Superconducting Phase Coherent Electron Transport in Proximity Conical Ferromagnets

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We report superconducting phase-periodic conductance oscillations in ferromagnetic wires with interfaces to conventional superconductors. The ferromagnetic wires were made of Ho, a conical ferromagnet. The distance between the interfaces was much larger than the singlet superconducting penetration depth. We explain the observed oscillations as due to the long-range penetration of an unusual helical triplet component of the order parameter that is generated at the superconductor/ferromagnet interfaces and maintained by the intrinsic rotating magnetization of Ho.

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The superconducting proximity effects in normal (N) nonmagnetic metals [1] can be described by the three characteristic length scales: the coherence length ξ_N, the electron phase breaking length L_ϕ, and the electron mean free path l. The normal metal may become superconducting within ξ_N from the normal/superconducting (N/S) interfaces, while the superconductor-induced changes in the normal conductance may survive even longer distances up to L_ϕ. Here we specialize in proximity effects in thin films, where l belongs to the nanometer scale due to the elastic electron scattering. The values of $\xi_N = \sqrt{\hbar D/k_B T}$ and $L_\phi = \sqrt{D\tau_\phi}$ may reach the micrometer scale at low temperatures; $D = v_F l/3$ is the electron diffusion coefficient, k_B is Boltzmann constant, T is the temperature, v_F is the Fermi velocity, and τ_ϕ^{-1} is the electron phase breaking rate. Normal proximity conductors with the dimensions less than L_ϕ are, in essence, electron quantum interferometers showing periodic conductance oscillations as a function of the superconducting phase difference between the N/S interfaces [2].

When the normal metal is substituted with a ferromagnet (F), the singlet superconducting correlations and phase coherent effects are destroyed by an exchange field within the ferromagnetic coherence length ξ_{F0} which is equal to $\sqrt{\hbar D/I}$ when $I\tau/\hbar < 1$ and equal to l when $I\tau/\hbar > 1$, where I is the exchange splitting of conduction bands and τ is the elastic scattering time [3,4]. This happens because of the splitting of spin-up and spin-down conduction bands so that a wave function of a singlet-state electron pair acquires a fast oscillating part due to the net momentum of the pair. The impurity scattering smears these oscillations and leads to the exponential decay of the condensate function.

The possibility of a long-range condensate penetration at distances larger than ξ_{F0} was not discussed until recently when resistance anomalies in F/S structures were reported [5–7]. The experiments were of two kinds: those with the current flowing through the F/S interfaces and those with the measuring current flowing through the ferromagnetic conductors bypassing the superconductors. It was shown experimentally [8] and theoretically [9] that most resistance anomalies reported in experiments of the first kind can be explained by the contribution of the F/S interfaces. On the other hand, the second type of experiments was proposed to be explained by a long-distance domain redistribution at the onset of superconductivity [10]. However, the latter effect was shown to be too small to account for the superconductor-induced changes in the resistance [11] reviving the question of anomalous long-range penetration of superconducting correlations into F. Several theories were put forward recently suggesting fundamentally new long-range proximity effects (see [12], and references therein).

A mechanism of the triplet proximity effect which is not destroyed by scattering on impurities was first suggested in Ref. [13]. The mechanism is operational in the presence of a rotating magnetization similar to that in the Bloch domain walls. To date, the attempts to observe coherent effects due to presence of domain walls near interfaces have failed because of difficulty to control domain structure at the submicrometer scale. Instead, we chose to build an Andreev interferometer with a ferromagnetic part made of Ho, a rare-earth metal with intrinsic helical magnetic structure. In order to prove experimentally the presence of the triplet proximity effect, one has to observe a coherent transport through F and to exclude the possibility of the singlet proximity effect by choosing $L_F \gg \xi_{F0}$. Observation of conductance oscillations in a F/S interferometer as a function of the superconducting phase difference is an unambiguous proof of coherent electron transport through F.

In this Letter, we report the first measurements of the superconducting phase-periodic conductance oscillations in proximity F wires with the distance between the F/S interfaces more than one order in magnitude larger than the singlet magnetic coherence length ξ_{F0}. We explain the observed oscillations as a direct result of the long-range triplet proximity effect predicted in Ref. [13].

The magnetic structure of Ho is shown in Fig. 1(a) [14]. The total magnetic moment of $10.34\mu_B$ (μ_B is the Bohr magneton) in Ho belongs to a cone with the opening angle $\alpha = 80°$ and rotates, making a helix along the c axis with a turning angle $\theta = 30°$ per interatomic layer with a net moment of $1.7\mu_B$ per atom. Above 21 K, the conical ferromagnetic structure transforms into a spiral antiferromagnetic structure ($\alpha = 90°$) with a Néel temperature of 133 K.

The measured structure had the geometry of the Andreev interferometer shown in Fig. 1(b). The S wires were made of Al, a conventional s-wave superconductor. Six samples were fabricated with the distance L_F between the F/S interfaces equal to 50, 120, 150, 160, 200, and 250 nm. The samples were fabricated using electron-beam lithography and the shadow evaporation technique. This method allowed us to make the whole structure without breaking vacuum, thus avoiding the formation of Ho oxide barriers at the Ho/Al interfaces. The films were thermally deposited at 6×10^{-7} mbar. First, 40 nm of Ho was evaporated at an angle $-14°$, then 60 nm of Al at $+14°$. Thus, an Al loop is created with a Ho segment. The area of the superconducting loop was close to 20 μm^2. Figure 1(b) shows sample geometry and a scanning electron microscope (SEM) micrograph of the F/S part in one of the measured samples. The resistance of the structure was measured using standard four probe technique with electrodes connected as shown in Fig. 1(b), at low frequencies and temperatures between 0.27 and 40 K with a magnetic field up to 2 T applied perpendicular to the substrate. The resistivity ρ was in the range 80–90 $\mu\Omega$ cm for Ho and 0.5–0.6 $\mu\Omega$ cm for Al.

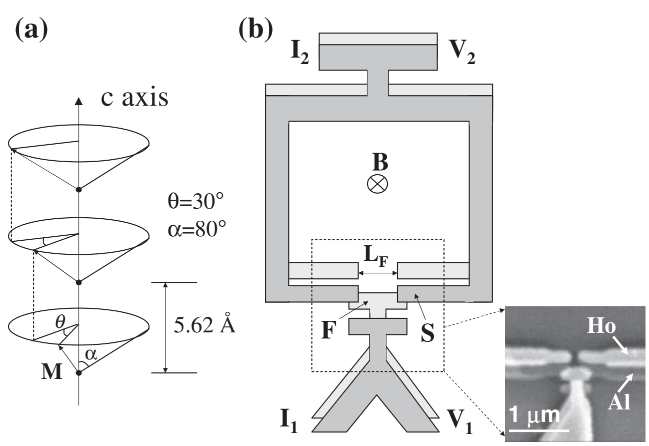

FIG. 1. (a) Magnetic structure of Ho: Magnetization M rotates by 30° each atomic layer along the c axis at an angle of 80° to this axis. (b) Experimental setup and SEM micrograph of the $S/F/S$ junction area prepared by shadow evaporation. Sample with $L_F = 120$ nm.

Direct measurements of the magnetoresistance of Ho films *detached* from superconductors (otherwise deposited in the same conditions as our hybrid structures) showed the ferromagnetic behavior observed earlier [15]. The magnetoresistance shows change of the sign between 10 and 0.3 K [see Fig. 2(a)], which is typical for Ho. The inset in Fig. 2(b) shows the temperature dependence of the resistance of our hybrid structure [Fig. 1(b)]. The sharp drop at $T = 1.2$ K corresponds to the superconducting transition of Al wire and shows that its superconducting properties are practically not affected by the underlying Ho film. At lower temperatures, the resistance increases as shown in Fig. 2(b). This increase correlates with the interface resistance: The lower is the interface transparency; the higher is the increase in resistance as in Ref. [7]. Figure 2(c) shows the differential resistance of the hybrid structure at various temperatures. At low temperatures, an increase in the differential resistance at decreasing voltage is seen, similar to the increase at lowering temperature of Fig. 2(b). The resistance increase at low temperatures or voltages is characteristic to the structures with low transparency interfaces [7–9].

The results of measurements of the oscillations in the resistance of Ho wires as a function of the superconducting phase difference between their interfaces to superconducting Al are shown in Fig. 3(a). The phase difference was created by magnetic flux through the S loop with the F segment [Fig. 1(b)]. The period of oscillations corresponded to the flux quantum $\Phi_0 = hc/2e = 2 \times 10^{-7}$ G cm^2 through the area of the loop with the corresponding sharp peak in the Fourier spectrum of oscillations [Fig. 3(b)]. The zero-field resistance of the structure in Fig. 3 with Al in a superconducting state was $R = 94.3\ \Omega$. This resistance can be written as a sum of contributions from three Ho/Al barriers R_b and that from ferromagnetic wires R_F; $R \approx R_F + (3/2)R_b$. In our structures, R_F is approximately equal to the sheet resistance that is measured to be 20 Ω, so the relative amplitude of conductance oscillations is estimated as $\Delta R/R_F \approx 10^{-4}$. The oscillations have maximum resistance in zero magnetic fields. Specially fabricated test N/S structures with similar geometry and R_b values also showed maximum resistance in a zero magnetic field. This reversal can be explained by the influence of interface resistance; see, e.g., [16]. Out of six measured F/S structures, the oscillations were observed in three samples with L_F equal to 50, 120, and 150 nm. The longer wires of 160, 200, and 250 nm did not show oscillations within our experimental sensitivity of $\Delta R/R$ about 10^{-5}.

To estimate l, we use the value of $\rho l \approx 5 \times 10^{-5}\ \mu\Omega\,\mathrm{cm}^2$ calculated using data [17,18]. This gives us $l \approx 6$ nm. Another estimate can be made using the residual resistance ratio (RRR) of our Ho films. The measured $RRR = R(300\ \mathrm{K})/R(0.3\ \mathrm{K}) \approx 1.5$, suggesting that l in our samples is approximately equal to the electron-phonon mean free path at room temperature that is on the same order as the above estimate. Using $v_F = 10^8$ cm/s, we also obtain electron elastic scattering time

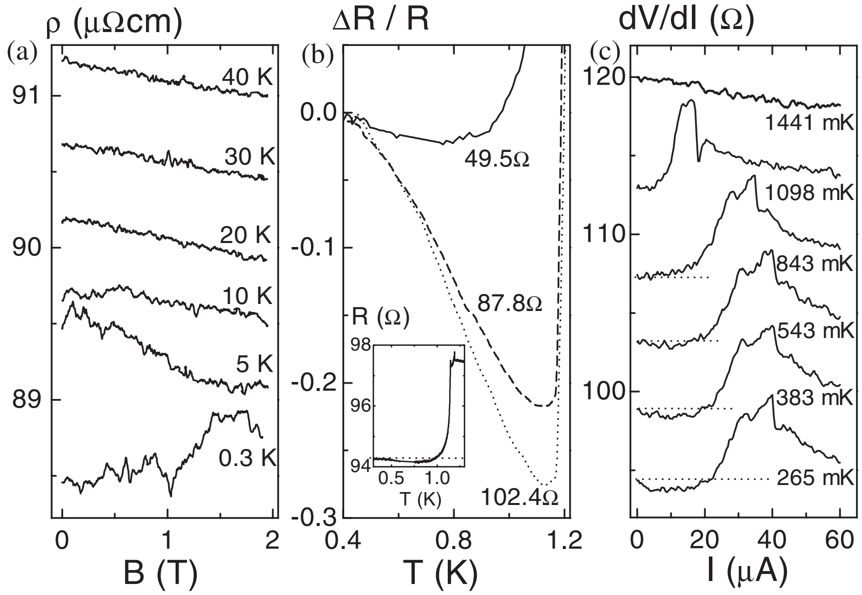

FIG. 2. (a) Magnetoresistance of detached Ho films at different temperatures. (b) Temperature dependence of the normalized resistance at different Ho/Al interface resistance taken at $H = 0$. Solid line: $R_b = 49.5\ \Omega$ ($R = 94.3\ \Omega$, $L_F = 120$ nm); dashed line: $R_b = 87.8\ \Omega$ ($R = 146.7\ \Omega$, $L_F = 50$ nm); dotted line: $R_b = 102.4\ \Omega$ ($R = 183.6\ \Omega$, $L_F = 200$ nm). Inset: Temperature dependence of the resistance of the hybrid Ho/Al structure of Fig. 1(b) at $H = 0$. (c) Differential voltage-current characteristics at various temperatures at $H = 0$ for the sample shown in Fig. 1(b). Curves are offset for clarity.

$\tau = 6 \times 10^{-15}$ s and diffusion constant $D = 20\ \mathrm{cm}^2/\mathrm{s}$. We estimate the value of I in Ho using experimental data for the spin polarization P of conduction electrons in Ho $P = 7\%$ [19]. In the free electron model, we get $I = 2P\epsilon_F$, where $\epsilon_F = 7.7$ eV is the Fermi energy of Ho. Thus, we obtain $I \approx 1.1$ eV. Another estimation of I can be made using the value of the magnetic moment per atom due to conduction electrons, $\mu_{\mathrm{CE}} = 0.34\mu_B$ [19], which gives $I \approx 0.84$ eV in reasonable agreement with the above estimation. Thus, in our case the parameter $I\tau/\hbar \approx 10$, corresponding to the clean limit, so that $\xi_{F0} \approx l$.

The observed resistance oscillations prove that the phase coherence has been established in our ferromagnetic wires through the length L_F up to 150 nm. Such a long-range phase coherence cannot be explained by a proximity effect involving penetration of a singlet order parameter. The upper limit for the singlet penetration length ξ_{F0}, as we mentioned before, is equal to l. Such a short penetration depth rules out a singlet proximity effect as it is attenuated by the factor $\exp(-L_F/\xi_{F0}) = 2 \times 10^{-9}$ at $L_F = 120$ nm. This is in line with the value of $\xi_{F0} = 1.2$ nm obtained from experiments on *SFS* Josephson junctions with a ferromagnetic layer made of Gd [20], another rare-earth element with ferromagnetism due to localized $4f$ electrons and indirect exchange splitting of conduction bands similar to Ho, however, without a ferrocone magnetization structure.

The "helical" triplet superconductivity contains states with spin projections $s = \pm 1$ that are insensitive to the exchange field, as are all other triplet mechanisms considered recently [21–23]. However, its condensate function being odd in the Matsubara frequency (the so called *odd* triplet superconductivity) is even in the momentum p and, therefore, unlike other unconventional condensates, is not destroyed by the presence of nonmagnetic impurities, thus surviving much longer distances than the mean free path of quasiparticles. It is generated in the presence of inhomogeneity of magnetization at the F/S interfaces. Such inhomogeneities, hence the effect, can in principle exist in "usual" ferromagnets within the domain walls explaining experiments [5–7]. However, no existing technology can create them in a controlled way at the F/S interfaces with nanoscale precision. In our Ho conductors, the helical

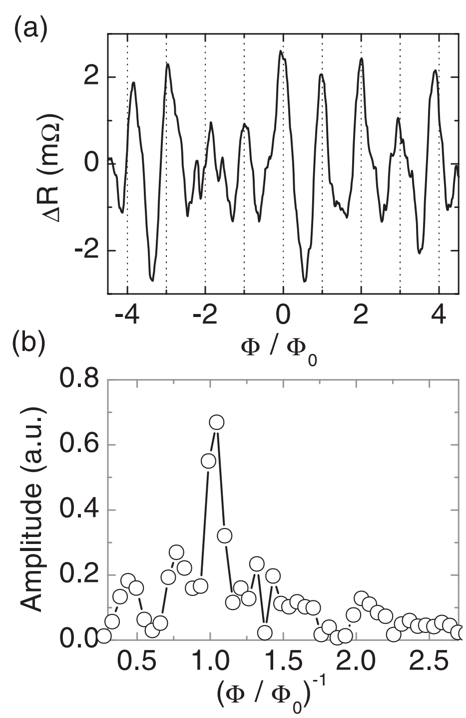

FIG. 3. (a) Magnetoresistance oscillations of the sample shown in Fig. 1(b) measured at $T = 0.27$ K as a function of a normalized external flux through the loop. Sample resistance is $94.3\ \Omega$. $L_F = 120$ nm. (b) Fourier spectrum of the oscillations confirming the $hc/2e$ periodicity.

magnetization is an intrinsic property. The value of the helical triplet coherence length ξ_{F1}, depends on I, τ, the period of magnetization rotation in space L_M, and their relationships. The generalized formula for ξ_{F1} valid for an arbitrary relation between I and $\hbar\tau^{-1}$ can be written as [24]:

$$\xi_{F1}^{-1} = \sqrt{2\pi k_B T/\hbar D + Q^2/(1 + (2I\tau/\hbar)^2)}, \qquad (1)$$

where $Q = 2\pi/L_M$, and L_M is the period of magnetization rotation. In Ho, $L_M = 6.74$ nm. Substituting values for I and τ in (1), we estimate $\xi_{F1} \approx 24$ nm.

The amplitude of the triplet component f_{F1} of the condensate function can be written as [24]

$$f_{F1}(x) = \frac{R_F}{R_b}\frac{\hbar Q v_F}{2I}\exp\left(-\frac{x}{\xi_{F1}}\right), \qquad (2)$$

where x is a space coordinate away from an F/S interface and $x = 0$ at the interface. The factor $\exp(-x/\xi_{F1})$ accounts for exponential decay of the condensate wave function over the characteristic length ξ_{F1} from the F/S interface, a good approximation when $x \gg \xi_{F1}$. We use this form because $L_F > \xi_{F1}$. The amplitude of resistance oscillations depends on interference of the condensate wave functions generated at both interfaces and can be estimated as [24]

$$\Delta R = R_F\left(\frac{R_F}{R_b}\right)^2\left(\frac{l}{L_M}\frac{\pi\hbar}{I\tau}\right)^2\exp(-L_F/\xi_{F1}). \qquad (3)$$

Substituting experimental values in (3), we obtain $\Delta R = 1.2$ mΩ in reasonable agreement with experimentally observed amplitudes [see Fig. 3(a)].

Note that Eq. (3) is obtained by averaging the squared amplitude of the triplet component over angles and the length of the ferromagnet. This averaged quantity does not depend on the orientation of the c axis with respect to the direction of the current, and it is determined mainly by rotation of the magnetization over a length of the order of $2\pi/Q$. Therefore, a possible domain structure cannot affect the result for ΔR (at least qualitatively) provided the period of a domain structure exceeds the period of the spiral $2\pi/Q$. The width δ of domain wall is of the order of $\sqrt{A/K}$, where A is the exchange stiffness constant and K is an anisotropy constant [25]. Using values $A = 8.6 \times 10^{-11}$ J/m and $K = 4 \times 10^4$ J/m^3 [26], we get $\delta \approx 50$ nm. So the above condition is fulfilled in Ho.

In conclusion, we have observed superconducting phase-periodic conductance oscillations in ferromagnetic wires with helical magnetization coupled to a singlet superconductor. The length of the wires was much larger than the singlet order parameter penetration depth, ruling out the conventional proximity effect. We explain the oscillations as due to the long-range penetration of an unusual helical triplet component of superconductivity that is generated in ferromagnetic conductors in the presence of rotating magnetization.

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Note added in proof.—We became aware of the observation of a triplet supercurrent through a SFS junction with a half-metallic ferromagnet by Keizer *et al.* [27].

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