[4]J. Rosenblatt, J. J. Hauser, and J. P. Burger, to be published.
[5]G. Deutscher and F. Meunier, Phys. Rev. Letters 22, 395 (1969).
[6]J. J. Hauser, H. C. Theuerer, and N. R. Werthamer, Phys. Rev. 142, 118 (1966).

EFFECT OF FLUCTUATIONS IN THE SUPERCONDUCTING ORDER PARAMETER ON THE TUNNELING DENSITY OF STATES

Roger W. Cohen, B. Abeles, and C. R. Fuselier
RCA Laboratories, Princeton, New Jersey 08540
(Received 28 April 1969)

Fluctuations in the superconducting order parameter above the transition temperature result in structure in the tunneling density of states. From the shape of this structure a value is derived for the decay rate of the fluctuations.

Fluctuations in the superconducting order parameter above the transition temperature are predicted to affect a variety of experimental quantities, e.g., electrical conductivity,[1] specific heat,[2] and diamagnetic susceptibility.[3] Fluctuation effects have so far been reported only in the electrical conductivity.[4,5] We have observed the effect of fluctuations on the electronic density of states (EDS), derived from tunneling measurements.

The measurements were carried out on M_1-I-M_2 junctions in which M_1 is a metal film in the fluctuation state above the transition temperature T_{c1}, M_2 is a film in the superconducting state, and I is a thin insulating layer. We find that the EDS in the fluctuation regime of M_1 exhibits a depression from the normal-state value over a small energy range E_c centered about the Fermi level. As the temperature is raised the EDS approaches that of the normal metal. As the temperature is lowered the depression in the EDS deepens and for $T < T_{c1}$ develops into an energy gap in the quasiparticle spectrum. From the observed structure in the EDS the decay rate of fluctuations in the Cooper-pair density is deduced.

Metal M_1 was granular Al, a material which has been used previously by several workers to study fluctuation phenomena.[5-7] The granular Al was deposited onto glass slides by using two different techniques: evaporation in an oxygen atmosphere[8] or cosputtering[9] Al and SiO_2. The insulating layer I was Al_xO_y which was grown by exposing the granular Al to laboratory air for several minutes. The film M_2 was In, Pb, or the alloy[10] $Pb_{0.7}Bi_{0.3}$, approximately 3000 Å thick, evaporated on top of the insulating layer. The parameters of the three junctions reported in this Letter are given in Table I. Here, R_j is the junction resistance and $\Delta_j(0)$ is the energy gap of metal M_j at $T = 0°K$. The quantity ρ_0 is the normal resistivity and d is the thickness of the granular Al film. The length $(\xi_0 l)^{1/2}$ (ξ_0 is the Pippard coherence length and l is the effective mean free path[11]) appears in the expression for the temperature-dependent Landau-Ginzburg coherence length[12] $\xi(T) = 0.85(\xi_0 l)^{1/2}\epsilon^{-1/2}(T)$, where $\epsilon(T) = (T - T_{c1})/T_{c1}$.

The first derivative dI/dV and the second derivative d^2I/dV^2 of the junction I-V characteristics were determined by the conventional technique of superposing a small audio-frequency voltage v on the dc bias V and measuring the resulting ac cur-

Table I. Properties of junctions.

Junction	R_j (Ω)	M_2	Δ_2 (mV)	M_1	T_{c1} (°K)	$2\Delta_1(0)/k_BT_{c1}$	ρ_0 (10^{-6} Ω cm)	d (Å)	$(\xi_0 l)^{1/2}$ (Å) [a]
I	990	$Pb_{0.7}Bi_{0.3}$	1.794	Sput.	2.59	3.45	840	2460	120
II	14.4	$Pb_{0.7}Bi_{0.3}$	1.766	Evap.	2.36	3.54	72	550	420
III	14.7	Pb	1.280	Evap.	2.12	3.47	38	800	580

[a]The Pippard coherence lengths ξ_0 were computed by multiplying the value $\xi_0 = 16\,000$ Å for ordinary Al ($T_c = 1.18°K$) by ratio of the energy gap $\Delta_1(0)$ of ordinary Al to that of the granular film. The effective mean free paths l were calculated from the formula $\rho_0 l = 1.6 \times 10^{-11}$ Ω cm^2 [J. L. Olson, Electron Transport in Metals (Interscience Publishers, Inc., New York, 1962), p. 84].

rent by means of a lock-in detector. The modulation voltage v must be smaller than the width of any structure to be resolved. At the highest temperatures, this condition could not be fulfilled because of signal-to-noise limitations. In these cases, a correction procedure was applied to account for the distortions in the derivatives.[13] Another source of distortion arises when the series resistance due to M_1 is not negligible with respect to R_j. To minimize the high strip resistance of the granular aluminum films, a modification of the usual cross-strip junction geometry was employed (see insert of Fig. 1). The special feature of this geometry is the Au current contact which was evaporated directly on M_1 as close as possible to the tunneling area.

Our primary experimental observation was that samples of granular Al, which exhibited fluctuation effects in the resistive transitions, displayed in the same temperature range structure in the dI/dV and d^2I/dV^2 tunneling characteristics. On the other hand, as ρ_0 of the granular Al was decreased, the resistive transitions sharpened and the magnitude of the structure decreased rapidly. An example of the structure in the d^2I/dV^2 of junction I is shown in Fig. 1 by the upper five curves measured at various temperatures in the fluctuation regime $T > T_{c1}$. Above $T/T_{c1} \approx 1.6$, the d^2I/dV^2 curve was characteristic of a normal-metal–superconductor tunneling junction. As the temperature is lowered, structure appears around the bias voltage $V = \Delta_2$. This structure becomes more pronounced with decreasing temperature. For $T/T_{c1} \leq 0.99$, the d^2I/dV^2 and dI/dV curves were characteristic of a superconductor-superconductor tunnel junction in which the granular Al had a well-defined energy gap $\Delta_1(T)$, and the values[14] of $\Delta_1(T)$ could be fitted by the reduced BCS temperature-dependent gap function[15] $\Delta_1(T/T_{c1})/\Delta_1(0)$. The values of T_{c1} and $2\Delta_1(0)/k_B T_{c1}$ determined in this manner are given in Table I.

The resistive transitions of the Al samples had the following properties: Al film I exhibited a transition which was characteristic of fluctuations in the three-dimensional regime[6] with $\xi(T) \ll d$. Al films II and III displayed transitions characteristic of fluctuations in the two-dimensional regime[4,5] with $\xi(T) \gg d$. The transition temperatures determined from the resistive transitions agreed with those determined from the tunneling measurements.

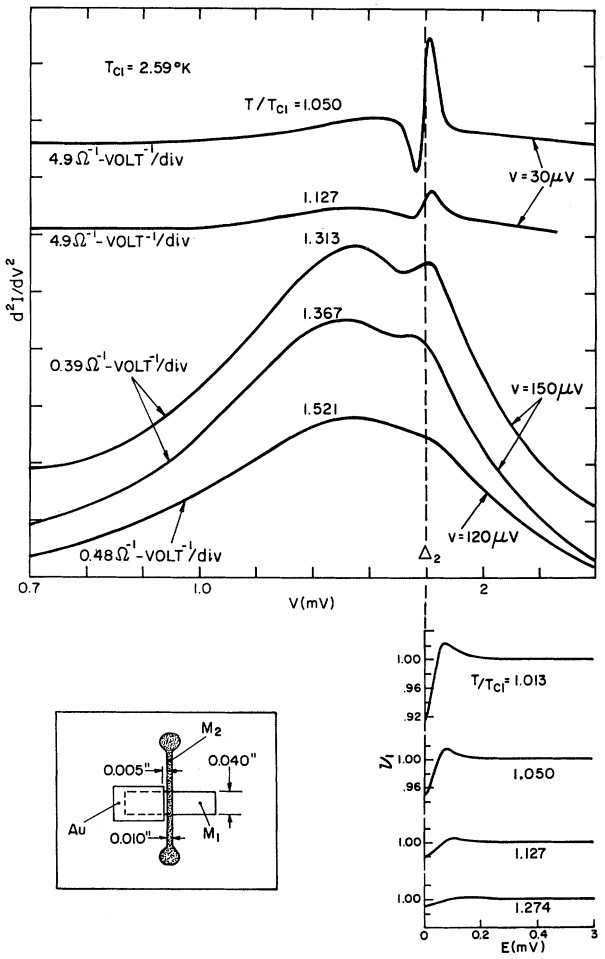

FIG. 1. The upper five curves are the second derivatives d^2I/dV^2 for Al-Al_xO_y-$Pb_{0.7}Bi_{0.3}$ junction I at various temperatures above T_c. The curves are displaced vertically, and the reduced temperatures, the vertical scales, and the ac modulation voltage v are indicated. The bias voltage $V = \Delta_2$ (energy gap of M_2) is indicated by the vertical dashed line. The lower four curves are the normalized electronic density of states of the aluminum $\nu_1(E)$ obtained from the expression for the tunneling current Eq. (2). The reduced temperatures are indicated. In the insert is shown schematically the junction M_1-I-M_2 with a Au current contact. All indicated dimensions are in inches.

To determine the EDS of the granular Al it is necessary to invert the measured dI/dV and d^2I/dV^2 characteristics using the formula for the tunneling current[16]:

$$I(V,T) = R_j^{-1} \int_{-\infty}^{+\infty} \nu_2(E,T)\nu_1(E-V,T)[f(E-V)-f(E)]\,dE. \quad (1)$$

Here E is the quasiparticle energy and $f(E)=[\exp(E/k_BT)+1]^{-1}$. The quantity $\nu_j(E,T)$ is the normalized EDS of metal M_j such that $\nu_j(E,T)=1$ when M_j is normal. The tunneling current may be regarded as being made up of two contributions $I(V,T)=I_0(V,T)+\delta I(V,T)$, where I_0 is the current in the absence of fluctuations $\nu_1=1$ and δI is the current due to the perturbation $\delta\nu_1(E,T)=\nu_1(E,T)-1$ in the EDS of M_1. The derivatives of $\delta I(V,T)$ with respect to V are

$$\frac{d^{1,2}}{dV^{1,2}}\delta I(V,T)=R_j^{-1}\frac{d^{1,2}}{dV^{1,2}}\int_{-\infty}^{+\infty}\nu_2(E,T)\delta\nu_1(E-V,T)[f(E-V)-f(E)]\,dE. \tag{2}$$

To compute $\delta\nu_1(E,T)$ from Eq. (2) we require the EDS $\nu_2(E,T)$ of M_2, the values of R_j, and $d^{1,2}\delta I/dV^{1,2}=(d^{1,2}I/dV^{1,2})-(d^{1,2}I_0/dV^{1,2})$. The values of R_j were measured at $V\gg(\Delta_1+\Delta_2)$. To determine $d^{1,2}I_0(V)/dV^{1,2}$, measurements were made on junctions in which the Al was sufficiently clean so that no fluctuation effects were detectable. For $\nu_2(E,T)$ we chose the BCS density of states[15] $\nu_2(E,T)=E[E^2-\Delta_2^2(T)]^{-1/2}$, where $\Delta_2(T)$ was determined from the tunneling measurements. The computed EDS for junction I at four temperatures above T_{c1} are given in the lower part of Fig. 1. The EDS $\nu_1(E,T)$ exhibits a depression $|\delta\nu_1(0,T)|$ at the Fermi level $E=0$ and rises to the normal-state value $\nu_1=1$ at the crossover point $E_c(T)$. For $E>E_c(T)$, $\nu_1(E,T)$ goes through a maximum and approaches asymptotically the value $\nu_1=1$. We use the crossover point $E_c(T)$ and the depression $|\delta\nu_1(0,T)|$ to characterize the shape of the EDS curves. In Fig. 2 are plotted the values of the quantities $E_c/\Delta_1(0)$ and $|\delta\nu_1(0,T)|$ for the three junctions.

We propose that the observed structure in the derivative and corresponding structure in the EDS of the granular Al for $T>T_{c1}$ arise from fluctuations in the superconducting order parameter. A theoretical treatment using the microscopic theory is in preparation.[17] For the purposes of the present Letter we choose a simple semiphenomenological approach. For the EDS we assume the form[18]

$$\nu_1(E)=1+\tfrac{1}{2}\Delta_1^2[E^2-\Gamma^2(T)][E^2+\Gamma^2(T)]^{-2}, \tag{3}$$

in which $\Gamma(T)/h$ is the decay rate of the Cooper-pair density. Equation (3) was derived[18] for the case $T<T_{c1}$ and the condition

$$\Delta_1^2/\Gamma^2(T)\ll 1. \tag{4}$$

We apply Eq. (3) to temperatures $T>T_{c1}$ by replacing Δ_1^2 by the thermodynamic mean-square value of the gap parameter $\langle|\Delta_1(T)|^2\rangle$ and setting $\Gamma(T)/h$ equal to the decay rate of zero-wave-number fluctuations[19]:

$$\Gamma(T)=(8/\pi)k_BT_{c1}\epsilon(T). \tag{5}$$

We compute the value of $\langle|\Delta_1^2|\rangle$ using the Landau-Ginzburg free-energy function in the Boltzmann factor[20] to weight fluctuations of wave numbers less than[21] $k_{max}=(\xi_0 l)^{-1/2}$, the maximum wave number allowed in the Landau-Ginzburg theory. In the classical fluctuation regime the quartic term in the Landau-Ginzburg expansion can be neglected, and one readily obtains

$$\langle|\Delta_1^2(T)|^2\rangle=k_BT_c[\,2\pi N_0\xi^2(T)d\epsilon(T)]^{-1}\ln[\xi(T)/(\xi_0 l)^{1/2}]\ \text{for}\ d\ll\xi(T), \tag{6a}$$

$$=k_BT_c[\,2\pi^2N_0\xi^3(T)\epsilon(T)]^{-1}\{\xi(T)/(\xi_0 l)^{1/2}-\tan^{-1}[\xi(T)/(\xi_0 l)^{1/2}]\}\ \text{for}\ d\gg\xi(T). \tag{6b}$$

The form of the EDS function Eq. (3) is similar to that obtained from tunneling measurements. According to Eq. (3), the crossover point $E_c(T)$ for which $\nu_1(E)=1$ is $\Gamma(T)$, and the value of the depression of the EDS at the Fermi level is $|\delta\nu_1(0,T)|=\frac{1}{2}\langle|\Delta_1^2(T)|\rangle/\Gamma^2(T)$. The theoretical curves for $\Gamma(T)/\Delta_1(0)$ and $|\delta\nu_1(0,T)|$ given in Figs. 2(a) and 2(b) were computed from Eqs. (3), (5), and (6) using the parameters given in Table I and the value of the normal[22] EDS for Al, $N_0=1.09\times10^{34}\ \text{erg}^{-1}\ \text{cm}^{-3}$. For sample I we used $\langle|\Delta_1(T)|^2\rangle$ appropriate to the case of fluctuations in the three-dimensional regime given by Eq. (6b), while for samples II and III we employed Eq. (6a) for the two-dimensional regime. This choice was consistent with the character of the observed resistive transitions. As can be seen in Fig. 2, the quan-

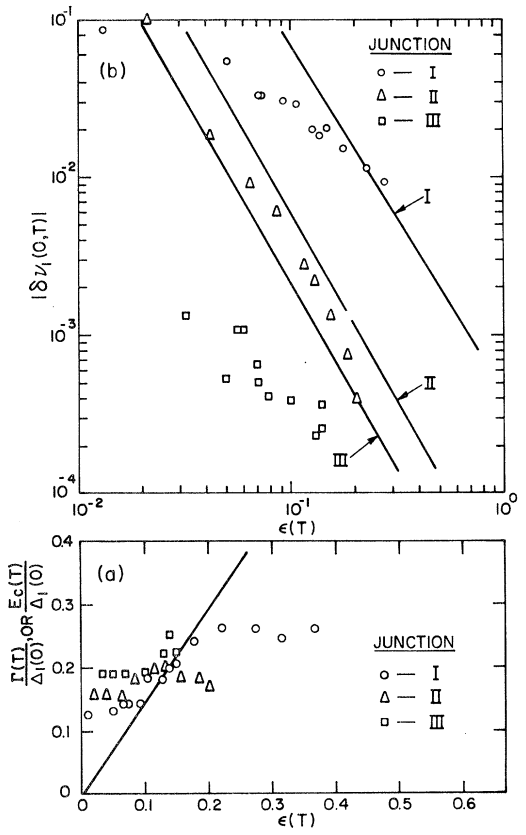

FIG. 2. The reduced crossover point energy $E_c/\Delta_1(0)$ [and the reduced decay rate $\Gamma(T)/\Delta_1(0)$] and the depression $|\delta\nu_1(0,T)|$ in the EDS at the Fermi energy versus ϵ. The points were determined from the tunneling measurements on junctions I, II, and III; the curves were computed from Eqs. (3), (5), and (6) using the parameters given in Table I.

tities $\Gamma(T)$ and E_c coincide only over a limited range of $\epsilon(T)$. For small values of $\epsilon(T) \lesssim 0.1$, condition (4) is not met, and we are not able to identify the crossover point E_c with $\Gamma(T)$. For large values of ϵ, the values of E_c appear to saturate, whereas $\Gamma(T)$ increases linearly with $\epsilon(T)$. The source of this discrepancy is not known. The calculated values of $|\delta\nu_1(0,T)|$ for samples I and II (M_2 was $Pb_{0.7}Bi_{0.3}$) are about a factor of 2 larger than the experimental values, and for sample III (M_2 was Pb) this factor is about one order of magnitude. Some of this discrepancy is believed to be due to the assumption of the BCS function for the EDS of M_2. The actual EDS's of Pb and $Pb_{0.7}Bi_{0.3}$ contain strong coupling effects and, in the case of Pb, there is the further complication of multiple gaps.[10] Overall, the qualitative agreement between theory and experiment indicates that fluctuations account for the observed structure in the tunneling characteristics.

We thank Dr. J. J. Hanak for supplying us with the sputtered Al films and Dr. G. D. Cody and Professor E. Abrahams for helpful discussions.

[1]L. G. Aslamazov and A. I. Larkin, Phys. Letters 26A, 238 (1968).

[2]R. A. Ferrell, in Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968, edited by J. F. Allen, D. M. Findlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1968), p. 802.

[3]H. Schmidt, Z. Physik 216, 336 (1968).

[4]R. E. Glover, Phys. Letters 25A, 544 (1967).

[5]M. Strongin, O. F. Kammerer, J. Crow, R. S. Thompson, and H. L. Fine, Phys. Rev. Letters 20, 922 (1968).

[6]J. I. Gittleman and Roger W. Cohen, to be published.

[7]R. V. D'Aiello and S. J. Freedman, Phys. Rev. Letters 22, 515 (1969); S. Marčelja, W. E. Masker, and R. D. Parks, Phys. Rev. Letters 22, 124 (1969).

[8]Roger W. Cohen and B. Abeles, Phys. Rev. 168, 444 (1968).

[9]J. J. Hanak, J. I. Gittleman, J. P. Pellicane, and S. Bozowski, to be published.

[10]The Pb-Bi alloys [J. G. Adler and S. C. Ng, Can. J. Phys. 43, 594 (1965)] were used because they exhibit one sharp gap whereas Pb has multiple energy gaps; e.g., see J. I. Rochlin and D. H. Douglass, Phys. Rev. Letters 16, 359 (1966).

[11]The concept of an effective mean free path in a granular superconductor enables one to apply the equations of a dirty type-II superconductor; see B. Abeles, R. W. Cohen, and W. C. Stowell, Phys. Rev. Letters 18, 902 (1967).

[12]P. G. de Gennes, Superconductivity of Metals and Alloys (W. A. Benjamin Inc., New York, 1966), p. 255.

[13]The measured apparent second derivative is given by $(d^2I/dV^2)_{\text{meas}} = (8/v^2)I_2(vD)I(V)$, where I_2 is the series expansion for the modified Bessel function of order 2 and D is the differential operator d/dV. The actual d^2I/dV^2 can be found from $(d^2I/dV^2)_{\text{meas}}$ by Fourier analyzing the above equation.

[14]To obtain the energy gaps we employed the criterion of Douglass and Meservey [D. H. Douglass, Jr., and R. Meservey, Phys. Rev. 135, A19 (1964)].

[15]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 128, 1175 (1957).

[16]I. Giaever, Phys. Rev. Letters 5, 147, 464 (1960).

[17]E. Abrahams (private communication).

[18]See de Gennes, Ref. 12, p. 266.

[19]E. Abrahams and T. Tsuneto, Phys. Rev. 152, 416 (1966). We neglect the wave number dependence of $\Gamma(T)$. This dependence is included in an approximate way, by the use of the $k_{\max}$ cutoff in the calculation of $\langle|\Delta(T)|^2\rangle$.

[20]E. Abrahams and J. W. F. Woo, Phys. Letters 27A, 117 (1968).

[21]S. Marčelja, Phys. Letters 28A, 180 (1968).

[22]Computed from the specific heat given in the Handbook of Chemistry and Physics, edited by C. D. Hodgman (Chemical Rubber Publishing Company, Cleveland, Ohio, 1965), 46th ed., p. E-67.