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0 and π phase Josephson coupling through an insulating barrier with magnetic impurities

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We have studied the temperature and field dependencies of the critical current I_C in the Nb-$Fe_{0.1}Si_{0.9}$-Nb Josephson junction with a tunneling barrier formed by a paramagnetic insulator. We demonstrate that in these junctions coexistence of both the 0 and the π states within one tunnel junction occurs, and leads to the appearance of a sharp cusp in the temperature dependence $I_C(T)$, similar to the $I_C(T)$ cusp found for the 0-π transition in metallic π junctions. This cusp is not related to the 0-π temperature-induced transition itself, but is caused by the different temperature dependencies of the opposing 0 and π supercurrents through the barrier.

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As first predicted by Josephson,[1] the supercurrent I_S through the tunnel barrier is driven by the phase difference φ across the junction applied to the superconducting wave function. In conventional Josephson junctions (JJs) this current is described by the relation $I_S = I_C \sin \varphi$, where I_C is the critical current. Recently, considerable attention has been devoted to the investigation of π JJs.[2–4] In this case the relation between the supercurrent and the phase difference is $I_S = I_C \sin(\varphi+\pi) = -I_C \sin \varphi$.[5] One of the possible realizations of a π junction is the superconductor–ferromagnetic metal–superconductor tunnel junction, wherein spatial oscillations of the superconducting order parameter occur in the ferromagnetic metal as a consequence of the exchange splitting of the conduction band.[6] The transition between the 0 and π states was experimentally observed as the vanishing of the Josephson current. The 0-π transition can be induced by varying the barrier thickness[2,4] or the temperature.[3,4] As the absolute value of the current is measured, e.g., for proper values of the ferromagnetic barrier thickness, a sharp cusp in the temperature dependence of the critical current $I_C(T)$ is observed as a consequence of the 0-π transition.

It was also predicted that JJs with magnetic impurities within an *insulating barrier* can produce the π state.[5] Later on, the possibilities of observing π junctions in JJs with ferromagnetic insulating or semiconducting barriers were analyzed theoretically in Refs. 7 and 8. In such types of JJs the proximity effect in the barrier is much weaker, as compared with the ferromagnetic metal junctions, and can be disregarded. In this case the formation of the π junction is caused by quasiparticle scattering on a magnetically active interface.[8] This can result in the splitting of the Andreev interface bound-state energies into two spin channels.[7] Theoretically, if these channels compensate each other the 0-π transition is observed. Up to now the π state in JJs with an insulating magnetic barrier has not been found experimentally.

In this Rapid Communication we present experimental evidence for the existence of the π state in a Josephson junction with magnetic impurities in the insulating barrier. We also demonstrate that the coexistence of both the 0 and π states within one tunnel junction leads to the appearance of a cusp in the temperature dependence of I_C like the one for the 0-π transition in metallic π junctions. The origin of this cusp, however, is not related to the 0-π transition itself, but rather to a simultaneous 0 and π Josephson tunneling through the insulating barrier with magnetic impurities. In this case, quite generally, the tunneling through the insulating barrier itself gives rise to a positive contribution to the critical current I_{C0} (0 part), whereas the tunneling via scattering on magnetic impurities generates a negative $I_{C\pi}$ (π part). These two currents have opposite signs and different temperature dependencies of the critical currents, which results in their complete mutual cancellation $|I_{C0} - I_{C\pi}| = 0$ at a certain temperature where a sharp depression of the critical current has been observed.

We studied a Nb-$Fe_{0.1}Si_{0.9}$-Nb tunnel junction, with the amorphous $Fe_{0.1}Si_{0.9}$ alloy as the barrier. Amorphous magnetic materials here have certain advantages compared to polycrystalline materials, because of the lack of crystalline defects and better composition homogeneity at a microscopic level. Moreover, the $Fe_{0.1}Si_{0.9}$ alloy is additionally favorable because it is an insulator at low temperatures, with the resistance a few orders of magnitude higher than that of metallic alloys. The $Fe_{0.1}Si_{0.9}$ alloy is a paramagnetic material,[9] but the amorphous structure does not rule out completely the possible existence of a local ferromagnetic exchange field at low temperatures. A molecular dynamics *ab initio* simulation reveals that nearest-neighbor positions of the Fe atoms are also quite probable. From that point of view, the formation of microscopic regions with ferromagnetic exchange coupling seems to be possible. In this case the barrier can be thought of as a "nanocomposite," containing regions with and without magnetic exchange coupling.

The Nb-$Fe_{0.1}Si_{0.9}$-Nb junctions were prepared by a sputtering technique under conditions similar to those used for the fabrication of the Nb-Si-Nb junctions.[10] The area of the junction is $20 \times 20\ \mu m^2$. The structure and composition homogeneity of the 6-nm-thick barrier were investigated by transmission electron microscopy (TEM) in a cross-sectional specimen (Fig. 1). These studies have clearly confirmed that the barrier is very well defined. No indications of either strong interdiffusion or local Nb shorts were found. The $Fe_{0.1}Si_{0.9}$ barrier is very homogeneous in thickness as well as in composition.

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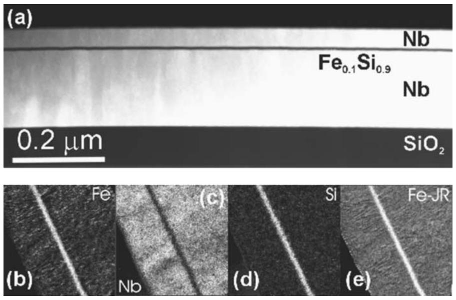

FIG. 1. Cross-sectional micrographs of the Nb-$Fe_{0.1}Si_{0.9}$-Nb tunnel junction obtained in various modes of transmission electron microscope: (a) z-contrast annular dark-field micrograph; (b), (c), and (d) energy-filtered TEM elemental maps of Fe, Nb, and Si, respectively; (e) jump-ratio Fe elemental map. The thickness of the $Fe_{0.1}Si_{0.9}$ barrier is 6 nm.

The current-voltage (IV) characteristics of the Nb-$Fe_{0.1}Si_{0.9}$-Nb junction were measured (Fig. 2), and subsequently the differential conductance (dI/dV) versus the bias voltage was determined numerically (Fig. 3). At both temperatures 4.8 and 2 K peaks are observed at voltages $V=1.77$ and 2.09 mV, respectively. They correspond to the sum of the superconducting gaps related to the individual Nb electrodes of the tunnel junction. The reduced value of the sum of the superconducting gaps is most probably due to a nonideal upper Nb electrode (see below).

At the temperature of 2 K, the critical current is $I_C=17\ \mu$A. As the I_C value is finite, the derivative of the current-voltage curve at the zero bias is infinite. Such IV and dI/dV curves are typical for temperatures and magnetic fields where the junction has a finite I_C value.

The dI/dV curve measured at 4.8 K corresponds to the applied magnetic flux $\Phi/\Phi_0=0.7$ where the maximum of the zero-bias peak was observed (see Fig. 5). This type of dI/dV curve is typical for temperatures and magnetic fields where there is no measurable critical current.

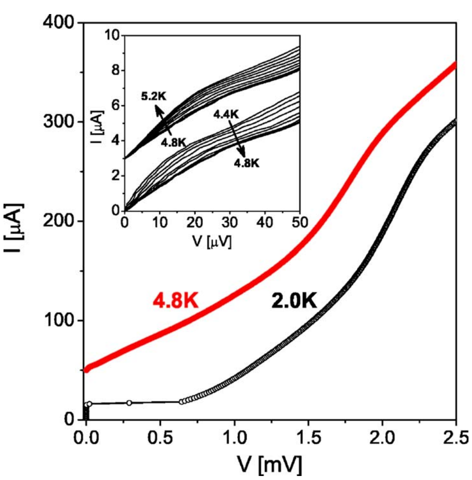

FIG. 2. (Color online) Current-voltage characteristics of the Nb-$Fe_{0.1}Si_{0.9}$-Nb tunnel junction at 4.8 and 2.0 K. The curves are shifted by 50 μA for clarity. Inset: The temperature-dependent change of the slope of the IV curves around 4.8 K. The curves for the range from 4.8 to 5.2 K are shifted by 3 μA for clarity.

For the reference junction Nb-Si-Nb,[10] the upper Nb electrode contains a thin ($\sim$2 nm) sublayer of amorphous Nb adjacent to the silicon barrier. This junction behaves like the SINS system, where N represents the amorphous part of the Nb electrode. Due to the nonequal atomic condensation of Nb on $Fe_{0.1}Si_{0.9}$, a similar amorphous Nb sublayer was also found in Nb-$Fe_{0.1}Si_{0.9}$-Nb junctions. The lack of a measurable critical current at zero bias is caused by the decay of the superconducting order parameter in this amorphous part of the Nb electrode as well as by the Andreev scattering at the interface of the polycrystalline and amorphous Nb. A similar zero-bias peak in dI/dV was described by Klapwijk,[11] and in his case (a Nb-Si-Nb junction) was considered as a precursor of the fully developed supercurrent observed for thinner barriers. In our case the zero-bias peak in dI/dV can be interpreted as a precursor of the supercurrent for the thinner amorphous part of the upper Nb electrode. To obtain information about the precursor of the I_C we consider the integrated zero-bias peak (IZBP) amplitude given by

$$\int_0^{V_C} [dI/dV(V) - dI/dV_{offset}]dV, \qquad (1)$$

where V_C is the voltage criterion (we used $V_C=5\ \mu$V), and dI/dV_{offset} is the reference conductance value. From the IZBP(T) and IZBP(Φ) data (see below) we confirmed that the zero-bias peak is the precursor of I_C.

Direct measurements of I_C at zero magnetic field reveal some finite values up to a temperature around 4 K (see Fig. 4). Above this temperature, instead of I_C the zero-bias peak is observed in the dI/dV curves. To find out what happens above 4 K, we have determined the IZBP for each used temperature $T>4$ K. As can be seen from Fig. 4, the proposed method reveals a sharp IZBP cusp at the temperature of 4.8 K [compare the temperature-dependent change of the slope of the IV curves around zero bias in the inset in Fig. 2], a maximum at approximately 6 K, and then a decrease down to zero at $T\approx7$ K which is the critical temperature of our Nb-$Fe_{0.1}Si_{0.9}$-Nb JJ.

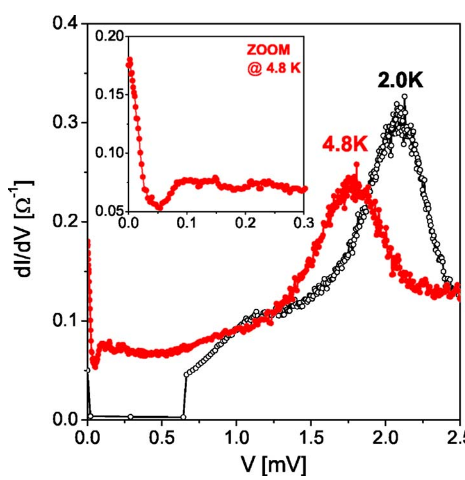

FIG. 3. (Color online) Differential conductance vs bias voltage characteristics of the Nb-$Fe_{0.1}Si_{0.9}$-Nb junctions at 4.8 and 2.0 K. Inset: Zoom, showing the zero-bias conductance peak at 4.8 K.

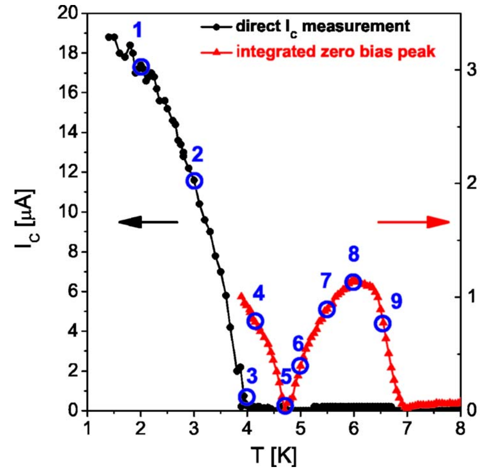

FIG. 4. (Color online) The temperature dependencies of the critical current $I_C(T)$ and the integrated zero-bias peak IZBP(T) of the Nb-$Fe_{0.1}Si_{0.9}$-Nb tunnel junction in zero magnetic field. IZBP(T) can be considered as a zoom of the $I_C(T)$ dependence in the temperature range from 4 up to 7 K. The circles denotes the points where $I_C(\Phi)$ was measured (see Fig. 5).

Figure 5 shows the critical current vs applied magnetic flux $I_C(\Phi)$ dependencies for the temperatures marked in Fig. 4 by circles. Curves 1, 2, and 3 show a peak around zero field with finite values of I_C. Elsewhere, the $I_C(\Phi)$ dependence is suppressed and was, therefore, obtained using the IZBP versus the applied magnetic flux. As for the IZBP versus temperature, the IZBP was found for each value of the magnetic flux. In what follows the IZBP together with the I_C temperature and magnetic field dependencies will be referred to as a single $I_C(T)$ or $I_C(\Phi)$ dependence.

The unusual behavior of the junction in a magnetic field is clearly seen from Fig. 5. As the temperature decreases, the shape of $I_C(\Phi)$ changes. In particular, in the interval of magnetic flux $\Phi \in \langle -\Phi_0;\Phi_0\rangle$ it is clear that the middle peak gradually vanishes as the temperature increases (curves 1–4). At the temperature of 4.8 K (curve 5) a minimum of the critical current at zero applied flux $I_C(0)$ is observed. Such behavior with a minimum of critical current at $\Phi=0$ is typical for 0-π JJs.[12] Then for temperatures $4.8 < T < 6$ K the shape of the $I_C(\Phi)$ curves is changing again and $I_C(0)$ recovers its zero-field maximum (curves 6–9). It is worth noting that in a reference Nb-Si-Nb JJ $I_C(\Phi)$ shows a well-defined conventional Fraunhofer-like pattern.[13]

As predicted by Bulaevskii *et al.*[12] a perfectly flat and homogeneous insulating barrier with magnetic impurities can induce the formation of an admixture of π and 0 junctions ("vortex states") for a certain range of barrier parameters and temperatures. Since the 0-π phase boundary corresponds to the nucleation of a semifluxon, this vortex phase is, in fact, a collection of semifluxons formed at the 0-π barrier boundaries. Another possible reason for the coexistence of the π and 0 phases could be the barrier thickness modulation or/and formation of the Fe clusters. However, taking into account the very homogeneous and flat boundaries (Fig. 1), the pure paramagnetic behavior, and the lack of electron diffraction rings typical for nanocrystallites in an amorphous ma-

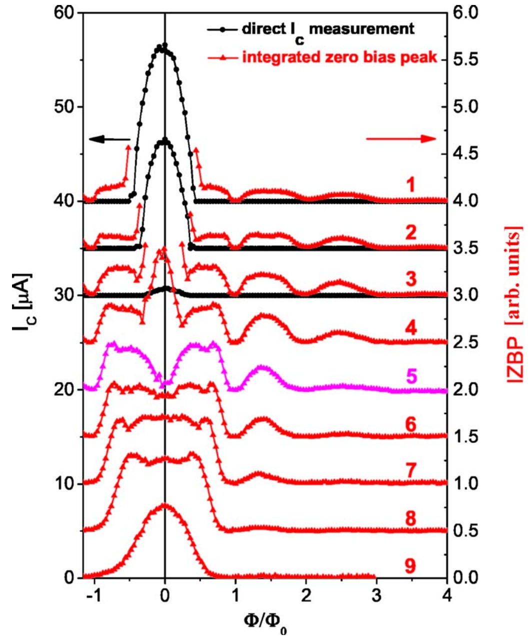

FIG. 5. (Color online) The applied magnetic flux dependencies of the critical current $I_C(\Phi)$ and integrated zero-bias peak IZBP(Φ) of the Nb-$Fe_{0.1}Si_{0.9}$-Nb tunnel junction for temperatures marked in Fig. 4.

trix, the latter scenario seems to be less probable.

In our case the coexistence of the 0 and π junctions can be simulated as a JJ with a nonuniform spatial distribution of the critical current density and with additional polarity alternations. The assumption about the simultaneous presence of 0 and π tunneling is confirmed by the unusual shape of the $I_C(\Phi)$ curves (Fig. 5). When both 0 and π phases of the Josephson supercurrent coexist in one Nb-$Fe_{0.1}Si_{0.9}$-Nb junction, two $I_C(T)$ dependencies must be taken into account: $I_{C0}(T)$ and $I_{C\pi}(T)$ (Fig. 6). Due to the lack of adequate theo-

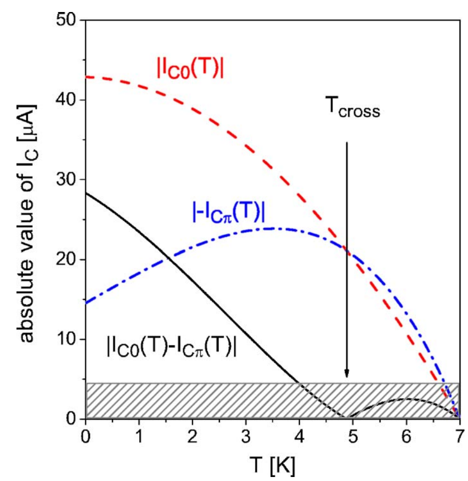

FIG. 6. (Color online) Simplified illustrative model of the Nb-$Fe_{0.1}Si_{0.9}$-Nb junction: The theoretical partial temperature dependencies of the critical current for the 0 and π parts and their sum, showing the cusp in $I_C(T)$ caused by crossing of the $I_{C0}(T)$ and $I_{C\pi}(T)$ curves. Shaded area: The levels of the I_C measured by the IZBP due to the absence of a measurable critical current.

ries for this kind of junction the $I_{C0}(T)$ and $I_{C\pi}(T)$ dependencies were calculated by using the theory of π junctions with a metallic barrier.[14,15] In this illustrative simulation $I_{C0}(T)$ and the $I_{C\pi}(T)$ are taken for the same barrier thickness but with different values of the ferromagnetic exchange energy, the decay length ξ_{F1}, and the oscillation period of the order parameter $2\pi\xi_{F2}$ (details will be provided elsewhere). The sum of these two currents of opposite polarities (positive for I_{C0} and negative for $I_{C\pi}$) gives the $I_C(T)$ dependence, which is similar to the one we found (compare Figs. 4 and 6). The minimum of the $I_C(T)$ dependence, T_{cross}, corresponds to the crossing point of the $|-I_{C\pi}(T)|$ and $|I_{C0}(T)|$ dependencies (see Fig. 6).

In conclusion, we have observed the coexistence of 0 and π states in Nb-$Fe_{0.1}Si_{0.9}$-Nb Josephson junctions with a paramagnetic insulating barrier formed by amorphous $Fe_{0.1}Si_{0.9}$. The different temperature dependencies of the I_{C0} and $I_{C\pi}$ currents and their opposite signs lead to the appearance of a very sharp cusp in the $I_C(T)$ curve at about 4.8 K where these two currents cancel each other completely. The simultaneous presence of both the 0 and π phases in the Nb-$Fe_{0.1}Si_{0.9}$-Nb junction has been interpreted in terms of the vortex state model proposed by Bulaevskii *et al.* for JJs with an insulating barrier with magnetic impurities. An adequate detailed theory that fully describes our experimental data is currently lacking, and further interactions between theory and experiment are needed to reveal the nature of the phase-shifting effect in JJs with an insulating paramagnetic barrier.

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