Penetration of a magnetic field into a regular 3D array of superconductor nanoparticles

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An oscillatory component of the critical current and the resistance as a function of the magnetic field has been detected in a regular 3D array of close-packed, weakly linked superconductor nanoparticles. The observed oscillation spectrum is interpreted as being related to a quantization of magnetic flux in a system of loops which can be distinguished in such a lattice.

A 3D ensemble of small superconductor nanoparticles is a material whose properties are a composite of the properties of the individual structural elements of the spatial network. These elements may be individual particles, weak links between particles, and closed loops which the particles form in the lattice. The properties of inhomogeneous superconductors whose dimensions are comparable to the coherence length oscillate as the external magnetic field is varied, as a result of a quantization of the magnetic field penetrating into them. These oscillations are manifested in the properties of both small, doubly connected entities (Little–Parks oscillations[1,2]) and Josephson junctions.[3] In disordered 3D ensembles of superconductor particles (granular superconductors), oscillations of this sort are smoothed out when an average is taken over the entire ensemble. In the case of ordered systems, in contrast, we would expect the properties of the entire ensemble as a whole to oscillate.

In this letter we are reporting a study of the behavior of the critical current I_c and the resistance $\Delta R/R$ near the superconducting transition temperature as a function of the magnetic field for 3D regular arrays of superconductor nanoparticles. The arrays were fabricated by incorporating metal in the cavities of an artificial, ordered, insulating matrix simulating the structure of a noble opal. The characteristics of the structure and the physical properties of the system are described in Refs. 4 and 5. We will refer to this material below as O–Me, where Me is Sn or In.

The electrical properties of the O–Me samples were measured by a four-contact method by the procedure described in Ref. 6. The rectangular samples had typical dimensions of $3\times1\times0.5$ cm and were equipped with four Ag contacts, which were grown electrochemically on one of the wide faces of the sample.

Figure 1 shows a typical plot of $I_c(H)$. The critical current falls off monotonically with increasing magnetic field, $I_c \propto 1/H$. Superimposed on the monotonic component is a component whose amplitude varies with the field. To determine the nature of the observed oscillations, we plotted the positions of the extrema of the curves, found by interpolating the experimental points (the solid curves in Figs. 1a and 1b), versus the extremum number. Figure 1b shows a broken curve consisting of several regions with progressively increasing slopes. The reason for this behavior is that there are discrete

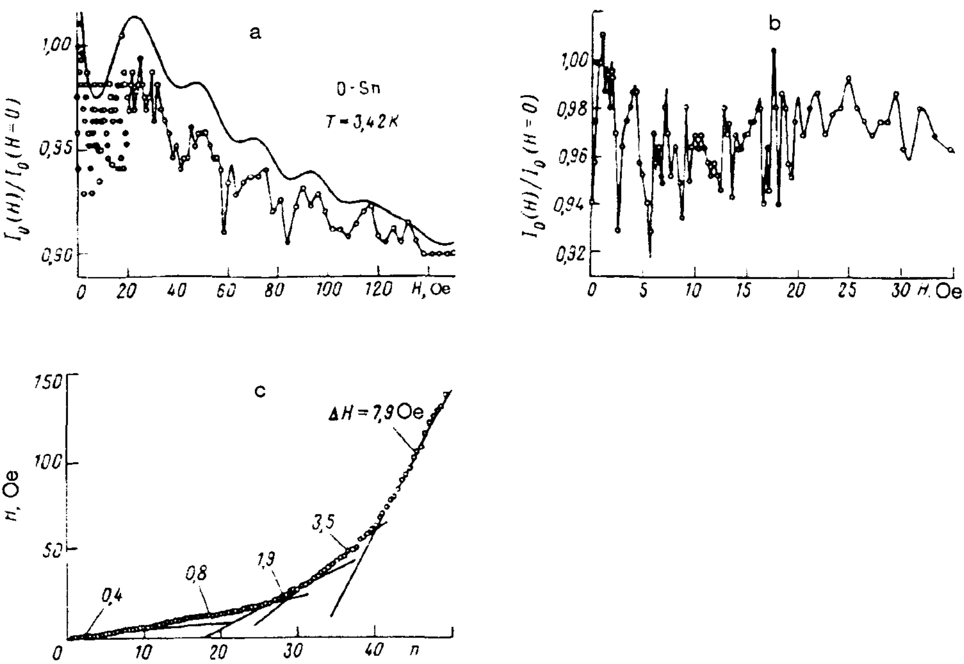

FIG. 1. a: Critical current versus the magnetic field. The solid curve drawn through the experimental points was found by an interpolation of these points. The upper curve was found by smoothing out the small-scale oscillations and shifting upward (for clarity). b: The same, in the weak-field region. c: Positions of the extrema, $I_c(H)$, versus the extremum number. The numbers are the oscillation periods.

changes in the $I_c(H)$ measurement step with increasing field. It thus becomes possible to see both small and large oscillation periods, which correspond to the slope of the linear regions of this dependence. The values of these periods are shown in Fig. 1c. In addition, the $I_c(H)$ curve is modulated by an oscillation with a period of 22 Oe. This modulation is clearly seen when the small-scale oscillations are smoothed out (Fig. 1a). The values of ΔH change slightly with the procedure used to interpolate $I_c(H)$, but the spectrum remains discrete. Consequently, the variable component of $I_c(H)$ can be represented as a superposition of several oscillations which are periodic in the field.

Figure 2 shows the magnetoresistance of an O–In sample near the superconducting transition temperature T_c. This is a complicated dependence, because of the simultaneous operation of several processes, e.g., a suppression of superconducting ordering (a positive magnetoresistance near $H=0$), a suppression of the resistive anomaly at the transition edge (a negative magnetoresistance), and the superposition of an oscillatory component. The first two of these features were discussed in Ref. 6. Since the density of measurement points does not change during a scan of the field in this case, we determined the oscillation spectrum by subtracting "supports" with a progressively decreasing degree of smoothness from the experimental curve. It turns out that the spectrum is of the same nature as in the case of $I_c(H)$. With decreasing measurement step, oscillations with

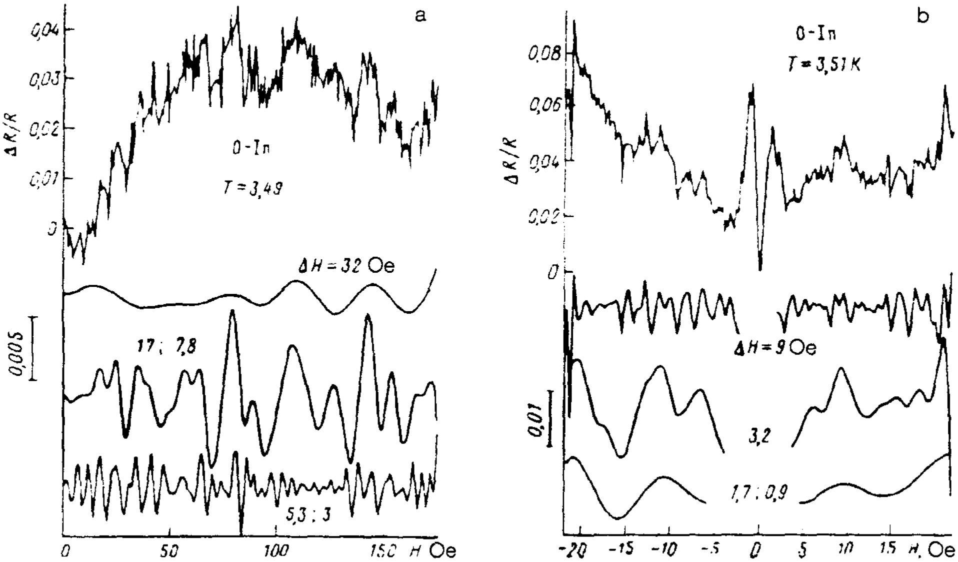

FIG. 2. a—Magnetoresistance of the material in the region near the superconducting transition (upper curve) and extraction of oscillatory behavior from it; b—the same, in the low-field region.

a smaller period arise; they can be seen in Fig. 2b. Below, we treat approximately equal values of the oscillation periods (7.8 and 9 Oe; and also 3 and 3.2 Oe) in Fig. 2 as being identical within the experimental error.

The oscillatory behavior of the magnetoresistance of the materials at low temperatures stems from an interference of the wave functions of the carriers, which have acquired phase increments during the penetration of flux quanta into the contours around which they flow.[7] We can cite several arguments which can be used to determine the mechanism responsible for the oscillatory behavior of this system: 1) The 3D lattice can be thought of as a set of closed contours. 2) The similarity of the oscillation spectra of two quite different characteristics, the critical supercurrent being one, suggests that the oscillations are associated with the superconductivity of the system. 3) The thickness of the generatrix of the contour is smaller than the magnetic-field penetration depth. Furthermore, at least two weak links are connected in each branch of the contour, so a weak field penetrates the O–Me sample. 4) The possible quantization of the flux in large-perimeter contours (at $T \simeq T_c$) and the large number of contours in the lattice make it necessary to deal with the intrinsic inductance of the contours. Under these conditions, the most probable mechanism for the observed oscillations is a quantization of a fluxoid in closed contours, which can be identified in the multiply connected 3D array of superconductor nanoparticles. When an external magnetic field is imposed, circular screening currents arise in the contours in the lattice. The magnitude and direction of these currents are such that they maintain an integer number (N) of flux quanta in each contour. As soon as the current (the transport current plus the screening current) flowing through the Ith

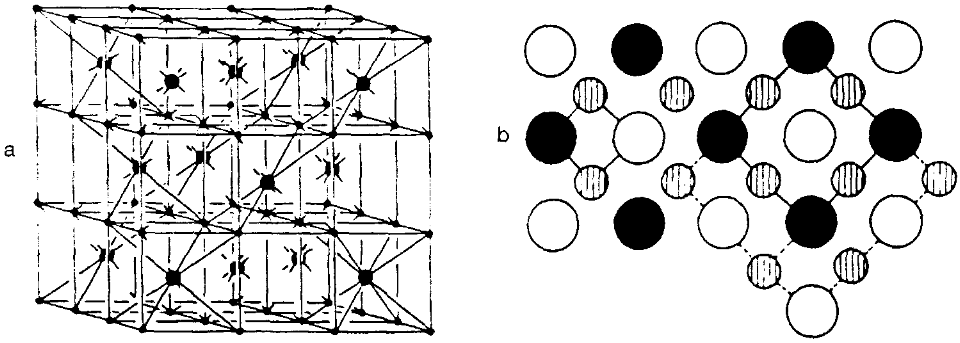

FIG. 3. a: Schematic diagram of the spatial lattice of the particles of a superconductor in an opal matrix (from Ref. 4). Large circles—Particles $0.41D$ in size; small circles—$0.23D$. b: Smallest possible contours in projection onto the {100} plane. The shading of a circle indicates the plane it belongs to. The contour of area D^2 is supplemented with a dashed line to a contour of area $1.5D^2$, and by a dotted line to $(1.5)^2D^2$.

individual weak link exceeds the critical value I_{c1}, the contour switches to a state with $N+m$ flux quanta (N and m are integers). Since the screening current periodically changes in magnitude and sign during the scanning along the field, the critical current and the measured sample resistance also oscillate.

Let us look at the structure of the material. This nanocomposite is a regular fcc packing of identical silicate balls of diameter D (the typical values of D are from 100 to 500 nm, with a scatter of less than 5% within a sample). A superconductor fills the cavities between these balls. The fcc array of balls consists of three plane layers. The upper of these layers lies in the depressions between the balls of the lower layer. Accordingly, there is no closed circumvention path in the lattice of cavities within any packing plane. Such a path would include at least two planes of the packing of balls. According to the geometry of the cavities, the superconducting component of the composite is an ensemble of alternating nanoparticles with sizes of $0.41D$ and $0.23D$, which are closely packed in an fcc lattice (with a density of about 10^{14} cm^{-3}) and which are separated by a distance of $0.61D$. Figure 3a shows the particle coordination scheme[4] in O–Me. The two main types of closed paths in this lattice are shown in Fig. 3b, in projection onto the {100} plane. By virtue of the translational symmetry, we can construct from them lattices which cover the entire plane. The areas of these contours are $D^2/4$ and D^2. The periods of the oscillations corresponding to these areas are, for $D=263$ nm, 1200 and 300 Oe. These figures go outside the range of the magnetic fields used.

If we compare several of the periods in Fig. 2 on the basis of the relation $\Delta H\cdot S=\Phi_0$ (where Φ_0 is the flux quantum) with the values of the area S, we find a sequence of numbers which differ by a factor of $3^{k/2}$ with a scatter of less than 10%, which is no worse than the error in the determination of the oscillation spectrum. Assuming the minimum of $\Delta R/R(H)$ near 150 Oe on the curve in Fig. 3a to be half the period of the quantization over a contour of area D^2, we find $\Delta H=32$ Oe. This result corresponds to a contour area of $3^{5/2}D^2$. The penetration of magnetic flux into O–Me can thus be thought of as a multistep process: When the external magnetic field is imposed, the magnetic flux induces circulating currents in contours with a large perimeter, and the properties of the material oscillate with a short period with increasing field. Screening currents are induced in smaller nested contours, and oscillations are manifested with a large period in the field and so forth, down to the smallest possible contour. The resulting oscillation spectrum is associated with a quantization of magnetic flux in a 3D regular lattice which is a multilevel lattice in terms of the size of the contours.

One reason for the small oscillation amplitude is the macroscopic structure of the samples. These samples are textured polycrystalline samples, whose crystallites are disoriented with respect to the direction of the magnetic field. A second reason is that the perimeter of the quantization contours is significant in comparison with the coherence length.[7]

The complex dynamics of the magnetic flux in a regular 3D lattice leads to several unusual properties of this O–Me material. These properties have been seen previously but not explained adequately.[5,8] There are also such effects as an increase in the critical current on the backward branch of the current–voltage characteristic in comparison with that on the forward branch, an increase in the absolute value of the critical current with a local decrease in the cross section of the sample, a decrease in the effect on the critical current of the low-frequency ac magnetic field with increasing frequency of this field, and the shape of the current–voltage characteristic, among others. Explaining the dynamic properties of the regular array of nanoparticles will apparently benefit from a consideration of the analogy between magnetic vortices in type-II superconductors and circulating currents in this system. In this case the motion of vortices corresponds to a jump of a circulating current from contour to contour, and an increase in the density of the vortex lattice with increasing field corresponds to the induction of such currents in contours of progressively smaller size. The time at which the quasivortex lattice begins to move (the critical current of the system and its hysteresis) depends on the magnitude of the trapped flux, i.e., on the particular level in terms of the dimensions of the contours at which the system of circulating currents is under the given conditions. The particular features associated with the motion of the magnetic flux are attributable to the commensurability of the lattice of circulating currents with pinning centers, in which the silicate matrix plays a role.

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[1] W. A. Little and R. D. Parks, Phys. Rev. Lett. **9**, 9 (1962).
[2] V. V. Moshchalkov *et al.*, Phys. Scr. T **45**, 226 (1992).
[3] A. Barone and G. Paternò, *Physics and Application of the Josephson Effect* (Wiley-Interscience, New York, 1981)
[4] V. G. Balakirev *et al.*, Kristallografiya **38**, 371 (1993).
[5] V. N. Bogomolov *et al.*, Phys. C **208**, 371 (1993).
[6] S. G. Romanov *et al.*, Fiz. Tekh. Poluprovodn. **58**, 883 (1993) [Sov. Phys. Semicond. **58**, 824 (1993)].
[7] A. G. Aronov and Yu. V. Sharvin, Rev. Mod. Phys. **59**, 755 (1987).
[8] V. N. Bogomolov *et al.*, JETP Lett. **36**, 365 (1982).

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