ON THE NATURE OF THE CRITICAL CURRENTS IN A SUPERCONDUCTING SURFACE LAYER

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An induction technique was used to measure critical surface currents in niobium superconductors having different degrees of purity, in fields $H_{c2} < H < H_{c3}$. In a very pure sample (having the resistance ratio 14 000) the critical current was negligible (< 0.002 A/cm). The proposed explanation is that under the influence of an arbitrarily small normal magnetic field component the surface layer undergoes a transition to the vortical state. Data are presented on the angular dependence of the critical current and of the field H_{c3}, along with temperature measurements.

A type II superconductor placed in an increasing magnetic field H experiences several successive phase transitions. At the point H_{c1} a second order phase transition is observed; in the bulk of the sample there arises a triangular lattice of quantized vortices with cores that are in the normal state.[1] At the point H_{c2} the entire bulk enters the normal state. However, a surface layer whose thickness is of the order of the coherence length remains superconductive up to $H_{c3} = 1.69\, H_{c2}$.[2] According to the conceptions prevailing at the present time the order parameter is of uniform structure in the surface layer, where the critical point is finite and can be calculated.[3-7] These properties of the surface layer were deduced while assuming a magnetic field that is strictly parallel to the superconductor-vacuum interface. In any actual experiment, however, a transverse component of the magnetic field always exists because of inaccurate orientation of the sample, imperfections of the sample's shape, magnetic field inhomogeneities etc. Therefore the magnetic field always forms a finite angle with the surface.

In the present work the existence of this angle has been taken into account and it has been shown that an inclined magnetic field converts a uniform order parameter of a surface layer into a periodic parameter. This effect has been confirmed by experiments showing that finite currents are observed in the surface layer only when impurities or other lattice defects are present.

1. SURFACE SUPERCONDUCTIVITY IN AN INCLINED MAGNETIC FIELD

Let a field H form an angle θ with the plane of the sample. We now face the customary problem of determining the highest field $H_{c3}(\theta)$ in which the linearized Ginzburg-Landau equation for the order parameter,[8]

$$\left(\frac{i\nabla}{\varkappa}+\mathbf{A}\right)^2\psi=\psi,$$

$$\mathbf{n}\left(\frac{i\nabla}{\varkappa}+\mathbf{A}\right)\psi=0 \quad \text{(on the surface of the sample),} \qquad (1)$$

possesses a nontrivial solution.

We arrange the Cartesian coordinates xyz in such a way that the yz plane coincides with the plane of the sample ($x \geq 0$) and the field **H** lies in the xz plane. The vector potential **A** is given the convenient form

$$\mathbf{A}=(0,\ Hx\cos\theta - Hz\sin\theta,\ 0). \qquad (2)$$

Since our problem is invariant along the y axis, we shall seek a solution in the form

$$\psi(x, y, z) = e^{iky}\psi(x, z).$$

Therefore

$$-\frac{\partial^2\psi}{\partial\xi^2}-\frac{\partial^2\psi}{\partial\eta^2}+(\xi\cos\theta-\eta\sin\theta-x_0)^2\psi=\varepsilon\psi,$$

$$\left.\frac{\partial\psi}{\partial\xi}\right|_{\xi=0}=0, \qquad (3)$$

where

$$\xi=\sqrt{\varkappa H}x,\quad \eta=\sqrt{\varkappa H}z,\quad x_0=k/\sqrt{\varkappa H},\quad \varepsilon=\varkappa/H. \qquad (3')$$

The highest value of the field, $H = H_{c3}(\theta)$, corresponds to the smallest eigenvalue $\epsilon_0(\theta)$ of Eqs. (3) and (3′). Equation (3) immediately reveals an important difference between the solutions for $\theta = 0$ and $\theta \neq 0$. In the first case the solution $\psi(x)$ is independent of z and the eigenvalue ϵ_0 is a function of a free parameter x_0 (i.e., k). Saint-James and de Gennes[2] have found by means of numerical integration that the minimal value $\epsilon_0 \approx 0.59$ occurs when $x_0 \approx 0.77$.[1)]

When $\theta \neq 0$ the eigenvalue ϵ_0 is entirely independent of x_0 (k degeneracy) and the solution for ψ falls off at large values of x and z. The presence of the degeneracy shows that all points of the surface are equivalent for the formation of a single nucleus.

Since the problem cannot be solved exactly even for $\theta = 0$, we shall utilize the approximation method developed by Schmidt[9] for $\theta = 0$. This method is based on the fact that the exact value $x_0 = 0.77$ is smaller than unity and can be used in calculating ϵ_0 by means of ordinary perturbation theory. In our case of $\theta \neq 0$ we have no parameter x_0. However, we can construct a rapidly converging series for $\epsilon_0(\theta)$ (for not too large angles θ) by taking the "potential" $V(x, z) = -2\xi\eta\sin\theta\cos\theta$ as a perturbation. When $\theta = 0$ our

[1)] It can be shown easily that the minimization of $\epsilon_0(x_0)$ with respect to x_0 corresponds to the selection of a ground state with zero total current.

result coincides with that of Schmidt.[9]

In the zeroth approximation the wave functions are

$$\psi_{nm}^{(0)} = \text{const} \times (\sin\theta\cos\theta)^{1/4} \exp\left(-\frac{\xi^2}{2}\cos\theta - \frac{\eta^2}{2}\sin\theta\right) \times H_n(\xi\sqrt{\cos\theta})H_m(\eta\sqrt{\sin\theta})$$

$(0 \le \xi < \infty, -\infty < \eta < \infty)$ and correspond to the energy levels

$$\varepsilon_{nm} = (2n+1)\cos\theta + (2m+1)\sin\theta. \qquad (4)$$

It follows from (3′) that only even values of n are allowed. In the second order of the perturbation the ground-state energy is

$$\varepsilon_0^{(2)} = \varepsilon_{00} + 4\sin^2\theta\cos^2\theta\,\langle 0|\eta|1\rangle^2 \sum_{n=0}^{\infty} \frac{\langle 0|\xi|n\rangle^2}{\varepsilon_{00}-\varepsilon_{n1}}. \qquad (5)$$

After calculating the matrix elements we have

$$\varepsilon_0^{(2)} = \frac{\varkappa}{H_{c3}(\theta)} = \left(1-\frac{1}{\pi}\right)\cos\theta + \sin\theta - \frac{\sin\theta\cos\theta}{\pi}\sum_{k=1}^{\infty}\frac{(2k)!}{(2^k k!)^2}\frac{1}{(2k-1)^2}\frac{1}{2k\cos\theta+\sin\theta}. \qquad (6)$$

For not too large angles $(\theta \lesssim 1)$ we can limit the summation to its first member:

$$\frac{\varkappa}{H_{c3}(\theta)} \approx \left(1-\frac{1}{\pi}\right)\cos\theta + \left(1-\frac{\cos\theta}{4\pi}\right)\sin\theta. \qquad (7)$$

When $\theta = 0$ Eq. (7) gives $H_{c3} = 1.5\ H_{c2}$ (in the conventional units) instead of the exact value $1.69\ H_{c2}$. In fourth order we obtain $H_{c3}(0) = 1.6\ H_{c2}$.

The initial slope of the $H_{c3}(\theta)$ curve is of interest in practice. By means of (7) we obtain $(dH_{c3}/d\theta)_{\theta=0} \approx -1.4\ H_{c3}(0)$, in agreement with the numerical calculation in[10]. To determine the structure of the state for $H < H_{c3}(\theta)$ we must solve simultaneously a nonlinear Ginzburg-Landau equation and a Maxwellian equation. For fields close to $H_{c3}(\theta)$ the solution for ψ is found as a superposition of degenerate solutions of the linearized problem subject to the condition that ψ is a periodic function in the yz plane.[1] It is reasonable to assume that the period d of the formed structure is of the same order of magnitude as a nucleus in the z direction. From (3) we obtain the period $d \sim 1/\kappa\sqrt{\sin\theta}$. When $\theta = 0$ we obtain the uniform layer of Saint-James and de Gennes $(d = \infty)$.[2)]

2. SAMPLE PREPARATION AND MEASUREMENT TECHNIQUE

A. Samples. Our work was done with niobium, which can be purified better than any other type II superconductor.[12] The starting material was niobium pentachloride that was purified by twofold rectification and reduced by hydrogen. A spectrochemical analysis revealed that the pentachloride contained 1×10^{-5} wt.% of Cu and of Mn; the accuracy of this analysis was 2×10^{-6}% for Cu and 5×10^{-6}% for Mn. The following kinds of impurities were not registered (the sensitivity of the analysis in wt.% is indicated within parentheses): Ta(5×10^{-4}), Ti(1×10^{-4}), Si(2×10^{-3}), Al(5×10^{-3}), Fe(5×10^{-4}), Bi(1×10^{-5}), Cd(2×10^{-5}), Zn(5×10^{-5}), Pb(1×10^{-5}), Co(1×10^{-5}), Ni(1×10^{-5}), W(2×10^{-3}), and Mo(5×10^{-4}). The starting material, consisting of small crystals with volumes of a few mm^3, was melted in an electron-bombardment oven. The wire drawn from the ingot was annealed, following removal of the surface layer, in an oilfree vacuum at $10^{-8}-5 \times 10^{-11}$ Torr. The annealing conditions are given in the table.

The prepared samples had a 0.5-mm diameter and were 10–30 mm long. They consisted of monocrystalline blocks, which under 20 × magnification were seen to be regular hexagons with specular surfaces. The purity of the samples was monitored through their residual resistance, which was measured in a 3–6-kOe magnetic field at 4.2°K (see the table), and through the shapes of their magnetization curves.

B. Measurement technique. The critical currents in the surface layer were determined from the real part of the magnetic permeability.[13,14] Figure 1 is a block diagram of the apparatus. To the sample 1, which was placed within one of two opposing measuring coils 2, we applied two longitudinal magnetic fields simultaneously—a constant field $H_{c2} < H < H_{c3}$ and an alternating field $h_0 \cos\omega_0 t$. The emf induced in the measuring coils was amplified, and was registered with an EPP-09M instrument following synchronous detection.

The measurement technique was based on the fact that the surface layer of a cylindrical sample com-

Properties of the samples

Sample (batch label)	Method of production*	Annealing conditions			$R(300°K)/R(4.2°K)$
		T, °C	Time, hrs	Pressure, Torr	
Nb-N-2	ER	2000	6	$2 \cdot 10^{-7}$	300
Nb-N-2	HR	2100	6	$2 \cdot 10^{-7}$	300
Nb-N-3	»	1700	6	$7 \cdot 10^{-8}$	300
Nb-N-3a	»	2000	6	10^{-7}—10^{-8}	—
Nb-H-3c	»	2200	6	10^{-7}—10^{-8}	700
Nb-Sh-1	EB	2100	10	$1.5 \cdot 10^{-10}$	80
Nb-Sh-2	HR	2100	9	$5 \cdot 10^{-11}$	14000

*Abbreviations: ER - electrolytic reduction, HR-hydrogen reduction, EB - electron-bombardment furnace.

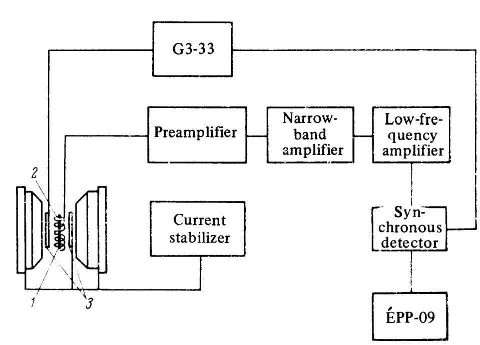

FIG. 1. Block diagram of apparatus for measuring critical currents in a superconducting surface layer. 1 - sample, 2 - measuring coils, 3 - modulating coils.

[2)] The structure was calculated by I. O. Kulik, [11] to whom the present authors are grateful for communicating his results before publication.

prises an ideal superconducting shield against a field $h_0 \cos \omega_0 t$. The shielding is complete so long as the current induced in the surface layer is below the critical value. As soon as the value J_c is exceeded the shielding ceases to be complete. For any given value of H_0 we determine J_c at the time when the field h_0 begins to penetrate into the sample:

$$h_0(H_0) = \frac{4\pi}{c} J_c.$$

When the synchronous detector is tuned in phase with the alternating field the measured emf induced in the measuring coils is

$$\Delta E_1 = -h_0\omega_0 Sn(\mu' - 1) = -\frac{4\pi\omega_0 Sn}{c} J_c,$$

where ω_0 is the alternating field frequency, S is the cross sectional area of the sample, n is the number of turns in a measuring coil, μ' is the amplitude of the first harmonic of the real part of the magnetic permeability, and J_c is the critical current density per unit length of the sample's surface. The transition of the surface layer from the superconducting state to the normal state was recorded for a fixed value of h_0 while H increased at a constant rate. The sensitivity of the apparatus to the preamplifier input was 10^{-8} V. Each measuring coil had 4000 turns. The relative accuracy of the J_c measurements, which depended on the ratio H_0/H_{c2}, was about 3% in the interval $1 \leq H_0/H_{c2} \leq 1.5$, and reached 5–7% near H_{c3}. The smallest value determined for the critical current in the surface layer was 0.002 A/cm.

The magnetic fields were generated by a superconducting solenoid and by a standard electromagnet that was used to investigate EPR. The electromagnet was calibrated by means of proton resonance while the solenoid was calibrated ballistically, and both were powered by a stabilized current source consisting of semiconductor elements. The described apparatus was used to measure the dependence of J_c on the magnetic field strength, temperature, and the angle between the external magnetic field and the axis of the sample.

A small inverted Dewar vessel was used for the intermediate temperature measurements.[15] The temperatures were measured with a carbon thermometer of 0.015°K accuracy. In recording $J_c(\theta)$ a measuring coil and sample were placed in a horizontal position and were rotated at a uniform rate in a fixed magnetic field between the poles of the electromagnet, at 30 Hz and 120 Hz.

3. EXPERIMENTAL RESULTS

A. Dependence of the critical current in the surface layer on sample purity. All samples, except in the case of Nb-Sh-1, which was made of ordinary niobium melted by electron bombardment, were prepared from the same starting material and had identical specular surfaces. However, the degree of outgassing varied, depending on the vacuum in which the high-temperature anneal took place. The residual concentrations of impurities were determined from the resistance ratio $\Gamma = R(300°K)/R(4.2°K)$ (see the table) and from the shapes of the magnetization curves, which were almost fully reversible. Since the diameters of the samples were small it could be assumed that during prolonged annealing the impurities were distributed uniformly throughout their volumes. Therefore the purity of the surface layer can be considered identical with that of the bulk.

Figure 2 shows the dependence of the critical current densities in the surface layer on the magnetic field strength, for the described samples. From the values of J_c near H_{c2} it is seen that the critical current density diminishes as Γ increases. In the purest samples J_c was below the sensitivity threshold of the measuring equipment in the entire region $H_{c2} < H < H_{c3}$.

B. Effect of surface defects on the critical current. It was of interest to determine how J_c is affected by surface deformations and impurities. For this purpose

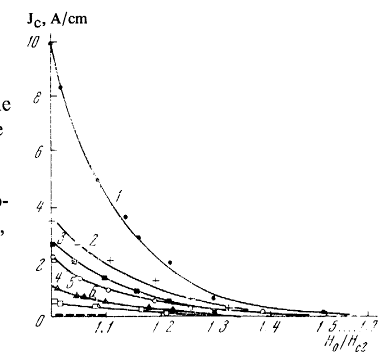

FIG. 2. Dependence of the critical current density on the magnetic field for different niobium samples (at 4.2°K). 1 - Nb-N-1, 2 - Nb-N-2, 3 - Nb-N-3, 4 - Nb-N-3a, 5 - Nb-N-3c, 6 - Nb-Sh-1; dashed line - Nb-Sh-2 ($J_c < 0.002$ A/cm).

an Nb-Sh-2 sample was carefully rubbed with an abrasive having grains that did not exceed 14 μ. Figure 3 shows the $J_c(H)$ measurements that were recorded after roughening. The introduction of defect is seen to have enhanced the critical current by four orders of magnitude.[3)]

The surface layer was subsequently etched and $J_c(H)$ was remeasured; the results are shown in Fig. 3, where we observe that J_c was reduced tenfold and never returned to its previous level. A microscopic examination of the sample following etching showed that the surface was covered with etch pits, so that it had lost its previous specular appearance.

It was thus shown that both impurities distributed throughout the bulk and surface defects lead to an enhancement of the critical current in a surface layer by several orders of magnitude.

C. Temperature dependence of J_c. The influence of defects on critical currents in the mixed state ($H_{c1} < H < H_{c2}$) is well known[17] and is similar to the above-described effect. In order to determine the degree of similarity between the properties of surface and bulk currents we investigated the dependence of J_c on temperature in the region $H_{c2} < H < H_{c3}$; the results are shown in Fig. 4.

In the temperature interval $4.2°K \leq T \leq T_c$ the surface current density is seen to depend linearly on T. A similar linear relationship was observed in[17],

3) The simultaneous enhancement of H_{c3} can be attributed to a reduction of the mean free path because of plastic deformation and the associated increase of the parameter κ [16] near the surface.

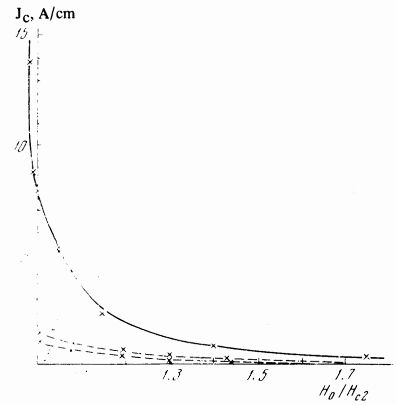

FIG. 3. Dependence of the critical current density in an Nb-Sh-2 sample on the treatment of the surface. 1 - after roughening with an abrasive (grain size < 14μ), 2 - after roughening followed by etching, 3 - after roughening and double etching.

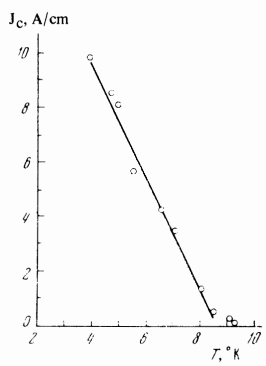

FIG. 4. Dependence of the critical current density on temperature for an Nb-N-1 sample.

where the temperature dependence of the bulk current was investigated in the mixed state of niobium ($H_{c1} < H < H_{c2}$).

D. Dependence of J_c on the angle θ between the magnetic field and the axis of the sample. For the purpose of evaluating the influence of the transverse magnetic field component on J_c we obtained $J_c(\theta)$ curves, which were recorded for fixed values of H and h_0 and are shown in Fig. 5. For all the curves we had $h_0 = 0.5$ Oe and f = 120 Hz. For the given sample $h_0(H_{c2}) = 4\pi c^{-1} J_c = 4.27$ Oe.

Curves 1 and 2 have flat maxima. The other curves lie in the angular interval 0–4.5°. The flatness of the maxima observed on curves 1 and 2 appears to be accounted for by the existence of a threshold value for the product $h_{0c}H_{\perp c}$, where the critical current begins to fall off rapidly. On the flat portions $h_0 H_\perp < h_{0c}H_{\perp c}$; on the steep portions $h_0 H_\perp > h_{0c}H_{\perp c}$.

It is interesting to note that according to Saint-James and de Gennes[2] the critical magnetic field of a superconducting surface layer is H_{c3} when $\theta = 0$ and H_{c2} when $\theta = \pi/2$. Accordingly, if while the sample was rotated J_c decreased as $H_{c3}(\theta)$ diminished, J_c should disappear when $\theta \rightarrow \pi/2$. The decrease actually occurred within the angular interval 4.5°. The angular dependence of J_c that we obtained for niobium (Fig. 5) agrees qualitatively with the results of Hart and

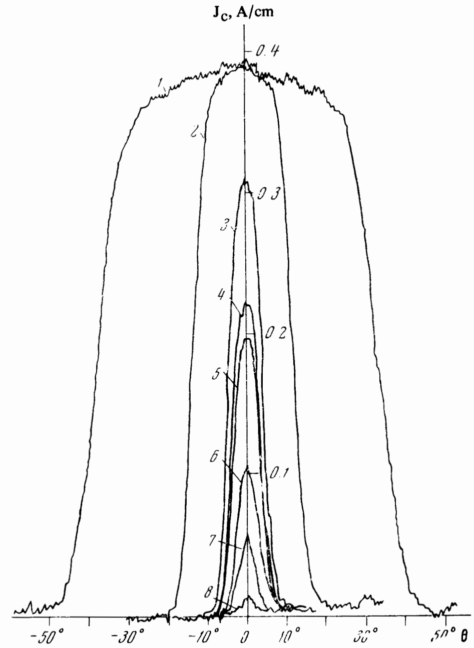

FIG. 5. Critical current density versus the angle between the magnetic field and the axis of the sample, for different field strengths: 1 - 2940 Oe, 2 - 3620 Oe, 3 - 4330 Oe, 4 - 4395 Oe, 5 - 4445 Oe, 6 - 4560 Oe, 7 - 4730 Oe, 8 - 5070 Oe. Nb-N-3c sample; T = 4.2°K.

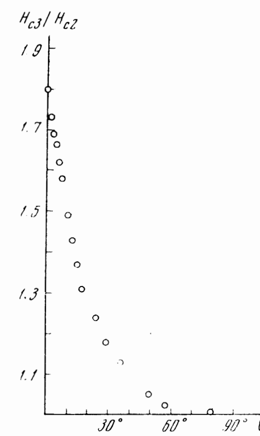

FIG. 6. Dependence of the field H_{c3} on the angle between the magnetic field and the axis of the sample. Nb-N-3c sample; T = 4.2°K.

Swartz,[18] who investigated the behavior of $J_c(\theta)$ in flat films of the alloy Pb + 2% In ($H_{c2} < H < H_{c3}$).

E. Dependence of H_{c3} on the angle θ between the magnetic field and the axis of the sample. It is of interest to make a comparison between the initial slope $(dH_{c3}/d\theta)_{\theta=0}/H_{c3}$ calculated in Section 1 and the experimental results shown in Fig. 6. The theoretical initial slope of the $H_{c3}(\theta)/H_{c2}$ curve is $\theta_0 \approx 22°$, while the experimental result is 20°. In view of the approximate nature of the calculations the agreement must be considered satisfactory.

4. DISCUSSION

Critical currents in the superconducting surface layers of cylindrical samples have been calculated by

Park[4,5] and by Fink.[7] We shall compare the theoretical and experimental results, assuming $\kappa \approx 1$, which corresponds to niobium at 4.2°K. Park's calculated value of $J_C(H_{C2})$ is 15 A/cm,[4,5] while Fink[17] calculated 20 A/cm. For Nb-Sh-2 samples we obtained 0.002 A/cm. This large discrepancy, which is four orders of magnitude, shows that theories which neglect the transverse component of the magnetic field are practically useless.

We performed the calculation described in Sec. 1, where it was shown that even a very small transverse component of the magnetic field transforms a uniform order parameter to a periodic form (with the period $d \sim 1/\kappa\sqrt{\sin\theta}$). The periodicity appears to signify that in a surface layer whose thickness is of the order of the coherence length a lattice of quantized vortices appears, similar to the Abrikosov vortices in the mixed state. Therefore an analogue of the mixed state should exist in the region $H_{C2} < H < H_{C2}(\theta)$ when the surface layer is placed in an inclined magnetic field.

Bardeen and Stephen[19] have shown that in an ideally pure superconductor even the smallest measuring current induces motion of the vortices; this is the equivalent of a resistance. However, numerous experimental studies have shown that the presence of defects stabilizes the Abrikosov vortex lattice and results in a finite critical current that can attain the volume density 10^6-10^{17} A/cm^2. Our experimental work, in agreement with the calculations of Sec. 1, indicates that under actual experimental conditions a superconducting surface layer exhibits properties similar to those observed in the mixed state of a type II superconductor. Indeed, as the purity of the niobium sample is enhanced the critical current in the surface layer is reduced, and in the purest samples it approaches the vanishing point within the entire region $H_{C2} < H < H_{C3}$. When defects are introduced artificially into the surface the critical current of surface superconductivity increases by several orders of magnitude.

The critical current density in the surface layer of impure and deformed niobium samples can reach 10^6 A/cm^2 (per unit area of a transverse cross section of the surface layer). This is comparable with the bulk density in highly deformed niobium-zirconium alloys within the region $H_{C1} < H < H_{C2}$. The decisive role of the periodic structure is also indicated by the strong angular dependence of $J_C(\theta)$, since the angle governs the period of the structure that appears in the surface layer.

We note in conclusion that it would be interesting to observe directly the magnetic field distribution on the surface of pure niobium, using the magnetic powder technique or the Faraday effect.

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