Asian Journal of Chemistry Vol. 12, No. 1 (2000), 85–88

Study of Some Organic Superconductors Using BCS-Theory

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Using the conventional phonon exchange mechanism of superconductivity, we have succeeded in reproducing the transition temperature T_c of some newly discovered organic superconductors by introducing certain modifications to the well known BCS-formulae for T_c. Taking the value of density of states at the Fermi surface, N(o), calculated from d.c. paramagnetic measurement, the values of $\Delta_0(o)$ and $H_c(o)$ have been evaluated in the weak coupling limit.

INTRODUCTION

A new class of materials[1, 2] based on the sulphur containing organic donor bis-(ethylidene-2-thio)-tetra-thiofulvalence (BEDT-TTF) has yielded over ten organic super-conductors with the critical temperature $T_c < 8$ K. Sugano[3] has discovered a new ambient-pressure organic superconductor $(BEDT\text{-}TTF)_2[Cu(NCS)_2]$ with $T_c = 10.4 \pm 0.3$ K by means of dc magnetic suspeptibility measurements[4, 5] and confirmed by resistivity measurements[6]. This material consists of sheets of the BEDT–TTF molecules with a zigzag arrangement of molecular dimers in the crystallographic bc plane and the sheets are interleaved by sheets of the $[Cu(NCS)_2]^-$ ions[7]. Thus $(BEDT\text{-}TTF)_2[Cu(NCS)_2]$ has a unique structure that does not form face to face stacking columns as found commonly in conventional organic conductors. It forms a two-dimensional interaction network where one dimer is nearly perpendicular to the neighbouring dimer. Since there is an unpaired electron per dimer, the unit dimer in $[BEDT\text{-}TTF]_2[Cu(NCS)_2]$ may be regarded as an atom having one valence electron like alkali metals. Therefore $(BEDT\text{-}TTF)_2[Cu(NCS)_2]$ is of great interest in the sense that it is not only the organic superconductor with highest T_c at present, but also the organic conductor with unique structural feature. Graebnes *et al.*[8] has performed high resolution specific heat-measurements on a single crystal of $K\text{-}(BEDT\text{-}TTF)_2Cu(NCS)_2$ in magnetic field up to ST reveal a jump SC at T_c. Comparison with $\gamma = 34$ ms/mol k^2 derived from Pauli paramagnetism yields $\Delta_c/\gamma T_c = 1.50 \pm 0.15$ within experimental error of the BCS value of 1.43. However, the magnetic penetration depth by muon-spin relaxation measurements[9], suggests the superconducting

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pairing with line nodes in the energy gap. It is a big question whether the mechanism of organic superconductor can be explained by the BCS[10] theory or not. If one sticks to the measurements of surface impedance by Klein *et al.*[11] and Harshman *et al.*[12], then these measurements suggest the conventional wave pairing.

In this paper, using conventional phonon exchange mechanism of superconductivity, we have succeeded in reproducing the transition temperature T_c of a large number of newly discovered organic superconductors. We have introduced certain modifications to the well known BCS-formula for T_c.

Derivation of the modified BCS Formula for T_c

We start with the two familiar equations one for the superconducting energy gap parameters $\Delta_0(o)$ and the other for the transition temperature T_c of the BCS theory which are given as[13]

$$\Delta_0(o) = \hbar w_D / \sin \hbar \left[\frac{1}{N(o)v}\right], \quad (1)$$

and

$$\left(\frac{1}{N(o)V}\right) = \int_0^{\hbar w_D} \frac{dx}{x} \tan\hbar \left(\frac{x}{2K_\beta T_c}\right), \quad (2)$$

For simple materials like Al, Cd etc. the values of Debye frequency $\hbar w_D$ and the energy gap parameter at zero temperature are known from phonon dispersion measurements and the tunnelling measurements. With the help of (1) the value of the strength parameter g = [N(o)V] is determined. N(o) is the density of states at Fermi surface and V is the electron-phonon interaction potential. If one evaluates the integral of (2) keeping both T_c and $\hbar w_D$ constant then N(o)V can be reproduced. In the case of organic superconductors $\hbar w_D$ is not known experimentally. We conjecture that for organic superconductor the value of Debye temperature θ_D at $T = T_c$ is different from the value at T = 0 K. Such temperature variation of θ_D has been known for metallic systems like Al and Mg[14]. In order to account for the temperature variation of θ_D, we assume that

$$\theta_D^{(c)}(T) = \theta_D^{(o)} \left[1 - \gamma \frac{T}{\theta_D^{(o)}}\right] \text{ for } T < T_c, \quad (3)$$

where γ is an adjustable parameter, $\theta_D^{(c)}$ and $\theta_D^{(o)}$ are the value of Debye temperature at $T = T_c$ and T = 0 K. The BCS formula for T_c in the weak coupling limit is given by

$$T_c = 1.14\theta_D \exp[-1/g], \quad (4)$$

with the help of (3), the BCS formula (4) may be modified as

$$T_c^{(MBCS)} = \frac{1.14\,\theta_D^{(o)} \exp[-1/g]}{[1 + 1.14\gamma \exp[-1/g]]}, \quad (5)$$

Using eqn. (5) we have calculated the transition temperature of organic superconductor. This has been given in Table-1. Using the BCS weak coupling limit, we have also calculated Δ_0 and $GH_c(o)$ for these superconductors given in Table-2.

TABLE-1

Organic Superconductor	T_c (Expt) (K)	$\theta_D^{(o)}$ (K)	$g = N(o)V$	$\theta_D^{(o)}$ (K)	γ	TC[MBCS] (K)
$(BEDT-TTF)_2$ $Cu[NCS]_2$	10.4	154	0.40	112	4.0	10.48
$(BEDT-TTF)_2$ $Cu[N(CN)_2]CN$	10.7	157	0.40	114	4.0	10.69
$(BEDT-TTF)_2$ $Cu[N(CN)_2]Br$	11.6	171	0.40	124	4.0	11.64
$(BEDT-TTF)_2$ $Cu[N(CN)_2]Cl$	12.8	188	0.40	137	4.0	12.80

TABLE-2

DENSITY OF STATE $N(o) = 5.8 \times 10^{33}$ $exp^{-1}cm^{-3}$

Organic Superconductor	Year of disovery	Country	$\Delta_0(o) = 1.76K_\beta T_c$ (lrg)	$H_c^2(o) = 4\pi\Delta_0^2(o)N(o)$ (gauss)	T_c (K)
$(BEDT-TTF)_2$ $Cu[NCS]_2$	1988	U.S.A.	2.52×10^{-15}	680	10.4
$(BEDT-TTF)_2$ $Cu[H(CN)_2]CN$	1991	Japan	2.60×10^{-15}	702	10.7
$(BEDT-TTF)_2$ $Cu[N(CN)_2]Br$	1990	U.S.A.	2.80×10^{-15}	756	11.6
$(BEDT-TTF)_2$ $Cu[N(CN)_2]Al$	1990	U.S.A.	3.10×10^{-15}	837	12.8

RESULTS AND DISCUSSION

In the present calculation, we have been able to obtain the transition temperature of some of the organic super-conductors by suitably modifying the well known BCS formulae for T_c. Taking the value of density of states N(o) calculated from d.c. paramagnetic susceptibility measurement the value of energy gap parameter $\Delta_o(o)$ and critical field $H_c(o)$ are determined. The phonon exchange mechanism can however be checked by evaluating various normal state properties of organic superconductors.

ACKNOWLEDGEMENT

Authors are thankful to Dr. N.M. Sahay, Professor and Ex-Head of the Department of Chemistry, Veer Kunwar Singh University, Ara. We give our sincere thanks to Prof. I.B. Singh, former Head of the Department of Chemistry, Magadh University, Bodh Gaya and Ex-Vice-Chancellor, Bhagalpur University for the encouragement and valuable suggestions.

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(*Received*: 28 June 1999; *Accepted*: 5 October 1999) AJC- 1855