Observation of vortex structure in MgB$_2$ single crystals by Bitter decoration technique

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We report the observation of superconducting vortices in pure and lightly Al doped MgB$_2$ single crystals. Low field experiments allow for the estimation of the London penetration depth, $\lambda$ $= 1900 \text{ Å}$ for $T \sim 6 \text{ K}$. Experiments in higher fields (e.g. 200 Oe) clearly show a triangular vortex lattice in both real space (13 $\mu$m by 13 $\mu$m Bitter decoration image of over 1000 vortices) and reciprocal space.

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The recent discovery of superconductivity with $T_c \approx 39 \text{ K}$ in the simple intermetallic compound, magnesium diboride, has caused an explosion of experimental and theoretical works with a major part of the measurements being performed on polycrystalline samples. Until now only a few groups have been able to grow (small, sub-mm size) single crystals of MgB$_2$. From the very earliest data it became clear that MgB$_2$ is a type-II superconductor with its electromagnetic properties described within the framework of the vortex state. So far, for the most part, bulk techniques (magnetization, magneto-transport, etc.) that evaluate the ”average” properties of the sample were used for studies of the superconducting state of MgB$_2$. On the other hand, direct imaging of the vortices is more of a local probe that can evaluate the homogeneity and strength of pinning for different parts of the sample. In addition this technique allows for the determination of basic superconducting properties such as anisotropy and London penetration depth $\lambda$.

In this work we use one of the direct techniques for the imaging of vortices: high resolution Bitter decoration, a technique that allows for the observation of the individual vortices (in small applied magnetic field) as well as for the imaging of vortex structures in a wide range of magnetic fields (up to 2kOe). It should be emphasized, though, that this technique requires that the surface of the sample be very clean and optically smooth. To achieve this degree of surface perfection, single crystals have to be used.

The single crystals used for our decoration experiments were grown using a high pressure cubic anvil technique from a mixture of Mg and B in a BN container (see [13] for details). The samples used for decoration were plates with the approximate dimensions $0.4 \times 0.6 \times 0.05 \text{mm}^3$. In addition to pure MgB$_2$ single crystals ($T_c = 38.4 \text{ K}$, $\Delta T_c = 0.9 \text{ K}$), crystals of nominal composition $\text{Mg}_{0.99}\text{Al}_{0.01}\text{B}_2$ and $\text{Mg}_{0.98}\text{Al}_{0.02}\text{B}_2$ (both with $T_c = 35.6 \text{ K}$, $\Delta T_c = 0.6 \text{ K}$) were studied.

The decoration was performed in the field-cooled (frozen flux) regime in applied magnetic fields ($H \parallel c$) from several Oersted to 200 Oe. The temperature of the sample before the decoration was either 1.4 K or 4.2 K, during the decoration process the temperature increased several degrees; up to 4-5 K or 6-8 K respectively by the end of the decoration process. The vortex structures were observed on the as grown surfaces of the crystals using field emission scanning electron microscope in the secondary electron emission regime to locate the small islands of iron.

Figure 1 shows the structure of vortices in MgB$_2$ for a small applied magnetic field ($B \approx 4.4 \text{ G}$). The observed structure is a collection of weakly interacting individual vortices that do not form a regular triangular lattice (and do not have long range order). The diameter, $d$, of the image of a single vortex ("vortex diameter") is, on av-

![FIG. 1: SEM imaging of the vortices at small magnetic field in MgB$_2$ single crystal.](image-url)
verage, 0.77±0.2 µm, a value that is much less than the distance between vortices. Taking into account the vortex expansion near the surface of a superconductor, the London penetration depth can be estimated as \( \lambda = kd \) with \( k \approx 1/4 \) that results in \( \lambda \approx 1900 \AA \) for the temperature of the decoration experiment \( T \approx 6K \). Common techniques for the measurements of London penetration depth in superconductors usually give very precise relative changes of \( \lambda \) as a function of temperature and/or applied magnetic field. At the same time the accuracy in the absolute value of \( \lambda \) is usually around several tens-of-percents. Since the range of the experimental values of the penetration depth for MgB\(_2\) obtained using different techniques is rather wide (≈ 600 – 3000 Å)\( ^{16,17,18,21} \) our estimate of \( \lambda \) from the size of the vortex image is useful by virtue of giving a reliable absolute value to the upper limit of the penetration depth. Several issues should be remembered in the course of such estimate. It is important to have the density of magnetic particles high enough to fill the region of the magnetic flux penetration in the vicinity of the vortex. From Fig. 1 it is clear that this condition is satisfied since the magnetic particles are observed in the area between the vortices. The correct estimate of the value of the coefficient \( k \) that accounts for the vortex expansion near the surface of a superconductor is apparently the main source of uncertainty in our estimate of \( \lambda \). Empirically, a comparison between the values for the penetration depth from decoration experiments and those obtained by other techniques in different materials where the values of \( \lambda \) are considered to be reliable (Nb, YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\), NbSe\(_2\))\( ^{19,20} \) give the upper limit of errors in \( \lambda \) from Bitter decoration of ≈ 30% using \( k = 1/4 \).

In higher applied magnetic fields a regular triangular lattice is clearly observed (see Fig. 2). No difference in vortex structure was seen between pure and Al-doped MgB\(_2\) crystals. Fig. 2 shows the vortex lattice for MgB\(_2\) single crystal in 200 Oe applied field. The remarkably high quality of the vortex lattice can be seen in the real-space image as well as from the fast Fourier transform (FFT) pattern (inset fig.2). Autocorrelation function allows to estimate the translational length as 8–9 intervortex spacings. Our observation of the hexagonal vortex lattice is in a good agreement with the imaging of vortex unit cell by STM at the magnetic fields of 2kG\( ^{13} \) as well as 5 kGs. It is worth noting that, given the high degree of order shown by the flux line lattice in Fig. 2, MgB\(_2\) should prove to be an excellent system for small angle neutron scattering (SANS) measurements.

FIG. 2: Triangular vortex lattice at magnetic field 200 Oe in MgB\(_2\) single crystal, inset: FFT pattern in an arbitrary scale.

FIG. 3: The vortex lattice in vicinity of the step at magnetic field 200 Oe (top panel), full image of the surface of the MgB\(_2\) single crystal (bottom panel). Note that the step shown in (top panel) can bee seen in the lower half of (top panel) making an 45 degree angle to the horizontal.

In pure and doped magnesium diboride single crystals
Meissner rims (narrow regions free from vortices) were often observed (see Fig. 3) near the top of growth steps and crystal edges. In the latter case the width of the stripe was several tens of microns that is comparable with the thickness of the crystals. Meissner rims were observed in several other superconductors, with weak volume pinning in which case interaction of vortices with the lateral surface of the crystal becomes significant. The observation of a regular triangular lattice and Meissner rims in pure and Al-doped magnesium diboride single crystals point to weak volume pinning and therefore to high quality, with regards to pinning, of the crystals used in the decoration experiments.

As part of our search for effects caused by the expected anisotropy of the London penetration depth, we made several attempts to perform Bitter decoration experiments in a tilted magnetic field using the same single crystals that were utilized for $H \parallel c$ measurements shown above (after cleaning the samples from magnetic particles). However imperfections of the observed vortex structure (smeared maxima in the FFT pattern) did not allow us to reach any unambiguous conclusions about the anisotropy of $\lambda$. These imperfections could be caused, at least in part, by possible damage of the surface of the crystals during the cleaning process. Decoration experiments in tilted and perpendicular to $c$ magnetic field will be part of the future studies and still do have the potential of addressing important issues of superconducting anisotropies in MgB$_2$.

In summary, a clear triangular vortex lattice was observed in an applied field of $\approx 200$ Oe ($H \parallel c$) for Mg$_1-x$Al$_x$B$_2$ single crystals ($x = 0$, 0.01, 0.02) by Bitter decoration technique. The Meissner rims seen near the growth steps and crystal edges suggest very small volume pinning in these crystals. And an upper limit of the London penetration depth $\lambda \approx 1900 \AA$ at $T \approx 6$ K was estimated from decoration experiments in very low fields.

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