NMR studies of Insulating, Metallic and Superconducting Fulleride Compounds: Importance of Correlations and Jahn-Teller Distortions

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Although high temperature superconductivity has been only found so far for *n* =3 in cubic A_nC_{60} compounds, it is often admitted that it results from a classical BCS electron-phonon mechanism. On the other hand the variation with *n* of the electronic properties of this series of compounds cannot be explained by a simple progressive band filling of the C_{60} six-fold degenerate t_{1u} molecular level. This has been ascribed to the influence of electron correlations and Jahn Teller Distortions (JTD) of the C_{60} ball which would enegetically favor evenly charged C_{60} molecules.

While JTD had never been observed directly, we have provided the first experimental observation which supports such a strong influence of JTD. Indeed ^{133}Cs NMR has allowed us [1] to evidence the existence of a charge disproportionation in the low T quenched cubic phase of CsC_{60}. About 12% of the C_{60} balls are in a doubly charged C_{60}^{2-}singlet ground state, which confirms that Hund's rule is disfavoured by the JTD. These balls act as "impurities" in a correlated metallic background with less than one electron per C_{60}. We provide observations which indicate why a completely segregated insulating ground state with doubly charged and neutral balls does not occur [1].

Although the insulating behaviour of A_4C_{60} compounds could result from a peculiarity of their *bct* structure, it has for long been considered that it rather results from a similar stabilization of the C_{60}^{4-} molecular level by a JTD. A small spin-gap is detected by NMR in these systems and is ascribed to a transition from the fundamental spin singlet state of C_{60}^{4-} to an excited spin triplet state. We shall present comparisons of the NMR properties of Na_2C_{60} and K_4C_{60} compounds which confirm that this scheme applies quite generally to evenly charged balls and does not depend critically on the actual crystal structure. The insulating state of evenly charged compounds therefore results from the effective repulsion between electrons induced by the JTD. It will be shown that these even electron systems are in the vicinity of a metal insulator transition [2], [3].

In superconducting *n*=3 compounds a static charge segregation does not occur. However a comparative study of the ^{13}C spin lattice relaxation T_1 of *n*=2 and *n*=3 compounds [2], [3] indicates that molecular excitations survive in the metallic state . This can be interpreted as the occurrence of dynamic charge segregation favouring C_{60}^{2-} and C_{60}^{4-}. This would indicate that the interplay betweeen electron correlations and JTD yields an effective attractive interaction which would enhance pairing, and would explain why superconductivity is restricted to *n*=3 compounds.

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