

VORTEX MATTER, DYNAMICS AND PINNING IN SUPERCONDUCTING MATERIALS

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**UNIVERSITY OF
BIRMINGHAM**

Body of work submitted to The University of Birmingham
for the Higher Doctorate degree of

DOCTOR OF SCIENCE

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Synopsis

The enclosed body of work represents a compilation of published papers covering work carried out in the Universities of Birmingham, Bath and Southampton, UK, in the Second University of Rome “Tor Vergata”, Italy, in the Nanoelectronics Research Institute (former Electrotechnical Laboratory) of AIST Tsukuba, Japan, and in the National Institute of Materials Physics (former Institute of Physics and Technology of Materials), Bucharest, Romania, during my research career over more than twenty years. The thesis comprises mainly research work on science and technology of high temperature superconducting cuprates of various compositions (Y-based, Bi-based, Tl-based, Hg-based, (Cu,C)-based) and morphologies (polycrystalline, single crystals, thin films, tapes, artificial superconducting superlattices). The major theme common to most of the research done on the above-mentioned materials is the study of vortex matter, dynamics, and pinning, which determine a large number of superconducting properties for various applications.

The submitted work presents the results of experimental and fundamental studies of superconducting materials in six main areas. These comprise: (i) synthesis and characterization (current-voltage characteristics, dissipation mechanisms, interaction between inter-and intra-grain vortices) of Y- and Bi-based superconducting ceramics; (ii) experimental and theoretical studies of current-induced unbinding of thermally-created vortex-antivortex pairs; (iii) fabrication and characterization of artificial superconducting superlattices; (iv) fabrication and characterization of anomalous superconductors, two- and multi-component superconductors, and exotic vortex matter; (v) study of vortex matter and dynamics by Scanning Hall Probe Microscopy; and (vi) science and technology of vortex pinning, including through self-assembling nanotechnology of pinning centres. There are a small number of publications that do not fall in the above-mentioned areas, but they are also in the larger field of science and technology of superconducting materials. The papers are presented essentially chronologically, with no attempt to group them into separate research areas, since the common philosophy and approach to the investigation of various superconducting materials is to determine how vortex matter, dynamics and pinning influence their physical properties.

The first group of papers [2-4, 6-9, 11-12, 15] concerns the fabrication and studies of superconducting properties from transport measurements of Y- and Bi-based polycrystalline superconductors, in the first decade after the discovery of high critical temperature

superconductivity, when many groups around the world were deeply involved in the comprehensive research of these materials and corresponds to the subject of my PhD studies (work done in Bucharest). In their ceramic form, in Y- and Bi-based cuprates the factor that is limiting the transport current capability is the thermal activation of inter-grain Josephson vortices [3,4], the peak-effect in the magnetic field dependence of the transport critical current density is due to the attractive interaction between the intra-grain Abrikosov vortices and the inter-grain Josephson vortices [9], while in moderate magnetic fields the current voltage characteristics are well described by a modified Kim-Anderson model [6].

The second group of papers [10, 13, 16-17, 19] concerns the investigation of dissipation in zero applied magnetic field in various superconducting materials and various morphologies (single-crystals, epitaxial thin films, ceramics with and without preferentially-oriented grains, mono- and multi-filamentary tapes). The work on the subject started in Rome in 1994, and continued in Bucharest and Southampton for other 3 years. Even in zero applied magnetic field, the critical current density was found to be much smaller than the de-pairing critical current. The reason for this fact is that, due to higher thermal energy available around liquid nitrogen temperature, vortex-antivortex pairs are spontaneously created, and a transport current in the system leads to dissipation due to the Lorentz force. From I-V characteristics of Bi:2212 films, we showed that the power law valid for 2D systems does not correctly described the data, and interlayer coupling must be taken into account. We observed a cross-over from 2D to 3D behaviour as the current is reduced. Furthermore, by annealing treatments in redox atmospheres, we changed the interlayer coupling, resulting in an increase of the J_c and a decrease of anisotropy factor with increasing O_2 content in the film. We quantitatively evaluated these effects in terms of current-induced unbinding of the thermally-created vortex-antivortex pairs in a quasi-2D approach. However, at T close to T_c and in the low-voltage region, we observed an additional dissipation, which we explained in terms of fluctuations of the phase of the order parameter [10]. We also found that vortex fluctuations are limited also by size effects and by additional pinning. As a consequence of these findings, I was invited to write a chapter on this subject in a book for an international book series published by Nova Science Publishers [24].

The third area of work involves the fabrication and characterization of $(BaCuO_2)_m/(CaCuO_2)_n$ artificial superconducting superlattices grown by layer-by-layer PLD [20-23], the work being done in Rome (1998-1999) under a contract with the National Institute of Physics of Matter, Italy. The thickness of the Charge Reservoir Layer, and the number of superconducting CuO_2 planes could be adjusted at will, and allowed the growth of artificial superconductors that can not be obtained in normal conditions, hence allowed us to play with

various superconducting parameters for fundamental research. From various transport measurements, I have clarified for the first time the peculiarities of the vortex dynamics, melting transitions, anisotropy and critical current densities in these artificial structures [21], and have demonstrated that the unusual properties of these artificial superlattices (high value of resistivity at room temperature, negative curvature and pronounced rounding well above the transition of $R(T)$ experimental curves, broadening of the transition and strong enhancement of fluctuations contribution to conductivity) can be explained if the high degree of structural disorder is taking into account [22]. Furthermore we have shown that normal state resistivity behaviour, enhanced thermodynamic fluctuations, decrease of T_c can be explained by a single numerical value of the localization parameter $\varepsilon_{FT} \approx 3\hbar$.

The fourth area of work involves the fabrication by high-pressure synthesis and characterization of anomalous, exotic superconductors in the (Cu,C)-, Tl-, Hg-, and F-doped cuprate families [26, 35-37, 56-58, 71, 73-78, 80-87, 92-93, 98]. The experimental work was done in Tsukuba (2000-2002, and 2006-2007), while some of the results were analyzed (and papers submitted) in Bucharest, Bath and Birmingham. I have imagined and developed a new, straight-forward method for determining the vortex melting lines by using the third-harmonic susceptibility response [58]. Since most of these superconductors are two-component (two-gap), they have a number of quite exotic properties. For example, due to the very small inter-band coupling, they may have an anomalous vortex melting line [71], or they may have superconducting CuO₂ outer planes and antiferromagnetic CuO₂ inner planes without the destruction of superconductivity along the c-axis [73]. I have discovered two new phases of vortex matter: magnetically-coupled pancake-vortex molecules in super-multi-layered Hg-based cuprates [80] and non-axis-symmetric (non-Abrikosov) vortices composed of two fractional flux quanta glued by a inter-band phase difference soliton [74]. For the last case, theoretical models and complex phase diagrams were proposed [75, 78, 98, 113]. After the discovery of the new pnictides and chalcogenides superconductors, the interest in two- and multi-component superconductors has increased, so we continued our theoretical analysis [105,107,117] of such materials, based on our previous work on *i*-solitons; most important results being on topological structures of solitons and two-band superconductors, and chiral domains in multi-band superconductors.

The fifth group of papers [59-63, 67-70, 79, 116] concerns the study of vortex matter and dynamics by Scanning Hall Probe Microscopy, the experimental work being done in Bath (2002-2004), while some of the later analysis of results and papers writing being done in Birmingham. We have observed and explained a local hysteresis inversion near the edge of the film [69-60], we have investigated the flux structures in mesoscopic discs [68],

interacting crossing vortex lattices in single crystals in the presence of quenched disorder [61] and in thin films [69], and flux lenses effects [62]. We have also observed and modelled flux channelling and streaming in superconducting films with regular array of micron-sized antidots (holes) [67] and demonstrated the manipulation of pancake vortices by a rotating Josephson vortex lattice [79]. In $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ thin films in very low perpendicular magnetic fields SHPM showed vortices that are intermittently trapped on strong pinning centres. This state shares many of the signatures of the re-entrant vortex liquid phase that has been theoretically predicted in these highly anisotropic materials at very low vortex densities. This is the first direct experimental evidence for the existence of a dynamic liquid-like vortex state in this highly anisotropic material at very low magnetic induction [116].

The final research area concerns the science and technology of vortex (flux) pinning. In applied magnetic fields, superconducting materials are subject to additional dissipation due to Lorentz forces, hence their critical current density decreases strongly with increasing magnetic field. To limit this decrease (*i.e.*, to increase critical current in magnetic fields) artificial defects (pinning centres) are needed. These strong artificial pinning centres were introduced (and their influence studied), by doping/substitutions with BaZrO_3 [29-31, 38], Zn [39], LiF [54-55] or by heavy-ion or neutron irradiation [43-47], the work being done in Bucharest and Tsukuba (2000-2002). The most significant result in this research area is the invention of self-assembled nanotechnology of pinning centres in superconducting films and devices, which led to a new field in the science and technology of superconducting materials, and to winning of a Marie Curie Excellence Grant with University of Birmingham as host institution. This was the first cost-effective method for introducing extended, correlated artificial pinning centres in films, which led to impressive increase of critical current and other superconducting properties in magnetic fields. The principle of the idea is that by growing an incomplete layer of nano-islands prior to the films deposition, correlated, extended defects that act as very efficient pinning centres form on top of nano-islands. Several variations of the patented method are now used. The work started in Japan in 2001 on Tl-based films [32-33, 52-53] and continued in Birmingham (2007 - till now) on YBCO films using various materials as nanodots, and several variations of the initial approach [72, 88-90, 94-97, 99-104, 106, 108-112, 114, 115]. Due to the catalytic effect, noble-metal nano-dots in substrate decoration and quasi-multilayers approaches allowed the growth of thick YBCO films (4-6 μm) with a less pronounced decrease of J_c with thickness as compared with pure YBCO films, allowing critical currents as high as 800-900 A/cm-w in self field and 77K [95, 97, 104], very close to the best results in the world (USA, Japan) obtained after many years of research, in large, very well funded laboratories or companies. The improvement in

applied fields is even larger. Since for cost-effective, energy-efficient electro-technical devices made from coated conductor tapes, the parameter of interest is I_c (superconducting film thickness is negligible in comparison with the substrate thickness), this will have a big impact for the future applications of superconductors. In addition to alleviating the thickness problem, noble metals nanodots promote a closely-packed columnar growth of YBCO, with column boundaries acting as extended, 2-dimensional pinning centres [94, **106**]. The columns diameter (and, consequently, the density of the pinning centres) is well correlated with the dimensions and surface density of the noble metal nanodots. The best results occurred from combining Ag nanodots in substrate decoration and quasi-multilayers with nanocrystalline targets containing 2-4% wt. BaZrO₃ (BZO) [109, 114], and showed that Ag helped maintaining a very high T_c (in YBCO/BZO nanocomposite T_c is depressed by few degrees) and promoted the growth of YBCO nanocolumns (nanothreads) entangled with BZO nanorods. The resulting thick nanocomposite films have large critical current both in self field and in applied fields (European record, to our best knowledge) and have a very small anisotropy of critical current for a broad range of applied fields, which is very important in practical applications. As for the theoretical contributions to the study of artificial pinning centres, we have shown [90] that the current dependence of the pinning potential in superconducting nano-composites are not described by the Anderson-Kim or collective pinning models, but instead it is described by the logarithmic law as proposed by Zeldov.

The published work comprises 117 papers, the full list being presented before copies of individual papers.

The most important papers, which made the largest impact and contribution to the development of the knowledge and technology in the field, are marked by bold fonts.

Statement indicating the nature and contribution in papers involving joint authorship.

1. I am single author of papers 5-7, 9 and 24
2. As first (principal) author of papers 4, 8, 12-13, 15-16, 19, 21, 23, 26, 28, 32-33, 39, 43-44, 52, 57-60, 67-69, 71-74, 79-82, 88, 90-91, 96, 100, 103, 113, 116, I had the ideas, performed all or part of the measurements and data analysis, wrote the largest parts of the papers, submitted (and revised when needed) for publications. I contributed less to sample preparations.

3. As Group Leader (in Bucharest) or Marie Curie Team Leader (in Birmingham), papers 29-31, 34, 38, 54, 65, 89, 94-95, 97, 101-102, 104, 106, 108-112,114, I coordinated the research, performed in some cases measurements, performed data analysis, wrote or supervised/corrected the final version of the paper.
4. As member of research groups I had contributions to measurements, usually all the data analysis, and I contributed to paper writing (in Japan, I usually wrote/corrected the final version of the papers): 1-3, 10-11, 14, 17-18, 20, 22, 25, 27, 35-37, 40-42, 45-51, 53, 55-56, 61-64, 66, 70, 75-78, 83-87, 92-93, 98-99, 105, 107, 115, 117.

LIST OF PUBLICATIONS

D. Sc.

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