent have been performed in Josephson junction qubits and resonators (see, e.g., Refs. [3,7–9]). All of them use Eq. (6) or (10) as a guideline for the experimental data analysis in a broad temperature range including the temperatures where the spectral diffusion is significant ($30 < T < 200$ mK). In the present work, we demonstrate that these equations are not applicable in this regime and should be modified according to Eq. (17). The experimental observations, indeed, show the intensity dependence different from $1/\sqrt{I}$ [Eqs. (6) and (10)] in the nonlinear regime. However, they show weakening compared to this dependence while the present work predicts its strengthening. In our opinion, this discrepancy can be understood assuming that the standard model of interacting TLSs in three dimensions is not quite relevant for quantum two-level systems in Josephson junctions. Particularly, amorphous films used in Josephson junctions possess the reduced dimensionality that can affect the interaction between TLSs and/or their statistics [43,44]. The theory should be modified accordingly, which is beyond the scope of the present work.

V. CONCLUSION

The present work suggests the resolution of the long-standing problem of nonlinear absorption by interacting two-level systems (TLSs) in low-temperature amorphous solids. The solution to this problem is obtained using the master equation formalism developed for the spectral diffusion of TLS resonant frequencies induced by their long-range interactions. It is demonstrated that the spectral diffusion extends the domain of a linear absorption compared to the noninteracting model, Eq. (6), to Rabi frequencies of the order of the TLS phase decoherence rate [$\Omega_{R0} \sim 1/T_{\rm sd}$; see Eq. (19)] provided that this rate exceeds the relaxation rate $1/T_{10}$. At larger Rabi frequencies, $1/T_{\rm sd} < \Omega_{R0} < T_{20}/T_{\rm sd}^2$, the loss tangent decreases inversely proportionally to the intensity of the external field [Ω_{R0}^{-2}; see Eq. (19)]. This new behavior can be understood assuming that the absorption by almost all TLSs, passing the resonance due to the spectral diffusion, is saturated for slow passage $\Omega_{R0} T_{\rm sd} > 1$ (cf. Refs. [31,45], where the slow resonance passage due to a bias sweep leads to similar behavior). TLSs passing the resonance absorb nearly the same energy $\hbar\omega$, while the absorption by other TLSs is negligible. Consequently, the absorbed energy is weakly sensitive to the field intensity, and the loss tangent is inversely proportional to that intensity. At larger intensities, the resonant domain size Ω_R exceeds the spectral diffusion range $T_{20}/T_{\rm sd}^2$ [17] and its increase with the intensity restores the earlier predicted behavior $\tan(\delta) \propto \Omega_{R0}^{-1}$, Eq. (6). For the typical microwave frequency $\omega/(2\pi) \sim 5$ GHz, the spectral diffusion remains

significant ($T_{10}/T_{sd} > 1$) at temperatures exceeding 30 mK [see Eqs. (3) and (7) and Ref. [31]]. The results of the consideration are summarized in Table I.

The theory is fully extendable to the nonlinear absorption of acoustic waves. The nonlinear internal friction can be described replacing the Rabi frequency in Eq. (19) with the product of the TLS-phonon interaction constant and the strain field ($\gamma\epsilon$; see reviews [20,33]). Although the present theory is not directly applicable to high temperatures $T > \hbar\omega/k_B$ where the thermal energy exceeds the external field quantization energy, the similar behavior of the nonlinear absorption is expected in this regime as well with modified crossover intensity as described in Sec. IV C and Table I.

The predictions of our theory can be verified experimentally by measuring the nonlinear absorption of acoustic or electromagnetic waves in "ordinary" glasses where the TLS model [1,2,36] is relevant. The analytical expression for the loss tangent given by Eq. (17) can be used as a guide line for the data analysis in a broad domain of parameters (see Table I). The results of previous measurements seem to be inconclusive (see the discussion in Sec. IV D) because the range of field intensities is insufficiently broad to distinguish between different intensity dependencies. According to our analysis, the conclusive measurements can be performed varying the external field intensity by one or two orders of magnitude in the nonlinear regime.

The predicted strengthening of the loss tangent intensity dependence in the nonlinear regime [$\propto I^{-1}$, Eq. (19)] contrasts with the observed weakening of the intensity dependence of microwave absorption in Josephson junction qubits [7,9,17,43,46,47]. The generalization of the present theory to low dimensions and modified TLS distribution compared to the standard tunneling model are possibly needed to interpret those experiments.

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