

Measurement of the intrinsic higher harmonic current-phase relation in NbN/NiCu/NbN Josephson junctions

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We report on the measurement of the intrinsic higher harmonic current-phase relation for epitaxial NbN/NiCu/NbN (SFS) Josephson junctions. We investigated the π -0 phase transition as a function of temperature for SFS Josephson junctions with a 3.2-nm-thick NiCu layer, and fitted the phase transition temperatures $T_{\pi-0}$ with a theoretical expression derived from the Usadel equations. We found that junctions on the same chip and with the same dimension showed different phase transition temperatures depending on the values of the fitting parameter α . The fitting parameter α indicates the magnetic scattering intensity, which mainly derives from spin-flip or spin-flop scattering in the NiCu layer. At the π -0 crossover, the intrinsic positive second harmonic current-phase relation was confirmed in a junction with moderate magnetic scattering by the nonvanishing critical current, half-integer Shapiro steps, and half-periodic Fraunhofer modulation.

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In ballistic and diffusive Josephson junctions, such as superconductor–normal metal–superconductor (SNS) or superconductor-ferromagnet-superconductor (SFS) junctions, the current-phase relation (CPR) is quite different from that in superconductor-insulator-superconductor (SIS) tunneling junctions because of Andreev reflection at the interface [1]. The phase difference $\phi = \pi$ in SFS Josephson junctions with a minimum Josephson coupling energy has drawn considerable attention, especially after the recommendation of applying both to superconducting digital [2–4] and quantum circuits [5–7]. The so-called π junctions have been realized by varying the thickness of the ferromagnet layer [8,9] or temperature [10,11]. The extraordinary CPR is not confined to the π -phase shift, but higher harmonics, such as $\sin(2\phi)$, have also been theoretically predicted [12,13] and verified by experiments [14–16]. For a more general CPR expression, the Josephson critical current can be written as $I_c = I_1 \sin \phi + I_2 \sin(2\phi)$ with $I_2 \ll I_1$ [17,18]. However, not all SFS Josephson junctions show a $\sin(2\phi)$ dependence near $T_{\pi-0}$ [14], because the CPR in a SFS Josephson junction depends not only on the thickness of the ferromagnet layer but also on the magnetic properties of the ferromagnet layer, such as magnetic scattering, exchange energy, interface transparency, and so on [19]. Although magnetic scattering has been taken into consideration when fitting the critical current dependence on ferromagnet thickness [20,21] and temperature [11], reports on the influence of magnetic scattering intensity on the phase transition temperature and the observation of higher harmonic CPR are lacking.

In this Rapid Communication, we report that magnetic scattering can markedly affect the π -0 crossover temperature

and the observation of the intrinsic positive second harmonic CPR in a NbN/NiCu/NbN Josephson junction. The damping length of the second harmonic, which is half that of the basic sinusoidal component in the CPR, decreases sharply under heavy magnetic scattering. Moderate magnetic scattering enabled the observation of a non-negligible positive $\sin(2\phi)$ component in our NbN-based SFS Josephson junction, as confirmed indirectly by a nonzero critical current, a half-integer Shapiro step under 300-MHz microwave radiation, and the half-periodic Fraunhofer modulation at the π -0 crossover.

The spatial distribution of the superconducting order parameter in the ferromagnet contributes to the oscillation of the Josephson critical current. In an SFS Josephson junction, the phase transition depends on the relative values of damping length ξ_{F1} and oscillating length ξ_{F2} as shown by Eq. (1), where D is the electron diffusion constant, k_B is the Boltzmann constant, and E_{ex} is the exchange energy. The π -0 phase transition in SFS Josephson junctions can be observed by changing the F layer thickness d_F around half of the order parameter spatial oscillation period $\sim 2\pi\xi_{F2}$. From Eq. (1), ξ_{F1} and ξ_{F2} are not only related to the exchange energy E_{ex} , but are also determined by temperature T . As temperature increases, the damping length ξ_{F1} decreases whereas ξ_{F2} increases. Therefore, the π -0 phase transitions in SFS Josephson junctions can also be measured with varying temperature,

$$\xi_{F1,2} = \sqrt{\frac{\hbar D}{[E_{ex}^2 + (\pi k_B T)^2]^{1/2} \pm k_B T}}. \quad (1)$$

Our NbN-based SFS Josephson junctions were prepared through a multistep fabrication process by magnetic sputtering and optical lithography. Full details of this process have been previously reported [22]. All the junctions referred to in this

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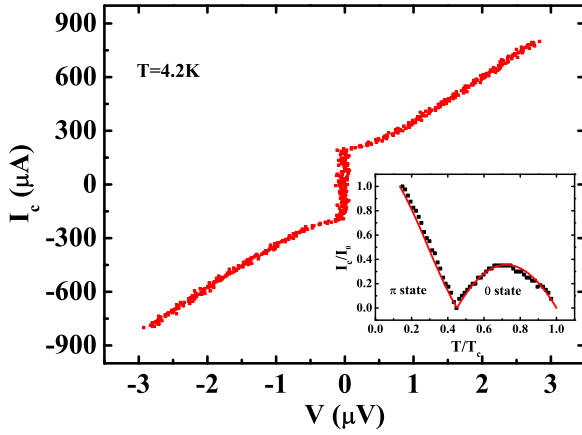


FIG. 1. I - V curve of an SFS Josephson junction with a 3.2-nm NiCu layer measured at 4.2 K. The inset shows the temperature dependence of the critical current of the same junction and the red line is fitted by Eq. (2).

Rapid Communication are $2 \mu\text{m} \times 2 \mu\text{m}$ in size. Figure 1 shows the I - V curve of the junction with a 3.2-nm-thick NiCu layer measured at 4.2 K. The critical current of this junction is $180 \mu\text{A}$. The inset shows the temperature dependence of I_c of the same junction, and for clarity both the temperature T and critical current I_c have been normalized. The critical current formally decreased to zero and then changed sign at $T = 0.46T_c$, indicating a phase transition from the π - to 0-coupling state. The red line was fitted with Eq. (2), a theoretical expression of I_c in SFS Josephson junctions derived from the Usadel equations [9,23]

$$I_c = I_{c0} \left(\frac{T}{T_c} \right) \text{Re} \left(\sum_{n=0}^{\infty} \frac{\mathcal{F}(n) q_1(n) \exp(-\frac{q_1(n) d_F}{\xi_F})}{[\sqrt{q_2(n) \mathcal{F}(n) + 1} + 1]^2} \right), \quad (2)$$

with functions

$$\begin{aligned} \mathcal{F}(n) &= \Delta^2(T) / [\omega_n + \sqrt{\omega_n^2 + \Delta(T)^2}]^2, \\ q_1(n) &= \sqrt{2(i + \alpha + \tilde{\omega}_n)}, \\ q_2(n) &= (i + \tilde{\omega}_n) / (i + \alpha + \tilde{\omega}_n), \\ \tilde{\omega}_n &= \omega_n / E_{\text{ex}} = \pi(2n + 1)k_B T / E_{\text{ex}}, \\ \alpha &= \hbar / (\tau_s E_{\text{ex}}), \end{aligned}$$

where I_{c0} is a constant prefactor, $\xi_F = 1 \text{ nm}$ is the coherence length adopted for the ferromagnetic layer, n is an integer, $\Delta(T)$ is the superconducting energy gap, τ_s is the magnetic scattering time, and \hbar is the reduced Planck constant. The value of $\alpha = 1.27$ indicates the degree of the magnetic scattering effect. Because the crossover temperature is much smaller than T_{Curie} , the exchange energy E_{ex} dependence on temperature can be neglected and assumed to be a constant parameter. As will be shown below, the transparency of our interfaces is high, so that we need not consider a transparency parameter.

Figure 2 demonstrates the temperature dependence of the critical current of four SFS Josephson junctions fabricated on the same chip with a 3.2-nm NiCu layer. All of those junctions have the same junction dimension of $2 \mu\text{m} \times 2 \mu\text{m}$.

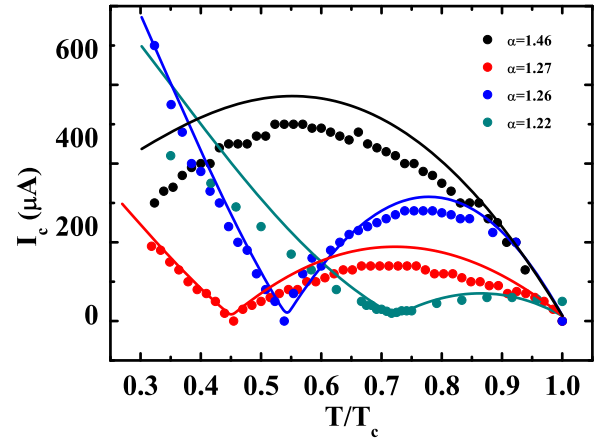


FIG. 2. Temperature dependence of the critical current of four SFS Josephson junctions with the same scale and NiCu layer thickness (3.2 nm) on the same chip. Lines are fittings with different magnetic scattering parameters α . $T_c = 13 \text{ K}$ due to the influence of the wiring NbN layer.

It is evident, as shown in Fig. 2, the junctions have different critical currents and phase transition temperatures, in spite of these junctions having the same junction size and being fabricated on the same chip. The spatial variation of the NiCu barrier thickness or inhomogeneity of the NiCu exchange energy might lead to the critical current deviations in SFS Josephson junctions. For example, a 0.6-nm deviation of the F layer thickness can result in a completely different coupling state [14]. However, this thickness effect should not be a factor for our SFS Josephson junctions, because the surface roughness of our NiCu film deposited on 200-nm NbN was only 0.24 nm. Magnetic scattering derives from spin-flip or spin-flop scattering owing to anisotropy in ferromagnets as a non-negligible element in SFS Josephson junctions, which can reduce the damping length ξ_{F1} and increase the oscillating length ξ_{F2} , and ultimately affect the critical current behaviors. The role of spin-orbit scattering can be neglected in our experiment for relatively small atomic numbers Z of NiCu.

As a weak magnetic alloy, the magnetic scattering in the NiCu barrier may heavily influence the phase transition process in our SFS Josephson junction. The inverse scattering time $\hbar\tau_s^{-1}$ is of the order of the average exchange field E_{ex} or even greater, which might considerably modify the proximity effect in the SF structure and in turn affect the phase transition temperature. We fitted the temperature dependence of those four junctions in Fig. 2 according to Eq. (2) with different values of α . A larger α value indicates a heavier magnetic scattering in the NiCu layer with the same thickness. Since magnetic scattering has the same effect as increasing temperature on a decrease of ξ_{F1} and an increase of ξ_{F2} , so SFS Josephson junctions on the same chip but with heavier magnetic scattering can realize a phase transition at much lower temperatures. We see that when the value of α is 1.46, there will be no phase transition observable (above 4.2 K), as shown in Fig. 2.

At the crossover of the phase transition, the first term of $I_c = I_1 \sin \phi + I_2 \sin(2\phi)$ must tend to zero for changing sign, and the second order I_{c2} should become the dominate

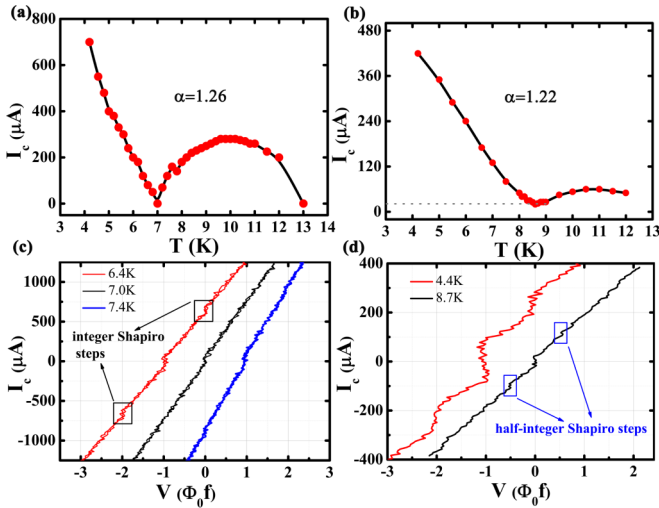


FIG. 3. $I_c(T)$ of SFS Josephson junction (a) with high magnetic scattering and (b) with moderate magnetic scattering. (c), (d) I - V curves measured under 300-MHz microwave radiation. The integer Shapiro steps highlighted by the black frame were observable near $T_{0-\pi}$ for the junction fitted with $\alpha = 1.26$ at 6.4 K, whereas a half-integer Shapiro step highlighted by the blue rectangle was observed for the junction fitted with $\alpha = 1.22$ at 8.7 K.

component. However, I_{c2} makes such a small contribution to the total critical current I_c that it is usually difficult to measure [13,24]. For a SFS Josephson junction, the higher harmonic CPR could be destroyed owing to a short damping length and become unobservable under heavy magnetic scattering. As the junctions showed in Fig. 2, the critical currents decrease to zero at the crossover, preventing the observation of higher harmonics in the CPR. However, for the junction with the smallest α value, there was a nonzero critical current at the crossover, which might indicate the existence of a higher harmonic component. Further measurements might be able separate the higher harmonic CPR and testing error.

Figures 3(a) and 3(b) (i.e., some of the results from Fig. 2 before normalization) show the $I_c(T)$ of junctions that are fitted with parameters $\alpha = 1.26$ and $\alpha = 1.22$, respectively. The junction with heavier magnetic scattering transferred from the π - to 0-coupling state at 7 K and the critical current decreased to zero, whereas the junction with moderate scattering transferred at a much higher temperature (~ 8.5 K) with a finite critical current of approximately $20 \mu\text{A}$. The half-integer Shapiro steps near $T_{\pi-0}$ are convincing evidence of a second harmonic CPR but can only be detected over a narrow temperature range [16]; thus, we measured the Shapiro step near the crossover in a temperature with steps of 0.1 K. Under 300-MHz microwave irradiation, we observed integer Shapiro steps near the crossover, as highlighted by the black frame in Fig. 3(c), based on the same junction shown in Fig. 3(a). The blue rectangle in Fig. 3(d) indicates the positions of the half-integer Shapiro steps at 8.7 K for the same junction shown in Fig. 3(b). The half-integer Shapiro steps are almost obscured by noise because of the environment conditions and poor coupling between the microwave and device. For clarity, the lines are shifted by $\pm\Phi_0 f$.

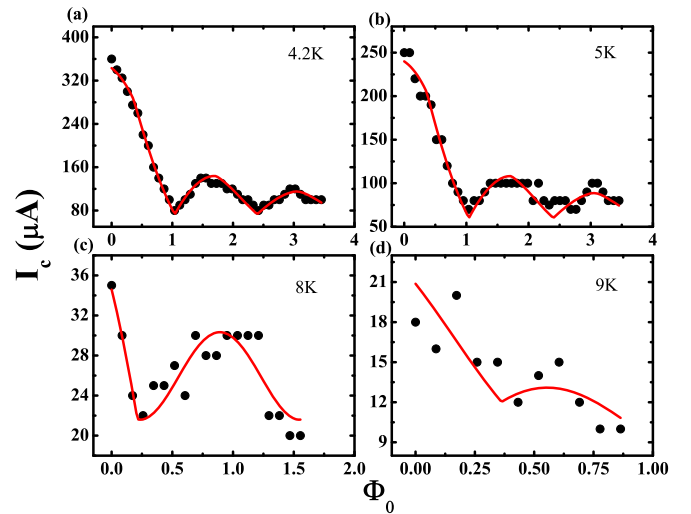


FIG. 4. Magnetic diffraction patterns (in plane) at different temperatures for the junction fitted with $\alpha = 1.22$ and with nonzero critical current at crossover. (a) Magnetic diffraction at 4.2 K, (b) 5 K, (c) 8 K, and (d) 9 K. Red lines indicate fits to the data by $\sin \phi$ and $\sin(2\phi)$ expressions. Minima at the half-integer flux and maxima at zero magnetic flux indicate the positive intrinsic second harmonic CPR.

The normal state resistances R_n , which mainly derive from the interface resistance in our SFS Josephson junctions, are approximately 1.2 and 3.5 $\text{m}\Omega$ for the junctions in Figs. 3(c) and 3(d), respectively; hence, the critical current in Fig. 3(a) is much larger than that in Fig. 3(b) at 4.2 K, although with a heavier magnetic scattering. The interface resistance due to elastic scattering originates from a deviation of the lattice orientation between NbN and NiCu films. The interface transparency parameters calculated by $\gamma_B = R_B/\rho_F \xi^*$ were 0.65 and 2.5, respectively, where R_B is the interface resistance, $\rho_F = 51 \mu\Omega \text{cm}$ is the NiCu resistivity, and the characteristic spatial scale $\xi^* = \sqrt{(\hbar D)/2\pi k_B T_c} = 1.2 \text{ nm}$. The interface transparencies are sufficiently high to support the fitting approximation mentioned above [25].

The phase transition should be discontinuous for SFS Josephson junctions with a uniform ferromagnetic barrier thickness, because the intrinsic second harmonic is always positive at the crossover [13,26]. For a nonuniform F layer, there will be a negative second harmonic term because of the spontaneous circulating supercurrent between the π - and 0-coupling segments [14]. The nonzero critical current and half-integer Shapiro steps at the crossover are not sufficient to determine the sign of the second harmonic component.

The magnetic interference pattern can also manifest the existence of the dominant second harmonic CPR at the crossover [15,27]. For the same junction shown in Fig. 3(b), we measured the critical current behavior with an in-plane magnetic field according to the easy magnetization axis of NiCu film through a vector field coil at different temperatures. The critical current measured at 4.2 K with a zero magnetic flux is slightly different from that in Fig. 3(b) because of the two different measurement systems. Figures 4(a) and 4(b) show a normal Fraunhofer-like pattern at 4.2 and 5 K, which

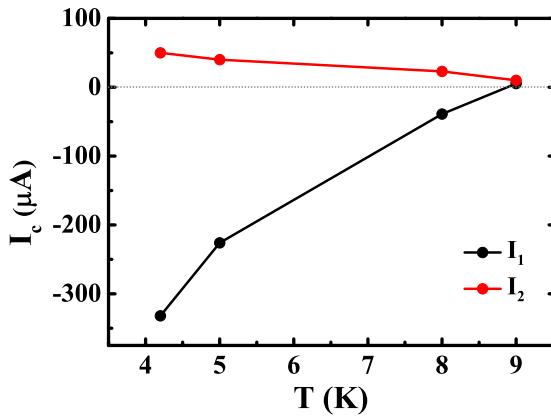


FIG. 5. Temperature dependence of I_1 and I_2 extracted from Fig. 4 for the junction fitted with $\alpha = 1.22$. The solid lines simply join the points and are guides to the eye.

is far away from $T_{\pi-0}$, but the ratio of $I_c(H)/I_c(0)$ at the first-side maximum is approximately 0.4 larger than that of $2/3\pi$ for a conventional Josephson junction, which indicates a weak higher harmonic component [26]. When the first sinusoidal term decreased to zero and the sign inverted at the crossover, the $I_c(H)$ dependence in Figs. 4(c) and 4(d) showed clear half-periodic diffractions at 8 and 9 K with critical current minima at $\Phi_0/2$. The temperatures were maintained by pulling the probe out of liquid helium; thus, there may be some deviation from the real values. Both I_1 and I_2 were extracted by fitting those measured results in Fig. 4 based on the current-phase relation $I_c = I_1 \sin \phi + I_2 \sin(2\phi)$. Figure 5 shows the temperature dependence of I_1 and I_2 . The second harmonic changed weakly and had a positive sign over the entire temperature range, whereas the sign of I_1 changed from negative to positive in the temperature range of 8–9 K due to a phase transition. The magnitude of I_2 was consistent with the finite value in Fig. 3(b), and also consistent with $I_2/I_1 \sim 0.1 \exp(-d/\xi_{F1})$ [13] if we take the same value $\xi_{F1} = 4$ nm as the same parameter in fitting the NiCu thickness dependence of the first sinusoidal critical current [28]. The abnormal ratio of $I_c(H)/I_c(0)$ and first minima at $\Phi = \Phi_0/2$ of the Fraunhofer patterns at the crossover indicates the existence of second harmonic CPR, the maximum critical currents at zero magnetic flux, and the positive sign of I_2 over the

entire temperature range, confirming the intrinsic origin of the second harmonic CPR.

In summary, we have measured and discussed the temperature dependence of the critical current of NbN/NiCu/NbN SFS Josephson junctions. For junctions on the same chip, the phase transition temperatures decreased under heavy magnetic scattering. The difference of magnetic scattering intensity for junctions on the same chip can be ascribed to magnetic anisotropy and an inhomogeneous distribution of the Ni element in the NiCu layer. The finite critical current and half-integer Shapiro steps at the crossover can only be detectable in a junction with moderate magnetic scattering, indicating the existence of a fragile higher harmonic CPR. Through fitting the Fraunhofer patterns at different temperatures with $\sin \phi$ and $\sin(2\phi)$ dependence, we extracted the positive second harmonic critical current over the entire temperature range, which means it is an intrinsic second harmonic CPR rather than originating from the spontaneous supercurrent flowing around the interfaces of the π - and 0-coupling segments. By measuring the nonzero critical current, half-integer Shapiro steps, and half-periodic Fraunhofer modulation of a single NbN-based SFS Josephson junction with moderate magnetic scattering, we verified the existence of the intrinsic positive second harmonic CPR indirectly with a much simpler construction. Nowadays, we are also working on a direct measurement of the CPR of NbN-based SFS Josephson junctions with asymmetric superconducting quantum interference devices (SQUIDs) and other structures, consisting of NbN-based SIS and SFS Josephson junctions [29,30]. The second harmonic current may be another noise source in the digital and quantum circuits that incorporated π Josephson junctions, and heavier magnetic scattering may be demanded in order to suppress this kind of noise. The confirmation of the existence of the second harmonic CPR may accelerate a comprehensive understanding of its origin and dissipation. It may also shed light on the investigation of spin-triplet current which can also give birth to the second harmonic current in nonhomogeneous SFS Josephson junctions.

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