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## Theoretical Model to Explain Excess of Quasiparticles in Superconductors

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Experimentally, the concentration of quasiparticles in gapped superconductors always largely exceeds the equilibrium one at low temperatures. Since these quasiparticles are detrimental for many applications, it is important to understand theoretically the origin of the excess. We demonstrate in detail that the dynamics of quasiparticles localized at spatial fluctuations of the gap edge becomes exponentially slow. This gives rise to the observed excess in the presence of a vanishingly weak nonequilibrium agent.

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Naïvely, the superconducting gap  $\Delta$  should ensure an exponentially small quasiparticle concentration at low temperatures. However, various experiments indicate that a long-lived, nonequilibrium quasiparticle population persists in the superconductor [1–7]. The so-called quasiparticle poisoning [8], whereby an unwanted quasiparticle is trapped in a bound state, is an important factor harming the ideal operation of superconducting devices [9]. Unwanted quasiparticles also forbid tempting perspectives to use Majorana states in superconductors for topologically protected quantum computing [10–12]. The poisoning rates have been quantified [13–17], and much experimental work is directed on protection from poisoning, with important advances in this direction [18–22]. The nonequilibrium quasiparticles are produced by some nonequilibrium agent, which is most likely related to the absorption of electromagnetic irradiation from the high-temperature environment [23] and/or electromagnetic fields applied to the setup in the course of its measurement and operation. Surprisingly, the efforts to reduce the intensity of this nonequilibrium agent are not entirely satisfying: the experiments give a substantial residual quasiparticle concentration, even if all efforts are performed [24,25].

In this Letter, we make a step toward the solution of this long-standing puzzle that impedes the successful implementation of superconducting quantum information processors. We investigate the peculiar dynamics of the annihilation of quasiparticles localized at the spatial fluctuations of the gap edge. Such fluctuations exist in all superconductors owing to inevitable disorder. Importantly, we find that the average distance between the quasiparticles depends only *logarithmically* on the intensity of the nonequilibrium agent, which is a consequence of the exponential dependence of the annihilation rate on the distance between two quasiparticles. It results in the quasiparticle concentration

$$c = \frac{C_p}{(4\pi/3)r^3} \quad \text{with} \quad \frac{r}{r_c} \approx \ln\left(\frac{\bar{\Gamma}}{Ar_c^6}\right), \quad (1)$$

valid at small  $A \ll \bar{\Gamma}/r_c^6$  [A more accurate estimate for  $r$  is given by Eq. (9)]. Here,  $r_c$  is the relevant radius of the localized quasiparticle state to be estimated in detail below: for practical circumstances, it exceeds the superconducting coherence length  $\xi_0$  by not more than an order of magnitude. Furthermore,  $A$  is the rate of nonequilibrium generation of quasiparticles per unit volume, and  $\bar{\Gamma}$  is a material constant characterizing the inelastic quasiparticle relaxation due to electron-phonon interaction. The packing coefficient  $C_p \approx 0.605 \pm 0.008$  can be derived from a simple *bursting bubbles* model outlined below. Equation (1) predicts a substantial concentration of quasiparticles, as well as the inefficiency of efforts to reduce it by shielding the device, consistent with the experimental observations. More work is needed to quantitatively describe the data.

Let us outline the derivation of the above relations. The relevant quasiparticles have energies close to the gap edge, and they annihilate by emitting a phonon with energy  $\sim 2\Delta$ . Assuming the “dirty” limit  $\ell \ll v_F/\Delta$ , where  $v_F$  is the Fermi velocity, and the phonon wavelength not exceeding the mean free path  $\ell$ , we derive a remarkably simple relation for the annihilation rate of two quasiparticles [26]

$$\Gamma_{12} = \bar{\Gamma} \int d\mathbf{r} p_1(\mathbf{r}) p_2(\mathbf{r}). \quad (2)$$

Here,  $p_{1,2}(\mathbf{r})$  are the normalized probability densities to find the quasiparticles 1,2 at position  $\mathbf{r}$ . Furthermore, we find [26]  $\bar{\Gamma} = 24\gamma(\Delta)/(\nu_0\Delta)$ , where  $\nu_0$  is the normal-metal density of states and  $\gamma(\Delta)$  is the normal-metal electron-phonon relaxation rate at energy  $\Delta$ . For aluminum, this yields  $\bar{\Gamma} \approx 40 \text{ s}^{-1} \mu\text{m}^3$ . Equation (2) is valid for localized as well as for delocalized states.

For large enough quasiparticle concentrations (in particular, for delocalized states), one can neglect the correlations in their positions. In that case, a simple mean-field calculation [30] shows that the balance between generation of nonequilibrium quasiparticles and their annihilation  $A = \bar{\Gamma}c^2$  results in the nonequilibrium concentration  $c = (A/\bar{\Gamma})^{1/2}$ . In this regime, a generation rate  $A \approx 4 \times 10^3 \text{ s}^{-1} \mu\text{m}^{-3}$  would thus result in  $c \approx 10 \mu\text{m}^{-3}$ . However, the annihilation itself reduces the probability for quasiparticles to be close to each other. Therefore, it boosts the nonequilibrium concentration. This effect is most pronounced if the quasiparticles are in localized states and do not move.

The description of the quasiparticle bound states is provided in Ref. [31] and has been recently revisited [32] in the context of strongly disordered superconductors. The main results can be summarized as follows. The short-range fluctuations of the pairing potential shift the gap edge  $E_g = \Delta - \varepsilon_g$ , by  $\varepsilon_g \ll \Delta$ , and smooth the density of delocalized states on the same scale  $\varepsilon_g$ . The long-range fluctuations of the pairing potential generate a tail of localized states at energies  $E < E_g$ . As the typical extent of these localized states is much larger than the correlation length of the pairing potential fluctuations, the latter can be regarded as point correlated, with the two-point correlation function  $\langle\langle \Delta(\mathbf{r})\Delta(\mathbf{r}') \rangle\rangle = (\delta\Delta)^2 \delta(\mathbf{r} - \mathbf{r}')$ . The intensity of the fluctuations is conveniently characterized by a dimensionless parameter  $F = a_1(\delta\Delta)^2/(\Delta^2\varepsilon_0^3)$ , where  $a_1 \approx 0.045$  [33],  $\varepsilon_0 = \sqrt{\hbar D/\Delta}$  is the diffusive coherence length, and  $D$  is the diffusion constant in the normal metal. For a typical localized state with energy  $E < E_g$ , the energy distance from the edge  $\varepsilon = E_g - E$  is of the order of the typical fluctuation  $\delta\Delta/L^{3/2}(\varepsilon)$ , on the length scale  $L(\varepsilon)$  of this state. The length scale itself depends on energy,  $L(\varepsilon) = \xi_0[2\Delta/(3\varepsilon)]^{1/4}$ . From this, one derives the energy scale  $\varepsilon_T = F^{4/5}\Delta$  of the exponential tail and the corresponding length scale  $L_T = L(\varepsilon_T) \approx 0.90\xi_0/F^{1/5}$ . Strong disorder would result in  $\varepsilon_T, \varepsilon_g \sim \Delta$ . However, it is typically not the case in standard superconductors for which  $\varepsilon_T \ll \varepsilon_g \ll \Delta$ . At  $\varepsilon_T \ll \varepsilon \ll \varepsilon_g$ , the density of states reaches an exponential asymptotics

$$\nu(\varepsilon) \approx \nu_T(\varepsilon/\varepsilon_T)^{9/8} \exp[-(\varepsilon/\varepsilon_T)^{5/4}], \quad (3)$$

where  $\nu_T = a_2\nu_0\sqrt{\varepsilon_T\Delta/\varepsilon_g^2}$  and  $a_2 \approx 0.79$  [33], and the most probable shape of the localized state is given by

$$p_{\text{LO}}(\mathbf{r}) = \frac{f(r/L(\varepsilon))}{2\pi L^3(\varepsilon)} \quad \text{with} \quad f(x) \equiv \frac{\sinh x}{x \cosh^3 x}. \quad (4)$$

Let us consider a quasiparticle generated by a non-equilibrium agent. Typically, its energy is much larger than  $\Delta$ . However, it loses its energy quickly due to low-energy

electron-phonon interactions before annihilating with another quasiparticle. At some stage, the quasiparticle reaches the gap edge and becomes localized at  $\varepsilon \approx \varepsilon_T$ . It is important for us to understand that its relaxation does not stop here. One can estimate the number of localized states that overlap with a given state and have a lower energy as

$$N(\varepsilon) \equiv L^3(\varepsilon) \int_{\varepsilon}^{\infty} d\varepsilon' \nu(\varepsilon') \approx N_T(\varepsilon/\varepsilon_T)^{1/8} \exp[-(\varepsilon/\varepsilon_T)^{5/4}], \quad (5)$$

with  $N_T = (4/5)\nu_T\varepsilon_T L_T^3$ . This number is likely to be big at  $\varepsilon \approx \varepsilon_T$ , where  $N(\varepsilon_T) \sim N_T \sim g^4\sqrt{\varepsilon_T^3\Delta}/\varepsilon_g \gg 1$  [34]. Here,  $g = \pi\nu_0\Delta\xi_0^3$  is the number of Cooper pairs in a cube of size  $\xi_0$ . Thus, the quasiparticle will relax further from these states, and the relaxation stops only at a rather definite energy  $\varepsilon_c$  [36] defined by  $N(\varepsilon_c) \approx 1$ ,  $\varepsilon_c \approx \varepsilon_T(\ln N_T)^{4/5}$ . Therefore, we come to a rather unexpected conclusion: the quasiparticles end up their random relaxation process at a rather definite radius  $r_c \equiv L(\varepsilon_c)/2$ , that is,

$$r_c \approx 0.45\xi_0(\varepsilon_T/\Delta)^{-1/4}(\ln N_T)^{-1/5}, \quad (6)$$

as illustrated in Fig. 1. Taking standard parameters for Al [37], we expect that scale to be only slightly larger than half the coherence length  $\xi_0 \approx 100 \text{ nm}$ . For instance, taking  $\varepsilon_T/\Delta = 10^{-2}$  and  $\varepsilon_g/\Delta = 10^{-4}$ , we find  $r_c \approx \xi_0$  and  $3\xi_0$ , respectively.

Using these results, we can formulate a model of stochastic quasiparticle dynamics [38]. The quasiparticles appear in random points with the rate  $A$ , keep their positions, and annihilate pairwise with a rate  $\Gamma(\mathbf{R})$  that is a function of their mutual distance  $\mathbf{R}$ . The rate is obtained from Eqs. (2) and (4). Namely,

$$\Gamma(\mathbf{R}) = \bar{\Gamma} \int d\mathbf{r} p_{\text{LO}}(\mathbf{r}) p_{\text{LO}}(\mathbf{r} + \mathbf{R}) \equiv \bar{\Gamma} r_c^{-3} g(R/r_c), \quad (7)$$

where

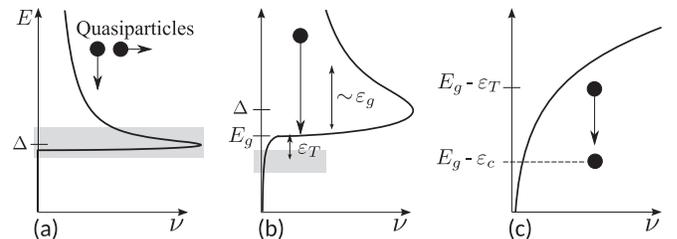


FIG. 1. The density of states and single quasiparticle relaxation in a superconductor. (a) The density of states is BCS like, except near the gap edge. (b) Near the gap edge, the singularity is rounded at an energy scale  $\varepsilon_g$ , and a tail of localized states within the gap develops at an energy scale  $\varepsilon_T$ . (c) The relaxation of a single quasiparticle stops at an energy scale  $\varepsilon_c > \varepsilon_T$ , where the localized states with lower energy no longer overlap.

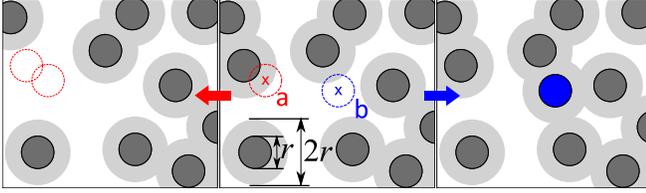


FIG. 2. Illustration of the bursting bubbles model. Each particle is represented by a bubble with diameter  $r$  (dark gray circles). If a new particle appears centered in the gray area with diameter  $2r$  (case a, central panel), it immediately annihilates with another particle (left panel). If the particle appears in the white area (case b), it is simply added to the system (right panel).

$g(2x) = (16\pi\sinh^4x)^{-1}(3 + 2\sinh^2x - 3\cosh x \sinh x/x)$ . In particular,  $g(x) \approx 1/(60\pi)$  at  $x \ll 1$  and  $g(x) \approx 1/(2\pi)\exp[-x]$  at  $x \gg 1$ .

The behavior of the model is governed by a single dimensionless parameter  $Ar_c^6/\bar{\Gamma}$ . At large values of this parameter, the typical distance between quasiparticles  $r$  is much smaller than  $r_c$ , and correlations are negligible as  $\Gamma(r) \sim \bar{\Gamma}r_c^{-3}$  is constant on that length scale. In this limit, we recover the mean-field result given above  $c = (A/\bar{\Gamma})^{1/2}$ , which is independent of  $r_c$  and does not rely on tail states. At small values of the parameter  $Ar_c^6/\bar{\Gamma}$ ,  $r$  is much larger than  $r_c$ . In this limit, it can be estimated from the competition of the annihilation rate  $\sim \bar{\Gamma}r_c^{-3}\exp[-r/r_c]$  and the generation rate within the typical volume of a quasiparticle  $\sim Ar^3$ . Thus,  $r \approx r_c \ln[\bar{\Gamma}/(Ar_c^6)]$ .

Because of the exponential dependence of the annihilation rate on the typical distance, one of the rates prevails over the other completely if the distance is changed by  $\delta r \sim r_c \ll r$ . This allows one to introduce a simplified model of bursting bubbles, see Fig. 2. Regardless the details of  $\Gamma(\mathbf{R})$ , we can consider the quasiparticles as spherical bubbles of radius  $r/2$ . If two bubbles overlap, the particles

annihilate. This model is easily simulated: we add bubbles to the system at random points. If the added bubble does not overlap with the existing ones, the number of quasiparticles is increased by 1. If there is an overlap, two bubbles burst, decreasing the number by 1. Equilibrium is achieved when these two outcomes happen with equal probabilities. This is the case when the volume covered by spheres of radius  $r$  centered around the quasiparticles equals half of the whole volume. If we rather naïvely assume that the spheres do not overlap, the volume covered is  $4\pi r^3/3$  per quasiparticle and the concentration is  $c = C_p(4\pi r^3/3)^{-1}$  with  $C_p = 0.5$ . In reality, some spheres overlap, so the simulation yields a slightly bigger packing coefficient, see Eq. (1).

To improve upon the logarithmic estimation of  $r$ , we performed simulations of the full model taking into account the details of  $\Gamma(\mathbf{R})$  [26]. The stationary concentrations are shown in Fig. 3.

In the limit  $\tilde{r} \equiv r/r_c \gg 1$ , the dynamics of the quasiparticle concentration is given by an evolution equation  $\dot{c}(t) = A - \Gamma_{\text{fit}}c(t)$ , with the effective asymptotic relaxation rate  $\Gamma_{\text{fit}}(r) = 4\pi/(3C_p)b\bar{\Gamma}r_c^{-3}\tilde{r}^\beta e^{-\tilde{r}}$ . Expressing  $c(t) = C_p/[4\pi(\tilde{r}r_c)^3/3]$  and introducing dimensionless time in units of  $9C_p r_c^3/(4\pi\bar{\Gamma})$ , this equation simplifies to

$$\dot{\tilde{r}} = (Ar_c^6/\bar{\Gamma})\tilde{r}^4 - b\tilde{r}^{\beta+1}e^{-\tilde{r}}. \quad (8)$$

The parameters  $b$  and  $\beta$  can be obtained by fitting the simulation at small values of  $Ar_c^6/\bar{\Gamma}$  with the stationary solution of Eq. (8) determined from

$$Ar_c^6/\bar{\Gamma} = b\tilde{r}^{\beta-3}e^{-\tilde{r}} \quad (9)$$

that improves the accuracy of Eq. (1). We find  $\beta = 0.41$  and  $b = 0.008$  [26]. At larger values of  $Ar_c^6/\bar{\Gamma}$ , corresponding to  $\tilde{r} \lesssim 3.0$ , the dependence of the concentration crosses over to the square-root law discussed above.

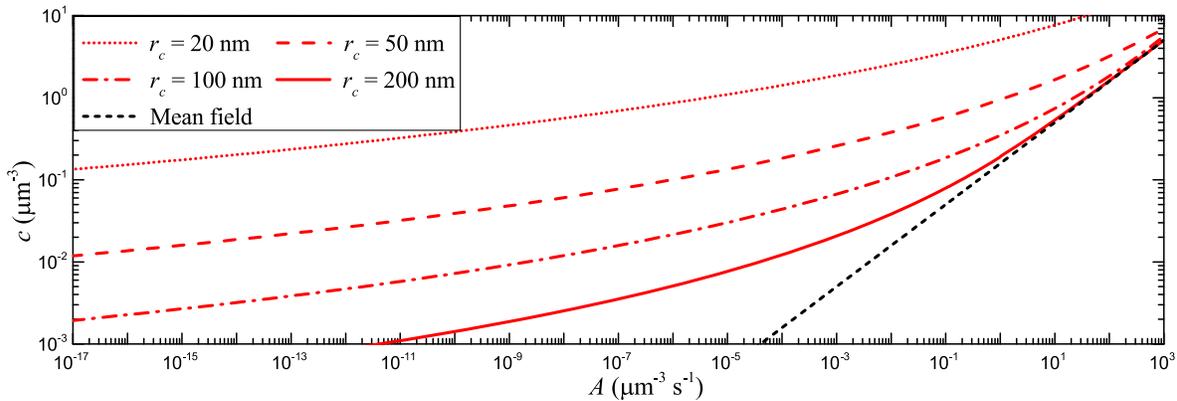


FIG. 3. Concentration  $c$  as a function of the generation rate  $A$  for quasiparticles annihilating pairwise with the rate given by Eq. (7) with  $\bar{\Gamma} = 40\text{s}^{-1}\mu\text{m}^3$  and several values of the quasiparticle localization radius  $r_c$ . The straight line shows the mean-field estimate  $c = \sqrt{A/\bar{\Gamma}}$  for comparison.

If the nonequilibrium agent ceases to work  $A = 0$ , the quasiparticle concentration relaxes very slowly. In particular, Eq. (8) yields the estimate  $\tilde{r}(t) \propto \ln(\bar{\Gamma}t/r_c^3)$ . Beyond the logarithmic approximation, the results of the simulation [26] are consistent with those obtained from the stationary solution.

Using realistic values for the generation rate, we thus can give accurate estimates of the quasiparticle concentration. In particular, cosmic radiation at the sea level is dominated by muons with energy in the GeV range and a flux of  $\sim 1 \text{ min}^{-1} \text{ cm}^{-2}$  [39]. The stopping power of GeV muons in aluminum is  $\sim 1 \text{ MeV cm}^{-1}$  [39]. Assuming an efficient conversion of the deposited energy into quasiparticles [40], we thus find a generation rate  $A \sim 10^{-4} \text{ s}^{-1} \mu\text{m}^{-3}$  ( $\sim 10 \text{ day}^{-1} \mu\text{m}^{-3}$ ). At  $r_c \sim 0.1 \mu\text{m}$ , it yields a quasiparticle concentration  $c \sim 0.01 \mu\text{m}^{-3}$ , which is close to the one measured in two recent experiments [6,24], where best efforts were performed in screening electromagnetic radiation.

In the above considerations, we have assumed that the annihilation rate does not depend on the spin state of two quasiparticles. This is valid in two cases: (i) the localization radius  $r_c$  exceeds the spin-orbit relaxation length, which may be relevant for heavy-atom metals, and (ii) the spin coherence time of an isolated quasiparticle is shorter than the (exponentially long) time scale  $\Gamma_{\text{fit}}$  for annihilation. In the opposite regimes, the quasiparticles could only annihilate if in a spin-singlet state.

To account for the spin structure is a challenging task owing to complex quantum entanglement of the spins of the overlapping quasiparticles that survive the annihilation. As a simplifying description, we considered an extension of the bursting bubbles model in which each bubble is assigned a classical spin degree of freedom. Whenever two bubbles with opposite spins overlap, they burst. The result of our simulation [26] is an enhanced  $C_p \approx 2.19 \pm 0.05$ . When spin-flip processes are added, the concentration decreases down to  $C_p \approx 0.61$  upon increasing the spin-flip rate, in agreement with the above considerations for the spinless case.

The validity of our estimation is limited by a variety of complex factors that can influence the nonequilibrium quasiparticle dynamics in superconductors. In particular, we ignored the possible formation of deep quasiparticle traps at the surface of the superconductor and quasiparticle accumulation in these traps. We also assumed immobile quasiparticles, which is valid in the limit of a vanishing temperature. At finite temperature, the quasiparticles could diffuse owing to inelastic transitions, even if they reside in localized states. This would favor their annihilation, as they would come closer to each other. As a result, the estimate for the concentration given in this Letter is rather an upper bound at a given generation rate. The evaluation of the diffusion of localized quasiparticles, as well as its complex temperature dependence, would be a subject of interesting research that is needed to understand the details of their dynamics.

In conclusion, our work provides a crucial element in the understanding of the excess quasiparticles in superconducting devices. The quasiparticles trapped at fluctuations of the gap edge certainly contribute to low-frequency absorption in the bulk. Furthermore, the possibility of their activated motion in the vicinity of a Josephson junction is expected to provide a deleterious effect on the coherence properties of a superconducting qubit by tunneling [41]. Thus, taking into account the physical phenomenon discussed in this work is essential for a correct interpretation of several experiments mentioned above, for planning new ones, and ultimately for the solution to quasiparticle poisoning.

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