Non-quantized penetration of magnetic field in the vortex state of superconductors

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As first pointed out by Bardeen and Ginzburg in the early sixties[1,2], the amount of magnetic flux carried by vortices in superconducting materials depends on their distance from the sample edge, and can be smaller than one flux quantum, $\phi_0 = h/2e$ (where h is Planck's constant and e is the electronic charge). In bulk superconductors, this reduction of flux becomes negligible at submicrometre distances from the edge, but in thin films the effect may survive much farther into the material[3,4]. But the effect has not been observed experimentally, and it is often assumed that magnetic field enters type II superconductors in units of ϕ_0. Here we measure the amount of flux introduced by individual vortices in a superconducting film, finding that the flux always differs substantially from ϕ_0. We have observed vortices that carry as little as $0.001\phi_0$, as well as 'negative vortices', whose penetration leads to the expulsion of magnetic field. We distinguish two phenomena responsible for non-quantized flux penetration: the finite-size effect[1–4] and a nonlinear screening of the magnetic field due to the presence of a surface barrier. The latter effect has not been considered previously, but is likely to cause non-quantized penetration in most cases.

The magnetic properties of a superconductor, including its current-carrying capacity, are determined by the motion of flux through that superconductor as a whole; this motion involves propagation of flux not only through the bulk but also through the superconductor's edge. Because of the inevitable pinning in real superconductors, vortices can initially penetrate only at a finite (usually, mesoscopic) distance from the edge. This effectively creates an edge layer that serves as a reservoir of vortices that are subsequently injected further into the bulk, and there is growing evidence that such a layer significantly influences global superconducting properties[5,6]. On the other hand, near-edge vortices are not exactly the same as vortices in the bulk because the distribution of electric currents around a vortex (that is, the vortex's structure) has to change owing to the presence of the edge[1–4].

One of the most directly observable consequences of the influence of an edge on a vortex is that its flux is no longer quantized and becomes smaller than ϕ_0 (refs 1–4). This effect is particularly important in the case of thin films, where the screening is strongly suppressed and non-exponential[3,4]. Although this flux reduction has been known theoretically for several decades, such vortices (carrying a fraction of ϕ_0) have never been observed or inferred in an experiment. This provided the original motivation for our work, as we found a way to address the issue by making use of ballistic Hall magnetometry[7,8]. This technique allows accurate magnetization measurements on micrometre-sized superconductors, where the edge effects can be dominant.

Figure 1 shows typical behaviour that we observed for the initial stages of field penetration in relatively large (15-μm), thin-film superconductors. Curve a shows magnetic flux penetrating inside a sample in a sequence of steps, such that each step corresponds to a vortex or a number of vortices jumping inside; such behaviour is in agreement with general expectations. However, a more careful look reveals that the step height is not quantized, and that some jumps are smaller than ϕ_0. We postpone a discussion of this observation and now refer to another (nominally similar) sample in Fig. 1. Curve b (for this sample) reveals a completely different picture, which we have observed for many other samples. Here, after the initial region of the full Meissner effect, the flux enters the film relatively smoothly and, only after several flux jumps, the behaviour becomes qualitatively similar to the one shown in curve a. On the smooth part of curve b, the flux jumps correspond to a minor fraction of ϕ_0. Moreover, the first two jumps are negative, indicating that the superconductor expels magnetic field when a vortex jumps inside. The influence of the edge[1–4] discussed above can decrease the amplitude of flux jumps and is partly (see below) responsible for non-quantized steps. However, the existence of negative flux jumps is unexpected and seemingly makes no sense.

To understand the origin of the negative jumps as well as the reason why similar samples exhibit such different behaviour, we performed a number of experiments using various sample geometries. The results are summarized in Fig. 2, where we try to simplify the situation as much as possible by using relatively small disks and by examining only the penetration of the first vortex. The advantage of using such small samples is that bulk pinning becomes negligible compared to interaction of vortices with the edge and, as a result, the first vortex comes right to the disk's centre[8,9]. Therefore, we can study the penetration of an individual vortex at the same, well defined, distance ($R = D/2$) from the edge. As seen from Fig. 2, the amount of flux carried by vortices entering the disk depends on the roughness of the edge of the disk. The disk shown in Fig. 2a, with a smooth edge, exhibits a negative flux jump when the first vortex

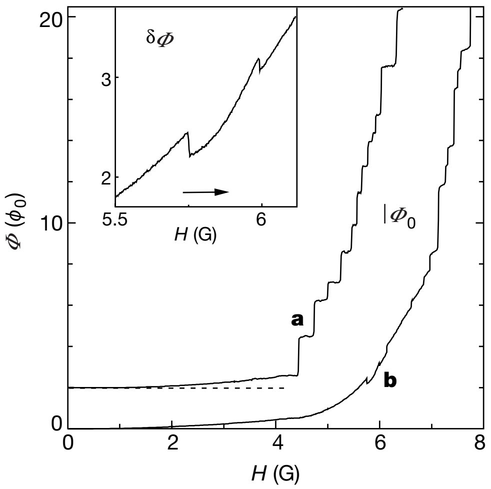

Figure 1 Penetration of perpendicular magnetic field in a thin superconducting film. The curves show the amount of flux Φ, as a function of increasing field H, inside two aluminium disks of diameter $D \approx 15\ \mu m$ and thickness $h \approx 0.1\ \mu m$ at $T \approx 0.5$ K. Due to bulk pinning, which is rather weak[8] but still present, entering vortices jump no farther than a few micrometres from the edge (we observe hysteresis due to bulk pinning if $D > 4\ \mu m$). This makes measurements for the larger disks essentially equivalent to a study of flux penetration in a πD-long strip of an identical macroscopic film. Initially, the samples were cooled in zero field. Special care was taken to avoid 'freezing-in' any vortices; the absence of such vortices was verified by observing a symmetric response for the opposite field direction. The measurements were performed using ballistic Hall magnetometry (Fig. 2). For convenience, we define Φ so that it has zero slope in the low-field limit where $M \propto H$ (such a notation ignores the amount of flux in the λ-layer for the ideal Meissner state; taking the latter flux into account would only lead to an additional, constant slope for Φ–H curves). The absolute scale along the Φ axis is determined with an experimental accuracy of about 10%. Curve a is shifted for clarity. Inset, magnified view of part of curve b, exhibiting negative flux jumps.

enters it. On the other hand, if we introduce a sharp defect at the edge of the disk (as in the disk shown in Fig. 2c), the more-or-less expected behaviour is recovered. Figure 2b shows an intermediate case, where a vortex brings in practically no flux. We note that the leaving vortices carry away approximately the same amount of flux (about $0.7\phi_0$), independent of edge roughness. If no special care is taken, our disks usually have a few tiny cuts at the edge[7–9] that occur because of ripping of the evaporated film during the standard lift-off procedure. To avoid this, the disks in Fig. 2 were fabricated using a double-layer resist. A retrospective analysis has shown that the sample that produced curve a in Fig. 1 has a jagged edge, while the edge of the sample that produced curve b is smooth.

Figure 2 shows clearly that the negative jumps are somehow connected with the stronger bending of the flux–field (Φ–H) curves and with the increase of the penetration field H_p for the first vortex. The smoother the edge, the longer the Meissner state persists and the more strongly the Meissner curve bends upwards. The bending means that the magnetic field penetrating in the near-edge layer cannot be described by the London model, which requires $M \propto H$ and $\Phi(H) = 0$ (see Fig. 1 legend). The nonlinear Meissner effect observed for samples with smooth edges is due to the presence of a high surface barrier that allows superconductors to persist in a metastable (superheated) state[8–10]. Figure 2 shows that the edge roughness strongly suppresses the barrier for vortex entry, while the exit barrier is influenced relatively weakly.

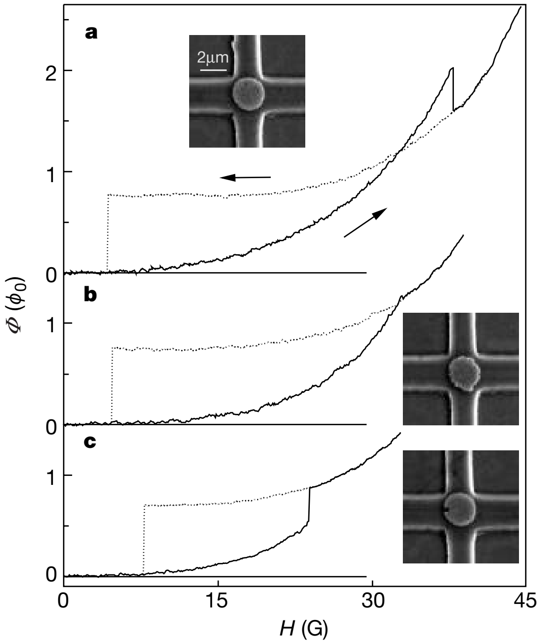

Figure 2 Penetration and expulsion of the first vortex in superconductors with various edge roughnesses. Solid and dotted curves were obtained by sweeping the magnetic field up and down, respectively. The hysteresis is due to a surface barrier and, until a vortex enters or leaves the disks, the curves are reproducible for sweeps in both directions. Insets, micrographs of the studied Al disks ($D \approx 2$ µm and $h \approx 0.15$ µm; $T \approx 0.5$ K). The rough edges for the disks shown in **b** and **c** were intentionally drawn by electron-beam lithography. The disks are placed on top of Hall crosses (see the micrographs) made from a high-mobility two-dimensional electron gas. Such probes measure the average magnetic field in the central area of the Hall cross[7]. The Hall magnetometers can be considered as flux meters with a square detection loop, in the centre of which a superconducting sample is placed. The superconducting coherence length $\xi(T = 0\,\mathrm{K})$ for the disks' material is about 0.25 µm and the magnetic penetration length $\lambda(0) \approx 70$ nm, that is, the material is a type I superconductor (Ginzburg–Landau parameter, $\kappa = \lambda/\xi \approx 0.3$). We note that thin films in a perpendicular magnetic field behave more like type II superconductors, and exhibit vortex structures[8–11]. We can move into the true type II regime by using less-pure Al, and have observed the discussed behaviour also for $\kappa > 1$. However, the reduced screening due to unavoidably larger λ for large κ led to rapid deterioration of our experimental resolution, and here we present the data for lower κ.

In order to explain the origin of negative flux jumps, we will refer to the theoretical curves in Fig. 3a that show the magnetization response for a disk with parameters close to those in Fig. 2a. The important feature to notice is the intersection of magnetization curves for different vortex states. The curve crossings occur in a metastable regime and mean that, as the field increases, the preceding vortex state can accommodate more flux (that is, can be less diamagnetic) than the following, more energetically favourable, state with a larger L. This leads to the possibility of vortex jumps that can bring inside a superconductor any amount of flux from zero to about $\pm\,\phi_0$, depending on H_p. The theory allows metastable states, but is not able to predict at what value of H they become unstable and a vortex jumps in or out[10,11]. In our experiments, only a part of the full length of the theoretical curves is realized. Indeed, we have never seen the intersection of magnetization curves with decreasing H, which could lead to an alternative situation where the flux inside a superconductor increases when a vortex leaves.

To elucidate the physical processes behind the observed negative jumps, we consider the distribution of magnetic field before and after such jumps. In Fig. 3b we plot the field distribution for the superheated Meissner state and the state with one vortex inside at the same applied field. For clarity, we will discuss the case of a superconducting cylinder rather than a disk. The former geometry keeps the essential physical processes intact, but allows us to avoid obscuring demagnetization effects. This also serves to illustrate the fact that negative flux jumps are not exclusive to the thin-film geometry. In the superheated state, the external field strongly suppresses the order parameter near the edge, so that the field penetrates inside the cylinder by as much as a few λ deeper than the

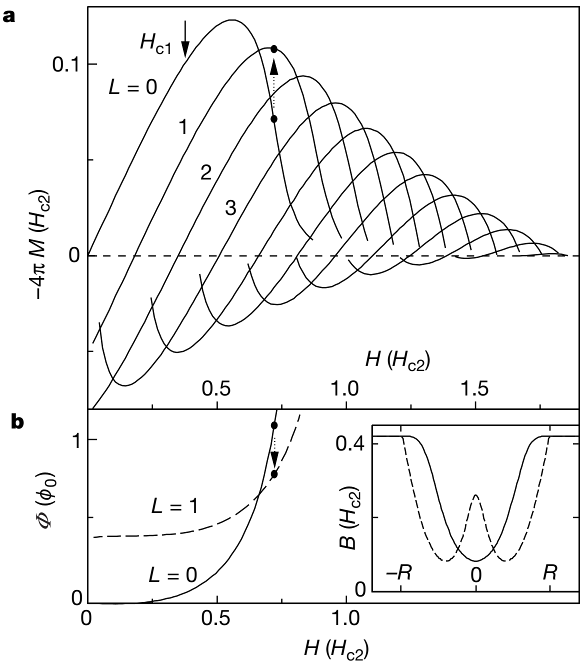

Figure 3 Magnetic response of mesoscopic superconductors found theoretically. **a**, Calculated magnetization response for a disk with $D = 8\xi$, $h = 0.1\xi$ and $\lambda = 0.28\xi$. The curves are found by solving numerically the full three-dimensional set of Ginzburg–Landau equations[10]. L is the fluxoid number which, for simplicity, can be considered as the number of vortices inside the disk. **b**, The amount of flux Φ inside the disk replotted from the M–H dependence. The solid arrow marks the low critical field H_{c1} where the Meissner state ($L = 0$) and the state with one vortex ($L = 1$) have the same free energy. Dots on the curves show an example of the situation where vortex entry could lead to flux expulsion. The inset shows the radial distribution of magnetic field B inside a superconducting cylinder ($D = 20\xi$ and $\lambda = 2\xi$) for a similar situation, in which an entering vortex reduces the amount of flux inside the cylinder. The Meissner state and the vortex state are shown by the solid and the dashed curve, respectively.

London model allows. The higher the applied field, the more strongly the order parameter is suppressed and the more flux is accumulated in the near-edge layer. On the other hand, when the vortex jumps inside, the screening is restored to a significant extent and, accordingly, there is less flux in the near-edge layer (Fig. 3). The competition between the flux expelled from this layer and the vortex's flux determines the sign and amplitude of flux jumps. Despite the deceptive simplicity of this explanation, there is no simple way to explain why the vortex entry restores screening while the field at the edge remains the same. This is a nonlinear property of superconductors.

We now turn to the question of why the flux jumps are not quantized, even when the surface barrier is suppressed by edge roughness, and why the distance between the curves with and without a vortex is less than ϕ_0 (see Fig. 2). The latter implies that the vortex's flux even in equilibrium (that is, not only the corresponding flux jumps) is considerably less than ϕ_0. This observation can be explained by the changes in the structure of near-edge vortices predicted in refs 1–4. Figure 4 plots the measured amount of flux ϕ carried by a vortex versus its distance from the disk's edge. We can see that all our data for different samples and temperatures fall on a single curve, if plotted in units of the effective penetration length, $\lambda_{\mathrm{eff}} = \lambda^2/h$. There is also excellent agreement with the corresponding theoretical dependence. We note that, for a typical experimental situation, h and λ are about 0.1 μm, and it is very unlikely that a vortex can jump farther than 1 μm from the edge before being stopped by pinning, even in samples with low pinning. According to Fig. 4, in such a case the flux carried by vortices is reduced to about $0.5\phi_0$. Only vortices located as far as 100 μm away from the film edge have their flux quantized with an accuracy better than 1%.

We have shown that there are two independent effects that lead to non-quantized penetration of magnetic field in type II superconductors. The first (theoretically established a long time ago, but never observed and often perceived as small) arises due to changes in the structure of near-edge vortices. This should be important in thin films and, in our opinion, may account for a number of unexplained observations. The second, unexpected, effect is more general, and appears owing to the inevitable presence of barriers for flux motion through a superconducting boundary (for example, Bean–Livingston barriers). If such barriers are sufficiently high, nonlinear screening can lead to the extreme situation, causing 'negative vortices'; but if this is not the case, surface barriers can still prevent the quantized penetration. One or both of the above effects can be expected in many—if not most—relevant experimental situations. □

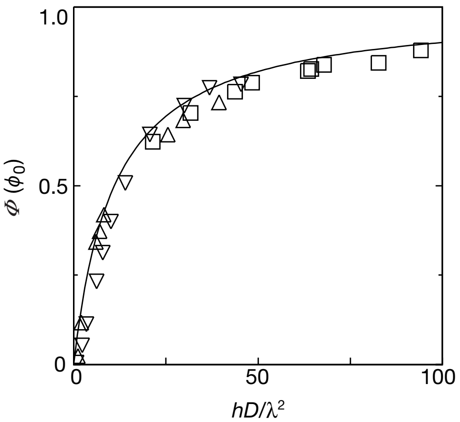

Figure 4 The amount of magnetic flux associated with a vortex in the centre of a thin superconducting disk in equilibrium. The solid curve is an approximate theoretical dependence, $\phi \approx \phi_0\gamma/(\alpha + \gamma)$, found numerically in the limit $h \ll \lambda$, where $\gamma = hD/\lambda^2$ and $\alpha \approx 11.0$ (ref. 3, and V.A.S. *et al.*, manuscript in preparation). Different symbols show experimental data for three disks with D (in μm) ~2 (upright triangles), ~2.4 (inverted triangles) and ~4 (squares), h from about 0.13 to 0.17 μm and the superconducting parameters as in Fig. 2. Because of the surface barrier that is always present (even for a rough edge), we cannot directly determine H_{c1} and, therefore, the amount of flux carried by a vortex in equilibrium. To this end, we notice that the theoretical curves (M–H and Φ–H) for $L = 0$ and 1 are nearly parallel below H_{c1} (Fig. 3) and, hence, the amount of flux associated with vortex exit is sufficiently close—within our experimental uncertainty of 10%—to the vortex's flux in equilibrium. So we have measured the amplitude of flux jumps for the vortex exit. To obtain different data points for each of the disks, we varied the penetration length $\lambda(T)$ by changing the temperature from 0.4 K to close to $T_c \approx 1.25$ K. No fitting parameters were used, except for a slight adjustment (≤10%) of the absolute scale along the ϕ axis for each of the disks.

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Nanomechanical oscillations in a single-C_{60} transistor

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The motion of electrons through quantum dots is strongly modified by single-electron charging and the quantization of energy levels[1,2]. Much effort has been directed towards extending studies of electron transport to chemical nanostructures, including molecules[3–8], nanocrystals[9–13] and nanotubes[14–17]. Here we report the fabrication of single-molecule transistors based on individual C_{60} molecules connected to gold electrodes. We perform transport measurements that provide evidence for a coupling between the centre-of-mass motion of the C_{60} molecules and single-electron hopping[18]—a conduction mechanism that has not been observed previously in quantum dot studies. The coupling is manifest as quantized nano-mechanical oscillations of the C_{60} molecule against the gold surface, with a frequency of about 1.2 THz. This value is in good agreement with a simple theoretical

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