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Dynamical Spin Polarization of Excess Quasiparticles in Superconductors

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We show that the annihilation dynamics of excess quasiparticles in superconductors may result in the spontaneous formation of large spin-polarized clusters. This presents a novel scenario for spontaneous spin polarization. We estimate the relevant scales for aluminum, finding the feasibility of clusters with total spin $S \simeq 10^4 \hbar$ that could be spread over microns. The fluctuation dynamics of such large spins may be detected by measuring the flux noise in a loop hosting a cluster.

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Various experiments using superconductors have been interpreted in terms of a long-lived, nonequilibrium quasiparticle population that persists at low temperatures [1–21]. Such quasiparticles may be created, for example, by Cooper pair breaking due to the absorption of stray photons or cosmic rays—the dominant mechanism is not clear at the moment. The bottleneck for their evacuation is the two-particle recombination mediated by the electron-phonon interaction. A simple balance predicts a residual quasiparticle density $n \sim c_0 = (2A/\bar{\Gamma})^{1/2}$, where A is the rate of nonequilibrium generation of quasiparticles per unit volume, and $\bar{\Gamma}$ is a material constant characterizing the inelastic quasi-particle relaxation due to the electron-phonon interaction. The subject has attracted much interest recently as excess quasiparticles will ultimately limit the performance of many superconducting devices [3,5,6,9–11,13]. Therefore one needs to deepen earlier studies on quasiparticle relaxation as, e.g., Ref. [23]. Several strategies, such as quasiparticle trapping in normal islands or vortices [1,9,12,18] and quasiparticle pumping with microwave pulse sequences [24], can be used to evacuate quasiparticles from the region of interest and lead to a better device performance. By contrast, unintentional trapping of quasiparticles in bound states below the superconducting gap edge, present in disordered superconductors, may slow down the relaxation dramatically at low concentrations [25] since the recombination requires two quasiparticles and thus is exponentially suppressed for those in distant bound states.

All above considerations neglect the quasiparticle spin. We note the spin selectivity of the recombination process in the absence of interactions violating spin conservation, the recombination only proceeds if two quasiparticles are in a spin-singlet state. In this Letter, we show that this spin selectivity may become a mechanism of nonequilibrium spin polarization. The quasiparticles align their spins

forming a polarized cluster with greatly enhanced concentration, the number of particles in the cluster and its size being limited by spin relaxation processes. We derive the corresponding conditions for a fully gapped superconductor, such as aluminum, showing the feasibility of the clusters of $\sim 10^4$ quasiparticles that could be spread over microns. The polarization of the cluster slowly fluctuates in time, and we propose a simple setup where the resulting noise can be utilized for the experimental observation of the phenomenon.

A cluster consists of an ensemble of quasiparticles with mutually overlapping wave functions. In the presence of spin-singlet recombination, a cluster of N quasiparticles is stable only if no pair of quasiparticles has an overlap with a spin-singlet state. This is the case if the cluster is in a maximal spin state, with total spin $S = N/2$ (in units with $\hbar = 1$). Let us align the z axis with the cluster polarization.

If a new quasiparticle is added to such a cluster, the number of quasiparticles changes by 1: $N \rightarrow N' = N + 1$, whereas the total spin changes by $\pm 1/2$: $S \rightarrow S' = S \pm 1/2 = (N \pm 1)/2$. The z projection of the spin is $S'_z = N/2 + s_z$, where $s_z = \pm 1/2$ is the z projection of the spin of the incoming particle. Thus, if $s_z = 1/2$, we obtain the maximal spin state $|S' = (N + 1)/2, S'_z = (N + 1)/2\rangle$. By contrast, if $s_z = -1/2$, there are two possible spin states: $|S' = (N \pm 1)/2, S'_z = (N - 1)/2\rangle$. The relative probabilities of these two possibilities are determined by the corresponding Clebsch-Gordan coefficients, which are given in Sec. I of the Supplemental Material [26]. Note that $|S' = (N + 1)/2, S'_z = (N - 1)/2\rangle$ is also a maximal spin state, though its polarization is not along the z axis anymore. Since the orientation of the incoming spin is random, the probabilities for being and not being in a maximal spin state are thus given as $[1 + 1/(N + 1)]/2$ and $[1 - 1/(N + 1)]/2$, respectively. As a consequence, the probability that the new cluster is

stable is larger than the probability that the new cluster can decay. This asymmetry thus favors the growth of spontaneously polarized clusters of quasiparticles. The polarization axis of such a cluster is not fixed, but it changes randomly and slightly with each new quasiparticle added.

From this consideration, we construct a simple model for the spin dynamics of excess quasiparticles. To do so, we consider N quasiparticles in a volume V . We assume $N/V \ll \nu\Delta$, where ν is the normal density of states and Δ is the superconducting gap. This condition ensures that the quasiparticles occupy a tiny fraction of the available states within the cluster's volume. (The results presented below remain well within this regime.) We also assume that the diffusion of the particles is sufficiently fast that the spatial structure of their wave functions does not affect the spin dynamics [27] and concentrate on spin effects only. Let us consider clusters that are close to the stable configuration with maximal spin $S = N/2$. We choose the instantaneous spin quantization axis such that $S_z = S$ and describe the cluster's deviation from the maximal spin state with the integer $m = N/2 - S$, $m \ll N, S$, which is the number of flipped spins with respect to the cluster's magnetization direction.

We consider four different processes that can change the state (N, m) of the cluster. (1) Quasiparticle injection: quasiparticles are injected with a rate AV and arbitrary spin. Thus, half of them are aligned with the polarization axis of the existing cluster, whereas half of them are antialigned. If the spin is antialigned, we find that the probability of creating an additional spin flip, $m \rightarrow m + 1$, is $(N - m)/(N - m + 1)$. The possible processes are thus $(N, m) \rightarrow (N + 1, m)$ with rate $AV[1 + 1/(N - m + 1)]/2$ and $(N, m) \rightarrow (N + 1, m + 1)$ with rate $AV[1 - 1/(N - m + 1)]/2$. (2) Singlet annihilation: such annihilation processes are possible only if the system is not in a maximal spin state. At a small concentration of spin flips, $m \ll N$, the corresponding rate is, thus, proportional to m . In particular, the process $(N, m) \rightarrow (N - 2, m - 1)$ happens with rate $\bar{\Gamma}(N - m)m/V$. (3) Spin flips: spin-orbit coupling admits for inelastic spin-flips via the electron-phonon interaction. We assume that each spin may flip independently. As for the injection process, a spin flip does not necessarily change the total spin; however, we will neglect the corresponding $1/N$ -corrections to the rates. The rate for the process $(N, m) \rightarrow (N, m + 1)$ is then given as $(N - m)/\tau_s$, where $1/\tau_s$ is the spin flip rate for a single spin. Similarly the process $(N, m) \rightarrow (N, m - 1)$ has the rate m/τ_s . And (4) triplet annihilation: in the presence of spin-orbit coupling, pairs of quasiparticles may annihilate even when they are in a spin-triplet state. To account for such processes, we introduce a weak spin-independent annihilation, $\bar{\Gamma}_t \ll \bar{\Gamma}$. Taking into account all possible orientations of the spins of the annihilated particles, this adds the following processes: $(N, m) \rightarrow (N - 2, m)$ with rate $\bar{\Gamma}_t(N - m)^2/(2V)$ as well as $(N, m) \rightarrow (N - 2, m - 1)$

with rate $\bar{\Gamma}_t(N - m)m/V$ and $(N, m) \rightarrow (N - 2, m - 2)$ with rate $\bar{\Gamma}_t m^2/(2V)$.

With this, the dynamics are described by a master equation explicitly given in Sec. II of the Supplemental Material [26]. As a first step, we derive the mean field solutions for the most probable N and m . The evolution equations for these quantities read:

$$\frac{dN}{dt} = AV - 2\frac{\bar{\Gamma}}{V}(N - m)m - \frac{\bar{\Gamma}_t}{V}N^2, \quad (1)$$

$$\begin{aligned} \frac{dm}{dt} = & \frac{AV}{2} \left(1 - \frac{1}{N - m} \right) - \frac{\bar{\Gamma}}{V}(N - m)m \\ & - \frac{\bar{\Gamma}_t}{V}Nm + \frac{1}{\tau_s}(N - 2m). \end{aligned} \quad (2)$$

Assuming $m \ll N$, Eq. (1) yields the stationary solution

$$m_0 = \frac{AV^2 - \bar{\Gamma}_t N_0}{2\bar{\Gamma}N_0}. \quad (3)$$

Substitution into Eq. (2) gives an equation for the average N in the cluster:

$$0 = \frac{AV}{N_0} - \frac{2}{\tau_s}N_0 - \frac{\bar{\Gamma}_t}{V}N_0^2. \quad (4)$$

We can distinguish two regimes, depending on whether spin relaxation (SR) or triplet annihilation (TA) dominates. In the SR regime, Eq. (4) yields $N_0^{(s)} = \sqrt{AV\tau_s/2}$, while in the TA regime, one finds $N_0^{(t)} = (AV^2/\bar{\Gamma}_t)^{1/3}$. The corresponding values for m are given as $m_0^{(s)} = (V/\bar{\Gamma}\tau_s)N_0^{(s)}$ and $m_0^{(t)} = [(AV^2\bar{\Gamma}_t^2)^{1/3}/2\bar{\Gamma}]N_0^{(t)}$, respectively. Comparing the two expressions for N_0 , we conclude that the TA regime requires $A > V/(\bar{\Gamma}_t^2\tau_s^3)$, that is, a sufficiently high injection rate at any given volume.

The above equations allow us to derive the requirements for the cluster to be highly polarized, that is, $N_0 \gg m_0$. Let us first consider a small A such the cluster is in the SR regime. In this case, a sufficiently small volume $V \ll V_c \equiv \bar{\Gamma}\tau_s$ is required. If at a given $V < V_c$ we increase A , and therefore the number of particles in the cluster, we cross-over to the TA regime, and a high polarization persists up to $A \simeq A_c(V_c/V)^2$ with $A_c \equiv \bar{\Gamma}/(\bar{\Gamma}_t\tau_s)^2$. This requirement is convenient to express in terms of the number of particles in the cluster, $N \lesssim N_c \equiv \bar{\Gamma}/\bar{\Gamma}_t$ [28].

Note that, in a polarized cluster, N_0 largely exceeds the value $N_{\text{unpol}} = c_0V$ expected for an unpolarized system. It is constructive to express the concentrations as follows: in the SR regime,

$$N_0^{(s)}/V = \frac{c_0}{2}\zeta_s^{-1/2}, \quad m_0^{(s)}/V = \frac{c_0}{2}\zeta_s^{1/2}, \quad (5)$$

with $\zeta_s \equiv V/V_c \ll 1$, and in the TA regime,

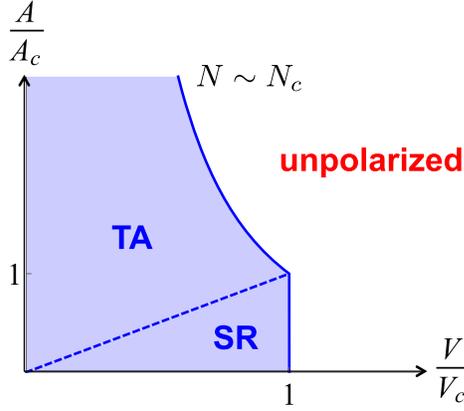


FIG. 1. Unpolarized and polarized regimes versus the cluster volume and the injection rate, according to Eqs. (5) and (6). Here $V_c = \bar{\Gamma}\tau_s$, $A_c = \bar{\Gamma}/(\bar{\Gamma}_t\tau_s)^2$, and $N_c = \bar{\Gamma}/\bar{\Gamma}_t$.

$$N_0^{(t)}/V = \frac{c_0}{2}\zeta_t^{-1/2}, \quad m_0^{(t)}/V = \frac{c_0}{2}\zeta_t^{1/2}, \quad (6)$$

with $\zeta_t \equiv (AV^2/A_cV_c^2)^{1/3}/2 \ll 1$. The regions where a polarized state is expected are illustrated in Fig. 1, see also Sec. III of the Supplemental Material [26] for more details.

Let us estimate the relevant material parameters $\bar{\Gamma}$, $\bar{\Gamma}_t$, and τ_s . In aluminum, the phonon-assisted recombination rate for quasiparticles near the gap edge is characterized by $\bar{\Gamma} \simeq 18 \text{ s}^{-1} \mu\text{m}^3$ [29]. As to the triplet annihilation rate, it involves a phonon emission accompanied by a spin-flip, and is estimated as $\bar{\Gamma}_t \sim \alpha_{\text{so}}^2 K \bar{\Gamma}$, where $\alpha_{\text{so}} \sim 10^{-2}$ is the dimensionless spin-orbit strength, and the suppression factor K reflects the smallness of the momentum transfer in the course of the emission. As such, K crucially depends on the wave vector q of the phonon involved that is set by the energy $\sim \Delta$ released, $cq \simeq \Delta$, c being the sound velocity. In the absence of disorder, $K \simeq (qa)^2$ [30], a being the interatomic distance scale. With the disorder setting a mean free path l , $K \simeq (ql)^{-1}$ for $1 \lesssim ql \lesssim (l/a)^{2/3}$, $K \simeq ql$ for $q \lesssim l^{-1}$ [31]. To have a disorder-independent estimation, we resort to the least suppressed case, $K = 1$. This gives $\bar{\Gamma}_t \sim \alpha_{\text{so}}^2 \bar{\Gamma} \sim 10^{-4} \bar{\Gamma}$.

It may seem that the relevant spin-flip rate is determined by elastic spin-orbit processes as it is usual in the context of spin transport [32], $1/\tau_{\text{so}} \sim \alpha_{\text{so}}^2 (\delta\epsilon/\Delta)^{1/2}/\tau_{\text{el}}$, where τ_{el} is the elastic scattering time, and $\delta\epsilon \lesssim \Delta$ characterizes the energy window for the excess quasiparticles above the superconducting gap [33], which is sensitive to the temperature. However, this estimation holds for propagating electron waves rather than for the localized states we are dealing with. As explained in [34], elastic spin-orbit interaction is inefficient in relaxing the spin of localized states, not lifting the Kramers degeneracy. Therefore the spin flips should involve inelastic processes. We assume that the dominant spin-flip process is the phonon emission or absorption in the presence of spin-orbit

coupling. The corresponding rate is then estimated as $1/\tau_s \sim \alpha_{\text{so}}^2 (\delta\epsilon/\Delta)^{7/2} K/\tau_0$, where $\tau_0 \sim 400 \text{ ns}$ in Al is the normal-state inelastic phonon scattering time at energy $\sim \Delta$ [23]. The first and second suppression factors reflect the smallness of the spin-orbit interaction and the reduction of the density of states [23], and the factor K now corresponds to the energy transfer $\delta\epsilon \simeq cq$. As above, we resort to the least suppressed choice $K = 1$. Even this choice gives very long spin-flip times: at $\delta\epsilon \simeq 0.1\Delta$ we estimate $\tau_s \simeq 10 \text{ s}$.

With this, we estimate the critical volume $V_c = \tau_s \bar{\Gamma} \sim 180 \mu\text{m}^3$. This implies that the spin-polarized cluster can be spread over micron lengths and V_c is not a very restrictive parameter. In particular, we do not expect our results to be very sensitive on the temperature in the currently studied aluminum-based devices. A more severe restriction comes from the triplet annihilation that sets the maximum number of particles in the cluster, $N_c = \bar{\Gamma}/\bar{\Gamma}_t \sim 10^4$. The critical injection rate, where the crossover from spin-flip limited to triplet-annihilation limited clusters size takes place, is then estimated as $A_c \sim 10^5 \text{ s}^{-1} \mu\text{m}^{-3}$. (A similar injection rate was reported in Ref. [17].) The quasiparticle density is enhanced compared to the unpolarized case, if $V < V_c$ and $A < A_c(V_c/V)^2$.

It is important to note that the number of particles in the cluster strongly fluctuates. The mean-field solution gives the most probable number of particles in the cluster, N_0 , while $\langle N \rangle$ differs from N_0 by a factor and the fluctuations $\langle\langle N^2 \rangle\rangle = \langle N^2 \rangle - \langle N \rangle^2$ are of the order N_0^2 . To quantify the fluctuations, we utilize a Fokker-Planck equation, cf. Sec. II of the Supplemental Material [26], which gives the distribution function

$$P(N) = CN^2 \exp \left[-\frac{2\bar{\Gamma}_t}{3AV^2} N^3 - \frac{2}{AV\tau_s} N^2 \right], \quad (7)$$

where the constant C ensures the normalization. For the SR and TA regimes, this gives, respectively,

$$\langle N \rangle^{(s)} = \frac{2}{\sqrt{\pi}} N_0^{(s)}, \quad \langle N^2 \rangle^{(s)} = \frac{3}{2} (N_0^{(s)})^2, \quad (8)$$

$$\langle N \rangle^{(t)} = \frac{\Gamma(\frac{1}{3})}{18^{1/3}} N_0^{(t)}, \quad \langle N^2 \rangle^{(t)} = \frac{2^{4/3} \pi}{3^{5/6} \Gamma(\frac{1}{3})} (N_0^{(t)})^2. \quad (9)$$

A comparison with the classical model [25] is provided in Sec. IV of the Supplemental Material [26].

While the quasiparticle system is polarized, its spin quantization axis is not fixed but diffuses with time, along with the number of the polarized particles. This produces a measurable spin noise that can be utilized for the experimental verification of the polarization, using the setup sketched in Fig. 2 as explained below. The spin noise for a certain spin component can be estimated in terms of the noise of the number of particles S_N , $S_{\text{spin}} \simeq \frac{1}{3} S_N \simeq \langle\langle N^2 \rangle\rangle t_f$.

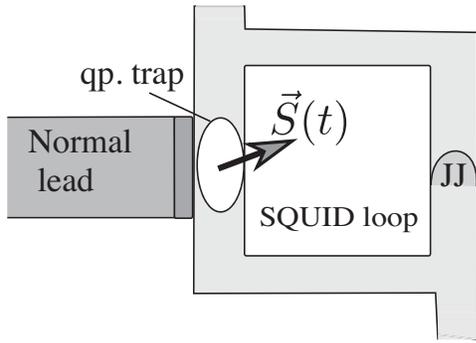


FIG. 2. Experimental setup for the detection of the polarized state. The quasiparticles are injected from a normal lead into a trap embedded in the arm of a SQUID loop. The fluctuations of their common spin produce in the loop the flux noise to be measured. The polarization is seen as an enhanced noise $\propto N_0^2$.

Here, t_f is a characteristic timescale for the fluctuations, which is estimated as $t_f \simeq N_0(N_0/AV)$; it yields $t_f \simeq \tau_s$ and $t_f \simeq (A\bar{\Gamma}_t/V)^{-1/3}$ in the SR and TA regimes, respectively. We have evaluated numerically the particle number zero-frequency noise in these two regimes to find $S_N(0) = 0.5\langle\langle N^2 \rangle\rangle\tau_s$ and $S_N(0) = 0.6\langle\langle N^2 \rangle\rangle(A\bar{\Gamma}_t^2/V)^{-1/3}$ where the variances $\langle\langle N^2 \rangle\rangle$ in the regimes are given by Eqs. (8) and (9).

A flux noise of substantial amplitude $S_\Phi \simeq 10^{-12}\Phi_0^2/\text{Hz}$ at low frequencies is routinely measured in superconducting quantum interference devices (SQUIDs), here Φ_0 is the flux quantum. This noise limits the performance of superconducting qubits, that motivated its thorough investigation [35–37]. Nowadays its origin is commonly attributed to the slow dynamics of localized spins at the surface of a superconductor [38–40]. We note that the spins of nonequilibrium quasiparticles may also contribute to this noise. In fact, the polarization mechanism predicted in this Letter make these spins very effective noise sources: N quasiparticle spins combined in a polarized cluster produce the same noise as N^2 localized spins, provided the timescale of their dynamics is the same. In distinction from localized spins, the quasiparticles can be brought to the superconductor in a controllable way, for instance, by injection through a normal lead separated from the superconductor by a tunnel barrier [41].

This leads us to the suggestion of a concrete experimental setup to observe the predicted polarized state. As depicted in Fig. 2, one makes a quasiparticle trap embedded in the arm of a superconducting loop by reducing the superconducting gap locally, and injects the quasiparticles into the trap from a normal lead that is biased at a voltage that slightly exceeds the reduced gap. The flux noise is monitored at different injection rates corresponding to different quasiparticle numbers N_0 . Assuming a width of 100 nm for the SQUID arm, one spin induces a flux $\simeq 10^{-7}\Phi_0$ through the SQUID loop [35]. For the following estimations, we assume that triplet annihilation dominates,

$N_0 = 10^4$, and the trap volume is $(100 \text{ nm})^3$. At these conditions, $t_f \simeq 0.5 \times 10^{-4} \text{ s}$, and the fluctuations of the polarized state produce the noise $S_\Phi \simeq 10^{-12}\Phi_0^2/\text{Hz}$, that exceeds the commonly observed level. If the particles were not polarized, the flux noise would be four orders of magnitude lower. An advantage of the setup is that the number of quasiparticles induced, as well as the fluctuations of this number, can be monitored through the high-frequency inductance and inductance noise of the superconducting sample [42].

In this work, we assume that a possible external magnetic field does not polarize the quasiparticle spins. This is valid provided the corresponding Zeeman energy $E_Z \ll \delta\epsilon$. On the level of the master equation, the polarizing effect of the magnetic field can be taken into account by assigning an anisotropy to the spin relaxation, but we have not investigated this.

In conclusion, we propose a novel scenario for spontaneous spin polarization of a finite system under out-of-equilibrium conditions. We predict that, owing to the spin selectivity of recombination, the excess quasiparticles in a superconductor may spontaneously polarize in clusters. The underlying mechanism differs from that considered in Ref. [43] for homogeneous quasiparticle states. For parameters of Al, such a polarized cluster may contain 10^4 quasiparticles and spread over microns. We show that the polarization can be detected as an excess flux noise.

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