Spin currents in superconductors

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(Dated: June 6, 2018)

It is argued that experiments on rotating superconductors provide evidence for the existence of macroscopic spin currents in superconductors in the absence of applied external fields. Furthermore it is shown that the model of hole superconductivity predicts the existence of such currents in all superconductors. In addition it is pointed out that spin currents are required within a related macroscopic (London-like) electrodynamic description of superconductors recently proposed. The spin current arises through an intrinsic spin Hall effect when negative charge is expelled from the interior of the metal upon the transition to the superconducting state.

PACS numbers:

There is currently great interest in the study of the role of the electron spin degree of freedom in transport in solids\cite{1}. In particular the possibility of generating a pure spin current in the absence of charge currents and magnetization has been suggested\cite{2, 3, 4}, and it was recently reported that it may have been achieved by optical techniques\cite{5}.

Another area of recent interest is the possible existence of novel states of matter that involve spontaneous currents, induced by electronic correlations between electrons. Early work on superconductivity predicted the existence of such macroscopic charge currents\cite{6}, but this was proven impossible by a so-called 'Bloch's theorem'\cite{7}. More recently, states with local charge currents that spontaneously break time reversal invariance have been proposed to describe high temperature cuprate superconductors\cite{8, 9}.

Combining the above two concepts, we proposed several years ago that the low temperature phase of certain metals and in particular Chromium may be a novel state of matter induced by electronic correlations ('spin-split state') where parity is spontaneously broken but time reversal invariance is preserved\cite{10}, in which the system carries a macroscopic spin current but no charge current\cite{11}. Such spin currents give rise to electric fields which should be experimentally detectable\cite{12, 13}. Recently Wu and Zhang discussed a variety of interesting possible states of metals arising from spontaneous generation of spin-orbit coupling\cite{13}, one of which is equivalent to the proposed spin-split state.

As pointed out in our discussion of spin-split states\cite{11}, such spin currents should be insensitive to degradation by non-magnetic disorder (hence we termed them 'spin supercurrents') for the same reason as supercurrents in superconductors\cite{14}: in the presence of weak disorder, quasiparticles giving rise to the spin current state should be interpreted as time-reversed extended eigenstates of the disordered system rather than states with definite crystal momentum $k$. The question then naturally arises whether spin currents also occur in superconductors.

Note that a single Cooper pair $(k \uparrow, -k \downarrow)$ carries a spin current but not a charge current, and that the form naturally allows for the existence of a spin current if $(u_k, v_k) \neq (u_{-k}, v_{-k})$. It is then natural to ask whether such symmetry breaking will lower the free energy of the system. In this paper we show that while this is not the case for the attractive Hubbard model, and likely also not the case for electron-phonon models of superconductivity, it is the case when superconductivity is driven by a kinetic-energy-lowering interaction as in the model of hole superconductivity\cite{15}, which has been proposed to describe all superconducting materials\cite{16}. In the model of hole superconductivity, pairing is driven by an off-diagonal matrix element of the Coulomb interaction; another off-diagonal matrix element of the Coulomb interaction was shown to favor the spin-split state\cite{10}. A qualitative picture of the situation envisaged is shown in Figure 1.

Before entering into the microscopic modeling we present a macroscopic argument. Consider a rotating
simply connected superconducting body. A uniform magnetic field ('London field') develops in its interior, in the absence of any applied external field, given by

$$\vec{B} = -\frac{2m_e c}{e} \vec{\omega}$$  \hspace{1cm} (2)

with $m_e$ and $e$ the free electron mass and charge respectively, and $\vec{\omega}$ the angular velocity of rotation. This has been verified experimentally for both conventional and high $T_c$ superconductors, and can be 'explained' theoretically from the London equation

$$\vec{J} = -\frac{n_s e^2}{m_e c} \vec{A}$$  \hspace{1cm} (3)

using

$$\vec{J} = e n_s \vec{v}_s$$  \hspace{1cm} (4a)

$$\vec{v}_s = \vec{\omega} \times \vec{r}$$  \hspace{1cm} (4b)

and $\vec{B} = \nabla \times \vec{A}$, with $n_s$ the superfluid density. Eq. (4b) says that in the interior of the superconductor the superfluid rotates with the same velocity ($\vec{v}_s$) as the body. Instead, within a London penetration depth of the surface the superfluid 'lags' and this relative motion between it and the body provides the electric current that generates the magnetic field Eq. (2).

However a superfluid electron rotating with angular velocity $\vec{\omega}$ requires a centripetal force

$$\vec{F}_c(\vec{\omega}) = -\frac{m_e v_s^2}{r} \hat{r} = \frac{e}{2c} \vec{v}_s \times \vec{B}$$  \hspace{1cm} (5)

which is only half of the Lorentz force provided by the magnetic field $\vec{B}$ in Eq. (2). Hence this situation requires a compensating outward electric force on the electron to balance the radial force, which would require a higher density of negative charge in the interior of the superconductor than near the surface. There is no logical reason why such a inhomogeneous charge density would develop in a rotating superconductor, in particular for why the electrons in a rotating normal metal would suddenly move in when the metal is cooled into the superconducting state.

Instead we propose the following solution of this conundrum. Assume the speed of electrons of spin $\sigma$ in the rotating superfluid is given by

$$\vec{v}_\sigma = \sigma \vec{v}_0 + \vec{\omega} \times \vec{r}$$  \hspace{1cm} (6)

for a given spin quantization axis, with $v_0 >> \omega r$. We assume furthermore that the vector $\vec{v}_0$ is parallel or antiparallel to $\vec{\omega} \times \vec{r}$. The difference in the centripetal force for a rotating and non-rotating superconductor is

$$\vec{F}_c(\vec{\omega}) - \vec{F}_c(0) = -2m_e v_\sigma \omega \hat{r} = \frac{e}{c} \vec{v}_\sigma \times \vec{B}$$  \hspace{1cm} (7)

i.e. precisely the Lorentz force provided by the generated London field in the rotating superconductor, Eq. (2). Eq. (6) implies that the charge current in the rotating superconductor is given by Eq. (4) and in particular is zero in the absence of body rotation, and that a spin current exists

$$\vec{J}_{\text{spin}} =\frac{n_s}{2}(\vec{v}_1 - \vec{v}_1) = n_s \vec{v}_0$$  \hspace{1cm} (8)

which is independent of the body rotation and in particular is non-zero when the body is not rotating. The spin current field Eq. (8) is a function of position.

The existence of such a 'spontaneous' spin current in the absence of body rotation and magnetic field requires the existence of an electrostatic field $\vec{E}$ in the interior of superconductors, related to $\vec{v}_0$ by Newton's equation

$$\frac{d\vec{v}_\sigma}{dt} = \frac{\vec{v}_0^2}{2} - \vec{v}_0 \times (\vec{\nabla} \times \vec{v}_0) = \frac{e}{m_e c} \vec{E}$$  \hspace{1cm} (9)

Note that the right-hand-side of Eq. (9) is independent of $\sigma$ because the velocity appears squared. We have recently proposed a new macroscopic electrodynamic description of superconductors which allows for the existence of such an electrostatic field, satisfying the equation

$$\nabla^2(\vec{E} - \vec{E}_0) = \frac{1}{\lambda_L}(\vec{E} - \vec{E}_0)$$  \hspace{1cm} (10)

with $\lambda_L$ the London penetration depth, and $\vec{E}_0$ the electrostatic field originating in a uniform positive charge density $\rho_0$ throughout the volume of the superconductor.

Conversely we may argue that if an electrostatic field exists in the interior of superconductors as predicted by the theory of hole superconductivity, this requires the existence of a velocity field for the supercarriers, and consequently requires that each superfluid carrier have a 'time-reversed' partner of opposite spin moving with opposite velocity so that no charge current results in the absence of magnetic fields, and hence leads naturally to the Cooper pair concept as well as to the conclusion that macroscopic spin currents should exist in the superconducting state.

Next we turn to the microscopic theory. The pairing interaction in the model of hole superconductivity in a hole representation is given by

$$V_{kk'} = U + 2\alpha(\epsilon_k + \epsilon_{k'})$$  \hspace{1cm} (11)

where the 'kinetic interaction' term $\alpha \equiv \Delta t/t$ originates in the correlated hopping term in the Hamiltonian with amplitude $\Delta$, and where $t$ is the single hole hopping amplitude and $\epsilon_k$ is the hole kinetic energy.

We have argued elsewhere that the model of hole superconductivity predicts that negative charge is expelled from the interior of the superconductor towards the surface in the superconducting state. This gives rise to a macroscopic outward pointing electric field in the interior of the superconductor, and in the presence of such a field
the electronic potential no longer has local inversion symmetry even if the underlying lattice structure does, hence the spin-orbit interaction necessarily leads to splitting of \((k, -k)\) degeneracy. This then leads necessarily to parity breaking and \((u_k, \nu_k) \neq (u_{-k}, \nu_{-k})\) in the BCS state.

For simplicity we consider a two-dimensional or quasi-twodimensional system in a sample of cylindrical symmetry and quantize the spin in the perpendicular direction. To allow for the possibility of spin splitting we assume at the outset for the kinetic energy of a hole of spin \(\sigma\) the form

\[
\epsilon_{k\sigma} = -t \sum_{\nu=x,y} (\cos k_{\nu} + \sigma \sin k_{\nu})
\]

(12)

where we assume unit lattice spacing. The kinetic energy Eq. (12) is time-reversal invariant but breaks parity for \(b \neq 0\). In addition to being induced by charge expulsion, such a ‘spin-split’ state is favored by a nearest neighbor off-diagonal matrix element of the Coulomb interaction

\[
H_J = J \sum_{<ij>\sigma} c_{i\sigma}^\dagger c_{j\sigma} c_{i,-\sigma}^\dagger c_{j,-\sigma}
\]

(13)

with \(J > 0\), which exists in all solids as discussed in [10]. Of course it can also arise in a crystal without inversion symmetry through ordinary spin-orbit coupling.

The kinetic energy in Eq. (11) is then \(\epsilon_{k\uparrow}\) or \(\epsilon_{-k\downarrow}\) which are identical, and solution to the BCS equations proceeds identically to the case in the absence of spin splitting, except that in the energy versus \(k\) relation the form Eq. (12) is to be taken. The gap function is of the form

\[
\Delta_k = \Delta_m \left(-\frac{\epsilon_k}{D/2} + c\right)
\]

(14)

with \(\Delta_m\), \(c\) constants. Hence \(\Delta_k \neq \Delta_{-k}\) in general, unless \(\Delta_m \to 0\), \(\Delta_m c \neq 0\) which only occurs for \(\alpha \to 0\), \(U < 0\) for the interaction Eq. (11).

We show in Figure 2 results for the critical temperature versus hole concentration for a case with parameters appropriate to the physics of the model of hole superconductivity (\(U > 0\), \(\alpha > 0\)) compared with an attractive Hubbard model (\(U < 0\), \(\alpha = 0\)). Notably it is seen in Fig. 2 that spin-splitting enhances the tendency to superconductivity when the attraction leading to Cooper pairing is of kinetic origin, and suppresses it when the attraction is of potential origin as in the attractive Hubbard model. It is likely that the same qualitative behavior as in the attractive Hubbard model will result for models with more extended attractive density-density interactions (e.g., Hubbard model with attractive nearest neighbor interactions) as well as for conventional electron-phonon interaction models (Holstein, SSH, Frohlich) where the effective electron-electron interaction albeit retarded also leads to potential energy lowering.

In the superconducting state, we find that spin splitting raises the condensation energy in the model of hole superconductivity and lowers it in the attractive Hubbard model. Results are shown in Fig. 3 for two values of the hole concentration. We find that for the entire concentration range where \(T_c\) is not zero spin splitting increases the condensation energy in the model of hole superconductivity and decreases it in the attractive Hubbard model.

Furthermore in the presence of spin splitting the relation between chemical potential and carrier concentration changes. Figure 4 shows the hole concentration in the model of hole superconductivity as a function of spin splitting for fixed value of the chemical potential, showing that spin splitting causes an increase in the hole concentration for fixed chemical potential. We interpret this result to mean that spin splitting enhances the tendency for the superconductor to expel electrons from its interior [23]. This tendency exists for all hole concentrations.
Of course the model considered here does not take into account the large charging energy that would result if the hole concentration changed by the amounts shown in Fig. 4. In fact, as electrons move out of the interior towards the surface the resulting cost in Coulomb energy will stabilize the excess electron concentration near the surface at a value corresponding to just a few electrons per million atoms, as discussed in ref. [23].

Furthermore as charge expulsion takes place the resulting internal electric field that builds up further enhances the tendency to spin splitting through the spin-orbit interaction that occurs between the superfluid electrons in macroscopic orbits and the macroscopic electric field that builds up, as would occur in a 'giant atom' [24].

Finally, our physical picture provides a simple way to understand how the macroscopic spin current builds up when the system makes the transition to the superconducting state. As negative charge is expelled from the interior of the superconductor in the radial direction, the interaction of the moving magnetic moment of the electron with the positive background gives rise to a force in direction perpendicular to the motion whose sign depends on the spin orientation. Such an effect was discussed by us in connection with a dissipationless intrinsic mechanism for the spin Hall effect and the anomalous Hall effect in ferromagnetic metals [27], and may also be predicted by a universal intrinsic spin Hall effect recently discussed [26, 27]. This force deflects the electron in opposite azimuthal directions for up and down spin electrons [22] such as to give rise to the macroscopic spin current. This is shown schematically in Figure 5.

In summary we have provided macroscopic and microscopic arguments in favor of the suggestion that macroscopic spin supercurrents exist in the superconducting state of all metals. As already pointed out by Bohm [7], 'Bloch’s theorem' does not invalidate such possibilities. The direction of flow of these currents is determined by the sample geometry through spin-orbit coupling to the electrostatic field that results from solution of the macroscopic electrodynamics equations [22]. Spin currents further stabilize the superconducting state in the model of hole superconductivity and explain the value of the 'London field' in rotating superconductors. When an external magnetic field is applied the pre-existent spin currents will be modified so that they no longer exactly cancel giving rise to a Meissner current that screens the applied magnetic field. Experimental detection of macroscopic spin currents in superconductors may be possible through spin-resolved neutron scattering experiments as discussed in ref. [10] or angle resolved photoemission experiments with circularly polarized photons as proposed in ref. [4] or optical [3] or other experiments [24]. It has also been suggested that the physics discussed here could have relevance to aromatic molecules [28, 29].

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