Intercalation and High Temperature Superconductivity of Fullerides II

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Based on the mechanism described by the authors in Ref. [1], we predict that the compound C_{60}/CHI_{3} will have \( T_{c} \approx 140 \text{K} \), which would be the highest value obtained so far for intercalated materials.

This paper is based on our recent analysis [1] of high-temperature superconductivity observed in intercalated fullerides [2] (see also Ref. [3]). Following the approach presented in Ref. [1] we predict that the compound C_{60}/CHI_{3} has a value of \( T_{c} \) which is presently the highest for intercalated superconductors and second after the high-\( T_{c} \) cuprates.

The authors of Ref. [2] have discovered recently that by intercalating CHCl_{3} into the hole-doped C_{60} fulleride (via field-effect doping, see Ref. [4]) one obtains a critical temperature of \( T_{c} \approx 80 \text{K} \). Intercalating CHBr_{3} molecule leads to a further increase to \( T_{c} \approx 117 \text{K}(!) \).

It is believed that the superconducting state of fullerides is caused by the coupling of the carriers with intramolecular vibrations of the C_{60} units (see e.g. Ref. [5]). With the field-effect technique one is able to dope the parent compound with either electrons or holes. In the latter case the critical temperature of the fulleride \( T_{c} \approx 52 \text{K} \) is higher than for usual n-doped materials, because the hole band has a larger density of states [6].

The presence of intercalated molecules in the hole-doped parent compound leads to a drastic increase in \( T_{c} \). As described in Ref. [1] we think that this increase is caused by the additional interaction of the carriers with the vibrational manifold of the intercalated molecules CHA_{3} (A \equiv \text{Cl,Br}). The mechanism caused by the addition of polyatomic molecules and consequently of new phonon modes, was introduced by one of the authors in Ref. [6]. The beauty of fullerides is that their molecular structure allows to perform the intercalation (see Fig. [1]). Our analysis [1] was based on the vibrational spectra of CHCl_{3} and CHBr_{3} molecules, (Fig. [2] see [8]). The increase in \( T_{c} \) caused by the CHCl_{3} \rightarrow CHBr_{3} substitution is due to the “softening mechanism” (see, e.g., Ref. [9]), that is to the decrease in characteristic vibrational frequencies and corresponding increase in the coupling constant \( \lambda \). Note that in the system under consideration the characteristic mass entering the expression of the coupling constant is \( M \approx M_{C} \), where \( M_{C} \) is the mass of the Carbon ion [10].

We propose here to intercalate another molecule in the parent fulleride (see Ref. [3]), namely iodoform. That is, we suggest to make the compound C_{60}/CHI_{3}. The CHI_{3} molecule has the same structure as CHA_{3} (A \equiv \text{Cl,Br}; cf. Fig. [3]). It has similar five vibrational modes (see Ref. [11]): \( \Omega_{1}^{j} = 110 \text{cm}^{-1} \), \( \Omega_{2}^{j} = 154 \text{cm}^{-1} \), \( \Omega_{3}^{j} = 425 \text{cm}^{-1} \), \( \Omega_{4}^{j} = 578 \text{cm}^{-1} \), \( \Omega_{5}^{j} = 1068 \text{cm}^{-1} \) (the notation is that of Ref. [11]).

One can compare these values with those for CHCl_{3} and CHBr_{3} and directly see the additional “softening” occurring in CHI_{3}. We have determined in Ref. [10], the additional coupling constants (relative to the parent compound) for the C_{60}/CHCl_{3} and C_{60}/CHBr_{3} compounds and found the values \( \lambda_{C}^{j} \approx 0.2 \) and \( \lambda_{Br}^{j} \approx 0.55 \). Because of the softening effect, the coupling constant for the C_{60}/CHI_{3} compound is even larger and
equals to \( \lambda_1' \simeq 1.1 \). As a result, we obtain \( T_c \simeq 140\text{K} \). The nature of the approximations \( [2] \) is such that this value is a lower bound for the \( T_c \) of the \( C_{60}/\text{CHI}_3 \) compound.

The structure of the \( \text{CHI}_3 \) molecule is similar to that of \( \text{CHA}_3 \) (\( A \equiv \text{Cl,Br} \)), and the intercalation thus seems realistic from the point of view of chemistry. Since the \( T_c \) obtained theoretically is the highest for this class of materials it would be very interesting to synthesize the \( C_{60}/\text{CHI}_3 \) compound and measure its \( T_c \).

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[1] A. Bill and V.Z. Kresin, cond-mat/0109553.
[2] J.H. Schön, Ch. Kloc, and B. Batlogg, Science 293, 2432 (2001).

[3] R.F. Service, Science 293, 1570 (2001); B. Levy, Physics Today, 54, 19 (2001).
[4] J.H. Schön, Ch. Kloc, B. Batlogg, Nature 408, 549 (2000).
[5] O. Gunnarsson, Rev. of Mod. Phys. 69, 575 (1997).
[6] V. Kresin, Sov.Phys.-Doklady 10, 1194 (1966); Phys.Lett. 49A, 117(1974).
[7] M. Jansen and G. Waidmann, Z. anorg. allg. Chem. 621, 14 (1995).
[8] G. Herzberg, Molecular Spectra and Molecular Structure. II. Infared and Raman Spectra of Polyatomic Molecules, (D.Van Nostrand, Princeton, 1964).
[9] W. McMillan, Phys.Rev. 187, 331 (1968).
[10] N. Neto, O. Oehler, and R.M. Hexter, J. Chem. Phys. 58, 5661 (1973); P. Dawson and B.J. Berenblut, Spectrochimica Acta 31A, 1049 (1975).
[11] The value of \( \mu^* \) used here and in Ref. [1] is \( \mu^* \simeq 0.15 \); note the misprint in Ref. [1]