sin(2\(\varphi\)) current-phase relation in SFS junctions with decoherence in the ferromagnet

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Abstract. – We propose a theoretical description of the sin(2\(\varphi\)) current-phase relation in SFS junctions at the 0-\(\pi\) crossover obtained in recent experiments by Sellier et al. (Phys. Rev. Lett., 92 (2004) 257005, cond-mat/0406236), where it was suggested that a strong decoherence in the magnetic alloy can explain the magnitude of the residual supercurrent at the 0-\(\pi\) crossover. To describe the interplay between decoherence and elastic scattering in the ferromagnet, we use an analogy with crossed Andreev reflection in the presence of disorder. The supercurrent as a function of the length \(R\) of the ferromagnet decays exponentially over a length \(\xi\), larger than the elastic scattering length \(l_d\) in the absence of decoherence, and smaller than the coherence length \(l_\varphi\) in the absence of elastic scattering on impurities. The best fit leads to \(\xi \approx \xi_d^{(\text{diff})}/3\), where \(\xi_d^{(\text{diff})}\) is the exchange length of the diffusive system without decoherence (also equal to \(\xi\) in the absence of decoherence). The fit of experiments works well for the amplitude of both the sin \(\varphi\) and sin(2\(\varphi\)) harmonics.

Introduction. – When Cooper pairs from a superconductor (S) penetrate a ferromagnet (F) at a SF interface, the spin-up electron decreases its spin energy because of Zeeman splitting, whereas the spin-down electron increases its spin energy. As a consequence, the spin-up electron accelerates and the spin-down electron decelerates so that Cooper pairs acquire a finite center-of-mass momentum \(\Delta k\). This induces spatial oscillations of the pair amplitude in the ferromagnet. If the length \(R\) of the ferromagnet is well chosen, the Josephson relation of a SFS junction can be inverted, giving rise to a \(\pi\)-junction [1–4]. The determination of the current-phase relation in SNS (where N is a normal metal) and SFS junctions is a long-standing problem (see the recent review by Golubov et al. [5]). The oscillations of the transition temperature of FS superlattices [6] as a function of the thickness of the ferromagnet are another consequence of the \(\pi\)-coupling.

SFS \(\pi\)-junction have been probed recently in experiments in which the ferromagnet is a magnetic alloy with a sufficiently small exchange field [7–11], so that the period of the spatial oscillations of the pair amplitude is large enough. In the following we discuss a recent
experiment [12] in which half-integer Shapiro steps were observed in a Nb/CuNi/Nb junction at the 0-π crossover, indicating a \( \sin(2\varphi) \) current-phase relation. For highly transparent interfaces and without decoherence, the current-phase relation is related to the derivative of the energy of the Andreev bound states with respect to the superconducting phase difference between the two superconductors [2–5]. The exchange field in the ferromagnet generates a Zeeman splitting of the spectrum of Andreev bound states, which was the explanation of the \( \sin(2\varphi) \) harmonics at the 0-π crossover proposed by Sellier et al. [12]. To explain the tiny magnitude of the residual supercurrent at the 0-π crossover, Sellier et al. [12] suggested the existence of a strong decoherence in the magnetic alloy. However, within this assumption the Andreev bound states are broadened so that the supercurrent cannot be any longer evaluated as the derivative of the bound-state energies. This calls for a specific modeling of π junction involving decoherence in the ferromagnet.

The origin of decoherence in a magnetic alloy is not well characterized experimentally, because it is not possible to carry out weak localization and universal conductance fluctuations as a function of an applied magnetic field. Quantum coherence in a ferromagnet was however studied in a recent experiment using time-dependent universal conductance fluctuations [13]. One source of decoherence in a ferromagnet are spin waves, but other effects can play a role, such as spatial heterogeneities of the exchange field or domain wall motion [14,15].

We discuss here the effect of decoherence in the magnetic alloy, motivated by the experimental observations of a very small residual supercurrent at the 0-π crossover and more generally to a huge reduction of the supercurrent compared to a model not involving decoherence [11,12]. We show that decoherence in the ferromagnet implies the existence of a length \( \xi \), intermediate between i) the elastic mean free path \( l_d \) due to elastic scattering on impurities in the absence of decoherence, and ii) the coherence length \( l_\varphi \) due to decoherence in the magnetic alloy in the absence of elastic scattering on impurities. The \( \sin \varphi \) harmonics due to the coherent transfer of a charge-\((2e)\) is proportional to \( \exp[-R/\xi] \), and the \( \sin(2\varphi) \) harmonics due to the coherence transfer of a charge-\((4e)\) is proportional to \( \exp[-2R/\xi] \), where \( R \) is the length of the ferromagnet. The length scale \( \xi \) can be smaller than \( R \) even though \( l_\varphi \) is larger than \( R \). Therefore we base our description on the first two terms of an expansion in the coherent transfer of multiples of a charge-\((2e)\). Similarly to ref. [16], the dressing by multiple local Andreev reflections is described non-perturbatively so that our description is valid for highly transparent interfaces. We also treat rigorously the geometrical effects related to propagation parallel to the interfaces.

**The models.** – We consider a model in which a 3D ferromagnet is connected to two 3D superconductors (see fig. 1(a)). The superconducting electrodes are described by the BCS Hamiltonian

\[
\mathcal{H}_{\text{BCS}} = \sum_{(\alpha,\beta),\sigma} -t \left( c_{\alpha,\sigma}^\dagger c_{\beta,\sigma} + c_{\beta,\sigma}^\dagger c_{\alpha,\sigma} \right) + \Delta \sum_\alpha \left( c_{\alpha,\uparrow}^\dagger c_{\alpha,\downarrow} + c_{\alpha,\downarrow}^\dagger c_{\alpha,\uparrow} \right),
\]

where \( \alpha \) and \( \beta \) correspond to neighboring sites on a cubic lattice. The ferromagnetic electrode is described by the Stoner model

\[
\mathcal{H}_{\text{Stoner}} = \sum_{(\alpha,\beta),\sigma} -t \left( c_{\alpha,\sigma}^\dagger c_{\beta,\sigma} + c_{\beta,\sigma}^\dagger c_{\alpha,\sigma} \right) - h_{\text{ex}} \sum_\alpha \left( c_{\alpha,\uparrow}^\dagger c_{\alpha,\downarrow} - c_{\alpha,\downarrow}^\dagger c_{\alpha,\uparrow} \right),
\]

where \( h_{\text{ex}} \) is the exchange field. We add to (2) a term describing elastic scattering by impurities. The ferromagnet is connected to the superconducting layers by the Hamiltonian

\[
W_{a(b)} = \sum_{\alpha,\sigma} -t_{a(b)} \left( c_{\alpha,\sigma,a(b)}^\dagger c_{\alpha,\sigma,a} + c_{\alpha,\sigma,a}^\dagger c_{\alpha,\sigma,a(b)} \right),
\]

where the label \( a \) runs over all sites at the interface.
To describe propagation in the ferromagnet, we use the advanced Green’s function of a bulk ballistic 3D ferromagnet,

\[ g_{1,1,A}^{a,b} = -\frac{1}{l_F} \frac{1}{k_F R} e^{-i k_F R} e^{-i \varphi / l}, \]  

(4)

where \( l \) is the phase coherence length. A similar expression is obtained for \( g_{2,2,A}^{a,b} \). We will discuss how to incorporate disorder in the ferromagnetic electrode. The Green’s functions of a superconductor can be found in the literature \[17\].

The Nambu representation of the tunnel amplitudes is given by

\[ \hat{t}_{a,\alpha} = \hat{t}_{\alpha,\alpha}^* = t_a \begin{pmatrix} e^{i \varphi / 4} & 0 \\ 0 & e^{-i \varphi / 4} \end{pmatrix}, \]  

(5)

and similar equations are obtained for \( \hat{t}_{b,\beta} \) and \( \hat{t}_{\beta,b} \).

**Supercurrent.** - The supercurrent is given by

\[ I_S = \frac{e}{\hbar} \int_0^{+\infty} \text{Tr} \left\{ \hat{\sigma}^z \left[ \hat{\tau}_{a,\alpha} \left( \hat{G}_{a,a}^A - \hat{G}_{a,a}^R \right) - \hat{\tau}_{a,\alpha} \left( \hat{G}_{\alpha,a}^A - \hat{G}_{\alpha,a}^R \right) \right] \right\} d\omega, \]  

(6)

where the trace corresponds to a summation over the “11” and “22” matrix elements in the Nambu representation, a summation over the two spin orientations (formally equivalent to summing over the “33” and “44” components in the spin \( \otimes \) Nambu representation \[18\]), and a summation over the channel labels. Equation (6) can be derived from Keldysh formalism \[19, 20\] by noting that in equilibrium the Keldysh Green’s function takes the simple form \( \hat{G}^{+, -} = n_F(\omega) [\hat{G}_A^A - \hat{G}_R^R] \). The fully dressed Green’s functions \( G_{i,j} \) are determined through the Dyson equation \( \hat{G} = \hat{g} + \hat{g} \otimes \hat{\Sigma} \otimes \hat{G} \), where \( \hat{\Sigma} \) is the self-energy corresponding to the bonds \( t_{a,\alpha} \) and \( t_{b,\beta} \) and \( \otimes \) denotes a summation over the sites \( \alpha_i, a_i, b_i, \beta_i \) (see fig. 1) and a convolution
over the time arguments that becomes a simple product once the Dyson equation is Fourier-transformed to the energy variable \( \omega \). The supercurrent takes the simpler form

\[
I_S = \frac{e}{\hbar} \int_0^{+\infty} d\omega \sum_{a,b} \{ \text{Tr} \left[ \tilde{g}_{a,b} \tilde{V}_{a,b} \tilde{g}_{b,a} \tilde{W}_{a,a} \right] + \text{Tr} \left[ \tilde{g}_{a,b} \tilde{V}_{b,a} \tilde{g}_{b,a} \tilde{W}'_{a,a} \right] \},
\]

(7)

where \([\cdot]\) is an anticommutator and \(\tilde{\sigma}^{z}\) one of the Pauli matrices.

Perturbative expansion of the supercurrent. – Now we describe a perturbative expansion in which we include the coherent transfer of a charge-\((2e)\) and charge-\((4e)\) object while keeping a non-perturbative description of local processes. The Green’s function \(\hat{G}_{a,a}\) can be expanded according to \(\hat{G}_{a,a} = \sum_n \hat{G}^{(n)}_{a,a}\), where \(\hat{G}^{(n)}_{a,a}\) describes the \(\sin(n\varphi)\) harmonics due to the coherent transfer of a charge-\((2ne)\) object. We obtain \(\hat{G}^{(0)}_{a,a} = \hat{K}_{a,a} \hat{g}_{a,a}\),

\[
\hat{G}^{(1)}_{a,a} = \hat{K}_{a,a} \hat{X}_{a,b} \hat{K}_{b,b} \hat{X}_{b,a} \left[ \hat{g}_{b,a} + \hat{X}_{b,a} \hat{K}_{a,a} \hat{g}_{a,a} \right],
\]

(8)

\[
\hat{G}^{(2)}_{a,a} = \hat{K}_{a,a} \hat{X}_{a,b} \hat{K}_{b,b} \hat{X}_{b,a} \hat{K}_{a,a} \hat{X}_{a,b} \hat{K}_{b,b} \left[ \hat{g}_{b,a} + \hat{X}_{b,a} \hat{K}_{a,a} \hat{g}_{a,a} \right],
\]

(9)

with \(\hat{X}_{a,a} = \hat{g}_{a,a} \hat{t}_{a,a} \hat{g}_{a,a} \hat{t}_{a,a}\), \(\hat{K}_{a,a} = [\hat{I} - \hat{X}_{a,a}]^{-1}\), and with similar expressions for \(\hat{K}_{b,b}, \hat{K}_{a,b}, \hat{K}_{b,a}\). The channels labels are kept implicit.

The \(\sin \varphi\) harmonics of the supercurrent is given by \(I_S^{(1)} \sin \varphi\), with

\[
I_S^{(1)} = \frac{e}{\hbar} \int_0^{+\infty} d\omega \sum_{a,b} \left\{ \text{Tr} \left[ \hat{g}_{a,b} \hat{V}_{b,b} \hat{g}_{b,a} \hat{W}_{a,a} \right] + \text{Tr} \left[ \hat{g}_{a,b} \hat{V}_{b,b} \hat{g}_{b,a} \hat{W}'_{a,a} \right] \right\},
\]

(10)

where the sum over \(a\) and \(b\) is a sum over all channels at the two interfaces. The vertices \(\hat{V}, \hat{W}\), and \(\hat{W}'\) containing information about the dressing by local processes are given by

\[
\hat{V}_{a,a} = \hat{t}_{a,a} \hat{g}_{a,a} \hat{t}_{a,a} \hat{K}_{a,a},
\]

(11)

\[
\hat{W}_{a,a} = \hat{t}_{a,a} \left[ \hat{g}_{a,a} \hat{\sigma}^{z} \right] \hat{t}_{a,a} \hat{K}_{a,a},
\]

(12)

\[
\hat{W}'_{a,a} = \hat{t}_{a,a} \hat{g}_{a,a} \hat{t}_{a,a} \hat{K}_{a,a} \hat{g}_{a,a} \hat{t}_{a,a} \left[ \hat{g}_{a,a} \hat{\sigma}^{z} \right] \hat{t}_{a,a} \hat{K}_{a,a}.
\]

(13)

The same perturbative expansion can be applied to the amplitude \(I_S^{(2)}\) of the \(\sin(2\varphi)\) harmonics. The three diagrams in fig. 1(c), (d), (e) give rise to 24 terms that can all be evaluated explicitly. One of these terms is

\[
\frac{e}{\hbar} \int_0^{+\infty} d\omega \sum_{a_1, b_1, a_2, b_2} g_{a_1, b_1}^{1,1} V_{b_1, b_1}^{1,2} \left[ g_{b_1, a_1}^{2,2} V_{a_1, a_2}^{2,1} g_{a_2, b_2}^{1,1} V_{b_2, b_2}^{1,2} g_{b_2, a_2}^{2,2} W_{a_2, a_1}^{2,1} \right].
\]

(14)

The quantities \(V_{b_1, b_1}^{1,2}\) and \(\int V^{2,1}(R) W^{2,1}(R) d\mathbf{R}\) (with \(\mathbf{R}\) the vector between \(a_1\) and \(a_2\)) are evaluated by a Fourier transform. \(V_{b_1, b_1}^{1,2}\) is proportional to \(\int f^{1,2}(k_{||}) d\mathbf{k}_{||}\) and \(\int V^{2,1}(R) W^{2,1}(R) d\mathbf{R}\) is proportional to \(\int V^{2,1}(k_{||} = 0) W^{2,1}(k_{||} = 0)\). We now evaluate the average over disorder of \(g_{a_1, b_1}^{1,1} g_{b_1, a_1}^{2,2}\) and show that, as for crossed Andreev reflection [21, 22], disorder changes the value of the exponent in the geometrical prefactor compared to the ballistic case and therefore enhances the supercurrent through the ferromagnet compared to the ballistic case. We also calculate the coherence length \(\xi\) and the effective Fermi wave vector mismatch \(\Delta K\) in the presence of both disorder and decoherence.
Effect of disorder in the ferromagnet. – The average Green’s function as a function of separation $d$ decays like $g_{a,b}^{1,1,A}(d) = g_{a,b}^{1,1,A}(d) \exp[-d/l_d]$ [17], where $g_{a,b}^{1,1,A}(d)$ is given by eq. (4). The mean free path $l_d$ due to elastic scattering on impurities is supposed to be much smaller than the ballistic coherence length $l_\varphi$. The calculation of the diffusion probability

$$P(d) = \frac{t_F}{a_0} g_{1,1}^A(d) g_{2,2}^A(d),$$

with $a_0$ the lattice spacing, is formally analogous to the calculation of the subgap conductance of a disordered superconductor [21,22], except for different phase factors. Following ref. [21] we obtain

$$P(d) = \frac{1}{4\pi \hbar D d} \exp\left[-\frac{d}{\xi}\right],$$

with $D = v_F l_d/3$ the diffusion constant, and with

$$1/\xi = \sqrt{\frac{3}{2l_d}} \left[ \frac{2}{l_\varphi} + i\Delta k \right].$$

(17)

We deduce from eq. (17) the following expression of the effective wave vector mismatch $\Delta K$ and coherence length $\xi$:

$$\Delta K = \sqrt{\frac{3}{2l_d}} \sqrt{\left(\frac{2}{l_\varphi}\right)^2 + (\Delta k)^2 - \frac{2}{l_\varphi}},$$

(18)

$$\frac{1}{\xi} = \sqrt{\frac{3}{2l_d}} \sqrt{\left(\frac{2}{l_\varphi}\right)^2 + (\Delta k)^2 + \frac{2}{l_\varphi}},$$

(19)

Integrating over all channels at the two interfaces we obtain

$$\sum_{a,b} g_{a,b}^{1,1,A}(d) g_{b,a}^{2,2,A}(d) = -N_{ch} a_0 \frac{1}{4\pi \hbar D} \int \frac{2\pi y \, dy}{\sqrt{R^2 + y^2}} \exp[-d/\xi] \exp[i\Delta K d]$$

(20)

$$= -N_{ch} a_0 \frac{1}{2\hbar D} \frac{\exp[-R/\xi]}{1/\xi - i\Delta K},$$

(21)

with $d = \sqrt{R^2 + y^2}$.

Fit of the experiments. – Now we use the model discussed above to fit quantitatively the recent experiments by Sellier et al. [12]. The value of the bulk hopping amplitudes $t_F$ and $t_S$ are chosen equal to avoid multiplying the number of parameters, and such that the diffusion constant of the ferromagnet is close to $D = 4 \text{ cm}^2\text{s}^{-1}$ [12]. We use $t_S = t_F = 5 \times 10^5 \text{ K}$ and the elastic mean free path is $l_d = 10 \text{ Å}$ [11]. The Fermi wave vector $k_F$ in the superconductor is chosen equal to $k_F = 1 \text{ Å}^{-1}$, and the lattice parameter is chosen equal to $a_0 = 1 \text{ Å}$. Since the interfaces are highly transparent we choose $t_a = t_b = t_S = t_F$. We fix the ratio between $l_\varphi$ and $\xi_{h}^{(\text{ball})} = \hbar v_F / \hbar_{\text{ex}}$ in the ballistic system to be smaller than unity, and determine the exchange field in such a way that the $\sin \varphi$ harmonics vanishes for the same value of $R$ as in the experiment. We first tried a fit with $l_\varphi = \xi_{h}^{(\text{ball})} = 7150 \text{ Å}, \hbar_{\text{ex}} = 70 \text{ K}$, and with $\xi = 100 \text{ Å}$. This fit is not compatible with the three experimental points with the largest values of $R$ and the residual value of the supercurrent at the 0-π crossover is far too large (curve (a) in
Fig. 2 – Variation of the amplitudes $I_S^{(1)}$ and $I_S^{(2)}$ of the sin$\varphi$ and sin(2$\varphi$) harmonics as a function of the thickness $R$ of the ferromagnet. The experimental points [11, 12] are indicated in the left panel. We use $D = 3.5$ cm$^2$s$^{-1}$, $d = 10$ Å. Panel (a) corresponds to $h_{ex} = 70$ K, $l_{\varphi} = 7150$ Å, $\xi = 100$ Å. Panel (b) corresponds to $h_{ex} = 190$ K, $l_{\varphi} = 1300$ Å, $\xi = 45$ Å. Panel (c) corresponds to $h_{ex} = 460$ K, $l_{\varphi} = 270$ Å, $\xi = 20$ Å.

To obtain a better agreement with experiments, we increase $h_{ex}$ and decrease $l_{\varphi}$. The fit (b) corresponds to $h_{ex} = 190$ K, $l_{\varphi} = 1300$ Å, $\xi = 45$ Å. The fit (c) in fig. 2 is even better, with $h_{ex} = 460$ K, $l_{\varphi} = 270$ Å, $\xi = 20$ Å. The values of $\xi$ can be compared to the values of $\xi_{h_{\text{diff}}}^{(\text{diff})} = \sqrt{hD/h_{ex}}$ for the diffusive system in the absence of decoherence. The values of $\xi_{h_{\text{diff}}}^{(\text{diff})}/\xi$ are approximately equal to 1.5 (fit (a)), 2 (fit (b)) and 3 (fit (c)), whereas $\xi_{h_{\text{diff}}}^{(\text{diff})}/\xi = 1$ in the absence of decoherence.

The amplitude of the sin(2$\varphi$) harmonics at the 0-π crossover is 4 $\mu$A in experiments. The fits (a) and (b) lead to much larger values, whereas the fit (c) has the correct order of magnitude (see fig. 2). The agreement of the fits of the amplitudes of both the sin$\varphi$ and sin(2$\varphi$) harmonics suggests the validity of the fit (c). The renormalization of the coherence length $\xi$ compared to its value $\xi_{h_{\text{diff}}}^{(\text{diff})}$ in the absence of decoherence may depend on the quality of the magnetic layer, since it is expected qualitatively that decoherence is reduced with less inhomogeneities in the exchange field.

Conclusion. – To conclude, we have provided a modeling of the sin(2$\varphi$) current-phase relation in SFS junctions at the 0-π crossover. This model is motivated by the fact that the residual supercurrent at the 0-π crossover is very small, therefore suggesting that decoherence in the ferromagnet plays a relevant role, as suggested by Sellier et al. [12]. Decoherence was introduced through a phenomenological coherence length $l_{\varphi}$. Elucidating precisely its microscopic origin is difficult experimentally, but several factors (spin waves, inhomogeneities in the exchange field, motion of domain walls) may play a role. The supercurrent was calculated through an expansion in the number of non-local processes connecting the two interfaces. Lowest-order processes correspond to a coherent transfer of a charge-(2$e$) contributing to the sin$\varphi$ harmonics and the next order corresponds to a coherent transfer of a charge-(4$e$) contributing to the sin(2$\varphi$) harmonics. As for crossed Andreev reflection [16], these non-local processes are dressed by multiple local Andreev reflections that were included in a non-perturbative fashion. We also included the geometrical effect of propagation parallel to the interfaces.
We found that in the ferromagnet disorder effects (characterized by the elastic scattering length $l_d$ in the absence of decoherence) and decoherence (characterized by the coherence length $l_\phi$ in the ballistic limit) generate a new coherence length $\xi$, intermediate between $l_d$ and $l_\phi$, and smaller than the exchange length $\xi_{\text{h}}^{(\text{diff})}$ evaluated in the diffusive limit in the absence of decoherence. The amplitude of the $\sin \varphi$ harmonics is proportional to $\exp[-R/\xi]$ and the amplitude of the $\sin(2\varphi)$ harmonics is proportional to $\exp[-2R/\xi]$ so that $\exp[-R/\xi]$ is a small parameter in the expansion in non-local Andreev processes. The approach is consistent in the sense that the fit to experiments shows that $\exp[-R/\xi]$ is a very small parameter (we have $R/\xi = 8.75$ for the fit (c) in fig. 2) so that the $\sin(3\varphi)$ and higher harmonics can indeed be neglected.

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