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Experimental determination of the (H, T) phase diagram of a superconducting network

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Résumé. — Nous avons mesuré la température critique résistive T_c d'un réseau régulier à deux dimensions en nid d'abeilles d'indium supraconducteur sous faible champ magnétique. Nous observons des oscillations de T_c de période égale à un quantum de flux ($\phi_0 = hc/2e$) par cellule du réseau. Des structures bien définies apparaissent aussi aux valeurs rationnelles du flux réduit $\phi/\phi_0 = 1/3$ et 2/3. Ces résultats sont comparés aux prédictions théoriques pour la ligne critique (H, T) du réseau étudié.

Abstract. — We have measured the resistive critical temperature T_c of a regular two dimensional network of superconducting indium at low magnetic fields. We find T_c oscillations with a period equal to one quantum flux $\phi_0 = hc/2e$ per lattice cell. Well-defined structures also appear at the rational values of the reduced flux $\phi/\phi_0 = 1/3$ and 2/3. These results are compared with theoretical predictions for the (H, T) critical line of the studied network.

The attractive properties of superconducting networks in a magnetic field have stimulated extensive theoretical studies in the last two years. The magnetic properties of such networks are expected to be very sensitive to the topology, and particularly to the connectedness of the multiply connected structure. The networks considered fall in three categories : simple finite geometries [1-3], infinite regular lattices [4, 5] and disordered structures [4, 6]. Up to now only two extreme cases have been investigated experimentally. The first corresponds to the single loop geometry considered in the original experiment of Little and Parks [7]. The second is given by the recent study of granular superconductors by Deutscher et al. [8], and represents the disordered structure situation. Besides these two extreme situations, extended periodic networks have not received much attention in the past, despite of the richness of the theoretically predicted phase diagram [4, 5]. Josephson junction arrays are expected to bear the same physics, but seem to involve more complicated behaviours [9]. One of the attractive properties of regular networks is the occurrence of dips predicted theoretically in the upper critical field $H_c(T)$. Profound depressions are expected at rational values of the reduced flux ϕ/ϕ_0 which correspond to flux quantization phenomena in these networks, and regular organization of vortices inside the structure ($\phi_0 = hc/2e$ is the quantum flux). In this letter, we report an experimental and theoretical study of the critical line $H_{c}(T)$ for a two dimensional honeycomb network made of submicron indium wires. Our experimental results demonstrate the above flux quantization phenomenon as predicted by the solution of linearized Ginzburg-Landau equations.

Samples were prepared by thermal evaporation of pure indium (5 N grade) film onto the polished surface of a microchannel glass plate. The plate is made with a lattice of close-packed parallel

Nº 20

hollow glass fibres. The studied network is simply obtained by direct deposition of the metal at the edge of the glass walls. We have investigated networks of indium wires, 1 000 Å thick, of width ~ 4 000 Å (as defined by the glass wall thickness) obtained by this method on a honeycomb lattice of period ~ 2.4 µm. For measurements, electrical contacts were made by evaporation of thick indium pads through a metal mask which defined the overall size of the network : length 500 µm, width 430 µm. The studied network contains ~ 200 × 150 elementary hexagonal loops. Resistance measurements were performed by using a 4-probe bridge with typical bias current of 4 µA. The normal resistance at 4.2 K is $R_n = 0.67 \Omega$.

The upper critical line $H_c(T)$ obtained directly from a X-Y recorder is shown in figure 1. In this measurement, the temperature of the sample was monitored in order to keep the resistance of the indium network at a constant value (0.2 Ω). The horizontal signal is the temperature measured by a carbon resistor [10] corresponding to a scanning of the magnetic field axis. As can be seen, a non monotonic phase boundary is observed, with a succession of cusps appearing at periodic values of the magnetic field. This modulation, of period $\Delta H = 3.62$ Oe corresponds to one quantum flux ϕ_0 per elementary hexagonal loop in the network. This modulation reflects the effect of one loop quantization phenomena similar to that of Little and Parks [7]. The enclosed area inside elementary loops, estimated from this magnetic period, is found 5.72 μ m² and corresponds to hexagonal cells of side $a = 1.48 \mu$ m. This value is closed to that extracted from S.E.M. micrographs : $a = 1.4 \pm 0.2 \mu$ m. The envelope of the observed modulation represents the change of the critical field of the strands, varying as $H_{c_0} \lambda/d \sim (T_0 - T)^{1/2}$. Here H_{c_0} denotes the thermodynamical critical field, λ is the penetration depth, d the width of the strands and T_0 the bulk critical temperature of indium at zero field.

The data shown in figure 1 call for the following comments, all reflecting a network behaviour. For this it is useful to compare the shape of the observed modulation with that of the one-loop-geometry shown in the insert and obtained for the same material. First of all, the initial slope dH/dT at zero field is finite for the network and infinite for the one-loop-geometry. Secondly, the



Fig. 1. — Critical line (H, T) of a regular honeycomb network of superconducting indium, defined as explained in the text. The arrows indicate the value of the magnetic field corresponding to one quantum flux $\phi_0 = hc/2e$ per elementary loop of the lattice. For comparison, the insert shows the critical line for an indium hollow cylinder of 1 000 Å thickness evaporated on a 3.3 µm diameter quartz fibre. Comparison of absolute values of the temperatures is meaningless [10].

observed cusps at *half integer* values of the reduced flux ϕ/ϕ_0 in the one-loop-geometry disappear in the network case. New cusps occurring at *integer* values of ϕ/ϕ_0 take place in the latter case. Note, in addition, the inversion of the concavity of the modulation. Finally, new well defined structures (see Fig. 2) appear in the network geometry and were absent in the one-loop case. All these features show a collective behaviour, as will be shown theoretically below, in the network geometry.



Fig. 2. — Plot of the resistance R as function of the applied magnetic field in the range $0 \le \phi \le \phi_0$ close to the bulk critical temperature T_0 . In addition to the fundamental dip at $\phi = \phi_0$, the secondary dips at $\phi = \phi_0/3$ and $2\phi_0/3$ are indicated by the corresponding arrows. Less marked dips are also observed at other well defined value of the reduced flux. The insert shows the honeycomb structure.

In order to study the new structures on the $H_c(T)$ line, a magnified view of the first period is shown in figure 2. In this figure, we have reported directly the measured resistance R as function of the magnetic field at fixed temperature. The width of the superconducting transition in zero field is larger ($\sim 10^{-2}$ K) than the amplitude of the T_c oscillations. Therefore, this curve is equivalent of the $H_c(T)$ plot of figure 1 as far as R(T) remains linear, which is strictly true in the region of our interest. This procedure provides an improved resolution, up to $\sim 10^{-4}$ K, well sufficient to distinguish the relevant physical features. One of the salient features seen in figure 2 is the occurrence of well defined dips, particularly at $\phi/\phi_0 = 1/3$ and 2/3 symmetrically located around $\phi/\phi_0 = 1/2$. Furthermore, other reproductible well defined structures appear also at other values of ϕ/ϕ_0 , and will be discussed more extensively in a forthcoming paper [11]. Note however that the expected dip at $\phi/\phi_0 = 1/2$ is not seen in figure 2. From the modulation amplitude we can estimate [11] the value of the coherence length $\zeta_s(T = 0)$. The obtained value is $\simeq 1000$ Å which is reasonable value for thin superconducting indium.

The basic concepts of treating superconducting network near the second order phase boundary go back to Alexander [4] and de Gennes [1]. The linearized Ginzburg-Landau equation leads in general to an eigenvalue problem which is best expressed in terms of the order parameter values at the nodes. If node α is linked to *n* nodes *via* strands of length $L_{\alpha\beta}$ ($\beta = 1$ to *n*), the basic equation, at node α may be written :

$$\sum_{\beta=1}^{n} \left(-\Delta_{\alpha} \operatorname{cotg} \frac{L_{\alpha\beta}}{\xi_{s}} + \Delta_{\beta} \frac{\mathrm{e}^{-i\gamma_{\alpha\beta}}}{\sin \frac{L_{\alpha\beta}}{\xi_{s}}} \right) = 0 \tag{1}$$

where Δ_{β} is the value of the order parameter at node β , and $\gamma_{\alpha\beta} = \frac{2\pi}{\phi_0} \int_{\alpha}^{\beta} \mathbf{A} \, d\mathbf{l}$ is the circulation of the vector potential along the strand linking α and β .

For a regular lattice (square, triangular, honeycomb, ...), where $L_{\alpha\beta} \equiv a$ is the same for all

$$\left(z\cos\frac{a}{\xi_{\rm s}}\right)\Delta_{\alpha} = \sum_{\beta}\Delta_{\beta}\,{\rm e}^{-i\gamma_{\alpha\beta}} \tag{2}$$

where z denotes the coordination of the lattice. Thus, the problem reduces to that of Landau level structure of a free-electron gas in the same geometry. The energy of Bloch electrons is given by $\varepsilon = z \cos \frac{a}{\xi_s}$. The spectrum associated with equation 2 has been studied in great detail [12] for the square and triangular lattices. As pointed out by Alexander [4], the upper critical field of the superconducting network is given by the band edge of the spectrum of equation 2. For a honeycomb lattice (z = 3), there are no eigenvalues outside the range : $-3 \le \varepsilon \le 3$, and the spectrum shows invariance properties in ε and $\gamma = 2 \pi \frac{\phi}{\phi_0}$, where $\phi = H \times 6 \times \frac{a^2 \sqrt{3}}{4}$ is the magnetic

flux through the elementary hexagonal loop of side a.

For small γ we can calculate the position of the band edge by using a continuum approximation. The result is simply :

$$3 - \varepsilon \simeq \frac{\gamma}{2\sqrt{3}}.$$
 (3)

In the limit $\gamma \ll 1$, the corrections to (3) are exponentially small and

$$H_{c_2} \simeq \frac{\phi_0}{\pi \xi_s^2} \tag{4}$$

which is valid only at $a/\xi_s \ll 1$, i.e. continuum result.

The whole spectrum of (2) for a honeycomb lattice can be calculated following a procedure similar to that used for the square and the triangular lattices. For convenience we choose a Landau gauge

$$A_{\rm x} = 0, \quad A_{\rm z} = 0, \quad A_{\rm y} = H_{\rm x}.$$
 (5)

The elementary cell contains two sites A and B and two equations 2 are to be considered. A rather cumbersome algebra, to be reported elsewhere [13], leads finally to the following difference equation :

$$(\varepsilon^{2} - 3)\psi_{i} = 2\cos\left[\frac{\gamma}{2}\left(i + \frac{1}{2}\right) + \alpha\right]\psi_{i+1} + 2\cos\left[\frac{\gamma}{2}\left(i - \frac{1}{2}\right) + \alpha\right]\psi_{i-1} + 2\cos\left[\gamma\left(i + \frac{1}{2}\right) + 2\alpha\right]\psi_{i} \quad (6)$$

where α denotes a Floquet factor. The energy spectrum of equation 6 is quite complex, and the rationality of ϕ/ϕ_0 plays an essential rôle.

The calculation can only be performed for rational ϕ/ϕ_0 . For $\phi/\phi_0 = p/q$ (p, q integers prime to each other), (6) reduces to q independent linear equations.

strands, equation 1 reduces to

Nº 20

In general, the associated determinantal equation takes the following form :

$$P_a(\lambda) + W = 0 \tag{7}$$

where W takes its values in [-3, 6] and $P_q(\lambda)$ denotes a polynomial of degree q in $\lambda = \varepsilon^2 - 3$.

In figure 3, we have shown only the band edge of the spectrum, corresponding to values of q up to q = 15. As can be seen, this graph displays the same striking pattern, as for the square and triangular lattices. At low field values, we recover the continuum limit result (Eq. 3) as expected. The irregular line so obtained shows pronounced dips at $\phi/\phi_0 = 1/2$, 1/3, 2/5, ... etc. and must be compared with figure 2.



Fig. 3. — Calculated values of the reduced variable $\varepsilon = 3 \cos a/\xi_s$ as function of the reduced flux ϕ/ϕ_0 . Here *a* denotes the length of a side of the hexagonal cell and ξ_s is the temperature dependent superconducting coherence length. Different points correspond to rational values of $\phi/\phi_0 = p/q$ with $q \le 15$. Noticeable dips appear at $\phi/\phi_0 = 1/3$, 2/5, 1/2, 3/5, 2/3, ... etc... As can be seen the more pronounced depressions are at 1/3 and 2/3. For $a/\xi_s \le 1$ this curve represents the variation of $\Delta T_c/T_0$ and must be compared with figure 2.

Other less marked dips occur also at other rational values of ϕ/ϕ_0 . In contrast with the square lattice case [12], the more pronounced dips occur at $\phi/\phi_0 = 1/3$, 2/3 instead of $\phi/\phi_0 = 1/2$. This peculiar behaviour is intimately related to the topology of the honeycomb lattice and compares very well to the experimental result. The origin of this behaviour is associated to the nature of the dual lattice in each case. Rational values of the reduced flux ϕ/ϕ_0 correspond to a regular organization of vortices on the dual lattice; and are responsible for the lowering of the magnetic energy. The dual of the honeycomb (i.e., triangular lattice) is more frustrating than that (i.e., square) of the square lattice.

In conclusion, we have reported in this letter the first experimental study of the (H, T) phase diagram of a superconducting network. Flux quantization at integer values of ϕ/ϕ_0 has been observed, as expected. In addition, other quantizations have been discovered at the rational values $\phi/\phi_0 = 1/3$ and 2/3 in agreement with the theoretical analysis. Quantization at other rational values of the reduced flux such as 1/2, 2/5, 3/5, ... are predicted to produce weaker depression of the line (H, T) and deserve more accurate measurements, now in progress.

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