Type-II superconductors, vortex lattice and role of an Indian scientist

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More than 40 years ago, research on superconductivity in India was in a rudimentary stage and mostly confined to theoretical and empirical studies. A few Indian scientists were engaged in research on superconductivity abroad, mainly in Europe and USA. The first indirect measurement of the vortex lattice structure inferred from neutron diffraction experiments of superconducting niobium (Nb) was reported[1] in 1964 by a group of four researchers from Saclay, France. Here lies a story of an Indian scientist and experimentalist. In fact, Madhav Rao from the then Atomic Energy Establishment, Trombay (AEET) was one of them. He was then visiting the Service de Physique du Solide et de Resonance Magnetique, Centre d'Etudes Nucleaires de Saclay (S&C), France and was actively engaged in experimental research. The superconductivity community of the world also learnt about this experiment from two contemporary conferences held in the later months of the year, where Cribier and collaborators presented two papers[2,3]. To understand the importance of the above experiment, it is worth recalling first the behaviour of a type-II superconductor in the Shubnikov phase and then how neutron diffraction technique helps in understanding the properties of materials.

In 1933, Meissner and Ochensfeld showed that in the presence of a small magnetic field the transition from normal to superconducting state took place with complete expulsion of magnetic flux from the bulk of the sample (Meissner effect). In pure metals, such a state of complete diamagnetism persisted until the external field reached the critical thermodynamics value (H_c) when abrupt field penetration occurred and the material reverted to normal state. In 1935, Shubnikov observed that the Meissner effect and ideal diamagnetism were in general observed in lead–thallium ($PbTl_2$) alloy only up to a much smaller field (H_{c1}). Beyond that the field penetration was gradual and it continued until a much higher critical field (H_{c2}) was applied, when bulk of the material turned normal. The state in which a superconducting alloy remains in the field region between H_{c1} and H_{c2} is referred to as the Shubnikov phase.

In 1950, Ginzburg and Landau examined superconducting transition as a phase transition of the second kind and introduced a formal order parameter which characterizes this transition. In the case of a phase transition, a change in the free energy near the transition temperature may be expanded into a series in terms of the order parameter. In 1957, Abrikosov investigated the case when the ratio of two characteristic lengths, defined in the above parameter, was large instead of small. This led to a negative surface energy, so that the subdivision into domains would process until it is limited by a fixed microscopic length, below which the gradient energy term would become excessive. Abrikosov called these superconductors as type-II, to distinguish them from the classical (type-I) superconductors. He showed that in the Shubnikov phase, the flux should not penetrate in laminar domains but in a regular array of flux tubes, each carrying a quantum of flux. Within each unit cell of the array there is a vortex of supercurrent concentrating the flux towards the vortex centre.

Neutron diffraction or scattering is a technique that reveals the position and structure of atoms in a material. When a beam of neutrons is directed at a given material, the neutrons are scattered by the atoms in the sample being investigated. From the change in the direction of neutrons, depending on the position of the atoms which they hit, and a diffraction pattern of the atoms, position and structure of the atoms can then be obtained. Understanding the exact position of atoms in a material and interactions among them is the key to understanding the properties of materials.

Due to unfortunate isolation of Soviet science from the rest of the world, the experiment of Shubnikov (1935), theoretical explanation of Ginzburg and Landau (1950) and of Abrikosov's extension (1957) were not appreciated in the West for sometime. The work of Soviet physicists was studied systematically in France under the leadership of theorist Pierre-Gilles de Gennes around early 1960s. Physics research in France in those days was not significant, except a few branches of physics, e.g. nuclear physics led by Irene and Frederic Joliot-Curie. Saclay, Grenoble, Paris and other smaller centres sometimes performed extremely sophisticated nuclear physics experiments during that period.

Low-energy neutron diffraction and other cutting-edge technologies were used in Saclay to solve research problems of solid state, atomic and nuclear physics. In Saclay, the neutron source was the reactor EL3, with a cold moderator which increases the flux of long-wavelength neutrons. The neutron beam passes a 20 cm beryllium (Be) filter, which filters out neutrons with a wavelength smaller than 4 Å. The neutron, being sensitive to the magnetic field, will be diffracted by the regular structure of the field which is present in the Shubnikov phase. Considering this, de Gennes and Matricon[4] suggested to check the existence of a regular array of vortex lines in a type-II superconductor by neutron diffraction or scattering studies.

In the 1960s, research on superconductivity gained momentum and entered into a new era known as the golden era of superconductivity. During this period experimental findings from different countries nourished the progress of the field. At this juncture, Cribier *et al.*[1] reported the discovery of the vortex structure of type-II superconductors by low-energy neutron diffraction studies. Although Abrikosov predicted a square array, later the neutron diffraction studies of the above group showed that a triangular array should have a slightly lower free energy. Cribier and collaborators[5,6] reported this at a conference on magnetism and magnetic materials in USA. They were not only the first to confirm Abrikosov's prediction by experimental observation of the structure of the vortex lattice, but they also made a small numerical correction in the theory so that it could satisfy all conditions and become universally acceptable (Box 1).

Madhav Rao visited Saclay in the early 1960s to acquire knowledge of the sophisticated technologies used in the then nuclear physics experiments. He was not only an author of the landmark paper, but also all the related papers[2,3,5,6] of the group bear his name. Thus, it could be ascertained that the success of the experiments was not due to the contribution of any single individual, but the result of a col-

Box 1.

L. Madhav Rao wrote:

'The day I joined Saclay, Jacrot asked me whether I had worked on superconductivity and I said, no. By then I was able to converse fluently in French (after spending three full months at the Alliance Francaise at Paris), which helped immensely in building an instant rapport with not only Jacrot but also with Cribier, Farnoux, etc. I was asked to spend the next ten days at the library to soak in as much as I could on superconductivity. A fortnight later Jacrot called me into his cabin and said that I should attend a briefing by de Gennes to take place the next day in his cabin. Beside the four of us, there was Herpin (boss of Jacrot and the one who gave the first theoretical interpretation of the helical magnetic structure in $MnAu_2$).There were two theoretical models on the nature of the Schubnikov phase: the vortex structure by Abrikosov and the laminary structure by B. B. Goodman, a British physicist working at Grenoble (Gorter too had proposed independently the laminary structure.). de Gennes, in his inimitable style spoke at length on these models and urged us to resolve this issue by doing a small-angle neutron diffraction experiment on a type-II superconductor. Compounds were out of question because of their large kappa values, so we had to choose only a type-II superconductor element, which are only two: Nb and V. Nb was the obvious choice. This was not magnetic scattering in the conventional sense, for here, neutrons had to be scattered by magnetic lines of force and not moments, for which there was no theoretical backing and we had no idea at all about the cross-sections involved. Herpin interjected, saying he will come up with a cross-section expression in a couple of days and he was true to his word! Putting the right numbers and the expected cold neutron flux at EL3 reactor, we found that it was feasible. We realised that we needed to have a very tight collimation of 1.5 minutes of arc before and after the sample. We could not achieve this from the conventional Soller collimators due to total reflection from cadmium! I suggested to Jacrot, brashly I thought then, that we should instead think of etching very fine horizontal slits on a Cd-coated Al plate and place a series of them over a metre length (both before and after the sample) to achieve this resolution. Fortunately, for me, Jacrot caught onto it and we got what we wanted (It was very generous of Jacrot to have acknowledged my role in this whenever he had the occasion to mention it). But our woes did not end here. At first we took a polycrystalline sphere of Nb and no matter what the field was, we got only one peak. We quickly realised that what was happening was that the vortex structure was getting pinned onto the defects and hence not responding to the external magnetic field. We then replaced this by a series of very well annealed single-crystal cylinders of Nb (to keep the demagnetizing coefficient to a minimum) and got those thrilling results. Since our set-up was not automated, we had to gather data manually round-the-clock, and since I was the only one who was unmarried in this group of four, I gladly opted to stay at the set-up during nights. We all wanted to publish our exciting results (which clearly vindicated Abrikosov) in *PRL*. Anatole Abragam, who was the super-chief, vetoed it. Jacrot told me that around that time *PRL* had rejected one of Abragam's paper and he was very furious with the Editorial Board of *PRL*. He compelled us to publish it in *Physics Letters* and in French! Gorter was the Chief Editor of the series *Progress in Low Temperature Physics*, published annually by North-Holland. Gorter invited us to write a full account of our work in this series and it appeared as Chapter IV in volume V as 'Study of the superconductive mixed state by neutron diffraction' by D. Cribier, B. Jacrot, L. Madhav Rao and B. Farnoux.

lective effort. The initial paper[1], which was published in French has more than 100 citations till date as recorded in the *Web of Science* database. Due to oversight, the note indicating the affiliation of Madhav Rao did not appear in the first page (p. 106) after the title of the paper. It finally appeared in the Errata (p. 318) of the same volume of the journal. Later, vortex lattice was observed directly by Essaman and Traüble in 1967 using decorative methods consisting of deposition of small ion particles on a superconductor in a magnetic field. After 25 years, scanning tunnelling microscope measurements also confirmed the existence of the vortex array in 1989.

Madhav Rao returned to India in the mid-1960s and was engaged in experimental solid state and nuclear physics research using neutron diffraction, scattering, polarization and depolarization techniques at AEET. From 12 January 1967, AEET was renamed as Bhabha Atomic Research Centre (BARC). Madhav Rao not only actively participated in different research programmes of the Nuclear Physics Division of BARC, but also collaborated in a number of experiments with other Department of Atomic Energy (DAE) institutions, such as Saha Institute of Nuclear Physics, Kolkata, etc. He retired from the BARC in the mid-1990s and became engaged in experimental neutron physics research along with faculty and scholars from the universities in western India through UGC–DAE Consortium for Scientific Research erstwhile known as the Inter University Consortium for DAE Facilities, Mumbai Centre. Madhav Rao belongs to those few rare physicists who participated in experimental superconductivity research in the 1960s golden era of superconductivity.

In his Nobel lecture[7] on 8 December 2003, Abrikosov acknowledged the contribution of Cribier and his collaborators for experimental verification of his theory. Madhav Rao was awarded the Medal of the Materials Research Society of India in 1993 in recognition of his excellence in research in materials science.

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