Molecular Excitations Detected by NMR in Alkali Fullerides

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Electrons can be transferred to the C_{60} lowest unoccupied molecular level (t_{1u}) by intercalating alkali ions into the structure of pure C_{60}. A family of compounds with formula A_nC_{60} is then obtained, in which the role of electronic correlations can be questioned, because of the narrowness of the t_{1u} level. Indeed, both insulating and metallic behaviors are observed as a function of doping, whereas a simple band picture would always predict a metallic behavior.

I will present NMR investigations of different stoechiometries of alkali fullerides that suggest that Jahn-Teller distortions (JTD) of the C_{60} molecule play an important role in this behavior, by inducing in some cases a form of electronic attraction that opposes the Coulomb repulsion.

Because it is triply degenerated, the t_{1u} level of the C_{60} molecule is subject to Jahn-Teller distortions, such as the ones represented on fig. 1. Schematically, this would lift the degeneracy of the three t_{1u} levels, as represented on fig. 2. This latter figure illustrates that the strength of the JTD is very sensitive to the charge of the C_{60} molecule and that the gain of energy is *larger* for *evenly* charged molecules. If JTD persists in the solids, one could speculate that they would favor the occurrence of evenly charge C_{60}.

Indeed, we have detected in a cubic quenched phase of CsC_{60} [1] the presence of spin-singlets on about 10 % of the C_{60} balls, that we believe are associated to the larger stability of these units. The separation between 'metallic balls' and balls wit a localized spin-singlet is represented on fig. 3.

Recently, we have also found evidence for the presence of JTD distortions on very short time scales in superconducting fullerides [2], so that it could be a general feature of these materials.

[1] V. Brouet, H. Alloul, F. Quere, G. Baumgartner and L. Forro, *Phys Rev. Letters* **82** , 2131 (1999)
[2] V. Brouet, H. Alloul, L. Thien-Nga, S. Garaj and L. Forro, *Phys Rev. Letters* **86**, 4680 (2001)

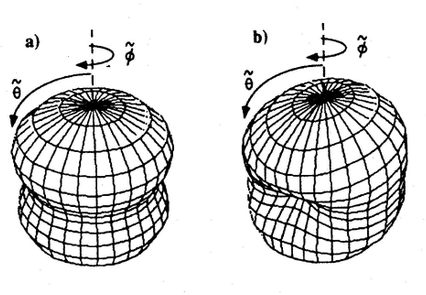

Fig. 1 : Examples of Jahn-Teller distortions predicted for a C_{60} molecule (see N. Manini *et al.*, *Phys. Rev. B* **49**, 13008 (1994)

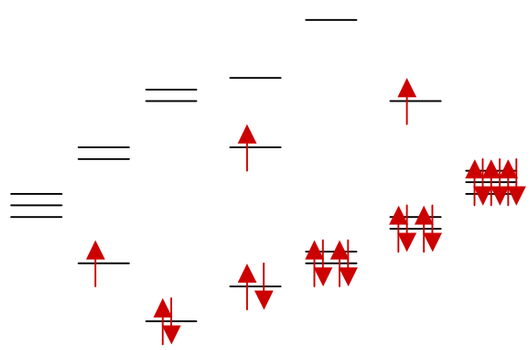

Fig. 2 : Representation of the gap induced by a JTD between the three t_{1u} levels as a function of the number of electrons per C_{60}.

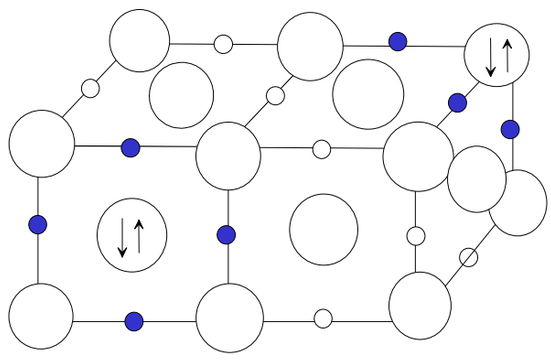

Fig. 3 : In a cubic quenched phase of CsC_{60}, NMR suggests that, while the compound is mostly metallic, some spn-singlets are localized on a few C_{60} balls. This is detected by the appearance of two types of Cs sites in the NMR spectra (represented here as closed or open circles).