Charging Effects and Quantum Coherence in Regular Josephson Junction Arrays

L. J. Geerligs, M. Peters, L. E. M. de Groot,[(a)] A. Verbruggen,[(a)] and J. E. Mooij
Department of Applied Physics, Delft University of Technology, P.O. Box 5046, 2600 GA Delft, The Netherlands
(Received 17 April 1989)

Two-dimensional arrays of very-small-capacitance Josephson junctions have been studied. At low temperatures the arrays show a transition from superconducting to insulating behavior when the ratio of charging energy to Josephson-coupling energy exceeds the value 1. Insulating behavior coincides with the occurrence of a charging gap inside the BCS gap, with an S-shaped I-V characteristic. This so far unobserved S shape is predicted to arise from macroscopic quantum coherent effects including Bloch oscillations.

PACS numbers: 74.50.+r, 05.30.−d, 74.40.+k

In the last few years the effects of charging energy in small Josephson junctions have been the subject of intensive theoretical study.[1] As noted by several authors,[2] experiments on very small junctions can provide important information about the validity of quantum mechanics on a macroscopic scale. From microscopic theory it has been derived[3] that a Josephson junction can be described as a quantum particle of mass $\hbar^2/8E_C$ in a periodic potential $-E_J\cos(\phi)$. Here the charging energy is $E_C \equiv e^2/2C$, C is the capacitance of the junction, E_J is the Josephson-coupling energy, and ϕ is the phase difference across the junction. With increasing ratio $x \equiv E_C/E_J$ the quantum-mechanical behavior of the junction (delocalization in phase-coordinate space) should become more noticeable. So far only low-x effects, i.e., macroscopic quantum tunneling and energy-level quantization,[4] have been observed convincingly. For high x the behavior of a junction should be governed by a band energy spectrum. External current causes a sweep of the junction Bloch state through this band spectrum. For increasing current the voltage is subsequently dominated by single-electron tunneling, Bloch oscillations, and finally Zener tunneling, resulting in a characteristic S shape of the I-V curve.[5] Although high-x junctions have been fabricated before,[6–8] this S shape was not observed. Yoshihiro *et al.*[9] reported on microwave-induced voltage steps in granular superconducting films which they interpreted as due to Bloch oscillations. Owing to the undefined nature of their samples this interpretation has not been generally accepted. Iansiti *et al.*[7] find in superconducting junctions a knee in the I-V characteristics which may be related to the above effects. The knee only occurs when E_j is suppressed by a magnetic field.

In this Letter experiments are reported on two-dimensional arrays of well-defined high-x junctions that prominently exhibit the predicted current-voltage dependence. We consider this as the clearest observation so far of macroscopic quantum coherent effects. The prominent negative slope is also a manifestation of the more general phenomenon of Bloch oscillations.

In addition the arrays provide the opportunity to test the effects of charging energy on coherence in two-dimensional systems. Generally quantum fluctuations of the phase destroy global superconductivity for high x.[10] Experiments on granular films[11] suggested that apart from the parameter x the junction dissipation, i.e., coupling to external degrees of freedom which is proportional to the quasiparticle conductance, has a strong influence on macroscopic quantum effects. It appeared that whether or not a granular film became superconducting depended on dissipation only, and not on x. In this Letter we compare our experimental results with predictions of phase diagrams for 2D systems.[10,12–14] Owing to the fabrication by nanolithographic methods, reliable estimates of E_J and E_C are available and percolation effects are absent. In short, results for $T=0$ show a phase transition from insulating behavior at high x to superconducting at low x, with at most a small dependence on dissipation. Preliminary results were published in Ref. 15.

The junctions in the arrays are arranged in a square network. The arrays are 190 junctions long and 60 junctions wide. The junctions are made of aluminum, and have an area of 0.01 or 0.04 μm^2. The area of the aluminum islands is approximately 1.9 μm^2. Since we found shielding of magnetic and electrical interference to be critical, we give some details of our experimental setup. The experiments on the arrays in the superconducting state were performed inside a magnetic shield. A magnetic field of 4.1 G corresponded to a flux quantum $\Phi_0 = h/2e$ per unit cell. (The area of the elementary cell is 4.9 μm^2.) The typical remanent field was between 0.04 and 0.004 G. In this paper the field is indicated as the frustration f, the flux per cell divided by Φ_0. The leads to the arrays were filtered at the entrance to the cryostat with rf interference feedthrough filters. At mixing-chamber temperature the leads were filtered by RC filters and microwave filters[4] before entering the electrically shielded case containing the arrays. For recording the charging gap, in a separate experiment we put 10-MΩ resistors in the leads close to the arrays. All the measuring methods were standard except for the addition of analog optical decoupling between the current source and preamplifier and the rest of the equipment.

As the critical current was too small to be measured directly, we calculated E_J with I_C given by the Ambegaokar-Baratoff equation, i.e., $E_J = \pi\hbar\Delta/4e^2R_n$ at zero temperature, using experimental values of the normal-state resistance R_n and the critical temperature T_c and assuming $\Delta(0)=1.76k_BT_c$. Results on larger single junctions justify this procedure.

For $T > T_c$ or in a large magnetic field, i.e., in the normal state, the arrays show the effect of charging energy as the "Coulomb gap" in the I-V curve. This is a voltage offset of magnitude $e/2C$ for a single junction and $Le/2C$ for an array, where L is the length of the array. For details we refer to Ref. 15. From this offset the capacitance is calculated. It is about 1.1 fF for a 0.01-μm^2 junction, and proportionally larger for the larger junctions.

Figure 1 shows $R(T)$ curves for several arrays in zero magnetic field, measured with a lock-in amplifier and current bias. The current was chosen small enough that the resistance was linear for increasing current, typically 0.1 to 1 nA. The resistance given is the measured resistance divided by the length/width ratio 3.14 of the array. For the five arrays shown, $E_C \approx 0.84$ K is constant and R_n varies from 4.8 to 36 kΩ. Since the critical temperature of the aluminum was also approximately constant, $T_c = 1.37$ K, this causes x to vary from 0.53 to 3.9. In

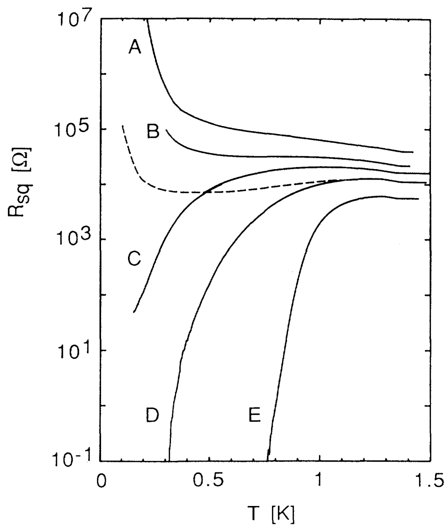

FIG. 1. $R(T)$ curves for arrays of 0.01-μm^2 junctions ($E_C \approx 0.84$ K). R_{sq} is the resistance divided by the length/width ratio 3.14. Each solid curve corresponds to an array with a particular normal-state resistance R_n in zero field. The dashed curve is for array D with $f \approx \frac{1}{2}$. Values of R_n in kΩ, E_J/k_B in K, and $x = E_J/E_C$ are, sample A: 36, 0.22, 3.9; B: 15.3, 0.51, 1.8; C: 14.1, 0.55, 1.5; D: 9.7, 0.80, 1.0; E: 4.8, 1.6, 0.53.

the figure caption the relevant data are given for each array. The arrays with the smallest x, which are not all included in Fig. 1, show the Kosterlitz-Thouless transition in the form of the square-root cusp behavior $R(\tau) = R_{qp}\exp[-b/(\tau-\tau_c)^{0.5}]$,[16] where $\tau = kT/E_J(T)$, and R_{qp} is the temperature-dependent quasiparticle resistance. The $R(T)$ curves for the arrays with significant charging energy show deviations from a Kosterlitz-Thouless transition. For arrays with $x \lesssim 1.0$ (curves D and E in Fig. 1), the resistance decreases to zero within experimental accuracy (about 0.01 Ω), but the transition temperature is significantly lower than the Kosterlitz-Thouless temperature. For array C, with $x \approx 1.5$, the resistance decreases in a similar way down to 0.1 K. At that temperature the "supercurrent" in the I-V curve becomes noisy with voltage spikes, the effect getting worse for lower temperature. It is therefore impossible to attribute a resistance to this array below 0.1 K. Arrays with still higher x show at low temperatures a strongly increasing resistance with no sign of flattening off.

In earlier experiments[15] we found a flattening off of the resistance at low temperatures. This feature has completely disappeared with the addition of the special cryogenic microwave filtering to the experiments.

At first sight all I-V curves show the same general features, similar to those of classical arrays. Figure 2(a) gives an example for $x \approx 3.9$ (array A). For increasing current there is first a supercurrentlike part. Then the voltage increases from near zero to a value equal to the length of the array times the single-junction BCS sum gap. Finally, after the gap edge, the voltage increases with the normal-state resistance of the array.

The new phenomenon of these quantum arrays, with $x > 1$, is the existence of a small second gap, in the supercurrentlike part of the I-V characteristic at low temperatures. This gap, of order 1 mV, is situated *inside* the BCS gap (80 mV in our 190-junction-long arrays). In the following we denote it as the charging gap. Figure 2(b) shows it for $T \approx 20$ mK. At this temperature the resistance in the gap is larger than 5 GΩ. The occurrence of the charging gap is responsible for the strong increase of resistance at low T for the high-x arrays in Fig. 1. In arrays with $E_C \approx 1$ K the charging gap becomes visible below 0.5 K; for $E_C \approx 0.4$ K, below 0.2 K. Below 0.1 K the gap edge of the high-x arrays develops a negative-resistance region. In an array with Hall contacts we verified that the gap is present proportionally in both halves of the array. This indicates that the gap is distributed over the length of the array, instead of being localized in certain cross rows. The charging gap is present in the I-V curves of the highest-x arrays at zero magnetic field. It is also present in arrays with smaller x (down to 0.5) at low temperatures if the array is frustrated in a magnetic field. For frustrated arrays the gap can cause quasireentrant behavior of the $R(T)$ curve (dashed curve in Fig. 1). No quasireentrant behavior

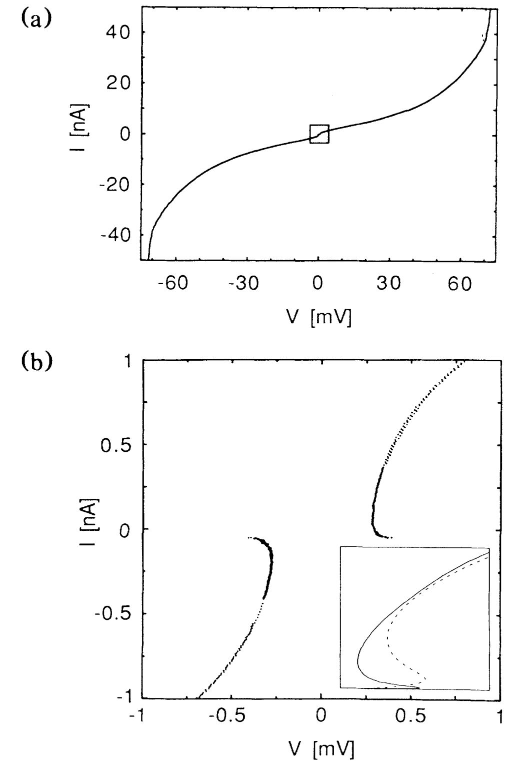

FIG. 2. I-V characteristic for sample A at 20 mK. (a) At large scale showing the BCS sum gap of the array, with a small "supercurrent." (b) Small-current region [box in (a)] with voltage measured over 95 junctions, showing effects of Bloch oscillations and Zener tunneling. Inset: Calculated I-V curves (Ref. 17) for a single junction (dashed curve) and of a circuit of one junction parallel to two junctions (solid curve). The junction parameters are chosen to be the estimated parameters for sample A. The axes are in arbitrary units but identical for the two calculated curves.

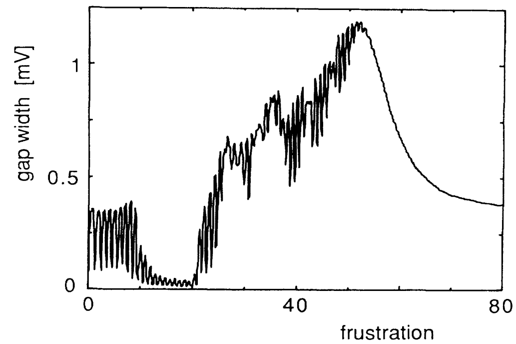

FIG. 3. Voltage for $I = 10$ pA in array C. This voltage is indicative of the charging gap and oscillates with a period of one flux quantum per cell.

was found in zero field. The width of the charging gap is modulated by the frustration with period 1. For large fields the width gradually increases and the gap changes into the normal-state Coulomb gap as the superconductivity in the islands is destroyed. This behavior is shown in Fig. 3.

Macroscopic quantum behavior of single high-x junctions is predicted to yield an S-shaped I-V curve because of band-spectrum effects.[5] For low current, quasiparticle tunneling confines the junction to the center of the first Brillouin zone, and the I-V curve follows a high-resistance branch. For higher currents, Bloch oscillations, which can be regarded as coherent Cooper pair tunneling, become important, decreasing the mean voltage. The resulting low-current part of the I-V curve is known as the "Bloch nose." For qualitative comparison, the inset of Fig. 2(b) shows calculated I-V curves[17] for a single junction and a series-parallel arrangement of three junctions. The trend of a sharpening up of the Bloch nose with increasing number of degrees of freedom is consistent with the experimental I-V curve. A quantitative comparison will have to wait for a more similar theoretical system.

The superconductor-insulator transition of Fig. 1 can be compared with theory. Quantum XY models which do not include dissipation[10] generally predict a transition from superconducting to nonsuperconducting behavior near $x = 1$. Several theoretical calculations[12] have shown that quasiparticle dissipation significantly influences superconductivity in Josephson junction arrays. Quasiparticle tunneling in addition leads to a renormalization of the capacitance[13,14,18] which even at low temperatures depends on the *normal-state* resistance. In our junctions the subgap resistance is very high so that quasiparticle dissipation is negligible. This leaves only capacitance renormalization to be considered in addition to bare-charging effects.

Array D, clearly showing superconducting behavior, has a value $x = 1.04$, calculated from the normal-state Coulomb gap. Possibly this Coulomb gap is suppressed by heating.[15] We estimate that x is between 1.0 and 1.3. Similarly, array C has $1.5 < x < 2.0$. It appears to go superconducting but develops the above described noisy supercurrent below 0.1 K. Array B, the first insulating sample, has $1.8 < x < 2.5$. So, the experimental transition is close to $x = 1.5$. The variation in the prediction for the critical x from various bare-charging theories is large and $x = 1.5$ lies in their range. In contrast, the transition observed in granular films[11] occurred at $x \gg 1$.

The phase diagram of a 2D array of Josephson junctions, influenced by capacitance renormalization, was evaluated by Chakravarty *et al.*[13] and Ferrell and Mirhashem.[14] The experimental results for granular films, where the capacitance can only be estimated, are in reasonable agreement with that phase diagram. In our arrays the capacitance is well known. Applying the phase diagram of Ref. 13, the transition should occur for $R_n = 13\ \mathrm{k\Omega}$. This is in excellent agreement with our experimental data. The same holds for the similar treatment in Ref. 14.

We thank J. Martinis and M. Devoret for explaining details of their cryogenic filtering, U. Geigenmüller for providing his results of *I-V* calculations, and H. van der Zant, G. Schön, U. Geigenmüller, and H. Jaeger for valuable discussions. This work was supported by the Dutch Foundation for Fundamental Research on Matter (FOM).

[(a)]Centre for Submicron Technology.

[1]See, e.g., A. Widom *et al.*, J. Phys. A **15**, 3877 (1982); K. K. Likharev and A. B. Zorin, J. Low Temp. Phys. **59**, 347 (1985); E. Ben-Jacob and Y. Gefen, Phys. Lett. A **108**, 289 (1985); F. Guinea and G. Schön, J. Low Temp. Phys. **69**, 219 (1987); M. Büttiker, Phys. Rev. B **36**, 3548 (1987); S. V. Panyukov and A. D. Zaikin, J. Low Temp. Phys. **73**, 1 (1988).

[2]A. J. Legget, Suppl. Prog. Theor. Phys. **69**, 80 (1980); A. I. Larkin, K. K. Likharev, and Yu. N. Ovchinnikov, Physica (Amsterdam) **126B**, 414 (1984).

[3]V. Ambegaokar, U. Eckern, and G. Schön, Phys. Rev. Lett. **48**, 1745 (1982); U. Eckern, V. Ambegaokar, and G. Schön, Phys. Rev. B **30**, 6419 (1984).

[4]J. M. Martinis, M. H. Devoret, and J. Clarke, Phys. Rev. B **35**, 4682 (1987).

[5]D. V. Averin, A. B. Zorin, and K. K. Likharev, Zh. Eksp. Teor. Fiz. **88**, 692 (1985) [Sov. Phys. JETP **61**, 407 (1985)]; U. Geigenmüller and G. Schön, Physica (Amsterdam) **152B**, 186 (1988); A. D. Zaikin, and I. N. Kosarev, Phys. Lett. A **131**, 125 (1988).

[6]T. A. Fulton and G. J. Dolan, Phys. Rev. Lett. **59**, 109 (1987).

[7]M. Iansiti *et al*, Phys. Rev. Lett. **59**, 489 (1987); Phys. Rev. Lett. **60**, 2414 (1988).

[8]L. S. Kuzmin *et al.*, Phys. Rev. Lett. **62**, 2539 (1989).

[9]K. Yoshihiro *et al.*, Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 1379 (1987).

[10]B. Abeles, Phys. Rev. B **15**, 2828 (1977); K. B. Efetov, Zh. Eksp. Teor. Fiz. **78**, 2017 (1980) [Sov. Phys. JETP **51**, 1015 (1980)]; S. Doniach, Phys. Rev. B **24**, 5063 (1981); E. Simanek, Phys. Rev. B **25**, 237 (1982); L. Jacobs and J. V. José, Physica (Amsterdam) **152B**, 148 (1988); R. Fazio, G. Falci, and G. Giaquinta, Physica (Amsterdam) **152B**, 257 (1988).

[11]B. G. Orr, H. M. Jaeger, and A. M. Goldman, Phys. Rev. B **32**, 7586 (1985); A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **33**, 3549 (1986); H. M. Jaeger *et al.*, Phys. Rev. B **34**, 4920 (1986); N. Yoshikawa *et al.*, Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 949 (1987); S. Kobayashi, Physica (Amsterdam) **152B**, 223 (1988).

[12]See, e.g., M. P. A. Fisher, Phys. Rev. B **36**, 1917 (1987); A. Kampf and G. Schön, Phys. Rev. B **36**, 3651 (1987); Physica (Amsterdam) **152B**, 239 (1988); S. Chakravarty *et al.*, Phys. Rev. B **37**, 3283 (1988); W. Zwerger, J. Low Temp. Phys. **72**, 291 (1988); A. D. Zaikin, Physica (Amsterdam) **152B**, 251 (1988); S. E. Korshunov, Physica (Amsterdam) **152B**, 261 (1988); A. I. Larkin, Yu. N. Ovchinnikov, and A. Schmid, Physica (Amsterdam) **152B**, 266 (1988).

[13]S. Chakravarty, S. Kivelson, G. Zimanyi, and B. I. Halperin, Phys. Rev. B **35**, 7256 (1987).

[14]R. A. Ferrell and B. Mirhashem, Phys. Rev. B **37**, 648 (1988); B. Mirhashem and R. A. Ferrell, Physica (Amsterdam) **152C**, 361 (1988).

[15]L. J. Geerligs, and J. E. Mooij, Physica (Amsterdam) **152B**, 212 (1988).

[16]C. J. Lobb, D. W. Abraham, and M. Tinkham, Phys. Rev. B **36**, 150 (1983).

[17]U. Geigenmüller and G. Schön, in Ref. 5; (private communication).

[18]V. Ambegaokar, in *Percolation, Localization, and Superconductivity,* Proceedings of the Nato Advanced Study Institute, Les Arcs, France, 1983, edited by A. M. Goldman and S. A. Wolf (Plenum, New York, 1984), p. 43.