

Spectrum of Andreev Bound States in a Molecule Embedded Inside a Microwave-Excited Superconducting Junction

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Nondissipative Josephson current through nanoscale superconducting constrictions is carried by spectroscopically sharp energy states, the so-called Andreev states. Although theoretically predicted almost 40 years ago, no direct spectroscopic evidence of these Andreev bound states exists to date. We propose a novel type of spectroscopy based on embedding a superconducting constriction, formed by a single-level molecule junction, in a microwave QED cavity environment. In the electron-dressed cavity spectrum we find a polariton excitation at twice the Andreev bound state energy, and a superconducting-phase-dependent ac Stark shift of the cavity frequency. Dispersive measurement of this frequency shift can be used for Andreev bound state spectroscopy.

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Supercurrents through mesoscopic or nanosized Josephson junctions are mainly carried by spectroscopically sharp subgap bound states, so-called Andreev bound states (ABS) [1], as predicted theoretically in Refs. [2,3]. When the weak link is a quantum dot with a single or a few discrete levels, supercurrents are also predicted to flow mainly through ABS [4]. ABS come in pairs, one state above and one below the Fermi level. The two ABS of the pair have opposite dispersion with the superconducting phase difference over the junction and carry supercurrent in opposite directions across the junction. These two states form a well-defined two-level system that has been suggested as a qubit [5,6]. Before such a qubit can be realized, experimental detection and characterization of this engineered two-level system should be carried out. To our best knowledge, however, no one has to date reported experiments with direct spectroscopic proof for the existence of Josephson current carrying ABS [7].

A single-wall carbon nanotube (SWCNT) embedded between two superconducting metal leads can support a supercurrent [8–13]. The electronic energy levels of the SWCNT, formed through size quantization, can be tuned in and out of resonance with the lead Fermi levels by gating the SWCNT. Such superconducting SWCNT transistors [11] can further be switched from a Coulomb blockade regime to a Kondo regime to a weakly interacting Fabry-Perot regime by changing a back gate voltage [12]. The potential for applications of such SWCNT quantum-level junctions was demonstrated through the fabrication and detailed functional control of a nano-SQUID, involving two gated SWCNT junctions with controlled on-off states as well as controlled $0-\pi$ SQUID states [13]. Similar control has been demonstrated using semiconducting nanowire junctions [14–16].

We propose a method for direct ABS spectroscopy, based on dispersive measurement of a polaritonic state formed by the ABS strongly coupled to a QED cavity mode. Consider two superconducting leads connected by

a molecule with one resonant level, as shown in Fig. 1. This setup can be realized in the SWCNT transistor by tuning the voltage of the gate electrode [11–13]. The superconducting proximity effect leads to a split of the resonant level into a pair of ABS, where the ABS level splitting can be controlled by tuning the superconducting phase difference in a SQUID setup. When the gate electrode is coupled to an LC oscillator, induced quantum fluctuations of the gate potential lead to dressed Andreev bound states. By tuning the ABS into resonance with the cavity, and measuring the shift of the cavity base frequency, the ABS energy can be observed. The complete ABS energy dispersion with respect to the superconducting phase can be extracted by using a cavity with a variable base frequency

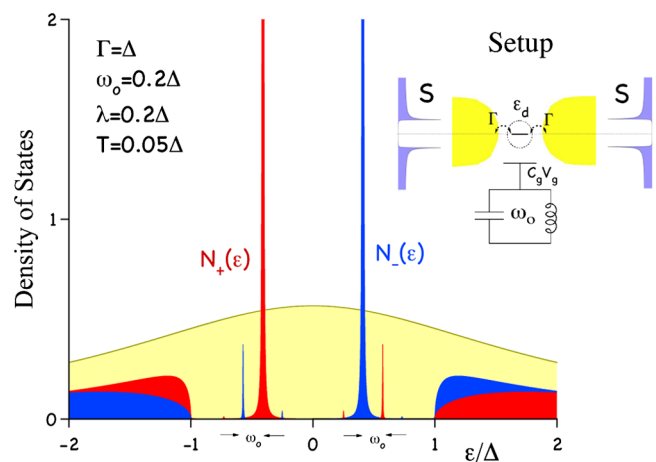


FIG. 1 (color online). Superconducting density of states for $\varphi = 0.6\pi$ resolved into right, $N_+(\epsilon)$, and left, $N_-(\epsilon)$, current-carrying branches. The lightly shaded background is the density of states of the resonant level when the contacts are in the normal state. The electron-oscillator coupling leads to dressed Andreev bound states with sidebands separated in energy by the oscillator frequency ω_0 . The inset shows a sketch of the considered system.

[17–21]. The suggested dispersive method does not involve real interlevel transitions and could be simpler to realize than methods based on microwave absorption [22]. Another advantage of the suggested spectroscopy is that it makes it possible to directly access and characterize individual ABS without measuring dc contact current-voltage characteristics [23]. A similar dispersive measurement method has earlier been applied to detect quantum states of superconducting charge qubits [24].

The Hamiltonian for the coupled electron-oscillator system depicted in the inset of Fig. 1 is

$$\hat{H} = \hat{H}_L + \hat{H}_R + \hat{H}_{\text{level}} + \hat{H}_T + \hat{H}_{\text{osc}} + \hat{H}_{\text{level-osc}} \quad (1)$$

and will be quantified in terms of creation operators for the three excitations of the system: reservoir electrons, $\hat{c}_{k\sigma,\alpha}^\dagger$ (reservoir $\alpha = L, R$, momentum k , and spin σ); dot electrons, \hat{d}_σ^\dagger ; and the oscillator mode \hat{b}^\dagger . The two superconducting reservoirs are described by a standard BCS Hamiltonian $\hat{H}_\alpha = \sum_{k\sigma} \xi_k \hat{c}_{k\sigma,\alpha}^\dagger \hat{c}_{k\sigma,\alpha} + \sum_k (\Delta_\alpha \hat{c}_{k\uparrow,\alpha}^\dagger \hat{c}_{-k\downarrow,\alpha}^\dagger + \text{H.c.})$, where H.c. denotes Hermitian conjugate, ξ_k is the quasiparticle dispersion, and $\Delta_\alpha = \Delta(T) e^{i\varphi_\alpha}$ are the order parameters of the reservoirs that have the same temperature dependent gap $\Delta(T)$, but have a tunable superconducting phase difference $\varphi = \varphi_R - \varphi_L$ between them. The reservoirs are coupled by tunneling through a single noninteracting molecular level described by $\hat{H}_{\text{level}} = \sum_\sigma \varepsilon_d \hat{d}_\sigma^\dagger \hat{d}_\sigma$, where the level energy $\varepsilon_d(V_g)$ is tunable by the gate voltage. The lead-to-level tunneling is described by $\hat{H}_T = \sum_{k\sigma,\alpha} (v_{k\sigma,\alpha} \hat{c}_{k\sigma,\alpha}^\dagger \hat{d}_\sigma + \text{H.c.})$. We consider symmetric, spin-independent coupling between the level and the two leads, $v_{k\sigma,L} = v_{k\sigma,R} \equiv v \delta_{k,k_F}$, which corresponds to an effective tunneling rate at each barrier $\Gamma = \frac{|v|^2}{2\pi} \mathcal{N}_F$, where \mathcal{N}_F is the normal state density of states of the leads at the Fermi level. The oscillator mode with frequency ω_0 is described by the Hamiltonian $\hat{H}_{\text{osc}} = \omega_0 \hat{b}^\dagger \hat{b}$. The coupling of the molecular level to the oscillator is described by a linear interaction $\hat{H}_{\text{level-osc}} = \sum_\sigma \lambda (\hat{b} + \hat{b}^\dagger) \hat{d}_\sigma^\dagger \hat{d}_\sigma$, where λ is the coupling strength. We will assume throughout this Letter that the molecular level is aligned with the Fermi levels of the leads, i.e., $\varepsilon_d = 0$.

We solve the Hamiltonian (1) treating the tunneling to infinite order in the hopping [25–27] and the electron-oscillator coupling perturbatively in a self-consistent Born approximation [28–33]. The problem reduces to considering a molecular level coupled to an oscillator. Superconductivity modifies the electronic state on the molecular level by splitting it into two branches ($s = \pm 1$) each described by a retarded Green function

$$G_s^R(\varepsilon) = \frac{\bar{\Omega}(\varepsilon)}{z_s^R(\varepsilon)\varepsilon^R + s\Delta \cos(\varphi/2)}, \quad (2)$$

where the energy-renormalization factor $z_s(\varepsilon)$ is defined by $z_s^R(\varepsilon)\varepsilon = \tilde{\varepsilon}^R + \bar{\Omega}(\varepsilon)[\varepsilon^R - \Sigma_{-s}^R(\varepsilon)]$, with $\bar{\Omega}(\varepsilon) = \sqrt{|\Delta|^2 - (\tilde{\varepsilon}^R)^2}/2\Gamma$. The state of the oscillator is given by the retarded Green function

$$D^R(\omega) = \frac{2\omega_0}{(\tilde{\omega}^R)^2 - \omega_0^2 - 2\omega_0 \Pi^R(\omega)}. \quad (3)$$

The electron-oscillator coupling enters via the retarded self-energies $\Sigma_s^R(\varepsilon)$ and $\Pi^R(\omega)$ which are determined self-consistently by numerical iteration. $\Sigma_s^R(\varepsilon)$ and $\Pi^R(\omega)$ are functionals of $G_s^R(\varepsilon)$ and $D^R(\omega)$ [and of $G_s^K(\varepsilon) = -2i \text{Im} G_s^R(\varepsilon) n_e(\varepsilon)$, with $n_e(\varepsilon) = \tanh(\varepsilon/2T)$ and $D^K(\omega) = -2i \text{Im} D^R(\omega) n_b(\omega)$, with $n_b(\omega) = \coth(\omega/2T)$, $G_s^A = (G_s^R)^*$] and defined as

$$\Sigma_s^R(\varepsilon) = i \frac{\lambda^2}{2} [D^K(\omega) \circ G_s^R(\varepsilon - \omega) + D^R(\omega) \circ G_s^K(\varepsilon - \omega)], \quad (4)$$

$$\Pi^R(\omega) = -i\lambda^2 \sum_{s=\pm 1} [G_{-s}^R(\varepsilon) \circ G_s^K(\varepsilon - \omega) + G_{-s}^K(\varepsilon) \circ G_s^A(\varepsilon - \omega)], \quad (5)$$

where the convolution is defined as $a(x) \circ b(x - y) = \int_{-\infty}^{\infty} \frac{dx}{2\pi} a(x)b(x - y)$. Upon reaching self-consistency the Josephson current is calculated as

$$I(\varphi) = \frac{e}{\hbar} \Delta \sin \frac{\varphi}{2} \sum_{s=\pm 1} \int_{-\infty}^{\infty} \frac{d\varepsilon}{2\pi} s \text{Im} \left[\frac{G_s^R(\varepsilon)}{\bar{\Omega}(\varepsilon)} \right] n_e(\varepsilon). \quad (6)$$

Inelastic coupling to the environment is introduced phenomenologically above by $\tilde{\varepsilon}^R = \varepsilon^R + i\gamma$ and $\tilde{\omega}^R = \omega^R + i\kappa$. The parameter $\gamma \ll \Delta$ describes a residual phase-breaking scattering rate in the superconducting reservoirs. In the absence of an electron-oscillator coupling it is γ that limits the lifetime of the ABS. κ describes the finite (long) lifetime of the mode originating from a finite (but high) quality factor Q , such that $\kappa = \omega_0/Q \ll \omega_0$. In the present calculations we set $\gamma = \kappa = 10^{-3}\Delta$.

Without coupling to the oscillator, the Josephson effect in this system is well known [4]. The Green function amplitudes G_s^R describe two different quasiparticle branches that form on the dot: one having an ABS below ($s = +1$) and one having an ABS above ($s = -1$) the Fermi level; see Fig. 1. The two branches contribute to the Josephson current (6) in opposite manner. For subgap energies $G_s^R(|\varepsilon| < \Delta)$ contributes to the current in the positive direction for $s = 1$ (left to right over the junction) and vice versa for the $s = -1$ branch. The continuum part of $G_s^R(|\varepsilon| > \Delta)$ contributes to the current in the opposite direction compared with its corresponding subgap part.

The electron-oscillator interaction couples the two quasiparticle branches. This is seen explicitly in the energy-renormalization factor z_s of one branch which is modified by the self-energy Σ_{-s}^R of the other branch. In Fig. 2 we show a fully self-consistent calculation of the Andreev spectrum as a function of superconducting phase difference. For the chosen parameters there is a resonance between the oscillator and the ABS, i.e., $2E_A(\varphi) = \omega_0$, at $\varphi \approx 0.9\pi$. Away from resonance, the ABS is shifted

$$E_A \rightarrow \bar{E}_A = E_A + \frac{\lambda^2 \mathcal{A}_E^2}{2} \left(\frac{\Phi_+}{2E_A + \omega_0} + \frac{\Phi_-}{2E_A - \omega_0} \right) \quad (7)$$

as compared with the case without coupling to the oscillator, but with retained phase-dependent spectral weight \mathcal{A}_E of the state at $\varepsilon = \pm E_A(\varphi)$. The effective electron-oscillator coupling in the subgap region is given by the product $\lambda \mathcal{A}_E$. In Eq. (7) thermal occupation factors enter in the combinations $\Phi_{\pm} = n_b(\omega_0) \pm n_e(E_A)$. Apart from the shifted ABS we find satellite resonances at $\varepsilon = s\bar{E}_A \pm \omega_0$ with spectral weights $\frac{1}{2}\lambda^2 \mathcal{A}_E^3 \Phi_{\pm s} / (2sE_A \pm \omega_0)^2$. It is only at resonance, $2E_A = \omega_0$, the satellite on either branch with spectral weight $\propto \Phi_{-}$ interferes with the main ABS of the same branch index. This is seen as a precursor of an avoided crossing in Fig. 2.

The Josephson current-phase relation is presented in Figs. 2(b) and 2(c) for the case that the electron and oscillator systems are in thermal equilibrium. Because of the different magnitudes of the thermal factors Φ_{\pm} ($\Phi_{+} \approx 2$ and $\Phi_{-} \approx 0$ for $T \lesssim E_A, \omega_0$), the current contribution of the satellites is dominated by the satellite with weight $\propto \Phi_{+}$. This satellite reduces the Josephson current by $\sim \lambda^2 \mathcal{A}_E^2 / (2E_A + \omega_0)^2$, which is of the order of a few percent of the full current for our parameter values. There is no dramatic signature in the current-phase relation of an emerging anticrossing at resonance because interference occurs between states carrying current in the same direction, and moreover, these states have spectral weights shared between them drawn from the original ABS, and the population of the states is largely phase-independent.

The Josephson current through the level modifies the oscillator spectrum giving it a phase dependence that is shown in Fig. 3(a) for the same parameters as for the ABS spectrum in Fig. 2. The polarization $\Pi^R(\omega)$ gives the

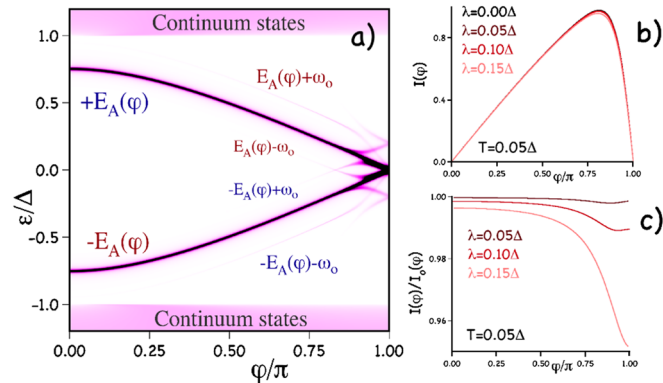


FIG. 2 (color online). (a) The spectrum of Andreev bound states as function of superconducting phase difference (φ) for electron-oscillator coupling $\lambda = 0.1\Delta$ and oscillator frequency $\omega_0 = 0.2\Delta$. The tunneling rate is $\Gamma = \Delta$ and the temperature is $T = 0.05\Delta$. The sidebands to each ABS are due to the dressing of the ABS by the electron-oscillator coupling. The sidebands of the ABS with branch index $s = \pm 1$ belong to the quasiparticle branch with index $s = \mp 1$. (b) The current-phase relation for different electron-oscillator couplings for $T = 0.05\Delta$. (c) The reduction of current due to coupling to the oscillator as a function of phase for the same temperature and electron-oscillator couplings as in (b).

possible collective excitations that are supported by the electronic system. Iterating the self-consistency equations once we get

$$\Pi^R(\omega) = 2\lambda^2 \frac{\mathcal{A}_E^2}{(\omega^R)^2 - 4E_A^2} n_e(E_A) \quad (8)$$

for the retarded phonon self-energy. As may be expected there is a mode with the phase dispersion $\omega_A = 2E_A(\varphi)$ originating from the transitions between the two ABS and the subsequent emission or absorption of the energy ω_A . A spectral weight $\sim 2\lambda^2 \mathcal{A}_E^2 [4\omega_0^2 / (4E_A^2 - \omega^2)] n_e(E_A)$ for this collective excitation is vanishingly small away from resonance. Near resonance the oscillator mode and the excitation interact strongly and an avoided crossing appears with a frequency split at $2E_A = \omega_0$

$$\omega_0(\varphi) = \omega_0 + \frac{\delta}{2} \pm \sqrt{\left(\frac{\delta}{2}\right)^2 + 2\lambda^2 \mathcal{A}_E^2 n_e(E_A)}, \quad (9)$$

where $\delta = 2E_A - \omega_0$ is the detuning. The analytic estimate of the split, $2\lambda \mathcal{A}_E \sqrt{2n_e(E_A)} \approx 0.096$, for a nearly phase-independent $\mathcal{A}_E \sim 0.34$ when $\Gamma = \Delta$, is in good agreement with the self-consistently determined split, $\approx 0.1\Delta$, extracted from the inset in Fig. 3(a) at $\varphi = 0.9\pi$. It is important to note that this split is significantly larger than the intrinsic broadening of the spectral features ($\sim \kappa$) and signals a strong-coupling regime in the sense of cavity QED, i.e., $\lambda \mathcal{A}_E \gg \kappa$.

Away from resonance, the electron-oscillator coupling gives a negative phase-dependent Stark shift of the oscillator base frequency. We plot this shift in Figs. 3(b) and 3(c) both as a function of electron-oscillator coupling strength λ for $T = 0.05\Delta$ and as a function of temperature

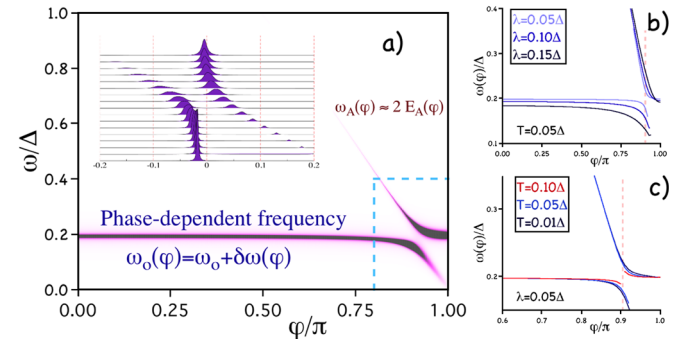


FIG. 3 (color online). (a) The spectra of the oscillator plotted as function of superconducting phase difference with the same parameters as in Fig. 2. A mode emerges at $\omega_A(\varphi) = 2E_A(\varphi)$ and develops to a polariton when in resonance with the oscillator (inset, $0.8\pi \leq \varphi \leq \pi$ in steps of 0.0125π). The dashed box indicates where the spectra in the inset are taken. Away from resonance there is a Stark shift $\delta\omega(\varphi)$ of the base frequency. (b) The Stark shift is shown at different electron-oscillator coupling strengths for $T = 0.05\Delta$ and (c) at different temperatures for an electron-oscillator coupling strength $\lambda = 0.05\Delta$. In both cases $\omega_0 = 0.2\Delta$. The dashed line in (b) and (c) indicates the resonant phase.

for $\lambda = 0.05\Delta$. Our analytic estimate in Eq. (9) gives in the limit of large detuning

$$\omega_0(\varphi) = \omega_0 + \delta\omega(\varphi) = \omega_0 - 2\frac{\lambda^2 \mathcal{A}_E^2 n_e(E_A)}{2E_A - \omega_0}. \quad (10)$$

This analytical expression for $\delta\omega(\varphi)$ is a good approximation when the continuum contributions can be neglected, i.e., for $\Gamma/\Delta \ll 1$, while in general one must use numerics to extract $\delta\omega(\varphi)$. Dispersive measurement of the acquired phase-dependent resonance frequency of the oscillator, $\omega_0(\varphi)$, gives a possibility to detect the position of the polariton resonance and hence define the energy of the ABS. Furthermore, by sweeping the base frequency of the oscillator the phase dispersion of the ABS energy can be detected.

Assume that we bridge two aluminum superconductors by a gated SWCNT, as shown in the inset of Fig. 1. The gate is part of an LC circuit which leads to an oscillation of the gate voltage with frequency $\omega_0 = 1/\sqrt{LC}$. A practical value for the resonator frequency is ~ 10 GHz, for which the polariton resonance is located well inside the superconducting gap (for aluminum $\Delta_{Al} \approx 50$ GHz), and at the same time the ABS energy splitting is large compared to the temperature below 100 mK. In our calculations we neglect electron-electron interactions. This can be done if Γ is large compared to Δ , which can experimentally be realized by tuning Γ by a back gate [12] to approach the weakly interacting Fabry-Perot regime. The main modification of the one-iteration approximation in the case $\Gamma \gg \Delta$ is that \mathcal{A}_E becomes strongly phase dependent, tending to $(\Delta/2\Gamma)\sin(\varphi/2)$ as Γ/Δ grows. This gives the effective coupling $\lambda\mathcal{A}_E \sim \lambda\Delta/\Gamma$ close to the resonance. This coupling must be large compared to the intrinsic oscillator damping, $\lambda\Delta/\Gamma \gg \kappa = \omega_0/Q$, in order to resolve $\delta\omega(\varphi)$. The strength of the bare coupling λ is determined by the capacitive interaction between the gate and the dot, and it is proportional to the ratio of corresponding capacitances, C_g/C_Σ , and can be expressed through the oscillator frequency as $\lambda = (C_g/C_\Sigma)(E_C/8E_L)^{1/4}\omega_0$ (E_C and E_L are charging and inductive energies of the oscillator, respectively). For SWCNT superconducting contacts the gate capacitance can be comparable with the capacitances of the contacts to the leads, having values of tens of aF [34]. Thus the coupling λ can be on the order of 10% of the oscillator frequency as assumed in our calculation. When the gate of the contact is connected to a superconducting cavity, the quality factor of the oscillator can be of the order of 1000–10000 [17–20]. Given our calculated value for effective ABS-oscillator coupling, $\lambda\mathcal{A}_E$, we get the ratio $\lambda\mathcal{A}_E/\kappa \approx 100$ –1000, indicating that the strong-coupling regime is indeed feasible, the resolution of the proposed spectroscopy should be very favorable to encourage experiments.

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