Unbinding of Charge-Anticharge Pairs in Two-Dimensional Arrays of Small Tunnel Junctions

J. E. Mooij, B. J. van Wees, L. J. Geerligs, M. Peters, R. Fazio,[a] and G. Schön

Department of Applied Physics, Delft University of Technology, Delft, The Netherlands

(Received 28 February 1990)

We describe the behavior of charges in two-dimensional arrays of normal-metal tunnel junctions with very small capacitance. A Kosterlitz-Thouless-Berezinskii phase transition with unbinding of charge-anticharge pairs occurs at a transition temperature of about $T_c = e^2/8\pi Ck_B$, with C the junction capacitance. We calculate the influence of tunneling conductance; T_c is reduced with increasing conductance and no transition occurs for junction conductance above $(14 \text{ k}\Omega)^{-1}$. In the superconducting state a similar transition occurs at a 4 times higher T_c. We present the first experimental results on the conductive transition of an array in the normal and superconducting states.

With modern lithographic techniques it is possible to fabricate metal-insulator-metal tunnel junctions with an area below $(100 \text{ nm})^2$, and consequently a capacitance of less than 10^{-15} F. When one electron crosses the tunneling barrier, the charging energy $E_C = e^2/2C$ is about 1 K and cannot be neglected at low temperatures. This has given access to a new area of mesoscopic physics. A series of effects have been predicted theoretically, and some have recently been observed. A review is given by Averin and Likharev.[1] Most experimental effort has been directed at single junctions, circuits with two or three junctions, and longer linear arrays. Only one experimental paper[2] has appeared on fabricated 2D arrays of small junctions. In that paper a transition is reported, similar to that seen in granular films, between insulating and superconducting behavior at $T = 0$ for samples with a normal-state sheet resistance above or below the quantum resistance. A large number of theoretical papers have been devoted to this subject.[3] In the present Letter, we discuss a different aspect of 2D arrays: For certain reasonable values of the parameters the interaction between single charges on islands depends logarithmically on their separation. A real Coulomb gas with 2D interaction can be realized, and a Kosterlitz-Thouless-Berezinskii (KTB) phase transition[4] should occur at a critical temperature T_c. Below T_c, only bound charge-anticharge pairs are present; above T_c free charges $+e$ and $-e$ are generated. We calculate the influence of dissipation on this charge-unbinding transition. It leads to a suppression of T_c when the tunnel-junction resistance is lower than the quantum resistance. As we will discuss later, present-day techniques only allow fabrication of samples in which the logarithmic interaction extends over a limited number of cell distances (10–100), with a consequent rounding of the transition. In the superconducting state, a similar transition is expected at a 4 times higher temperature, where bound $(+2e)$-$(-2e)$ pairs unbind. The possibility of a charge KTB transitions in a superconducting 2D granular materials has been indicated by Sugahara and Yoshikawa[5] and by Widom and Badjou.[6]

Single-electron charge solitons in 1D chains have been discussed in detail by Averin and Likharev[1] and others.[7] A simple exact solution is available for the dependence of the island potential on position. When the nearest-neighbor capacitance is C and the self-capacitance of an island is C_0, the screening length is $\Lambda = (C/C_0)^{1/2}$. When Λ is small, the solitons are independent for low density. Solitons repel (attract) each other when they have equal (opposite) charge. We adopt a similar picture for the 2D array, concentrating on the behavior within the screening length. It should be noted that the 1D array does not show a phase transition at a finite temperature.

Consider a square 2D array of small tunnel junctions (see Fig. 1) with capacitance C, connecting "islands"
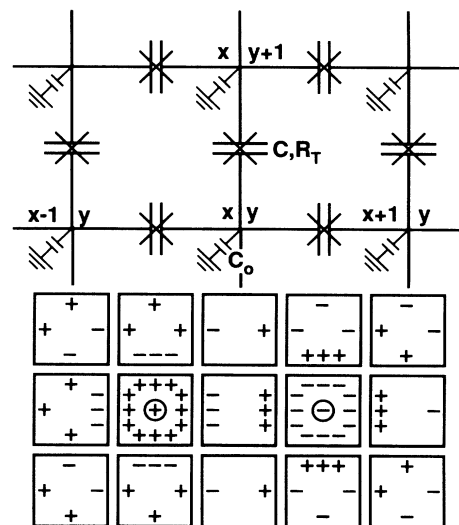

FIG. 1. Top: Approximation scheme of a square 2D network. Tunnel junctions are represented as crossed capacitances. "Islands" are positioned at integer values (x,y). Bottom: Schematic distribution of charges in the neighborhood of a charge-anticharge pair. Only two islands in the picture contain a net charge.

(x,y) with their nearest neighbors at distance 1 (lengths are dimensionless). Each island in addition has a capacitance C_0 to ground. The non-nearest-neighbor elements of the capacitance matrix are neglected. The electrical potential of island x,y is indicated as $\Phi_{x,y}$. The charge distribution for the case of a positively charged island near a negatively charged island is schematically indicated in Fig. 1. The charge on island x,y is equal to

$$q_{x,y} = C_0\Phi_{x,y} + C(4\Phi_{x,y} - \Phi_{x-1,y}$$
$$- \Phi_{x+1,y} - \Phi_{x,y-1} - \Phi_{x,y+1}) .$$

When $q_{0,0} = e$ and all other $q_{x,y}$ are zero, the potential for $r = (x^2 + y^2)^{1/2} \gg 1$ can be approximately solved in a quasicontinuous approximation from $\nabla^2\Phi(r) - (C_0/C) \times \Phi(r) = 0$, with the solution

$$\Phi(r) = \alpha K_0(r/\Lambda), \quad \Lambda = (C/C_0)^{1/2} . \tag{1}$$

The modified Bessel function $K_0(r/\Lambda)$ falls off exponentially for $r/\Lambda \gg 1$. For $r/\Lambda \ll 1$ it is approximately equal to $-\ln(r/\Lambda)$. In this regime we have $\Phi(r) = -\alpha\ln(r/\Lambda)$, which is the same potential as for the 2D Coulomb gas. From Gauss's law in the 2D medium with effective dielectric constant C we find $\alpha = e/2\pi C$. The free energy of a pair of charges $+e$ and $-e$ at a mutual distance r, for $1 \ll r \ll \Lambda$, is equal to

$$U_p = 2\mu_{core} + (E_C/\pi)\ln r . \tag{2}$$

The constant $2\mu_{core}$ is the free energy of a pair with separation 1 and includes an entropy term. Without the

latter it has a value of about $0.42E_C$. The form of (2) is the same as for vortex-antivortex pairs in the 2D X-Y model[4] or in arrays of superconducting Josephson junctions.[8] The ratio between μ_{core} and the prefactor of the logarithmic term in (2) is also very similar to the ratio in those systems. A KTB phase transition occurs at a temperature

$$k_B T_c = E_C/4\pi\varepsilon_c , \tag{3}$$

where ε_c is a nonuniversal constant slightly larger than 1. Above T_c, free charges of either sign, $\pm e$, will be present. Near T_c, their density should be given by the well-known square-root cusp formula, $n_e = K\exp\{-2b \times (T/T_c - 1)^{-1/2}\}$, where K and b are constants of order 1.

Above we concentrated on the interactions between the charges. We did not account explicitly for the tunneling of electrons between the islands, except that we assumed that it establishes the equilibrium charge distribution. However, if the tunneling conductance, characterized by the parameter $\alpha_T = (h/4e^2)/R_T$, where R_T is the junction resistance, is not small, this picture is no longer sufficient. We can investigate the influence of arbitrarily strong tunneling by means of the microscopic theory.[9] The partition function

$$Z = \int \prod_i D\varphi_i \exp\{-A[\varphi]\}$$

can be expressed as a path integral over the fields φ_i, which are related to the electric potential by $d\varphi_i/dt = e\Phi_i$. The action is[9]

$$A[\varphi] = \frac{1}{4E_C}\sum_{\langle ij\rangle}\int_0^\beta d\tau\left[\frac{d\varphi_{ij}}{d\tau}\right]^2 - \sum_{\langle ij\rangle}\int_0^\beta d\tau\int_0^\beta d\tau' \alpha(\tau-\tau')\cos[\varphi_{ij}(\tau) - \varphi_{ij}(\tau')] . \tag{4}$$

The first term represents the charging energy (here for simplicity we drop the self-capacitance and put $\hbar = 1$, $\beta = 1/k_B T$), and the second is due to the tunneling. The islands are labeled by the subscripts i, and $\varphi_{ij} = \varphi_i - \varphi_j$ refers to nearest neighbors. The dissipative kernel is $\alpha(\tau) = \alpha_T[\beta\sin(\pi\tau/\beta)]^{-2}$. The fields φ_i are conjugate to the charges and the limits of the integration in the partition function depend on the allowed charge states of the system. Since here the total charges on the islands are quantized, the integrals include a summation over the winding numbers $\varphi_i(\beta) = \varphi_i(0) + 2\pi n_i$. To proceed we decompose the phase as $\varphi_i(\tau) = \varphi_i(0) + \vartheta_i(\tau) + 2\pi n_i\tau/\beta$, where $\vartheta_i(0) = \vartheta_i(\beta) = 0$. In lowest order we consider the charging energy only. The winding-number contribution then leads to the so-called discrete Gaussian model (DGM). This model exhibits the KTB transition at the critical temperature given by (3). This result was also obtained (by a different method) in Ref. 6 where the case of a 2D Josephson array was considered. The present method allows us to extend the analysis to evaluate the influence of the dissipation by tunneling on the transition temperature. Details of the calculations will

be presented elsewhere.[10] For small α_T the dissipation can be treated perturbatively. The first-order correction to the transition temperature is

$$T_c(\alpha_T) = (E_C/4\pi\varepsilon_c)(1 - 0.1\alpha_T) . \tag{5}$$

On the other hand, for strong dissipation T_c is almost reduced to zero. In this limit it is possible to map the problem onto the absolute solid-on-solid (ASOS) model,[11] in which the coupling constant (in the limit $T \to 0$) is proportional to α_T. The critical value of dissipation determined from Monte Carlo calculations[12] is

$$\alpha_{T,crit} \approx 0.45 . \tag{6}$$

Above this critical value the Coulomb gas is always in the disordered phase. All roughening models (such as the DGM and ASOS models) belong to the same class of universality so that the transition is of the KTB type everywhere in the phase diagram (Fig. 2). The value 0.45 corresponds to a critical junction resistance of 14 $k\Omega$.
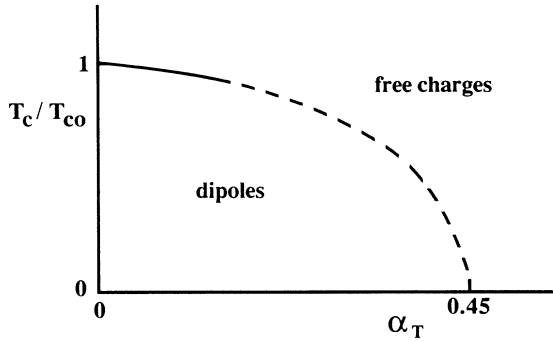
FIG. 2. Phase diagram of the normal-metal tunnel junction array. T_c is the transition temperature, $\alpha_T = (6.45 \text{ k}\Omega)/R_T$. Values of T_c have been calculated on both axes and close to the temperature axis.

The mobility of the charges is determined by the tunneling rate in the junctions. Applying the global rules, where the energies of the whole system before and after tunneling count, in a large system at low density the charge energy is independent of position and the charges should be mobile. Without driving voltage, they will diffuse around. With a voltage V over the length L of the array, the net tunneling rate is $r_t = (eR_T)^{-1}V/L$, as long as $L \ll \Lambda$. This leads to a current $I = n_e W e r_t$, where W is the array width. Consequently, the conductance of the whole array is

$$G = \left[\frac{W}{L} \right] R_T^{-1} n_e$$

$$= \left[\frac{W}{L} \right] R_T^{-1} K \exp \left[- \frac{2b}{(T/T_c - 1)^{1/2}} \right] . \qquad (7)$$

Below T_c, $G = 0$. Above T_c, the conductivity should start to rise according to Eq. (7). In practice, the screening length Λ or the array size will limit the scale over which charge-anticharge pairs exist. To have an ideal KTB transition, one needs conditions in which the logarithm of the pair separation can be much larger than 1. For finite array size or finite screening length, the transition will be rounded off.

Because of the complementarity of phase and charge as well as the similarity of the Hamiltonians involved, a high degree of correspondence exists between charges in arrays of low-capacitance superconducting or normal-metal tunnel junctions and vortices in arrays of superconducting junctions where charging effects can be ignored. In classical two-dimensional Josephson-junctions arrays, a KTB transition occurs where vortex-antivortex pairs dissociate. The resistance is zero below T_c and grows with a square-root cusp equation similar to Eq. (7) above T_c. Voltage, conductance, and charge are replaced by current, resistance, and vortex.

When the potential of both end electrodes is increased to V_g with respect to ground (V_g is much larger than V

used for measuring G) and the array is shorter than Λ, the capacitive coupling to ground leads to an induced charge $C_0 V_g$ on each island. The effect of this induced charge is a "frustration," similar to the frustration induced in classical Josephson-junction arrays by a perpendicular magnetic field. There, f is equal to the flux per cell divided by the superconducting flux quantum $h/2e$. In the 2D charge system the frustration is

$$f = C_0 V_g / e . \qquad (8)$$

The properties of the array should be periodic in f with period 1. In practical fabricated arrays random fractional charges will sometimes be induced on islands by trapped charges in the barriers or on the film surfaces. Their presence leads to a random initial additional value of f for each island.

In the superconducting state, if there are no quasiparticles, the unit of charge is $2e$ and the charging energies are larger by a factor of 4. This is also true for the KTB temperature, which should now be $k_B T_{cs} = E_C/\pi \varepsilon_c$. Because of the presence of Josephson tunneling, and because the charges have equal energy on all islands, the charges will not be localized. However, the calculation of the conductance in the highly correlated superconducting state is more complicated. Also, the influence of dissipation on T_{cs} is different from the normal-state case. Widom and Badjou[6] previously indicated the possibility of a charge KTB transition in granular superconducting films, and gave the same (unrenormalized) transition temperature. From the correspondence with classical 2D Josephson-junction arrays, Sugahara and Yoshikawa also qualitatively predicted the charge transition in superconducting films. It is clear that for large Josephson coupling energy E_J, the superconducting phase coherence dominates at low temperatures and the resistance is zero. When $E_C \gg E_J$, on the other hand, the conductance is zero at low T. This implies that a zero-temperature transition should occur between a superconducting and an insulating phase when E_C is of order E_J. This is exactly the type of transition that we reported on in Ref. 2, which had to be studied by fabricating a series of samples with varying resistance and E_J. In those samples that become insulating at $T = 0$, we expect the charge-pair-unbinding transition to occur when the temperature is increased to T_{cs}.

We have experimentally investigated this transition in an aluminum array with $(100 \text{ nm})^2$ junctions, an island size of about $(0.5 \text{ } \mu\text{m})^2$, and a cell size of $(2 \text{ } \mu\text{m})^2$. The junction resistance R_T is 15.3 kΩ. The array length is 190 cells, and the width 60 cells. We estimate the self-capacitance to ground to be about 3×10^{-18} F. The junction capacitance is near 10^{-15} F, so Λ is about 18 cells. The array is considerably larger than Λ, which should lead to significant rounding of the transition. According to Eq. (3) the normal-state T_{c0} should be near 60 mK (for ε_c about 1.2). For this array, with a junction
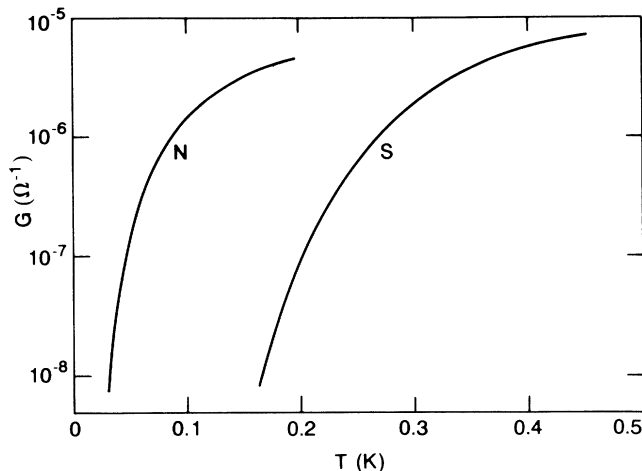
FIG. 3. Measured conductance of an array of $(100 \text{ nm})^2$ aluminum tunnel junctions, 190 cells long and 60 cells wide. N is in the normal state (magnetic field of 3T applied); S in the superconducting state.

resistance such that a_T is just above the critical value (6), we expect the normal-state transition temperature to be considerably reduced below T_{c0}. For the same sample, the conductance in the normal state and in the superconducting state is given in Fig. 3. The normal state is achieved by application of a 3-T magnetic field. As shown, the conductance is zero at low temperatures and increases sharply above about 20 mK in the normal state and 160 mK in the superconducting state, clearly showing the conductive transition.

We consider the value of the transition temperature in the normal state to be in good agreement with the theoretical prediction, including effects of dissipation. The functional dependence of G on T does not follow the square-root cusp dependence, due to the limited screening length. The transition in the superconducting state at 160 mK is to be compared with the theoretical value T_{cs}, about 240 mK without taking dissipation into account. It appears from the experiment that the influence of dissipation is smaller in the superconducting state (no theoretical calculation is available as yet).

We want to draw attention to the remakable fact that the conductance is orders of magnitude smaller in the su-

perconducting state compared with the normal state. This directly demonstrates that charging effects are dominating. We expect that it is possible to fabricate arrays with smaller islands and an order of magnitude smaller junction capacitance. The increased screening length will allow a closer test of the theory.

(a)On leave from Faculty of Engineering, Institute of Physics, Catania, Italy.

[1] D. V. Averin and K. K. Likharev, in "Quantum Effects in Small Disordered Systems," edited by B. L. Altshuler, P. A. Lee, and R. A. Webb (to be published).

[2] L. J. Geerligs, M. Peters, L. E. M. de Groot, A. Verbruggen, and J. E. Mooij, Phys. Rev. Lett. 63, 326 (1989).

[3] See Proceedings of the NATO Advanced Research Workshop on Coherence in Superconducting Networks, edited by J. E. Mooij and G. B. J. Schön [Physica (Amsterdam) 152B, 1–302 (1988)].

[4] J. M. Kosterlitz and D. J. Thouless, J. Phys. C 6, 1181 (1973); V. L. Berezinskii, Zh. Eksp. Teor. Fiz. 59, 907 (1970) [Sov. Phys. JETP 32, 493 (1971)].

[5] M. Sugahara and N. Yoshikawa, in Extended Abstracts of the International Superconductivity Electronics Conference (ISEC '87), Tokyo, 1987 (unpublished), p. 341; N. Yoshikawa, T. Akeyoshi, M. Kojima, and M. Sugahara, Jpn. J. Appl. Phys. 26, 949 (1987).

[6] A. Widom and S. Badjou, Phys. Rev. B 37, 7915 (1988).

[7] M. Amman, E. Ben-Jacob, and K. Mullen, Phys. Lett. A 142, 431 (1989).

[8] C. J. Lobb, D. W. Abraham, and M. Tinkham, Phys. Rev. B 27, 150 (1983).

[9] V. Ambegaokar, U. Eckern, and G. Schön, Phys. Rev. Lett. 48, 1745 (1982); E. Ben-Jacob, E. Mottola, and G. Schön, Phys. Rev. Lett. 51, 2064 (1983); G. Schön and A. D. Zaikin, Physica (Amsterdam) 152B, 203 (1988).

[10] R. Fazio, U. Geigenmüller, and G. Schön (to be published).

[11] Y. Saito and H. Müller-Krumbhaar, in Applications of the Monte Carlo Method, edited by K. Binder (Springer-Verlag, Berlin, 1984).

[12] R. H. Swendsen, Phys. Rev. B 15, 5421 (1977).