Artificial pinning centers using the barrier layer of ordered nanoporous alumina templates

X Hallet¹, S Mátéfi-Tempfli¹, M Mátéfi-Tempfli¹, S Michotte¹, L Piraux¹, J Vanacken², V V Moshchalkov²

¹ Unité de Physico-Chimie et Physique des matériaux, Université Catholique de Louvain, Place Croix du Sud 1, 1348 Louvain-la-Neuve, Belgium
² Institute for Nanoscale Physics and Chemistry, Katholieke Universiteit Leuven, Celestijnenlaan 200D, 3001 Leuven, Belgium
E-mail: xavier.hallet@uclouvain.be

Abstract. The barrier layer of self-ordered anodized aluminium oxide, which is grown from an aluminium foil, has been revealed by a selective chemical etching of the remaining aluminium. The surface obtained in this way consists of a triangular lattice of bumps with 100nm spacing, and heights of approximately 50nm. Using this surface as a template for controlling the pinning in thin superconducting films, superconducting Nb was deposited with different thicknesses and under different deposition angles. The evaporation under a 30° angle shows an asymmetric pinning potential composed of two triangular lattices having different pinning strengths. Matching effects are observed up to 1T. Matching effects are also maintained at relatively low temperature.

1. Introduction
When a current is applied to a superconductor in the mixed state, vortices move perpendicularly to this current and generate energy loss. This dissipation weakens the superconducting state and the critical current is generally reached much below the theoretical pair-breaking current. Vortex pinning has therefore been extensively studied in the last decades because of the technological and scientific interest.

Abrikosov has shown [1] that in defect free superconductors, the vortices tend to form a triangular lattice with a lattice constant depending on the applied magnetic field (equation 1). Therefore, random pinning sites do not provide an optimized vortex lattice pinning. Since the development of lithographic techniques, several artificially created lattices of pinning sites have been investigated [2-7]. The commensurability of the vortex lattice with the periodic pinning array (PPA) generates a strong stabilization of the vortex lattice. This results in an increase of the critical current, a decrease of the magnetoresistance and a reduction of the flux creep. More recently, magnetic defects attracted much attention. The interaction between the superconductor and the magnetic field of the dots produces asymmetric matching effects [6,8] and field-induced superconductivity [9].

Lithography enables to create a tremendous variety of PPA’s. However, this technique is very expensive and time-consuming. Nowadays, matching effect from lithographically introduced PPA’s was demonstrated only to about hundreds of gauss. However, practical applications usually require much higher fields.
A breakthrough was made by Welp et al. [10] with the use of anodized aluminium oxide (AAO) templates to imprint a triangular lattice of holes in a thin Nb film increasing the first matching field by about more than one order of magnitude compared to previous lithographic techniques results. Another self-assembled approach was proposed by Vinckx et al. [11]. In that case, a triangular lattice of polystyrene spheres was used to modulate the thickness of a Nb thin film inducing a hexagonal lattice of pinning sites with matching effects up to 90 mT.

We present in this paper, a novel method to make hexagonal lattices of pinning centres with a very small lattice parameter. Highly ordered AAO templates are created by a two step anodization of a high purity Al foil. The oxide barrier layer is then revealed providing a triangular lattice of bumps (almost half-spheres) on which the evaporation of a thin Nb layer is made under different angles. This induces a periodic thickness modulation of the Nb film and thus creates a periodic lattice of pinning sites.

Pinning centres form a hexagonal arrangement since they are supposed to be localised on the interstitial sites of the bump lattice. This configuration of pinning centres allows us to increase by two the number of pinned vortices compared to the configuration obtained when the opposite face (with the holes) is chosen such as in [10,12]. In our case, the evaporation under an angle provides an interesting asymmetric pinning potential, which can be seen as two interpenetrating triangular pinning lattices of different strength. They can thus pin twice as many vortices before the first matching field is reached. The formation of undesirable interstitial vortices is thus delayed to higher fields. Moreover, the highly ordered structure obtained in this way remarkably shows matching effects up to 1T.

2. Experimental

The non-supported AAO templates used in this work were fabricated by a two step anodization process (Figure 1). An ultra-pure (99.999 %) aluminium foil (500µm) is first partially anodized in 0.3M oxalic acid at 2°C under a constant voltage of 40V (fig. 1a) leading to a mean interpore distance of 100nm (fig.1b). The first anodization step was performed to a depth of 50µm to promote ordering. Then this first anodized layer is selectively removed with a mixture of H$_3$PO$_4$ 6wt% and chromic acid 1.8wt% at 60°C (fig. 1c). A second anodization step is then applied (fig. 1d) in order to obtain a highly ordered template.

The pores form a triangular lattice of long channels closed at one end by round-shaped Al$_2$O$_3$ barrier layers. The selective removal of the unanodized aluminium with CuCl$_2$ reveals a nanoporous template with contrasting surfaces (fig. 1e). Indeed, the top surface presents a triangular lattice of holes, while the bottom surface shows a triangular lattice of bumps. These two lattices have the same periodicity because the bumps represent the barrier layer at the bottom of the pores.

![Figure 1](image-url)

**Figure 1.** (a) Anodization system (b) Aluminium + alumina layer after the first anodization (c) Aluminium layer after the Al$_2$O$_3$ removal (d) After the second anodization (e) Aluminium oxide template after the aluminium removal.
The use of triangular lattices of holes to create antidots in thin superconducting films has been reported recently [10,12]. In this present paper, we present results obtained using the bottom face of the template (conjured barrier layer) to modulate laterally thin superconducting films of Nb. These films were deposited using molecular beam epitaxy (MBE) at room temperature at a working pressure of $10^{-10}$ torr and an evaporation rate of $1.5 \, \text{Ås}^{-1}$. The SEM image in figure 2 shows the bottom surface of a highly ordered sample but yet not a perfectly one. Vacancies and deformed bumps are observed throughout the sample at the boundaries of the ordered domains having different orientations. Average domain size [13] is about 1 $\mu$m², which is enough for the reported behaviour.

![Figure 2. SEM image of the bottom surface (bumps) of a highly-ordered alumina template](image)

Six different Nb films were deposited on the same bottom surface. Two different nominal thicknesses (30 and 50nm) and three different angles (0°, 30° and 60° from normal) were tried. Real thicknesses are obtained by multiplying the evaporated thicknesses by the cosine of the corresponding evaporation angle.

### 3. Results and discussion

We first discuss the thickness modulation under the different evaporation conditions in order to underline the importance of angular evaporation on our template. Then, we present and discuss the experimental results for the best samples that are those evaporated under 30° (30 and 50 nm nominal thickness).

The bottom surface looks like a triangular lattice of half spheres of radius of ~50nm. The hemispheres are “slightly” deformed into a hexagonal shape. The deepest points are the interstitial sites between 3 adjacent bumps. These sites form a hexagonal lattice. The number of these sites is twice the number of the bumps. Between two interstitial sites, there is a small maximum which is in fact a saddle point.

In the case of a perpendicular evaporation, the thickness of the deposited layer is theoretically the same everywhere on the surface. Figure 3a (upper panel) is a SEM image of the AAO surface. A transversal cut of the 50 nm thick layer along the dotted line is represented on the down scheme. There is no thickness modulation meaning that the volume of the superconductor occupied by a vortex is the same wherever the vortex is placed. However, the surface induces a shape modulation of the superconducting film. At the interstitial sites, the Nb layer has a sharp angle, which can be penetrated more easily by an applied magnetic field. Vortices will tend to go on these sites where the vortex energy is lowered. Therefore, the layer forms a hexagonal lattice of pinning centres. The matching field expected for this hexagonal lattice can be easily calculated. Indeed, the number of interstitial sites is twice the number of bumps. But the first matching field for a triangular lattice (bumps) is equal to:
\[ H_1^\Delta = \frac{2\Phi_0}{\sqrt{3} \cdot d^2} \cong 237 mT \]  

where \( d \approx 100 \text{nm} \) is the bumps spacing and \( \Phi_0 = h/2e \) is the magnetic flux quantum. Therefore, the first matching field in the case of interstitial pinning is simply twice this value:

\[ H_1 = 2 \cdot H_1^\Delta \cong 474 mT \]

Figure 3a (top) represents the location of the vortices on the AAO surface in the case of perpendicular evaporation (0°). Vortices most favourable location is indicated. The pinning strength in this case is weak (sharp angle part is small compared to the total thickness).

The evaporation under an angle produces a thickness modulation, which depends on the direction of evaporation. As seen in figure 2, the bump lattice does not present a unique direction. In fact, the template is split in domains of tens of bumps, which have their own orientations. Therefore, it is not possible to match the evaporation direction with the same lattice direction for all the domains.

The thickness modulation will depend on the orientation of the evaporation axis compared to the hexagonal lattice. Because of the symmetry of the system we only have to consider directions spread over 30°. The figures 3b and c represent two extremes of the considered angular distribution. The first direction is located on a row of bumps (indicated by the arrow in Fig. 3b – upper frame). The other direction is along one side of a hexagonal unit cell (Fig. 3c – upper frame). It is enough to consider only the set of directions in the 30° angle formed by the two extreme ones. As a first approximation, we guess that the thickness modulation in an intermediate direction will be close to the average of the two principal directions. Figure 3b (lower frame) represents the superconducting film thickness (along the dotted line) in the case of an evaporation in the direction of the bumps row. One can see that the angular evaporation induces a pronounced thickness modulation. Indeed, the thickness is proportional to the cosine of the angle between the normal to the surface and the direction of evaporation (30°). Moreover shadow effects also play an important role. Vortex pinning sites will be situated close to every interstitial site. In this case, all pinning sites are equivalent and therefore the pinning lattice has a hexagonal symmetry.

If the evaporation is performed along the direction of a hexagon side, the six interstitial sites are not equivalent anymore. Indeed, half of the sites are situated behind a bump and the other half are behind a small bump in the middle of a hexagon side (saddle point). The simulated layer deposited along this
direction is given in figure 3c (lower panel). It shows that the layer thickness is now strongly varying, which induces an asymmetric vortex pinning potential. In this case, the hexagonal lattice of pinning sites consists of a double triangular lattice of different pinning strength. In figure 3c (upper panel), the energy of the vortices is indicated with their colour. Gray means a low energy (strong pinning) and black a higher energy (weaker pinning).

The behaviour of the whole sample should be somewhat intermediate between these two limiting cases. Therefore, the pinning lattice should consist of two interpenetrating triangular lattices with different pinning strength. If the pinning strengths of the two lattices are similar, the pinning pattern will behave like a hexagonal lattice. On the other hand, if for one of the lattices, pinning strength is negligible compared to the other, it will behave more like a single triangular lattice.

From this moment, we will only discuss the results obtained with 30° evaporation. Critical current measurements have been performed on the 30 nm (nominal) sample (Figure 4a). In this case, the first matching field would correspond to the filling of both triangular pinning lattices forming together a hexagonal lattice. Well-pronounced matching effects are observed every 0.2T. This is clearly visible on the differentiated curve (t=0.83 of figure 4a) up to $H_{3/2} = 1T$ (figure 4b). This result is consistent with theoretical [14] as well as experimental [7,11] results obtained on hexagonal lattice of pinning centres. Indeed, these works showed that periodic anomalies (of different amplitude) appear every half-integer matching field, which corresponds here to 0.2T. In particular, $H_{1/2} = 0.2T$ corresponds to the filling of the strongest triangular lattice. As the triangular arrangement of vortices is known to be the most stable [1], $H_{1/2}$ is well-pronounced and even more than $H_1$ as predicted theoretically [14]. One can also observe $H_{1/4} = 0.1T$, which is only predicted for hexagonal pinning lattice. The matching effect is more pronounced at high temperature which is due to a decrease of intrinsic pinning but an important matching effect is maintained at low temperatures (down to t=0.75) contrary to lithographically made samples.

The figure 5a shows the magnetoresistance curves of our sample (50nm nominal deposited under 30°) for reduced temperatures from 0.89 to 0.99 by step of 0.01. Several matching effects are clearly seen in the entire range of reduced temperature. As observed previously on the 30nm thick sample, periodic anomalies are present every 0.2T. This periodicity corresponds to a triangular lattice of bumps spaced by 108nm, which is consistent with our anodization conditions.

As seen on figure 4b, the 50nm layer also shows a weak fractional matching effects at 0.1T corresponding to $H_{1/4}$. The matching effects are more pronounced for a 50nm nominal thickness than for 30nm.

Fig. 4: 30nm (nominal) Nb film deposited under 30° on bumps of AAO: (a) Normalized critical current versus magnetic field curves for several reduced temperatures ($t = T/T_c$). The critical current is determined from the onset of a measurable resistance. At high temperature only $H_{1/2}$ is visible. At low temperature, matching effects are observed every 0.2T. (b) Differentiated curve of figure 4a at t=0.83. One can see apparent periodic behaviour up to 1T. $H_{1/4} = 0.1T$ is also clearly seen.

The figure 5a shows the magnetoresistance curves of our sample (50nm nominal deposited under 30°) for reduced temperatures from 0.89 to 0.99 by step of 0.01. Several matching effects are clearly seen in the entire range of reduced temperature. As observed previously on the 30nm thick sample, periodic anomalies are present every 0.2T. This periodicity corresponds to a triangular lattice of bumps spaced by 108nm, which is consistent with our anodization conditions.

As seen on figure 4b, the 50nm layer also shows a weak fractional matching effects at 0.1T corresponding to $H_{1/4}$. The matching effects are more pronounced for a 50nm nominal thickness than for 30nm.
At lower temperature (figure 5b) matching effects are preserved (zoom of the dashed frame of figure 5a). The matching effects are still seen every 0.2T at very low resistance. The commensurability effect at the first and second matching field results in a drop in the magnetoresistance curves. At the third matching field the commensurability effect appears as a change in the slope of the magnetoresistance curves. The sharp increase of the resistance from the first matching field is consistent with the appearance of interstitial vortices situated at the centre of the hexagon. The matching effect observed at $H_{3/2} = 0.6T$ then corresponds to the filling of each hexagon centre producing a triangular lattice. The configurations at the different matching field can be found in [14].

4. Conclusion
An original new technique leading to a laterally modulated thin film has been presented. We used self-ordered alumina templates prepared by a two step anodization of an Al foil. Two modulated surfaces on each side of the template are obtained and both can be used for superconducting vortices pinning. The top surface of the template contains a triangular array of holes, while the bottom surface shows a triangular array of bumps. The latter (bottom surface) is revealed after the removal of the remaining Al substrate and was used here as a substrate for the evaporation of a thin Nb layer. Two thicknesses and three different angles were compared. The depositions under 30° from normal show matching effects up to ~1T, which is a big improvement of the magnetic field range for matching effects compared to previous works [11,12]. The periodic matching effects observed are consistent with a quasi-hexagonal lattice of pinning centres that is composed of two interpenetrating triangular sublattices of different pinning strength. This results from the particular thickness modulation obtained using angle deposition over such a template.

This work was supported by the Interuniversity Attraction Pole Program (P6/42) - Belgian State - Belgian Science Policy. X. H. acknowledges financial support of the Fund for Training in Research in Industry and Agriculture. S.M. is a post-doctoral researcher of the National Fund for Scientific Research. The authors wish to thank Bas Opperdoes for the MBE growth of the Nb thin films.

References
[1] Abrikosov A A 1957 Sov. Phys. JETP 5 1174
[2] Daldini O, Martinoli P, Olsen J L and Berner G 1974 Phys. Rev. Lett. 32 218
[3] Fiory A T, Hebard A F and Somekh S 1978 Appl. Phys. Lett. 32 73
[4] Baert M, Metlushko V V, Jonckheere R, Moshchalkov V V and Bruinseraede Y 1995 Phys. Rev. Lett. 74 3269 ()
[5] Bezryadin A and Pannetier B 1995 J. Low Temp. Phys. 98 251
[6] Morgan D J and Ketterson J B 1998 Phys. Rev. Lett. 80 3614
[7] Wu T C, Wang J C, Horng L, Wu J C and Yang T J 2005 J. Appl. Phys. 97 10B102
[8] Lange M, Van Bael M J, Bruinseraede Y and Moshchalkov V V 2003 Phys. Rev. Lett. 90 197006
[9] Lange M, Van Bael M J, Moshchalkov V V and Bruinseraede Y 2002 J. Magn. Magn. Mat. 240 595
[10] Welp U, Xiao Z L, Jiang J S, Vlasko-Vlasov V K, Bader S D, Crabtree G W, Liang J, Chik H and Xu J M 2002 Phys. Rev. B 66 212507
[11] Vinckx W, Vanacken J and Moshchalkov V V 2006 J. Appl. Phys. 100 044307
[12] Vinckx W, Vanacken J, Moshchalkov V V, Mátéfi-Tempfli S, Mátéfi-Tempfli M, Michotte S and Piraux L 2006 Eur. Phys. J. B 53 199
[13] Mátéfi-Tempfli S, Mátéfi-Tempfli M and Piraux L 2008 Thin Solid Films 516 3735
[14] Reichhardt C and Olson Reichhardt C J 2007 Phys. Rev. B 76 064523