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OPTICAL PROBING AND ELIASHBERG CALCULATION OF THE SUPERCONDUCTING STATE IN K_3C_{60} AND Rb_3C_{60}

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Abstract We have evaluated the optical conductivity over a broad spectral range of single phase K_3C_{60} and Rb_3C_{60} compounds both below and above the superconducting transition temperature. In the superconducting state we found gap values consistent with the BCS weak coupling limit. Moreover, we calculated the electrodynamic response with the standard Eliashberg electron-phonon theory of superconductivity, which strongly supports a pairing mechanism mediated by high frequency intramolecular phonon modes.

Introduction

The nature of the pairing mechanism in the superconducting alkali-doped C_{60} systems has remained for quite a long time an unsolved and controversial problem. There was quite a bit of debate with respect to arguments, favouring electron-phonon interactions with low frequency inter-molecular vibrations [1] or with high frequency intra-molecular modes [2,3].

The energy scales of the various modes which mediate the e-ph coupling are different, and therefore, which of these are important, could in principle be decided by examining whether the weak or strong coupling limit applies. In the case of high frequency phonons for example, one expects,

within the mean field Bardeen-Cooper-Schrieffer (BCS) theory, the weak coupling-limit to be appropriate with the ratio of the single particle gap Δ and the transition temperature T_c given by $2\Delta/k_BT_c = 3.52$. In the case of low frequency vibrations, on the other hand, $2\Delta/k_BT_c$ is expected to exceed the value which is appropriate for the weak coupling limit. In this context, the optical investigation is a suitable tool in order to single out whether the weak or strong coupling limit of the electron-phonon coupling mechanism is more appropriate to describe the superconducting state.

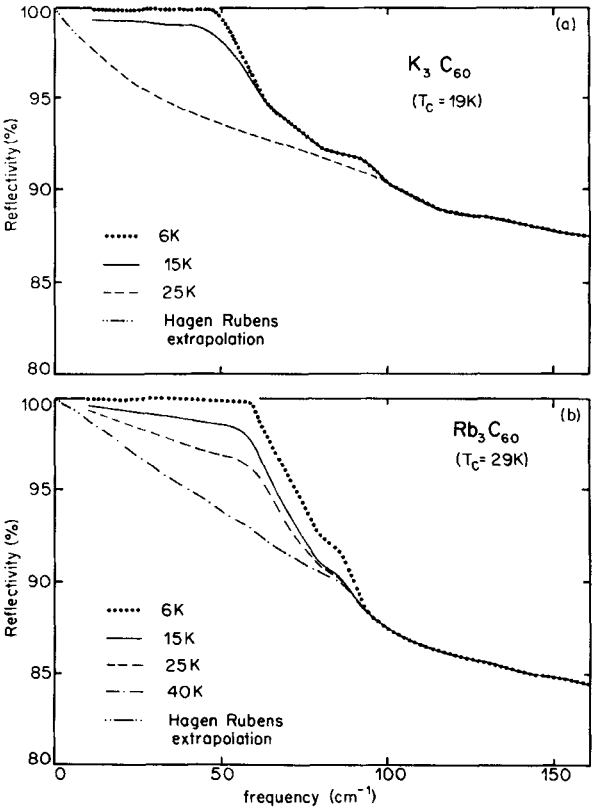

Fig.1: Optical reflectivity at several temperatures in the infrared spectral range for (a) K_3C_{60} and (b) Rb_3C_{60}.

Experiment and Results

The K_3C_{60} and Rb_3C_{60} specimens were prepared by a gas-solid reaction between C_{60} and gaseous alkali metal, in a way similar to that reported earlier [4]. The samples have a superconducting phase transition

temperature T_c=29 K and 19 K for the Rb- and K-fullerene, respectively. Reflectivity measurements (R(ν)) were performed on pressed pellet of about 3 mm diameter and 1 mm thick between 14 and 5x10^4 cm^{-1} using three different spectrometers with overlapping frequency ranges, as described in previous work [4]. Here, we just note that, since the materials are highly air sensitive, precautions and care have been taken in order to measure the compounds in an oxygen free environment.

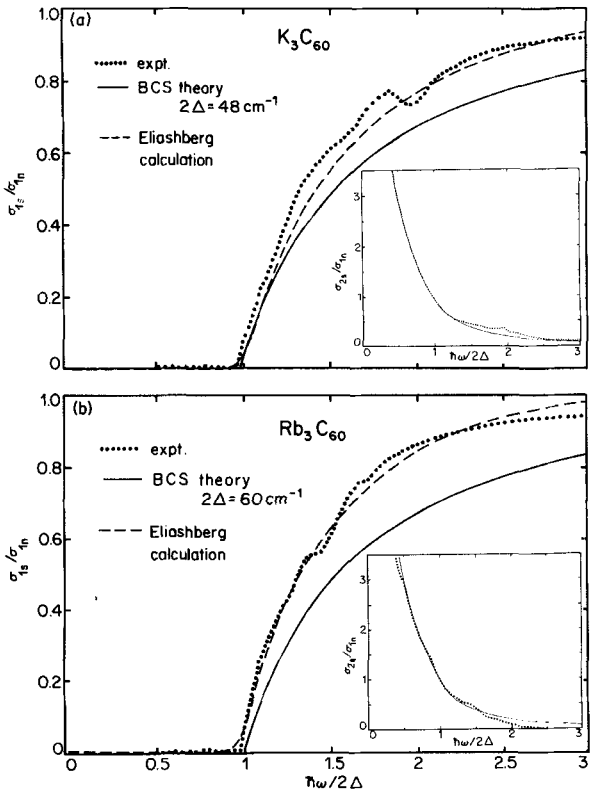

Fig.2: The measured optical conductivity together with $\sigma_{1,s}(\nu)/\sigma_{1,n}(\nu)$ calculated using the Mattis-Bardeen theory (BCS). The insets show the imaginary part $\sigma_{2,s}(\nu)/\sigma_{1,n}(\nu)$. The dashed curve corresponds to the fit within the Eliashberg approach (see text, model 1 and 2) with an impurity parameter of $1/\tau$= 200 cm^{-1} and 194 cm^{-1} for K_3C_{60} and Rb_3C_{60}, respectively.

The complete electrodynamic response has been already fully and thoroughly discussed elsewhere [4]. Therefore, we limit our attention to the frequency range, which is relevant for the expected optical fingerprints of the

superconducting state. In Fig. 1 we present the optical reflectivity measured above and below the superconducting transition temperature, in the spectral range which covers the superconducting gap values. While at low frequencies the reflectivity in the normal state is well described by the so called Hagen-Rubens extrapolation (see dashed-double-dotted line in Fig. 1), we observe below T_c a clear enhancement of R(ν) in FIR at frequencies below 100 cm^{-1}, with an onset as a function of temperature coincident with T_c. The optical conductivity is then obtained through Kramers-Kronig (KK) transformation of R(ν) [4]. Fig. 2 displays the optical conductivity in the superconducting state normalized to the optical conductivity in the normal state.

Discussion

From Fig. 2, it is clearly shown that for both compounds the conductivity is zero up to a threshold frequency which we identify as the superconducting gaps. We obtain Δ= 24 cm^{-1} for the K- and 30 cm^{-1} for the Rb- compound. Together with the superconducting transition temperatures, this leads to the ratio $2\Delta/k_BT_c$= 3.6 and 2.98 for K_3C_{60} and Rb_3C_{60}, with both values in good agreement with the weak coupling BCS results of $2\Delta/k_BT_c$= 3.52. This would suggest that the relevant excitation, responsible for superconductivity pairing, is significantly larger than the energy associated with the single particle gap in these materials. Furthermore, we note that our recent optical investigations on K_3C_{60} and Rb_3C_{60} single crystals (to be reported elsewhere) fully confirm our result on the pellet specimens, thus implying the intrinsic nature of the optical properties.

Several experimental results obtained with various techniques were also found to be in very good agreement with the weak coupling limit of the BCS theory. Particularly, we quote the specific heat, isotope effect and normal state susceptibility investigations by Ramirez et al. [5,6], the pressure and temperature dependence of the nuclear spin-lattice relaxation by Quirion et al. [7] and finally the muon-spin relaxation (μSR) experiments by Kiefl et al. [8].

It is of interest to compare now 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 use of the standard Eliashberg theory to calculate the optical conductivity for arbitrary impurity scattering in the local approximation. 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 [9]. While it reduces to the standard weak coupling BCS theory in the limit of the average phonon frequency being much greater than T_c and the renormalization parameter $\lambda=0$, the full theory takes into account the full details of the retarded electron-phonon interaction. With regard to the optical properties, a good agreement with experiment has been achieved beyond the Mattis-Bardeen theory [10] with regard to Holstein structure and impurity scattering. Note that the conventional Mattis-Bardeen theory of optical conductivity corresponds to a weak coupling superconductor with no phonon structure and in the dirty limit ($1/\tau \rightarrow \infty$). Development of formalism for the calculation of optical conductivity for arbitrary impurity scattering in the superconducting state has only recently been put forward [11]. This recent work includes both impurity- and phonon-assisted absorption processes whereby a photon creates a hole-particle pair in the metal and this pair relaxes through impurity or phonon scattering (this latter process being termed the Holstein process). The complex optical conductivity is thus calculated from the current-current correlation function [4,11].

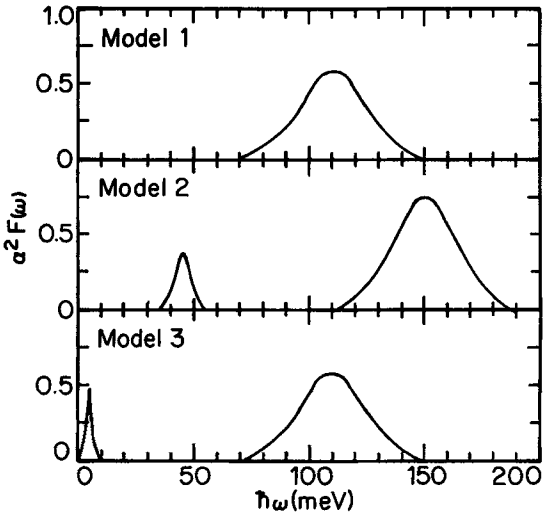

Fig.3: The model electron-phonon spectral functions $\alpha^2F(\omega)$ for both compounds which was used for the calculations described in the text and shown in Fig. 2, and 4.

Our primary goal here 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? We simply want to examine three models. In one we place a peak at high energy typical of intramolecular phonons and calculate the conductivity, in a second we add an extra peak to the spectrum of the first model at 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 very low energy to simulate librational or intermolecular phonons.

Fig. 3 shows the typical $\alpha^2F(\omega)$ spectra which we have used in our calculations, presented in Fig. 2 and 4. First of all, we have coupling to high energy intramolecular phonons (model 1) which we have simulated by placing a Lorentzian centered at ω_0= 0.15 eV for both K_3C_{60} and Rb_3C_{60} [4]. 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., $<\omega> > T_c$). The lower peak (omitted in the phonon spectrum of Rb_3C_{60}) is centered at ω_0= 0.045 eV (model 2). This peak was introduced only to provide a fit to the temperature dependence of the resistivity of K_3C_{60} [4]. Omitting this lower energy peak does not change our conclusions regarding the optical properties. Used as input in the Eliashberg equations and scaling this spectrum to give the correct T_c of 19 K and 29 K, we find that Δ=23.6 cm^{-1} and 35.9 cm^{-1}, $2\Delta/k_BT_c$=3.57 and 3.56, and λ=0.513 and 0.514 for K_3C_{60} and Rb_3C_{60}, respectively. Fig. 2 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 [10] is significantly improved.

Finally, we would like to briefly investigate the role of low frequency intermolecular or librational modes. By introducing a peak in the $\alpha^2F(\omega)$ spectrum at very low frequency (model 3), 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. The result of the calculation of the model 3 is presented in Fig. 4 for Rb_3C_{60} (a similar

result is obtained for K_3C_{60}). For clean to nearly dirty values of $1/\tau$, the theoretical optical conductivity displayed large Holstein structure in stark disagreement with the data and for $1/\tau$ approaching the dirty limit, the Holstein structure damped down but the theoretical conductivity ratio was always below the experimental data following more closely the Mattis-Bardeen curve. 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.

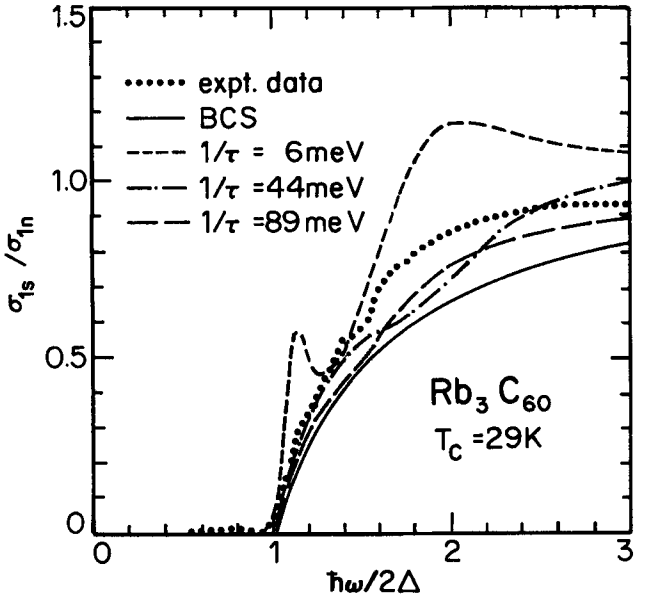

Fig.4: Optical conductivity in the superconducting state compared with the Eliashberg calculation after model 3 (Fig. 3) for different impurity parameters τ.

Conclusion

From our optical investigations on the alkali-doped K_3C_{60} and Rb_3C_{60} superconductors, we have found that the electrodynamic response of the superconducting ground state is in full agreement with that of a BCS singlet superconductor, and the magnitude of the single particle gaps also agrees with the weak coupling limit. Furthermore, beyond the standard BCS

approach, we have applied the conventional Eliashberg electron-phonon theory of superconductivity and found evidence for a pairing mechanism mainly involving high frequency intramolecular modes.

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