INTELBIOMAT - Exchange Grant Scientific Report

NMR Study of the Mott transition to superconductivity in cesium fulleride Cs_3C_{60}

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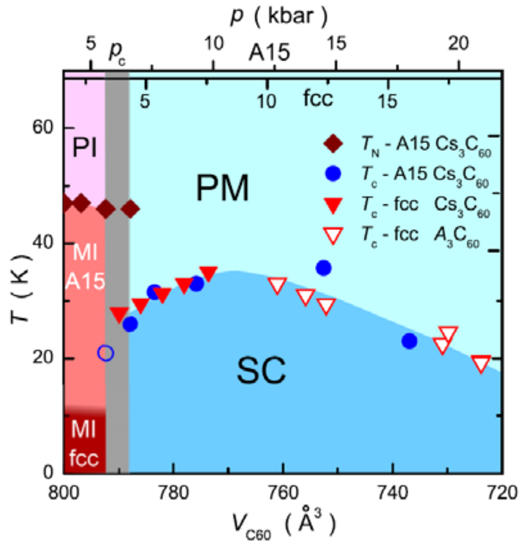
1) Purpose of the visit and introduction of my study

The main purpose of my visit to Laboratoire de Physique des Solides, Université Paris-Sud 11 was to finalize the work that I had performed since last year on the recently discovered cesium fulleride Cs_3C_{60}. The superconductivity in the class of fullerene had been thought to be described within a standard Bardeen-Cooper-Schrieffer (BCS) theory. However, the success in expanding the inter-fullerene distances in A_3C_{60} (A= alkali ion) by intercalation with large Cs ion had given one a new aspect of this field. Namely, Cs_3C_{60} is the first fulleride whose ground state changes from an antiferromagnetic insulating (AFI) state at ambient pressure to a superconducting (SC) state with transition temperature Tc reaching a maximum of 38K under pressure above ~ 4 kbar. The emergence of the superconductivity in the vicinity of the insulator (magnetic) to metal (paramagnetic) transition [1,2] is reminiscent of the superconductivity in high-Tc cuprates, iron oxyarsenides, and organic compounds. Cs_3C_{60} also brings another interest that the insulator - metal transition in this compound is a genuine Mott transition. Cs_3C_{60} is one of few good materials to observe the Mott transition itself and to investigate the relations between the superconductivity, magnetism and Mott transition due to its highly symmetric crystalline structure and the precisely assigned molecular valence. This is unlike in other superconductors such as high Tc cuprates with additional ambiguities of low dimensionality and variable numbers of carriers.

I had visited Université Paris-Sud 11 from April to September in 2011 for my scientific research abroad (sabbatical). During my stay, I took part in the project to study this compound which had been directed by Prof. H. Alloul. In order to investigate the AFI - SC transition of Cs_3C_{60}, high pressure technique is inevitable. Besides, the use of microscopic experimental probe is quite useful, because the synthesized products of this compound is up to date a mixture of three phases, Cs_3C_{60} (cubic A15 structure), Cs_3C_{60} (face centered cubic), and Cs_4C_{60} (body centered orthorhombic). Prof. Alloul and his collaborators succeeded in obtaining distinct information on each phase by nuclear magnetic resonance (NMR) measurements at Cs and C sites [3.4]. Then the main purpose of my study has been to investigate in detail the physical properties of the A15 Cs_3C_{60} near its phase boundaries by using a new A15-rich sample. The sample was provided by the group of Unversità di Parma in Italy headed by Prof. M. Riccò. NMR and ac-susceptibility measurements were performed with help of Prof. P. Wzietek. This study is quite important from physical point of view and is suited to me because I have also had rich experience in performing NMR measurements under high pressure as well as developing high pressure techniques.



Phase diagram of Cs_3C_{60} proposed by previous NMR and ac-susceptibility experiments (Ihara, Wzietek, Alloul, and their collaborators.) Please see Ref. [4] for details. The sample used in the present project, which contains smaller amount of the Cs_4C_{60} phase, gives more detailed information about the A15 phase.

[1] A. Y. Ganin *et al.*, Nature Mater. **7**, 367 (2008).
[2] Y. Takabayashi *et al.*, Science **323**, 1585 (2009).
[3] Y. Ihara *et al.*, Phys. Rev. Lett. **104**, 256402 (2010).
[4] Y. Ihara *et al.*, Europhysics Lett. **94**, 37007 (2011).

2) The work carried out during the visit

During my stay in France in 2011, we could obtain systematic series of ^{133}Cs- and ^{13}C-NMR data (spectrum and T_1 (spin-lattice relaxation time)) and ac-susceptibility data for the A15-rich sample by changing temperature and pressure. All the NMR measurements were performed by using a nine tesla surperconducting magnet, and ac-susceptibility were done at zero field. For high pressure measurements, we used an externally controlled system in which a pressure generator is connected to the pressure cell through a capillary tubing.

The practical missions of this visit are to analyze carefully the data obtained at eight different pressures below and above critical pressure (Pc~5kbar) so as to extract intrinsic properties of this compound from the data, and to prepare appropriate graphs to present the results. In order to complete these works as much as possible during my stay, I had frequent discussions with Profs. Alloul and Wzietek almost every day.

3) Main results obtained

From the careful analyses of the spectra and T_1 data, we could get rich information as listed below;

- Both of the Knight shift, estimated from the NMR spectra, and $1/T_1T$ show their unique temperature dependences below and above a characteristic temperature, respectively, at each pressure. From their changes from one phase to another, we could estimate Mott transition temperature as a function of pressure,
- From the NMR spectra and the ac-susceptibility data, we could determine AFI ordering temperature T_N at relatively lower pressures and SC transition T_C at higher pressures. Moreover these measurements allowed us to estimate the volume fractions of the AFI and SC phases. We determined Pc and found that the transition between the two phases is rather narrow in pressure range, indicative of the high quality of present sample.
- The mechanism of Cooper pair in the SC state was discussed by the temperature dependence of $1/T_1$ and the Knight shift obtained from the ^{133}Cs- and ^{13}C-NMR measurements.
- A pressure vs temperature phase diagram was completed for our A15-rich Cs_3C_{60} sample near the nonmagnetic - magnetic phase boundary, in which the AFI, SC and Mott transitions are clearly shown.
- The ^{133}Cs-NMR line shape was well reproduced with proper fitting parameters, *i.e.* nuclear quadrupole resonance frequency ν_Q and anisotropic Knight shift. The temperature dependence of ν_Q gives us information about local structural symmetry or the lattice parameters at the Mott transition.

4) Future collaboration with host institution

As for the study of cesium fulleride Cs_3C_{60}, some detailed analyses of the data are still remained, and further experiments at a higher pressure are now in progress by Prof. Wzietek. Besides, this project can be developed into other experimental techniques than NMR, for example diffraction measurements for the precise determination of crystalline structure under pressure. Therefore our collaboration will surely continue. Also, we will be able to find some other new projects to collaborate in future. I am now planning to invite Prof. Wzietek to Japan next year with help of other organization in order to discuss the studies of strongly correlated electron systems, including Cs_3C_{60}, and to exchange useful information about the latest experimental techniques. This may lead us to new collaborations in future.

5) Projected publications / articles

Our project is now a pre-publication stage and we are planning to submit some reports to journals for condensed matter physics soon.