

## The electrodynamic response of $K_3C_{60}$ and $Rb_3C_{60}$ single crystals

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### Abstract

We present the optical response of  $K_3C_{60}$  and  $Rb_3C_{60}$  single crystals in their normal and superconducting phase. Above  $T_c$  the optical conductivity is characterized by two components: a Drude one due to the free charge carriers and a mid-infrared absorption. Below  $T_c$  the data clearly indicates a superconducting gap in agreement with the weak coupling limit of the BCS theory, suggesting furthermore that high frequency intramolecular phonon modes are relevant for the pairing mechanism.

Superconductivity in alkali-metal doped fullerenes  $C_{60}$  raised a controversy debate with respect to the electron-pairing mechanisms. Electron-phonon coupling to both low-frequency (intermolecular) and high frequency (intramolecular) vibrations - the strong-coupling and weak-coupling cases respectively - have been proposed. These two mechanisms have different associated energy scales, which is reflected in the magnitude of the superconducting gap. Optical investigations are a powerful experimental tool in order to determine this important energy scale. We present here our recent results regarding the superconducting and normal state properties, as well, obtained on single crystals. Some of the results were already presented elsewhere [1,2].

High quality single crystals of  $K_3C_{60}$  and  $Rb_3C_{60}$  were prepared by doping vapour-transport-grown  $C_{60}$  crystals with alkali metals following a method similar to that described previously and had a  $T_c$  of 20.3 and 30.5 K, respectively [2]. Reflectivity measurements ( $R(\nu)$ ) were performed between 2 meV and 6 eV as a function of temperature using three different spectrometers, with overlapping frequency ranges as described in previous work [2]. The absolute accuracy of the measurement is about 0.8%, while the relative one (important for the temperature dependence of  $R(\nu)$ ) is better than 0.3%. The optical conductivity is obtained with Kramers-Kronig transformations [1,2].

The reflectivity of  $Rb_3C_{60}$  in the normal state is displayed in Fig. 1a (results for  $K_3C_{60}$  are similar); besides the overall metallic behaviour (see Ref. 2 for a detailed discussion), we recognize in the mid-infrared frequency range two well defined absorptions at 0.07 and 0.17 eV, which are ascribed to the IR-active  $T_{1u}$  phonon modes [3]. Their appearance and the fair agreement with the expected frequency of these modes for this alkali-metal doping level

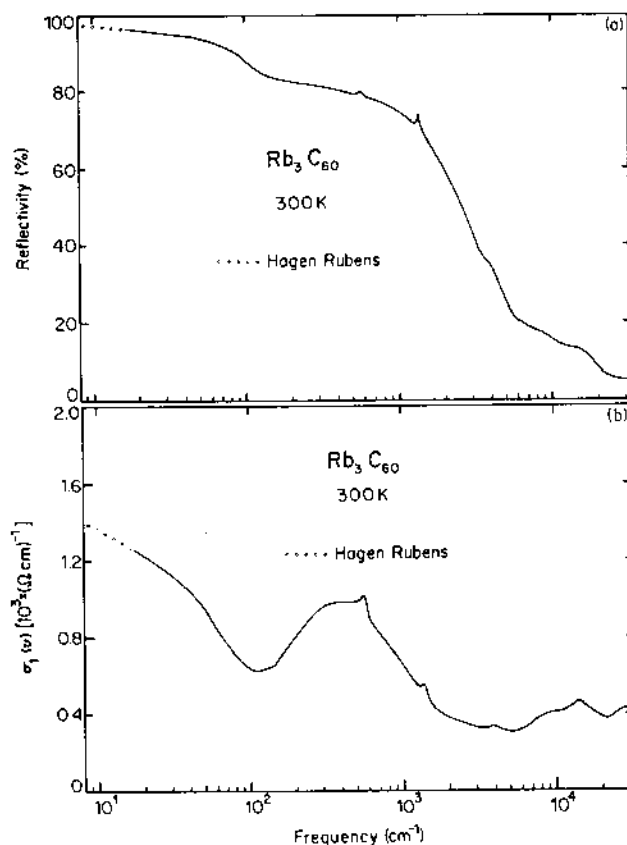


Fig.1: Normal state reflectivity spectrum (a) and optical conductivity (b) of  $Rb_3C_{60}$  single crystal (please note the logarithmic scale).

[3] call for the very good quality of the samples and for the outstanding resolution of these optical measurements.

Fig. 1b displays the optical conductivity in the normal state. The low frequency part of  $\sigma_1(\nu)$  (i.e., from FIR up to mid-IR) is characterized by two main features: the broad band centred at approximately  $400\text{-}800\text{ cm}^{-1}$  and the Drude-like contribution to  $\sigma_1(\nu)$  at the far-infrared (FIR) frequencies. In view of these features we model the dielectric function with a Drude like-model for the low frequencies part of  $\sigma_1(\nu)$  and a harmonic oscillator for the mid-infrared absorption [2]. There are two main directions that can be followed to interpret our data. The first one, which we will call the one-component picture, is based on the assumption that the low frequency Drude-like behaviour and the mid-IR absorption are due to the response of the conduction band, and crudely speaking the charge carriers behave as free carriers at low frequencies and as bound ones at high frequencies. The second way is to consider the two contributions independently, using a two-component picture. Then, the Drude part is characterized by a reduced plasma frequency and the harmonic oscillator is viewed as a separate mid-IR excitation. We note, however, that in both cases the scattering rate of the Drude component is larger than the expected superconducting gap  $2\Delta$ . This suggests that the  $\text{K}_3\text{C}_{60}$  and  $\text{Rb}_3\text{C}_{60}$  compounds are in the dirty or nearly dirty limit.

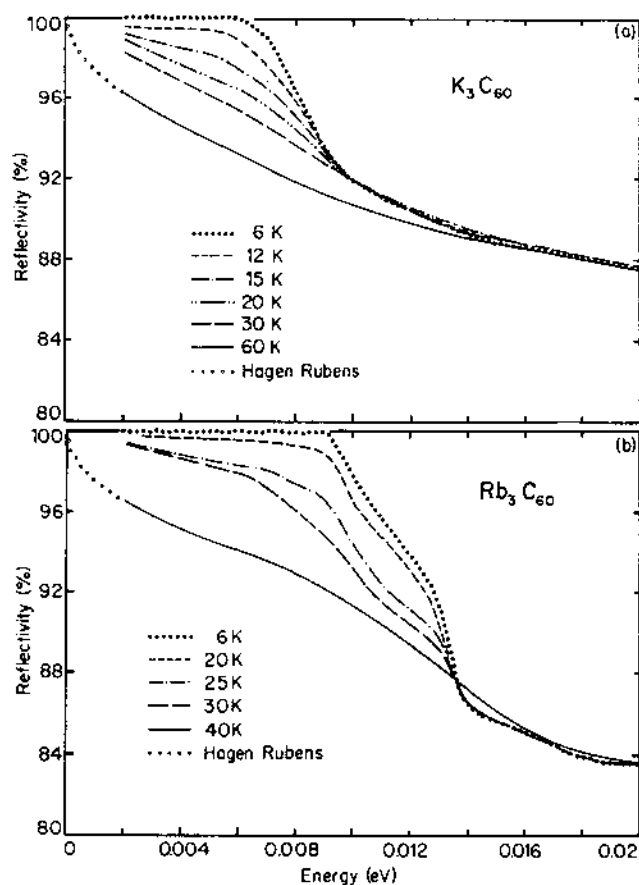


Fig. 2: Far-infrared reflectivity spectra above and below  $T_c$  of (a)  $\text{K}_3\text{C}_{60}$  and (b)  $\text{Rb}_3\text{C}_{60}$ .

Fig. 2 presents the reflectivity spectra in FIR of  $\text{K}_3\text{C}_{60}$  and  $\text{Rb}_3\text{C}_{60}$  single crystals. It is clearly seen that below  $0.014\text{ eV}$   $R(\nu)$  is enhanced and the onset of this BCS-like enhancement is coincident with  $T_c$ . At the lowest temperature (i.e.,  $\sim 6\text{ K}$ ) the reflectivity is, within experimental error, 100% below a threshold frequency of about 6 and 9 meV for  $\text{K}_3\text{C}_{60}$  and  $\text{Rb}_3\text{C}_{60}$ , respectively. These values correspond to a reduced gap ratio of  $2\Delta/k_B T_c = 3.44$  and  $3.45$  for  $\text{K}_3\text{C}_{60}$  and  $\text{Rb}_3\text{C}_{60}$ , respectively, which suggests a weak coupling BCS limit. We also note that the presence of the mid-infrared absorption might lead to another explanation of the data: that the behaviour of  $R(\nu)$  below  $T_c$  is due to a sort of plasma-edge effect [4]. The coincidence with  $T_c$  of the onset of the temperature dependence of  $R(\nu)$  and particularly the absence of any zero crossing of the real part of the dielectric function  $\epsilon_1(\nu)$  (which would define the screened plasma frequency) [4] in the FIR frequency range clearly rule out this explanation.

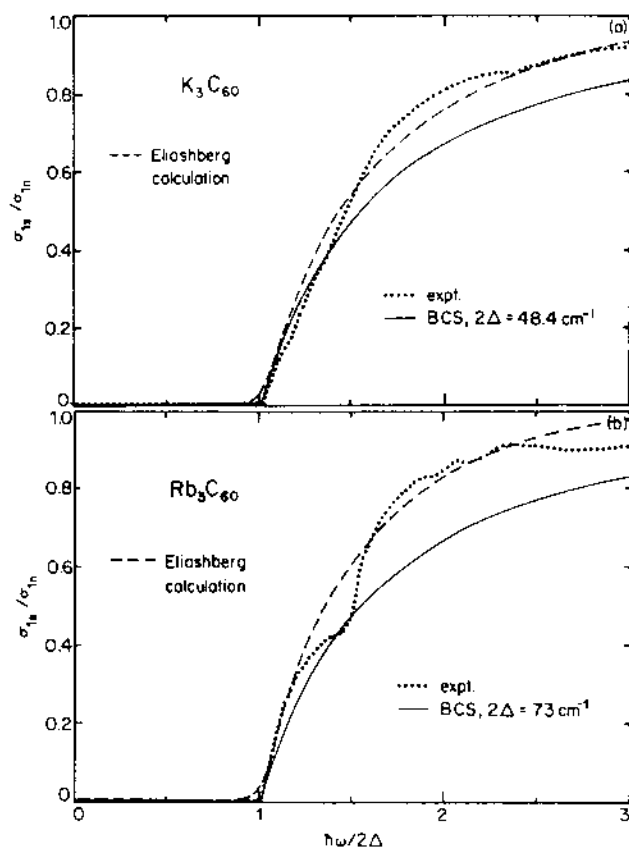


Fig. 3: The measured optical conductivity together with  $\sigma_{1,s}(\nu)/\sigma_{1,n}(\nu)$  calculated using the Mattis-Bardeen theory (BCS) with the single particle gap values given in the text. The dashed curve corresponds to the fit within the Eliashberg approach.

It is instructive at this point to first compare the electrodynamic response in the superconducting state with the conventional calculation of Mattis and Bardeen [5] within the BCS framework. This is shown in Fig. 3, where

the agreement between our experimental results of  $\sigma_{1s}(\omega, 6K)/\sigma_{1n}(\omega, T > T_c)$  (obtained by KK transformations of the spectra in Fig. 2) and the theory based on a BCS ground state is excellent. We also note, that the functional form reflects the so called case II coherence factors.

One can also compare 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 make use of the standard Eliashberg theory to calculate the optical conductivity for arbitrary impurity scattering in the local approximation [2]. 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. While it reduces to the standard weak coupling BCS theory in the limit of the average phonon frequency being much greater than  $T_c$ , the full theory takes into account the full details of the retarded electron-phonon interaction. The complex optical conductivity is thus calculated from the current-current correlation function [2].

Our primary goal here is to decide if the pairing is due to intermolecular phonons at low frequencies or intramolecular phonons at high frequencies or both. We considered several models for the electron-phonon spectrum, in order to take into account these different scenarios [2]. For the case, where we have coupling to high energy intramolecular phonons (simulated by placing a Lorentzian centered at  $\omega_0 = 0.15$  eV for both  $K_3C_{60}$  and  $Rb_3C_{60}$  [2]) as input in the Eliashberg equations and scaling this spectrum to give the correct  $T_c$  of 20 K and 30 K, we find that  $\Delta = 23.6$  cm<sup>-1</sup> and 35.9 cm<sup>-1</sup>,  $2\Delta/k_B T_c = 3.57$  and 3.56, and  $\lambda = 0.513$  and 0.514 for  $K_3C_{60}$  and  $Rb_3C_{60}$ , respectively. Fig. 3 presents the calculation of the optical conductivity within the Eliashberg formalism. For both compounds the reproducibility of the experimental data within this model is very good and the Mattis-Bardeen BCS result [5] is significantly improved.

On the other hand, by introducing a peak in the phonon spectrum at very low frequency, we found that we could not have significant coupling to these modes and explain the data at the same time. The low frequency modes 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. In addition, ignoring that issue and still trying to fit the normalized conductivity, it was not possible to find a value of the impurity parameter which would provide a good fit to the data [2]. Therefore, we definitely conclude that the experimental data does not support coupling to very low energy phonon modes for the pairing mechanism in the doped fullerenes.

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