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RATIO BETWEEN THE JUMP IN THE SPECIFIC HEAT AND THE THERMODYNAMIC CRITICAL FIELD FOR THE DOPED FULLERENE SUPERCONDUCTORS

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ABSTRACT

The jump in the specific heat at T_c *and the critical fields of Type II superconductors are connected through a thermodynamic relation. The relationship leads to* $\Delta C/T_c = [dH_{c2}(T_c)/dT]^2/8\pi^2\kappa^2$ *where* $H_{c2}(T)$ *is the upper critical field and* κ *is the ratio between the penetration depth and coherence length. Application of this equation to the critical field data for the doped fullerene superconductor* K_3C_{60} *yields a value for* $\Delta C/T_c$ *of 380 mJ/mole/K*^{2} *which is far above the measured value of 68 ± 13 mJ/mole/K*^{2}*. We find that this discrepancy could be due to the electrons in the alkali doped fullerene superconductor being coupled to both a high intramolecular vibration mode and a low lying intermolecular vibration mode. The strong coupling Eliashberg formalism is used.*

INTRODUCTION

The discovery of superconductivity in the alkali-doped fullerene has created much excitement since it was so unexpected.[1] Their discovery has provided theorists with a new system to test various theories on. The values of the coherence length and penetration depth of these superconductors[2] place them into the category of being extreme Type II superconductors. The values[3] of the ratio $2\Delta(0)/k_BT_c$ for K_3C_{60} and Rb_3C_{60} (5.2 and 5.3 respectively) and the value[4] of the ratio between the jump of the specific heat at T_c and the normal phase specific heat at T_c for K_3C_{60} (2.0 ± 0.5) point to these superconductors being strong coupling (SC) superconductors. Our interest in these superconductors concerns the last feature. In particular, we are interested in the influence of strong el-ph (electron-phonon) coupling on the magnetic properties of alkali-doped fullerene superconductors.

It is the purpose of the present note to look at the connection between the critical fields and the jump in the specific heat at T_c within the framework of a strong coupling description of the superconducting state of the alkali-doped fullerene superconductors. In the Ginzburg-Landau (GL) theory,[5] the ratio between the jump in the specific heat and the critical temperature can be obtained from the value of the slope at T_c of the upper critical field dependence on T, *i.e.*,

$$\frac{\Delta C}{T_c} = \frac{1}{8\pi^2\kappa^2}\left(\frac{dH_{c2}(T)}{dT}\right)^2 \tag{1}$$

where $H_{c2}(T)$ is the upper critical field and κ is the ratio between the penetration depth and the coherence length. Applying this relationship to the measured critical field data for K_3C_{60} and Rb_3C_{60},[2,6] Dolgov and Mazin[5] obtained values for the jump in the specific heat of 380 mJ/mole/K^2 and 230 mJ/mole/K^2, respectively. Direct measurement of the specific heat jump at T_c of K_3C_{60} yielded a value of 68 ± 13 mJ/mole/K^2 for this ratio.[4] Dolgov and Mazin attributed the disorenacy between the calculated and the measured values to the presence of a renormalization factor (which is the coefficient of ϕ^4 in the expansion of the GL free energy) in the expression for the jump in the specific heat.

Substituting the relationship between the slope of the upper critical field, $dH_{c2}(T_c)/dT$, and the value of the upper critical field at $T=0\ K$, *i.e.*, the Werthamer-Helfand-Hohenberg formula.[7]

$$H_{c2}(T=0K) = \frac{0.69}{T_c}\left\{\frac{dH_{c2}(T)}{dT}\right\}_{T=T_c} \tag{2}$$

and the relation between the upper critical field $H_{c2}(T)$ and the thermodynamic critical field $H_c(T)$,[8]

$$H_{c2}(t=T/T_c) = 2^{1/2}\ H_c(t)\kappa_1 \tag{3}$$

where κ_1 is the temperature dependent parameter which is equal to 1.20κ at $T=0$ K, into equation (1), we obtain

$$\frac{\Delta C}{T_c} = 0.213\left[\frac{H_c(0)}{T_c}\right]^2 \tag{4}$$

The ratio between $\Delta C/T_c$ and $[H_c(0)/T_c]^2$, where $H_c(0)$ is the value of the thermodynamic critical field at $T=0$ K, is an universal constant in the microscopic BCS theory.[9] In the BCS theory, the value of this ratio is 0.24. This, however, differs from the GL value by only 11%. The difference in the predicted value of $\Delta C/T_c$ (380 mJ/mole/K^2) using the GL approach and of the observed value (68 mJ/mole/K^2) for K_3C_{60} could be an indication that the weak coupling BCS theory is inappropriate for the doped fullerene superconductors. Strong coupling corrections may have to be taken into consideration.

In a series of papers, Carbotte and co-workers[10] obtained the strong coupling corrections to various BCS ratios. They showed that the corrections were proportional to the ratio T_c/ω_0, where ω_0 is the phonon frequency characteristic of the system. The corrections are the results of expanding the frequency dependent el-ph coupling parameter in terms of the moments of the spectra density function. They found that the SC expressions for the BCS ratios would be in the form

$$\text{BCS Ratio} = \text{BCS value}\left\{1+a\left[\frac{T_c}{\omega_0}\right]^2 ln\left[\frac{\omega_0}{bT_c}\right]\right\}$$

If the electrons are coupled only to the intramolecular $H_g(8)$ mode as is suggested by Varma *et al.*,[11] the corrections would be negligible since the ratio $(T_c/\omega_H)^2 \sim (10^0/10^2)^2 \sim 0$. The value of $(T_c/\omega_H)^2$ for the case of coupling to the $H_g(8)$ mode is close to the values for the weak coupling superconductors, which are well described by the BCS theory.

To reconcile several contradictory properties seen in the fullerene superconductors, *i.e.*, the non BCS behaviours of the critical field, penetration depth and the NMR relaxation rates, Mazin *et al.*,[12] introduced an additional coupling to the low frequency mode. Crespi and Cohen[13] have analyzed the temperature dependent normal phase resistivity of single crystal Rd_3C_{60} and came to the conclusion that the electrons were also coupled to a low frequency mode. Varelogiannis[14] has looked at the effects of the spectral function being the sum of two Einstein peaks on the strong coupling theory. Recently, the present author[15] showed that the observed deviation of other ratios of thermodynamic properties of the fullerene superconductors from their BCS values could be due to the el-ph coupling to a low frequency mode.

$[T_c/H_c(0)]^2$ RATIO

The difference between the free energy of the normal and superconducting phases is equal to the square of the thermodynamic critical field divided by 8π. Bardeen and Stephen[16] showed that the difference in the free energies is given by

$$F/N(0) = -\pi k_B T\sum_m\left\{\left(\omega_m^2+\Delta_m^2\right)^{1/2} - |\omega_m|\right\}\times\left\{Z_s(m) - Z_N(m)\frac{\omega_m}{\left[\omega_m^2+\Delta_m^2\right]^{1/2}}\right\} \tag{5}$$

where $Z_S(m)$ and $Z_N(m)$ are the normalization constants for the superconducting and normal phases and which have been calculated for the present model in ref. 15. The additional coupling to a low frequency mode can be incorporated into Eliashberg equations using a spectral function $\alpha^2F(\Omega)$ which is the sum of two δ function peaks, *i.e.*,

$$A_L\delta(\Omega-\omega_L)+A_H\delta(\Omega-\omega_H) \qquad (6)$$

in the expressions for the various moments of the frequency dependent el-ph coupling parameter. In the above, ω_H is the frequency of the intramolecular $H_g(8)$ vibration mode (1,400 cm^{-1}); ω_L, frequency of a low lying intermolecular vibration mode (<100 cm^{-1}) and A_L and A_H are the weights of the two distribution. Substituting the two peak spectral functions into the moments appearing in the normalization constant $Z_S(T)$, we get

$$Z_s(T)=1+\lambda+\Delta_0^2\left\{A_L\left[\frac{1}{\omega_L}\right]^3 ln\left[\frac{T_c}{\omega_L}\right]+A_H\left[\frac{1}{\omega_H}\right]^3 ln\left[\frac{T_c}{\omega_H}\right]\right\} \qquad (7)$$

where

$$\lambda = \lambda_L + \lambda_H \text{ with } \lambda_{L(H)} = A_{L(H)}/\omega_{L(H)} \qquad (8)$$

If one has non delta function distributions centered at ω_L and ω_H, one would have to replace the factors $A_i(T_c/\omega_i)^2 ln(\omega_i/T_c)$ appearing in equation (7) by more complicated expressions. Carbotte and co-workers[10] replaced the factors by the approximation $a(T_c/\omega_i)^2 ln(\omega_i/bT_c)$, where a and b are phenomenological constants. Making similar approximation, equation (7) becomes

$$Z_s(m)=1+\lambda+a_1\left[\frac{T_c}{\omega_L}\right]^2\lambda_L ln\left[\frac{\omega_L}{b_1T_c}\right]+a_2\left[\frac{T_c}{\omega_H}\right]^2\lambda_H ln\left[\frac{\omega_H}{b_2T_c}\right] \qquad (9)$$

$Z_N(m)$ is obtained by setting Δ in equation (7) to zero.

Equation (5) can be written as

$$\Delta F/N(0)=-\pi k_BT\sum_m\left\{Z_s(m)\omega_m\left[\left(1+1/U_m^2\right)^{1/2}-1\right]+Z_N(m)\omega_m\left[\left(1+1/U_m^2\right)^{-1/2}-1\right]\right\} \qquad (10)$$

where $U_m = \Delta_m/\omega_m$. Close to T_c, $1/U_m^2$ is small and so we can expand the RHS of equation (10) in powers of $1/U_m^2$. Doing this, we find that the leading term is of fourth order in the order parameter Δ_0. Expansion to higher orders is needed only if one is interested in the difference in the free energies at temperatures below T_c.

Carrying out the expansion we obtain

$$\frac{\Delta F}{N(0)}=-\pi k_BT\sum_m\left\{\frac{1}{8}\left[\frac{1}{U_m}\right]^4(Z_s-3Z_N)\omega_m-\frac{1}{2}\left[\frac{1}{U_m}\right]^2(Z_s-Z_N)\omega_m\right\} \qquad (11)$$

Performing the summation, we get

$$F/N(0)=(1/4)B_1(T)\Delta_0^4/(\pi k_B T)^2 \tag{12}$$

where

$$B_1(T)=(7/8)(1+\lambda)\zeta(3)+(\pi k_B T)^2\{c(T)-a(T)+b(T)/4\} \tag{13}$$

with $\zeta(3)$ being the Reimann zeta function and with $a(T)$, $b(T)$ and $c(T)$ being the moments of the spectral density function defined as

$$a(T)=\int_0^{\infty}\frac{d\Omega\alpha^2F(\Omega)}{\Omega^3}ln\left[\frac{1.13\Omega}{k_BT}\right] \tag{14a}$$

$$b(T)=\int_0^{\infty}\frac{d\Omega\alpha^2F(\Omega)}{\Omega^3} \tag{14b}$$

and

$$c(T)=\int_0^{\infty}\frac{d\Omega\alpha^2F(\Omega)}{\Omega^3}ln^2\left[\frac{1.13\Omega}{k_BT}\right] \tag{14c}$$

The moments $a(T)$ and $b(T)$ appear when summing $(3Z_N-Z_S)/|\omega_m|^3$, while the moment $c(T)$ appear when summing $(Z_S-Z_N)/|\omega_m|$.

Substituting the two peak spectral function into the integrand of the three moments, we find that the moments are the sums of two contributions, one from the high frequency mode and the other from the low frequency mode. Varelogiannis[14] replaces the sums by weighted averages. We will, however, retain the moments as the sums of two contributions. The SC corrections are obtained by multiplying the moments by $(k_BT)^2$. Putting everything together, we get

$$\begin{aligned}H_c^2(0)/8\pi=(N(0)/4)\{\Delta_0^4/(\pi k_BT_c)^2\}\{\frac{7}{8}(1+\lambda)\zeta(3)+\\ \hat{a}\left[\frac{T_c}{\omega_L}\right]^2\lambda_L\,ln^2\left[\frac{\omega_L}{bT_c}\right]-\hat{a}'\left[\frac{T_c}{\omega_L}\right]^2\lambda_L\,ln\left[\frac{\omega_L}{b'T_c}\right]+(\hat{a}''/4)\left[\frac{T_c}{\omega_L}\right]^2\lambda_L+\\ \hat{b}\left[\frac{T_c}{\omega_H}\right]^2\lambda_H\,ln^2\left[\frac{\omega_H}{b'T_c}\right]-\hat{b}'\left[\frac{T_c}{\omega_H}\right]^2\lambda_H\,ln\left[\frac{\omega_H}{b'T_c}\right]+(\hat{b}''/4)\left[\frac{T_c}{\omega_H}\right]^2\lambda_H\}\end{aligned} \tag{15}$$

where $\hat{a}(\hat{b})$, $\hat{a}'(\hat{b}')$ and $\hat{a}''(\hat{b}'')$ are phenomenological constants. Noting that $T_c \sim 10^0$ meV, $\omega_H \sim 100$ meV and $\omega_L \sim 10^0$ meV, we have at T_c

$$(T_c/\omega_H)^2 << (T_c/\omega_L)^2$$

We can therefore drop the terms proportional to $(T_c/\omega_H)^2$ in equation (15). Then using the expression for the ratio between $\Delta(0)$ and T_c given in ref. 15, we obtain

$$\frac{\gamma T_c^2}{H_c^2(0)} = 0.168\left\{1 - \left[\frac{T_c}{\omega_L}\right]^2 \lambda_L \ln\left[\frac{\omega_L}{bT_c}\right]\right\} \tag{16}$$

In ref. 15, we showed that the ratio between the jump in the specific heat at T_c and the normal phase specific heat at T_c is given by

$$\frac{\Delta C}{\gamma T_c} = 1.43\left\{1 + a_L\left[\frac{T_c}{\omega_L}\right]^2 \lambda_L \ln\left[\frac{\omega_L}{bT_c}\right]\right\} \tag{17}$$

Combining equations *(16)* and *(17)* together, we get

$$\frac{\Delta C}{T_c} = 0.24\left\{1 - a\left[\frac{T_c}{\omega_L}\right]^2 \lambda_L \ln\left[\frac{\omega_L}{bT_c}\right]\right\}\left[\frac{H_c(0)}{T_c}\right]^2 \tag{18}$$

Equation *(18)* shows that the ratio between C/T_c and $[H_c(0)/T_c]^2$ is less than 0.24, the BCS value. This reduction of the value of the ratio occurs only if the electrons are also coupled to low frequency mode. If the coupling is only to high frequency $H_g(8)$ intramolecular mode, the value of the ratio would not change from the BCS value and the discrepancy between the observed and predicted values of C/T_c of K_3C_{60} would remain unexplained.

DISCUSSION

One of the first studies on the alkali-doped fullerene superconductors, found that the increase in the T_c's of the A_3C_{60} superconductors correlated with the increase in the density of states occurring when larger ions were inserted into the fcc structure of the C_{60} system. Fleming *et al.*,[17] hypothesize that the observed correlation meant that the el-ph coupling was little effected by the alkali ions or that the intermolecular phonon modes are weakly affected by changes in the lattice parameter. This would imply that the strong coupling corrections appearing in equation *(14)* are controlled by the changes in T_c (through its appearance in the ratio (T_c/ω_L)). Therefore, strong coupling corrections for Rb_3C_{60} would be greater than that for K_3C_{60}. Since the strong coupling correction in Rb_3C_{60} is stronger, the ratio between the specific heat jump and T_c in this doped fullerene superconductor is expected to be less than 41 mJ/mole/K^2, the value if the correction in Rb_3C_{60} is the same as that in K_3C_{60}.

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