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Relationship between positron lifetime and temperature in C_{60}

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Abstract. The positron lifetime is related to the temperature in C_{60}, i.e. the positron lifetime increases as a function of increasing temperature between 10 and 300 K. In this paper we adopted the extended Su–Schrieffer–Heeger model; by using the method of the Green function at a finite temperature, we then deduce a relation between the positron lifetime and temperature. The results show that the theoretical calculation may be in accordance with experiment.

Recently, the discovery of C_{60} has generated great interest in research in condensed-matter sciences [1,2]. Its caged molecular structure has special significance in the synthesis of new chemical compounds and of high-temperature superconducting materials. As is well known, positron annihilation spectroscopy is a special probe for the characterization of the electronic and defect structures of solids [3].

Experiments have discovered a monotonic increase in the positron lifetime of C_{60} as the temperature increases between 10 and 300 K [4]. The aim of the present paper is to derive the form of the above function from theoretical calculation. Although there might be other possible forms of Hamiltonian in the C_{60} system, we consider that used by Harigaya and Terai [5]. Because the electron–phonon coupling interaction is assumed to be intense, Coulomb interactions between electron and electron or between electron and positron can be neglected. In terms of the extended Su–Schrieffer–Heeger (SSH) [6] model, the Hamiltonian is given by

$$H = \sum_{\langle ij\rangle,s}[t_0 - \alpha(u_i^{(j)} + u_j^{(i)})](C_{is}^{+}C_{js} + hc) + \sum_{\langle ij\rangle,s}[t_0' - \alpha'(u_i^{(j)} + u_j^{(i)})](d_{is}^{+}d_{js} + hc)$$
$$+\frac{K}{2}\sum_{\langle ij\rangle}(u_i^{(j)} + u_j^{(i)})^2 + \tfrac{1}{4}\sum_{i,j} M[(\dot{u}_i^{(j)})^2 + (\dot{u}_j^{(i)})^2] \tag{1}$$

where t_0 and t_0' are the hopping integrals of the electron and positron, respectively, α and α' are the electron–phonon and positron–phonon coupling constants, respectively, C_{is} (d_{is}) and C_{is}^{+} (d_{is}^{+}) are the annihilation and creation operators, respectively, of an electron (a positron) at the ith carbon atom, $u_i^{(j)}$ is the displacement of the ith atom in the direction opposite to the jth atom, and the quantity $u_i^{(j)} + u_j^{(i)}$ is the change in bond length between the ith and jth atoms. The sum is taken over all nearest-neighbour pairs $\langle ij\rangle$ of carbon atoms.

Let

$$\Psi_{1s}(r_n) = \frac{1}{\sqrt{2}}(-1)^n(\mathrm{i}C_{2n,s} + C_{2n+1,s})$$
$$\Psi_{2s}(r_n) = \frac{1}{\sqrt{2}}(-1)^n(C_{2n,s} + \mathrm{i}C_{2n+1,s}) \tag{2}$$

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and

$$\Phi_{1s}(r_n) = \frac{1}{\sqrt{2}}(-1)^n(\mathrm{i}d_{2n,s} + d_{2n+1,s})$$
$$\Phi_{2s}(r_n) = \frac{1}{\sqrt{2}}(-1)^n(d_{2n,s} + \mathrm{i}d_{2n+1,s}). \tag{3}$$

By means of equations (2) and (3), the Hamiltonian can be rewritten as

$$\begin{aligned} H = &-\sum_{n,s} \mathrm{i}t_0 \sum_{i=1}^{3} A_i \left(\Psi_{1s}^{+}(r_n)\frac{\mathrm{d}\Psi_{1s}(r_n)}{\mathrm{d}X_{n,i}} - \Psi_{2s}^{+}(r_n)\frac{\mathrm{d}\Psi_{2s}(r_n)}{\mathrm{d}X_{n,i}} \right) \\ &- \sum_{n,s} \mathrm{i}t_0' \sum_{i=1}^{3} A_i \left(\Phi_{1s}^{+}(r_n)\frac{\mathrm{d}\Phi_{1s}(r_n)}{\mathrm{d}X_{n,i}} - \Phi_{2s}^{+}(r_n)\frac{\mathrm{d}\Phi_{2s}(r_n)}{\mathrm{d}X_{n,i}} \right) \\ &+ \sum_{n,s} \Delta(r_n)[\Psi_{1s}^{+}(r_n)\Psi_{2s}(r_n) + \Psi_{2s}^{+}(r_n)\Psi_{1s}(r_n)] \\ &+ \sum_{n,s} \frac{\alpha'}{\alpha}\Delta(r_n)[\Phi_{1s}^{+}(r_n)\Phi_{2s}(r_n) + \Phi_{2s}^{+}(r_n)\Phi_{1s}(r_n)] + \frac{K}{2\alpha^2}\sum_n \Delta^2(r_n) \\ &+ \frac{M}{8\alpha^2}\sum_n [\dot{\Delta}(r_n)]^2 \end{aligned} \tag{4}$$

where

$$A_i = (X_{n+1} - X_n)_i$$
$$\Delta(r_n) = -\alpha(u_{2n}^{(2n+1)} + u_{2n+1}^{(2n)}) \tag{5}$$

and

$$r_n = (X_{n,1}, X_{n,2}, X_{n,3}). \tag{6}$$

Making use of

$$r_n \to r \qquad \sum_n \to \frac{1}{\Omega}\int \mathrm{d}^3 r \tag{7}$$

the quantity H in (4) becomes

$$\begin{aligned} H = &-\frac{\mathrm{i}t_0}{\Omega}\sum_s \int \mathrm{d}^3 r \left(\Psi_{1s}^{+}(r)\sum_{i=1}^{3}\frac{\mathrm{d}\Psi_{1s}(r)}{\mathrm{d}x_i}A_i - \Psi_{2s}^{+}(r)\sum_{i=1}^{3}\frac{\mathrm{d}\Psi_{2s}(r)}{\mathrm{d}x_i}A_i \right) \\ &- \frac{\mathrm{i}t_0'}{\Omega}\sum_s \int \mathrm{d}^3 r \left(\Phi_{1s}^{+}(r)\sum_{i=1}^{3}\frac{\mathrm{d}\Phi_{1s}(r)}{\mathrm{d}x_i}A_i - \Phi_{2s}^{+}(r)\sum_{i=1}^{3}\frac{\mathrm{d}\Phi_{2s}(r)}{\mathrm{d}x_i}A_i \right) \\ &+ \frac{1}{\Omega}\sum_s \int \mathrm{d}^3 r \Delta(r) \Big\{ [\Psi_{1s}^{+}(r)\Psi_{2s}(r) + \Psi_{2s}^{+}(r)\Psi_{1s}(r)] \\ &+ \frac{\alpha'}{\alpha}[\Phi_{1s}^{+}(r)\Phi_{2s}(r) + \Phi_{2s}^{+}(r)\Phi_{1s}(r)] \Big\} \\ &+ \frac{K}{2\alpha^2\Omega}\int \mathrm{d}^3 r \Delta^2(r) + \frac{M}{8\alpha^2\Omega}\int \mathrm{d}^3 r [\dot{\Delta}(r)]^2. \end{aligned} \tag{8}$$

The phonon field $\Delta(r)$ is

$$\Delta(r) = \frac{1}{\sqrt{\Omega}}\sum_q (a_q \exp(\mathrm{i}q \cdot r) + a_q^{+}\exp(-\mathrm{i}q \cdot r)) \tag{9}$$

where a_q and a_q^+ are annihilation and creation operators, respectively, of a phonon to wavevector q.

In the case of a finite temperature, the forms of field operators become

$$\begin{aligned}\Psi_{\alpha s}(r,\tau) &= \exp(k\tau/h)\Psi_{\alpha s}(r)\exp(-k\tau/h)\\ \Psi^+_{\alpha s}(r,\tau) &= \exp(k\tau/h)\Psi^+_{\alpha s}(r)\exp(-k\tau/h)\end{aligned} \tag{10}$$

and

$$\begin{aligned}\Phi_{\alpha s}(r,\tau) &= \exp(k\tau/h)\Phi_{\alpha s}(r)\exp(-k\tau/h)\\ \Phi^+_{\alpha s}(r,\tau) &= \exp(k\tau/h)\Phi^+_{\alpha s}(r)\exp(-k\tau/h)\end{aligned} \tag{11}$$

where

$$k = H - \mu_- N_- - \mu_+ N_+ \tag{12}$$

N_- (N_+) is the number operator of the electron (positron), and μ_- (μ_+) is the chemical potential of the electron (positron).

The Green function at finite temperatures [7] is defined by

$$g_{e,\alpha\beta}(r\tau, r'\tau') = -\exp(\beta\Omega)\,\mathrm{Tr}\left\{\exp[-\beta(H-\mu_-N_- - \mu_+N_+)] \times T_\tau \sum_s [\Psi_{\alpha,s}(r,\tau)\Psi^+_{\beta,s}(r',\tau')]\right\} \tag{13}$$

$$g_{p,\alpha\beta}(r\tau, r'\tau') = -\exp(\beta\Omega)\,\mathrm{Tr}\left\{\exp[-\beta(H-\mu_-N_- - \mu_+N_+)] \times T_\tau \sum_s [\Phi_{\alpha,s}(r,\tau)\Phi^+_{\beta,s}(r',\tau')]\right\}. \tag{14}$$

The positron partial annihilation rate $R(P)$ can be described by the two-particle Green function at zero temperature [8], which may be popularized to the case of non-zero temperature, namely

$$R(P) = \frac{\lambda}{\Omega}\int \mathrm{d}^3r\,\mathrm{d}^3r' \exp[\mathrm{i}P\cdot(r-r')]g_{ep}(r\tau, r'\tau; r\tau^+, r'\tau^+) \tag{15}$$

where the two-particle Green function g_{ep} at the temperature is given by

$$g_{ep}(x,x';y,y') = \frac{\mathrm{Tr}\{\exp[-\beta(H_0-\mu_-N_- - \mu_+N_+)]T_\tau \sum_\alpha [\Psi_\alpha(x)\Phi_\alpha(x')\Phi^+_\alpha(y')\Psi^+_\alpha(y)]\}}{\mathrm{Tr}\{\exp[-\beta(H_0-\mu_-N_- - \mu_+N_+)]u(\beta,0)\}} \tag{16}$$

where $X = (x,\tau)$, $X' = (x',\tau')$.

The electron–positron Green function g_{ep} has the well known perturbation expansion in terms of Feynman graphs. The major contribution to the annihilation rate is given by the diagrams shown in figure 1. The single solid lines represent the free-electron propagator and the double solid lines the free-positron propagator. The ripply lines represent the phonon propagator.

The total annihilation rate of positron in C_{60} is

$$\Gamma = \int \mathrm{d}^3P\, R(P). \tag{17}$$

The contribution of figure 1 to Γ obtained from (15)–(17) can be expressed as

$$\Gamma \approx B - CT \tag{18}$$

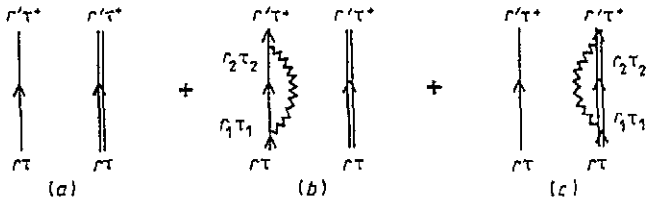

Figure 1. Feynman diagram for positron annihilation in C_{60}. The single solid lines, double solid lines and ripply lines denote electron, positron and phonon lines, respectively.

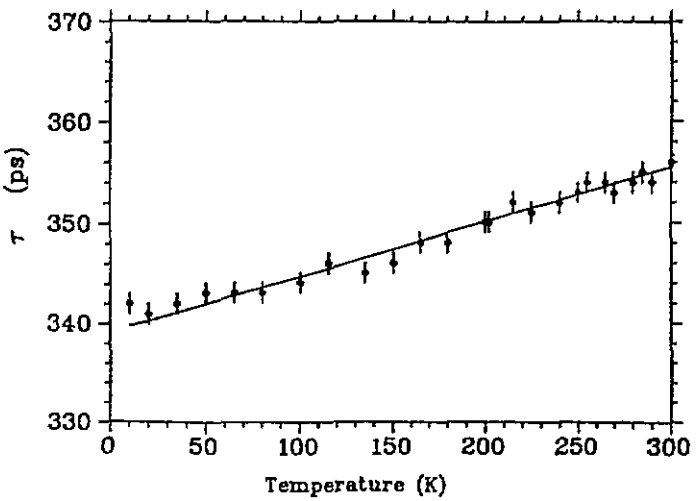

Figure 2. Positron lifetime of C_{60} versus temperature. The solid line shows the theoretical results of this paper. The experimental data points measured in [4] are given for comparison.

where

$$B = \lambda N^2(0) E_F^2 \left((2\pi)^3 + \frac{2N(0)E_F}{\pi(2\pi)^3} \right)$$
$$C = \lambda N^2(0) k_B \left(2(2\pi)^3 \mu_- + \frac{2E_F^2 N(0) \ln 2}{\pi(2\pi)^4} \right) \tag{19}$$

and here $N(0)$ is the average electron density in C_{60}, k_B the Boltzmann constant and E_F the Fermi energy.

From $\tau = \Gamma^{-1}$, the values of B and C can be calculated using the different two sets of experimental values in [4]. For example, when choosing $\tau = 343$ and 350 ps for $T = 65$ and 200 K, respectively, from [4],

$$B \approx 0.002\,94 \text{ ps}^{-1} \qquad C \approx 0.000\,000\,43 \text{ K}^{-1} \text{ ps}^{-1}. \tag{20}$$

Since $CB^{-1} \approx 0.000\,15$ K^{-1} and $CB^{-2}T \gg \frac{1}{2}C^2B^{-3}T^2$ between 10 and 300 K, and using equation (18),

$$\tau \approx C_1 + C_2 T \tag{21}$$

where

$$C_1 = B^{-1} \qquad C_2 = CB^{-2}. \tag{22}$$

According to equations (21) and (22), the lifetime results for C_{60} as a function of temperature between 10 and 300 K are as plotted in figure 2. Obviously, our calculations obtained from (21) are in excellent agreement with the experimental values in [4].

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