On the Limit of T_c in C_{60}-Based Superconductivity

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Abstract

Ionic superconductors (C_{60}^{3-}, C_{60}^{3+}) by gate-induced doping are explained in the unified picture with alkali ion-doped A_3C_{60} superconductors. For the weak-coupling s-wave superconductivity, the extended BCS-like theory is presented in analyzing roles by some kinds of phonons playing on the transition temperature T_c. The most interesting point is the existence of a phonon to adjust whether assisting or preventing T_c. In electron- and hole-doped C_{60} superconductors, the possibility of higher T_c is discussed for C_{60}-based superconductors.

Key words: C_{60}, superconductivity, phonon, device

1. Introduction

Since the discovery of C_{60} superconductors in field-effect transistor (FET) device [1,2], Fullerenes superconductivity becomes an attractive problem again, because hole-doped C_{60}^{3+} and the expanded C_{60} materials might produce higher transition temperature T_c than well-known A_3C_{60} superconductors. How high T_c would be realized is a challenging theme within the theory of phonon-mediated superconductivity.

First, we reconsider C_{60}-based superconductivity with various types of phonons, and propose an equation for T_c with the different form from McMillan's equation in order to solve the remaining and newly observed problems. Then we make clear the roles of major phonons playing in the T_c-equation, and within one theory, we will explain the mechanism of superconductivity for both chemical-doped and gate-induced C_{60} synthetically. In spite of the difference between 3-dimensional ($3D$) bulk and 2-dimensional ($2D$) FET, C_{60}-based superconductors reveals to be the same one, and therefore, the values of T_c in electron- and hole-doped C_{60} superconductors are analyzed collectively to compare with the observed results and to predict unknown data. After all, we would be able to explain which phonon part plays no role for gate-induced C_{60} and to guess the possible high T_c values.

2. Equation for T_c

Although superconductivity in A_3C_{60} is explained by using McMillan's equation in an early stage [3], we venture to propose another T_c-equation to make clear the role of various types of phonons. Applying site representation formalism developed by Appel and Kohn [4] for phonon-mediated superconductivity, we got the generalized BCS-like equation of T_c for C_{60}-based systems as [5]:

$$k_B T_c = 1.13\omega_a \exp[-1/N(0)I_{eff}], \quad (1)$$

$$I_{eff} = (I_a + I_H) + \frac{I_o - U^*}{1-(I_o - U^*)N(0)\ln(\omega_o/\omega_t)}, \quad (2)$$

$$U^* = \frac{U}{1+N(0)U\ln(\omega_F/\omega_t)}. \quad (3)$$

Within the weak-coupling theory, we introduced the effective electron-electron interaction vertex I_{eff} composed of three kinds of (acoustic, optical, high frequency) phonons as (I_a, I_o, I_H) with each cutoff frequency (ω_a=50 cm^{-1}, ω_o=100 cm^{-1}, ω_t=1500 cm^{-1}, respectively) below which the inter-electron interaction becomes attractive: I_a and I_H are concerned with C_{60} molecules with different ω for inter- and intra-molecular phonons, whereas I_o is related to optical phonon at alkali-ion sites. I_a plays the role to make the basic mechanism of phonon-mediated superconductivity, and I_H to make high T_c, and I_o the role of promoting or preventing T_c. Low ω_a takes the lead in T_c-equation, and therefore, Migdal theorem is conserved because of $\omega_a \ll \epsilon_F (\sim 0.2\,\mathrm{eV})$. Thus Cooper pairing through phonons originated from the different mass and site, can make the various physical properties depending on the optical phonon contribution, I_o/I_H(because $I_a + I_H \sim I_H$). Here, it is worth to note the role of the last term in the equation (2). It is guessed that T_c is enhanced by I_o and reduced by a factor $\ln(\omega_o/\omega_t)$ in the denominator. This is an important role of doped alkali-ions in superconductivity except for supplying electrons.

3. Gate-induced superconductivity

Now let us consider the gate-induced C_{60} system without alkali-ions. Most interesting point is whether the absence of alkali-ions promotes or disturbs the superconductivity. Contrary to I_H given by 37.2 meV [3],

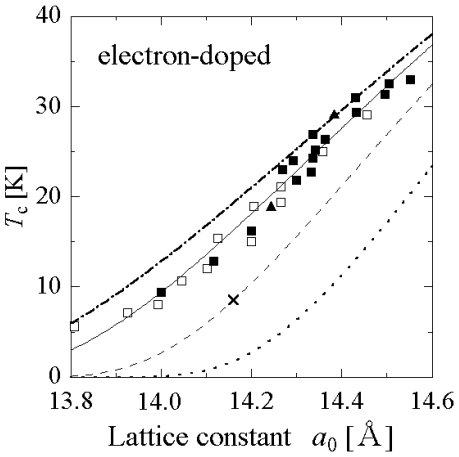

Fig. 1. Calculated T_c-a_0 relation for electron-doped C_{60}; solid line: I_o/I_H=0.6 where experimental data are plotted by various symbols, broken line: I_o/I_H=0.18 for no contribution of optical phonon (× is predicted for C_{60}^{3-} with a_0=14.16 Å), dotted line: I_o/I_H=0 for the case that optical phonon disturbs T_c extremely, and dash-and-dotted line: I_o/I_H=1 for the maximum contribution of I_o to assist T_c.

I_o is difficult to observe by any experimental means. When I_o/I_H exceeds 0.18, I_{eff} becomes larger than I_H. If I_o/I_H becomes smaller than 0.18, the existence of optical phonon disturbs the superconductivity. And at the point of I_o/I_H=0.18, no contribution of chemical doping would be expected for T_c.

Here T_c is calculated by using the density of states $N(0)$-lattice constant a_0 relation referred from the band calculation [6]. As shown in Fig.1, we got the reasonable ratio I_o/I_H=0.6 as the solid line to induce T_c-a_0 relation in good agreement with experimental data for various A_3C_{60}-type superconductors [7]. It means that the optical phonon assists T_c in A_3C_{60} with I_o/I_H=0.6. The broken line is given as I_o/I_H=0.18 for gate-induced electron-doping C_{60}^{3-}. By estimating T_c on the broken line for a_0=14.16 Å, we predict $T_c(C_{60}^{3-})\sim$9 K in good agreement with the observed $T_c\sim$11 K [1] and is considerably lower than $T_c(A_3C_{60})\sim$16 K on the solid line. Thus the superconductivity by electron doping (C_{60}^{3-}, A_3C_{60}) can be explained in the same mechanism. In 3D A_3C_{60} systems, the doped alkali-ions disturb the direct C_{60}-C_{60} interaction to keep away FCC→SC phase transition [8], and to assist T_c. In 2D FET-device, systems are probably composed of a sheet of C_{60} molecules, and therefore, C_{60}-C_{60} orientation ordering ought not to take place from the outset.

Hence T_c can be varied by changing the band filling (doping level) or the band width (lattice parameter). In gate-induced C_{60}, $N(0)$ is controlled by the applied gate voltage. As C_{60} is strongly electronegative, adding holes to C_{60}(eg. C_{60}^{3+}) is expected to be very difficult. However, if this would be performed, $T_c(C_{60}^{3+}) > T_c(C_{60}^{3-})$ might be predicted, because

Table 1
Expected T_c values for some C_{60} compounds with a_o=14.16 Å in comparison to experimental T_c(obs). Optimum T_c^{opt} is estimated for the limit of I_o/I_H=1 and a_0=14.6 Å.

doping	example	I_o/I_H	T_c(cal)	T_c^{opt}	T_c(obs)
electron	A_3C_{60}	0.60	16 K	38 K	16 K
	C_{60}^{3-}	0.18	9 K	38 K	11 K
hole	C_{60}^{3+}	0.18	32 K	54 K	52 K

C_{60} has large $N(0)$ in valence band than that in conduction band [9]. By estimating $N(0)$ for C_{60}^{3+} as twice of $N(0)$=14 states/eV for C_{60}^{3-} with a_o=14.16 Å, it can be presented to be $T_c(C_{60}^{3+})\sim$32 K where E_F lies probably at the maximum $N(0)$. This is certainly larger than $T_c(C_{60}^{3-})$ and also than $T_c(A_3C_{60})$, but is exceptionally smaller than the observed T_c=52 K [2]. When the crystal structure is quite different between 3D bulk and 2D FET, a_0 should be changed. If anticipating the optimum T_c, we will suggest $T_c\sim$54 K for the limit that I_o/I_H tends to 1 (because of $I_o\leq I_H$) and that a_0 becomes 14.6 Å just before Mott transition. In Table I, the expected and the optimum T_c in hole- and electron-doped C_{60} are summarized in comparison with experimental data. A challenge to make B_xC_{60}(ex. B=F^-) by chemical doping with the valence of -1 would be propose the higher T_c similar to C_{60}^{3+}.

4. DISCUSSION

Within the framework of s-wave superconductivity, we made clear important properties in C_{60}-based superconductors. Whether optical phonon assists or disturbs T_c, gives an interesting contribution to C_{60} systems in addition to the conventional mechanism. In the present theory, T_c has two aspects: (i) The different phonon modes (ω_a, ω_t) are made at the same C_{60} site. If C_{60} molecule is treated as a huge atom without intramolecular phonons, I_H=0 and ω_t=ω_a lead to T_c=0. (ii) The optical phonon is made mainly at A(A') site. When I_o/I_H larger than 0.18, the optical phonon contributes to enhance T_c. Basing on this theory, we will prove large Coulomb repulsion μ^*=$N(0)U^*$, anomalous $^{12}C\rightarrow^{13}C$ isotope effect and so on. The 2D nature of the channel is the same condition with so-called high-T_c cuprates, and low interface density of states of FET will be useful to the next research on Cu-oxides.

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