Fast Track Communication

Temperature dependence of clusters with attracting vortices in superconducting niobium studied by neutron scattering

A Pautrat¹ and A Brûlet²

¹ Laboratoire CRISMAT, UMR 6508 CNRS-ENSI Caen, 6 Bd Maréchal Juin, 14050 Caen, France
² Laboratoire Léon Brillouin, UMR 12 CEA-CNRS, CE Saclay, 91191 Gif sur Yvette, France

E-mail: alain.pautrat@ensicaen.fr

Received 17 March 2014, revised 1 April 2014
Accepted for publication 3 April 2014
Published 9 May 2014

Abstract

We investigated the intermediate mixed state of a superconducting niobium sample using very small angle neutron scattering. We show that this state is stabilized through a sequence where a regular vortex lattice appears, which then coexists with vortex clusters before vanishing at low temperature. Vortices in clusters have a constant periodicity regardless of the applied field and exhibit a temperature dependence close to the one of the penetration depth. The clusters disappear in the high temperature limit. All the results agree with an explanation in terms of vortex attraction due to non-local effects and indicate a negligible role for pinning. Phase coexistence between the Abrikosov vortex lattice and vortex clusters is reported, showing the first-order nature of the boundary line.

Keywords: vortex, superconductors, attractive interactions, neutron scattering

Online supplementary data available from stacks.iop.org/J.PhysCM/26/232201/mmedia

(Some figures may appear in colour only in the online journal)
type 1.5 superconductivity, was proposed to arise from two coherence lengths associated with each of the superconducting electron bands of MgB$_2$ [11]. A similar interpretation in terms of a multicomponent system was also addressed for Sr$_2$RuO$_4$ [12] and reviewed in [13]. Some debate exists, however, on the genuine interpretation of the observed topologies [6, 7, 14].

As a matter of fact, the mixture of Meissner and vortex states has been reported for small magnetic fields of tens of gauss or a few gauss. Most observations were made with surface studies (magnetic decoration, magneto-optical imaging or local SQUID magnetometry techniques), whose experimental resolutions are well suited for measurements at such low fields. Magnetic decoration studies with a single vortex resolution have been especially efficient in producing beautiful real space pictures of the intermediate mixed state [15], but understanding the origin of the observed structures is not straightforward. Measurements are made after cooling the sample from above $T_c$ with the applied field (field cooling (FC)). First, pinning effects cannot be excluded and may explain the existence of vortices under a very low field ($B < (1 - D)B_{c1}$ with $D$ the demagnetization coefficient) where they are normally not expected [16]. Under FC conditions, the observed structures are frozen at an ill-defined temperature, estimated to be close to $T(B_{c2})$ for low $T_c$'s [17]. This implies that the thermal variation of these structures is unknown and that the roles for pinning and thermodynamics are hard to quantify.

One of the known candidates for explaining the attractive interaction between vortices in a low $\kappa$ superconductor is the non-local correction. In a non-local model, the potential $A(r)$ is no longer simply proportional to the current density $J_s(r)$ (the position) [18]. One consequence is that the supercurrent efficiency is reduced compared to the pure local limit, and field penetration is allowed for lengths longer than the London penetration length $\lambda_L$. In a low $\kappa$ and single-gap superconductor, this non-local effect may cause an attractive interaction between two vortices for a certain $(B, T)$ range [19]. Interestingly, this non-local correction should no longer be effective close to the critical temperature where the London length reaches large values, and a phase boundary between the low temperature attractive region and the high temperature repulsive region is expected [19, 20]. Field- and temperature-dependent measurements are then required and are possible with small angle neutron scattering (SANS). SANS also has the advantage of being a bulk probe, allowing direct measurement of the lattice periodicity and of its disorder. The experimental resolution of SANS is, however, best suited for vortex lattices created by fields larger than typically 100 G. Neutron scattering experiments at very low fields have been scarcely reported. The intermediate state was observed at $T = 3.6$ K for magnetic fields between 100 G and 300 G in pure Nb cylinders [21]. Neutron grating interferometry has also been very recently used to visualize the morphology of heterogeneous vortex states for $B \geq 100$ G [22].

Here, we have used the new very small angle neutron scattering (VSANS) spectrometer très petits angles (TPA) at the Laboratoire Léon Brillouin (LLB; Saclay, France),
allowing us to reach low values for the scattering vector \( Q \), such as \( 6.10^{-4} \) Å\(^{-1} \) in our experiments with an optimized \( Q \) resolution [23]. We clarify how the vortex structures are formed at low temperature when the sample is field cooled. We report on the high temperature boundary for attractive vortices and show a phase coexistence between a vortex lattice with regular periodicity and clusters of attracting vortices.

A large slab of pure niobium (\( T_c = 9.17 \pm 0.05 \) K) was used for the neutron scattering experiment (dimensions: \( L = 35 \) mm, \( w = 17 \) mm, \( t = 1.5 \) mm). A small replica with the same aspect ratio was cut for magnetic characterizations in a Magnetic Property Measurement System (MPMS) SQUID magnetometer. Comparison of the sample characteristics with those in the literature [24, 25] indicates good purity and a small number of interstitial defects (\( B_{c2} = 2950 \) G, \( B_{c1} = 1400 \) G at \( T = 4.2 \) K consistent with \( \kappa = 0.9 \)). A significant critical current exists, however (\( J_c(4 \) K, \( 0 \) G) = \( 10^4 \) A cm\(^{-2} \)), largely due to the unpolished surfaces leading to important pinning [26, 27].

Due to the long strip shape of the sample, the first penetration field for vortices is largely lower than \( B_{c1} \) because of a significant demagnetizing field [28]. Measurements were performed using the VSANS spectrometer TPA at LLB. The originality of this new spectrometer is its multibeam collimator, which has a large number of very small drilled holes (about 1 mm in diameter) and which produces a small convergent beam at one point on the detector. This latter is an image plate produced by Marresearch. It is equipped with a neutron converter. It has very small pixels, 0.15 × 0.15 mm\(^2 \), i.e. it has high spatial resolution even at low \( Q \) [23]. The neutron wavelength used was 6 Å with a full width at half maximum distribution of 14%. The sample-to-detector distance was 6.187 m. The magnetic field \( B \) was parallel to the neutron beam and perpendicular to the large facets of the Nb sample. A magnetic field from 50 G to 200 G produced with a SpectroMag (10 T) superconducting magnet was applied. The background scattering was measured in the normal state (\( T = 10 \) K > \( T_c \)) and subtracted from the raw data in order to reveal the scattering arising from the superconducting structures.

A typical SANS pattern obtained just below \( T_c \) at a temperature of \( T = 8.5 \) K and with an applied field \( B = 100 \) G is shown in figure 1.

![Figure 1](image1.jpg)

**Figure 1.** Variation of \( Q_1 \) as a function of temperature for different applied magnetic fields (\( B = 75, 100, 125 \) and 200 G).

When the temperature decreases, \( T < 8 \) K, a second peak emerges at \( Q_2 \) (figure 3).

![Figure 2](image2.jpg)

**Figure 2.** Variation of \( Q_2 \) as a function of temperature for different applied magnetic fields (\( B = 75, 100, 125 \) and 200 G).

As \( T \) decreases, \( T < 5 \) K, a second peak appears at \( Q_2 \) (figure 3).

![Figure 3](image3.jpg)

**Figure 3.** Variation of \( Q_2 \) as a function of temperature for different applied magnetic fields (\( B = 75, 100, 125 \) and 200 G).

A typical SANS pattern obtained just below \( T_c \) at a temperature of \( T = 8.5 \) K and with an applied field \( B = 100 \) G is shown in figure 1.
we find \( Q_1 = 2\pi(B\rho_0)^{1/2} \) as expected for a square Abrikosov lattice. The \( Q_1 \) value at 75 G is the only one close to that of a hexagonal lattice. Both square and hexagonal lattices have been reported for high purity Nb close to \( T_c \), albeit at larger field for the square lattice [21, 31]. We will return to this point later. On reducing the temperature further, the intensity of the first peak at \( Q_1 \) decreases before vanishing, and only the second peak at \( Q_2 \) remains at low temperature as shown in figure 4.

We note that the peaks at both \( Q_1 \) and \( Q_2 \) are observed simultaneously over a substantial temperature range (figure 5), demonstrating a phase coexistence of two different lattices with different periodicities.

We report in figure 6 the \( Q_2 \) variation as a function of temperature for different applied fields (\( B = 75–200 \) G). Note that no scattered intensity (in excess of the background) was observed for \( B = 50 \) G, indicating that a full Meissner state was likely obtained.

All the curves in figure 6 are superimposed (within the error bars at high temperature): \( Q_2 \) is then field independent for the whole temperature range. The large value of \( Q_2 \) implies an attractive interaction between the flux lines. The corresponding periodicity, \( d_2 = 2\pi/Q_2 \), can be interpreted as the upper limit for the lattice spacing as expected in this regime [32].

A similar conclusion was reported from the analysis of the induction jump at \( B_{1c} \) in a (pinning-free) low \( \kappa \) superconductor with highly reversible magnetization [33–36] and from a neutron diffraction experiment for temperatures close to 4 K [8, 31]. Figure 7 shows the corresponding lattice periodicity \( d_2 \) as a function of temperature, independent of the applied field up to 200 G. The value \( d_2 = 1660 \) Å measured at 4 K is in good agreement with the values reported by Schelten et al. (=1800 Å) and Muhlbauser et al. (=1600 Å) [8, 31].

Theoretical calculations predict that the temperature dependence of the equilibrium distance between attracting vortices is governed by the London penetration depth [6, 33]. We tried the Casimir–Gorter variation \( y = (1 - t^3)^{-1/2} \) with \( t = T/T_c \) [37], and an analytic form \( y = (1 - t^{3-\gamma})^{-1/2} \), which closely follows the BCS variation [38]. As shown in figure 7, the latter expression corresponds well to the thermal variation of \( d_2 \) up to the temperature \( T \) where the \( Q_2 \) peak vanishes. In our experiments, the temperature interval over which the recorded data is relatively large and \( T \) is not very precisely determined. However, we found \( T/T_c \approx 0.87–0.92 \), in reasonable agreement with the boundary line between the dominating vortex attraction and the dominating vortex repulsion regimes predicted by non-local models [19, 20, 34]. The phase coexistence observed here is a bulk probe of the first-order nature of the boundary line between the two regimes, and is in agreement with interpretations of the magnetization discontinuity in superconductors with small \( \kappa \) parameters [34]. Since we observe that the attractive interaction disappears at high temperature, this cannot be the mechanism that stabilizes the square lattice at low field in our sample. The effect of fourfold Fermi surface symmetry is already relevant [39]. The differences in the lattice symmetry at low field and high temperature reported here and in [31] confirm the close competition between the cubic crystal symmetry and the tendency of the vortex to form a hexagonal lattice, as in niobium, and that small differences of quality between samples apparently change the dominant interaction.

Analysis of the \( Q_2 \) width shows a peak broadening much above the experimental resolution and then a non-perfect positional ordering. To extract a quantitative value of the positional correlation length \( r_2 \), the instrumental resolution has to be accounted for. The \( Q \) resolution of the non-standard multibeam VSANS instrument TPA and the calculation of \( r_2 \) are given in the supplementary material (stacks.iop.org/J.Phys.CM/26/232201/media). The correlation length \( r_2 \) is found to be close to 1.2 μm at low temperature (see figure 8). This is similar to the typical cluster size reported from magnetic decorations experiments [15]. We can then conclude that the bulk structures are not strongly different from the structures observed at the surfaces, and at least that the Landau branching of vortex domains at the surface is small. We observe that \( r_2 \) is roughly constant only below \( T \approx T_c/2 \), which is the temperature below which there are no significant changes of the condensate parameters, such as the penetration depth. Thus, no pinning effects are required to explain the quasi-constant cluster size of attractive vortices at low temperature.

Since our sample has a significant critical current, we have to discuss whether bulk pinning may contribute to disordering of the lattice and to the existence of vortex clusters, withstanding the attractive interaction. The dominant pinning mechanism in pure Nb is generally surface pinning [26, 27], which has no direct relation with a bulk disordering of the lattice [40]. As a consequence, significant critical current can be present without a direct relation with vortex disorder due to bulk pinning. This is the situation that makes most sense here considering the good bulk purity of our sample, which has superconducting parameters very close to the intrinsic values of Nb. We have also shown in this experiment that the clusters have characteristics that closely follow equilibrium theories. All this indicates that pinning plays only a minor role for stabilizing the vortex structures observed here.

In conclusion, we have measured the temperature dependence of clusters formed by vortices under attractive interaction using neutron scattering. This interaction is no longer effective close to \( T_c \), showing that it originates from non-local effects. The phase coexistence between the Abrikosov lattice and vortex clusters demonstrates the first-order nature of the transition. Since the temperature dependence of the vortex periodicity in these clusters follows that of the penetration depth, pinning does not play the leading role in the stabilization of the vortex cluster structures observed at low temperature and low field.

References

[1] Abrikosov A A 1957 Sov. Phys.—JETP 5 1174
[2] Trüible H and Essmann U 1967 Phys. Status Solidi A 20 95
[3] Krägeloh U 1969 Phys. Lett. A 28 565
[4] Eilenberger G and Butner H 1969 Z. Phys. 224 335
[5] Halbritter J 1971 Z. Phys. 243 201
[6] Leung M C 1973 J. Low Temp. Phys. 12 215
[7] Brandt E H and Das M P 2011 J. Supercond. Novel Magn. 24 57
[8] Babaev E and Silaev M 2013 J. Supercond Novel Magn. 26 2045
