resonance produces no noteworthy structure in μ. We have found that the behavior of σ_T above $ka = 0.6$ is completely insensitive to a 10% variation in γ_0, and consequently, μT^3 is unchanged above 0.33°K. However, a smaller γ_0 produces a larger μT^3 at lower temperatures. We also tried $\gamma_2 = 3.92$ and found that over the range $0.3°K \leq T < 0.5°K$ μT^3 is flat like the experimental data, but its value is 10% larger than for $\gamma_2 = 2.22$. An experimental determination of μ at lower temperatures would be useful in fixing the parameters more precisely.

We are indebted to Professor T. M. Sanders for several discussions, and particularly for emphasizing the importance of the s-wave resonance, and suggesting the treatment of the $l \geq 3$ partial waves. We have also had beneficial discussions with Dr. B. Györffy and Dr. J. Lekner. David Saul has kindly assisted us with the numerical calculations.

*Research supported by National Science Foundation Grant No. NSF-GP 7765.

†Research supported in part by the Advanced Research Projects Agency under Contract No. SD-131.

‡On leave of absence from Magdalen College, Oxford, England. Research supported in part by the U. S. Air Force Office of Scientific Research Contract No. AF 328-67.

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FREQUENCY-PULLING EFFECTS IN JOSEPHSON RADIATION*

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Using techniques developed in the quantum theory of the laser, we show that the frequency of the radiation emitted from a superconducting tunnel junction may be pulled slightly from $2eV/\hbar$ and present a physical discussion and numerical estimate of the effect.

Recent experimental[1] activity has been directed toward measuring the ratio of the electronic charge to Planck's constant. The basis[2] for these measurements is the observation that when a pair tunnels between two superconductors maintained at a potential difference V, a photon is emitted having a frequency $\omega = 2eV/\hbar$. It is the purpose of this communication to indicate a slight shift in the frequency of the emitted radiation due to cavity pulling effects. Techniques developed in treating the theory of laser oscillators[3,4] will be applied to this problem.

In our model we consider the junction to be at absolute zero and to consist of two identical thin films separated by an oxide barrier. When pairs tunnel across the barrier they excite a single mode of the electromagnetic field having an eigenfrequency Ω and a bandwidth ν/Q. In order to maintain charge neutrality pairs must be removed from one side of the barrier and added to the other by means of an external circuit.

We treat the problem as an exercise in nonequilibrium statistical mechanics in analogy with the quantum theory of the laser. The radiation-superconductor system will be coupled to reservoirs which represent the effects of a finite cavity Q in the case of the field, while in the case of the superconductors the reservoirs represent the wires (of normal metal) which are necessary in order to maintain charge neutrality. After demonstrating the frequency-pulling effect, a simple physical argument will be given which leads to the same result. It is hoped that for the reader not interested in the details of the calculation, this latter discussion will provide a satisfactory physical explanation of the frequency pulling.

We now outline a calculation[5] of the pulling effect. The state of the superconductors will be taken as that corresponding to strongly coupled BCS states. In this limit the electronic kinetic energy is neglected in comparison with the interaction energy between electrons. Furthermore, we consider only the electron pairs within a narrow region ($\epsilon_F \pm \hbar\omega_D$) of the Fermi surface. We are interested in states of the coupled-superconductor system in which there are k excess pairs on the left-hand side. As a result of the voltage established by an external battery, the mean value of $\langle k\rangle$ is equal to the difference between the mean number on the left, N_L, and the right, N_R. The state of the superconducting junction will then be represented by

$$|\mathrm{s}\rangle = \sum_k a_k |k\rangle, \tag{1}$$

where a_k is the probability amplitude for having k excess pairs. We expect the state of the radiation field to be very close to that of a pure coherent state,

$$|\alpha\rangle = \sum_n [\alpha^n (n!)^{-\frac{1}{2}}] |n\rangle e^{-\frac{1}{2}|\alpha|^2}, \tag{2}$$

where α is a complex c number and $|n\rangle$ is an eigenstate of the photon-number operator. Throughout this discussion we make the Ansatz that the superconductor-radiation density matrix is of the factorized form

$$\rho(t) = \sigma(t) \otimes r(t), \tag{3}$$

where σ is the density operator for the superconductor system and $r = |\alpha\rangle\langle\alpha|$. While the factorization Ansatz is quite adequate for the purposes of demonstrating a frequency shift, it will be removed in future publication.

The tunneling of a pair from left to right is expressed in terms of the spin operators[6] as $S_L S_R^\dagger$. The inverse process (from right to left) is given by $S_R S_L^\dagger$. The operators S_L and S_R are sums over the individual pair operators

$$S_{L,R} = \sum_p s_{L,R}^{\;p},$$

where $s_{L,R}^{\;p}$ is the spin-flip operator which effectively removes a pair of momentum p from L or R. The Hamiltonian for the superconductor-radiation system in which the left-hand superconductor is maintained at a voltage[7] V while the right-hand side is kept at zero voltage is

$$H = H_0 + \hbar\Omega a^\dagger a + [2eV] S_{Lz} + g(S^\dagger a + S a^\dagger), \tag{4}$$

where H_0 is the usual Hamiltonian for the right- and left-hand superconductors, Ω is the cavity eigenfrequency, $a^\dagger$ and a are the creation and annihilation operators for the field, S_{Lz} is the z component of the spin operator for the left-hand side, g is the coupling constant between the electron pairs and the radiation field, and the operator S equals $S_L S_R^\dagger$. The total time evolution of the density matrix is given by

$$\dot\rho = -(i/\hbar)[H,\rho] + (\partial\rho/\partial t)_{\text{cavity loss}} + (\partial\rho/\partial t)_{\text{wire}}. \tag{5}$$

The cavity-loss term due to the leaking of radiation out of the cavity is given by[8]

$$(\partial\rho/\partial t)_{\text{cavity loss}} = -\tfrac{1}{2}(\nu/Q)[a^\dagger a\rho + \rho a^\dagger a] + (\nu/Q)[a\rho a^\dagger], \tag{6}$$

where ν/Q is the passive cavity bandwidth. The effect of the wire in removing electrons from the right-hand superconductor and placing them in the left-hand superconductor is found[9] to be

$$(\partial\rho/\partial t)_{\text{wire}} = -\tfrac{1}{2}A[SS^\dagger\rho + \rho SS^\dagger] + A[S^\dagger\rho S], \tag{7}$$

where A is a constant which may be expressed in terms of the dc current and other known parameters.

Upon taking the superconductor states in Eq. (5) we obtain an equation of motion for the radiation density matrix in an interaction picture:

$$\dot r = -i\Omega[a^\dagger a, r] - i(g/\hbar)\{\langle S^\dagger\rangle[a,r] + \langle S\rangle[a^\dagger, r]\} - \tfrac{1}{2}(\nu/Q)[a^\dagger a r + r a^\dagger a] + (\nu/Q)[a r a^\dagger], \tag{8}$$

where

$$\langle S\rangle = \mathrm{Tr}(S\sigma).$$

Likewise an equation of motion for the coupled-superconductor density matrix is obtained from (5) by taking the trace over the radiation:

$$\dot\sigma = -i(2eV/\hbar)[S_{Lz},\sigma] - i(g/\hbar)\{\alpha[S^\dagger,\sigma] + \alpha^*[S,\sigma]\} - \tfrac{1}{2}A[SS^\dagger\sigma + \sigma SS^\dagger] + A[S^\dagger\sigma S], \tag{9}$$

where we have used the fact that

$$\alpha = \mathrm{Tr}(ar).$$

We may obtain an expression for $\langle S\rangle$ as it appears in Eq. (8) by multiplying Eq. (9) by S and taking the trace:

$$d\langle S\rangle/dt=-i(2eV/\hbar)\langle S\rangle-i(g/\hbar)\alpha(t)\Delta+\langle\dot{S}\rangle_{\text{wire}}, \qquad (10)$$

where

$$\Delta=\langle S_{Lz}S_R^{\dagger}S_R\rangle-\langle S_{Rz}S_L^{\dagger}S_L\rangle.$$

Using Eq. (7) it is found that to a good approximation the last term in (10) is given by

$$\langle\dot{S}\rangle_{\text{wire}}=-\Gamma\langle S\rangle, \qquad (11)$$

where

$$\Gamma=(I/2e)(N_L^{-1}-N_R^{-1}). \qquad (12)$$

The expression for Γ is obtained by relating the coefficient A as it appears in (7) to the dc current. We separate out the high-frequency time dependence of $\langle S(t)\rangle$ by writing

$$\langle S(t)\rangle=e^{-i\nu t}\langle S(t)\rangle',$$

where the frequency ν is the same as that of the radiation. That is, the frequency of $\langle S(t)\rangle$ and the frequency of the emitted radiation are locked together just as the frequency of the atomic polarization and that of the electromagnetic radiation are locked together in the laser problem. Noting that $\langle S(t)\rangle$ is oscillating at a frequency ν and considering the steady-state situation in which $d\langle S(t)\rangle'/dt=0$, Eqs. (10)-(12) lead to the following determination of $\langle S\rangle$:

$$\langle S\rangle=-ig\alpha\Delta D(\omega-\nu), \qquad (13)$$

where

$$D(\omega-\nu)=[i(\omega-\nu)+\Gamma]^{-1}.$$

Substituting Eq. (13) into Eq. (8) we obtain the following equation of motion for the Josephson-radiation density matrix:

$$\dot{r}=-i\Omega[a^{\dagger}a,r]+(g/\hbar)^2\Delta\times\{\alpha^*D^*(\omega-\nu)[a,r]-\alpha D(\omega-\nu)[a^{\dagger},r]\}-\tfrac{1}{2}(\nu/Q)[a^{\dagger}ar+ra^{\dagger}a]+(\nu/Q)[ara^{\dagger}]. \qquad (14)$$

From Eq. (14) it may be shown[10] that in order to achieve oscillation we must have

$$\Gamma(g/\hbar)^2\Delta/[(\omega-\nu)^2+\Gamma^2]>\tfrac{1}{2}\nu/Q, \qquad (15)$$

and the frequency ν is given by

$$\nu=[\tfrac{1}{2}(\nu/Q)\omega+\Gamma\Omega][\tfrac{1}{2}\nu/Q+\Gamma]^{-1}. \qquad (16)$$

In the present case ν/Q greatly exceeds the rate Γ, and Eq. (16) becomes

$$\nu=\omega-\frac{\Gamma(\omega-\Omega)}{\nu/2Q}. \qquad (17)$$

Taking Γ from (12) this may be written as

$$\nu=\omega-(\nu/2Q)^{-1}(\omega-\Omega)(I/2e)(N_L^{-1}-N_R^{-1}).$$

The physical content of Eq. (18) may be understood in the following way. Consider an atom (atomic frequency ω) to be placed for a time T in a cavity characterized by an eigenfrequency Ω and a quality factor Q. The radiation emanating from the cavity-atom system is then at a frequency

$$\nu=[\tfrac{1}{2}(\nu/Q)\omega+(1/T)\Omega][\tfrac{1}{2}\nu/Q+1/T]^{-1}, \qquad (19)$$

which for $\nu/Q\gg T^{-1}$ becomes

$$\nu=\omega-(\nu/2Q)^{-1}(\omega-\Omega)(1/T). \qquad (20)$$

In the present problem electron pairs remain in the left superconductor (like an atom in its upper state) on the average for a time $T_L=D/v_L$, where v_L is the mean drift velocity of pairs in the left side and D is a characteristic dimension of the superconductor. In analogy to Eq. (20) we would then expect a shift of an amount

$$\nu=\omega-(\nu/2Q)^{-1}(\omega-\Omega)(1/T_L). \qquad (21a)$$

However, the pairs then pass into the right-hand superconductor (now act like an atom in its lower state) and tend to pull the frequency in the opposite direction. The pairs "remain" in the right side on the average for a time T_R given by

$$T_R=D/v_R,$$

where v_R is not the same as v_L since there are fewer electrons on the right-hand side carrying the same current; therefore we expect a shift in the opposite sense during this time T_R,

$$\nu=\omega+(\nu/2Q)^{-1}(\omega-\Omega)(1/T_R), \qquad (21b)$$

and the total frequency shift from (21a) and (21b) is

$$\nu=\omega-(\nu/2Q)^{-1}(\omega-\Omega)[T_L^{-1}-T_R^{-1}]. \qquad (22)$$

Now T_L^{-1} may be expressed in terms of the current and the number of carriers on the left-hand side as

$$T_L^{-1}=v_R/D=(I/2e)N_L^{-1}$$

with a similar expression for T_R^{-1}. The frequency (22) may then be written as

$$\nu = \omega - (\nu/2Q)^{-1}(\omega - \Omega)(I/2e)(N_L^{-1} - N_R^{-1}), \quad (23)$$

which is the result (18).

Finally, let us estimate the magnitude of the frequency pulling,

$$\nu - \omega = (I/2e)[(N_L - N_R)/N_L N_R](\omega - \Omega)(\nu/2Q)^{-1}.$$

For a current of 10^{-2} A, $I/2e$ is of the order of 10^{16}-10^{17}. The difference in the number of pairs $N_L - N_R$ may be estimated by knowing the capacitance of the junction ($C \approx 1\ \mu$F) and the voltage V ($\approx 10^{-5}$ V), to be of the order of 10^8 pairs. The magnitude of N_L or N_R for a volume 10^{-6} cm^3 is of the order of 10^{12}. From these numbers we find

$$\nu - \omega \approx (1\text{-}10) \times (\omega - \Omega)(\nu/2Q)^{-1};$$

so that if $(\omega - \Omega)$ is around $\nu/2Q$, the pulling would be between 1 and 10 Hz. As Eq. (18) seems to be very sensitive to the number of pairs on the right- and left-hand sides, it would be interesting to measure the frequency pulling for superconductive films of different thicknesses.

We would like to thank Professor L. W. Gruenberg and Professor P. C. Martin for helpful discussions, and Professor M. J. Stephen for collaboration on certain portions of this work.

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*Work supported by the Advanced Research Projects Agency under Contract No. SD-90.

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[7]The voltage is related to the number of excess pairs. This however is not a fixed number [See Eq. (10)], and one expects fluctuations in the voltage. M. J. Stephen has shown (private communication and to be published) that these voltage fluctuations lead to a linewidth in agreement with the recent experiments reported by W. H. Parker [in Fluctuations in Superconductors, edited by W. S. Goree and F. Chilton (Stanford Research Institute, Stanford, California, 1968)].

[8]The effects of a finite cavity Q are discussed in Ref. 4. There we were working in the number representation; however, it is clear that Eq. (6) of the present paper follows directly from Eq. (84) of Ref. 4.

[9]Equation (7) was derived by considering the wires connecting the superconductors as particle reservoirs of unpaired electrons. The coefficient A may be related to the dc current by noting that I equals $2e\langle \dot{S}_{Lz}\rangle$. The detailed development of Eq. (2) will be given in future publication.

[10]Calculations of this type are to be found in M. Scully, thesis, Yale University, 1966 (unpublished), Appendix V.

VARIATION OF CHARGE-CARRIER RELAXATION TIME IN BISMUTH WITH MAGNETIC FIELD*

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The attenuation of Alfvén waves in bismuth can be analyzed to show the detailed variation of the relaxation time with magnetic field. For the example presented, which has the magnetic field along the threefold axis, variations between 2.5×10^{-10} and 6.0×10^{-10} sec are found around an average value for τ of 4.3×10^{-10} sec.

The transmission of Alfvén waves in bismuth has been used in several laboratories to determine the effective masses and number of charge carriers.[1,2] In addition, the attenuation coefficient of Alfvén waves is directly related to the carrier relaxation times τ (which are, in general, tensor quantities). In this Letter we show that in addition to average values for τ, we can obtain good numerical values showing the variation in τ with magnetic field using data on the attenuation of Alfvén waves. This variation in τ with magnetic field is, of course, responsible for the de Haas-Shubnikov effect.

Numerical values of τ in bismuth from Alfvén-wave experiments have been obtained by McLachlan[2] and Marsten and Kao.[3] A theoretical study has been performed by Brownell and Hygh.[4] In each of these studies it is clear that the relaxation times are not isotropic. In each of the previous studies only the average value of the relax-