PHYSICAL REVIEW B VOLUME 62, NUMBER 10 1 SEPTEMBER 2000-II

Spin polarized carrier injection into high-T_c superconductors: A test for the superconductivity mechanism

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(Received 8 May 2000; revised manuscript received 5 July 2000)

We point out in this short paper, that be it an *s*-wave or *d*-wave superconductor, in a nonequilibrium situation (i.e., in the presence of excess unpolarized and polarized quasiparticles maintained by an injection current) the superconducting gap suppression by the presence of the same amount of excess quasiparticles, can at best differ by a factor of 2, for a conventional BCS superconductor. For the high-T_c superconductors on the other hand, there is a huge difference in gap suppression between unpolarized and polarized quasiparticle injection, as observed in the experiments [V. A. Vasko *et al.*, Phys. Rev. Lett. **78**, 1134 (1997); T. A. Venkatesan *et al.*, Appl. Phys. Lett. **71**, 1718 (1997)]. We argue that this large anomaly has a natural explanation within the *interlayer tunneling mechanism* of Wheatley, Hsu, and Anderson [J. M. Wheatley, T. C. Hsu, and P. W. Anderson, Phys. Rev. B **37**, 5897 (1988)], and is due to the excess polarized quasiparticles blocking the interlayer pair tunneling process. We also point out that spin polarized quasiparticle injection in a superconductor is a very easy way to distinguish between an *s*-wave or an anisotropic gap superconductor.

Spin polarized electron tunneling from ferromagnetic metals to superconductors[1] or strongly correlated metals has recently generated much interest. The focus is on trying to understand spin-dependent transport properties of electrons, spin relaxation, and possible superconducting devices. One is also interested in the mechanisms of superconductivity suppression (such as reduction in critical currents) due to tiny injected tunneling currents (unpolarized and polarized) across an insulating junction into the superconductor.

Though original experiments on spin polarized tunneling in superconductors were carried out in the 1970s by Tedrow and Meservey[2] and a theoretical attempt was made by Aronov,[3] recently this subject has caught the attention of the physics community due to the discovery of lanthanam manganite (CMR materials). In these materials the degree of polarization of the charge carriers is close to unity (almost perfect ferromagnetic metal).

Both of these experiments[1] are done in the three-layer structurelike, HTSC/I/manganite, where a high-T_c superconductor (HTSC) thin film is separated from a ferromagnetic metallic underlayer (manganite) by an insulating (I) layer. The critical current of the HTSC film is found to drop precipitously as a function of a tiny injection current pushed from the ferromagnetic metal through the insulating junction into the HTSC film. When manganite is replaced by Au (an ordinary paramagnetic metal), then it is found that for the same amount of injected current the drop in the critical current of the HTSC film is much smaller. The fractional change in critical current for polarized and unpolarized injection currents (over the same injection current interval) differs by an order of magnitude.

In this paper we try to show that the large difference between polarized and unpolarized injection currents is very special only to the high-T_c superconductors. We predict that if the high-T_c superconductor is replaced by an ordinary BCS superconductor then the relative difference in superconducting gap suppression in the two cases is not at all that striking (differs by at most a factor of 2). This is our main result. In other words spin polarized electron tunneling into high-T_c superconductors can be used to understand the mechanism of superconductivity in the high-T_c materials.

The suppression of the superconducting gap due to unpolarized quasiparticle overpopulation (a nonequilibrium situation) was investigated theoretically by Owen and Scalapino.[4] They examined a simple model of an electron gas containing both Cooper pairs and excited quasiparticles with the ratio of paired to unpaired electrons being artificially specified, instead of being uniquely determined by the temperature, as is usual, in the thermal equilibrium situation for an isolated superconductor. The system is considered as being in thermal equilibrium although the paired and unpaired electrons are not in chemical equilibrium. It is assumed that for a small amount of excess injected carriers, the time for pair recombination is much larger than the time for the injected quasiparticles to thermalize with the lattice. This is achieved by introducing a chemical potential for the quasiparticles different from the pair chemical potential.

The free energy of the superconductor F_s is given by

$$F_s=2\sum_k |(\epsilon_k-\mu)|(f_k-2f_kv_k^2+v_k^2)-\sum_{k,k'} V_{k,k'}u_kv_ku_{k'}v_{k'} \times(1-f_k)(1-f_{k'})-T\sum_k [f_k\ln f_k+(1-f_k)\ln(1-f_k)], \tag{1}$$

where u_k and v_k have their usual meaning, and $f_k=\langle \gamma^\dagger_{k,\sigma}\gamma_{k,\sigma}\rangle$ is the Fermi function for the superconducting quasiparticles and ϵ_k is the dispersion of the electrons in the normal state. However, in addition to the usual constraint equation for the total number of electrons $\Sigma_{k,\sigma}\langle c^\dagger_{k,\sigma}c_{k,\sigma}\rangle=N$ which is enforced by proper choice of the chemical potential μ, an additional constraint on quasiparticle excitation number will be imposed, $\Sigma_{k,\sigma}f_{k,\sigma}=n\ll N$. This can be done by introducing an extra chemical potential[5] μ^* so that now $f_k = [1 + e^{\beta(E_k - \mu^*)}]^{-1}$. The modified BCS gap equation is now,

$$\Delta_k = \sum_{k'}{}' \frac{V_{k,k'}\Delta_{k'}}{2E_{k'}}(1 - f_{k'\uparrow} - f_{-k'\downarrow})$$

$$= \sum_{k'}{}' \frac{V_{k,k'}\Delta_{k'}}{2E_{k'}} \tanh \beta/2(E_{k'} - \mu^*). \tag{2}$$

Assuming a momentum independent $V_{k,k'}$ (s-wave superconductor) and going from momentum summation to energy integration with a cutoff ω_D, we get

$$\frac{1}{N(E_F)V} = \int_{-\omega_D}^{\omega_D} \frac{d\epsilon}{2E} \tanh \frac{1}{2}\beta(E - \mu^*).$$

The number of excess quasiparticles is

$$n = 2\sum_k [f(E_k - \mu^*) - f(E_k)] = 4N(E_F)\int_0^\infty \frac{d\epsilon}{e^{\beta(E-\mu^*)} + 1}. \tag{3}$$

The factor 2 before the momentum summation is due to two spin species.

Defining $n_0 = n/4N(E_F)\Delta_0$, where Δ_0 is the gap at $T = 0$ and $\mu^* = 0$, we can easily solve for the gap value at any T and n from the above two equations. In the limit of zero temperature the gap value can be determined from the algebraic equation,

$$\Delta_0/\Delta = [n_0\,\Delta_0/\Delta + \sqrt{1 + n_0^2\,\Delta_0^2/\Delta^2}]^2. \tag{4}$$

This is the result of Owen and Scalapino, for excess unpolarized quasiparticles.

For spin polarized quasiparticle injection, we introduce a different chemical potential, for *only the up-spin quasiparticles,* i.e., assuming complete spin polarization of electrons in the ferromagnet. The gap equation (2) will be modified to

$$\Delta_k = \sum_{k'}{}' \frac{V_{k,k'}\Delta_{k'}}{2E_{k'}} \frac{1}{2}[\tanh \beta/2(E_{k'} - \mu^*) + \tanh \beta/2E_{k'}]. \tag{5}$$

The corresponding equation for the number of excess quasiparticles will be the same as Eq. (3) with the factor 2 missing before the momentum summation, because excess quasiparticles are of only one spin species. In the limit of zero temperature we get the following algebraic equation for the normalized gap value,

$$\Delta_0/\Delta = [2n_0\,\Delta_0/\Delta + \sqrt{1 + 4n_0^2\,\Delta_0^2/\Delta^2}]^2. \tag{6}$$

In Fig. 1 we show the normalized gap versus extra quasiparticles for an s-wave superconductor [solutions of Eqs. (3) and (4)]. We find that for s-wave superconductors, a first-order phase transition to normal metal occurs, for $n = 0.15$, $\Delta = 0.63\Delta_0$, and $n = 0.08$, $\Delta = 0.58\Delta_0$ for unpolarized and polarized quasiparticle injections. Beyond this critical concentration of injected carriers, the free energy of the perturbed superconductor becomes larger than normal-state free energy. Notice though that, for the same amount of injected carriers, the relative gap suppression in unpolarized and polarized carriers differs by at most by a factor of 2.

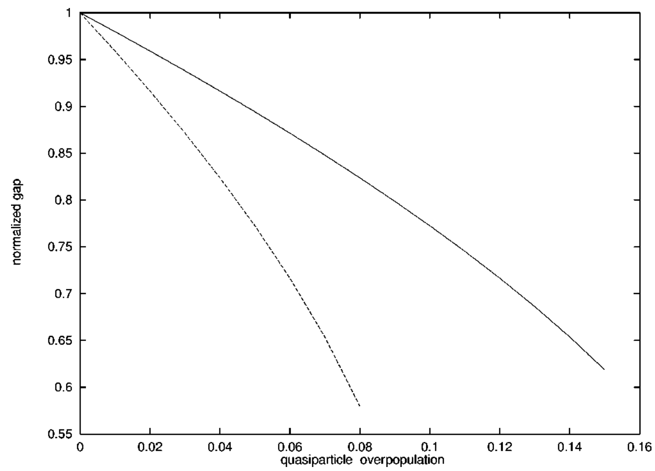

FIG. 1. $\Delta(T=0, n_0)/\Delta(T=0, n_0=0)$ versus n_0 (the quasiparticle overpopulation, as defined in the text) for an s wave superconductor [the solutions of Eqs. (3) and (4)].

Just as at finite temperatures quasiparticle excitations interfere with the pairing interaction and eventually destroy superconductivity at T_c, when an excess number of quasiparticles are injected into the superconductor, it basically reduces the phase space for the BCS pair scattering process and reduces the gap value. BCS interaction scatters pairs $(k\uparrow, -k\downarrow) \rightarrow (k'\uparrow, -k'\downarrow)$ across the Fermi surface. So any excess quasiparticle occupying these states limits the phase space for the BCS interaction. It is obvious that when the injected quasiparticles are polarized (all of one spin) they interfere with BCS interaction more severly and hence the gap value falls faster with quasiparticle overpopulation, compared to the unpolarized injection current.

We next investigate the effect of quasiparticle injection on superconductors with anisotropic gaps, specifically gaps of d-wave symmetry. The dispersion of electrons is chosen to be of the form

$$\epsilon(k) = -2t(\cos k_x + \cos k_y) + 4t' \cos k_x \cos k_y$$

$$-2t''(\cos 2k_x + \cos 2k_y), \tag{7}$$

with $t = 0.25$ eV, $t'/t = 0.45$, $t''/t = 0.2$. We also choose $\epsilon_F = -0.45$ eV corresponding to a Fermi surface which is closed around the Γ point. These choices are inspired by band-structure calculations[6] for the Y-Ba-Cu-O compound at optimal doping concentration. The cutoff of the pairing interaction is chosen to be $\omega_D = 30$ meV, and the pairing interaction strength $V_{k,k'} = V_0 f_k f_{k'}$ with $f_k = \cos(k_x) - \cos(k_y)$ and $V_0 = 2.8$ meV. With this choice of parameter the T_c comes out to be 30 K.

In Fig. 2 we plot the normalized momentum averaged value of the superconducting gap magnitude, $\Delta(n,T)/\Delta(0,T)$ versus n (quasiparticle overpopulation) for different temperatures. There are some interesting features to be noticed here.

(1) Notice the crossing of curves, for $T = 5$ and 25 K. The origin of this can be seen by looking at the gap equation (2). States of energy E_k less than μ^* interfere with the pairing process by giving a negative contribution to the binding. At

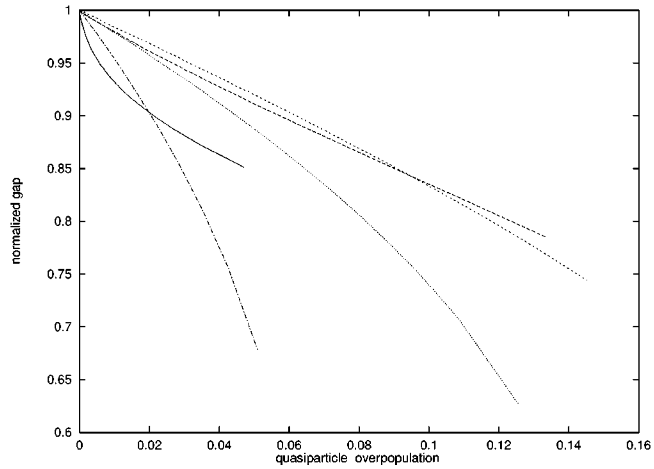

FIG. 2. The normalized momentum averaged gap $\Delta(T,n)/\Delta(T,n=0)$ of a d-wave superconductor, with $T_c=30$ K, versus injected carrier concentration. The solid line is for $T=5$ K, long dashes for $T=10$ K, short dashes for $T=15$ K, dotted line for $T=20$ K, and the dash-dotted line is for $T=25$ K.

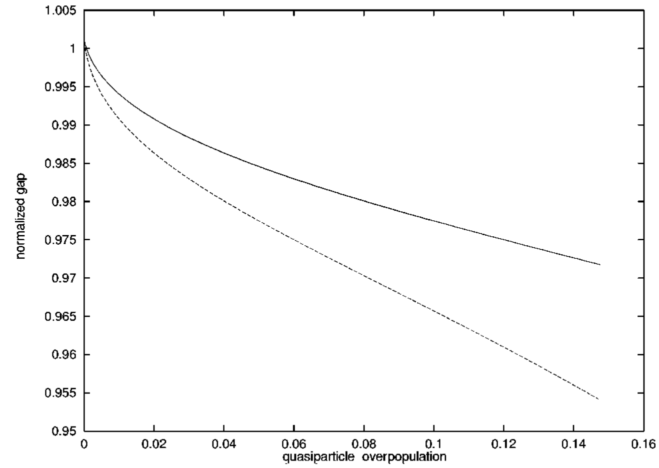

FIG. 3. The normalized momentum averaged gap $\Delta(T,n)/\Delta(T,n=0)$ of a d-wave superconductor, with $T_c=90$ K, versus injected carrier concentration at $T=10$ K. The solid and dashed lines are for unpolarized and polarized injected carriers.

low temperatures and small injection currents, the extra carriers occupy the states near the deep gap nodes, where the denominator E_k is also small giving rise to large reduction in binding energy. At larger temperatures (for the same amount of injected carriers) on the other hand, the quasiparticles are distributed over a larger range of energy (larger denominator) leading to smaller suppression of binding (and hence larger gap values). A larger concentration of injected carriers, of course, will ultimately be more damaging to superconductivity, because of the larger number of thermally excited quasiparticles that are already present. This leads to the crossing of curves as seen in Fig. 2. This will be a generic feature for any superconductor with deep gap nodes, and should be easily seen in spin injection experiments.

(2) Notice also that the critical concentration of excess quasiparticles that destroy superconductivity is not a monotonic function of temperature and goes through a maximum around $T_c/2$.

(3) Though it is not shown in the plot, injected current suppresses superconductivity more in the d-wave superconductors than for an s-wave superconductor having the same critical temperature, as one would expect for gap functions with deep nodes.

In Fig. 3, we have plotted the normalized momentum averaged gap magnitude of a d-wave superconductor, with a T_c of 90 K versus both unpolarized and polarized quasiparticle overpopulation (we go up to $n=0.15$ only). The difference in the reduction of gap values in the unpolarized and polarized tunneling is in the range of $2.5-5.0\,\%$ (i.e, a factor of 2 only). Clearly the large anomaly observed while tunneling into the high-T_c superconductor, as seen in the experiments,[1] has its origin in the superconducting mechanism in the high-T_c materials itself.

We shall explore here the *interlayer tunneling model* of superconductivity.[7,8]

We begin by writing the gap equation for interlayer tunneling model with unpolarized injected carriers as[8]

$$\Delta_k=T_J(k)\frac{\Delta_k}{2E_k}\tanh\frac{\beta(E_k-\mu^*)}{2}+\sum_{kk'}{}' V_{kk'}\frac{\Delta'_k}{2E_{k'}}\tanh\frac{\beta(E_{k'}-\mu^*)}{2}. \tag{8}$$

For polarized carrier injection, this equation has to be corrected as shown in Eq. (5).

This gap equation can be obtained by considering two close Cu-O layers as in Y-Ba-Cu-O coupled by a Josephson tunneling term of the form

$$H_J=-\frac{1}{t}\sum_k t_\perp^2(k)(c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger d_{-k\downarrow}d_{k\uparrow}+\text{H.c.}),$$

where $t_\perp(k)$ is the bare single-electron hopping term between the two coupled layers c and d and t is a band-structure parameter in the dispersion of electrons along the Cu-O plane. Finally, $T_J(k)$ in the right-hand side of Eq. (1) is given by $T_J(k)=t_\perp^2(k)/t$, where $t_\perp(k)=(t_\perp/4)[\cos(kx)-\cos(ky)]^2$. The dispersion of electrons along the Cu-O plane is given by Eq. (5). Note that the Josephson coupling term in H_J conserves the individual momenta of the electrons that get paired by hopping across the coupled layers. This is as opposed to a BCS scattering term which would only conserve the center-of-mass momenta of the pairs. This is the origin of all features that are unique to the interlayer tunneling mechanism. This term has a local U(1) invariance in k space and cannot by itself give a finite T_c. It needs an additional BCS-type nonlocal interaction in the planes which could be induced by phonons or residual correlations. Here we assume the in-plane pairing interaction to be d-wave kind, i.e, $V_{kk'}=V_0 f_k f_{k'}$ with $f_k=\cos k_x-\cos k_y$. $T_J(k)$ can be inferred from electronic structure calculations. As shown in Ref. 6, it is adequate to choose $t_\perp(k)=(t_\perp^2/4)(\cos k_x-\cos k_y)^2$, with $t_\perp/t\equiv 1/3$ to 1/5. According to Anderson, it is the k-space locality that leads to a scale of T_c that is linear in the interlayer pair tunneling matrix element. He finds that in the limit $T_J>V_{kk'}$, $k_BT_c\approx T_J/4$ and in the other limit,

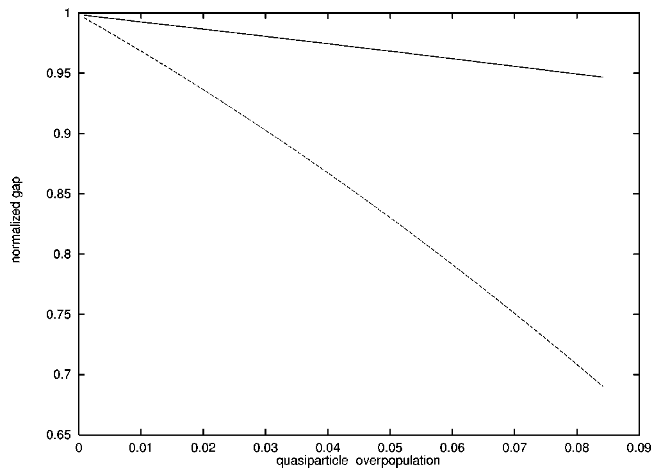

FIG. 4. The normalized momentum averaged gap $\Delta(T,n)/\Delta(T,n=0)$ of a d wave superconductor+interlayer tunneling, with $T_c=90$ K, versus injected carrier concentration at $T=10$ K. The solid and dashed lines are for unpolarized and polarized injected carriers.

$k_BT_c \approx \hbar\omega_D e^{-1/\rho_0 V_0}$, where ω_D, ρ_0, and V_0 are Debye frequency, density of states at the Fermi energy, and Fermi-surface average matrix element $V_{kk'}$. The important point is that even with a little help from the in-plane pairing interaction the interlayer tunneling term can provide a large scale of T_c. In this model the gap value is mainly controlled by the interlayer tunneling amplitude, rather than the in-plane pairing interaction, though the symmetry of the gap function is determined overwhelmingly by the in-plane BCS kind of interaction strength $V_{k,k'}$, and the gap value is very sensitive to the interlayer pair tunneling amplitude.[9]

In Fig. 4, we plot the normalized momentum averaged gap values versus both unpolarized and polarized quasiparticle overpopulation up to $n=0.085$. We find that for $n=0.085$, the reduction in gap value for unpolarized injection current is about 5%, whereas for the polarized carrier injection it is about 35% (that is a factor of 7).

The large difference between the two situations (as observed in the experiments[1] also) now shows up. We argue that when polarized carriers are injected, then, over and above the usual dynamical pair breaking (due to phase space blocking of BCS interaction) in the planes like in usual BCS superconductors, there is an added inhibition of interlayer pair tunneling. This is so because there are many fewer extra singlets near the Fermi surface, to tunnel from plane to plane. This effect is absent when unpolarized quasiparticles enter the planes. In this mechanism, *extra quasiparticles directly affect the interlayer pairing tunneling process, which is the main source of superconducting condensation energy gain*. This is over and above the interference in the binding process in the individual layers that we have discussed earlier in usual BCS superconductors.

The introduction of an extra chemical potential μ^* to tackle the nonequilibrium superconductivity, in the presence of artificially maintained quasiparticles, is a reasonable starting point, when the excess quasiparticles thermalize with low energy phonons more often than they recombine into pairs. In this limit the quasiparticles are in a steady state at T but have an excess number denoted by the increased chemical potential μ^*. One important ingredient of the mechanism of Anderson and co-workers[7] is that in the normal state the electrons are spin-charge separated and no quasiparticles in the Fermi liquid sense exist. We have assumed here that below the superconducting T_c somehow spin-charge separation is absent. This seems to be the case from photoemission experiments which show a clear signal of well-defined quasiparticles on the Fermi surface. In the superconducting state how exactly it happens is still not clear.

Throughout this analysis, we have assumed the following: (1) The London penetration depth is less than the superconducting film thickness, so that direct magnetic field of the ferromagnetic metal does not penetrate the superconductor much. (2) The spin diffusion length is much larger than the SC film thickness, so that no spatial variation of the gap has to be taken into account. This is certainly true when there is no magnetic impurity or for small spin orbit interaction, because the then extra spin density relaxes slowly. We have also not taken into account the case of finite recombination time for the excess quasiparticles.

Recently Yeh *et al.*,[10] have observed an initial, actual increase in critical current for low enough injection currents, when the insulating barrier thickness is small. They argue that some up-spin quasiparticles in the superconductors can diffuse into the magnetic materials, creating spin imbalance in the superconductor (more of down-spin quasiparticles). On the other hand, when the injection current is switched on, then up-spin electrons start coming into the superconductor, nullifying the spin imbalance in the superconductor. That is why T_c increases for small injection currents. For larger injection currents, of course, T_c falls drastically as is seen in experiments, and as we have argued to be natural within the interlayer tunneling mechanism of Anderson and co-workers.

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