Experimental Fine Tuning of Frustration: Two-Dimensional Superconducting Network in a Magnetic Field

B. Pannetier, J. Chaussy, and R. Rammal
Centre de Recherches sur les Très Basses Températures,
Centre National de la Recherche Scientifque, F-38042 Grenoble Cédex, France

and

J. C. Villegier
Laboratorie d'Electronique et de la Technologie de l'Informatique IRDI, F-38041 Grenoble-Cédex, France
(Received 7 May 1984)

This Letter reports measurements of the magnetic field dependence of the resistive critical temperature T_c of a regular square network of superconducting aluminum. We find new effects of flux quantization corresonding to both integral $(1,2,3,\ldots)$ and fractional $(\frac{1}{4},\frac{1}{3},\frac{2}{5},\frac{1}{2},\frac{3}{5},\frac{2}{3},\frac{3}{4})$ numbers of flux quanta per unit cell of the network. The fractal fine structure of the upper critical-field line is identified as the edge of the Landau-level spectrum for a tight-binding problem on a square lattice.

PACS numbers: 74.10.+v, 73.60.Ka, 74.40.+k

Frustration[1] means competing interactions and therefore unsatisfied states. Degeneracy, metastability, and sensitivity to topology or external parameters are just a few characteristic features of frustrated systems. The three basic concepts are those of gauge invariance, frustration function, and curvature. During the last years the manifestation of these concepts in various fields has been the object of intensive studies[2]: magnetic systems (spin-glasses), amorphous packing and random networks, optimization problems (traveling salesman problem), biological problems (neutral networks, content addressable memories, the origin of life), etc. However, in most of these examples frustration and disorder occur simultaneously, leading to new phenomena like erogodicity breaking or failure of linear-response theory, which are characteristics of spin-glasses.

In this Letter we propose a study of a physical system where frustration enters alone and in a controllable way. For this we investigate experimentally the magnetic properties of a two-dimensional (2D) superconducting regular network. The frustration is induced by the magnetic field. Continuous variation of the applied field allows for a fine tuning of the frustration that is not easy to achieve otherwise. Besides the proper superconducting properties, the studied system provides a simple physical realization of an almost periodic system described by the so-called almost-Mathieu equation,[3] which is relevant for various physical situations. Contact with other physical problems leaves us with a large program for further investigations.

The samples were prepared by electron-beam evaporation of pure aluminum on a silicon substrate. The Al film, 800 Å thick, has a sheet resistance 0.33 $\Omega/\square$ at room temperature and a residual resistivity ratio (RRR) of 5.2 between room temperature and 4.2 K. This corresponds to a resistivity $\rho = 5.08\times10^{-7}$ Ω cm at 4.2 K. The network pattern was made by reactive-ion etching by optical lithography. The investigated sample is a square network of Al strips 2 μm wide of periodicity 6 μm containing about 2.7 million identical square loops of medium area 36 μm^2. The overall size of the network is 1×1 cm^2. The sample is attached to a copper block thermally linked to the mixing chamber of a dilution refrigerator. Control of the temperature to better than 10^{-4} K is achieved thanks to a proportional, integral, derivative thermal regulation. The sample resistance is measured by a four-terminal resistance bridge at low injection current, typically 1 μA, which corresponds to a current density ~ 0.3 A/cm^2. The power dissipation at the interface between the Al film and the Si substrate is $\sim 10^{-12}$ W/cm^2. In order to monitor the critical line with accuracy, we use the sample resistance as the sensor for temperature regulation. As the magnetic field is increased, the resistance change is fed back to the temperature regulation in order to keep the network at a constant value R_0 $(=0.1\ \Omega)$ chosen at the steepest slope in the resistive transition. The field-dependent critical temperature $T_c(H)$ is defined as the temperature for which $R(T)=R_0$. By this method, the network is maintained at $T=T_c(H)$ during the sweep of the field. T_c is measured with the help of a calibrated germanium thermometer. A further improvement of the signal-to-noise ratio is obtained by averaging the data over a large number of field sweeps: 85 cycles for data shown in Fig. 1(b).

Figure 1(a) shows the field dependence of the network critical temperature. We observe well-defined oscillations superposed on a parabolic background. This parabolic envelope simply reflects the contribution of the bulk of the Al strands to the critical field, varying as $H_{c0}(\lambda/d) \sim (T_{c0}-T)^{1/2}$. Here H_{c0} denotes the thermodynamic critical field, λ is the penetration depth, d the width of the strands, and $T_{c0} = T_c(H=0)$ the bulk critical temperature of Al at zero field. The modulations observed in Fig. 1(a) are the results of a flux-quantization effect. We observe more than six periods for each polarity of the field. These oscillations are reminiscent of Little-Parks oscillations[4] resulting from one-loop quantization phenomena. The magnetic period 0.606 Oe corresponds to one-flux quantum $\phi_0 = hc/2e$ ($=2.07\times10^{-7}$ G cm^2) per unit cell. The enclosed area inside elementary loops estimated from this magnetic period is found to be 34.1 μm^2 and corresponds to a square cell of side $a = 5.84$ μm. This value compares very well to the value $a = 6.0 \pm 0.1$ μm deduced from microscope observation.

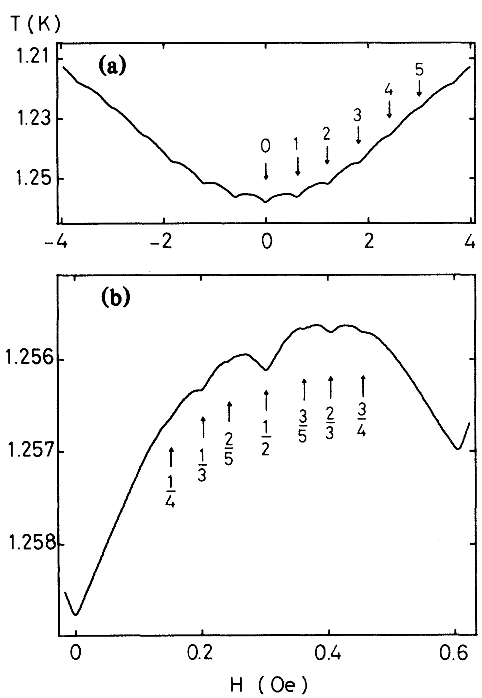

FIG. 1. (a) Critical temperature vs magnetic field for a square network made of superconducting Al. The arrows indicate the magnetic field values corresponding to integral number of flux quanta ϕ/ϕ_0 per unit cell of the network (b) Magnified view of the first period $(0 \leqslant \phi \leqslant \phi_0)$ of the critical line. In addition to the fundamental dip at $\phi/\phi_0 = 1$ the secondary dips at $\phi/\phi_0 = \frac{1}{4}, \frac{1}{3}, \frac{2}{5}, \frac{1}{2}, \frac{3}{5}, \frac{2}{3}$, and $\frac{3}{4}$ are indicated by the corresponding arrows.

Figure 1(b) is a magnification of the critical line discussed above. In addition to the single-loop effect, this plot reflects several features which are characteristic of the network behavior: finite slope dT_c/dH at $H=0$, downwards cusp at the half-integer value of the reduced flux ϕ/ϕ_0, and the down concavity of the modulation. All these features reflect the collective behavior which distinguishes the network from the single loop from the point of view of flux quantization. This distinction was emphasized in a previous study of a honeycomb network of superconducting indium.[5] In addition to these characteristic features, new structures of the critical line are shown in Figure 1(b). As can be seen, well-defined dips are observed at $\phi/\phi_0 = \frac{1}{4}, \frac{1}{3}, \frac{2}{5}, \frac{1}{2}, \frac{3}{5}, \frac{2}{3}$, and $\frac{3}{4}$, in addition to the fundamental dip at $\phi = \phi_0$. These salient features, which are absent in the case of one loop, correspond to new flux-quantization phenomena clearly observed with such details for the first time. Previous observations of one such feature, at $\phi/\phi_0 = \frac{1}{2}$, have been reported in the literature.[6]

It is possible to understand quantitatively the behavior of the critical line in the framework of recent theoretical works[7-9] on superconducting networks. The mean-field theory involved in the computation of the upper critical field of a superconducting network (neglecting fluctuations) consists of solving the linearized Ginzburg-Landau equation, which is essentially equivalent to the Schrödinger equation. For a regular lattice, the corresponding equation can be written

$$[z \cos(a/\xi_s)]\Delta_\alpha = \textstyle\sum_\beta \Delta_\beta \exp(-i\gamma_{\alpha\beta}) , \qquad (1)$$

where Δ_β is the value of the order parameter at node β (neighbor of α) and $\gamma_{\alpha\beta} = (2\pi/\phi_0)\int_\alpha^\beta \vec{A} \cdot d\vec{l}$ is the circulation of the vector potential along the strand, of length a, linking α to β. In Eq. (1), z denotes the coordination number of the lattice, and ξ_s is the superconducting coherence length. Thus the problem reduces to that of Landau levels of a tight-binding model in the same geometry. The energy of Bloch electrons is given by $\epsilon = z\cos(a/\xi_s)$. Note that for superconducting networks one is only interested in the lowest eigenvalue, i.e., the edge of the spectrum.

For a square lattice ($z=4$), there are no eigenvalues outside the range $-4 \leqslant \epsilon \leqslant 4$, and the spectrum shows invariance properties in ϵ and $\gamma = 2\pi\phi/\phi_0$, where $\phi = Ha^2$ is the magnetic flux through the elementary square loop of side a.

The Landau-level spectrum associated with Eq. (1), for a tight-binding problem on a square lattice, has been studied in detail by Hofstadter.[10] Using a Landau gauge and looking for periodic solutions in the y direction, one obtains the following equation

$$\epsilon\Delta_n = \Delta_{n+1} + \Delta_{n-1} + 2\cos(n\gamma - \alpha)\Delta_n \ , \qquad (2)$$

where α denotes a Floquet factor and $x = na$ (n integer) is the x coordinate of the lattice sites. The one-dimensional equation so obtained, known as the almost-Mathieu equation,[3] appears also in different physical problems like lattice dynamics (phonons) or electrons in a modulated crystal[11] and models of physical properties of incommensurate structures.

The upper critical field, calculated from Eq. (2) for different ratios p/q, is shown (dots) in Fig. 2, which permits a direct comparison with experiment. Otherwise, this figure must be compared with the lower edge of the whole spectrum, shown in Fig. 1 of Ref. 9.

The edge of the spectrum is a fractal continuous curve exhibiting self-similarity as was pointed out by Hofstadter.[10] The genesis of this curve has been discussed in Ref. 7. The variation of the lowest eigenvalue as function of ϕ determines the effect of frustration induced by the magnetic field on the superconducting transition temperature treated in mean-field theory. The frustration parameter is simply the reduced flux ϕ/ϕ_0. Each rational $\phi/\phi_0 = p/q$ corresponds to an amount of frustration in the network, giving rise to a particular periodic organization of vortices and currents. For instance, if $\xi_s >> a$ (continuous limit), the critical currents are exceeded locally, so that some strands are driven normal by frustration, corresponding to normal vortex cores in the continuum limit. The presence of these normal regions allow the network to accomodate flux continuously. On the contrary for $\xi_s << a$, vortices have no normal core[7] and the state of the system is described by a suitable distribution of vortex cores in the holes of the network. Physically, this corresponds to flux-quantization phenomena through superunit cells, resulting in a commensurate structure at rational ϕ/ϕ_0. This flux quantization is at the origin of the reduction of T_c, without any normal core. For instance, $\phi/\phi_0 = \frac{1}{2}$ is associated with a checkerboard lattice superstructure.

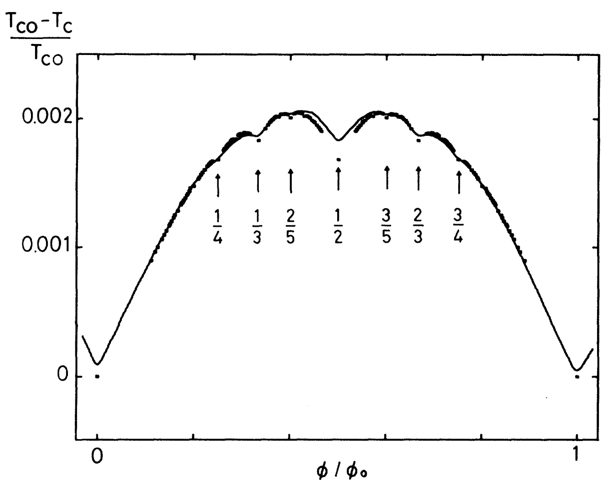

FIG. 2. Solid curve, same experimental curve as in Fig. 1, with reduced units, $\Delta T_c/T_{c0}$ vs ϕ/ϕ_0, where the parabolic background has been substracted; dotted curve, theoretical values calculated for rational $\phi/\phi_0 = p/q$ ($q \leq 30$). The best-fit parameters are $T_{c0} = 1.259$ K and $\xi_s(0) = 3050$ Å, respectively.

A close comparison between theory and experiment is illustrated in Fig. 2. The continuous curve is the critical line [same data as in Fig. 1(b)] plotted in reduced units $\Delta T_c/T_{c0}$ vs ϕ/ϕ_0, once the parabolic envelope ($T_{c0} - T_c = AH^2$) has been numerically subtracted. In the same figure is shown the theoretical values derived from Eq. (2) by

$$\frac{T_{c0}-T_c}{T_{c0}} = \frac{\Delta T_c}{T_{c0}} = \frac{\xi_s^2(0)}{a^2}\left(\arccos\frac{\epsilon}{4}\right)^2 . \qquad (3)$$

The best fit is obtained for values of the parameters $T_{c0} = 1.259$ K, $\xi_s(0) = 3050$ Å, and $A = 0.498$ K/Oe^2. The result obtained for the zero-temperature coherence length is in good agreement with that expected for dirty Al, with[12] $\xi_s(0) = 0.855\ (\xi_0 l)^{1/2}$ if one takes the mean free path $l = 800$ Å and the accepted value of the intrinsic coherence length $\xi_0 = 1.6\ \mu$m for Al.

As can be seen, the agreement is quite good and calls for some comments. In the above analysis, we have neglected superconducting fluctuations. We do not claim that this assumption is necessarily valid in all relevant situations. However, the perfect linear behavior of $H_c(T)$ near T_{c0} shows no sign of deviation from mean-field behavior. We believe that deviations from perfect regularity of the network are the main sources of discrepancy between theory and experiment. At least, for fully frustrated networks ($\phi/\phi_0 = \frac{1}{2}$) disorder will modify in a sensitive way the numerical value of $T_c(H)$. We expect, however, that disorder is not strong enough to modify $T_c(H)$ at small fields in the same way. This is actually the case at $\phi/\phi_0 \leq \frac{1}{3}$ as shown in Fig. 2. On the other hand, the finite width of the strands as well as the finite measurement current may smooth out some unobserved sharp singularities predicted by the theory.

Let us conclude by noting that Hofstadter[10] has pointed out that the question of irrationality is somewhat artificial in real materials. It would require an enormous magnetic field of about 10^6 kOe to let $\phi/\phi_0 \sim 1$ for a typical lattice spacing of the order of 2 Å. Nowadays, experimental limitations do not allow for magnetic fields higher than 10^3 kOe, which implies that the influence of the magnetic field on the motion of the electrons is very small and the rich spectrum is not felt experimentally in this regime. As shown in this Letter, the superconducting networks provide a powerful and simple example of a tunable frustrated system where some of the main features obtained by Hofstadter can be observed in realistic conditions. Other properties of this spectrum (gaps, nesting properties, etc.) are also relevant in other physical situations like the quantized Hall effect.[13]

Finally, it should be noted that close physics is provided by the magnetic behavior of Josephson-junction arrays. In the mean-field approximation, this problem reduces to ours. However, only one feature at $\phi/\phi_0 = \frac{1}{2}$ has been observed[6] in the resistance of these arrays as function of magnetic field. This feature was predicted theoretically by different authors.[14] This system is very promising for future studies. In fact, introducing disorder[15] is expected to produce a physical realization of the so-called "gauge-glass," where varying the magnetic field is equivalent to jumping from one "spin-glass replica" to another.

We wish to thank Dr. D. Kimhi for providing the mask of the network. We are indebted to P. Brosee-Maron and B. Picot for their expert technical assistance.

[1]G. Toulouse, Commun. Phys. (G.B.) **2**, 115 (1977).

[2]For a recent review, see G. Toulouse in *Heidelberg Colloquium on Spin-Glasses,* edited by J. L. Hemmen, Springer Lecture Notes in Physics, Vol. 192 (Springer-Verlag, Berlin, 1983).

[3]B. Simon, Adv. Appl. Math. **3**, 463 (1982).

[4]W. A. Little and R. Parks, Phys. Rev. A **133**, 97 (1964).

[5]B. Pannetier, J. Chaussy, and R. Rammal, J. Phys (Paris), Lett. **44**, L853 (1983).

[6]M. Tinkham, D. W. Abraham, and C. J. Lobb, Phys. Rev. B **28**, 6578 (1983), and references cited therein; D. Kimhi, F. Leyvraz, and D. Ariosa, Phys. Rev. B **29**, 1487 (1984); R. A. Webb, R. F. Voss, G. Grinstein, and P. M. Horn, Phys. Rev. Lett. **51**, 690 (1983).

[7]S. Alexander, Phys. Rev. B **27**, 1541 (1983); S. Alexander and E. Halevi, J. Phys. (Paris) **44**, 805 (1983).

[8]R. Rammal, T. C. Lubensky, and G. Toulouse, Phys. Rev. B **27**, 2820 (1983).

[9]G. Toulouse, "Frustration, Landau Levels and Superconducting Diamagnetism on Networks" (to be published).

[10]D. F. Hofstadter, Phys. Rev. B **14**, 2239 (1976); see F. H. Claro and G. H. Wannier, Phys. Rev. B **19**, 6068 (1979) for a similar study of the triangular network.

[11]C. de Lange and T. Janssen, J. Phys. C **14**, 5269 (1981).

[12]P. G. de Gennes, *Superconducting of Metals and Alloys* (Benjamin, New York, 1966).

[13]R. Rammal, G. Toulouse, M. T. Jaekel, and B. I. Halperin, Phys. Rev. B **27**, 5142 (1983).

[14]S. Teitel and C. Jayaprakash, Phys. Rev. Lett. **51**, 1999 (1983); W. Y. Shih and D. Stroud, Phys. Rev. B **28**, 6575 (1983).

[15]W. Y. Shih, E. Ebner, and D. Stroud, to be published.