

Reduction of Dissipative Nonlinear Conductivity of Superconductors by Static and Microwave Magnetic Fields

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A theory of dissipative nonlinear conductivity, $\sigma_1(\omega, H)$, of *s*-wave superconductors under strong electromagnetic fields at low temperatures is proposed. Closed-form expressions for $\sigma_1(H)$ and the surface resistance $R_s(\omega, H)$ are obtained in the nonequilibrium dirty limit for which $\sigma_1(H)$ has a significant minimum as a function of a low-frequency ($\hbar\omega \ll k_B T$) magnetic field H . The calculated microwave suppression of $R_s(H)$ is in good agreement with recent experiments on alloyed Nb resonator cavities. It is shown that superimposed dc and ac fields, $H = H_0 + H_a \cos \omega t$, can be used to reduce ac dissipation in thin film nanostructures by tuning $\sigma_1(H_0)$ with the dc field.

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One of the hallmarks of superconductivity is that static magnetic fields H induce screening currents that break Cooper pairs and reduce the transition temperature T_c [1]. This manifests itself in the nonlinear Meissner effect [2] and intermodulation [3], which have been observed on high- T_c cuprates [4,5]. Behavior of a superconductor becomes far more complex under the alternating field $H = H_a \cos \omega t$, which not only induces pairbreaking currents, but also drives the quasiparticles out of equilibrium, particularly if the frequency ω exceeds the superconducting gap Δ [6]. Microwave absorption can produce nonequilibrium states with higher T_c and the critical current I_c as has been observed on thin films and tunnel junctions [7,8]. The effect of nonequilibrium Andreev states on the Josephson current-phase relation and I_c in superconducting weak links and hybrid nanostructures has recently attracted much interest [9–11].

At low temperatures $T \ll T_c$ and frequencies $\omega \ll \Delta$, the small density of quasiparticles affects neither T_c nor the dynamics of superconducting condensate, yet the effects of oscillating superflow and nonequilibrium quasiparticle states on dissipative kinetic coefficients cause a strong field dependence of the surface resistance $R_s(H)$. Usually R_s increases with the amplitude of the rf field [4,5], consistent with the expected enhancement of dissipation by pairbreaking currents, electron overheating, penetration of vortices, etc. A remarkable departure from this conventional scenario is the puzzling reduction of R_s by the rf field, which has been observed on many superconductors. For instance, R_s measured on the Nb resonator cavities at 2 K and 1–2 GHz typically decreases by 10%–20% at $H \approx 20$ –30 mT and then increases at higher fields [12,13]. Moreover, the Nb resonators alloyed with Ti [14] or N [15] impurities can exhibit even stronger microwave suppression of R_s (by $\approx 50\%$ –70% at 2 K) which extends to the fields $H \approx 90$ –100 mT at which the density of screening

currents $J \approx H/\lambda$ reaches $\approx 50\%$ of the pairbreaking limit $J_d \approx H_c/\lambda$, where H_c is the thermodynamic critical field and λ is the London penetration depth (see Fig. 1). Reduction of R_s by dc or microwave fields has also been observed on thin films [16–18]. The behavior of $\sigma_1(H)$ at $T \ll T_c$ is related to the fundamental limits of dissipation which controls decoherence in Josephson qubits [19] or performance of resonator cavities for particle accelerators [12] or microresonators [20].

In this work a theory of nonlinear conductivity and the microwave suppression of R_s in dirty *s*-wave superconductors is proposed. Here the electromagnetic response at weak fields is described by the local Ohmic relation $\mathbf{J}(\mathbf{r}, \omega) = [\sigma_1(\omega) - i\sigma_2(\omega)]\mathbf{E}(\mathbf{r}, \omega)$, where $\sigma_2 = 1/\mu_0\lambda^2\omega$ and σ_1 is the quasiparticle conductivity [21],

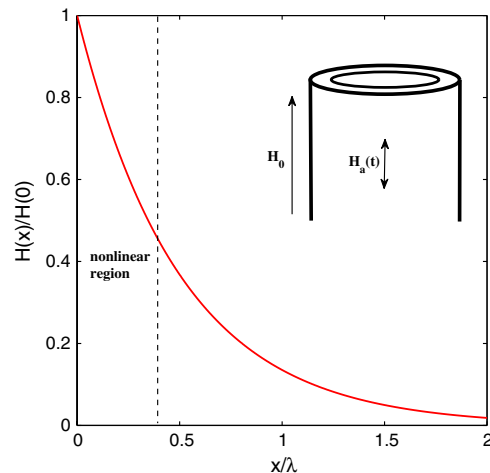


FIG. 1 (color online). Penetration of a parallel rf field into a superconductor. The dashed line depicts a layer where the current pairbreaking is essential. Inset shows a thin film ($d < \lambda$) deposited onto a cylindrical substrate in a superimposed dc and rf field.

$$\sigma_1 = (2\sigma_n \Delta/T) \ln(CT/\omega) e^{-\Delta/T}, \quad T \ll T_c, \quad (1)$$

where σ_n is the normal state conductivity, $C = 4e^{-\gamma} \approx 9/4$ and $\gamma = 0.577$. The logarithmic term in Eq. (1) comes from the convolution of the BCS density of states $\sigma_1 \propto \int_{\Delta}^{\infty} N(\epsilon)N(\epsilon + \omega)e^{-\epsilon/T} d\epsilon$, which diverges at $\omega = 0$ and $N(\epsilon) = N_0\epsilon(\epsilon^2 - \Delta^2)^{-1/2}$ [21], so smearing the gap singularities in $N(\epsilon)$ decreases σ_1 at $\omega \ll T$.

The broadening of the gap peaks in $N(\epsilon)$ and the reduction of a quasiparticle gap ϵ_g can be caused by current [1] or by magnetic impurities [22] which break the time reversal symmetry of pairing electrons. Particularly, the effect of dc current on $N(\epsilon)$ shown in Fig. 2 was observed by tunneling spectroscopy [23], in full agreement with the theory [24]. Under strong rf current, $N(\epsilon, t)$ oscillates between two solid curves in Fig. 2, so the peak in $\langle N(\epsilon) \rangle$ averaged over the rf period is smeared out within the energy region $\epsilon_g < \epsilon \lesssim \Delta$ of width $\delta\epsilon = \Delta - \epsilon_g \sim (J/J_d)^{4/3}\Delta$ at $J \ll J_d$ [23,24]. This picture gives insight into one of the mechanisms of microwave reduction of σ_1 : as the current-induced width $\delta\epsilon$ exceeds ω , the energy cutoff in the logarithmic term in Eq. (1) changes from ω to $\delta\epsilon$. Hence, $\sigma_1 \propto \ln[(J_d/J)^{4/3}T/T_c]$ decreases with J if $J > (\omega/\Delta)^{3/4}J_d$ and $\omega \ll T$, so that the decrease of ϵ_g in the Boltzmann factor $e^{-\epsilon_g/T}$ has a smaller effect on $\sigma_1(H)$ at $J < (T/\Delta)^{3/4}J_d$ since $\delta\epsilon < T$. For instance, $\omega/T \sim 2 \times 10^{-2}$ at 1 GHz at 2 K.

A theory of $\sigma_1(H)$ must address both the pairbreaking and nonequilibrium effects caused by microwaves. Most of the previous works have focused on nonequilibrium states caused by absorption of photons by quasiparticles while neglecting the effect of rf superflow on $N(\epsilon)$ at weak fields $H \ll (\omega/\Delta)^{3/4}H_c$ and $\omega \gtrsim T$ [7,8]. Here $\sigma_1(H)$ can be described by the linear response theory [21] but with a nonequilibrium quasiparticle distribution function $f(\epsilon, H)$

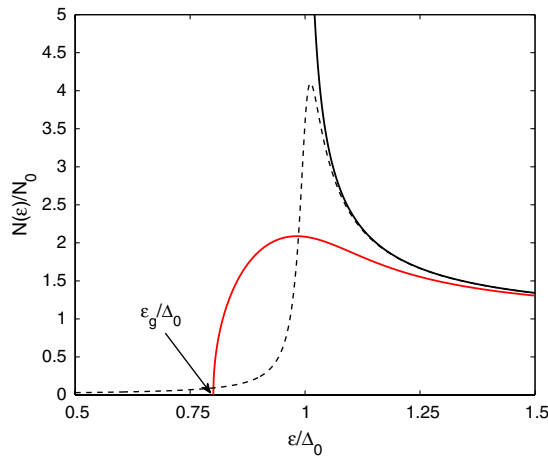


FIG. 2 (color online). The effect of current on the density of states calculated from Eqs. (6)–(8) at $s = 0.2$. The dashed line shows $N(\epsilon) = N_0 \text{Re}[(\epsilon - i\gamma)/\sqrt{(\epsilon - i\gamma)^2 - \Delta_0^2}]$ at $\gamma = 0.02\Delta_0$.

calculated from a kinetic equation. Using this approach, it was shown recently that $\sigma_1(H)$ can decrease with H_a as the quasiparticle population spreads to higher energies $\epsilon \gtrsim T$ [18], similar to the mechanism of stimulated superconductivity [25]. This result was used to explain the reduction of σ_1 with H_a observed on Al films at 5.3 GHz at 350 mK [18]. Here I consider a fundamentally different mechanism of microwave suppression of $\sigma_1(H)$ at strong, low-frequency fields with $\omega \ll T$ and $H > (\omega/\Delta)^{3/4}H_c$ for which the effect is due to the time-dependent $N(\epsilon, t)$ and a nonequilibrium distribution function controlled by oscillating superflow. In this case the Mattis-Bardeen theory is no longer applicable and $\sigma_1(H)$ is to be rederived using the Keldysh technique of nonequilibrium Green functions [6]. It is what was done in this work where the nonlinear conductivity $\sigma_1(H)$ was calculated for two cases: (1) a weak ac field superimposed onto the dc field $H(t) = H_0 + H_a \cos \omega t$, where the dc superflow can be used to tune $\sigma_1(H_0)$, and (2) parallel rf field $H(t) = H_a \cos \omega t$, as shown in Fig. 1.

In a type-II superconductor ($\lambda \gg \xi$) considered here, the rf field with $\omega \ll T$ does not generate new quasiparticles while $H(x, t)$ varies slowly over the coherence length ξ . In this case the dependence of $\mathbf{J}(\mathbf{r}, t)$ on the vector potential $\mathbf{A}(\mathbf{r}, t)$ is local but nonlinear and time dispersive. It can be expressed in terms of nonequilibrium matrix Green functions $\hat{G}(t, t', \mathbf{r})$ which satisfy the time-dependent Usadel equation coupled with kinetic equations taking into account scattering of quasiparticles on phonons [6,9,26]. The nonlinear conductivity $\sigma_1 = 2\langle \mathbf{J}\mathbf{E} \rangle / E_a^2$ is calculated in the Supplemental Material [27] by averaging the dissipated power over the rf period of slowly oscillating superflow at $\omega \ll T$ and $(H/H_c)^2 \ll 1$. Here $\mathbf{E} = -\partial_t \mathbf{A} = \mathbf{E}_a \sin \omega t$ is the electric field, $\hat{G}[e, Q(t)]$ depends on the local current density $\mathbf{J}(\mathbf{r}, t) = -\phi_0 \mathbf{Q}(\mathbf{r}, t) / 2\pi\mu_0\lambda^2$, where $\mathbf{Q} = \nabla\chi + 2\pi\mathbf{A}/\phi_0$, ϕ_0 is the flux quantum, χ is the phase of the order parameter, $\Delta(y) = \Delta e^{iQ(y)}$. The normal and anomalous Green functions are parametrized by $G^R = \cosh(u + iv)$ and $F^R = e^{iQy} \sinh(u + iv)$, where u and v satisfy the quasistatic Usadel equation [6,9]:

$$\epsilon + is \cosh(u + iv) = \Delta \coth(u + iv), \quad (2)$$

$$s(t) = DQ^2/2 = e^{-2x/\lambda} \beta(t) \Delta_0. \quad (3)$$

Here $\beta(x, t) = (H/2H_c)^2 = (J/2J_d)^2 \ll 1$, D is the electron diffusivity, $H_c = \phi_0/2^{3/2}\pi\mu_0\lambda\xi$, $\xi = (D/\Delta_0)^{1/2}$, and $\Delta_0 = \Delta(T=0, Q=0)$. A correction to Q due to the nonlinear Meissner effect [2] is disregarded.

The rf conductivity for the weak rf field superimposed onto the dc field is given by [27]

$$\sigma_1(H_0) = \frac{2\sigma_n}{\omega} (1 - e^{-\omega/T}) \int_{\epsilon_g}^{\infty} e^{-\epsilon/T} M(\epsilon, \omega, s) d\epsilon, \quad (4)$$

$$M(\epsilon, \omega, s) = \cos v_\epsilon \cos v_{\epsilon+\omega} \cosh(u_\epsilon + u_{\epsilon+\omega}), \quad (5)$$

where the spectral function $M(\epsilon, \omega, s)$ incorporates the effect of dc superflow on $N(\epsilon, Q)$ and the coherence factors. Here u_ϵ and v_ϵ are defined by the real and imaginary parts of Eq. (2), which yields the cubic equation $\sinh^3 2u + [(\epsilon^2 - \Delta^2)/s^2 + 1] \sinh 2u - 2\epsilon\Delta/s^2 = 0$ with the following Cardano solution:

$$\sinh 2u = [(r + \epsilon\Delta s)^{1/3} - (r - \epsilon\Delta s)^{1/3}]/s, \quad (6)$$

$$r = [\epsilon^2 \Delta^2 s^2 + (\epsilon^2 + s^2 - \Delta^2)^3/27]^{1/2}, \quad (7)$$

$$\sin v = [-\Delta + (\Delta^2 - s^2 \sinh^2 2u)^{1/2}]/2s \cosh u. \quad (8)$$

The quasiparticle density of states and the gap energy ϵ_g at which $N(\epsilon)$ vanishes (see Fig. 2) are given by $N(\epsilon) = N_0 \cosh u \cos v$, and [24]

$$\epsilon_g^{2/3} = \Delta^{2/3} - s^{2/3}, \quad \Delta = \Delta_0 - \pi s/4, \quad (9)$$

where Δ is obtained from the BCS gap equation at $T = 0$ in the first order in s [27]. Here $\epsilon_g(H_0)$ decreases with H_0 but remains finite ($\epsilon_g \approx 0.3\Delta_0$) even at the maximum superheating field $H_{sh} \approx 0.84H_c$ for the Meissner state [28]. Shown in Fig. 3 is the linear rf conductivity $\sigma_1(H_0)$ biased by a dc superflow calculated from Eqs. (4)–(9). At $H_0 = 0$ and $H_a \ll (\omega/\Delta)^{3/4}H_c$, Eqs. (4)–(9) reproduce Eq. (1), but at higher field $\sigma_1(H_0)$ has a minimum which becomes more pronounced as ω decreases. This behavior is due to interplay of the current-induced broadening of the gap peak in $N(\epsilon, s)$ and the reduction of ϵ_g shown in Fig. 2. As a result, σ_1 becomes dependent on H_0 if $H_0 > (\omega/\Delta)^{3/4}H_c$ and reaches minimum at $H_0 \sim (T/T_c)^{3/4}H_c \ll H_c$. The field region $(\omega/\Delta_0)^{3/4}H_c < H_0 < (T/\Delta_0)^{3/4}H_c$, where

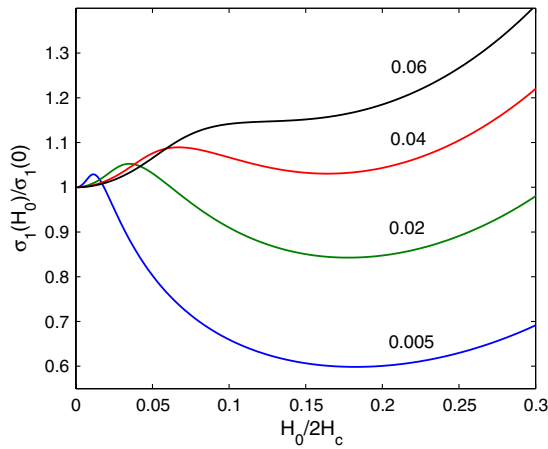


FIG. 3 (color online). Linear conductivity $\sigma_1(H_0)$ calculated from Eqs. (4)–(9) for ω/Δ_0 : 0.005, 0.02, 0.04, 0.06, and $T/\Delta_0 = 0.1$.

$\sigma_1(H_0)$ decreases with H_0 , shrinks as ω increases and disappears at $\omega > T$, as shown in Fig. 3.

Calculation of the nonlinear conductivity $\sigma_1(H_a)$ at a strong rf field $H(t) = H_a \cos \omega t$ requires taking temporal oscillations of $N(\epsilon, t)$ and $f(\epsilon, t)$ into account. Here $\sigma_1(H_a) = 2\langle \mathbf{J}\mathbf{E} \rangle / E_a^2$ is defined as before by averaging the power over the rf period [27]:

$$\sigma_1(H_a) = \frac{2\sigma_n}{\pi} \int_0^{\pi/\omega} dt \int_{\epsilon_g(t)}^{\infty} [f(\epsilon, s) - f(\epsilon + \omega, s)] M d\epsilon, \quad (10)$$

where $M[\epsilon, \omega, s(t)]$ is given by Eq. (5). Solving the kinetic equation for $f(\epsilon, s)$ with time-dependent parameters and the electron-phonon collision integral [6] is a very complicated problem, so I only consider here the case of $\min(\tau_r^{-1}, \tau_s^{-1}) \ll \omega \ll T$ for which the rf period is shorter than either the recombination time τ_r or the scattering time τ_s of quasiparticles on phonons [29]:

$$\tau_r = \tau_1(T_c/T)^{1/2} e^{\Delta/T}, \quad \tau_s = \tau_2(T_c/T)^{7/2}, \quad (11)$$

where τ_1 and τ_2 are materials constants. Taking $\Delta = 1.9T_c$, $T_c = 9.2$ K, $\tau_1 \approx 3 \times 10^{-12}$ s, and $\tau_2 \approx 8 \times 10^{-11}$ s for Nb [29] yields $\tau_r \sim 0.4 \mu\text{s}$ and $\tau_s \approx 1.7 \times 10^{-8}$ s at 2 K. The condition $\tau_s^{-1} < \omega < T$ that the quasiparticle density does not change during the rf period can be satisfied in a frequency range (0.06–44 GHz) relevant to many experiments [12,19,20].

The distribution function $f(\epsilon, t)$ can be obtained from the following consideration. As $s(t)$ increases, $N(\epsilon, s)$ extends to lower energies as shown in Fig. 2, but because the quasiparticles do not scatter during the rf period if $\omega\tau_s \gg 1$, the probability to occupy the energy state ϵ moved from the state $\tilde{\epsilon}$ at $s = 0$ does not change. The relation between $\tilde{\epsilon}$ and ϵ follows from the conservation of states: $\int_{\epsilon_g}^{\epsilon} N(\epsilon, s) d\epsilon = \int_{\Delta_0}^{\tilde{\epsilon}} N(\epsilon, 0) d\epsilon = N_0(\tilde{\epsilon}^2 - \Delta_0^2)^{1/2}$, giving $\tilde{\epsilon}^2 = \Delta_0^2 + [\int_{\epsilon_g}^{\epsilon} \cosh u \cos v d\epsilon]^2$. The function $f(\tilde{\epsilon})$ ensures that the quasiparticle density $n_{qp} = \int f(\tilde{\epsilon}) N(\epsilon, s) d\epsilon = \int_0^{\infty} f[\tilde{\epsilon}(\psi)] d\psi$ does not change during the rf period, where $\psi = \int_{\epsilon_g}^{\epsilon} N(\epsilon, s) d\epsilon$. Then the condition $f(\epsilon, s) = \exp(-\tilde{\epsilon}/T)$ at $s(t) = 0$ yields

$$f = \exp \left\{ -\frac{1}{T} \left(\Delta_0^2 + \left[\int_{\epsilon_g}^{\epsilon} \cos v_\epsilon \cosh u_\epsilon d\epsilon \right]^2 \right)^{1/2} \right\}. \quad (12)$$

The quasiparticle temperature T at $\omega\tau_s \gg 1$ is defined by the stationary power balance, $R_s H_a^2/2 = h(T_i - T_0) = Y(T - T_i)$. Here T_i is the lattice temperature, T_0 is the ambient temperature, $h = \kappa h_K / (dh_K + \kappa)$ accounts for heat transfer due to thermal conductivity κ and the Kapitza interface conductance h_K across a film of thickness d , and $Y(T)$ quantifies the energy transfer rate from

quasiparticle to phonons [30]. For weak overheating, the heat transfer may be linearized in $T - T_0 \ll T_0$:

$$T - T_0 = \frac{\alpha T_0}{R_{s0}} \left(\frac{H_a}{H_c} \right)^2 R_s(H_a, T), \quad (13)$$

$$\alpha = \frac{R_{s0} B_c^2}{2\mu_0^2 T_0} \left(\frac{1}{Y} + \frac{d}{\kappa} + \frac{1}{h_K} \right), \quad (14)$$

where Y , h , and $R_{s0} = R_s(T_0)$ are taken at $T = T_0$ and $H_a = 0$. The surface resistance $R_s(T, H_a)$ is calculated by integrating the local power $R_s H_a^2 / 2 = (\mu_0 \omega \lambda H_a)^2 \int_0^\infty e^{-2x/\lambda} \sigma_1(\beta) dx$. Changing here to integration over $\beta = \beta_0 e^{-2x/\lambda}$ defined by Eq. (3) yields

$$R_s = \frac{\mu_0^2 \omega^2 \lambda^3}{2\beta_0} \int_0^{\beta_0} \sigma_1(\beta) d\beta. \quad (15)$$

Equations (10) and (12)–(15) determine self-consistently $R_s(H_a, T)$ and $T(H_a)$. Shown in Fig. 4(a) is $\sigma_1(H_a)$ calculated from Eqs. (10)–(14) for different values of α . The field dependence of $\sigma_1(H_a)$ is similar to that of $\sigma_1(H_0)$ in Fig. 3: in both cases the current-induced smearing of $N(\epsilon)$ reduces $\sigma_1(H)$, but the mechanisms of the increase of $\sigma_1(H)$ at higher fields are different. For a weak rf field superimposed onto the dc field, the increase of $\sigma_1(H_0)$ results from the reduction of $\epsilon_g(H)$, while the increase of $\sigma_1(H_a)$ in Fig. 4(a) is due to overheating: the condition that the density of quasiparticles does not change during the rf cycle greatly enhances the microwave reduction of $\sigma_1(H_a)$. If $\omega \ll [\tau_s^{-1}(T), \tau_r^{-1}(T)]$, the minimum in $\sigma_1(H_a)$ is controlled by the field reduction of $\epsilon_g(H_a)$ as $f(\epsilon) \rightarrow \exp(-\epsilon/T)$ becomes more equilibrium.

Shown in Fig. 4(b) is $R_s(B_a)$ calculated from Eqs. (10) and (12)–(15) to fit the experimental data of Ref. [14] with only one adjustable parameter $\alpha = 0.91$ for which the overheating $T - T_0 \approx 0.17$ K calculated from Eq. (13) is indeed weak even at $B_a = 80$ mT, $R_s(B_a)/R_{s0} = 0.6$, and $T_0 = 2$ K. This theory describes well the microwave suppression of R_s observed on Ti-alloyed Nb cavities [14]. For $R_s = 20$ n Ω , $\kappa = 10$ W/mK, $h_K = 5$ kW/m² K at 2 K, and $d = 3$ mm [14], the phonon heat transfer in Eq. (14) can only account for $\alpha \approx 0.06$. The larger value of $\alpha = 0.91$ used to fit $R_s(H_a)$ in Fig. 4(b) indicates a significant role of electron overheating [27,30].

The parameters $Y(T)$, τ_s , and τ_r are not only controlled by the scattering and recombination of quasiparticles [29,30], but also by the smearing of the gap peak in $N(\epsilon)$ due to inhomogeneities, inelastic scattering, or impurities [22,31], which can make Y very sample dependent. Interplay of the subgap states and current pairbreaking can bring about competing mechanisms of nonlinearity of $R_s(H_a)$, since the subgap states can cause both a finite τ_r at $T \rightarrow 0$ [32] and a residual conductivity [33]. In any case, the microwave suppression of $R(H_a)$ is more pronounced

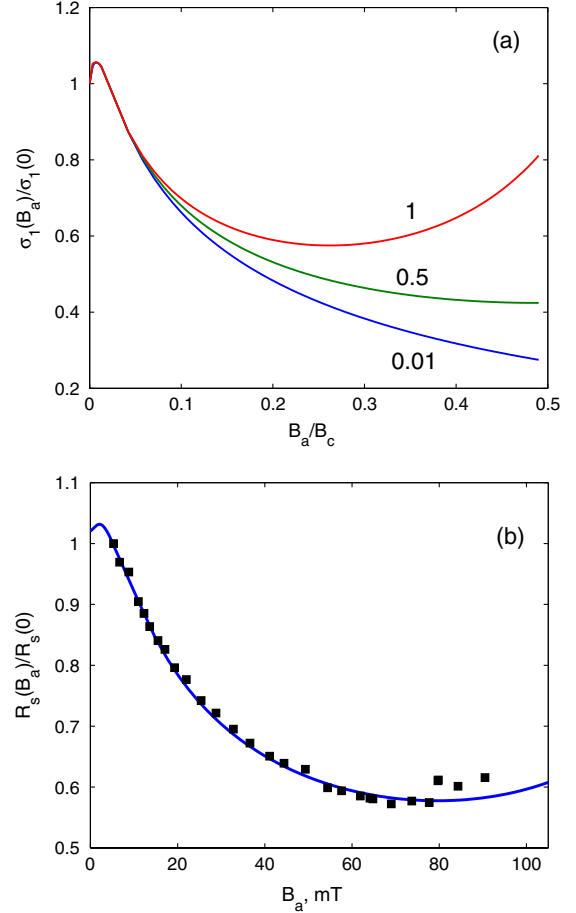


FIG. 4 (color online). (a) $\sigma_1(B_a)$ calculated from Eqs. (10)–(15) at $T/\Delta_0 = 0.1$, $\omega/\Delta_0 = 0.0025$, and $\alpha = 0.01, 0.5$, and 1 . (b) $R_s(B_a)$ calculated for $\alpha = 0.91$, $T_0 = 2$ K, $\Delta_0 = 17.5$ K, and $B_c = 200$ mT. The dots show the experimental data for the Nb cavity at 1.75 GHz [14]. The quench at $B_a \approx 90$ mT $\approx 0.5B_c$ is likely due to penetration of vortices at surface defects.

for sharper gap peaks in $N(\epsilon)$ at $H_a = 0$, so that τ_s and τ_r are not much reduced and the current-induced broadening of $N(\epsilon)$ takes over at comparatively low fields (see Fig. 2). This conclusion is consistent with the observed variability of the field-induced reduction of $R_s(H_a)$ [12–15] and the tunneling measurements [14], which revealed fewer subgap states in $N(\epsilon)$ for the Nb resonators exhibiting the significant minimum in $R_s(H_a)$ shown in Fig. 4(b). A dc field applied parallel to a thin film can be used to tune $\sigma_1(H_0)$ and separate current pairbreaking from nonequilibrium effects [16,34].

In conclusion, a theory of nonlinear conductivity of dirty superconductors at low temperatures and strong rf electromagnetic field is developed. The theory explains the effect of the field-induced suppression of surface resistance, in excellent agreement with recent experiments.

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