Geometry-guided flux behaviour in Superconducting Pb Microcrystals

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Abstract. Electrochemistry offers highly flexible routes to fabrication of a wide variety of mesostructures, including three-dimensional (3D) crystallites, thin films and nanowires. Using this method we have grown various 3D superconducting Pb mesostructures with vastly different morphologies. We present here results on a truncated(half)-icosahedron with a hexagonal base and a tripod structure with a triangular base. Using Hall probe magnetometry we have obtained magnetisation curves for these structures at several temperatures and see evidence of geometry-driven flux entry and exit as well as flux trapping caused by specific sample geometries. We also observe behaviour that we interpret in terms of the formation of giant vortices, bearing in mind that bulk Pb is a type-I superconducting material.

1. Introduction
Recent developments in electrodeposition have demonstrated that it is possible to tailor the shape of metallic mesostructures by careful control of deposition parameters [1]. Using this method, highly faceted 3D mesoscale Pb crystals with dramatically different shapes (ranging from regular polyhedra to nanowires and snowflakes) can be produced by electrodeposition onto a graphite substrate from Pb salt solutions at different reduction potentials.

Over the past decade, one of the major research directions in mesoscopic superconductivity was so-called “quantum tailoring”, i.e. the engineering of the superconducting properties of the sample by its size and shape [2,3]. The influence of sample geometry is particularly important for vortex matter, as individual vortices in type-II superconductors were found to strongly interact with the edge currents, due to the small size of the sample in units of characteristic lengthscales $\xi$ (order parameter coherence length) and $\lambda$ (magnetic field penetration depth). As a result, in superconducting squares, vortex states with total vorticity that are a multiple of four showed enhanced stability [4], and a similar analogy can be drawn for other polygons. However, previous experimental studies have been restricted to thin, flat samples, and electrodeposition enables us to take the next step in this research – to grow truly three-dimensional, highly faceted samples, where the sample as a whole has distinct geometry, and surfaces are composed of units of very pronounced symmetry. In addition, the material used here (Pb) is a type-I superconductor (in bulk), unless structured with size smaller than $\xi$ and $\lambda$, which makes our microcrystals very different from previously studied samples. We therefore expect that the properties of our samples would show a very complex interplay between the nucleation of surface superconductivity, surface barriers, geometrical shape, and temperature-dependent lengthscales and the Ginzburg-Landau (GL) parameter ($\kappa=\lambda/\xi$).
2. Methods

Two Pb structures have been investigated. The first was a half-icosahedron (figure 1a); an icosahedron that has been sliced through the centre so that it has a flat hexagonal base with a flat triangle on top. The total width of this crystal is about 1.5µm with each triangular face having a side length of about 1µm. The second was a tripod structure (figure 1b) that has a triangular base with three legs pointing upwards from each corner. In the studied crystal, each leg is roughly 5µm long and 0.5µm wide while the edge of the triangular base is 1µm long. The Pb crystals were grown by electrodeposition on a highly oriented pyrolytic graphite substrate (HOPG). An electrolyte of 5mM lead nitrate with a supporting electrolyte of 0.1M nitric acid was used. The half-icosahedron was grown by applying a reduction potential of -0.05V vs. a Pb wire reference electrode for 60s. The tripod was grown by applying a reduction potential of -0.15V for 60s (a typical SEM image of the result is shown in figure 2).

Measurements were carried out using a GaAs/AlGaAs heterostructure array of 1µm x 1µm Hall probes. The probes were operated with a 20µA 32Hz ac current and the Hall voltage detected with a lock-in amplifier. An external magnetic field was applied perpendicular to the Hall array using a superconducting magnet. The structures were placed on the Hall probes using a micromanipulator and stuck down with paraffin wax. The half-icosahedron was placed on top of the Hall probe, in its very centre, while the tripod was placed in the orientation depicted in figure 1b.

![Figure 1](image1.png)  
**Figure 1.** Sketches showing the geometry of the two Pb structures we have investigated. (b) also shows the orientation of the tripod on the Hall probe. The shaded square underneath represents the active area of the Hall probe.

![Figure 2](image2.png)  
**Figure 2.** SEM image of typical electrodeposited Pb tripod.

3. Results & Discussion

We characterize the superconducting properties of our samples by Hall magnetometry, where the magnetic response of the samples is measured in increasing and decreasing applied magnetic fields, all the way to saturation i.e. full destruction of superconductivity. As already known, flux entry and exit in the samples manifests in the M(H) curves in a step-like, quantized manner. Therefore, by measuring and comparing the size of the jumps in the magnetisation curve we are able to determine how many quanta of flux are entering or leaving the sample at a given applied field.

3.1. Half-icosahedron

Figure 3 shows a typical M(H) curve for the half-icosahedron, at 4.9K. As the field is gradually increased the sample exhibits clear Meissner screening up to applied field of 400Oe. Beyond that point, we observe flux penetration over a narrow range of fields before the sample is driven normal at 425Oe, which roughly corresponds to the critical field of bulk Pb at that temperature. First flux entry during the sweep-up is measured to be up to 3 flux quanta, corresponding to the jump labelled b in figure 3. A suggested likely flux distribution for this case is given in figure 5. The subsequent jump in the M(H) curve corresponds to the entry of 6 flux quanta, followed by another 6 before the sample
transits to the normal state. For the reverse sweep (sweep-down), superconductivity is first recovered at the lateral surfaces. This results in a gradual quantisation of the flux in decreasing fields, and vortex states nucleate in a “giant” form. Note also that our samples are made of type-I material, where the formation of multi-quanta vortices is favourable. The state labelled g we believe to represent a giant vortex where a large normal area is located in the centre of the sample, encircled by a superconducting area. As the field is decreased further, we back track the vortex states to the point of flux expulsion. Using the same methodology, we established that flux leaves in multiples of 3 flux-quanta, and we identified the states with vorticity 15 (state f), 12 (e), and 3 (b). The last observed state is actually a single-vortex state (a). Therefore, based on the combined results of up- and down- sweeps, we conclude that flux entry and exit are guided by the C3 symmetry of the upper facet of the crystal as well as the C3 symmetric arrangement of identical facets on the sloped sides of the sample. The single vortex state is recovered in the measurement as it never conflicts with the symmetry of flat polygonal samples.

To verify this, we repeated the measurement at a higher temperature, 5.6K. The M(H) curve shown in figure 4 does not show any flux penetration on the sweep up and the sample remains in the Meissner state up to the critical magnetic field. In the reverse sweep however, we do see very similar flux penetration as we did at 4.9K. The sequence is slightly different, and we found transitions in vorticity of \(15 \rightarrow 12 \rightarrow 6 \rightarrow 3\) and eventually back to the Meissner state. Therefore, at both temperatures the size of the flux jumps is found to be dependent on the geometry of the sample. As flux enters or leaves, it must do so in a way that preserves the symmetry of the vortex state with respect to the complex shape of the crystal. The latter shape is best described in terms of the C3 symmetry of the facets and their spatial arrangement, hence most jumps in M(H) characteristics correspond to multiples of 3 flux quanta.

![Figure 3](image1.png)  
**Figure 3.** Magnetisation curve for the half-icosahedron at 4.9K. Labelled steps refer to flux distribution shown in figure 5.  

![Figure 4](image2.png)  
**Figure 4.** Magnetisation curve for the half-icosahedron at 5.6K.


3.2. Tripod

Geometrically speaking, the tripod studied here is a far more complex structure than the half-icosahedron. Therefore, the magnetisation curves (see figures 6 and 7) exhibit extremely complicated behaviour, which is very difficult to interpret in terms of vortex arrangements. However, we do observe one prominent and novel result - the large amount of flux trapped in this structure as the applied field changes polarity. In addition, we observed that flux trapping diminishes with increasing temperature.

To shed light on this phenomenon, one must take into account the position of the structure of the Hall bar (see figure 1b), which strongly suggests that flux remains trapped in the vertically standing leg of the tripod. Based on our earlier experience, nanowires with similar dimensions to those of the tripod’s legs have shown no such flux pinning when they lie perpendicular to the direction of the applied field. At present, we can offer no plausible explanation of this behaviour, but we report this measurement as an illustration of the richness and complexity of the flux matter in truly 3D superconducting samples.

References

[1] Xiao Z L, Han C Y, Kwok W K, et al., 2004 J. Am. Chem. Soc. 126, 2317
[2] Geim A K, Dubonos S V, Grigorieva I V, et al., 2000 Nature 407, 55
[3] Chibotaru L F, Ceulemans A, Bruyndoncx V and Moshchalkov V V, 2000 Nature 408, 833
[4] Baclis B J and Peeters F M, 2002 Phys. Rev. B 65, 104515; Misko V R, Fomin V M, Devreese J T and Moshchalkov V V, 2003 Phys. Rev. Lett. 90, 147003