ELSEVIER

Physica B 219&220 (1996) 169–171

PHYSICA B

The Eliashberg electron–phonon theory for the superconducting alkali-metal-doped fullerenes

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Abstract

The electrodynamic response in the superconducting ground state of K_3C_{60} and Rb_3C_{60} is analysed within the standard Eliashberg electron–phonon theory of superconductivity. The results strongly support a pairing mechanism mediated by high-frequency intramolecular phonon modes.

The discovery [1,2] of superconductivity at relatively high temperatures in the alkali-metal C_{60} compounds has created considerable interest. The central question, of course, is about the nature of the pairing mechanism, which remained for a long time quite controversial. There has been debate about whether electron–electron [3] interactions on the C_{60} ball or electron–phonon (e–ph) coupling [4–6] mediate the pairing. Concerning the latter mechanism, there was quite a bit of controversy with respect to arguments, favouring electron–phonon interactions with low-frequency *inter*molecular vibrations [6] or with high-frequency *intra*molecular modes [4,5].

The energy scales of the various modes which mediate the e–ph coupling are different and are reflected in the magnitude of the superconducting gap. Therefore, which of these scales are important could, in principle, be decided by examining whether the weak or strong coupling limit applies. In this context, the optical investigations are a powerful experimental tool. In this paper, we discuss our experimental results within the standard Eliashberg electron–phonon theory of superconductivity. This analysis allows us to exploit fully the optical investigation and to single out the relevant phonon excitation for the pairing mechanism and thus for superconductivity.

Reflectivity measurements ($R(\omega)$) as a function of the temperature on high-quality single crystals were performed between 14 and 50 000 cm^{-1} using three different spectrometers. The optical conductivity is then obtained through Kramers–Kronig (KK) transformation of $R(\omega)$. Fig. 1 displays the measured optical conductivity together with the BCS calculation [7,8]. Given the fact that the calculation neglects the role played by mean-free path effects, the agreement between our results and the theory based on a BCS ground state is excellent. The values of the gap (see Fig. 1) correspond to a reduced gap ratio of about 3.5, which is in agreement with the weak coupling limit of the BCS theory. The sharp onset of absorption at the gap suggests an s-wave singlet superconducting ground state [7].

It is now of interest to exploit further the experimental data by comparing the experimental results with the predictions of the conventional electron–phonon theory of superconductivity, in a more complete fashion, however, than the BCS approach. To this end we describe our

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0921-4526/96/$15.00
SSDI 0921-4526(95)00685-0

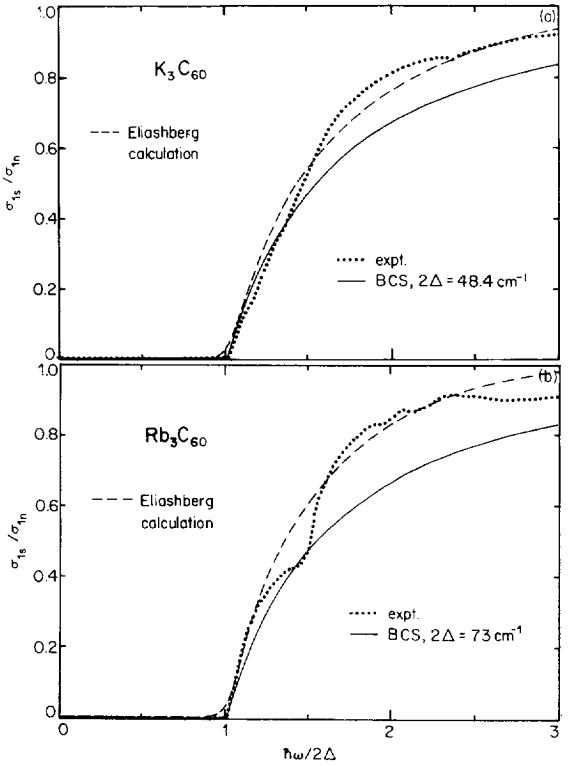

Fig. 1. The measured optical conductivity together with $\sigma_{1,s}(\omega)/\sigma_{1,n}(\omega)$ calculated using the Mattis–Bardeen theory (BCS) with the given single-particle gap values. The dashed curve corresponds to the fit within the Eliashberg approach (see text).

use of the standard Eliashberg theory to calculate the optical properties. Eliashberg theory describes all properties of the conventional superconductors to within a few percent and is considered to be one of the most exact theories in condensed matter physics. It reduces to the standard weak coupling BCS theory in the limit of the average phonon frequency being much greater than T_c. The complex optical conductivity is calculated from the current–current correlation function. A full development of the formalism was discussed elsewhere [7].

Here, our primary goal is to decide: If these materials are conventional electron–phonon superconductors, then what can the optical conductivity tell us about the phonon spectrum? Is the pairing due to intermolecular phonons at low frequencies or intramolecular phonons at high frequencies, or both? To this end we first adopt a model for the electron–phonon spectral function $\alpha^2F(\omega)$ with the form of a single truncated Lorentzian with peak position at ω_0 and width Γ_c and truncated at

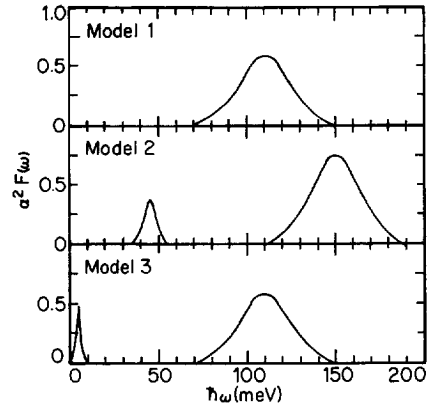

Fig. 2. The model electron–phonon spectral function $\alpha^2F(\omega)$ for both compounds which was used for the calculations described in the text and shown in Fig. 1.

Γ_c, i.e.,

$$\alpha^2F(\omega)\begin{cases}\propto \dfrac{1}{(\omega-\omega_0)^2+\Gamma_0^2}-\dfrac{1}{\Gamma_c^2+\Gamma_0^2}, & |\omega-\omega_0|<\Gamma_c,\\ =0, & |\omega-\omega_0|>\Gamma_c.\end{cases} \quad (1)$$

We do not attempt to fit an exact $\alpha^2F(\omega)$ as the variation of parameter space is quite large. Our aim is simply to examine three models. In the first model we place a peak at high-energy typical of intramolecular phonons and calculate the conductivity, in the second we add an extra peak to the spectrum of the first model at a slightly lower energy to see if we can describe the temperature dependence of the resistivity along with the optical data, and in the third we add a peak at a very low energy to simulate librational or intermolecular phonons.

Fig. 2 shows the typical model $\alpha^2F(\omega)$ spectra which we have used in our calculations. In the first case (Fig. 2(a)) we have coupling to high-energy intramolecular phonons which we have simulated by placing a Lorentzian centered at $\omega_0 = 1210\,\mathrm{cm}^{-1}$ and with the following parameters: $\Gamma_0 = 160\,\mathrm{cm}^{-1}$ and $\Gamma_c = 320\,\mathrm{cm}^{-1}$ for both K_3C_{60} and Rb_3C_{60}. Generally, as expected, shifting this peak in energy does not affect our conclusions in any way so long as the energy is much greater than T_c (i.e., $\langle\omega\rangle > T_c$). In model 2 (Fig. 2(b)), besides a high-frequency Lorentzian at 150 meV, we add a lower centered at $\omega_0 = 363\,\mathrm{cm}^{-1}$ with parameters: $\Gamma_0 = 40\,\mathrm{cm}^{-1}$ and $\Gamma_c = 80\,\mathrm{cm}^{-1}$. This peak was introduced only to provide the best possible fit also to the temperature dependence of the resistivity. Omitting this lower energy peak does not change our conclusions regarding the optical properties. Used as an input to the Eliashberg equations and scaling this spectrum to give

the correct T_c of 19 and 29 K, we find for both models (Figs. 2(a) and 2(b)) that $\Delta = 23.6\ cm^{-1}$ and $35.9\ cm^{-1}$, $2\Delta/k_BT_c = 3.57$ and 3.56, and $\lambda = 0.513$ and 0.514 for K_3C_{60} and Rb_3C_{60}, respectively [7]. The calculated gap values are in a good agreement with the experimental ones. The best fits are shown in Fig. 1 for both compounds. The agreement with the experimental findings is outstanding.

Finally, we would like to investigate briefly the role of low-frequency intermolecular or librational modes [7]. By introducing a peak in the $\alpha^2F(\omega)$ spectrum at very low frequency (model 3, Fig. 2(c)), we found that we could not have significant coupling to these modes and explain the data at the same time. The low-frequency modes at $\hbar\omega < 10$ meV produce strong coupling effects and therefore increase the gap ratio to order > 4 which is much greater than what is observed in the experimental data [7]. In addition, ignoring that issue and still trying to fit the normalized conductivity, it was not possible to provide a good fit to the data. The theoretical optical conductivity displayed large Holstein structure in stark disagreement with the experimental findings [7].

Therefore, we definitely conclude that the optical results do not support coupling to very low-energy phonon modes for the pairing mechanism in the doped fullerenes. The calculation within the Eliashberg formalism strongly suggests a pairing mechanism mediated by high-frequency intramolecular phonon modes, in accord with the implications of the weak coupling BCS limit. This conclusion also enforces the outcome of several experimental results, arrived at by applying a large variety of techniques [7].

Acknowledgements

The authors are grateful to L. Mihaly, M.J. Rice, T.M. Rice, and D.B. Tanner for fruitful discussions, to J. Müller and H.P. Staub for technical assistance. One of the authors (L.D.) also wishes to acknowledge the financial support of the Swiss National Foundation for the Scientific Research.

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