Nr. 46
Juli 2015

SPG MITTEILUNGEN
COMMUNICATIONS DE LA SSP

Progress in Physics (48)

Microstructured crystals of correlated electron systems

Philip J. W. Moll [1,2,3], Bertram Batlogg [3]
[1] University of California, Berkeley, USA
[2] Max-Planck-Institute for Chemical Physics of Solids, Dresden, Germany
[3] ETH Zürich, Switzerland

Progress in Physics (48)

Microstructured crystals of correlated electron systems

Philip J. W. Moll [1,2,3], *Bertram Batlogg* [3]
[1] *University of California, Berkeley, USA*
[2] *Max-Planck-Institute for Chemical Physics of Solids, Dresden, Germany*
[3] *ETH Zürich, Switzerland*

Introduction

When novel classes of materials are discovered that capture the interest of the condensed matter physics community, such as for example the iron-based high temperature superconductors in 2008 [1], the first experimental step towards understanding their physics is to establish their basic physical properties. Unfortunately these initial measurements are inevitably plagued by materials shortcomings, such as only minute crystal size, compositional inhomogeneity or presence of parasitic phases. Quite often, the pioneering measurements are later superseded by results on large and compositionally homogeneous crystals as the product of extensive synthesis efforts.

This traditional and time consuming approach can now be supplemented thanks to new tools developed for the micro- and nanosciences. Micro-analysis and –manipulation methods may be used to identify, separate and extract even micron-sized grains of high phase purity from the powder products of the first synthesis. We have developed and employed high-precision methods based on a Focused Ion Beam (FIB), mainly, mainly used in the semiconductor industry, as powerful tools to fabricate single-crystal microstructures from small grains of correlated electron compounds. The shape and electrical layout of the samples are tailored with sub-micrometer precision in order to tackle a particular scientific question.

When such small structures are prepared, the FIB technique is used in two main modes of operation: micro-cutting (milling) and deposition of electric contacts. The first mode starts with cutting a regular shape suitable for electric transport or micro-magnetic measurements out of a crystalline grain. Ga^{2+} ions are accelerated to 60 keV and focused to a nm-sized beam-spot on the target material. The focus spot can be precisely positioned on the sample surface where it locally removes the sample material. The sample may be rotated in situ around all axes and thus a properly oriented rectangular slab can be carved from any oddly-shaped crystallite. This pre-cut sample is then transferred onto a substrate with lithographically prepared contact pads.

In the second mode of operation, electric contact is made to this pre-cut slab. A metal-organic precursor gas containing Pt is introduced into the chamber and adsorbes onto the surface. The impinging ion beam destroys the organic molecule, binds the platinum to the surface and evaporates parts of the organic compounds into the chamber vacuum. Over time, the irradiated area is covered by a metallic film consisting of about 45% platinum, 50% carbon, and 5% gallium from the beam [2]. Alternatively, superconducting tungsten-based films with a T_c ~6 K can be deposited and open a new path to design superconductor-quantum material structures such as proximity-effect devices or phase-coherent loops with unconventional superconducting segments. This mask-less contacting technique usually leads to low contact resistances as the energy of the ion beam typically leads to the destruction of any potential surface barrier such as an oxide layer. In the final step, the sample is again milled into any shape desirable for a transport experiment such as multiple four-point structures along any selected crystal orientation, or a Hall-effect geometry. A few examples of the variety of shapes and sizes are shown in Figure 1.

$(V_2Sr_4O_6)Fe_2As_2$: A new class hosting intrinisic Josephson junctions

The discovery of the iron-based superconductors as the second class of high-T_c materials after the cuprates opened a new path to deepen and expand concepts about high-temperature superconductivity by comparing and contrast-

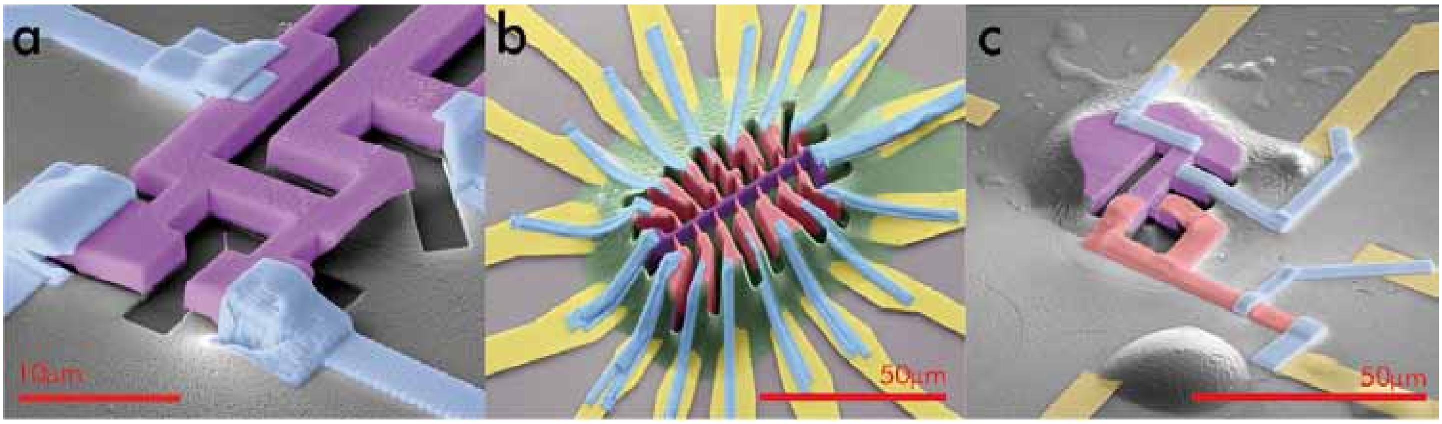


Figure 1 : Example of Focused Ion Beam microstructured crystals. a) Resistivity anisotropy device made from the heavy-fermion superconductor URu_2Si_2. b) Iron-based superconductor SmFeAs(O,F) with voltage probes every 3 µm to search for doping inhomogeneity. c) phase-coherent superconducting loop between LaFeAs(O,F) crystal (purple) and FIB-deposited lead (red).

ing these two distinct classes of materials. Both are layered compounds with pronounced electronic anistropy, and highlight the apparent importance of low-dimensionality or layeredness for high transition temperatures. In those cuprates with most pronounced electronic anisotropy the superconducting coherence length perpendicular to the layers is well known to be shorter than the interlayer distance, forming Josephson junctions within the crystal lattice. This intrinsic Josephson coupling is in many ways similar to that in artificially grown Josephson junctions. In particular, an ac-Josephson effect is observed when a constant voltage is sustained across a junction, leading to an oscillation of the relative phase between adjacent layers with a frequency given by the Josephson constant $K_J = 2e/h \approx 483.6$ GHz/V. This natural frequency range around 100 GHz - 1 THz is also interesting from a practical point of view as applications based on the intrinsic Josephson effect in cuprates have been shown to effectively generate radiation in this technologically important frequency range [3].

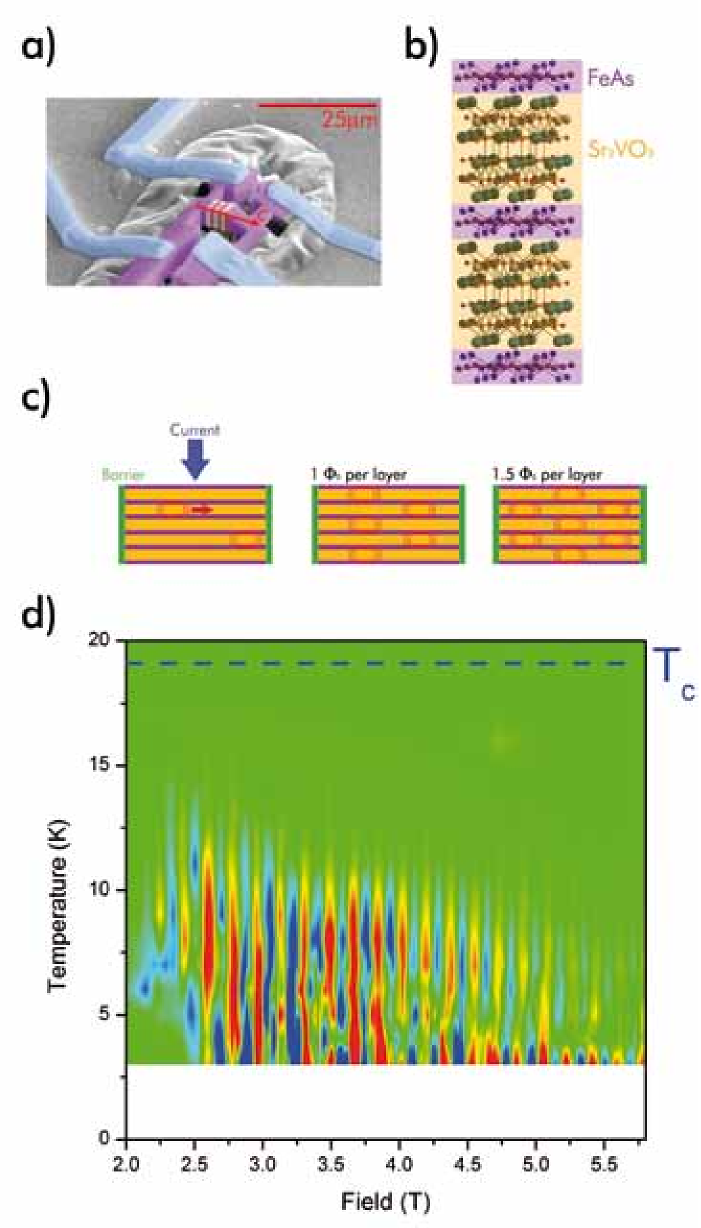


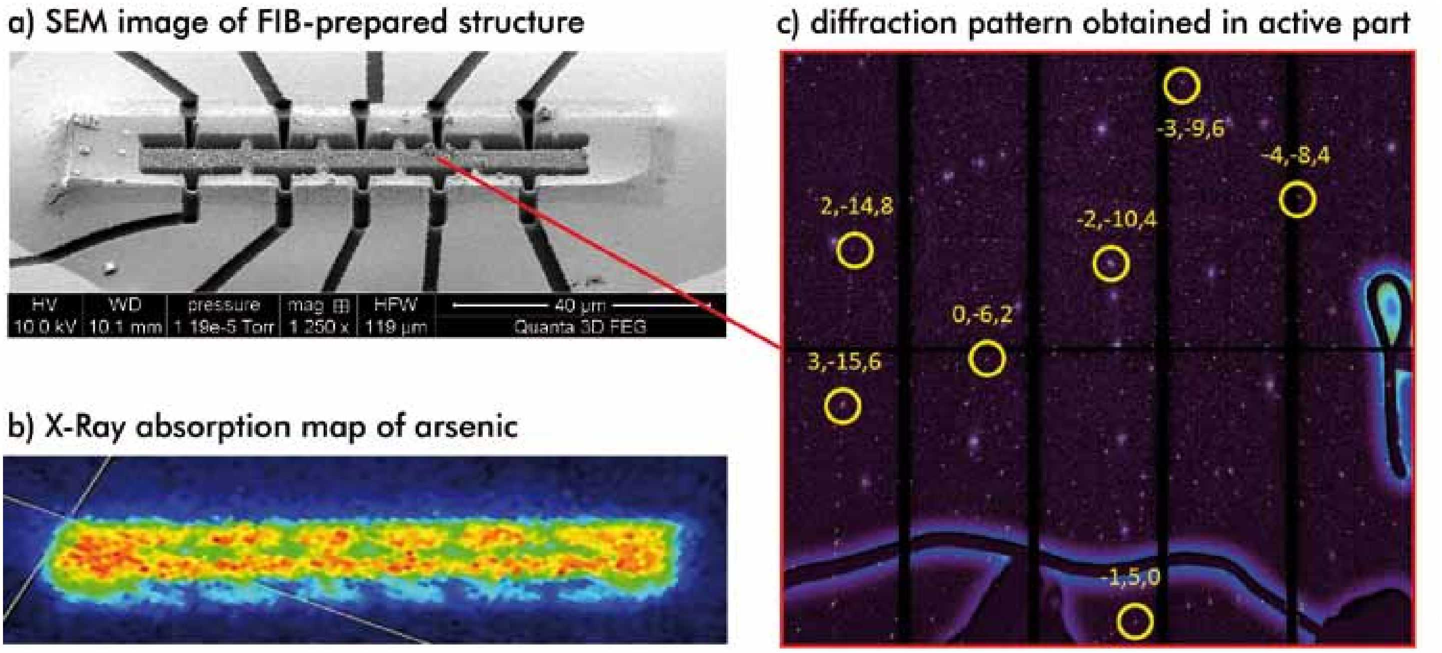
Figure 2 : Intrinsic Josephson Junctions in a FeAs-based superconductor. (a) Microstructured crystallite and (b) crystal structure of $(V_2Sr_4O_6)Fe_2As_2$. (c) Stable vortex configurations at integer numbers per double layer, that quantitatively explain the periodic modulations of the critical current $j_c(H)$ shown in (d).

With the advent of iron-based superconductors as another family of anisotropic, layered high-T_c materials [4–7], the investigation of their potential for THz applications shifted into focus as well. The most anisotropic Fe-superconductors consist of FeAs layers separated by a double-layer of Sr_2VO_3. Even as this insulating layer is comparatively thick (0.8 nm), the electronic anisotropy remains moderate ($\gamma \approx 51$) and suggest possible Josephson coupling. Unfortunately, the growth of large-scale single crystal of such a chemical complexity proves very difficult. Using the FIB technique, we have successfully identified a single crystal grain of high quality within a larger piece, extracted it and fabricated it into the microstructure optimized for four-point resistance measurements shown in Figure 2 [8].

If a magnetic field is applied parallel to the FeAs-planes, in-plane vortices of Josephson character form in-between adjacent layers. Unlike previous samples made from macroscopic crystals, this microstructure was of such high crystal quality that the vortices themselves form a regular lattice with a lattice spacing given by the applied magnetic field. Whenever the vortex lattice is commensurate with the crystal lattice, i.e. the vortex lattice vector is an integer multiple of the c-axis unit cell size of the $(V_2Sr_4O_6)Fe_2As_2$ crystal structure, the configuration is stable and the critical current reaches a maximum. The periodicity is linked to the crystal structure and can be well explained quantitatively by the theory developed for the intrinsic Josephson effect in the cuprates. Thereby we were able to show that the intrinsic Josephson effect exists in iron-based superconductors. In the future, we plan to investigate the efficiency of the emission of THz radiation from such prepared structures as well as investigate the theoretically predicted peculiarities of the vortex matter that arise from the additional Josephson tunneling channels expected in these multi-band superconductors. In particular, study of the Leggett-mode excitation in the vortex system may be possible [9,10].

Microanalysis

It is not always the case that the experiment itself provides such clear evidence of the crystallinity and phase purity as the previous example in $(V_2Sr_4O_6)Fe_2As_2$. In particular complex materials tend to form other stable compounds as impurity phases besides the desired phase in the growth product. These can be mixed into a piece of material on the micron scale, and thus parts of such a microstructure may consist of a different composition than the material of interest. We are thus exploring for new ways to confirm the crystal structure and composition within a microstructure in collaboration with the group of James Analytis at the University of California, Berkeley, and the Advanced Light Source (ALS) at the Lawrence Berkeley National Lab (LBNL). In particular, the high luminocity of X-Ray radiation now available from synchrotron sources allow fast X-Ray-diffraction experiments with sub-micrometer spot sizes. Figure 3 shows recent results of micro-diffraction experiments performed on a FIB prepared microstructure of the Dirac-semimetal Cd_3As_2. A scanning micrograph map of the crystal quality and lattice parameters within the electrically active regions between the voltage contacts can be obtained with high special resolution.


a) SEM image of FIB-prepared structure

b) X-Ray absorption map of arsenic

c) diffraction pattern obtained in active part

Figure 3 : A FIB prepared microstructure seen in different ways: SEM image, X-Ray absorption map and diffraction pattern. Local crystallographic analysis can clarify the phase and crystal quality of the actual micron-sized crystal piece under investigation and thus exclude extrinsic effects such as parasitic phases.

With these recent advances in microstructure fabrication and analysis experiments probing their electronic properties on the micron-scale in as-grown materials become possible that now complement experiments based on thin-films. Advances in material research are necessary to address technological challenges, from energy to information technology, yet understanding the behavior of these new materials on the sub-micron length scale at which they will be implemented into devices is equally important. High quality thin-film growth is an essential step for the eventual introduction of novel materials into technology, however its development is a time and cost intensive endeavor. Focused Ion Beam microstructuring, on the other hand, is a simple and fast process that is routinely adapted to new materials within days. Therefore this technique is well suited to bridge the gap between the forefront of materials science, fundamental condensed matter physics research and applications.

We thank our colleagues R. Puzniak, K. Rogacki, J. Karpinski, N. D. Zhigadlo, S. Galeski, J. Reuteler, K. Kunze, P. Gasser, F. Balakirev, B. Zeng, L. Balicas, E. Bauer, F. Ronning, R. McDonald, N. Nair, T. Helm, A. C. Potter, I. Kimchi, A. Vishvanath, J. Analytis, N. Tamura, X. Zhu, P. Cheng, H.-H. Wen for their important contributions.

References

[1] Takahashi, H. et al. Superconductivity at 43 K in an iron-based layered compound LaO(1-x)F(x)FeAs. *Nature* **453**, 376–8 (2008).

[2] Langford, R. M., Wang, T. X. & Ozkaya, D. Reducing the resistivity of electron and ion beam assisted deposited Pt. *Microelectron. Eng.* **84**, 784–788 (2007).

[3] Ozyuzer, L. et al. Emission of coherent THz radiation from superconductors. *Science* **318**, 1291–3 (2007).

[4] Moll, P. J. W. et al. High magnetic-field scales and critical currents in SmFeAs(O, F) crystals. *Nat. Mater.* **9**, 628–633 (2010).

[5] Moll, P. J. W. et al. Critical Current Oscillations in the Intrinsic Hybrid Vortex State of SmFeAs(O,F). *Phys. Rev. Lett.* **113**, 1–5 (2014).

[6] Jaroszynski, J. et al. Upper critical fields and thermally-activated transport of Nd($O_{0.7}F_{0.3}$)FeAs single crystal. *Phys. Rev. B* **78**, *174523* (2008).

[7] Moll, P. J. W. et al. Transition from slow Abrikosov to fast moving Josephson vortices in iron pnictide superconductors. *Nat. Mater.* **12**, 134–8 (2013).

[8] Moll, P. J. W., Zhu, X., Cheng, P., Wen, H.-H. & Batlogg, B. Intrinsic Josephson junctions in the iron-based multi-band superconductor ($V_2Sr_4O_6$)Fe_2As_2. *Nat. Phys.* **10**, 644–647 (2014).

[9] Ota, Y., Machida, M. & Koyama, T. Macroscopic quantum tunneling in multigap superconducting Josephson junctions: Enhancement of escape rate via quantum fluctuations of the Josephson-Leggett mode. *Phys. Rev. B* **83**, 060503 (2011).

[10] Leggett, A. J. Number-Phase Fluctuations in Two-Band Superconductors. *Prog. Theor. Phys. 36*, 901–930 (1966).

The basic steps of the FIB technique

The Focused Ion Beam (FIB) is an imaging and machining tool with sub-μm precision, commonly used in materials science, physics, biology and semiconductor industry. Most machines use Ga ion beams that can be raster-scanned across the surface. The impacting ions interact with the target and locally sputter the material with high spatial resolution, rendering the FIB a versatile tool for mask-less microstructuring. In addition, a variety of metalcontaining precursor gases can be introduced into the chamber (most common are Pt, C, W). The ion beam interaction releases the metal ion from the organic gas molecule, leading to the growth of thin, conductive films.