Understanding the Josephson Current through a Kondo-Correlated Quantum Dot

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(Received 25 January 2012; published 30 May 2012)

We study the Josephson current 0-π transition of a quantum dot tuned to the Kondo regime. The physics can be quantitatively captured by the numerically exact continuous time quantum Monte Carlo method applied to the single-impurity Anderson model with Bardeen-Cooper-Schrieffer superconducting leads. For a comparison to an experiment, the tunnel couplings are determined by fitting the normal-state linear conductance. Excellent agreement for the dependence of the critical Josephson current on the level energy is achieved. For increased tunnel couplings the Kondo scale becomes comparable to the superconducting gap, and the regime of the strongest competition between superconductivity and Kondo correlations is reached; we predict the gate voltage dependence of the critical current in this regime.

DOI: 10.1103/PhysRevLett.108.227001 PACS numbers: 74.50.+r, 05.60.Gg, 72.15.Qm, 73.21.La

Introduction.—Recently, hybrid superconductor-quantum dot devices have attracted much attention [1] due to their peculiar physical behavior determined by the interplay of superconductivity of the leads and the level characteristics of the dot. Applications in nanoelectronics or quantum-information processing are envisaged. Among other properties, dc Josephson transport [2–7] was intensively studied. Similar to the Josephson effect of ordinary tunnel junctions [8], a difference $\phi \neq 0, \pi$ of the order parameter phases of the two superconductors with gap Δ leads to an equilibrium Josephson current J running through the system [2–7]. The focus was on carbon nanotube dots [2,4–7] with well separated single-particle levels, i.e., level broadening Γ and temperature T much smaller than the level spacings, simplifying the modeling as a single-level dot with energy ϵ can be considered.

It is well established both theoretically [9] and experimentally [3–7] that the local Coulomb interaction, i.e., the dot charging energy U, can lead to a 0-π transition of the quantum dot Josephson junction, associated to a first-order (level-crossing) quantum phase transition from a singlet (0) to a doublet (π) ground state [10]. In fact, a variation of any of the system parameters U, ϵ, Δ, ϕ as well as the tunnel couplings $\Gamma_{L/R}$ (with $\Gamma = \Gamma_L + \Gamma_R$) can be used to tune the system across the phase boundary, if the others are taken from appropriate ranges. At $T = 0$, the transition leads to a jump in J from a large and positive (0 phase) to a small and negative value (π phase). At finite temperatures, it is smeared out and significantly diminished, yet the sign change of J is clearly observed in SQUID setups [3,4,7]. The experimental challenge in observing the true magnitude of the Josephson current to be compared with theoretical predictions consists in suppressing uncontrolled phase fluctuations, which can be achieved by using designed on-chip circuits [5,6]. In such experiments, J is tuned by a variation of a gate voltage V_g which translates into a rather controlled change of ϵ [11].

The physics becomes particularly interesting if the dot is tuned to a parameter regime in which Kondo correlations [12] become relevant for suppressed superconductivity. It is characterized by the appearance of the Kondo scale (at odd dot filling) $k_B T_K = \sqrt{\Gamma U/2}\exp(-\pi U/8\Gamma)$ [12]. Kondo physics is important if $k_B T \lesssim k_B T_K \ll \Gamma$, with k_B denoting the Boltzmann constant. In this regime perturbative methods in either U, such as self-consistent Hartree-Fock (HF) [13], or Γ [9] become uncontrolled. Even for $\Delta \gg k_B T_K$, at which superconductivity prevails, one expects Kondo correlations to have a significant impact on J. These were partly incorporated using a method developed for large Δ values [14]. Other techniques successfully used for Kondo-correlated quantum dots with normal leads, such as the noncrossing approximation (NCA) [15], numerical renormalization group (NRG) [16,17], (Hirsch-Fye) quantum Monte Carlo (QMC) calculations [18], and functional renormalization group (fRG) [17] were extended to the present setup. With superconducting leads, they suffer from significant conceptual or practical limitations such as half filling of the dot level (NRG) and high (NCA, QMC) or zero (fRG) temperature and, therefore, cannot be used for a quantitative comparison to experiments performed at temperatures on the order of a few tens of mK and with a wide span of gate voltages [5,6]. The regime of the strongest competition between superconductivity and Kondo correlations is reached for $\Delta \approx k_B T_K$. For typical experimental gap sizes of $\Delta \approx 0.1$ meV [5–7], in this regime $k_B T_K \ll \Gamma$ is no longer fulfilled. Still, even for

$k_B T_K \lesssim \Gamma$, a precursor of Kondo correlations is expected to stabilize the singlet phase and perturbative methods become unreliable.

Recently, the continuous time QMC method, called CT-INT in what follows, was introduced as a new tool to study correlated quantum dots with Bardeen-Cooper-Schrieffer (BCS) leads [19]. Here, we exploit the exceptional flexibility and accuracy of this approach and compute J as well as the normal-state linear conductance G for the parameters of the experiment of Ref. [5]. Our simultaneous analysis of J and G reveals that the dot shows significant Kondo correlations, but superconductivity prevails as $\Delta \approx 10 T_K$. In the normal state, it lies in the interesting and theoretically challenging parameter regime with $k_B T \approx k_B T_K \approx \mu_B h$, where $\mu_B h$ (with the Bohr magneton μ_B) denotes the scale associated to the applied Zeeman field h used to destroy superconductivity. Compared to previous approaches, we are now able to quantitatively study this experimentally relevant parameter regime with a numerically exact method and find excellent agreement between the experimentally measured critical current J_c and the numerically computed one for both the 0 and π phases (see Fig. 1). We show that due to the fairly large left-right asymmetry of the tunnel couplings and the finite temperature, the current-phase relation $J(\phi)$ is rather sinusoidal even close to the 0-π transition (see Fig. 2), providing an *a posteriori* justification of the extraction of J_c from the measured current-voltage characteristics of the on-chip circuits applying the extended resistively shunted-junction (RSJ) model [5,6]. Finally, using the parameters of the experiment, but increasing Γ such that $\Delta \approx k_B T_K$ we compute the gate voltage dependence of the current in the regime of the strongest competition between superconductivity and (precursors of) Kondo correlations (see Fig. 3).

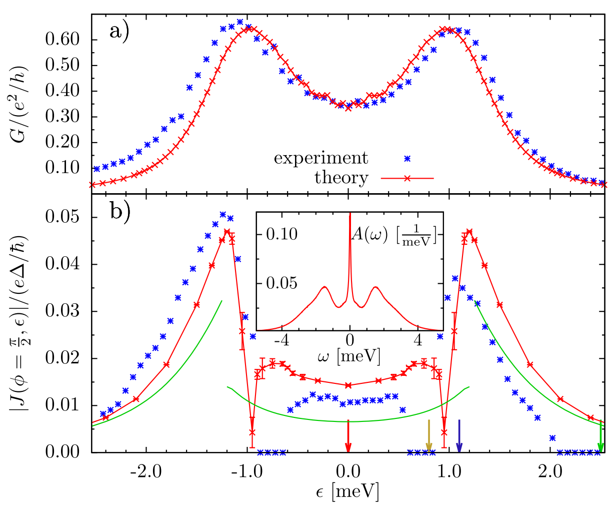

FIG. 1 (color online). Comparison of experimental data [5] with the numerically exact solution of the superconducting Anderson model. (a) Best fit of the normal-state linear conductance with applied magnetic field used for extracting values of Γ and Γ_L/Γ_R (for details see the main text). (b) Measured critical current vs theoretically calculated Josephson current at $\phi = \frac{\pi}{2}$ (CT-INT, symbols with line; self-consistent Hartree-Fock, thin lines). The arrows indicate the level positions for which the current phase relation is presented in Fig. 2. Inset: Normal-state spectral function at $\epsilon = 0$.

Model and method.—For the description of the single-level quantum dot with superconducting leads we use the Anderson impurity model with Hamiltonian $H = H^{\rm dot} + \sum_{s=L,R} H_s^{\rm lead} + \sum_{s=L,R} H_s^{\rm coup}$. The dot part reads

$$H^{\rm dot} = \sum_\sigma \epsilon_\sigma d_\sigma^\dagger d_\sigma + U\left(d_\uparrow^\dagger d_\uparrow - \frac{1}{2}\right)\left(d_\downarrow^\dagger d_\downarrow - \frac{1}{2}\right) \quad (1)$$

in standard second quantized notation. In the presence of a Zeeman field h, the single-particle energies depend on the orientation of the spin $\epsilon_\sigma = \epsilon + g\mu_B h\sigma/2$, with the Landé g factor $g = 2$ [20] and $\sigma = \pm 1$. The energy is shifted such that for $h = 0$, $\epsilon = 0$ corresponds to the point of half-filling of the dot. The left ($s = L$) and right ($s = R$) superconducting leads are modeled by BCS Hamiltonians

$$H_s^{\rm lead} = \sum_{k\sigma} \epsilon_{sk} c_{sk\sigma}^\dagger c_{sk\sigma} - \Delta \sum_k (e^{i\phi_s} c_{sk\uparrow}^\dagger c_{s-k\downarrow}^\dagger + {\rm H.c.}), \quad (2)$$

where (without loss of generality) $\phi_L = -\phi_R = \phi/2$. The quantum dot is coupled to the leads by $H_s^{\rm coup} = \sum_{k,\sigma}(t_{sk} c_{sk\sigma}^\dagger d_\sigma + {\rm H.c.})$. We assume energy-independent dot-lead hybridizations $\Gamma_s = \pi \sum_k |t_{sk}|^2 \delta(\epsilon_F - \epsilon_{sk})$, with the Fermi energy ϵ_F.

The CT-INT is based on an interaction expansion of the partition function in which *all* diagrams are summed up stochastically. The method is numerically exact and allows the calculation of thermodynamic observables with any required precision $\sigma_{\rm MC}$ (indicated by error bars in the figures) with the practical limitation that the computing time grows as $1/\sigma_{\rm MC}^2$. Details can be found in Ref. [19]. Here we go far beyond the proof-of-principle study of Ref. [19] by considering $\epsilon \neq 0$, larger U/Γ as well as left-right coupling asymmetries. Furthermore, we

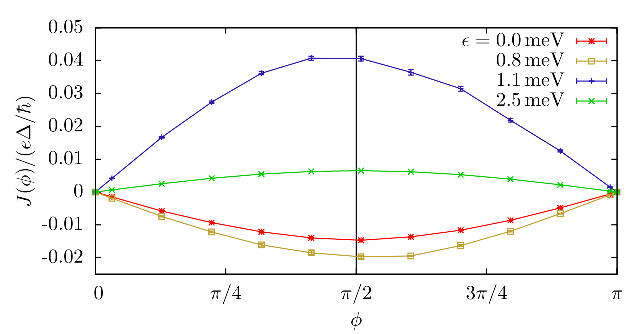

FIG. 2 (color online). Josephson current-phase relation for the parameters of the experiment [5] at values of ϵ indicated by arrows in Fig. 1. It is rather sinusoidal even very close to the critical value of ϵ (1.1 and 0.8 meV), and the critical current is thus well approximated by $J(\phi = \frac{\pi}{2})$.

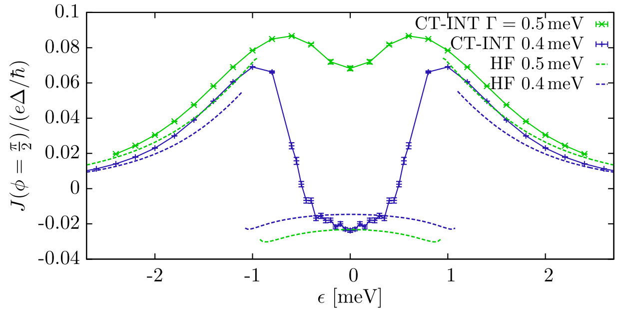

FIG. 3 (color online). Josephson current at $\phi = \frac{\pi}{2}$ for the parameters of the experiment (see Fig. 1) but with increased level broadening $\Gamma = 0.4$ and 0.5 meV (instead of 0.27 meV) and thus increased T_K. Self-consistent Hartree-Fock is obviously unable to describe the strong competition between superconductivity and Kondo correlations in this parameter regime and leads to a spurious π phase for parameters for which the numerical exact solution only shows a precursor close to half dot filling $\epsilon = 0$.

compute the normal-state linear conductance in the challenging regime $k_B T \approx k_B T_K \approx \mu_B h$.

The Josephson current is computed as the expectation value of the left (or right) current operator $J = ie/\hbar \sum_{k\sigma} \langle t_{Lk} c^\dagger_{Lk\sigma} d_\sigma - \text{H.c.}\rangle$. The noninteracting lead degrees of freedom are integrated out, and one arrives at a formula for the Josephson current in terms of the imaginary-frequency Nambu-Green function $\mathcal{G}(i\omega_n)$ of the dot only (directly accessible in CT-INT) [19]

$$J = 2\,\mathrm{Im\,Tr}\Bigg[\frac{1}{\beta}\sum_{i\omega_n} \frac{\Gamma_L}{\sqrt{\Delta^2 + \omega_n^2}} \times \begin{pmatrix} i\omega_n & -\Delta e^{-i\phi/2} \\ \Delta e^{i\phi/2} & -i\omega_n \end{pmatrix} \mathcal{G}(i\omega_n)\Bigg]. \quad (3)$$

As our second observable, we investigate the normal-state linear conductance $G = \sum_\sigma G_\sigma$ with

$$G_\sigma = \frac{e^2}{\hbar} \frac{2\Gamma_L \Gamma_R}{\Gamma_L + \Gamma_R} \int_{-\infty}^{\infty} A_\sigma(\omega)\left(-\frac{df(\omega)}{d\omega}\right) d\omega, \quad (4)$$

where A_σ denotes the normal-state dot spectral function and f the Fermi function. The computation of A_σ from the (normal-state) imaginary frequency Green function $\mathcal{G}_\sigma(i\omega_n)$ obtained numerically by CT-INT is based on analytical continuation. It is found that the calculation of G_σ is much more reliable if the method detailed in Ref. [21] is used. As shown there, the conductance can be written as

$$G_\sigma = \frac{e^2}{\hbar} \frac{2\Gamma_L \Gamma_R}{\Gamma_L + \Gamma_R} \frac{2}{\beta} \sum_{\alpha > 0} R_\alpha \,\mathrm{Im}\, \frac{d\mathcal{G}_\sigma(i\tilde{\omega}_\alpha)}{d\tilde{\omega}_\alpha}, \quad (5)$$

where the frequency derivative of the Green function has to be evaluated at imaginary frequencies $i\tilde{\omega}_\alpha$, which can differ from the Matsubara ones [22] given together with the weights R_α in Ref. [21]. Within CT-INT, $\mathcal{G}_\sigma$ is accessible only at the Matsubara frequencies. Therefore, we introduce a (real) Padé approximant $\mathcal{G}_P(\omega) = \sum_{j=0}^{M-1} a_j \omega^j / \sum_{j=0}^{M} b_j \omega^j$ of degree $(M, M+1)$ and minimize the function

$$\chi^2(\{a_i\},\{b_i\}) = \sum_{n,m} \{\mathcal{G}_P(\omega_n) - \mathrm{Im}[\mathcal{G}_\sigma(i\omega_n)]\} C^{-1}_{n,m} \{\mathcal{G}_P(\omega_m) - \mathrm{Im}[\mathcal{G}_\sigma(i\omega_m)]\},$$

where C is the carefully bootstrapped estimate of the covariance matrix of the QMC data $\mathrm{Im}[\mathcal{G}_\sigma(i\omega_n)]$. The degree of the Padé approximant $(M, M+1)$ is chosen such that the minimal χ^2 is not smaller than the number of degrees of freedom to obtain a statistically consistent fit and is found to be surprisingly small with $M = 3, \ldots, 6$. The Padé approximant may now be safely evaluated at the positions $i\tilde{\omega}_\alpha$, and Eq. (5) can be used.

Comparison to the experiment.—In experiments, the charging energy U can be determined accurately from the height of the Coulomb blockade diamonds obtained by bias spectroscopy in the normal state. The same type of measurement in the superconducting state reveals sharp features at the gap position from which Δ can be extracted [5,6]. In addition, T and h are known within tight bounds. The parameters which are most delicate to determine but strongly affect J are the level width Γ and the asymmetry Γ_L/Γ_R. Based on this insight, we proceed as follows. (i) The parameters Δ, U, T, and h are taken directly from the experiment. Those and the comparison of theoretical curves for the normal-state conductance $G(\epsilon)$ with the experimental ones are used for obtaining accurate estimates of Γ, Γ_L/Γ_R, and the gate conversion factor α which relates the change of ϵ to a variation of the gate voltage V_g according to $V_g = \alpha\epsilon$ [24]. (ii) For the complete parameter set determined this way, we compute the Josephson current and compare to the measured J_c.

We focus on the most symmetric conductance double peak presented in Fig. 4d of Ref. [5]. The experimental parameter estimates with errors of approximately 10% are $U \approx 3$ meV, $\Delta \approx 0.1$ meV, $T \approx 75$ mK, and $h \approx 150$ mT. In Fig. 1(a), we show our best fit of $G(\epsilon)$ to the experimental result from which we extract $\Gamma = 0.27$ meV, $\Gamma_L/\Gamma_R = 9.3$, and $\alpha = 0.011$ V/meV. At fixed U the peak separation and the peak to valley ratio are determined by Γ whereas the overall height is given by Γ_L/Γ_R, as can be inferred from Eq. (5) (in $\mathcal{G}_\sigma$, only $\Gamma = \Gamma_L + \Gamma_R$ enters). Note that Γ turns out to be significantly smaller and Γ_L/Γ_R significantly larger than the values extracted in Ref. [5] based on the assumption that the dot is in the Coulomb blockade regime. However, our analysis allowing for Kondo correlations clearly reveals that those are relevant for $U/\Gamma \approx 11.15$ and the Kondo scale $k_B T_K \approx 8$ μeV. It is roughly an order of magnitude smaller than Γ and of the order of the temperature ($k_B T = 6.5$ μeV) as well as the Zeeman energy ($\mu_B h = 8.7$ μeV). Therefore, neither T

nor h can be neglected when considering the normal state; the conductance is characterized by a split Kondo plateau (ridge) [25], not to be mistaken with the Coulomb blockade peaks which would be located at larger energies $\epsilon \approx \pm U/2 \approx \pm 1.5$ meV. As an inset we show, for illustration, the normal-state spectral function at $\epsilon = 0$ for the extracted parameters obtained from analytic continuation of CT-INT data onto the real frequency axis by the maximum entropy method [26]. The appearance of a sharp zero-energy resonance is a hallmark of Kondo correlations [12]. The splitting of the Kondo resonance by the Zeeman field is too small to be observable on the scale of the plot (but present in the data).

In the experiments [5,6] J_c, defined as the maximum of $|J(\phi)|$ over $\phi \in [0, \pi]$, is extracted from current-voltage characteristics of the on-chip circuits using an extension of the standard RSJ model [27]. In this analysis it is assumed that $J(\phi)$ is purely sinusoidal with its maximum at $\phi = \frac{\pi}{2}$. At first glance, this appears to be at odds with what is known theoretically for the current-phase relation of a Josephson quantum dot in the 0 phase (half-sinusoidal with maximum at $\phi \to \pi$) and the transition region (jump from $J > 0$ to $J < 0$ at $T = 0$, smeared out by $T > 0$) [14–18]. However, as it was shown already in Ref. [5] for an effective noninteracting model, the sizable left-right asymmetry and the finite temperatures of the experimental setups imply sinusoidal currents in the 0 and π phase apart from very narrow ranges around the 0-π transitions. This conclusion is confirmed by the numerically exact CT-INT in Fig. 2, where we present $J(\phi)$ for the above given parameters at the level positions indicated by the arrows in Fig. 1(b) showing $|J(\phi = \frac{\pi}{2}, \epsilon)|$. Apparently, only for ϵ values very close to the transition the ϕ position of the maximal current $|J|$ deviates observably from $\frac{\pi}{2}$, and yet the maximal value is still very close to that of $|J(\phi = \frac{\pi}{2})|$. This gives an *a posteriori* justification of the extraction of J_c using the extended RSJ model and allows us to focus on the current at $\phi = \frac{\pi}{2}$ when comparing to the gate voltage dependence of the critical current, as done in Fig. 1(b). Without any additional fitting parameters, we achieve excellent agreement in both the 0 (to the left and right of the peaks) and the π phase (central region with nearly ϵ-independent J_c). In addition, we show $|J(\phi = \frac{\pi}{2}, \epsilon)|$ obtained for the same parameters using the HF approach [13,17]. Whereas in the empty dot and doubly occupied regime $|\epsilon| \gtrsim 2$ meV this mean-field approximation gives decent agreement with the exact result (CT-INT; see also Fig. 3), it apparently fails in the mixed valence regime and for half dot filling ($\epsilon \approx 0$) in which Kondo correlations are crucial. Important features like the smoothing of the phase transition by the finite temperature and the smooth crossing through zero of $J(\frac{\pi}{2})$ cannot even be obtained qualitatively. This emphasizes that Kondo correlations are relevant even in the presence of prevailing superconductivity ($\Delta \approx 10 T_K$) [28].

Increasing T_K.—Considering $\Gamma = 0.4$ and 0.5 meV with all the other parameters fixed at the values given above, we finally investigate the regime $\Delta \approx k_B T_K$ of the strongest competition between superconductivity and (precursors of) Kondo correlations. In Fig. 3, $|J(\phi = \frac{\pi}{2}, \epsilon)|$ obtained by CT-INT is compared to HF results. Obviously, the singlet (0) phase is stabilized by the correlations—an effect which HF is unable to describe. For the largest T_K value (at $\Gamma = 0.5$), $|J(\phi = \frac{\pi}{2}, \epsilon)|$ computed by CT-INT only shows a precursor of the π phase (the slight suppression close to $\epsilon = 0$) whereas HF gives a spurious π phase. It would be very interesting to measure the gate voltage dependence of the critical current for dots falling into this parameter regime, which would confirm the predictive power of our calculations.

Summary.—We presented a thorough study of the Josephson-current 0-π transition of a quantum dot in the Kondo regime. A quantitative agreement to the measured dependence of the critical current on the gate voltage for a dot with Kondo correlations but prevailing superconductivity was achieved. This shows that our minimal model is sufficient to quantitatively capture the relevant physics and qualifies the CT-INT as a theoretical tool with predictive power for transport properties of correlated quantum dots. We further studied the regime of the strongest competition between superconductivity and Kondo correlations, confirming qualitatively that the latter stabilize the singlet state and thus the 0 phase and predicting quantitatively the supercurrent, which can be experimentally verified.

We are grateful to the authors of Refs. [5,6], in particular K. Grove-Rasmussen, R. Deblock, and A. Eichler for discussions. This work was supported by the DFG via FOR1346 (D. L. and F. A.) and KA3360-1/1 (C. K.) and by the Czech Science Foundation via Grant No. 204/11/J042 (T. N.).

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