Radiation-Induced Bistability in Josephson Junctions

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(Received 31 December 1979)

It is predicted that a nonequilibrium first-order phase transition analogous to optical bistability can occur when external coherent radiation is applied to a suitably prepared Josephson junction with an external resistance across it. The size of the hysteresis region can be tuned by varying the resistance.

PACS numbers: 74.50.+r, 74.30.Gn, 42.65.Gv

Recently there has been an upsurge of activity[1-5] in nonequilibrium steady states and nonequilibrium phase transitions in externally driven systems. Remarkable new physical effects are seen in such systems. For example, a superconductor irradiated by photons of less than pair-breaking energies is driven into a nonequilibrium steady state in which the order parameter is enhanced.[1] Optical bistability[3] is an example of nonequilibrium first-order phase transition that occurs when an absorbing medium is subjected to resonant coherent irradiation. These systems exhibit interesting switching and hysteresis effects as a function of external parameters.

In this Letter we predict that such effects controlled by external coherent radiation can occur in suitably prepared Josephson junctions. The coherent radiation produces a dc current in a circuit consisting of the junction and a resistance. This dc current, or equivalently, the dc voltage $V_{\rm dc}$ developed across the resistance, is shown to exhibit bistable behavior and hysteresis as a function of the external radiation intensity. The frequency of the associated ac Josephson current will also exhibit bistability. For a given junction, the size of the hysteresis region in the voltage-intensity plot can be tuned by varying the resistance. This example of nonequilibrium superconductivity differs from previous[1] ones in that the external drive acts directly on the pairs rather than directly on the quasiparticles. The absence of an applied voltage in the steady state distinguishes this effect from the Fiske and Shapiro steps[6] in the dc voltage.

The system considered is depicted schematically in Fig. 1. An external source of frequency Ω irradiates the junction with coherent photons. The oxide region[6,7] functions as a resonance cavity of frequency $\omega_c = c' n\pi/l_x$, with transverse junction dimensions l_x, l_y, the oxide having a dielectric constant ϵ and a thickness l_z. Here $c'/c = [l_z/\epsilon(2\lambda + l_z)]^{1/2}$. The (similar) superconducting electrodes have a penetration depth λ and gap Δ and are at temperature $T \ll T_c$ so that the quasiparticle tunneling current is negligible. For $\Omega < 2\Delta/\hbar$, the external radiation will not create quasiparticles.

The Hamiltonian of the junction can be written,[7-9] in terms of pseudospin operators and neglecting contributions from the bulk electrodes, as

$$H = \frac{1}{2}\frac{(2eS_z)^2}{C} - \frac{\hbar I_j}{4e}(S^- + S^+) + \hbar\omega_c a^\dagger a + i\hbar T(k,\omega)(a + a^\dagger)(S^- - S^+) + 2i\hbar T_{\rm ext} N^{1/2}\cos\Omega t(S^- - S^+). \tag{1}$$

The first and second terms describe the energy of the charged-junction capacitance $C = \epsilon l_x l_y/4\pi l_z$, and the pair-tunneling term leading to the Josephson current. The third and fourth terms represent the cavity photon energy and the absorption and/or emission of cavity photons by the Josephson current. The last term is the coupling of the Josephson current to an external coherent field of frequency Ω. We will assume that the external source has finite bandwidth, which is quite important as seen later. The coupling constant T is[7] $[K = (c'/c)k]$

$$T(k,\omega) = \frac{I_j}{(\hbar\omega_c C)^{1/2}}\frac{k_H \sin[(k_H - k)l_x]}{[k_H^2 - k^2]l_x}; \quad T_{\rm ext} = T(K,\Omega), \tag{2}$$

where $k_H = (2e/\hbar c)(2\lambda + l_z)H$ and H is an applied magnetic field that enables the current to couple to the standing-wave junction-cavity photons and selects one mode for $k_H \sim k$.

The pseudospin operators S^-, S^+, S_z are defined in terms of those of the left and right electrodes.[9] S^- (S^+) transfers pairs from left (right) to right (left), and S_z counts the number of excess pairs on the left. For the usual dc Josephson effect, $\langle S_z\rangle = 0$ and there is no pair imbalance of dc voltage across the junction. The commutator of S^+ and S^- is neglected.[7]

The equations for the expectation values of coupled spin-photon system follow from (1) and (2). We make the simplest "mean-field" type of approximation, replacing operators by their factored averages. This yields

$$\langle \dot{S}^- \rangle \equiv \frac{d}{dt} e^{-i\theta(t)}; \ \dot{\theta} = \frac{2eV(t)}{\hbar} = \frac{4e^2}{\hbar C}\langle S_z \rangle, \tag{3a}$$

$$\langle \dot{S}_z \rangle = \frac{\langle S_z \rangle}{RC} - \frac{I_j}{2e}\sin\theta - 2T\langle a + a^\dagger \rangle \cos\theta - 4T_{\rm ext}N^{1/2}\cos\Omega t \cos\theta, \tag{3b}$$

$$\langle \dot{a} \rangle = -\kappa\langle a \rangle - i\omega_c\langle a \rangle - 2iT\sin\theta, \tag{3c}$$

where decay terms have been added to represent resistance losses [in (3b)] and photon leakage (3c) from the cavity, of quality factory Q, at the rate $\kappa = \omega_c/2Q$.

We assume that a pair charge imbalance with a particular sign $\langle S_z \rangle = \mathrm{sgn}\dot{\theta} > 0$, for example, has already been set up and, as shown below, that it can be maintained by the radiation power. We substitute $\langle a \rangle = \langle \tilde{a} \rangle e^{-i\theta}$ into (3) and take $\theta(t) = \theta(0) + \overline{\omega}t$ $(t > 0)$. Keeping in mind the finite bandwidth of incident radiation, we time average Eq. (3). Further assuming that the photon mode is a fast mode,[5] i.e., $RC \gg \kappa^{-1}$, we put $\langle \dot{\tilde{a}} \rangle = 0$, which yields $\langle \tilde{a} \rangle_{\rm dc} = T/[\kappa + i(\omega_c - \overline{\omega})]$, where the subscript dc specifies dc quantities. Following this, a physically transparent result for the rate of change of the pair charge imbalance is obtained, i.e.,

$$2e\langle \dot{S}_{z_{\rm dc}} \rangle = -4eT_{\rm ext}N^{1/2}\cos\theta(0) - (V_{\rm dc}/R) - 4e\kappa|\langle \tilde{a} \rangle_{\rm dc}|^2, \tag{4}$$

where the phase difference $\theta(0)$ between the applied radiation and the resultant current can thus be absorbed into the (adjustable) phase difference between the two superconducting electrodes. N is the number of external photons *inside* the junctions. The escape of photons, otherwise fed back and forth between cavity and current, contributes to a pair imbalance decay $2e(2\kappa\langle a^\dagger a \rangle_{\rm dc}) \sim 4e\kappa \times |\langle \tilde{a} \rangle_{\rm dc}|^2$.

When drive and dissipation balance, the scaled steady-state voltage $f \equiv \overline{\omega}/\omega_c = 2eV_{\rm dc}/\hbar\omega_c$ is obtained from

$$f - \left(\frac{N}{N_c}\right)^{1/2} + \frac{2Q\alpha}{1 + 4Q^2(f-1)^2} = 0, \tag{5}$$

where we have set $\cos\theta(0) = -1$, as this can be shown to minimize the Ginzburg-Landau–like potential defined below. The dimensionless constants are

$$\alpha \equiv \frac{8e^2}{\hbar} R\left(\frac{T(k,\omega)}{\omega_c}\right)^2; \ N_c^{-1/2} = \frac{8e^2}{\hbar} R\left(\frac{T_{\rm ext}}{\omega_c}\right), \tag{6}$$

Equation (5) admits three real roots provided

$$Q^2\alpha > \tfrac{2}{3}\sqrt{3}, \tag{7a}$$

$$N_2 \geq N \geq N_1 > N_c, \tag{7b}$$

where $N_{1,2}$ are the roots of $dN/df = 0$. Equation (7a) is equivalent to requiring N to exceed a critical value. A plot, in the bistable regime of the roots $\overline{\omega}/\omega_c$ vs $(N/N_c)^{1/2}$, is given in Fig. 2. The hysteresis, as in other nonequilibrium systems,[3,4] is reminiscent of superheating and supercooling in equilibrium first-order transitions.

In the limit $Q^2\alpha = (2/3\sqrt{3})(1+\delta)$, $0 < \delta \ll 1$, we find $f_{1,2} = f_0 \pm \frac{1}{3}\delta^{1/2}/Q$ and $N_{1,2} = N_0^{1/2} \mp \frac{1}{9}\delta^{3/2}/Q$ where $f_0 \equiv 1 + \frac{1}{2}\sqrt{3}\,Q$, $(N_0/N_c)^{1/2} \equiv 1 + \sqrt{3}(1 + \frac{2}{3}\delta)/2Q$, and $\delta = (R/R_0 - 1)$, R_0 being defined implicitly above. For $\Omega \sim \omega_c$, the finite bandwidth can cover the small hysteresis region.[10] In general, for a given junction, the hysteresis size can be varied by changing R.

The relative stability of these three states can be studied by constructing a Ginzburg-Landau-like function $\Phi(f)$ whose minima are the multiple steady states and whose lowest minimum occurs at the most probable state.[2,5] The probability of

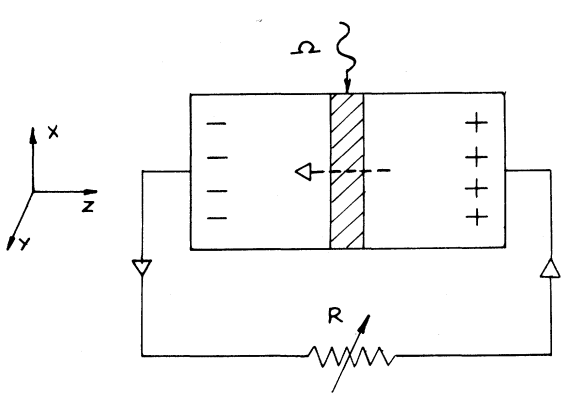

FIG. 1. Schematic diagram of the system, with the arrows denoting the direction of pair flow.

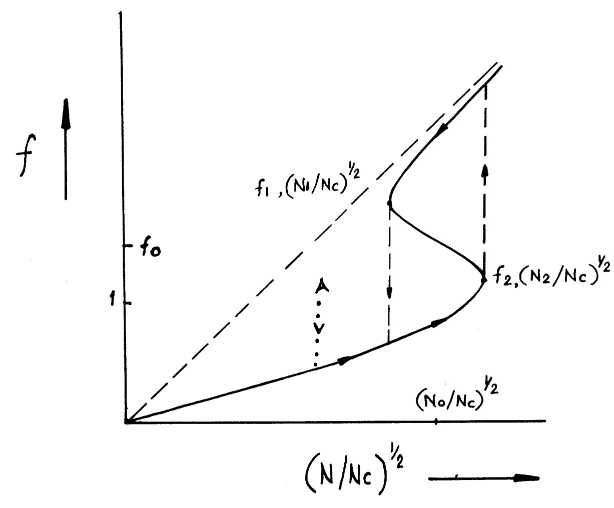

FIG. 2. Schematic plot of the scaled dc voltage $f = 2eV_{dc}/\hbar\omega_c$ vs the scaled (square root of) externally supplied photon number in the cavity $(N/N_c)^{1/2}$. The hysteresis region is bounded by the critical points $(f_{1,2}, N_{1,2}^{1/2})$.

a given f is $P(f) \propto \exp\{-2\tau_f \Phi(f)/RC\}$, where τ_f^{-1} is the strength of a delta-correlated random force in a stochastic equation for f, and the dimensionless function $\Phi(f)$ is just the integral of the left-hand side of (5) and can be related to the dissipation in the system.[11] The function Φ (Fig. 3) clearly shows the exchange of relative stabilities and hysteresis.

The potential is symmetric, $\Phi(f) = \Phi(-f)$ (with a cusp at $f = 0$), reflecting the basic symmetry of the circuit. The zero-voltage state at the cusp is unstable, and a spontaneous fluctuation may perhaps be sufficient to break the symmetry. The symmetry can also be externally broken if need be; e.g., as follows. Apply a dc voltage to the irradiated junction such that $2eV_{ext}/\hbar = \Omega = \omega_c$. This will result, through the Fiske step effect,[6] in a dc current in a direction fixed by V_{ext}, corresponding to a point A in Fig. 2. Now switch off V_{ext}, leaving the microwave power on. The voltage will fall (dotted line) till it reaches the steady-state curve.

The dc voltage variation of Fig. 2 also corresponds to a variation of the frequency $\overline{\omega} = 2eV/\hbar$ of an ac Josephson current component and of the emitted radiation. For a given circuit, the frequency can be intensity tuned and switched.

We now examine the feasibility of the bistability. Keeping in view that lossy cavities make the photon leakage dissipation significant and large resistances make the power requirements low for

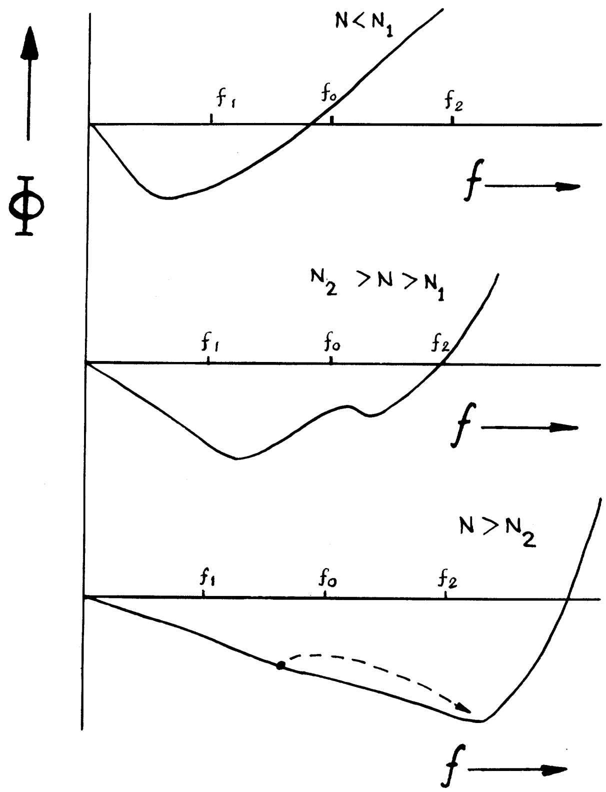

FIG. 3. Schematic plot of the Ginzburg-Landau-like potential $\Phi(f)$ vs the scaled voltage f. The dot denotes the initial steady state.

a given $V_{dc} \sim \hbar\omega_c/2e$, we take the typical values of the junction parameters as $l_x = l_y = 0.1$ cm ($\Omega = 10^{11}$ Hz), $l_z = 20$ Å, $\epsilon = 5$, $I_j = 30$ μA, $R = 100$ Ω, $Q = 5$, $\lambda = 4\times10^{-6}$ cm; we then find $N_c = 5\times10^4$ photons, and $\alpha = 0.02$. The external photon density in the cavity must be at least $N_c/(2\lambda + l_z)l_x l_y \times 10^{11}$ cm^{-3}. This corresponds to an available power of $N_c\hbar\Omega c'/l_x \sim 4\times10^{-9}$ W, and is less than the power required by resistance and cavity losses. The lifetimes κ^{-1} and RC justify the original fast-mode assumption for photons. For a 10% transmission from outside, this means an external power of at least 0.05 W/cm^2 is required. Voltages and currents are typically around 0.1 mV and 1 μA. The fractional pair imbalance established by the radiation is one pair in 10^9. The power requirement and the bistability condition of (7) are very sensitive to the junction dimensions and also depend on Q, R, and I_j. This could explain[12] why the predicted phenomena have not previously been seen, apart from that fact that here the junction must be "free running" with a self-consistently developed, rather than an applied,[6] voltage. In order for the bistability to be observable, the lifetimes of the states must be large. Estimating the photon fluctuation rate $\tau_c^{-1} \gtrsim \kappa \sim 10^{10}$ sec^{-1}, we find $\tau_f^{-1} \sim 0.1$ sec^{-1}. The scaled lifetime in the probability exponent is $\tau_f / RC \sim 10^6$, and the distribution is thus, indeed, sharply peaked.

We thank P. Chandrasekhar, P. A. Narayana, and V. S. S. Sastry for useful discussions.

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[10]For the case of hysteresis region larger than the bandwidth, one could consider two coherent sources with two frequencies, close to the higher and lower values at the transition, and with nonoverlapping bandwidths. Equation (6) would hold approximately for either source. After the intensity variation of one beam drives the system to the other frequency, the second beam takes over control and the first can be switched off.

[11]Cf. S. R. Shenoy, to be published.

[12]With systems parameters in the above range, one finds the neglected $I_j \sin\theta$ term yields corrections of order 1%. However, see J. T. Chen, R. J. Todd, and Y. W. Kim, Phys. Rev. B 5, 1843 (1972).

Theoretical Studies of a Light-Induced Change in Multiplet Satellites of 3*p*-Photoelectron Spectra of Transition-Metal Compounds

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(Received 31 December 1979)

It is shown that optical excitations of the *d* shell may induce changes in the multiplet structure of the 3*p*-photoelectron emission spectra of transition-metal compounds. The photoelectron spectra of Ni^{2+} and Cr^{3+} compounds in some excited states are calculated and compared with those in the ground states.

PACS numbers: 79.60.Eq

It is well known[1] that the initial *d*-electron state of a transition-metal compound is clearly reflected in multiplet satellites of photoelectron emission spectra (PES). In the present paper, we shall show that an optical excitation of the *d* shell, whose lifetime could be assumed extremely short (as little as 10^{-15} s), may induce a change in the multiplet structure of, mainly, 3*p* PES of transition-metal compounds. We should like to emphasize the point that observation of this change provides us with a unique method of detecting the excited state with a very short lifetime. As discussed later, a physically important point in this method is that the satellite lines associated with an excited state are still expected to be observed even if the state is extremely shortlived.

Let us first examine how short a change of the *d*-electron state could be and still be observed as a change in the multiplet structure. As well-known[2] shakeup satellites or multiplet satellites

