

Unconventional pairing in fullerides by nonadiabatic channels

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Abstract

In C_{60} compounds the frequencies of the phonon involved in the superconductive processes (intra-molecular modes with ω_{ph} up to 2300 K) are comparable with the typical electronic scale ($E_F = 2900$ K) and thus the two dynamics cannot be treated separately (breakdown of the adiabatic hypothesis). This implies a generalization of the standard theory to include nonadiabatic effects. The strong electronic correlation in the fullerenes causes the contribution of the nonadiabatic processes to be positive with consequent increase of the effective electron–phonon (el–ph) pairing λ and of T_c . We show that the inclusion of nonadiabatic terms in the el–ph interaction is a key element for the high values of T_c in fullerenes. In particular T_c as high as 100 K are shown to be compatible with moderate values of $\lambda \lesssim 1$. This implies that it should be possible to increase the value of λ , and so T_c , and still have a stable system.

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The superconductivity in C_{60} materials has been often assumed to be consistently described by the conventional Migdal–Eliashberg (ME) theory of the phonon mediated superconductivity. From the ME point of view the high values of the critical temperatures in fullerene compounds ($T_c \sim 30$ K in Rb_3C_{60}) have been interpreted in terms of a strong electron–phonon (el–ph) coupling λ generated mainly by intra-molecular phonon modes.

However, fullerene-based superconductors display also a variety of anomalous features making the ME picture problematic. In fact, like the high- T_c copper oxides, C_{60} compounds have extremely low charge carrier density [1], have a significant electron correlation and are close to a metal–insulator transition showing a strong dependence of T_c upon doping and disorder [1–3]. Within the ordinary ME framework, all these anomalous features tend to degrade superconductivity.

In addition to the above unconventional features of the C_{60} compounds, other hints against the ME scenario arise from a closer look to the relevant energy scales involved in the el–ph interaction. The C_{60} molecule has a rather wide range of phonon modes of energy extending from $\omega_{min} = 400$ K up to $\omega_{max} = 2300$ K, while the relevant electronic energy scale, for the alkali doped compounds (A_3C_{60}), is the Fermi energy $E_F \simeq 0.25$ eV $\simeq 2900$ K [2]. In this context the adiabatic parameter ω_{ph}/E_F appears to be not at all negligible, $\omega_{ph}/E_F \sim 0.3$ – 0.8 , and one of the assumptions of ME theory, the adiabatic hypothesis, breaks down [4]. The use of the conventional theory is thus intrinsically inconsistent and the explicit inclusion of nonadiabatic effects needs to be taken into account.

From a technical point of view the definition of an el–ph theory, valid in nonadiabatic regime where Migdal’s theorem does not apply, is a highly difficult task. An appropriate starting point is a perturbative scheme where nonadiabatic vertex diagrams, arising from the breakdown of Migdal’s theorem, are taken into account up to the first order. Although this approach is formally

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valid as far as the magnitude of the vertex corrections is small ($\lambda\omega_{\text{ph}}/E_{\text{F}} < 1$), it properly provides a qualitative insight on the metallic properties of the fullerenes in a broader nonadiabatic regime, where a Fermi liquid picture holds true. It is worth to stress that the inclusion of nonadiabatic effects implies however nontrivial deviations from the ideal Fermi liquid description.

A controlled theory of nonadiabatic superconductivity has been built up in the past years and a detailed derivation of such a generalized theory beyond the Migdal's limit has been reported in Refs. [5,6]. A crucial point is that the nonadiabatic corrections are strongly dependent on the particular range of momenta and frequencies actually probed by the el-ph interaction. In this regards the microscopic characteristics of the real materials are important. In particular the strong degree of electronic correlation in fullerenes has been shown to have an important and positive effect in nonadiabatic regime by favouring small momentum scattering [7]. In this situation the vertex corrections mainly enhance the superconductive pairing in C_{60} compounds with a consequent net increase of the critical temperatures with respect to the adiabatic ME predictions.

The concrete example of Rb_3C_{60} , studied in Ref. [6], can be useful to underline the substantial change of perspectives in fullerene compounds. In fact in a conventional ME theory the high- T_{c} value ($T_{\text{c}} \sim 30$ K) together with the quite small isotope effect ($\alpha_{T_{\text{c}}} \sim 0.21$) observed in Rb_3C_{60} can be reproduced only by assuming unrealistic values of the coupling constant $\lambda > 1$ [6]. In this λ 's regime the system is expected become unstable with respect to different kinds of lattice instabilities and a Fermi liquid picture of the systems breaks down. Moreover $\lambda > 1$, together with a not negligible adiabatic ratio $\omega_{\text{ph}}/E_{\text{F}}$ in fullerenes provide a sizable vertex function, of magnitude $\lambda\omega_{\text{ph}}/E_{\text{F}}$, which cannot thus be neglected. This point out the intrinsically inconsistent of ME picture of Rb_3C_{60} .

A more reasonable scenario is given by the nonadiabatic theory where the experimental data for T_{c} and $\alpha_{T_{\text{c}}}$ arise from much lower and more realistic values of the coupling, $\lambda \lesssim 1$ [6]. In the new framework of nonadiabatic theory of superconductivity the driving element of the large critical temperatures is thus the opening of new nonadiabatic channels of pairing more than an extremely large values of λ .

An interesting question that arises from the above discussion concerns the problem of the extent to which the critical temperatures can be enhanced by increasing λ in a nonadiabatic context before the lattice instabilities occur. From the theoretical point of view, one way to enhance T_{c} is given by considering the injection of holes as charge carriers instead of electrons. This should cause an enhancement of the density of the states and a corresponding increase of T_{c} .

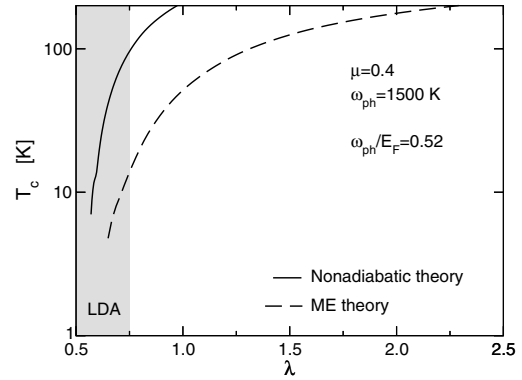


Fig. 1. T_{c} versus el-ph coupling λ for both ME theory (---) and nonadiabatic theory (—). The shaded area represents LDA calculations for λ in A_3C_{60} .

In Fig. 1 the critical temperatures are plotted as function of the el-ph coupling λ , both in the conventional theory (dashed line) and nonadiabatic theory (solid line), while the shaded area corresponds to LDA calculations of λ for the electron-doped C_{60} compounds. The nonadiabatic theory can naturally account for $T_{\text{c}} \sim 30$ K with moderate values of the coupling that fall in broad LDA region. In this situation the nonadiabatic theory predicts that λ can be further enhanced to get critical temperatures as high as 100 K remaining yet in a weak-intermediate regime, far away from the region in which structural instabilities occur. This implies that it should be possible to increase the value of λ , and so T_{c} , and still have a stable system. On the other hand, in the conventional framework $\lambda > 1.5$ are needed to give $T_{\text{c}} \sim 100$ K. These values appear unrealistic when compared with LDA calculations and problematic from the point of view of lattice instabilities and possible localized polaron formation.

The nonadiabatic theory provides also a new scenario wherein unconventional features of both the superconducting and normal states acquire a natural explanation. Specific predictions regard mainly the isotope effects on T_{c} [5,6], on the effective electronic mass m^* [8] and on the Pauli susceptibility χ [9].

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