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Theory of Josephson Radiation. I. General Theory

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We consider the problem of a Josephson junction interacting with a quantized electromagnetic field contained in a microwave cavity. Inasmuch as the cavity has a quality factor Q, and an external current J flows in and out of the junction, this is a problem in nonequilibrium statistical mechanics. We use a technique that eliminates adiabatically the radiation field directly from the total-density-matrix equation of motion. The resulting equation for the reduced density matrix is found to have a simple physical interpretation. The spectrum of the voltage fluctuations and the linewidth of the emitted radiation are then calculated for the case when the resonance cavity is at absolute zero.

I. INTRODUCTION

It is well known[1,2] that a superconducting tunnel junction can support a dc current up to a certain critical value without developing any voltage across the junction. However, when an external current in excess of this critical value is forced through the junction, charges pile up on one side of the barrier and a voltage $V_0 = q/C$ is devolped, where q is the excess charge on one side of the barrier and C is the capacitance of the junction. In the presence of the voltage, it was predicted that an ac current will oscillate in the junction and radiation will be emitted.

In previous work[3,4] we considered the interaction of such a tunnel junction with a radiation field contained in a resonance cavity possessing a quality factor Q. We concluded that there is a slight pulling of the frequency of the emitted radiation from $\omega = 2eV_0/\hbar$. In obtaining this result we formulated the problem in terms of the density matrix. Furthermore we factorized the density matrix into two separate parts corresponding to the superconductor and the radiation field at all times and considered only the mean equations of motion. Hence the treatment was semiclassical. In the present work we consider the time evolution of the total density matrix of the combined radiation and superconductor system and relax the factorization assumption. The linewidth of the emitted radiation is calculated and is found to arise from fluctuations in the voltage across the junction, which in turn give rise to a frequency modulation of the radiation. The voltage fluctuations are calculated by deriving an equation of motion for the reduced density matrix involving the superconductor coordinates alone. This is achieved by noting that the characteristic time of the radiation field $(\nu/Q)^{-1} \approx 10^{-8}$ sec is the fastest decay time in the problem. Consequently the radiation field can be eliminated adiabatically. The adiabatic elimination is performed directly using a density-matrix technique for the case of zero cavity temperature (Sec. III). The case of finite cavity temperature requires more involved techniques and will be treated in a later paper. An equation for the reduced density matrix is obtained which is interpreted physically as a flow of probability. The physical origin of the voltage fluctuations is then clearly demonstrated to be shot noise associated with the tunneling of electron pairs. The resulting linewidth agrees with that obtained by Stephen[5] using Langevin equations with noise operators. However, in Ref. 5, operators are treated as c numbers, and it is felt that the present quantum-mechanical treatment is more consistent.

In Sec. II we present a description of the tunnel junction and its interaction with a quantized radiation field. Insofar as this is a nonequilibrium problem we set up the equation of motion for the total density matrix including the effects of a finite quality factor Q of the cavity and the external current forced through the junction. In Sec. III we present a calculation of the linewidth of the radiation for the case of zero cavity temperature.

II. FORMULATION

A. Discussion of Tunneling Junction

In this section we consider a tunneling junction consisting of an oxide layer sandwiched between two superconductors. We shall investigate the properties of the superconductors responsible for a tunneling current and establish our notation. Let us define $c_{\vec{k}\sigma}$ to be the annihilation operator of an electron with momentum k and spin σ in the left-hand superconductor, and a similar $c_{\vec{q}\sigma}$ for an electron on the right. Tunneling across the junction is then described by the Hamiltonian[6]

$$H_1 = \sum_{\vec{k},\vec{q},\sigma} T_{\vec{k},\vec{q}}(c^\dagger_{\vec{k}\sigma}c_{\vec{q}\sigma} + c^\dagger_{\vec{q}\sigma}c_{\vec{k}\sigma}) \ . \tag{2.1}$$

In this paper we shall restrict our attention to tunneling of Cooper pairs. It is convenient to use the pseudospin notation of Anderson[7] and define

$$\sigma_{\vec{k}} = c^\dagger_{\vec{k}}c^\dagger_{-\vec{k}} \ , \quad \sigma_{z,\vec{k}} = \tfrac{1}{2}(1 - c^\dagger_{\vec{k}}c_{\vec{k}} - c^\dagger_{-\vec{k}}c_{-\vec{k}}) \ .$$

A similar set $\sigma_{\vec{q}}$, $\sigma_{z,\vec{q}}$ is defined for the superconductor on the right. Here $-\vec{k}$ indicates a momentum of $-\vec{k}$ and spin down in the usual manner. Following Wallace and Stavn,[8] we make a canonical transformation on Eq. (2.1) and obtain an equivalent tunneling Hamiltonian in terms of $\sigma_{\vec{k}}$, $\sigma_{\vec{q}}$:

$$H_T = \sum_{\vec{k},\vec{q}} g_{\vec{k},\vec{q}}(\sigma^\dagger_{\vec{k}}\sigma_{\vec{q}} + \sigma^\dagger_{\vec{q}}\sigma_{\vec{k}}) \ . \tag{2.2}$$

We shall assume that the dependence of $g_{\vec{k},\vec{q}}$ on $\vec{k}$, $\vec{q}$ is weak and replace it by the coupling constant g. To obtain the tunneling current J, we note that $J = -2e\dot{N}_L$, where N_L represents the number of electron pairs in the left-hand superconductor:

$$N_L = \tfrac{1}{2}\sum_{\vec{k}} (c^\dagger_{\vec{k}}c_{\vec{k}} + c^\dagger_{-\vec{k}}c_{-\vec{k}}) = \sum_{\vec{k}} (\tfrac{1}{2} - \sigma_{z,\vec{k}}) \ . \tag{2.3}$$

The tunneling current J is then given by

$$\begin{aligned} J &= -2e\dot{N}_L \\ &= -2e(i/\hbar)[H_T, N_L] \\ &= 2eg\frac{i}{\hbar}\sum_{\vec{k},\vec{q}} \left[(\sigma^\dagger_{\vec{k}}\sigma_{\vec{q}} + \sigma^\dagger_{\vec{q}}\sigma_{\vec{k}}),\ \sum_{\vec{k}'} \sigma_{z,\vec{k}'}\right] \\ &= -2eg\frac{i}{\hbar}\sum_{\vec{k},\vec{q}} (\sigma^\dagger_{\vec{k}}\sigma_{\vec{q}} - \sigma^\dagger_{\vec{q}}\sigma_{\vec{k}}) \ . \end{aligned} \tag{2.4}$$

To calculate the current to first order in g, we simply calculate the mean value of J as given in Eq. (2.4) with an initial wave function $|\Psi_0\rangle$. Thus if we take the wave function $|\Psi_0\rangle$ to represent two pieces of superconductors both in their ground states with phase ϕ_L on the left, and phase ϕ_R on the right, i.e.,

$$|\Psi_0\rangle = |\phi_L\rangle \otimes |\phi_R\rangle \ , \tag{2.5}$$

where

$$|\phi_L\rangle = \prod_{\vec{k}} (u_{\vec{k}} + v_{\vec{k}}e^{i\phi_L}\sigma_{\vec{k}})|0\rangle$$

and

$$|\phi_R\rangle = \prod_{\vec{q}} (u_{\vec{q}} + v_{\vec{q}}e^{i\phi_R}\sigma_{\vec{q}})|0\rangle \ , \tag{2.6}$$

the average current is given by

$$\begin{aligned} \langle\Psi_0|J|\Psi_0\rangle &= -2eg\frac{i}{\hbar}\sum_{\vec{k},\vec{q}} (\langle\phi_L|\sigma^\dagger_{\vec{k}}|\phi_L\rangle \\ &\quad \times \langle\phi_R|\sigma_{\vec{q}}|\phi_R\rangle - \text{c.c.}) \\ &= -2eg\frac{i}{\hbar}\sum_{\vec{k},\vec{q}} (u_{\vec{k}}v_{\vec{k}}u_{\vec{q}}v_{\vec{q}}e^{i(\phi_L-\phi_R)} - \text{c.c.}) \\ &= j_1\sin(\phi_L - \phi_R) \ , \end{aligned} \tag{2.7}$$

where

$$j_1 = 4e\frac{g}{\hbar}\sum_{\vec{k},\vec{q}} (u_{\vec{k}}v_{\vec{k}}u_{\vec{q}}v_{\vec{q}}) \ . \tag{2.8}$$

Thus we see that the current is nonzero as long as a relative phase its maintained between the superconductors. This current has a maximal magnitude of j_1 as given by Eq. (2.8). This is the dc Josephson effect.[1]

If we examine the above calculation of the tunneling current, it becomes apparant that the properties of the superconductor relevant to the tunneling problem can be described by three operators S, $S^\dagger$, S_z defined as follows:

$$S = S_L S_R^\dagger \ , \tag{2.9a}$$

$$S^\dagger = S_R S_L^\dagger \ , \tag{2.9b}$$

and

$$S_z = \tfrac{1}{2}(S_{z,L} - S_{z,R}) \ , \tag{2.9c}$$

where

$$\begin{aligned} S_L &= \frac{1}{\sqrt{2}m}\sum_{\vec{k}} \sigma^\dagger_{\vec{k}}, \quad S_R = \frac{1}{\sqrt{2}m}\sum_{\vec{q}} \sigma^\dagger_{\vec{q}} \ , \\ S_{z,L} &= -\sum_{\vec{k}} \sigma_{z,\vec{k}} \ , \quad S_{z,R} = -\sum_{\vec{q}} \sigma_{z,\vec{q}} \ . \end{aligned} \tag{2.10}$$

The normalization factor m is the number of electron momentum states with energy between $\epsilon_F - \hbar\omega_D$ and ϵ_F, ω_D being the Debye frequency. We recall here that the attractive interaction responsible for superconductivity involves electron states with energy in the range $\epsilon_F \pm \hbar\omega_D$, and the sum over k in Eq. (2.10) is over momentum states in this range. The operator S_L (S_R) destroys a coherent superposition of pairs on the left (right)-hand superconductor, and $S_{z,L}$ ($S_{z,R}$) describes the number of electron pairs in excess of the value required to maintain charge neutrality. Since the total charge is conserved, we conclude that $S_{z,L} + S_{z,R} = 0$ and by Eq. (2.9c), $S_z = S_{z,L}$. Consequently, S_z describes the number of excess electron pairs on the left superconductor, and S describes the coherent transfer of a pair from left to right. These operators obey the following commutation relations[9]:

$$[S, S_z] = S, \tag{2.11}$$

$$[S^\dagger, S_z] = -S^\dagger, \tag{2.12}$$

$$[S^\dagger, S] \approx (2/m^2)S_z. \tag{2.13}$$

It has been shown in Refs. 3 and 5 that the commutation relation in Eq. (2.13) leads to a small frequency pulling (of order $1/m^2$) of the radiation frequency away from the electrostatic potential. However, since we are presently most interested in obtaining a linewidth which is orders of magnitude broader than the magnitude of the frequency pulling, we shall neglect this small effect and set

$$[S, S^\dagger] = 0. \tag{2.14}$$

With this simplification, we note that the operator $SS^\dagger$ commutes with every operator in the system, and can therefore be treated as a c number. We have chosen a normalization such that

$$S^\dagger S = 1. \tag{2.15}$$

We can then define an operator θ by

$$S = e^{-i\theta}. \tag{2.16}$$

To reproduce the commutation relations (2.11) and (2.12), we shall require that

$$[\theta, S_z] = i. \tag{2.17}$$

The operator S is now recognized as the operator described in Josephson's original paper[1] which transfers a pair without disturbing the superconductors from their respective ground states. The angle θ is the operator corresponding to the macroscopic phase difference $(\phi_L - \phi_R)$, and S_z, as mentioned earlier, describes the number of excess electron pairs on the left-hand superconductor. Equation (2.2) can now be written as

$$H_T = -(\hbar j_1/2e)\tfrac{1}{2}(S^\dagger + S). \tag{2.18}$$

The coefficient $(\hbar j_1/4e)$ has been chosen to yield a correct dc tunneling current as is shown in the following. The current operator J is given by

$$\begin{aligned} J &= -2e\dot{S}_z = -2e(i/\hbar)[H_T, S_z] \\ &= j_1(S^\dagger - S)/2i = j_1 \sin\theta. \end{aligned} \tag{2.19}$$

The maximum value of $\langle J\rangle$ is then given by j_1 as it should be. Equation (2.19) is an operator equation, the mean value of which yields Eq. (2.7).

We next note that Eq. (2.19) describes an ac current when the operator θ has a linear time dependence, as is the case when a voltage is established across the junction. The superconductor-oxide-superconductor sandwich has a capacitance C, the value of which depends on the geometry, but for a typical junction, $C \simeq 1\ \mu$F. When a current in excess of j_1 is forced through the junction, charges pile up on one side of the oxide layer, and a voltage of $2eS_z/C$ develops across the junction. To describe this situation we will add to the Hamiltonian a term corresponding to the energy of the capacitor,

$$H_C = q^2/2C, \tag{2.20}$$

where $q = 2eS_z$ is the total charge on one side of the junction. The operator θ now obeys the following equation of motion:

$$\begin{aligned} \dot{\theta} &= (i/\hbar)[H, \theta] = (i/\hbar)[2e^2S_z^2/C, \theta] \\ &= [(2e)^2/\hbar C]S_z \end{aligned} \tag{2.21}$$

or

$$\theta(t) = [(2e)^2/\hbar C]S_z t + \theta_0. \tag{2.22}$$

We observe that the average voltage V across the junction is given by

$$V = 2e\langle S_z\rangle/C. \tag{2.23}$$

Taking the mean of Eq. (2.22), we obtain

$$\langle\theta(t)\rangle = (2eV/\hbar)t + \langle\theta_0\rangle. \tag{2.24}$$

Putting Eq. (2.24) into the mean equation obtained from Eq. (2.19), we conclude that when a voltage V is established across the junction, an ac current at the frequency $2eV/\hbar$ is produced. This current will in turn emit radiation at the same frequency. Typically this frequency is in the microwave or infrared region. The junction can either be placed in an external resonance cavity, or as it turns out, the junction itself provides a good resonance cavity. The modes of the cavity and the way the radiation field interacts with the superconducting pair current is discussed in Sec. II B.

B. Cavity Modes and Interaction between Radiation Field and Superconductors

Let us consider a Josephson junction consisting of an oxide layer sandwiched between two superconductors. The oxide layer bounded by superconducting surfaces serves as a resonance cavity for microwave radiation with $Q \simeq 10$–100. Let the superconductor surfaces be in the x-y plane. Typical dimensions of the cavity are $l_x \simeq l_y \simeq 0.1$ cm and $l_z \simeq 10^{-7}$ cm, l_z being the thickness of the oxide layer. While the ends of the cavity are open, there is a large impedance mismatch with the outside world that enables us to assume that $\mathcal{H} = 0$ at the boundary. We can then imagine that standing waves are supported in the x direction (or y direction), with wave vector $k_n = n\pi/l_x$. Such modes can be analyzed in detail by solving the usual macroscopic Maxwell equations in the oxide and the Maxwell equations combined with London's equation

$$\vec{J} = -(c/4\pi\lambda)\vec{A} \tag{2.25}$$

in the superconducting region, where λ is the penetration depth. One then proceeds by imposing continuity conditions on E_x and B_y across the oxide-superconductor surface. This has been done by several authors.[10] The main conclusions are: (i) The dominant component of the vector potential is A_z, i.e., the direction pointing into the superconductor. (ii) While the penetration depth λ is usually much larger than the thickness of the oxide

l_z, the bulk of the $\vec{A}$ field is confined to the oxide region. (iii) The oscillation has an effective phase velocity

$$\bar{c} = \left(\frac{l_z}{l_z + 2\lambda} \frac{1}{\epsilon} \right)^{1/2} c \,, \tag{2.26}$$

where ϵ is the dielectric constant of the oxide. Thus the penetration of the field into the superconductors causes the mode frequency to be reduced by as much as a factor of 10.

With these facts in mind, the normalized eigenfunction of the nth mode can roughly be written as

$$\phi_n(x, z) = \begin{cases} (2/l_x l_y l_z)^{1/2} \cos k_n x, & |z| < \frac{1}{2} l_z \\ 0, & \text{otherwise.} \end{cases} \tag{2.27}$$

The radiation field A_z can be quantized in the standard manner,

$$A_z(x, z, t) = \sum_n \left[\frac{2\pi\hbar c^2}{\epsilon \Omega_n} \right]^{1/2} \phi_n(x, z)(a_n e^{-i\Omega_n t} + a_n^\dagger e^{i\Omega_n t}),$$

where

$$\Omega_n = \bar{c} k_n \tag{2.28}$$

and the following commutation relation is obeyed:

$$[a_n, a_{n'}^\dagger] = \delta_{nn'} \,. \tag{2.29}$$

For a complete description of the radiation in the cavity, we need to take into account the loss of radiation through leakage to the outside world and dispersion in the oxide layer. This loss is described by a finite quality factor Q. This is further discussed in Sec. II C.

Next we will couple the field to the superconductors via interaction with the current, i.e.,

$$H_I = (1/c) \int d^3 r \, \vec{A}(\vec{r}) \cdot \vec{j}(\vec{r}) \,, \tag{2.30}$$

where $\vec{j}(r)$ is the current density.

Up to now we have not discussed the spatial variation of $\vec{j}(\vec{r})$, and Eq. (2.19) for the current should be interpreted as a local relation between the current and the local value of the phase $\theta(\vec{r})$. From the relation $\dot{\theta} = (2e/\hbar)V$ it is clear that a change of gauge $V \to V + (1/c)[\partial\chi(r)/\partial t]$, $\vec{A} \to \vec{A} + \nabla\chi$ implies a corresponding change in phase $\theta \to \theta + (2e/\hbar c)\chi(r)$. We then conclude that the equation describing the spatial variation of θ must be of the form

$$\nabla\theta = (2e/\hbar c)\vec{A} \tag{2.31}$$

in order to be gauge invariant. Let us suppose that a constant magnetic field $\vec{H}$ is applied to the junction along the y direction. Following Josephson the change of the phase θ along the barrier in the x direction can be obtained by integrating Eq. (2.31) along a loop in the x-z plane that extends deep inside both superconductors. The change in phase $\Delta\theta$ over a distance Δx is then proportional to the total magnetic flux $\Delta x\, dH$, where d is the barrier thickness l_z plus twice the penetration depth λ. One then obtains the following equation for the spatial variation of θ:

$$\frac{\partial\theta}{\partial x} = -\frac{2ed}{\hbar c} H \,. \tag{2.32}$$

Hence in the presence of a magnetic field H the phase has a spatial dependence of the form $\theta = -Kx$, where $K = 2edH/\hbar c$.

Returning to Eq. (2.30) we can take as a first approximation the expression (2.19) for the current $j(r) = j_1 \sin\theta = j_1 \sin Kx$. Furthermore, due to the small size of the cavity, the cavity modes are well separated in frequency, and we can assume that only one particular mode with frequency Ω is excited. The integral in Eq. (2.30) can then be evaluated, and we can write Eq. (2.30) as

$$H_I = i\hbar T(a^\dagger + a)(S - S^\dagger) \,,$$

where

$$T = j_1 \left(\frac{\pi l_z}{\hbar\epsilon\Omega l_x l_y} \right)^{1/2} \left[\frac{2K \sin[(K - k_n) l_x]}{K^2 - k_n^2} \right],$$

where the factor in [ ] comes from the interference between the spatial variation of the current $\vec{j}(r)$ and the mode function ϕ_n as given in Eq. (2.27). We note that the coupling strength T vanishes when $K = 0$. Hence it is necessary to have a uniform magnetic field in order to observe interaction between the Josephson current and the intrinsic electromagnetic modes of the junction. This is so because the region of interaction is comparable to the wavelength of the radiation field. However, in the case of a point contact placed inside a resonance cavity, the region of interaction becomes much smaller than the wavelength of the radiation. This situation is then more analogous to an atom sitting inside a cavity and a magnetic field would not be necessary for coupling to take place.

To summarize the total Hamiltonian for the interacting system is given by

$$H = \hbar\Omega a^\dagger a - (\hbar j_1/2e)\tfrac{1}{2}(S + S^\dagger) + i\hbar T(a^\dagger + a)(S - S^\dagger) + 2e^2 S_z^2/C. \tag{2.33}$$

Recall that $S = e^{-i\theta}$, and according to Eq. (2.22), $e^{-i\theta}$ and $e^{i\theta}$ oscillate at a frequency $2eV/\hbar$. At the same time, a and $a^\dagger$ oscillate at their natural frequency Ω. We may then throw out terms in the Hamiltonian which have this rapid oscillatory time dependence. Hence we will neglect the j_1 term which is associated with an ac current at frequency $2eV/\hbar$, and make a rotating wave approximation on the T term, keeping only the difference frequency components. Our working Hamiltonian is now

$$H = \hbar\Omega a^\dagger a + (2e^2/C)S_z^2 + i\hbar T(a^\dagger S - aS^\dagger) \ . \qquad (2.34)$$

C. Cavity Q and External Current

In order to set up a description of the nonequilibrium problem, we need, in addition to the Hamiltonian, a description of the finite Q of the resonance cavity, as well as that of the external current forced through the junction. A good model for the effect of the finite Q of the cavity is to couple the radiation field to a heat bath consisting of harmonic oscillators or two-level atoms at temperature T. [11,12] The result is a contribution to the time evolution of the density matrix in addition to that implied by the Hamiltonian given in Eq. (2.34):

$$\left(\frac{\partial\rho}{\partial t}\right)_Q = -\tfrac{1}{2}(\bar{n}+1)\,\frac{\nu}{Q}\,(a^\dagger a\,\rho + \rho a^\dagger a - 2a\rho a^\dagger)$$
$$-\tfrac{1}{2}\bar{n}\,\frac{\nu}{Q}(aa^\dagger\rho + \rho aa^\dagger - 2a^\dagger\rho a) \ , \qquad (2.35)$$

where $\bar{n}$ is the Planck function $(e^{\hbar\Omega/kT}-1)^{-1}$, or the average number of thermal photons in a blackbody cavity at temperature T. Equation (2.35) describes the absorption of energy from the field at the rate $(\nu/Q)(\bar{n}+1)$ and the flow of energy into the field at the rate $(\nu/Q)\,\bar{n}$, and hence implies a net loss of (ν/Q) photons per unit time.

A similar approach will be adopted to provide a model for the current flow. We will couple the pair creation and annihilation operators $S^\dagger$ and S to an external electron reservoir, S and $S^\dagger$ now playing the role of a and $a^\dagger$ in Eq. (2.35). The following equation then provides a description of the increase in the number of pairs on the left-hand superconductor:

$$(\dot{\rho})_{\text{current}} = -\tfrac{1}{2}A(SS^\dagger\rho + \rho SS^\dagger - 2S^\dagger\rho S) \ . \qquad (2.36)$$

We can calculate the rate of increase $\langle \dot{S}_z\rangle$:

$$\langle \dot{S}_z\rangle = -\tfrac{1}{2}A\,\mathrm{Tr}([S_z, S]S^\dagger\rho + S[S^\dagger, S_z]\rho) = A. \qquad (2.37)$$

Equation (2.36) then describes a flow of A pairs per unit time, or a current of $J_{\text{dc}} = 2eA$.

To summarize, our interacting system is described by a density matrix the time evolution of which is given by the following equation:

$$\dot{\rho} = -(i/\hbar)[H,\rho] + (\dot{\rho})_Q + (\dot{\rho})_{\text{current}} \ , \qquad (2.38)$$

where H is given by Eq. (2.34); $(\dot{\rho})_Q$ and $(\dot{\rho})_{\text{current}}$ are given by Eqs. (2.35) and (2.36), respectively. A pictorial representation of Eq. (2.38) is found in Fig. 1.

D. Mean Equations of Motion

To gain some insight into the physical implications of the equation of motion for the density

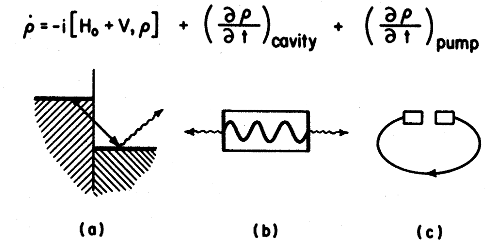

FIG. 1. Illustration of the density-matrix equation of motion. (a) Two superconductors separated by the barrier and maintained at different chemical potentials. An electron pair drops from the higher potential on the left to the lower potential on the right, and emits a single photon. (b) An imperfect resonance cavity having a finite quality factor Q. (c) Illustrates the role of the battery as electron pump, bringing the electrons from the right-hand superconductor back to the left via wires of normal metal.

matrix, we find it instructive to make use of Eq. (2.38) and write the mean equations of motion for $\langle a\rangle$, $\langle S\rangle$, and $\langle S_z\rangle$:

$$\langle \dot{a}\rangle = -[i\Omega + (\nu/2Q)]\langle a\rangle + T\langle S\rangle \ , \qquad (2.39)$$

$$\langle \dot{S}\rangle = -i[2e^2/\hbar C]\langle S_z S + SS_z\rangle$$
$$\approx -i[(2e)^2/\hbar C]\langle S_z\rangle\langle S\rangle$$
$$= -i(2eV/\hbar)\langle S\rangle \ , \qquad (2.40)$$

$$\langle \dot{S}_z\rangle = -T\langle aS^\dagger + a^\dagger S\rangle + A$$
$$\approx -T(\langle a\rangle\langle S\rangle^* + \langle a\rangle^*\langle S\rangle) + A. \qquad (2.41)$$

In deriving Eqs. (2.40) and (2.41), we have factorized the mean values of products of operators. Equation (2.39) implies that the average radiation field $\langle a\rangle$ is driven by the current $T\langle S\rangle$ which oscillates, according to (2.40), at a frequency $\omega = 2eV/\hbar$. The mean value $\langle a\rangle$ then oscillates at the same frequency ω and reaches the steady-state value

$$\langle a\rangle = \frac{T\langle S\rangle}{i(\Omega-\omega) + (\nu/2Q)} \ . \qquad (2.42)$$

We next recall that the total current J is given by $J = 2e\langle \dot{S}_z\rangle$. Equation (2.41) is then an expression for the total current through the junction. At steady state $\langle \dot{S}_z\rangle = 0$, and we obtain, using Eq. (2.42),

$$A = T(\langle a^\dagger\rangle\langle S\rangle + \langle a\rangle\langle S^\dagger\rangle)$$
$$= \frac{T^2(\nu/Q)\langle S\rangle\langle S^\dagger\rangle}{[\Omega - (2e/\hbar)V]^2 + (\nu/2Q)^2} \ . \qquad (2.43)$$

The presence of the radiation field then gives rise to a dc current which is balanced by the external current when steady state is reached.

E. Philosphy of Linewidth Calculation

It is of interest to note that while ω/V has been measured to such great accuracy as to give $e/\hbar$, the linewidth of the Josephson radiation is about one part in 10^7. Compared with the laser linewidth of one part in 10^{15}, the Josephson linewidth is rather broad. The physical explanation behind this is that when electron pairs tunnel across the junction via interaction with the radiation field, the charge number, and therefore the voltage, fluctuates about the steady-state mean value calculated earlier. This gives rise to a frequency modulation of the radiation, and is the main contribution to the linewidth.

In Sec. III we present a calculation of the linewidth for the case where the resonance cavity is assumed to be at temperature $T=0$. But first we would like to outline the idea behind the calculation. We recall that the system is described by the density matrix ρ which satisfies the equation of motion

$$\dot{\rho} = -(i/\hbar)[H,\rho] + (\dot{\rho})_Q + (\dot{\rho})_{\text{current}} , \qquad (2.44)$$

where

$$H = \hbar\,\Omega a^\dagger a + 2e^2 S_z^2/C + i\hbar T(a^\dagger e^{-i\theta} - a e^{i\theta}). \qquad (2.45)$$

Our model describes a radiation field interacting with the supercurrent $e^{-i\theta}$ in the same way as a damped harmonic oscillator driven by a classical external force. As is well known, the oscillator will oscillate at the frequency of the driving force. Now if we further assume that the damping constant is much greater than the linewidth $\Delta\nu$ of the driving force,

$$\nu/Q \gg \Delta\nu , \qquad (2.46)$$

the linewidth of the radiation will equal $\Delta\nu$ as given by the correlation function of the driving force,

$$G_\theta(t,t') = \langle e^{-i\theta(t)}\, e^{i\theta(t')}\rangle . \qquad (2.47)$$

The correlation function G_θ arises out of the operator nature of Eq. (2.21), which implies that the rate at which the phase angle θ rotates is sensitive to the local fluctuation of the pair population S_z in the junction area.

Let us define

$$\delta N = S_z - \langle S_z\rangle \qquad (2.48)$$

and

$$\delta\theta = \theta - [(2e)^2/\hbar C]\langle S_z\rangle t . \qquad (2.49)$$

Then

$$\delta\dot{\theta} = [(2e)^2/\hbar C]\delta N . \qquad (2.50)$$

Equation (2.47) becomes

$$G_\theta(t,t') = \exp[-i(2eV/\hbar)(t-t')]\langle e^{-i\delta\theta(t)}\, e^{i\delta\,\theta(t')}\rangle . \qquad (2.51)$$

Making the Gaussian approximation, we obtain

$$G_\theta(t,t') = \exp[-i(2eV/\hbar)(t-t')]\exp\{-\tfrac{1}{2}\langle[\delta\theta(t)-\delta\theta(t')]^2\rangle\}$$

$$= \exp[-i(2eV/\hbar)(t-t')]$$

$$\times \exp\{-\tfrac{1}{2}\langle\{\textstyle\int_{t'}^{t} dt_1\,[(2e)^2/\hbar C]\delta N(t_1)\}^2\rangle\}$$

$$= \exp[-i(2eV/\hbar)(t-t')]$$

$$\times\exp\left[-\tfrac{1}{2}\left(\frac{(2e)^2}{\hbar C}\right)^2\int_{t'}^{t}\int_{t'}^{t} dt_1\,dt_2\,\langle\delta N(t_1)\delta N(t_2)\rangle\right] . \qquad (2.52)$$

In order to obtain the linewidth of the emitted radiation, we must calculate the number correlation function

$$G_N(t_1,t_2) = \langle\delta N(t_1)\delta N(t_2)\rangle . \qquad (2.53)$$

This is done in Sec. III.

III. LINEWIDTH AT ZERO CAVITY TEMPERATURE

The description of the superconductors relevant to our discussion can be spanned by the eigenstates of S_z

$$S_z|k\rangle = k|k\rangle . \qquad (3.1)$$

From the commutation relations (2.11) and (2.12) and the normalization condition (2.15), we conclude that

$$S|k\rangle = |k-1\rangle , \quad S^\dagger|k\rangle = |k+1\rangle . \qquad (3.2)$$

The state vector $|k\rangle$ has the interpretation of representing two superconductors both in their respective BCS states with k excess electron pairs on the left-hand side. We can now write down the equation of motion for the density operator [Eq. (2.44)] in n representation for the field, and pair-number representation for the superconductors. In this section we shall restrict our attention to the case where $\bar{n}=0$, i.e., the cavity is at zero temperature. The basic physics can be brought out more easily in this case, and we shall treat the general case of $\bar{n}\neq 0$ which requires more involved techniques in a separate publication. Using Eqs. (2.34)–(2.36) and (2.38) and setting $\bar{n}=0$, we obtain

$$\dot{\rho}_{k,n;k',n'} = -i\left(\Omega(n-n') + \frac{(2e)^2}{\hbar C}\;\frac{k+k'}{2}\;(k-k')\right)\rho_{k,n;k',n'}$$

$$- T[(n+1)^{1/2}\rho_{k-1,n+1;k',n'} - (n')^{1/2}\rho_{k,n;k'+1,n'-1}]$$

$$+ T[(n)^{1/2}\rho_{k+1,n-1;k',n'} - (n'+1)^{1/2}\rho_{k,n;k'-1,n'+1}]$$

$$- (\nu/2Q)\{[(n+1)+(n'+1)]\rho_{k,n;k',n'}$$

$$-2[(n+1)(n'+1)]^{1/2}\rho_{k,n+1;k',n'+1}\}$$

$$-A(\rho_{k,n;k',n'}-\rho_{k-1,n;k'-1,n'}). \quad (3.3)$$

As discussed in Sec. II E we are interested in calculating the pair-number correlation function

$$G_N(t_1,t_2)=\langle\delta N(t_1)\delta N(t_2)\rangle \ .$$

This correlation function involves only the superconductor coordinates and hence can be calculated from the equation of motion of the reduced density $\sigma_{k,k'}$ obtained by tracing over the radiation-field coordinates as follows:

$$\sigma_{k,k'}=\sum_n \rho_{k,n;k',n} \ . \quad (3.4)$$

An equation of motion for the reduced density matrix can be obtained by performing an adiabatic elimination of the photon coordinates from Eq. (3.3). This is achieved by noting that the radiation-field characteristic time $(\nu/Q)^{-1}$ is the fastest decay time in the problem, and hence the radiation field follows the time development of the electron-pair population. The time development of the entire system is then described by the time evolution of the reduced density matrix $\sigma_{k,k'}$ alone. Details of the adiabatic elimination are discussed in Appendix A. The resulting equation of motion for the diagonal element of the reduced density matrix is the following:

$$\dot{\sigma}_{k,k}=-(1/2e)[J(k)\sigma_{k,k}-J(k+1)\sigma_{k+1,k+1}]$$
$$-A(\sigma_{k,k}-\sigma_{k-1,k-1}) \ , \quad (3.5)$$

where

$$J(k)=\frac{2eT^2(\nu/Q)}{\{\Omega-[(2e)^2/\hbar C](k-\frac{1}{2})\}^2+(\nu/2Q)^2} \ . \quad (3.6)$$

Equation (3.5) can be interpreted in terms of the flow of probability from the k-to-$(k-1)$ and the k-to-$(k+1)$ excess pair states at the rate A and $J(k)$, respectively. This is shown in Fig. 2. Furthermore, detailed balance can be used to obtain the steady-state solution for $\sigma_{k,k}$.

Next, let us define $\bar{k}$ as the mean number of

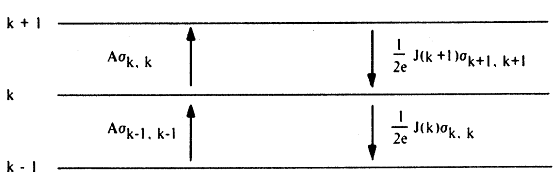

FIG. 2. Flow of probability for having k excess pairs on the left-hand side when the cavity temperature $T=0$. Terms proportional to A are due to external current and drive the system toward higher values of k, while terms resulting from tunneling tend to lower the number of excess pairs, as indicated by the arrows pointing down.

pairs at steady state,

$$\bar{k}=\sum_k k(\sigma_{k,k})_{\text{steady state}} \ . \quad (3.7)$$

We may approximate Eq. (3.6) for $J(k)$ by expanding $J(k)$ about the mean value $\bar{k}$:

$$J(k)=J(\bar{k})+\frac{\partial J}{\partial \bar{k}}(k-\bar{k})+\cdots \ . \quad (3.8)$$

Keeping only the first two terms of Eq. (3.8), Eq. (3.5) becomes

$$\dot{\sigma}_{k,k}=-\frac{J(\bar{k})}{2e}(\sigma_{k,k}-\sigma_{k+1,k+1})-A(\sigma_{k,k}-\sigma_{k-1,k-1})$$
$$-\frac{1}{2e}\frac{\partial J}{\partial \bar{k}}[(k-\bar{k})\sigma_{k,k}-(k+1-\bar{k})\sigma_{k+1,k+1}] \ . \quad (3.9)$$

Let us examine the mean equation of motion for S_z as implied by Eq. (3.9):

$$\langle\dot{S}_z\rangle=\sum k\dot{\sigma}_{k,k}=\left(A-\frac{J(\bar{k})}{2e}\right)-\frac{1}{2e}\frac{\partial J}{\partial \bar{k}}(\langle S_z\rangle-\bar{k}). \quad (3.10)$$

This leads us to identify the average tunneling current with the external current as follows:

$$J_{\text{dc}}=2eA=J(\bar{k}) \ . \quad (3.11)$$

After making this identification, Eq. (3.10) has the following solution:

$$[\langle S_z(t)\rangle-\bar{k}]=[\langle S_z(0)\rangle-\bar{k}]\exp\left(-\frac{1}{2e}\frac{\partial J}{\partial \bar{k}}t\right) \ . \quad (3.12)$$

This shows that $\langle S_z(t)\rangle$ decays to the steady-state value $\bar{k}$ at the rate $(1/2e)(\partial J/\partial\bar{k})$. Recalling that $V=(2e/C)\bar{k}$, the rate can be written in terms of more physical quantities,

$$\frac{1}{2e}\frac{\partial J}{\partial \bar{k}}=(R_D C)^{-1} \ , \quad (3.13)$$

where $R_D=(\partial J/\partial V)^{-1}$ is the dynamic resistance of the junction.

To calculate the linewidth $\Delta\nu$, we need, as noted in Sec. II,

$$G_N(t)=\langle\delta N(t)\delta N(0)\rangle \ ,$$

where

$$\delta N(t)=S_Z(t)-\bar{k} \ .$$

We shall have to apply the Onsager regression theorem,[13,14] which is based on the statement that at $t=0$, when we make the first measurement, the superconductor and radiation-field density matrix factorizes as follows:

$$\rho(0)=\sigma(0)\otimes r(0) \ . \quad (3.14)$$

The correlation function $G_N(t)$ then describes how the system evolves to time t.

With this additional assumption we obtain the following expression for $G_N(t)$:

$$G_N(t) = G_N(0)\, e^{-(R_D C)^{-1} t} , \tag{3.15}$$

where

$$G_N(0) = \mathrm{Tr}[(\delta N)^2 \sigma(0)] \tag{3.16}$$

and $\sigma(0)$ is the steady-state solution to Eq. (3.9). For completeness, a proof of the regression theorem applied to this particular situation is given in Appendix B.

Setting $\dot{\sigma}_{k,k} = 0$ in Eq. (3.9), we multiply both sides of the equation by $(k-\bar{k})^2$ and sum over k. After a straightforward calculation we obtain the following expression for $G_N(0)$:

$$\langle \delta N^2(0) \rangle = \left(\frac{J(\bar{k})}{2e} + A \right) \Big/ [2(R_D C)^{-1}] . \tag{3.17}$$

We note in this expression that the contribution from the term proportional to A arises entirely out of our model which has the external current flowing in discrete numbers. The shot noise that arises can be "smoothed out" in an experimental situation by placing a large resistance in the external circuit. The shot noise then becomes Johnson noise kT/R. A discussion of how shot noise and Johnson noise both arise out of the fact that the electric charge is a discrete quantity is found in Ref. 15. With a sufficiently large resistance in the external circuit, we will neglect the contribution to Eq. (3.17) arising from the external current and keep only the contribution from $J(\bar{k})$, which arises from the actual tunneling of electron pairs with the emission or absorption of quantized photons.

Our expression for $G_N(t)$ is then

$$G_N(t) = \{\tfrac{1}{2}[J(\bar{k})/2e] R_D C\} \exp[-(R_D C)^{-1} t] . \tag{3.18}$$

The knowledge of $G_N(t)$ enables us to calculate $G_\theta(t, t')$ according to Eq. (2.52). Standard FM theory[16] can be applied here; the derivation is outlined as follows:

$$G_\theta(t) = e^{-i(2eV/\hbar)t} \times \exp\left[-\frac{1}{2} \left(\frac{(2e)^2}{\hbar C} \right)^2 \int_0^t \int_0^t dt_1\, dt_2 \langle \delta N(t_1)\, \delta N(t_2) \rangle \right]. \tag{3.19}$$

We make the change of variables

$$\sigma = \tfrac{1}{2}(t_1 + t_2) , \qquad \tau = t_1 - t_2 . \tag{3.20}$$

Equation (3.19) becomes

$$G_\theta(t) = e^{-i(2eV/\hbar)t} \times \exp\left[-\frac{1}{2} \left(\frac{(2e)^2}{\hbar C} \right)^2 2 \int_0^{t/2} d\sigma \int_{-2\sigma}^{2\sigma} d\tau\, G_N(\tau) \right]. \tag{3.21}$$

We note that $G_N(\tau)$ decays to zero in the characteristic time $R_D C$ and contributes to the τ integral only in this range. Furthermore the resulting linewidth $\Delta\nu$ is expected to be much smaller than $(R_D C)^{-1}$. Hence we are interested in the behavior of $G_\theta(t)$ for

$$t \gg R_D C , \tag{3.22}$$

and we can extend the limits of the τ integration to infinity. We then obtain

$$\begin{aligned} G_\theta(t) &= e^{-i(2eV/\hbar)t} \exp\left[-\frac{1}{2} \left(\frac{(2e)^2}{\hbar C} \right)^2 \left(\int_{-\infty}^{\infty} d\tau\, G_N(\tau) \right) t \right] \\ &= e^{-i(2eV/\hbar)t} \exp\left[-\frac{1}{2} \left(\frac{8e^3}{\hbar^2} J_{\mathrm{dc}} R_D^2 \right) t \right] . \end{aligned} \tag{3.23}$$

The spectrum is Lorentzian centered at frequency

$$\omega = 2eV/\hbar , \tag{3.24}$$

with a full width at half-height $\Delta\nu$ in Hz,

$$\Delta\nu = \frac{8e^3}{2\pi\hbar^2} J_{\mathrm{dc}} R_D^2 . \tag{3.25}$$

APPENDIX A: DERIVATION OF EQUATION FOR REDUCED DENSITY MATRIX

In this Appendix we present a derivation of the equation of motion for the reduced density matrix [Eq. (3.5)]. From Eq. (3.3) in the text we have

$$\begin{aligned} \dot{\sigma}_{k,k} = \sum_n \dot{\rho}_{k,n;k,n} = &-T\left[\sum_n (n+1)^{1/2} \rho_{k-1,n+1;k,n} - \sum_n (n)^{1/2} \rho_{k,n;k+1,n-1}\right] + T\left[\sum_n (n)^{1/2} \rho_{k+1,n-1;k,n} \right. \\ &\left. - \sum_n (n+1)^{1/2} \rho_{k,n;k-1,n+1}\right] - A(\sigma_{k,k} - \sigma_{k-1,k-1}) . \end{aligned} \tag{A1}$$

Next we need an equation for $\sum_n (n+1)^{1/2} \rho_{k-1,n+1;k,n}$ as it appears in (A1). Hence we write

$$\frac{\partial}{\partial t}\left(\sum_n (n+1)^{1/2} \rho_{k-1,n+1;k,n} \right) = -\left[i\left(\Omega - \frac{(2e)^2}{\hbar C} \frac{(2k-1)}{2} \right) + \frac{\nu}{2Q} \right] \left(\sum_n (n+1)^{1/2} \rho_{k-1,n+1;k,n} \right) + T\left(\sum_n (n+1) \rho_{k,n;k,n} \right.$$

$$-\sum_n (n+1)\rho_{k,n+1;k,n+1}\Big) - T\Big(\sum_n [(n+2)(n+1)]^{1/2}\rho_{k-2,n+2;k,n} - \sum_n [(n+1)n]^{1/2}\rho_{k-1,n+1;k+1,n-1}\Big)$$

$$-A\Big(\sum_n (n+1)^{1/2}\rho_{k-1,n+1;k,n} - \sum_n (n+1)^{1/2}\rho_{k-2,n+1;k-1,n}\Big).$$

Equation (A2) can be written as

$$-\left[i\left(\Omega - \frac{(2e)^2}{\hbar C}(k-\tfrac{1}{2})\right) + \frac{\nu}{2Q}\right]\left(\sum_n (n+1)^{1/2}\rho_{k-1,n+1;k,n}\right) = T\sum_n \rho_{k,n;k,n} + \eta\,, \tag{A3}$$

where

$$\eta = -\sum_n (n+1)^{1/2}\dot{\rho}_{k-1,n+1;k,n} + T\sum_n n(\rho_{k,n;k,n} - \rho_{k-1,n;k-1,n}) - T\sum_n [(n+1)n]^{1/2}(\rho_{k-2,n+1;k,n-1} - \rho_{k-1,n+1;k+1,n-1})$$

$$-A\left[\sum_n (n+1)^{1/2}\rho_{k-1,n+1;k,n} - \sum_n (n+1)^{1/2}\rho_{k-2,n+1;k-1,n}\right]. \tag{A4}$$

We now note that for a given charge configuration k or k' the equation of motion for the density matrix [Eq. (3.3)] describes an oscillating current at a frequency $2e^2(k+k')/\hbar C$ driving the radiation field which has a natural frequency Ω and a damping constant ν/Q. Recall that the damping constant ν/Q is assumed to be large [Eq. (2.46)]. The characteristic times of the radiation field are then much faster than those of the superconductors. We can make the adiabatic approximation and assume that the field decays rapidly to the steady state corresponding to the driving force, i.e., for a given $k+k'$. Such a steady-state solution $\mathcal{S}(k+k')$ is well known. It is simply the coherent state[17] $|\beta\rangle\langle\beta|$, where

$$\beta(k+k') = \frac{T}{i\Delta(k+k') + (\nu/2Q)}\exp\{-[2e^2(k+k')/\hbar C]t\}$$

and

$$\Delta(k+k') = \Omega - \frac{(2e)^2}{\hbar C}\frac{(k+k')}{2}\,. \tag{A5}$$

In the number representation,

$$\mathcal{S}_{n,n'}(k+k') = \frac{\beta^n\beta^{*n'}}{(n!)^{1/2}(n'!)^{1/2}}e^{-|\beta|^2}\,. \tag{A6}$$

We will now make the ansatz that

$$\rho_{k,n;k',n'} = \sigma_{k,k'} \otimes \mathcal{S}_{n,n'}(k+k')\,. \tag{A7}$$

This is similar to the ansatz made in the Born-Oppenheimer treatment of the diatomic molecule. There, a wave-function description can be used and the approximation involves assuming a wave function $\Psi(r_i, R_i)$ of the form

$$\Psi(r_i, R_i) = f_k(R_i)\phi_k(r_i, R_i)\,, \tag{A8}$$

where $\phi_k(r_i, R_i)$ is the eigenstate with electron coordinates r_i for fixed nuclear coordinates R_i. An eigenvalue equation is then obtained for the function $f_k(R_i)$. Our treatment can be regarded as a generalization of the adiabatic approximation to a nonequilibrium situation.

Using this ansatz, the terms in Eq. (A4) can be written in terms of $\sigma_{k,k'}$. We work out a typical term as follows:

$$\sum_n n\rho_{k,n;k,n} = \sigma_{k,k}\sum_n n\frac{\beta^n\beta^{*n}}{(n!)^{1/2}(n!)^{1/2}}e^{-|\beta|^2}$$

$$= \sigma_{k,k}\beta\beta^*\sum_n \mathcal{S}_{n-1,n-1}$$

$$= \beta\beta^*\sigma_{k,k}\,. \tag{A9}$$

Proceeding in the same way for all the other terms in Eq. (A4), we obtain

$$\eta = \frac{\partial}{\partial t}\beta\sigma_{k-1,k} + T\beta\beta^*(\sigma_{k,k} - \sigma_{k-1,k-1})$$

$$-T\beta\beta(\sigma_{k-1,k} - \sigma_{k-1,k+1}) - A\beta(\sigma_{k-1,k} - \sigma_{k-2,k-1})\,. \tag{A10}$$

Next we note that $(\partial/\partial t)\sigma_{k-1,k}$ can be calculated from Eq. (3.3) in exactly the same way Eq. (A1) was obtained and we have the following expression:

$$\frac{\partial}{\partial t}(\beta\sigma_{k-1,k}) = \dot{\beta}\sigma_{k-1,k} + \beta\dot{\sigma}_{k-1,k}$$

$$= -T\beta\left[\sum_n (n+1)^{1/2}\rho_{k-2,n+1;k,n} - \sum_n (n)^{1/2}\rho_{k-1,n;k+1,n-1}\right]$$

$$+T\beta\left[\sum_n (n)^{1/2}\rho_{k,n-1;k,n} - \sum_n (n+1)^{1/2}\rho_{k-1,n;k-1,n+1}\right]$$

$$-A\beta(\sigma_{k-1,k} - \sigma_{k-2,k-1})\,. \tag{A11}$$

If we now use the ansatz to calculate the four terms proportional to T on the right-hand side, we obtain

$$\frac{\partial}{\partial t}(\beta\sigma_{k-1,k}) = -T\beta^2(\sigma_{k-2,k}-\sigma_{k-1,k+2}) + T\beta\beta^*(\sigma_{k,k}-\sigma_{k-1\,k-1}) - A\beta(\sigma_{k-1,k}-\sigma_{k-2,k-1}) . \tag{A12}$$

With Eq. (A12) we immediately see that the right-hand side of Eq. (A10) is zero. Thus $\eta = 0$ and Eq. (A3) becomes

$$\sum_n \left(n+1\right)^{1/2}\rho_{k-1,n+1;k,n} = \frac{T}{-i\{\Omega - [(2e)^2/\hbar C](k-\frac{1}{2})\} + (\nu/2Q)} \sum_n \rho_{k,n;k,n} . \tag{A13}$$

Replacing the terms on the right-hand side of Eq. (A1) with the aid of Eq. (A13), we obtain Eq. (3.5) in the text.

APPENDIX B: PROOF OF REGRESSION THEOREM

Our objective in this Appendix is to calculate, given the equation of motion for the reduced density matrix, the two-time correlation function $G_N(t)$:

$$G_N(t) = \langle \delta N(t)\delta N(0)\rangle \equiv \mathrm{Tr}_{R,S}[U^{-1}(t)\delta N(0)U(t)\delta N(0)\rho(0)] . \tag{B1}$$

In Eq. (B1),

$$U(t) = \exp[-(i/\hbar\, H_{\text{tot}}\, t] \tag{B2}$$

is the time-evolution operator for the combined radiation-superconductor system governed by the total Hamiltonian H_{tot}. The symbols R and S denote the vector spaces of the radiation field and the superconductors, respectively.

To make contact with our knowledge of the time evolution of the reduced density matrix, let us consider that at a time $t = 0$ a measurement is made and that the density operator for the combined system is

$$\rho(0) = \sigma(0) \otimes r(0) . \tag{B3}$$

Next we allow the system to evolve according to $U(t)$ to a time t, at which instant a second measurement is made. With the assumption (B3), Eq. (B1) can be written as

$$G_N(t) = \mathrm{Tr}_R\{\delta N\, \mathrm{Tr}_S[U(t)\delta N(\sigma(0)\otimes r(0))U^{-1}(t)]\}$$

$$= \sum_k (k-\bar{k}) \sum_{n,n',n''} \sum_{k'k''} \{U_{k,n;k'n'}(t)(k'-\bar{k}) \times \sigma_{k',k''}(0) r_{n',n''}(0)[U^{-1}(t)]_{k'',n'';k,n}\}$$

$$= \sum_k (k-\bar{k})\sigma'_{k,k}(t) , \tag{B4}$$

where

$$\sigma'_{k,k}(t) \equiv \sum_{k',k''} O_{k,k;k',k''}(t)(k'-\bar{k})\sigma_{k',k''}(0) \tag{B5}$$

and

$$O_{k,k;k',k''}(t) \equiv \sum_{n,n',n''} U_{k,n;k',n'}(t)\, r_{n',n''}\, [U^{-1}(t)]_{k'',n'';k,n} . \tag{B6}$$

At this point we recall that

$$\sigma_{k,k}(t) = \mathrm{Tr}_R(U(t)\rho(0)U^{-1}(t))$$

$$= \sum_{k',k''} \sum_{n,n',n''} U_{k,n;k',n'}(t) r_{n',n''}(0) \times \sigma_{k',k''}(0)[U^{-1}(t)]_{k'',n'';k,n}$$

$$= \sum_{k'k''} O_{k,k;k',k''}(t)\sigma_{k',k''}(0) . \tag{B7}$$

Noting that the only time dependence of both $\sigma'(t)$ and $\sigma(t)$ as given by Eqs. (B5) and (B7) is contained in the matrix $O(t)$, we conclude that $\sigma'(t)$ satisfies the same equation of motion as $\sigma(t)$, the only difference being that they have different initial conditions, i.e.,

$$\sigma'_{k,k}(0) = (k-\bar{k})\sigma_{k,k}(0) . \tag{B8}$$

Hence we can use Eq. (3.9) as an equation for $\sigma'_{k,k}(t)$,

$$\dot{\sigma}'_{k,k}(t) = -[J(\bar{k})/2e](\sigma'_{k,k} - \sigma'_{k+1,k+1}) - A(\sigma'_{k,k} - \sigma'_{k-1,k-1}) - \frac{1}{2e}\frac{\partial J}{\partial \bar{k}}[(k-\bar{k})\sigma'_{k,k} - (k+1-\bar{k})\sigma'_{k+1,k+1}] . \tag{B 9}$$

Multiplying both sides of Eq. (B9) with $(k-\bar{k})$ and summing over k, we obtain an equation of motion for $G_N(t)$,

$$\dot{G}_N(t) = -\frac{1}{2e}\frac{\partial J}{\partial \bar{k}} G_N(t) \tag{B10}$$

or

$$G_N(t) = G_N(0)\, e^{-(1/2e)(\partial J/\partial \bar{k})t} , \tag{B11}$$

with the initial condition $G_N(0)$ given by

$$G_N(0) = \sum_k (k-\bar{k})\sigma'_{k,k}(0) = \sum_k (k-\bar{k})^2\sigma_{k,k}(0) . \tag{B12}$$

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Specific Heat of Superconducting Transition Metals Containing Nonmagnetic Impurities at Low Temperatures*

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Green's-function theory is used for the calculation of the specific heat of superconducting transition metals containing nonmagnetic impurities at low temperatures, $T \gtrsim 0$. It is found that in this temperature region, the s-band specific heat is lowered by the presence of the nonmagnetic impurities. The logarithm of the s-band specific heat obeys the following relation: $\ln C_s = \ln C_s^{(0)} - \Gamma/k_B T$ for $T \gtrsim 0$, where $C_s^{(0)}$ is the s-band specific heat for a pure two-band superconductor, and Γ is proportional to the density of impurities, the density of states at the d-band Fermi surface, and the strength of the interband impurity scattering. This relation agrees very well with the low-temperature experimental data of Shen, Senozan, and Phillips.

I. INTRODUCTION

It was first proposed by Suhl, Matthias, and Walker (SMW)[1] that at low temperatures, both the s-band and the d-band electrons in transition metals, such as niobium and vanadium, can be in the superconducting phase. This model is known as the two-band model. Recently, the two-band model has received considerable attention, since it does succeed in explaining various physical properties of the superconducting transition metals.[2] On the side of experiments, particularly noteworthy are the specific-heat measurements made by Shen, Senozan, and Phillips[3] in the low-temperature region of the niobium superconductors. They notice that there are two slopes appearing in the logarithm of specific heat versus T^{-1} plot, a larger slope near the transition temperature and a smaller slope in the low-temperature region, $T \gtrsim 0$. Based on the BCS theory and the SMW model, they identify the two slopes as due to the existence of two order parameters in the two-band system, Δ_s and Δ_d, for s band and d band, respectively. The two-slope behavior in the $\ln C$ versus T^{-1} plot is not limited to *pure* niobium crystals. The same behavior is also observed in *impure* niobium crystals, but then the values of the specific heat are generally lowered by the impurities in the temperature region, $10^{-1} T_c > T > 0$. A simple analysis of the data for a *pure* niobium superconductor has been separately given by Sung and Shen.[4] The analysis is simple in the sense that they have only fitted the BCS theory for *pure* one-band superconductors to the data of Shen *et al.* to obtain the values of Δ_s and Δ_d. It should be pointed out that so far no systematic attempt has been made to interpret the specific-heat data of *impure* niobium superconductors, particularly in the most interesting low-temperature region, $10^{-1} T_c > T > 0$.

Recently, a detailed investigation of the thermo-