Phase-dependent Andreev spectrum in a diffusive SNS junction: Static and dynamic current response

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A long phase coherent normal (N) wire between superconductors (S) is characterized by a dense phase-dependent Andreev spectrum. We investigate the current response of Andreev states of an NS ring to a time-dependent Aharonov Bohm flux superimposed to a dc one. The ring is modeled with a tight-binding Hamiltonian including a superconducting region with a BCS coupling between electron and hole states, in contact with a normal region with on-site disorder. Both dc and ac currents are determined from the computed eigenstates and energies using a Kubo formula approach. Beside the well-known Josephson current, we identify different contributions to the ac response: a low-frequency one related to the dynamics of the thermal occupations of the Andreev states and a higher-frequency one related to microwave induced transitions between levels. Both are characterized by phase dependencies with a high-harmonic content, opposite to one another. Our findings are successfully compared to the results of recent experiments.

I. INTRODUCTION

Most properties of a nonsuperconducting N metal connected to two superconductors (an SNS junction) can be seen as resulting from the phase-dependent Andreev states (AS) in the N metal. These eigenstates are described by coherent combinations of electron and hole wave functions, determined by boundary conditions imposed by the superconducting contacts.[1] Whereas most equilibrium properties of SNS junctions are well understood theoretically and experimentally,[2–5] their high-frequency dynamics is a more complex issue, which has only been addressed very recently via the investigation[6,7] of NS rings submitted to a dc Aharonov Bohm flux Φ_{dc} with a small ac modulation $\delta\Phi_\omega \exp(-i\omega t)$. The quantity measured is the ac current response $\delta I\omega$ superimposed to the dc Josephson current. Within linear response, δI_ω is related to $\delta\Phi_\omega$ by the complex susceptibility $\chi(\omega) = \delta I\omega/\delta\Phi_\omega = i\omega Y$, where Y is the impedance of the NS ring. Our work is motivated by these recent experiments,[6,7] which revealed the dc flux, frequency, and temperature dependencies of the response function $\chi(\omega)$ and related them to the various relevant energy scales: the Thouless energy E_{Th} (inverse diffusion time through the N wire) and the relaxation rate of the population of the Andreev levels. On the theoretical side, the linear response of SNS junctions has been investigated using time-dependent Keldysh-Usadel equations.[8] Whereas a good agreement is found with experimental results in the frequency range dominated by relaxation processes of the population of the Andreev levels, the theoretical results obtained at higher frequency, i.e., in the regime where the dynamics is dominated by quasiresonant absorption of photons do not agree with experimental findings. In order to elucidate this disagreement, we have performed a Kubo analysis of the linear current response of an NS ring to an ac flux, calculated from the Andreev eigenstates and energies. The diffusive NS ring is described with a tight-binding Bogoliubov-de Gennes Hamiltonian. As detailed in Sec. II, this Hamiltonian describes a ring containing a superconducting region with a BCS coupling between electron and hole states, in contact with a normal region with on site (Anderson type) disorder and a

vector potential imposing the phase(φ)-dependent boundary condition. The eigenstate spectrum is obtained by numerical diagonalization. For a long diffusive N metallic wire (of length L greater than the superconducting coherence length ξ_s), we find that as expected the spectrum exhibits a phase-dependent gap $2E_g(\varphi)$.[2,9] This so-called minigap, much smaller than the superconducting gap Δ, is fully modulated by the phase difference of the superconducting order parameter φ across the N region. $E_g(\varphi)$ is maximal at $\varphi = 0$ with $E_g(0) \simeq 3.1 E_{Th}$ and goes linearly to zero at $\varphi = \pi$, approximatively like $E_g(\varphi) \simeq E_g(0)|\cos(\varphi/2)|$.[9,10] The phase-dependent Josephson current $I_J(\varphi)$ at equilibrium is calculated by summing the contributions of each AS of energy ϵ_n, via $i_n = -\frac{2e}{\hbar}\frac{\partial\epsilon_n}{\partial\varphi}$, the current carried by level n of thermal occupation factor $f_n(\varphi) = f(\epsilon_n(\varphi))$, where $f(\epsilon)$ is the Fermi-Dirac distribution function:

$$I_J(\varphi) = \sum_n f_n(\varphi)i_n(\varphi). \tag{1}$$

In Sec. III, we show how to compute from the Andreev levels and eigenstates, the ac linear response of the NS ring to an ac flux, using a Kubo formula similarly to what was previously done in normal Aharonov Bohm rings.[11–13] One can identify two main mechanisms responsible for the frequency dependence of the in-phase susceptibility and correlatively the existence of an out-of-phase dissipative response.

The first mechanism, discussed in Sec. IV, is the relaxation of the thermal populations of the Andreev levels with a time scale τ_{in}, the inelastic scattering time. It leads to a response χ_D that can be expressed with the diagonal matrix elements of the current operator. This mechanism is at the origin of a drastic increase of the harmonic content of the nondissipative response, in contrast with the zero-frequency susceptibility $\chi(0) = \chi_J = \partial I_J/\partial\Phi$, which is a pure cosine in the same regime of temperature. The dissipative response χ''_D is nearly π periodic with extra cusps at π that reflect the closing of the minigap.

The second mechanism, discussed in Sec. V, dominates at frequencies $\omega\tau_{in} \gg 1$. It corresponds to quasiresonant

transitions above the minigap within frequency scales of the order of $E_g(\varphi)/\hbar$. In the limit where $\hbar\omega \gg E_g \gg k_B T$, this phase-dependent dissipative response is simply proportional to the opposite of the minigap. In the other limit $k_B T \gg E_g \gg \hbar\omega$, this dissipative response is mainly determined by the flux dependence of the nondiagonal matrix elements of the current operator and is reversed in sign compared to the diagonal ones determining the low-frequency phase-dependent dissipation χ_D''. In Conclusion, we compare our results to recent experiments[6,7] and theoretical results based on frequency-dependent Usadel equations.[8]

II. TIGHT-BINDING HAMILTONIAN FOR A DIFFUSIVE SNS RING

We implement the Bogoliubov-de Gennes Hamiltonian described by the four blocks matrix:

$$\mathcal{H} = \begin{pmatrix} H - E_F & \boldsymbol{\Delta} \\ \boldsymbol{\Delta} & E_F - H^* \end{pmatrix}, \qquad (2)$$

where H and $-H^*$ are $N \times N$ matrices that describe respectively the electron and holelike wave function components of a hybrid NS ring within a tight-binding 2D model with on-site disorder:

$$H = \sum_{i=1}^{N} \epsilon_i |i\rangle\langle i| + \sum_{i \neq j} t_{ij} |i\rangle\langle j|. \qquad (3)$$

The ring has $N = N^N + N^S = N_x \times N_y$ sites on a square lattice of period a, with a normal portion of $N^N = N_x^N \times N_y$ sites in contact with a superconducting one ($N^S = N_x^S \times N_y$ sites). The on-site random energies ϵ_i of zero average and variance W^2 describe the disorder in the ring. The hopping matrix element between nearest neighbors reads $t_{ij} = t \exp i\varphi_{ij}$, where the phase factor is related to the superconducting phase difference through the normal junction via $\varphi_{ij} = (\pi/2\Phi_0) \int \vec{A}\,d\vec{l} = \varphi(x_i - x_j)/N_x^N$, which describes the effect of an Aharonov-Bohm flux $\Phi = AN_x^N a = \Phi_0\varphi/2\pi$, and $\Phi_0 = h/2e$ is the superconducting flux quantum. For sites in the S part, $\varphi_{ij} = 0$. The BCS diagonal matrix $\boldsymbol{\Delta}$ couples electron and hole states exclusively in the S part $\Delta_{i,i} = \Delta$ for $N_N + 1 \leqslant i \leqslant N$ and is zero otherwise. We have chosen the amplitude of the superconducting gap $\Delta = t/4$ such that the S coherence length $\xi_s = at/\Delta \ll N_x^S$ in order to avoid any reduction of the superconducting correlations in the S region (inverse proximity effect). The number of transverse channels and the amplitude of the disorder correspond to the diffusive regime where the length $N_x a$ of the normal region is longer than the elastic mean-free path l_e and shorter than the localization length $N_y l_e$. The length l_e is related to the amplitude of disorder by $l_e \simeq a15(t/W)^2$ at 2D.[14] We checked that the results do not depend on the position of the Fermi energy, typically chosen at filling 1/4. Hereafter, all energies are taken relatively to E_F.

A. Minigap and dc Josephson current

Typical flux-dependent spectra obtained upon diagonalization of the Hamiltonian $\mathcal{H}$ (2) are shown in Fig. 1. At energy well below the superconducting gap, energy levels exhibit a

mean level spacing $\delta_N = E_F/N^N$ characteristic of the normal part and a $\Phi_0 = h/2e$ periodicity. These constitute the Andreev spectrum. A denser spectrum is observed above the gap with the periodicity h/e as expected for a normal ring, see Figs. 1(a) and 1(b). By construction, the spectrum is perfectly symmetric with respect to the Fermi energy. We observe disorder-dependent fluctuations [see Fig. 1(c)] of the position of the energy levels in the spectrum. At low energy, the amplitude of these fluctuations is of the order of the mean level spacing δ_N in the N part of the ring in which Andreev levels are confined. The flux-dependent minigap closes linearly at $\pm\pi$ in the limit of a very dense spectrum and can be well described as expected by $E_g(\varphi) = E_g(0)|\cos(\varphi/2)|^9$ [see Fig. 1(d)]. In short junctions, this closing of the gap at $\varphi = \pi$ is directly related to the existence of conductance channels of transmission one in a large diffusive system.[15,16] In long junctions, the same qualitative behavior is observed even though Andreev levels and eigenvalues of the transmission matrix are not simply related and that the amplitude of the minigap is much smaller than the superconducting gap. As shown in Fig. 2, the flux dependence of the Josephson current $I_J(\varphi)$ and its flux derivative at low temperature are sensitive to the anharmonicity of the flux dependence of low-energy levels and exhibit a slight skewness. $I_J(\varphi)$ becomes sinusoidal at temperatures larger than the Thouless energy (of the order of 0.03Δ in the simulations) according to Ref. 2. We will see in the following that the ac current response is much more sensitive than the Josephson current to the strong anharmonicity of the flux-dependent minigap, and exhibits strong anomalies at π that survive at temperatures larger than the Thouless energy.

III. FINITE FREQUENCY LINEAR RESPONSE

We investigate the linear dynamics of the NS ring excited by an oscillating flux $\delta\Phi(t) = \delta\Phi \exp(-i\omega t)$ leading to the time-dependent Hamiltonian $H(t) = H_0 - \mathbf{J}\delta\Phi(t)$, where $\mathbf{J}$ is the current operator. Inspired by previous work on the dynamics of persistent currents in normal mesoscopic Aharonov Bohm rings,[12,13] we use as a starting point the master equation describing the relaxation of the density matrix towards equilibrium:

$$\partial\rho(t)/\partial(t) = (1/i\hbar)[H(t),\rho] - \Gamma[\rho(t) - \rho_{eq}(t)], \qquad (4)$$

where the equilibrium density matrix $\rho_{eq}(t) = \exp -H(t)/k_B T$ and the phenomenological relaxation tensor Γ describes the coupling of the system to a thermal reservoir. The diagonal elements $\gamma_{nn} = \gamma_D = \hbar/\tau_{in}$ describe the relaxation of the populations f_n of the Andreev states due to inelastic scattering such as electron-phonon or electron-electron collisions. Nondiagonal elements γ_{nm} describe the relaxation of the coherences $\rho_{nm}(t)$ due to interlevel transitions. We will mostly consider the limit where ω and $k_B T \gg \gamma_{nm} \gg \delta_N$, for which the response function is independent of the values of γ_{nm}. Following[12,13] the linear current response $\delta I(t) = \text{Tr}(J\delta\rho(t)) + \text{Tr}(\delta J(t)\rho_0)$ is expressed via the complex susceptibility $\chi(\omega) = \delta I(t)/\delta\Phi(t)$ ($\rho_0 = \sum_n f_n(\Phi_{dc})|n\rangle\langle n|$ is the unperturbed matrix density)
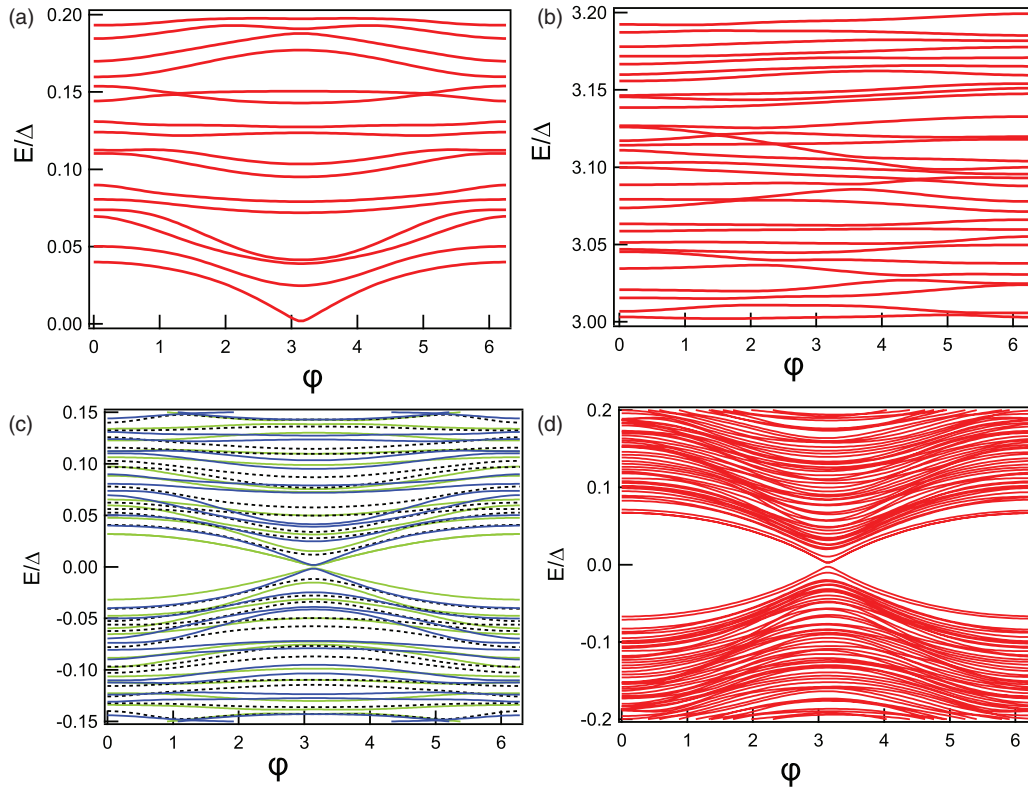
FIG. 1. (Color online) Phase-dependent spectrum of Andreev levels: (a) close to the Fermi energy and (b) above the superconducting gap for a diffusive ring with $N_x^N = 60 \times 24$ normal sites and on-site disorder of amplitude $W/t = 1.5$. (The number of S sites with $\Delta = t/4$ is $N^S = 50 \times 24$.) Note the denser spectrum above the minigap and the different periodicity. (c) Same as (a), for three different disorder configurations (different colors). Note the symmetry of the spectrum with respect to the Fermi level at zero energy as well as the opening of the phase-dependent minigap, whose amplitude scales with the Thouless energy, $E_{\text{Th}} = \delta_N N_y l_e/N^N a$, where δ_N is the energy level spacing in the normal region. (d) Diffusive ring with a larger number of transverse channels $N^N = N_x^N \times N_y = 20 \times 100$ normal sites for a single disorder configuration of amplitude $W/t = 1.5$. (The number of S sites is $N^S = 20 \times 100$.)

leading to

$$\chi(\omega) = -N\frac{e^2}{2mL^2} - \sum_n \frac{\partial f_n}{\partial \epsilon_n}|J_{nn}|^2 \frac{\gamma_D}{\gamma_D - i\omega}$$
$$- \sum_{n,m\neq n}|J_{nm}|^2 \frac{f_n - f_m}{\epsilon_n - \epsilon_m} \frac{i(\epsilon_n - \epsilon_m) + \hbar\gamma_{nm}}{i(\epsilon_n - \epsilon_m) - i\hbar\omega + \hbar\gamma_{nm}}.$$
$$(5)$$

J_{nm} is the matrix element of the current operator between the eigenstates n and m of the unperturbed Hamiltonian H_0 and $J_{nn} = i_n$. Using the sum rule derived from the second-order perturbation of H with respect to the perturbation $J\delta\varphi$:[12,17]

$$\sum_{m\neq n} \frac{|J_{nm}|^2}{(\epsilon_n - \epsilon_m)} = -\frac{1}{2}\frac{\partial i_n}{\partial \Phi} - \frac{e^2}{2mL^2}.$$
$$(6)$$

$\chi(\omega)$ can be expressed as

$$\chi(\omega) = \frac{\partial I_J}{\partial \Phi} - \sum_n i_n^2 \frac{\partial f_n}{\partial \epsilon_n}\frac{i\omega}{\gamma_D - i\omega} - \sum_{n,m\neq n}|J_{nm}|^2$$
$$\times \frac{f_n - f_m}{\epsilon_n - \epsilon_m}\frac{i\hbar\omega}{i(\epsilon_n - \epsilon_m) - i\hbar\omega + \hbar\gamma_{nm}}.$$
$$(7)$$

This second expression clearly yields the zero-frequency limit of the susceptibility $\chi(0) = \partial I_J/\partial \Phi$. This expression also emphasizes the two relaxation processes that cause frequency-dependent effects as discussed in the next sections.

IV. DIAGONAL SUSCEPTIBILITY AND RELAXATION OF ANDREEV LEVELS POPULATIONS

We discuss in the following the second term of expression (7) that we call χ_D and is the finite frequency nonadiabatic contribution due to the thermal relaxation of the populations f_n of the Andreev levels with the characteristic inelastic time τ_{in}.[20] As pointed out in Ref. 12, this term contains exclusively diagonal elements of the current operator and is, like χ_J, nonzero only in the Aharonov Bohm ring geometry. It is associated to the existence of a finite persistent current in a phase coherent ring at equilibrium. It is proportional to the sum over an energy range $k_B T$ around the Fermi energy of the square of the single-level current i_n^2. We recast χ_D into a product of a frequency-dependent term and a phase-dependent one:

$$\chi_D(\omega) = \frac{i\omega\tau_{\text{in}}}{1 - i\omega\tau_{\text{in}}}F(\varphi,T),$$
$$(8)$$

where $F(\varphi,T) = -\Sigma_n(i_n^2\frac{\partial f_n}{\partial \epsilon_n})$. We have numerically evaluated this function deriving i_n from the phase derivative of each eigenenergy pictured in Fig. 1. $F(\varphi)$ is shown for different
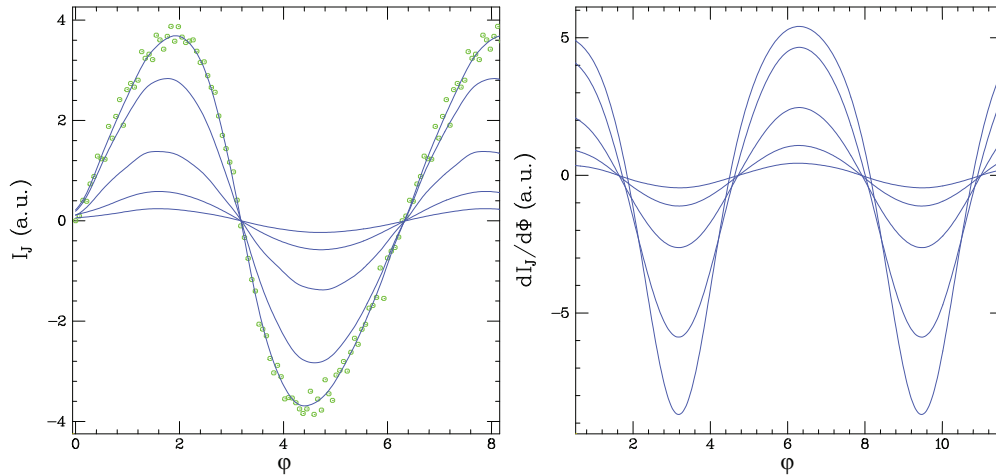
FIG. 2. (Color online) Phase-dependent Josephson current and susceptibility calculated from the Andreev spectrum shown in Fig. 1(left). The temperatures correspond to 0.01, 0.02, 0.04, 0.06, and 0.08 in the units of the superconducting gap Δ. The amplitude of the minigap is estimated to be 0.04 Δ. The anharmonicity is best revealed on the derivative $dI_J/d\phi$.

temperatures in Fig. 3. As expected, $F(\varphi)$ has a strong second-harmonic component and exhibits sharp anomalies in the vicinity of odd multiples of π for which the minigap closes. In the continuous spectrum limit and for $k_B T \gg E_{\text{Th}}$, $F(\varphi,T)$ can be written in terms of the spectral current $J(\epsilon)$ and the density $n(\epsilon)$ of Andreev levels as $F(\varphi,T) = \int J^2(\varphi,\epsilon)/[k_B T n(\epsilon)]d\epsilon$. This function, initially introduced by Lempitsky[21] to describe nonequilibrium effects in voltage biased SNS junctions, can be approximated by the analytical expression derived from Usadel equations in the limit where $k_B T \gg E_g(0)$:[6,8]

$$F_U(\varphi,T) \propto \left(\frac{1}{k_B T}\right)\left\{[-\pi + (\pi + \varphi)(2\pi)]\sin(\varphi) - \frac{|\sin(\varphi)|}{\pi}\sin^2(\varphi/2)\right\}. \quad (9)$$

As shown on Fig. 3, this analytical form describes well the phase dependence of the numerical results at temperatures

larger than $E_g(0)$. We find, however, that the $1/T$ decrease at large temperature predicted in Eq. (9) is only qualitatively obeyed for numerical results. As pointed out in the context of atomic point contacts,[18] the dissipative component of χ_D is related via the fluctuation dissipation theorem to the existence of a nonintuitive supercurrent low-frequency thermal noise.[18] This low-frequency noise due to the closing of the minigap at π does not exist in ordinary tunnel Josephson junctions.[19] One can associate to this dissipative response an effective phase-dependent conductance $\delta G_{\text{eff}}(\varphi) = \chi_D''(\varphi)/\omega$. The amplitude of $\delta G_{\text{eff}}(\varphi)$ at frequencies smaller than γ_D and temperatures of the order or larger than E_g is of the order $G_N E_g^2/(k_B T \hbar \gamma_D)$ and can be much larger than G_N, the normal state conductance. This component $\chi_D(\omega,\varphi)$ was recently experimentally measured on a mesoscopic NS ring[7] with a very good quantitative agreement with expressions (8) and (9).
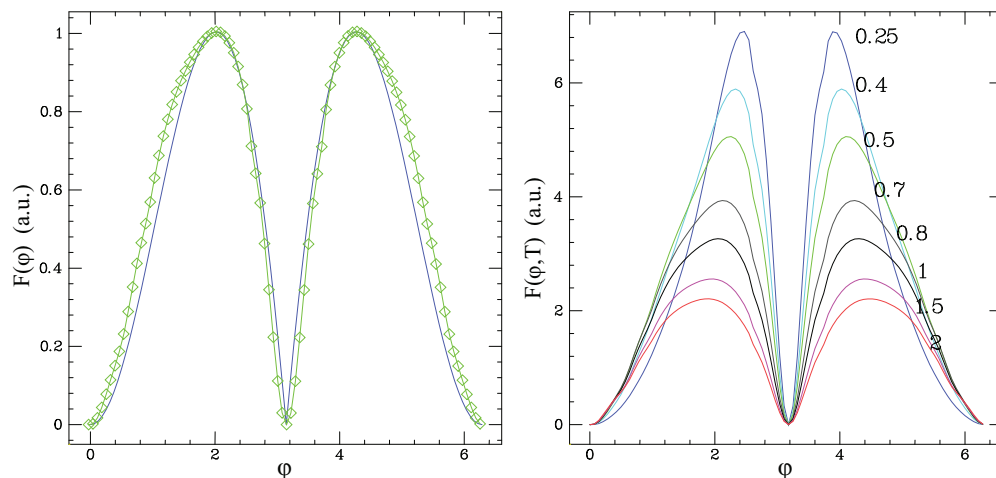

FIG. 3. (Color online) (Right) Phase dependence of the function F computed for different temperatures, increasing from the top to the bottom curves, in units of the minigap: $2E_g(0)$. (Left) Comparison of the numerical results (diamonds) with the analytical expression (9) (continuous line) at a temperature equal to the minigap 0.08Δ.

V. NONDIAGONAL SUSCEPTIBILITY AND MICROWAVE INDUCED TRANSITIONS IN THE ANDREEV SPECTRUM

A. Analytical considerations

We now consider the contributions of nondiagonal elements of the current operator, which describe the physics of microwave induced transitions within the Andreev spectrum,

$$\chi_{\mathrm{ND}} = \sum_{n,m\neq n} |J_{nm}|^2 \frac{f_n - f_m}{\epsilon_n - \epsilon_m} \frac{i\hbar\omega}{i(\epsilon_n - \epsilon_m) - i\hbar\omega + \hbar\gamma_{\mathrm{ND}}}, \quad (10)$$

where we have assumed that all γ_{nm} are identical given by a single γ_{ND}.

In the continuous spectrum limit, the average level spacing δ_N is much smaller than the energy scales γ_{ND}, $k_B T$, and $\hbar\omega$, so that one can write

$$\chi_{\mathrm{ND}} = -\int_{-E_M}^{E_M} |J_{\epsilon,\epsilon'}|^2 \frac{f(\epsilon) - f(\epsilon')}{\epsilon - \epsilon'} \frac{i\hbar\omega}{i(\epsilon - \epsilon') - i\hbar\omega + \gamma_{\mathrm{ND}}}$$
$$\times n(\epsilon)n(\epsilon')d\epsilon d\epsilon', \quad (11)$$

where E_M is a high-energy cutoff of the order of the bandwidth, from now on arbitrarily taken as unity, and $n(\epsilon)$ is the density of states at energy ϵ. In the limit where the induced minigap is very small compared to the superconducting gap Δ (long junction), one can approximate the density of states as a step function at $E_g(\varphi)$: $n(\epsilon,\varphi) = n_0[\theta(\epsilon - E_g(\varphi)) + \theta(-\epsilon - E_g(\varphi))]$, [with $\theta(x)$, the Heaviside function]. Below, we also assume that $|J_{\epsilon,\epsilon'}|^2$ can be approximated by a constant J^2. We will see that this approximation is valid when $k_B T \ll E_g < \hbar\omega$ where the dominant contribution comes from matrix elements nearly independent of φ. This leads to

$$\chi_{\mathrm{ND}} = -n_0^2 \iint_{|\epsilon|,|\epsilon'|\geqslant E_g(\varphi)} d\epsilon d\epsilon'$$
$$\times \left[|J|^2 \frac{f(\epsilon) - f(\epsilon')}{\epsilon - \epsilon'} \frac{i\hbar\omega}{i(\epsilon - \epsilon') - i\hbar\omega + \gamma_{\mathrm{ND}}} \right]. \quad (12)$$

We define $\delta\chi'_{\mathrm{ND}} = \chi'_{\mathrm{ND}}(\pi) - \chi'_{\mathrm{ND}}(0)$ and $\delta\chi''_{\mathrm{ND}} = \chi''_{\mathrm{ND}}(\pi) - \chi''_{\mathrm{ND}}(0)$ as the amplitudes of the flux-dependent components of the real and imaginary parts of $\chi_{\mathrm{ND}}(\Phi,\omega)$. The frequency dependence of these quantities is depicted in Fig. 4

for several values of the minigap larger than the temperature. We find that $\delta\chi'_{\mathrm{ND}}$ is negative and decreases slowly at low frequency with an inflexion point at $\omega = E_g(0)/\hbar$; $\delta\chi''_{\mathrm{ND}}$ is positive and increases linearly with frequency up to $\omega = E_g(0)/\hbar$ and is independent of frequency at larger values. These results, in agreement with Kramers-Kronig relations, show that the minigap is the fundamental frequency scale for $\chi_{\mathrm{ND}}(\varphi)$. In the limit where $\gamma_{\mathrm{ND}} \ll \omega$ and $\gamma_{\mathrm{ND}} \ll k_B T$, $\gamma_{\mathrm{ND}}/[(\epsilon - \epsilon' - \hbar\omega)^2 + \gamma_{\mathrm{ND}}^2)]$ entering in χ'' deduced from Eq. (12) can be approximated by the δ function: $\delta(\epsilon - \epsilon' - \hbar\omega)$. It is then possible to express simply $\chi''_{\mathrm{ND}}(\omega,\varphi)$ analytically as

$$\chi''_{\mathrm{ND}} = n_0^2 |J|^2 \int_{|\epsilon|\geqslant E_g(\varphi)} [f(\epsilon) - f(\epsilon + \hbar\omega)] d\epsilon. \quad (13)$$

Because the variation in φ is only contained in the integration limits, we find that in the frequency range where $\omega \gg k_B T$, $\chi''(\varphi)$ mimics the minigap (with a minus sign) in the flux domain where $\hbar\omega \geqslant 2E_g(\varphi)$ and reads $\chi''_{\mathrm{ND}}(\varphi,\omega) = G_N(\omega - 2E_g(\Phi)/\hbar)$. The normal state conductance $G_N = \chi''_{\mathrm{ND}}(\pi)/\omega$ (where the minigap closes) can be expressed as $G_N = |J|^2 n_0^2$. On the other hand, at low frequencies below $E_g(\varphi)$, $\chi''_{\mathrm{ND}}(\varphi)$ is equal to zero. As a result, when $\omega \ll E_g(0)$, the flux-dependent absorption exhibits sharp peaks at odd multiples of π whose amplitude scales linearly with ω as shown on Fig. 4. One finds that at low frequency, the ratio $\delta\chi''_{\mathrm{ND}}/\delta\chi_J = \delta\chi''/\delta\chi_J$ varies like $\hbar\omega/E_g$. There is, however, no simple analytical expression for the complete phase and frequency dependencies of $\chi'(\omega,\varphi)$ owing to the fact that according to Eq. (12), it explicitly depends logarithmically on the energy cutoff E_M.

In the opposite limit of high temperature $T \gg E_g \gg \hbar\omega$, we can easily find from Eq. (11) that the ratio $\delta\chi''_{\mathrm{ND}}/\delta\chi'_J(T)$ varies like $\hbar\omega/k_B T$, independently of the energy and phase dependence of $|J_{\epsilon,\epsilon'}|^2$. It is, however, not possible to use Eq. (13) to deduce the phase dependence of χ''_{ND}. This equation relies on a crude approximation neglecting the phase dependence of the nondiagonal matrix elements of the current operator. We will show in the next paragraph devoted to numerical calculations that this approximation is only reasonable at low temperature and large frequency where, in the expression of χ''_{ND}, only a small number of matrix elements
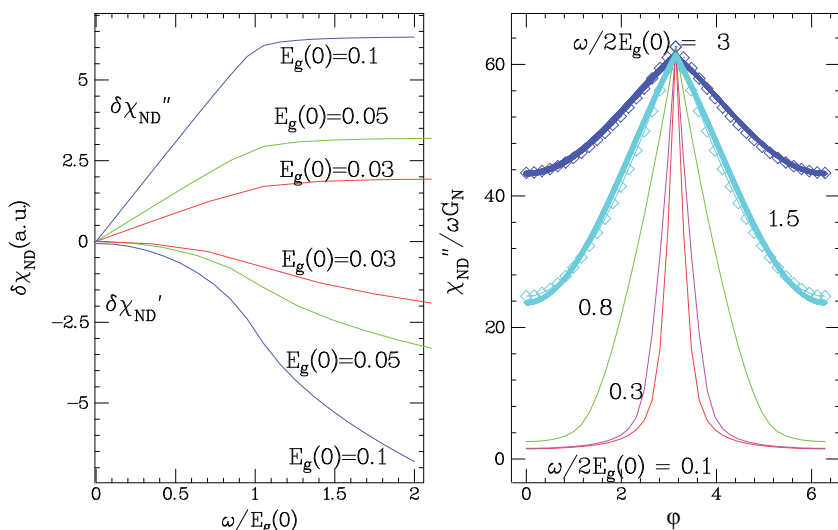

FIG. 4. (Color online) Nondiagonal susceptibility calculated assuming no phase dependence for the nondiagonal matrix elements of the current operator. The temperatures and frequencies investigated correspond to $T \ll \hbar\omega$. The values of γ_{ND} and $k_B T$ were both taken equal to 0.01, i.e., much smaller than the minigap $2E_g(0)$. (Left) Frequency dependence of $\delta\chi''_{\mathrm{ND}}$ and $\delta\chi'_{\mathrm{ND}}$ dissipative and nondissipative responses for different values of the minigap. (Right) Phase dependence of χ''_{ND} for different frequencies. The thick continuous lines correspond to a fit with a $-|\cos(\varphi/2)| \propto -E_g(\varphi)$ dependence.

contribute. These are matrix elements $|J_{\epsilon,\epsilon'}|^2$ coupling negative energy levels close to the minigap to positive energy levels much larger than E_g. These matrix elements have indeed only a very small phase dependence. On the other hand, at high temperature, $k_B T \gg E_g$, a large number of matrix elements $|J_{\epsilon,\epsilon'}|^2$ contribute to the integral in ϵ' in Eq. (11). We can then estimate their contribution to the phase dependence of χ''_{ND} using the fact that $\mathrm{Tr}(J^2) = \Sigma_n |J_{nn}|^2 + \Sigma_{n,m \neq n} |J_{nm}|^2$ does not depend on the Aharonov-Bohm phase just like $\mathrm{Tr}(\mathcal{H})$ (since the Aharonov Bohm phase only affects nondiagonal matrix elements of $\mathcal{H}$). The sum of all nondiagonal matrix elements $|J_{nm}(\varphi)|^2$ with $m \neq n$ is thus opposite in sign to the variation of $F(\varphi) \propto \sum_n |J_{nn}|^2$ at large T. Therefore, in the limit $T \gg \hbar\omega \simeq E_g(0)$, where the sum of a large number of nondiagonal matrix elements $|J_{nm}(\varphi)|^2$ with $m \neq n$ contributes to the phase dependence of χ''_{ND}, the phase dependencies of χ''_D and χ''_{ND} are thus expected to be reversed from one another. The results of the numerical simulations presented in the next section agree with this simple qualitative prediction.

B. Numerical results for the nondiagonal susceptibility

The nondiagonal matrix elements of the current operator $\vec{J} = (\hbar/i)\vec{\nabla} - q\vec{A}$ along the ring are calculated from the eigen-wave-functions according to

$$
\begin{aligned}
J_{nm} = \frac{\hbar}{i} \sum_j \Psi_n^{e*}(x_j, y_j) \big[\Psi_m^e(x_j + 1, y_j) \\
- \Psi_m^e(x_j, y_j) + eA(x_j) \big] + \Psi_n^{h*}(x_j, y_j) \\
\times \big[\Psi_m^h(x_j + 1, y_j) - \Psi_m^h(x_j, y_j) - eA(x_j) \big],
\end{aligned} \quad (14)
$$

where $\Psi_m^e(x_j, y_j)$ and $\Psi_m^h(x_j, y_j)$ correspond respectively to the electron and hole components of the wave function at point j of coordinates (x_j, y_j) in units of a. The phase dependence of the square modulus of these matrix elements is shown for various indices n and m in Figs. 5 and 6. The indices n and m are taken respectively positive above and negative below the
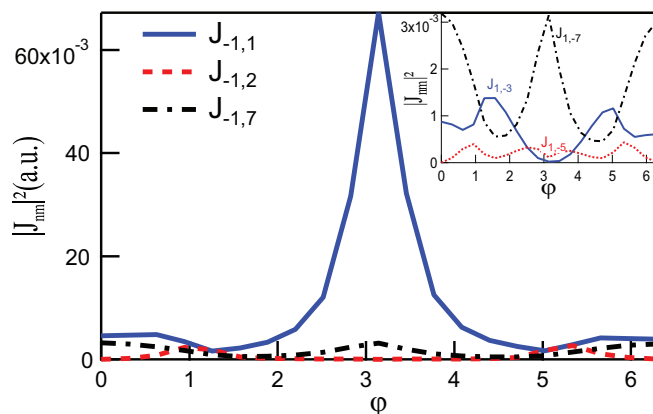

FIG. 5. (Color online) Phase dependence of the nondiagonal current matrix elements $|J_{-1,n}|^2$ coupling the highest level below the minigap to levels above the minigap. (Inset) Zoom on $|J_{-1,n}|^2$ with $n > 1$, which have a very small phase dependence compared to $|J_{-1,1}|^2$. They are obtained from the exact diagonalization of the spectrum of an NS ring whose normal region size is 90×30 and $W/t = 2$. The minigap amplitude is $2E_g(0) = 8\delta_N$.
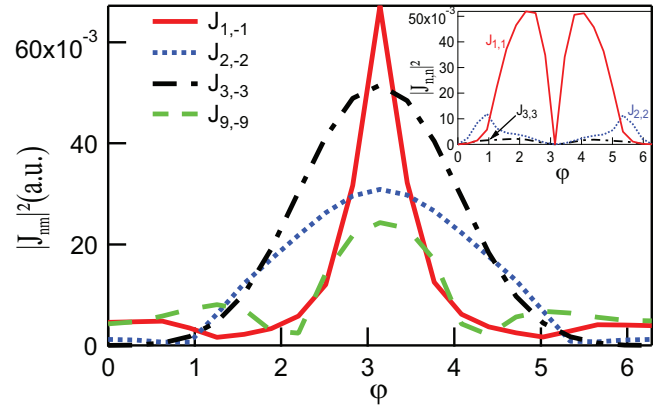

FIG. 6. (Color online) Phase dependencies of the electron hole symmetrical nondiagonal matrix elements compared to the diagonal ones shown in the inset. These matrix elements are obtained from the exact diagonalization of the spectrum of an NS ring whose normal region size is 90×30 and $W/t = 2$. The minigap amplitude is $2E_g(0) = 8\delta_N$.

minigap. Whereas $|J_{-11}(\varphi)|^2 = i_1^2(\varphi)$ exhibits a strong peak at $\varphi = \pi$, the amplitude of $|J_{-1n}(\varphi)|^2$ is much smaller at large n with a phase dependence that is smooth around π and a maximum around zero phase, see Fig. 5. On the other hand, matrix elements $|J_{-nn}(\varphi)|^2$ corresponding to states symmetric with respect to the minigap, i.e., electron hole symmetric states, keep a phase dependence peaked at π similar but reversed in sign compared to $|J_{1,1}|^2(\varphi)$. We can see in Fig. 6 that their amplitude decreases only slowly with n in contrast to the fast amplitude decrease of the diagonal matrix elements $J_{nn} = i_n$ shown in the inset. Whereas we can understand qualitatively that matrix elements between electron-hole symmetric states such a $|J_{-nn}|$ are much larger than matrix elements between nonsymmetrical states $|J_{-nm}|$ with $n \neq m$, it is difficult to explain why their decay with n is lower than the decay of the diagonal matrix elements $|J_{nn}|$. The difference between the phase dependence of $|J_{-nn}(\varphi)|^2$ compared to $|J_{-1n}(\varphi)|^2$ can qualitatively explain the evolution of the shape of $\chi_{ND}(\varphi)$ in the limit $\omega \gg k_B T$ compared to $\omega \ll k_B T$. In the first case, the main contribution stems from matrix elements $|J_{-1n}(\varphi)|^2$, where $n \gg 1$ with a very small phase dependence, whereas in the second case, a much larger number of matrix elements contribute to χ''_D, including the electron hole symmetrical ones $|J_{-nn}(\varphi)|^2$.

$\chi''_{ND}(\varphi)$ is computed from these matrix elements and the related energy spectrum following Eq. (10). We took $\gamma_{ND} = 3\delta_N$ in order to reproduce the continuous spectrum limit. The results concerning the imaginary component $\chi''_{ND}(\varphi)$ are shown in Figs. 7(a) and 7(b) for $\hbar\omega > k_B T$ and $\hbar\omega < k_B T$, respectively. In the first case, $\hbar\omega > k_B T$, we find good qualitative agreement with our analytical findings neglecting the flux dependence of the $|J_{nm}|^2$ in particular $\delta\chi''_{ND}(\varphi)$ is peaked at π and its amplitude increases linearly with frequency up to $\hbar\omega = 2E_g$, whereas in the second case, $\hbar\omega < k_B T$, we find that the shape of $\delta\chi''_{ND}(\varphi)$ is very similar to the opposite of the function $F(\varphi)$ (giving the phase dependence of the average square of the single level current) with a characteristic bump at $\varphi = 0$ [see Eq. (8)]. A similar behavior is found for $\delta\chi'_{ND}(\varphi)$.
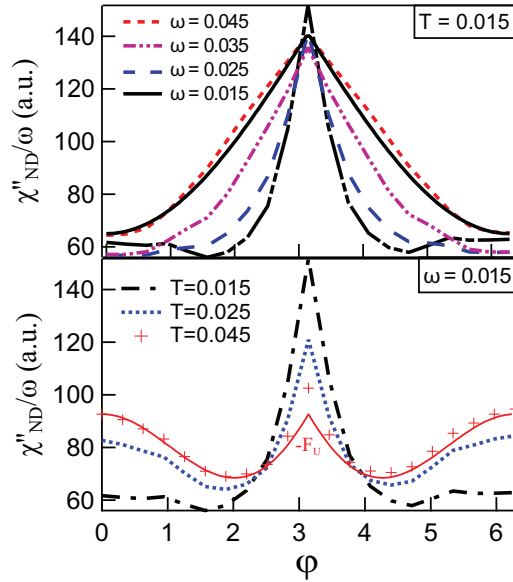
FIG. 7. (Color online) Evolution of the phase dependence of $G_{\mathrm{ND}} = \chi''_{\mathrm{ND}}/\omega$ obtained from the exact diagonalization of the spectrum of an NS ring (size 90×30) $W/t = 2$ using Eq. (10) $\gamma_{\mathrm{ND}} = 3\delta_N$. (Top) For $k_B T = 2\delta_N$ and different frequencies $\hbar\omega > k_B T$ below and of the order of the minigap, note the good agreement with the data obtained in Fig. 4, neglecting the phase dependence of the current matrix elements. (Bottom) $\hbar\omega = 2\delta_N$ and different temperatures $k_B T > \hbar\omega$ below and of the order of the minigap. For the largest temperature, the phase dependence observed is close to the opposite of the function $F_U(\varphi)$, continuous line.

VI. CONCLUSION

We have developed a simple model for the computation of the ac linear response of an NS diffusive ring to a high-frequency flux in the long junction limit. Starting from the dc phase-dependent Andreev spectrum and wave functions of the ring, we use a Kubo formula adapted for the Aharonov-Bohm geometry, which yields the complex susceptibility of the ring as a function of the energy levels and matrix elements of the current operator. We clearly identify two different finite frequency contributions superimposed to the dc response which is the flux derivative of the Josephson current. The first one, expressed in terms of the diagonal element of the current operator, can be understood as the Debye relaxation of the populations of the Andreev states. The second one, expressed in terms of the nondiagonal matrix elements of the current operator, describes interlevel transitions within the Andreev spectrum. It is striking that numerical simulations on small systems with less than ten levels in the energy scale corresponding
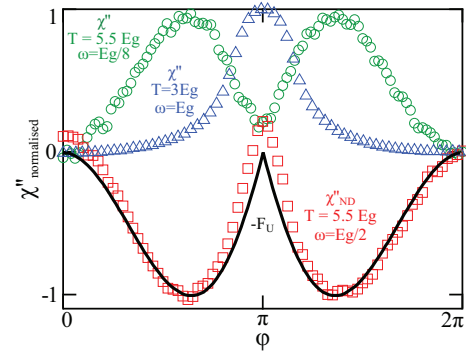

FIG. 8. (Color online) The phase-dependent dissipative response measured experimentally[7,22] is shown for three different regimes: circles: $\omega\tau_{\mathrm{in}} \simeq 1$, where χ''_D is the dominant contribution with a phase dependence following $F(\varphi)$. At frequencies and $k_B T$ of the order of $E_g(0)$, the dissipative response is dominated by χ''_{ND} (triangles) and we observe a phase dependence peaked at π, which resembles the minigap. In the limit where $k_B T > \omega \gtrsim E_g$, a phase dependence opposite to $F(\varphi)$ is found (squares) as expected for the contribution of the nondiagonal matrix elements of the current operator, in agreement with the results in Fig. 7. The continuous line is $-F_U(\varphi)$ calculated from expression (9).

to the minigap can reproduce the experiments[7,22] investigating the ac susceptibility of an NS ring where E_g/δ_N is of the order of 1000, as illustrated in Fig. 8. The phase-dependent dissipative response is shown for three different regimes: (i) $\omega\tau_{\mathrm{in}} \simeq 1$, where χ''_D is the dominant contribution with a phase dependence well described by $F(\varphi)$; (ii) at frequencies and $k_B T$ of the order of $E_g(0)$, we observe a phase dependence peaked at π, which resembles the minigap; (iii) finally, in the limit where $k_B T > \omega \gtrsim E_g$, a phase dependence opposite to $F_U(\varphi)$ is found, as expected for the contribution of the nondiagonal matrix elements of the current operator, in agreement with the numerical results in Fig. 7. On the other hand, whereas Usadel equations[8] provide an excellent agreement between the numerical and experimental findings for the diagonal contribution χ_D, the high-frequency regime yields different results. In particular, the predicted phase oscillations of the susceptibility do not reproduce our findings.

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