Coupled Superconducting Phase and Ferromagnetic Order Parameter Dynamics

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Via a direct coupling between the magnetic order parameter and the singlet Josephson supercurrent, we detect spin-wave resonances, and their dispersion, in ferromagnetic Josephson junctions in which the usual insulating or metallic barrier is replaced with a weak ferromagnet. The coupling arises within the Fraunhofer interferential description of the Josephson effect, because the magnetic layer acts as a time dependent phase plate. A spin-wave resonance at a frequency \( \omega_s \) implies a dissipation that is reflected as a depression in the current-voltage curve of the Josephson junction when \( h\omega_s = 2eV \). We have thereby performed a resonance experiment on only 107 Ni atoms.

The coupled dynamics of the electromagnetic field and a Josephson junction has a number of manifestations and is very well understood [1, 2, 3, 4]. When the usual insulating or metallic barrier is replaced with a weak ferromagnet there is a coupling to another field, namely the spontaneous magnetisation of the ferromagnet. Spin-waves are elementary spin excitations which are very well understood [1, 2, 3, 4]. When the ferromagnet is a Josephson junction has a number of manifestations as described by the Landau-Lifshitz equations [5]. The Josephson phase difference \( \phi \) between the two superconductors has its own dynamics. A bias voltage \( V_0 \) causes \( \phi \) to become time-dependent so that \( \phi(t) = \phi_0 + \omega_J t \), where \( \omega_J = (2e/\hbar)V_0 \) and \( \phi_0 \) is arbitrary. Corresponding to the ac Josephson effect [1], for our junctions, to a good approximation, the resulting ac Josephson current density is \( J_s = J_c \sin(\phi_0 + \omega_J t) \), where \( J_c \) is the critical current density.

In analogy with the A-phase [2] of 3He, coupled magnetic and phase oscillations should exist in ferromagnetic superconductors with triplet pairing, but have never been observed. We show here that a similar coupling for singlet superconductors can be realised in a Josephson junction with a ferromagnetic barrier. The dynamical coupling stems from the spatial interference of the Aharonov-Bohm phase caused by \( \mathbf{M}(t) \), resulting in the spatial dependence of \( \phi(r, t) \). The ac Josephson current produces an oscillating magnetic field \( \mathbf{H}(t) \) and when the Josephson frequency matches the spin wave frequency, \( \omega_J \approx \omega_s \), this resonantly excites \( \mathbf{M}(t) \). Due to the nonlinearity of the Josephson effect, there is a rectification of current across the junction, resulting in a dip in the average dc component of \( J_s \) at voltage \( V_s = (h/2e)\omega_s \). The principal result reported here is the observation of these coupled dynamics.

Magnetised Josephson junctions [7] require weak ferromagnetic materials and nanosized junction area to keep the overall magnetic flux in the junction smaller than the flux quantum \( \Phi_0 \). An electron microscope image of a typical ferromagnetic junction used in this study is shown in Fig. 1(a), while Fig. 1(b) is a schematic representation of the different layers. The superconducting electrodes comprise 50 nm of Nb (\( T_c = 7.6 \) K), while the barrier is 20 nm of Pd0.9Ni0.1 (\( T_{\text{Curie}} = 150 \) K). The current-voltage (IV) characteristics are measured using current bias and are reported, as function of the applied in-plane field, in the right insert of Fig. 2. The IV characteristics are not hysteretic, and overall they correspond closely to those expected for a junction with a conductive barrier [8, 9]. The junction normal resistance is \( R_n \approx 0.8 \) \( \Omega \), and the Josephson coupling is \( I_cR_n \approx 5 \mu \text{V} \), as expected for ferromagnetic junctions of this thickness [7], yielding the critical supercurrent of \( I_c \approx 7 \mu \text{A} \). The hostile nature of even a weak ferromagnetic environment for singlet Cooper pairs is illustrated by a similar junction with 70 nm of nonmagnetic Pd which, despite the almost four times larger thickness, has a larger critical current \( I_c \approx 44 \mu \text{A} \).

For a square junction of side \( L \), the total supercurrent is given by the integral [10]

\[
I_s = J_c \int_{-L/2}^{L/2} dx \int_{-L/2}^{L/2} dy \sin \phi(x, y, t)
\]

(1)

with

\[
\phi(r, t) = \phi_0 + \omega_J t - \frac{2e}{\hbar} \int \mathbf{A} \cdot d\mathbf{r},
\]

(2)

where the last term is the Aharonov-Bohm phase [11], involving the vector potential \( \mathbf{A} \). We use a gauge where
son junction. The ac Josephson current \( I(t) \) flows through the junction creating an rf magnetic field \( H(t) \), causing the magnetisation precession \( M(t) \), which in turn resonantly couples with the Josephson phase at frequency \( \omega_J \). Layers are respectively from the bottom: Nb (50 nm), Pd\(_{0.9}\)Ni\(_{0.1}\) (20 nm) and Nb (50 nm).

(c) The equivalent RSJ model and the sketch of the effect of the FMR on the current-voltage characteristics. The ferromagnetic layer is modeled as a series LCR\(_0\) oscillator, in parallel with the Josephson junction. Resistance \( R_0 \) is proportional to the imaginary part of the susceptibility \( \chi'' \).

\[
A = A(r, t) \hat{z}, \quad \text{the direction } \hat{z} \text{ being perpendicular to the junction surface [see Fig. 1].}
\]

Therefore \( \phi(r, t) = \phi_0 + kx + \omega_J t + \phi_m \), where \( \phi_m = (4ae/\hbar)A_{mz} \) reflects time dependent fields and \( k = (4ed/\mu_0)H + (4ea/\mu_0)M_{0y} \). Here \( M_{0y} \) is the \( y \) component of the static magnetisation \( M_0 \), the applied field \( H \) is in the \( y \) direction, \( 2a \) and \( 2d = 2(a + \lambda) \) are the actual and magnetic thickness of barrier and \( \lambda \) the London penetration depth. Equations 1 and 2 are used to describe both the statics and the dynamics of our junctions.

In the absence of a bias \( (V_0 = 0) \), we are dealing with static fields, and the Equations 1 and 2 lead to the Fraunhofer pattern \( I_s = J_L \int_{-L/2}^{L/2} dx \sin kx \) [1]. The magnetisation \( M_0 \) of the barrier has the same effect as inserting a wedge shaped phase plate in front of the slit, it displaces the diffraction pattern. Experimentally, the diffraction pattern is shifted to the right for increasing (positive \( M_{0y} \)), and left for decreasing (negative \( M_{0y} \)), fields. This illustrates the linear nature of the coupling to \( M \). In Fig. 2, the dotted curves are a fit using Eqs. 1 and 2, along with the magnetisation data measured on a trilayer with the same cross section as the junction [see the left insert of Fig. 2]. The periodicity and the asymptotic behaviour of the measured diffraction pattern attest to the high quality of our junctions. They confirm the close-to-uniform current distribution and single-harmonic current-phase relation, while the production of the shift with the two sweep directions, using experimental magnetostatic data, confirms the validity of our description.

The dynamical coupling reflects a similar phase contribution \( \phi(r, t) \) due to \( M(r, t) \), but which now has both a temporal and a spatial dependence, the equivalent of a phase plate in the optical analog with a similarly dependent refractive index \( n(r, t) \). The dynamics of the magnetisation (timescale of 1 ns) is much slower than the diffusion time through the ferromagnetic layer (0.5 ps). The Josephson coupling is thus adiabatic with respect to the magnetisation dynamics. This assumption is implicit in Eqs. 1 and 2. The signal is seen for \( V > I_cR \), implying Eq. 1 can be linearized. The dc magnetic signal then corresponds to 3.

\[
I_m = \frac{4ae}{\hbar} \int_{-L/2}^{L/2} dx \int_{-L/2}^{L/2} dy J_L \cos(kx + \omega_J t)\phi_m, \quad 3
\]

where the bar denotes a time average. Substituting for \( \phi_m = (4ae/\hbar)A_{mz} \) and using \( J = \nabla \times H \), following both a time and space integration by parts, the dc signal reflecting the magnetic resonance is

\[
I_m = \frac{1}{V_0} \int d\mathbf{r} \mathbf{H} \cdot \frac{d\mathbf{M}}{dt}, \quad 4
\]

with \( M_{i}(r,t) = \int dt' \chi_i(t-t')H_i(r,t') \), \( i = x, y, z \), where \( \chi_i(t) \) is the dynamic susceptibility. This has an appealing interpretation in terms of magnetic losses. Here, as illustrated by Fig. 1(b), \( \mathbf{H}(r, t) \) is the magnetic field which
circulates inside the junction by virtue of the ac Josephson current. The junction lateral size $L$ is smaller than both $\lambda$ and the skin depth for the frequencies involved. The displacement current is therefore negligible and all that is needed is to integrate Ampère’s law in order to determine $\mathbf{H}(\mathbf{r}, t)$. More details of these calculations are given elsewhere. The current due to $\mathbf{M}(\mathbf{r}, t)$ is

$$I_m = 2\pi I_c(0) \Phi_{rf} \left[ F_x \chi_x(\omega_J) + F_y \chi_y(\omega_J) \right],$$

where $\Phi_{rf} = (2aL)B_{rf} = (2aL)\mu_0I_c(0) L$ is the flux due to the radio frequency field and $F_x = (1/12)(I_c(B_0)/I_c)^2$ and $F_y = (2/\theta_L^2)[1 + \sin(\theta_L/2) \cos(\theta_L/2) + ((13/12) - (4/\theta_L^2)) \sin^2(\theta_L/2)]; \theta_L = kL$, reflect the geometrical structure of the coupling. As the equilibrium magnetisation is along the $z$ axis, the magnetic resonance signal is contained in $\chi_z(\omega_J)$ and $\chi_y(\omega_J)$, the Fourier transforms of the imaginary part of the susceptibility. Therefore, the total dc current within the Resistively Shunted Josephson junction (RSJ) model is

$$I = \frac{V_0}{R(0)} + \frac{I_c^2(B_0)}{2V_0} R(\omega_J) - I_m,$$

where $R(0)$ and $R(\omega_J) \approx R$ are the junction resistances for dc and frequency $\omega_J$. A simple physical argument can account for the three terms in Eq. (6). The average power dissipated in the junction is $IV_0$ and so the first term, $V_0^2/R$, corresponds to the Ohmic loss at dc, while $\frac{1}{2}I_c^2 R(\omega_J)$ is the similar loss at $\omega_J$. The key third term represents a self-inductance $L(M)$, stemming from the ferromagnet, in parallel with the junction and modeled as an LCR oscillator [see Fig 1(c)], where $R_0$ reflects the magnetic damping. At the magnetic resonance frequency, energy is absorbed by the ferromagnet, causing the oscillator to be lossy. This actually reduces the effective junction resistance, leading to a dip in $I(V)$. In this manner, the Josephson junction rectifies the self-induced magnetic resonance.

This coupling to the magnetic system is evident in the measured dynamical resistance $dV/dI$ curves reported in Fig. 3. We measure the dynamical resistance rather than the IV characteristics to improve amplitude resolution. The mode labeled FMR (Ferromagnetic Resonance) is seen only for ferromagnetic junctions. There is good agreement between the experiment, solid curves, and theory, dotted curves. The magnetic resonance mode observed in our experiments reflects the properties of a thin film of the ferromagnet Pd$_{65}$Ni$_{35}$/Nb, Magnetisation curves $M(H)$, measured directly for a large area Nb/PdNi/Nb trilayer with the same cross section as the junction, are shown in the insert of Fig. 2. They indicate that $\mathbf{M}$ is perpendicular to the junction plane, a conclusion reinforced by earlier anomalous Hall effect measurements on similar thin films. The FMR mode, shown in Fig. 3, occurs at $V_0 = 23 \mu V$. This is unambiguously identified as such, since the frequency $\omega_s = 2\pi V_0/h$ agrees, without fitting parameters, with the Kittel formula

$$\omega_s = \gamma_c \sqrt{(H_K - 4\pi M_S)^2 - H^2}$$

as in the plane magnetic field dependence of the uniform FMR mode when the anisotropy field is perpendicular to the plane. The anisotropy field $H_K = 4900 \text{ G}$ and the magnetisation at saturation $M_S = 930 \text{ G}$ are both determined directly from the static magnetisation data, and $\gamma_c = \mu_B/h$, where $\mu_B$ is the Bohr magneton. For comparison, the ferromagnetic resonance of a microscopic Nb/PdNi/Nb trilayer has been measured in a conventional 9.5 GHz cavity spectrometer at 10 K with field applied parallel to the substrate. The cavity FMR, shown in the bottom insert of Fig. 3, occurs at 2160 G, again exactly as predicted by Eq. (7). Displayed in the top insert of Fig. 3 is the comparison of the resonant mode in the Josephson junction (solid square) with the cavity measurement on a macroscopic trilayer (open square). The dotted curve is a parameter free fit of the FMR using the Kittel formula [Eq. (7)].

![Fig. 3: (color online). Dynamical resistance of the ferromagnetic Josephson junction (solid curve, SFS, bottom and left axis) shows resonances compared to a similar non-ferromagnetic junction (solid curve, SNS, top and right axis). Dotted curve is a fit to theory [Eq. (6)]. The mode at $23 \mu V$ is the ferromagnetic resonance (FMR). Bottom insert: Conventional cavity ferromagnetic resonance on a macroscopic trilayer. Top insert: Comparison between the field dependence of the FMR in the Josephson junction (solid square) with the cavity measurement on a macroscopic trilayer (open square). Dotted curve is a parameter free fit of the FMR using the Kittel formula [Eq. (7)].](image-url)
approximately 10, consistent with the FMR mode measured in a microwave cavity and reported in the bottom insert of Fig. 3.

In order to demonstrate that the magnetic system is coupled to the super- but not to the normal current, we have performed Shapiro step measurements, reported as the dynamical resistance $dV/dI$ in Fig. 4(a). The junction is irradiated with microwaves of frequency $\nu = 17.35$ GHz at 35 mK. The Shapiro steps arise from the mixing of the microwave signal with the ac Josephson effect and are smaller replicas of the zero-voltage current steps due to the drift of the amplifier. The solid curve is Eq. (7) when the spatial dependence of the FMR modes is taken into account. The spatial dependence of the spin-waves leads to an additional term to Eq. (7) given by $ak^2$, where $k=(\pi d/\Phi_0)H$ is the spin wave momentum and $a=E_{ex}b^2$, where $E_{ex} = 50$ meV is the PdNi exchange energy and $b = 0.1$ nm the lattice constant. Since the width of the junction is only about 500 nm, this leads to a small but finite correction to the uniform FMR energy which is larger than the direct effect of the applied dc field. Illustrated in this manner is the direct determination of spin-wave dispersion using the present technique.

In conclusion, we have demonstrated the dynamical coupling of the superconducting phase with the spin waves in a ferromagnet and measured their dispersion. We have performed a photon free FMR experiment on about $10^7$ Ni atoms, which would be infeasible with standard FMR techniques, and have illustrated a new methodology for the study of spin dynamics. There are direct and implied applications to spintronics and nanomagnetism.

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