Spin injection and polarization of excitations and nuclei in superconductors

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A spin density flux that leads to polarization of excitations and nuclei in a superconductor arises when a current is passed through a ferromagnet–superconductor tunnel junction. The stationary states produced by spin injection and the paramagnetic resonance with the excitations in the absence of an external field are investigated. It is shown that the plots of the degree of polarization of the nuclei and of the intensity of the ESR signal against the current reveal the presence of hysteresis.

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Spin electron resonance in bulky superconductors is impossible because of the small depth of penetration of the magnetic field and of its strongly inhomogeneous distribution near the boundary, meaning that the Overhauser effect and the effect of dynamic polarization of the nuclei are impossible. The possibility of polarization of excitations and of nuclei in superconductors is, however, very attractive, since it would make it possible to observe many new phenomena, nuclear magnetic resonance, and even electron spin resonance in bulky samples. This paper considers a new method of polarizing excitations and nuclei in superconductors.

Pikus and the author[1] have shown that when current passes through a junction between a ferromagnet and a semiconductor, the electron spins in the semiconductor become polarized. The sign of the polarization depends on the direction of the current. It is natural to assume that a similar effect can occur also when current flows through a ferromagnet–superconductor junction.

Consider a ferromagnet–superconductor tunnel junction. Let the ferromagnet be magnetized in such a way that the magnetization vector in entire sample has a single direction and lies in the plane of the junction. If current flows through the junction, then a spin-density flux is produced simultaneously with the current transport. Depending on the direction of the current, this flux is directed into the superconductor or out of the superconductor. The appearance of the spin flux through the boundary leads to the appearance of spin polarization in the superconductor parallel or antiparallel to the direction of the magnetization in the ferromagnet. This polarization of the spins extends into the interior of the junction over the spin diffusion length L_s, which can be much larger than even the diffusion length.

The degree of polarization of the excitations depends essentially on the degree of polarization of the tunnel current through the ferromagnet–superconductor junction. Tedrow and Meservey[3] obtained for different ferromagnets a tunnel-current polarization ranging from 11% for Ni to 34% for Fe. If the ferromagnet is a ferromagnetic semiconductor, then the degree of polarization of the conduction electrons in it, determined by the ratio of the volume splitting to the Fermi energy, can be easily made to reach 100%.

This paper deals with the dependences of the degree of polarization of the excitations on the intensity of the spin pumping and on the parameters of a superconductor, with the polarization of nuclei in a superconductor, and with electron spin resonance in a superconductor without an external magnetic field.

POLARIZATION OF EXCITATIONS IN A SUPERCONDUCTOR

At low temperatures, when the excitation thermalization time is short in comparison with the lifetime,[3] the energy distribution of the excitations is characterized by a quasi-equilibrium distribution with a nonzero chemical potential. The spin relaxation in the superconductor in the absence of paramagnetic impurities is connected with the spin-orbit interaction, and can therefore be very small if the g-factor of the electron in the metal differs little from the factor g_0 of the free electron. This makes it possible, when describing spin injection, to use phenomenological equations for the numbers of the excitations with spins along the magnetization direction (n_+) and against this direction (n_-) in the ferromagnet. The system of equations for n_+ and n_- takes the form

$$\frac{\partial n_+}{\partial t}=-\frac{n_+-n_-}{2\tau_s}-\frac{n_+n_--n_0^2/4}{n_0\tau_R}-\frac{n_+^2-n_0^2/4}{n_0\tau_R^s}+p_+, \tag{1}$$

$$\frac{\partial n_-}{\partial t}=-\frac{n_--n_+}{2\tau_s}-\frac{n_+n_--n_0^2/4}{n_0\tau_R}-\frac{n_-^2-n_0^2/4}{n_0\tau_R^s}+p_-. \tag{2}$$

Here τ_s is the excitation spin relaxation time in the superconductor, n_0 is the equilibrium concentration of the excitations, τ_R is the lifetime of the excitations relative to recombination of two quasiparticles with opposite spins,[1)] τ_R^s is the lifetime of the excitations relative to recombination of two quasiparticles with identical spins, and $p_\pm$ is the rate of pumping of an excitation with up or down spin in tunnel injection.

In superconductors, without allowance for the spin-orbit interaction, particles with identical spin directions cannot recombine. Therefore the time $(\tau_R^s)^{-1}$ contains, in comparison with the time τ_R^{-1}, an additional small quantity $(g-g_0)^2\ll g_0^2$. In Eqs. (1) and (2), we have neglected diffusion, assuming that the sample thickness is smaller than the diffusion length L. This means that the distribution of the spin density and of the excitation concentration is uniform over the sample. The penetration of the magnetic field into the superconductor from the ferromagnet can be disregarded, inasmuch as, firstly, it is directed parallel to the spin polarization and therefore does not lead to depolarization effects, and secondly, it penetrates only to a depth that is small in comparison with all the characteristic lengths. Here $\dot{p}_{\pm}=J_{\pm}/d$, where d is the thickness of the plate and $J_{\pm}$ is the density of the flux of electrons with a given spin through the junction.

From (1) and (2) we easily obtain equations for the concentration of the excitations n and for the polarization s. If we introduce $n_{\pm}=\frac{1}{2}(n+s)$, then

$$\frac{\partial n}{\partial t}=-\frac{n^2-n_0^2}{2n_0}\frac{1}{\tau_R}+\frac{s^2}{2n_0}\frac{1}{\tau_R}+\dot{n}, \tag{3a}$$

$$\frac{\partial s}{\partial t}=-\frac{s}{\tau_s}-\frac{n}{n_0}\frac{s}{\tau_R^s}+\dot{s}, \tag{3b}$$

where $\dot{n}=\dot{p}_{+}+\dot{p}_{-}$, and $\dot{s}=\dot{p}_{+}-\dot{p}_{-}$.

We have neglected τ_R^s in the derivation of the system (3). At not too strong pumping, we can neglect also the second term in the right-hand side of (3b), since $\tau_s/\tau_R^s\sim\tau_p/\tau_R\ll 1$, where τ_p is the momentum relaxation time. The system of equations can be easily solved in the stationary case:

$$n^2-n_0^2=2\tau_R n_0\dot{n}+(\dot{s}\tau_s)^2, \tag{4}$$

$$s=\dot{s}\tau_s. \tag{5}$$

The degree of polarization of the excitations is therefore

$$p_s=\frac{s}{n}=\frac{\dot{s}\tau_s}{n_0}\left[1+\frac{2\tau_R\dot{n}}{n_0}+\left(\frac{\dot{s}\tau_s}{n_0}\right)^2\right]^{-1/2}. \tag{6}$$

It is seen from (6) that if the spin relaxation time is so large that $(\tau_s\dot{s})^2\gg\tau_R\dot{n}n_0$, then the degree of polarization of the excitations is equal to unity.

Were we to take into account the possibility of changing the excitation concentration by means of processes with spin flip (i.e., the terms proportional to τ_R^s) then the degree of polarization would be defined by the relation

$$p_s=\left(\frac{\tau_R^s-\tau_R}{\tau_R^s+\tau_R}\right)^{1/2}. \tag{7}$$

In the case of weak pumping and short spin-relaxation times, the degree of polarization of the excitations is proportional to τ_s.

In concluding this section, we present final expressions for the density of the electron flux j through a ferromagnet–superconductor tunnel junction[2] and the spin-flux density $j_s=\dot{s}d$. If p is the degree of polarization of the electrons in a ferromagnet, then we have[2)] at $T=0$

$$j=\frac{1}{4e^2R\sigma}\operatorname{Re}\left\{\frac{1+p}{2}\sqrt{(eV+I)^2-\Delta^2}+\frac{1-p}{2}\sqrt{(eV-I)^2-\Delta^2}\right\}, \tag{8}$$

$$j_s=\frac{1}{4e^2R\sigma}\operatorname{Re}\left\{\frac{1+p}{2}\sqrt{(eV+I)^2-\Delta^2}-\frac{1-p}{2}\sqrt{(eV-I)^2-\Delta^2}\right\}. \tag{9}$$

Here R is the resistance of the junction, σ is the area of the junction, $2I$ is the spin splitting of the excitation states, and V is the voltage.

It is seen from (8) that the current-voltage characteristic is asymmetrical because of the polarization of the electrons in the ferromagnet. The spin flux density behaves in similar fashion, although it does reverse sign when the voltage sign is reversed, but $j_s(V)\neq j_s(-V)$. Therefore, by reversing the direction of the current, it is possible to polarize an excitation both parallel and antiparallel to the ferromagnet polarization.

POLARIZATION OF NUCLEI IN THE CASE OF SPIN INJECTION

If the electrons in a semiconductor are optically polarized, nuclear polarization takes place in an external magnetic field.[6] The transfer of the angular momentum to the nuclear subsystem is due to the hyperfine interaction of the electrons and nuclei. The fast dipole-dipole relaxation of the nuclei leads to establishment of a spin nuclear temperature, which vanishes within the time of the spin-lattice relaxation. D'yakonov and Perel'[7] have obtained an expression for the nuclear temperature Θ in an external field in the presence of oriented electrons:

$$\frac{1}{\Theta}=\frac{2J}{\mu_J}\frac{H}{H^2+\tilde{H}_L^2}\frac{p_s+\operatorname{th}(I/T)}{1+p_s\operatorname{th}(I/T)}, \tag{10}$$

and a nuclear polarization $\langle J_z\rangle$

$$\langle J_z\rangle=\mu_J H(J+1)/3\Theta, \tag{11}$$

$$\langle J_z\rangle=\frac{2}{3}J(J+1)\frac{H^2}{H^2+\tilde{H}_L^2}\frac{p_s+\operatorname{th}(I/T)}{1+p_s\operatorname{th}(I/T)}. \tag{12}$$

Here J is the angular momentum of the nucleus, μ_J is the nuclear magneton, H is the external magnetic field, $\tilde{H}_L^2$ is the mean squared effective local magnetic field produced at the nucleus by the surrounding nuclei and dependent in the general case on the state of the electrons. If $I\ll T$, then $\tilde{H}_L^2=bH_L^2$, where H_L^2 is the mean squared local field in the absence of an external magnetic field ($b=2$–3).

In the derivation of (10) and (12) it was assumed that the spin state of the electron subsystem is governed by external conditions (pumping). In a superconductor, the external magnetic field is equal to zero, so that it can be assumed that $\langle J_z\rangle=0$, $\Theta\to\infty$ ($\Theta=T$ when account is taken of the Zeeman energy of the nuclei), and there is no effect of nuclear polarization. This is not the case, however. Indeed, polarization excitations produce, as a result of the hyperfine interaction, an effective magnetic field H_s at the nuclei:

$$H_s=\frac{A}{2\mu_J}\frac{n}{N}p_s=H_e p_s. \tag{13}$$

Here A is the hyperfine interaction constant, n is the concentration of the excitations, and N is the concentration of nuclei with a given spin. In turn, the polarized nuclei, owing to a hyperfine interaction, split the spin states of the excitations in such a way that

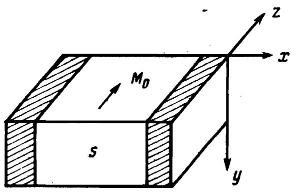

FIG. 1. Geometry of experiments on ESR in superconductors. The shaded regions on the left and on the right are, respectively, a ferromagnet and a normal metal.

$$2I=g\mu_e H_N=2A\langle J_z\rangle. \tag{14}$$

If $I/T \ll p_s < 1$, then in the absence of an external magnetic field nuclear polarization takes place in the superconductor, and its value according to (12) is

$$\langle J_z\rangle=\frac{2}{3}J(J+1)\frac{H_e^2}{H_e^2 p_s^2+bH_L^2}p_s^3. \tag{15}$$

Since usually $H_e \sim (10^6-10^7)n/N$ Oe, it follows that at $n \sim 10^{16}$ cm^{-3} the electron fields become comparable with the local field at $p_s \lesssim 1$. At lower excitation concentrations, the degree of polarization of the nuclei is proportional to the cube of the degree of polarization of the excitations.

It appears that it is easiest to register the hyperfine splitting of the energy spectrum of the excitations by using a second tunnel junction with a normal metal. Indeed, as is well known, the conductivity of an $S-I-N$ junction has a maximum at voltages $eV=\Delta$. In our case the plot of the tunnel conductivity against the voltage will have, owing to hyperfine splitting, two maxima at $eV=\Delta-I$ and $eV=\Delta+I$ (see expression (8)). Therefore, by measuring this splitting, we can measure directly the quantity I as a function of the spin pumping and obtain information on the spin relaxation times, which determine the width and the form of the maxima,[2,9] and even on the lifetimes.

ELECTRON SPIN RESONANCE IN SUPERCONDUCTORS

As shown in the preceding section, spin injection leads to the appearance of polarization of the spins of the excitations in the nuclei. The polarization of the nuclear spins on account of the hyperfine interaction leads to a splitting of the electronic states of the excitations. Therefore, if a microwave with frequency close to the frequency of the resonant transition is incident on a superconductor, resonant absorption of the microwave power is possible. In normal metals,[10,11] owing to the large spin-diffusion length, the anomalous skin effect is always encountered in ESR investigations. At resonance, the high-frequency magnetic field penetrates into the interior of the sample to a distance on the order of the spin diffusion length, which as already noted, has macroscopic dimensions. This leads to the Overhauser volume effect and to polarization of the nuclei.

In our case the situation turns out to be more complicated. Indeed, if the polarization of the nuclei at the initial instant of time is p_{N0}, then owing to the paramagnetic-resonance saturation effect it begins to decrease until the absorption changes to such an extent that the line shift offsets the decrease of the polarization of the nuclei. A self-consistent value of the degree of nuclear polarization sets in. This uncovers a new possibility of measuring an ESR signal, by investigating the line shape as the microwave pump intensity is varied. In this section we investigate all these phenomena and obtain the line shape and the microwave-field distribution in the sample.

To describe the ESR we can use Bloch's equation with a term that describes the spin-density diffusion. Since $I \ll T$, it follows that

$$\frac{\partial \mathbf{M}}{\partial t}=[\mathbf{M}\times\mathbf{\Omega}]-\frac{\mathbf{M}-\mathbf{M}_0}{\tau_s}+D\Delta\mathbf{M}. \tag{16}$$

Here $\mathbf{M}$ is the magnetization produced by the excitation and depends both on the rate of the spin injection and on the intensity of the microwave field, $\mathbf{M}_0$ is the initial magnetization, D is the diffusion coefficient, and $\mathbf{\Omega}$ is the spin-precession frequency in the field $\mathbf{H}$. If the coordinate axes are chosen in the manner shown in Fig. 1, then

$$M_0=M_{0z}=\mu_e n p_{s0},\quad \Omega=\Omega_0+\Omega_1,$$
$$\Omega_0=\Omega_{0z}=2I,\quad \Omega_1=g\mu_e H_1.$$

We introduce

$$me^{i\omega t}=M_{1x}+iM_{1y},\quad \Omega_1=-\Omega_{1y}+i\Omega_{1x},\quad M_{1z}=m_z.$$

The system of equations can then be represented in the form

$$\{1/\tau_s-D\Delta-i(\omega-\Omega_0)\}m=(m_z+M_0)\Omega_1,$$
$$\{1/\tau_s-D\Delta\}m_z=-\mathrm{Re}\,\Omega_1^* m. \tag{17}$$

It is obvious that to obtain a homogeneous nuclear polarization it is necessary to use samples of thickness $d \ll L_s$. As shown by Lifshitz *et al.*,[11] the boundary conditions have little effect on the character of the solution, and we shall therefore assume specular reflection of the excitations from both boundaries, corresponding to continuation of the microwave field to the outside of the sample in even fashion. The remaining steps of the solution are analogous to those given by Lifshitz *et al.*,[11] and we therefore present the final results without derivation, for a plate of thickness $\delta \ll d \ll L_{eff}$ (δ is the depth of the skin layer):

$$m=iM_0\frac{a_s[1+i(\omega-\Omega_0)\tau_s]}{1+|a_s|^2+(\omega-\Omega_0)^2\tau_s^2}. \tag{18}$$

$$m_z=-M_0\frac{|a_s|^2}{1+|a_s|^2+(\omega-\Omega_0)^2\tau_s^2}. \tag{19}$$

Here $a_s=g\mu_e cE(0)/\omega d$ and $E(0)$ is the electric field intensity inside the metal on the boundary with the vacuum.

Let us examine first the behavior of the degree of polarization of the nuclei, meaning also the position of the resonance, as functions of the rate of spin injection, as well as the microwave field intensity. Since the connection between the electron and nuclear polarizations is determined at $I \ll T$ and $H_s^2 \ll H_L^2$ by expression (15), it follows that, taking (14) into account, we

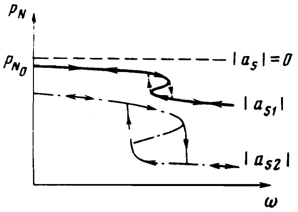

FIG. 2. Dependence of the degree of polarization of nuclei under ESR conditions on the frequency at different excitation levels $|a_{s1}| < |a_{s2}|$.

obtain an expression for the self-consistent value of I:

$$\left(\frac{I}{I_0}\frac{3H_L^2}{H_c^2}\right)^{1/2} = p_{s0}\left(1 - \frac{|a_s|^2}{1+|a_s|^2+(\omega-2I)^2\tau_s^2}\right). \qquad (20)$$

Here $I_0 = \frac{2}{3}AJ(J+1)$ is the maximum hyperfine splitting of the electron states in the case of complete polarization of the nuclei, and J is the spin of the nucleus. From (18) it is seen that under paramagnetic-resonance saturation conditions, i.e., at $|a_s|^2 \gg 1$, the degree of polarization of both the nuclei and the excitations tends to zero.

The solution of (20) at different microwave radiation powers is shown in Fig. 2. It is seen that the variations of the initial polarization of the nuclei are not the same in the forward and backward directions, i.e., hysteresis takes place. A similar hysteresis is observed also on the plot of the degree of nuclear polarization against the pump frequency. The hysteresis takes place, obviously, in the shape of the microwave absorption line, if we investigate it, for example, as a function of the spin-injection intensity. It is remarkable that, just as in a normal metal,[10] under resonance conditions the transparency of the superconducting plate increases sharply.

In conclusion let us estimate the order of magnitude of the quantities that determine the effects under consideration. We consider ^{27}Al, from which the tunnel junctions with the ferromagnet were prepared.[2] The spin of the nucleus is $J = \frac{5}{2}$. If it is assumed that the hyperfine interaction constant A for ^{27}Al is of the same order as for ^{23}Na, for which at $J \approx \frac{3}{2}$ the effective hyperfine field $H_N = 2I_0/g\mu_e$ is of the order of 200 Oe,[12] then we obtain for ^{27}Al an effective hyperfine field $H_N \approx 500$ Oe. At the same time, according to Shina,[13] the ratio of the spin-relaxation time to the momentum (or energy) relaxation time is of the order of 10^{+5}. This means that at $\tau_p \sim 10^{-10}$ sec (which is easily attained in pure aluminum) we have $\tau_s \sim 10^{-5}$ sec, and accordingly the spin diffusion length is $L_s \sim 1$ cm. The width of the ESR signal line at these values of τ_s turns out to be of the order of $\Delta H \sim 10^{-2}$ Oe.

Thus, for aluminum there are very favorable conditions for the observation of ESR in superconductors and of nuclear polarization in the case of spin injection.

In conclusion, I wish to express my gratitude to M. I. D'yakonov, G. E. Pikus, and B. Z. Spivak for useful discussions.

[1)]As shown in a number of papers,[4,5] the bottleneck may be caused by the escape of phonons of energy larger than 2Δ from the sample, rather than by the excitation recombination time. Such a mechanism leads to heating of the system if the energy of the injected quasiparticles exceeds T, and to cooling of the electron excitations if the energy of the injected quasiparticles is less than T.[4] Accordingly, in this case it is necessary to replace τ_R in the final equations by $\tau_{ef} = \tau_R(1 + d/l\eta)$, where η is the coefficient of phonon passage through the boundary, and l is the mean free path of the phonons with energy larger than 2Δ.

[2)]At finite temperatures, expressions (8) and (9) are very cumbersome, they depend on the explicit form of the distribution function of quasiparticles with given spin, and are therefore not presented in this article.

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