

Positron Energy Loss and the Interaction between Positrons and Soliton-Like Electron Density in Graphite-Alkali Metal Intercalation Compounds*

Yoshiki Satou, Tomoaki Sasaki,[†] Makoto Saito, Kohei Yamada, and Ikuzo Kanazawa

Department of Physics, Tokyo Gakugei University, Koganei-shi, Tokyo 184-8501, Japan

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We have analyzed the narrow components in the positron annihilation angular correlation spectra of stage-2 graphite-potassium intercalation by using the topological charge density of the skyrmion-like soliton, and have discussed the energy loss rate for low-energy positrons through “topological quasi-positronium” effect.

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I. INTRODUCTION

It is known that alkali-metal graphite intercalation compounds (AGIC) have catalytic activities for hydrogen layer of the compounds. The behaviors of these compounds have been known to depend on the structure, sort of metals and temperature. Hydrogens are physisorptively accommodated in the molecules in the interstices among the alkali-metal ions in intercalant layers of stage-2 compounds $C_{24}M$ ($M=K, Rb$ and Cs) at temperatures below 200 K [1–3]. Chemisorption of hydrogen takes place in both first and second stage compounds at higher temperatures [4, 5]. In order to clarify the mechanism of hydrogen chemisorption in the graphite compounds and properties of hydrogen-absorbed graphite compounds, several studies have been carried out. The change in magnetic susceptibilities as a function of hydrogen content has been investigated for hydrogen-chemisorbed C_8K [6]. It has been suggested that dissolved hydrogen in C_8K is paramagnetic by means of magnetic resonance studies [7]. The studies of means of ESR and electrical conductivity [8] have shown that in C_8K , $C_{24}K$, and $C_{24}Rb$, the hydride ions are stabilized after the dissociation of the hydrogen molecules into atoms and the subsequent charge transfer, and in C_8Rb hydrogens are absorbed in the atomic form. Recently the present authors have discussed the mechanism of anomalous magnetic effect due to hydrogen uptake in C_8RbH_x with the theoretical formula, which is extended from “topological quasi-hydrogen” model [24]. It is suggested strongly that the hydrogen state in C_8RbH_x might have the Kondo-like property [24].

Positron annihilation spectroscopy is a useful method in the study of electronic structures of materials. This spectroscopy has been used in the investigation of electronic structures in graphite and AGICs. The momentum distribution of σ and π -electrons in a graphite crystal have been studied by angular correlation of positron annihilation radiation (ACPAR) [9–12], and by Doppler-broadening positron annihilation radiation (DBPAR) [13]. ACPAR spectra of C_8K and $C_{24}K$, after subtraction of a broad contributions which accounts for annihilation with the graphite σ and π -electrons. Features of the nar-

row components in ACPAR spectra are in good agreement with the quasi-two dimensional electronic structures, which might correspond to the interlayer state with quasi-two dimensional free electron character parallel to the carbon planes [16]. In addition, hydrogen physisorption and chemisorption effects in AGICs have been studied by DBPAR [17–20]. The DBPAR spectra line shape of C_8K became sharp through hydrogen chemisorption at 300 K [17, 18], while the spectral line-shape of $C_{24}Cs$ became broad through the physisorption of hydrogen molecules at 77 K [18, 19]. The intensity of the narrow component of DBPAR spectrum of C_8Rb was suppressed through hydrogen absorption at 300 K, while it decreased at first and then increased through the absorption at the 373 K [20]. From the change in DBPAR spectral line-shape of C_8Rb , the hydrogen accommodated in C_8Rb is considered to be atomic at 300 K and to be in the form of the hydride ion at 373 K. There exist some problems in the assumption that the narrow component of positron annihilation spectra corresponds to the interlayer state in AGICs. That is, the energy level of the interlayer state in the second-stage AGICs is above Fermi energy E_F . Furthermore, it looks like that the narrow component is not attributed to the simple positronium Ps in AGICs, because simple Ps cannot exist in the metallic state of AGICs.

In this study, we analyze the ACPAR spectra in $C_{24}K$ [21] with the theoretical formula, which is extended from “topological quasi-positronium” model [24] and discuss the origin of the anisotropic narrow components in ACPAR spectra. In addition, we will discuss the positron energy loss due to the interaction between positrons and skyrmion-like electron density in graphite-alkali metal intercalation compounds.

II. TOPOLOGICAL QUASI-POSITRONIUM AND POSITRON ENERGY LOSS

The structure of the stage-1 and stage-2 alkali-metal-GIC's C_8K and $C_{24}K$ are shown in Figure 1. In the case of the Stage-1 compound C_8K , the alkali-metal atoms form a closed-packed two-dimensional triangular lattice structure between the graphite layers.

In the stage-2 GIC $C_{24}K$, the atomic density of potassium metal atoms intercalated between graphite layers is reduced to 2/3 of the density of the close-packed structure in the stage-1 compound, taking into account the difference in composition between the stage-1 compound

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[†] Corresponding author: f107143f@st.u-gakugei.ac.jp

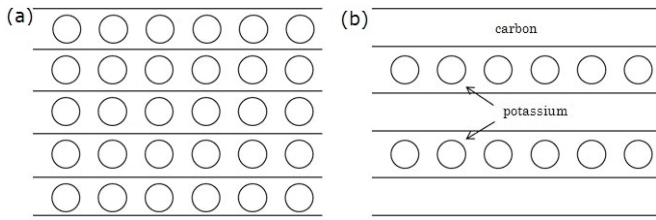


FIG. 1. The structure of the alkali-metal GIC's. (a) The stage-1 compound C_8K . (b) The stage-2 compound $C_{24}K$.

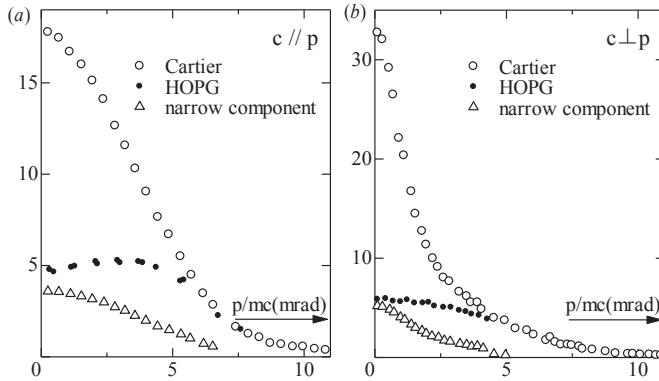


FIG. 2. $\circ$ is the positron annihilation angular correlation data in $C_{24}K$ [21], $\bullet$ is the positron annihilation angular correlation data in HOPG, $\triangle$ is the narrow component of the $\circ$.

C_8K and the stage-2 compound $C_{24}K$. According to the electronic structure model [16], there coexist quasi-two dimensional graphite π -bands, bands originating from potassium-metal s electrons, and the electronic interlayer state in the stage-2 GIC $C_{24}K$. The Fermi energy E_F and the location of the bands are determined by a balance between electronic and lattice energies. The electronic interlayer states were introduced by Posternak *et al.* [16]. These interlayer states, which exhibit free-electron character parallel to the layers, form a quasi-two dimensional band close to the Fermi energy and provide for alkali graphite intercalation compounds a new understanding of the origin of bands near the Fermi energy. Since the energy level of the interlayer state is close to the Fermi energy level, SU(2) pseudospin structure can be introduced by assigning “up” and “down” pseudospins to the electron on the interlayer state level and the Fermi energy level, respectively. A skyrmion is a topological soliton spread over two energy levels.

Now, we shall consider the quantum fluctuation in quasi (2+1) electron state's system. A charged skyrmion-like soliton in quasi (2+1) intercalate state in AGICs might be described as a topological soliton [22, 23]. “Topological quasi-positronium” is regarded as the skyrmion-like soliton, which traps a positron, of the electron-density. The electron number associated with the skyrmion-like soliton can be derived from the induced current J_{sky}^μ on the SU(2) pseudospin-like skyrmion configuration [25–28].

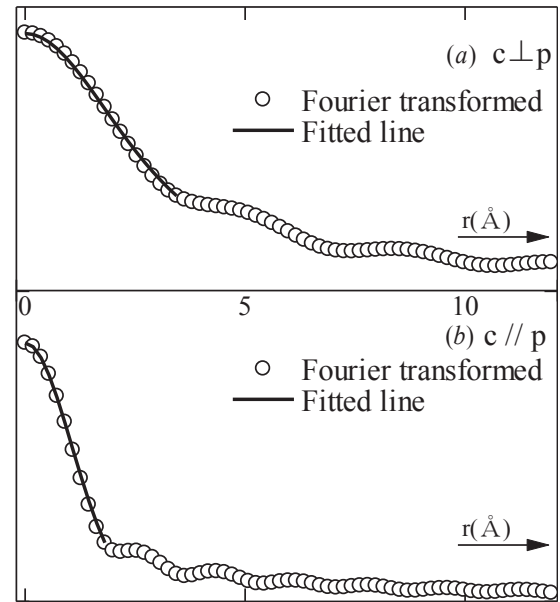


FIG. 3. $\circ$ is Fourier-transformed from the narrow component for pair-momentum p . The solid line is fitted one by (a) the function $J_{sky}^0(r)$, (b) Gaussian function $\propto \exp(-r^2/2\delta^2)$.

The electron number N_{sky} is

$$N = \int d^2x J_{sky}^\mu, \quad (1)$$

$$J_{sky}^\mu(r) = \frac{1}{\pi} \frac{4\xi^2}{[r^2 + 4\xi^2]^2}, \quad (2)$$

$$\text{where } r^2 = x^2 + y^2. \quad (3)$$

The electron-density distribution parallel to the quasi-two dimensional electron layer in the interlayer states in $C_{24}K$ can be represented by the SU(2) pseudospin-like skyrmion configuration.

Figure 2(a) shows the annihilation spectra for pair-momentum p perpendicular to the crystallographic c -axis in $C_{24}K$. The triangle in Figure 2(a) shows p -distribution of the narrow component ($p \perp c$), taking into account that p -distribution of the narrow component ($p \perp c$) of the annihilation spectra is isotropic approximately on the direction parallel to the graphite plane. Figure 2(b) shows the annihilation spectra for pair-momentum p parallel to the crystallographic c -axis in $C_{24}K$. The triangle in Figure 2(b) shows p -distribution of the narrow component ($p \parallel c$), assuming that p -distribution of the narrow component ($p \parallel c$) of the annihilation spectra is in elliptic circle approximately.

Figure 3(a) and (b) show the electron-density distribution perpendicular to the c -axis and parallel to the c -axis, which is Fourier-transformed from the narrow component for pair-momentum p perpendicular to the c -axis and parallel to the c -axis, respectively. The solid line in Figure 3(a) is fitted one by the function J_{sky}^0 in eq.(2). The value of ξ is estimated to be ~ 2.1 Å. The solid line in Figure 3(b) is fitted one by the Gaussian function $\propto \exp(-r^2/2\delta^2)$. The value of δ is estimated to be ~ 1.1 Å. As the origin of the anisotropic narrow component of ACPAR for AGICs, we can consider the contribution of “topological quasi-positronium”, from this

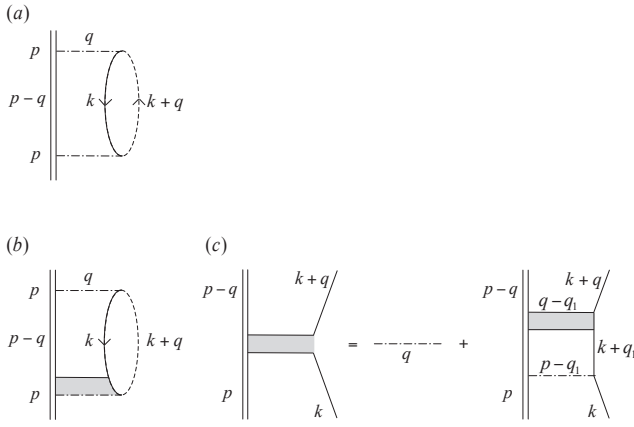


FIG. 4. A Feynman diagram of the positron proper self-energy within the random-phase approximation (RPA). A double solid line denotes a free positron Green's function. A single solid line denotes a hole Green's function. A single dotted-line denotes the electron Green's function. A dashed-dotted line denotes a bare positron-electron Coulomb interaction. (b) A Feynman diagram of the positron proper self-energy with the effective Coulomb interaction. (c) A Feynman diagram of the effective Coulomb interaction.

analysis. "Topological quasi-positronium" is regarded as the skyrmion-like soliton, which traps a positron, of the electron-density.

Now, we shall consider the positron energy loss due to the interaction between positrons and skyrmion-like electron density. The rate for the positron energy loss is defined by

$$\frac{dE_p^{\text{posi}}}{dt} = \int d^3q Wq(E_p^{\text{posi}} - E_{p-q}^{\text{posi}}). \quad (4)$$

E_p^{posi} is the positron energy with momentum p , in the case of low energies ($E_p^{\text{posi}} < \sim 2000$ eV). Wq is the transition probability. The transition probability Wq is related to the imaginary part of the positron self-energy $|\Sigma(p, E_p^{\text{posi}})|$ as follows.

$$\text{Im}\Sigma(p, E_p^{\text{posi}}) = \int \frac{d^3q}{2\pi^3} Wq. \quad (5)$$

This estimation has been made within the so-called random-phase approximation (RPA) as shown in Eq.(6). Figure 4(a), (b), (c) shows the approximate self-energy for low-energy positrons, where the dashed-dotted line represents the statically screened Coulomb potential. The

solid line shows the hole-propagator in π^* band in C_{24}K . The dashed line represents the electron-propagator in the skyrmion-like state, which is introduced in the present theoretical formula. In RPA approximation

$$\text{Im}\Sigma(p, E_p^{\text{posi}}) \sim 2 \int d^3q v_q^2 \text{Im} \sum_k \frac{f_k^{\pi^*} (1 - f_{k+q}^{\text{sky}})}{W - E_{k+q}^{\text{sky}} + E_k^{\pi^*}}, \quad (6)$$

$$\text{here } v_q = -\frac{4\pi e^2}{\varepsilon(q, 0)}. \quad (7)$$

$\varepsilon(q, 0)$ is the dielectric function due to π^* band. Since electron density of the Fermi energy in π^* band is low, it is suspected that the value of $\varepsilon(q, 0)$ is not large. But in the case of C_{24}K , the location of the interlayer state above E_F has not been established [16]. The energy level, E^{sky} , of the skyrmion-like state seems to be located in the energy region $\geq E_F$. Equation (6) shows a logarithmic diverging frequency $W \rightarrow 0$. That is, if $E^{\text{sky}} - E_F \sim W$ is very small, we might be able to observe the anomalous property of the positron energy loss in C_{24}K . When the effect of quasiparticle damping on the singularities is large, the singularities will not be observable.

To include the positron e^+e^- correlation in this regime we replace a single screened interaction by an energy-independent t matrix which sums the ladder diagrams shown in Figure 4(b). In this case,

$$Wq = 2v_q \sum_k t(p, k; p-q, k+q) f_k^{\pi^*} (1 - f_{k+q}^{\text{sky}}) \times \delta(w - E_p^{\text{posi}} + E_{p-q}^{\text{posi}}) \quad (8)$$

III. CONCLUSION

The static t matrix $t(p, k; p-q, k+q)$ is the scattering amplitude for a single e^+ of momentum p scattering from a single e^- with momentum k scattering to the state $p-q$ and $k+q$. In the case of C_{24}K , it is deduced that the interlayer state is located 0.2 eV above E_F [16].

The narrow component in the positron annihilation angular correlation spectra in the second stage graphite-potassium intercalation C_{24}K is analyzed from the viewpoint of the topological quasi-positronium model. We have considered the positron energy loss due to the interaction between positrons and skyrmion-like electron density within the random-phase approximation(RPA). The anomalous property of the positron energy loss in C_{24}K has been suggested.

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