Flux Quantization on Quasicrystalline Networks

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We have measured the superconducting transition temperature $T_c(H)$ as a function of magnetic field for a network of thin aluminum wires arranged in two quasicrystalline arrays, a Fibonacci sequence and the eightfold-symmetric version of a Penrose tiling. The quasicrystals have two periods whose ratio σ is irrational and are constructed of two tiles with irrationally related areas. We find a series of dips in $\delta T_c(H)$ corresponding to favorable arrangements of the flux lattice on the quasicrystalline substrate. The largest dips are found at σ^n and the dips approach the zero-field transition temperature as n increases.

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Flux-quantization experiments on periodic arrays of superconducting elements have shown a wealth of interesting and complex structure.[1,2] When the magnetic field is such that there is an integral number of flux quanta per unit cell, the superconducting transition temperature returns to its zero-field value since at these fields each and every cell satisfies fluxoid quantization[3] with no current through the filaments. For fields less than one flux quantum per cell there are cusps on the phase-boundary curve $T_c(H)$ at every rational field (that is, every rational fraction p/q of a flux quantum, $\Phi_0 = hc/2e$, per unit cell). This is indicative of the flux lattice's finding a commensurate configuration on the array.[4]

Using cells or tiles with relatively irrational areas will preclude the system from satisfying quantization in all cells simultaneously. If the system possesses quasiperiodic translational order, two or more incommensurate periods, this will greatly change the ability of the flux lattice to find a favorable "commensurate" configuration. Quasicrystalline arrays,[5,6] herein taken as quasiperiodic networks with a finite number of elementary tiles, should therefore exhibit phase boundaries which differ greatly from those of periodic (crystalline) arrays. However, quasicrystals are highly ordered structures, and so we would expect considerably more structure in the phase boundary than for a random network.

In this Letter we report on experiments on two patterns that we have generated at the National Research and Resource Facility for Submicron Structures at Cornell. One pattern, Fig. 1 inset, consists of 400 parallel lines equally spaced (2 μm) in the y direction crossed by 400 parallel lines arranged in a Fibonacci sequence[6] in the x direction [the small spacing is 1.45 μm and the large spacing is 2.34 μm, larger by a factor of $\tau = (\sqrt{5}+1)/2$, the golden mean]. The quasiperiodicity of the Fibonacci sequence and the ratio of the number of large to small intervals is also characterized by the number τ. The thickness of the aluminum wires is 500 Å and their width is 0.30 μm. The resistance of the samples is measured with a four-probe ac technique. Typically the width of the superconducting transition as a function of temperature in zero field is 2.5 mK. The sample resistance is stabilized at a fixed fraction of the normal-state value by use of a feedback loop to control the temperature. The magnetic field is then varied and the temperature variation monitored with a calibrated Allen-Bradley resistance thermometer.

The quasicrystalline patterns are intrinsically incommensurate with the flux lattice for any value of the magnetic field, in contrast to periodic patterns which are incommensurate only for irrational fields.[1,4,7] The incommensurability is reflected in the inability of the applied magnetic field fully to satisfy flux (rather than fluxoid) quantization on any subset of tiles which cover the system in a superlattice. One elementary way to see the difference between the periodic and quasicrystalline networks is the attempt to satisfy the quantization of the phase integral[3]

$$\int \nabla\phi \cdot dl = \frac{4\pi}{c\Phi_0}\int \lambda^2 \mathbf{J}\cdot dl + \frac{1}{\Phi_0}\int \mathbf{A}\cdot dl = 2\pi n \qquad (1)$$

with one flux quantum through every elementary tile (area a^2) and a minimal J. By elementary tile we refer to the smallest area enclosed by the wires in the network, a unit cell for the square lattice, either of the two rectangles from which the Fibonacci network can

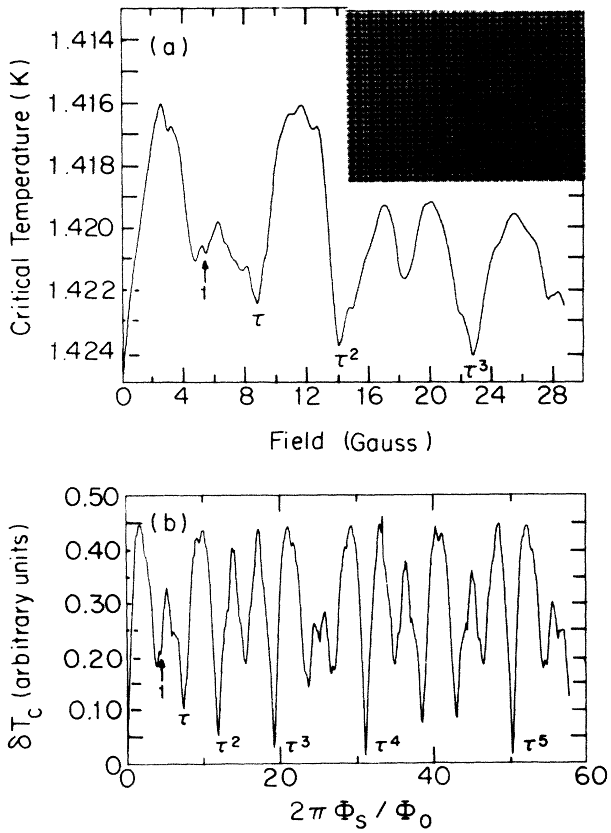

FIG. 1. Inset: Optical micrograph of a "Fibonacci pattern," parallel lines along y and a Fibonacci sequence along x. (a) Superconducting transition temperature vs magnetic field (after subtraction of a quadratic background). (b) Calculated T_c vs H from linearized Ginzburg-Landau theory for the pattern above. Φ_s is the flux through a small tile.

be constructed, or either the square or the rhombus which are the building blocks of the eightfold quasicrystal. In Eq. (1) **A** is the vector potential, λ the penetration depth, **J** the screening currents, n an integer, and ϕ the phase of the superconducting order parameter. For the square lattice one flux quantum per square corresponds to an applied field of Φ_0/a^2 and indeed each tile has the appropriate field to reduce the screening currents to zero, with the result that $\delta T_c \rightarrow 0$. For the Fibonacci sequence one flux quantum in every elementary tile corresponds to an applied field of $H_0 = (\Phi_0/a_s^2)(1+\tau)/(1+\tau^2)$ [total flux $=\Phi_0(n_s + n_l)$; total area $= n_s a_s^2 + n_l a_l^2$, $n_l/n_s = \tau$, $a_l^2/a_s^2 = \tau$; $n_{s,l}$ = number of small, large tiles; $a_{s,l}^2$ = the area of a small, large tile]. The magnetic field through neither of the tiles is a flux quantum and the screening currents for this situation are quite large. The kinetic energy associated with these currents makes the proposed field energetically unfavorable and δT_c remains large.

The measured transition temperture versus magnetic field is shown in Fig. 1(a). A quadratic background due to the finite thickness of the wires has been subtracted. The pattern is clearly not periodic. To get an idea of the correspondence between applied field and flux per tile, we have indicated as $H/H_0 = 1$ the field corresponding to a flux quantum in every tile. The small dip in the phase boundary near this field is shown in Fig. 1(a). As discussed above, this is not a particularly favorable field. There are many cusplike features, as in the periodic case, but not entirely at fields which correspond to rational values of H/H_0. Structure is also observed for $H/H_0 < 1$ indicating the favorable energy of certain flux-lattice configurations on the quasicrystalline substrate. There is also structure at $H/H_0 > 1$ indicating that the irrational areas can approximately satisfy flux quantization when there are several flux quanta in each tile.

The sharpest decreases in δT_c occur at *powers* of τ times a flux quantum per tile, i.e., $H/H_0 = \tau^n$, and δT_c approaches 0 more closely for larger n. Since $\tau^n = F_n + \tau F_{n+1}$ where F_n is the nth Fibonacci number,[6] we see that the strongest dips correspond to the case where successive Fibonacci numbers of flux quanta are found in small and large tiles, approximating the ratio of their areas, τ. If there are N_l flux quanta in every large tile and N_s flux quanta in every small tile then the required applied field or the average field is

$$H(N_s,N_l) = \Phi_0(n_s N_s + n_l N_l)/(n_s a_s^2 + n_l a_l^2)$$
$$= (\Phi_0/a_s^2)(N_s + \tau N_l)/(1+\tau^2).$$

Thus $H/H_0 = 1$, τ, τ^2, and τ^3 correspond to $(N_s,N_l) = (1,1)$, $(1,2)$, $(2,3)$, and $(3,5)$, respectively. (Note that since τ is a quadratic irrational, $p\tau^q$ and p/τ^q belong to the set $n + m\tau$ where m, n, p, and q are integers.)

To treat the problem theoretically we turn to the linearized Ginzburg-Landau equations appropriate near the phase boundary, written for a network with current conservation at the nodes or lattice points[8]:

$$-\Delta_i \textstyle\sum_{ij} \cot(\theta_{ij}) + \sum_{ij} \Delta_j e^{i\gamma_{ij}}/\sin(\theta_{ij}) = 0. \qquad (2)$$

Δ_i is the order parameter at node i, $\theta_{ij} = l_{ij}/\xi_s$ is the distance between nodes divided by the coherence length, and $\gamma_{ij} = \int^j (2e/\hbar c)\mathbf{A}\cdot d\mathbf{l}$. When there is only one value which l_{ij} can take, as in square, triangular, and hexagonal lattices, the $\sin(\theta)$ term cancels in the sums and the problem reduces to that of an electron on a similar lattice in a magnetic field.[1,8] For a particular value of H there is a maximum value of ξ, $\xi_c(H)$, for which there are nonzero solutions to Eq. (1). Since $\xi = \xi_0(\delta T/T_{c0})^{-1/2}$, $\delta T = (T_{c0} - T)$, the superconducting state is restricted to $\delta T/T_{c0} > (\xi/\xi_0)^2$ or $\delta T_c = [\xi_c(H)/\xi_0]^2$. This gives the phase boundary.

For the quasicrystalline case the solution to these network equations is considerably more difficult pre-

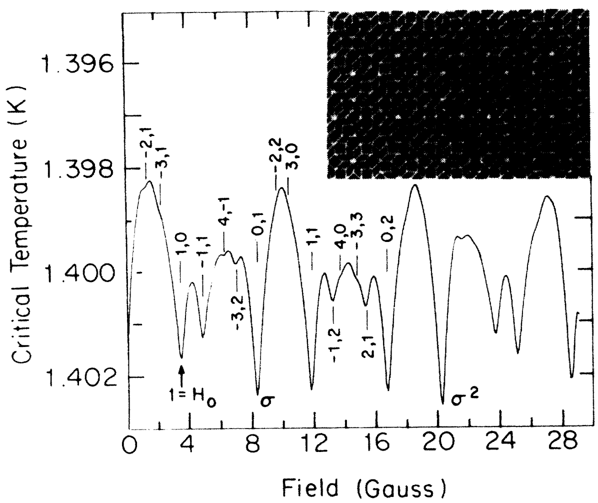

FIG. 2. Superconducting transition temperature vs magnetic field for the pattern above (after subtraction of a quadratic background). Numbers, n,m, indicate the fields $H = H_0(n+m\sigma)$, $\sigma = 1+\sqrt{2}$. Inset: Optical micrograph of an "eightfold Penrose tiling."

cisely as a result of the lack of periodicity. However, some of the structure, especially for $H/H_0 > 1$, can be obtained simply by solution of the Ginzburg-Landau equations on a pair of tiles whose areas are in an irrational ratio. This calculation gives a set of parabolic dips which correspond to the cusps at τ^n in the actual data. Thus the overall behavior may be viewed as a consequence of the irrational ratio of the areas of the tiles. To proceed further we need to solve the network equations on a larger grid. For the Fibonacci pattern, it is possible to solve these equations on rather large grids by using the periodicity in the transverse direction to introduce a Floquet variable which can be discretely varied.[9] The results of this calculation are shown in Fig. 1(b) for a 23×23 grid. The agreement with the experiment is excellent for both the gross and the fine structure.

The use of one periodic direction allows us to perform the calculation in a straightforward way. However, it also complicates the identification of the set of dips at different values of H/H_0. Some are due to periodicity while others are due to quasiperiodicity. For example, the dip at $H/H_0 = \frac{1}{2}$ in Fig. 1(a) corresponds to alternating rows in the periodic direction with large tiles occupied in one row and small tiles in the next. The deeper dip just preceding the one marked $H/H_0 = 1$ in Fig. 1(a) corresponds to $H = \Phi_0/a_l^2 = \Phi_0/\tau a_s^2$, the field which allows a flux quantum in an individual large tile. Although it is not apparent on the scale shown in the figure, an expanded study of this region both experimental and calculated shows that this dip is quadratic while the dips at $H = H_0$ and most other fields are cusplike. It therefore results largely from a single-tile effect rather than the coherence of the network. The calculated curve, Fig. 1(b), is shown on a compressed field scale to emphasize another unusual property of the phase boundary: the "quasireflection symmetry" about the fields $(\tau^n + \tau^{n+1})/2$, extending from 0 to $\tau^n + \tau^{n+1}$, and becoming more nearly perfect as n increases.

The second pattern studied (Fig. 2) was the eightfold-symmetric analog of the Penrose tilings (~20 000 elementary tiles were used). It is a two-dimensional quasicrystal with no simple periodicity. We were unable to produce a Penrose tiling because of restrictions in the microfabrication apparatus. The eightfold pattern was mathematically generated by an inflation scheme.[10] In the conventional Penrose tiling the ratio of the areas of the tiles, the ratio of the number of large to small tiles, and the "quasiperiodicity ratio" (the ratio of basic frequencies in the diffraction pattern, or the length-scale change associated with each inflation) are all characterized by the same irrational number, τ. In the present case the ratio of the areas is $\sqrt{2} = \Sigma$, there are more small tiles than big ones by the ratio Σ, while the quasiperiodicity is characterized by $\sigma \equiv \sqrt{2}+1$. Note, however, that σ and Σ belong to the same system of quadratic irrationals (i.e., Σ is a member of $n+m\sigma$). The lack of any periodic direction precludes the one-dimensionalization of the network equations with a Floquet factor.

The experimental $T_c(H)$ is shown in Fig. 2 with a quadratic background subtracted. The apparent decrease of $T_c(H)$ below $T_c(0)$ for some fields is due to the inadequacy of the quadratic subtraction for a network with some variation in the width of the wires. $\delta T_c(H)$ for the two-dimensional pattern bears a great deal of resemblance to that of the Fibonacci pattern. In particular, there are still sharp cusps with the greatest reduction in δT_c found at σ^n, a lack of periodicity, and a quasisymmetry about the midpoints of the cusps at σ^n and σ^{n+1}. The magnetic field which corresponds to one flux quantum in each elementary tile $H_0 = (1+\sqrt{2})\Phi_0/2a_s^2$ is again indicated by the arrow and does not lie at a low value of δT_c. The applied field for N_l (N_s) flux quanta in every large (small) tile is $H(N_l,N_s) = (\Phi_0/2a_s^2)(N_l + \sqrt{2}N_s)$. Thus the arrangements of the flux quanta in the different tiles is $(N_l,N_s) = (1,1)$, (3,2), and (7,5) for $H/H_0 = 1$, σ, and σ^2, respectively. Note that for these increasingly favorable configurations the ratios N_l/N_s are successive rational approximants to Σ (the relative areas) in a continued-fraction expansion, again similar to the "Fibonacci pattern" (where the continued-fraction expansion is given by the ratio of successive Fibonacci numbers). At low field there is structure which shows that the flux lattice can find energetically favorable arrangements on a quasicrystalline substrate. In Fig. 2

we have also indicated some of the fields corresponding to $n + m\sigma$ for $|n|, |m| < 4$ which describes the positions of most of the fine structure seen.

We have performed experiments on a number of other networks which are interesting in relation to those described above. One of the patterns consists of equally spaced parallel lines along y and lines along x which are spaced with either a or τa and with the same ratio of large to small tiles as in the Fibonacci sequence, but with their order randomized. The phase boundary $\delta T_c(H)$ for this pattern resembles that of the Fibonacci pattern (Fig. 1) at large fields where the major dips occur at fields corresponding to $H/H_0 \sim \tau^n$, and at low field resembles a superposition of independent tiles with area ratio τ. The dips appear more "quadratic" than cusplike and there is a complete lack of fine structure particularly evident in the low-field region $H/H_0 < \tau$. Thus the flux lattice cannot find a favorable arrangement on the randomized pattern, but the discreteness of the areas gives the lowering of δTc for rational approximants to an integral number of flux quanta per cell.

Another pattern consists of equal spacing of lines along y and a superposition of two incommensurate periodically spaced sets of lines along x. The result is a quasiperiodic set of tiles with a large number of different areas from a maximum given by the smallest periodic spacing to a minimum of zero. The phase boundary for this array shows cusplike structures, but they never reduce δTc significantly toward zero. The flux lines can find a configuration on the quasiperiodic lattice which lowers the energy but the wide variety of areas prevents any semblance of satisfying an integral number of flux quanta per cell with low kinetic energy from the screening currents. Hence δT_c remains large.

In conclusion we have measured the transition temperature versus magnetic field for a set of quasicrystalline networks. We find a set of cusplike structures reminiscent of those found in periodic systems. The major dips are found at $\sigma^n H_0$ (where σ characterizes the irrationality of the periods in the quasicrystal and H_0 the field for a flux quantum in each tile) rather than at nH_0 (for the periodic case). The fine structure observed at "rational fields" $(n/m)H_0$ in the periodic case are replaced by fine structure at $(m+n\sigma)H_0$ in the quasiperiodic case. The flux lattice can find favorable arrangements which lower its energy even on the incommensurate substrate. Thus there are preferred configurations, similar to epitaxy, on a quasicrystal.

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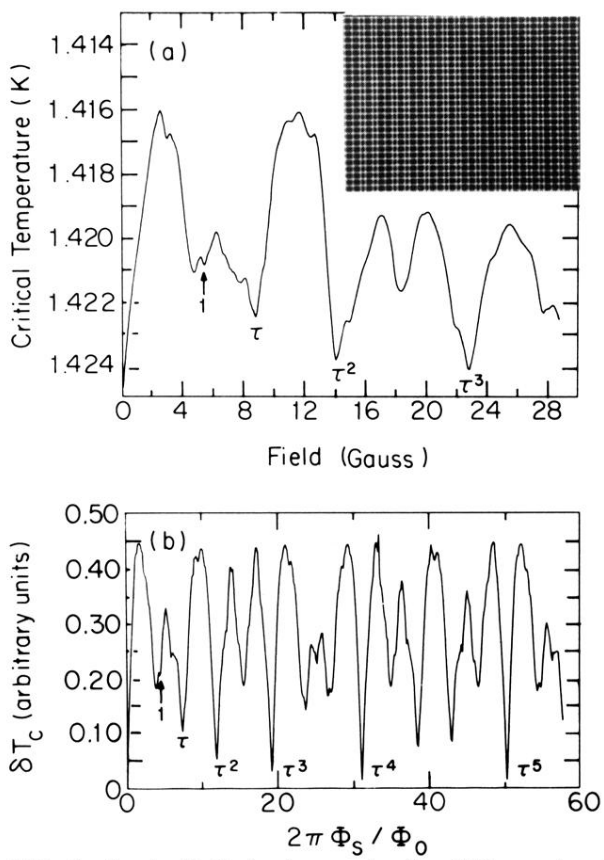

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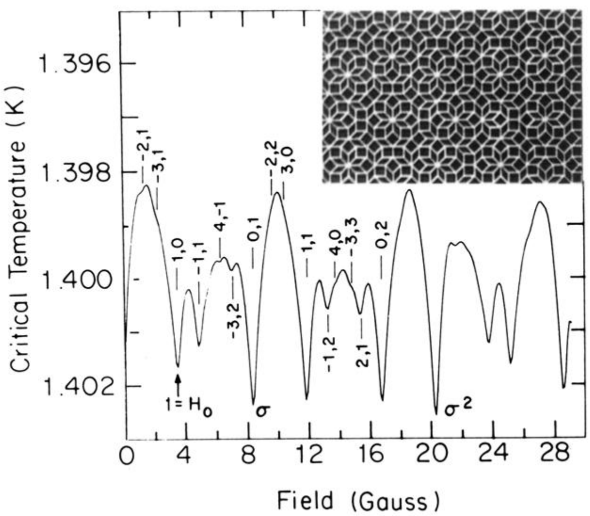

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