Positron and Ion Migrations and the Attractive Interactions between like Ion Pairs in the Liquids: Based on Studies with Slow Positron Beam

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Abstract. We have discussed positron and ion diffusions in liquids by using the gauge-invariant effective Lagrange density with the spontaneously broken density (the hedgehog-like density) with the internal non-linear gauge fields (Yang-Mills gauge fields), and have presented the relation to the Hubbard-Onsager theory.

1. Introduction
The investigation for ion mobility in the liquid phase has been one of the central area of physical chemistry. The electrohydrodynamic theory by Hubbard and Onsager [1,2] and the stochastic theory [3,4] for ionic conductivity in the liquid phase have been proposed. Since fluctuation from the equilibrium medium is preferable in liquid phase, localization of ions such as positrons is highly probable [5,6]. Free energy density functional theories [7,8] provide self-trapping as solution of the ions in a given host liquid. The sensitivity of positrons to changes induced by melting has already been reported by the positron annihilation angular correlation [9-11] and lifetime [12,13] studies for liquids. Gramsh et al. [14,15] have observed very different behavior of the diffusion length \( L_+ \) of positrons in liquid and solid metals. That is, on melting, \( L_+ \) decreases remarkably, and in the liquid phase, \( L_+ \) increases with temperature. Kanazawa [16-20] has introduced the effective Lagrangian in the gauge-invariant formula with spontaneous symmetry breaking for liquids and glasses, and has discussed the origin of the boson peak, the glass transition, and the viscosity of the supercooled liquids. Kanazawa and coworkers [21-24] proposed a qualitative explanation for the increase of the positron diffusion length \( L_+ \) with temperature in the liquid phase, by using the theoretical formula, which is based on the gauge-invariant effective Lagrangian with spontaneously broken density (the hedgehog like fluctuation) and the massive internal gauge fields. In addition Kanazawa et al. [28] have suggested one origin of the attractive interaction between like ion pairs in liquids, extending the theoretical formula [20-23]. Recently Yamada et al. [29] have explained the origin of the attractive interaction between the \( Cl^- - Cl^- \) pair in water, which is introduced by the integral equation method [1], by using the theoretical formula [28]. In this study, we have analyzed the positron diffusion in the solid and liquid Pb, and have discussed the relation to the Hubbard-Onsager’s dielectric theory for positrons and ions migrations in liquids.
2. The hedgehog-like density fluctuation and the positron diffusion

We shall introduce a field-theoretical model to treat three-dimensional liquids by using excited cluster (solitons). The connection on gauge field describes how the orientation of the fiber changes as one goes along a path in base space, or how the path in the base space is lifted into a path in the fiber bundle. We introduce a field-theoretical model to treat the problem of a charged particle in a fluid host in three spatial dimensions. It has been proposed that the parameter \( \rho(t, r, u) = \rho(t, x, y, z, u) \) in three dimensional liquids is specified by the rotation, which is related to the non-linear gauge fields (Yang Mills gauge fields) \( A_\mu^a \) of SO(4) symmetry of \( S^3 \). The curvature can be represented by using a component, \( u \), in the other-axis direction, if the three spatial dimensional axes are \( x, y, \) and \( z \). It is preferable that we think of the anomalous fluctuation around the charged particle in the three dimensional liquid as the hedgehog-like fluctuation (soliton), taking into account the curvature. We adopt the parameter \( \rho(r, u) = \rho^a(a = 1, 2, 3, 4) \), which is similar to that in the Sachdev and Nelson model [25].

The SO(4) quadruplet fields, \( A_\mu^a \), are spontaneously broken through the Higgs mechanism similar to the way in which the fluid host is broken around a charged particle. When the hedgehog-like fluctuation (soliton) around a charged particle is created, we set the symmetry breaking of the quadruplet fields, \( \langle 0 | \rho | 0 \rangle \), equal to \( (0, 0, 0, \nu_1) \). Now, we can introduce the approximate Lagrangian as follows,

\[
L = \psi^+ (i\partial_0 - g_2 T^a A^a_\mu) \psi - \frac{1}{2m} \psi^+ (i\nabla - g_2 T^a A^a_\mu \neq 0)^2 \psi
- \frac{1}{4} (\partial_\mu A^a_\mu - \partial_\mu A^a_\mu + g_1 e^{a b c} A^b_\mu A^c_\mu)^2
+ \frac{1}{2} (\partial_\mu \rho^a - e_{a b c} A^b_\mu \rho^c)^2 + \frac{m_2^2}{2} (\langle A^a_\mu \rangle^2 + \langle A^a_\mu \rangle^2) + m_1 (A^1_\mu \partial_\mu \rho^2 - A^2_\mu \partial_\mu \rho^1) + m_1 (A^3_\mu \partial_\mu \rho^3 - A^3_\mu \partial_\mu \rho^3) + m_1 (A^3_\mu \partial_\mu \rho^1 - A^1_\mu \partial_\mu \rho^3)
+ g_1 m_1 \{ \rho^4 \langle A^1_\mu \rangle^2 + \langle A^2_\mu \rangle^2 + \langle A^3_\mu \rangle^2 \} - A^4_\mu (\rho^1 A^1_\mu + \rho^2 A^2_\mu + \rho^3 A^3_\mu)
- \frac{m_2^2}{2} (\rho^1)^2 - \frac{m_2 g}{2m_1} (\rho^2)^2 - \frac{m_2^2 g^2}{8m_1^2} (\rho^3)^2.
\]

(1)

where \( m_1 \) is \( \nu_1 g \) and \( m_2 \) is \( 2\sqrt{2} \lambda_3 \nu_1 \). The effective Lagrange density, \( L_{\text{eff}} \), represents three massive vector fields \( A^1_\mu, A^2_\mu, A^3_\mu \), and one massless vector field \( A^4_\mu \). The generation function \( Z[J] \) for Green’s functions is shown as follows;

\[
Z[J] = \int \mathcal{D}A \mathcal{D}B D\rho Dc D\bar{c} D\psi^+ D\psi \cdot \exp \left[ \int_0^\beta d\tau \int d^3x (\mathcal{L}_{\text{DF+FP}} + \mathcal{L}_{\text{GF+FP}} + J \cdot \Phi) \right],
\]

(2)

\[
\mathcal{L}_{\text{DF+FP}} = B^\alpha \partial^\mu A^a_\mu + \frac{1}{2} \alpha B^\mu B^a + i e^{a b c} \partial^\mu D^b c^c,
\]

(3)

where \( B^\alpha \) and \( c^a \) are the Nakanishi-Lautrup(NL) fields and Faddeev-Popov fictitious fields, respectively, and \( \beta = 1/T \), whose \( T \) is Temperature.

\[
J \cdot \Phi \equiv J^a_\mu A^a_\mu + J^B_\mu B^a + J^\rho \cdot \rho^a + J^\eta \psi + J^{\bar{\psi}} \bar{\psi} + J^c_\mu c^a + J^{\bar{c}}_\mu \bar{c}^a
\]

(4)

BRS-quartet \([26,27]\) in the present theoretical formula are \((\phi_1, B^1, C^1, \bar{C}^1), (\phi_2, B^2, C^2, \bar{C}^2), (\phi_3, B^3, C^3, \bar{C}^3), (A^4_{\mu \mu}, B^4, C^4, \bar{C}^4)\) where \( A^4_{\mu \mu} \) is the longitudinal component of \( A^4_\mu \). So we
need these fields are unobservable and fictitious ones. Since these masses are created through the Higgs mechanism by introducing a charged particle in the fluid host, the massive gauge fields $A_1^\mu, A_2^\mu, A_3^\mu$ are localized around the charged particle. In metallic liquids, $A_4^\mu$ also becomes massive due to screening effect of conduction electrons.

Kanazawa [21,22] has proposed the explanation for the increase of the positron diffusion length with temperature in liquid metals. That is, as the temperature increases, the effective mass of the positron decreases due to the restoration of the spontaneously broken density around the positrons in the liquid phase. The diffusivity $D_+$ of positron is introduced approximately from the previous theoretical formula[21,22],

$$D_+ = \frac{2}{3} \frac{k_B T}{M_{\text{eff}}} \tau_r \propto \frac{k_B T \cdot \tau_r}{\left[ 4 c_3^2 - \left( \frac{1}{3} \lambda_3 + \frac{g^2}{2} \right) T^2 \right]^\frac{1}{2}}$$

(5)

Where $\tau_r$ is the relaxation time, for example, the phonon relaxation time is $\tau_{ph} \sim T^3$. Restoration of the spontaneously broken density reduces the effective mass $M_{\text{eff}}$ as temperature increases as shown in Figure 1. From eq.(5), it is seen that the diffusivity $D_+$ increases with temperature. From Fig.2, it is seen that eq.(5) is consistent with experimental results [15].

![Figure 1](image)

**Figure 1.** Intuitive picture of the reduction of the positron effective mass due to restoration of the spontaneously broken density around positron, with increase in temperature.

Now, we recall that the electro-hydrodynamic stress tensor $S_{el}$. is given in the Hubbard-Onsager theory [3,4] as $8 \pi S_{el} = DE + ED - E \cdot D I$, where $I$ is the unit tensor and $D = E + 4 \pi P$. In the presence of a uniform periodic external field $E_{ext}$, the induction $D$, and the total electric field are given by

$$D = \epsilon(\omega) E_{ext} + \epsilon(\omega = 0) E_0$$

$$E = E_{ext} + E_0,$$

where $\epsilon$ is the dielectric constant, $E_0$ is the field due to an ion field in a dielectric continuum. In the present theoretical formula, $E_0 \propto -\partial_t A_{\mu}(\mu \neq 0) - \partial_{\mu}(\mu \neq 0) A_0^\mu$. $A_0^\mu$ is the massless gauge field. The divergence of electrical stress $S_{el}$ defines a local force density $f$ which we write as

$$f = \frac{1}{2} \left\{ \frac{\epsilon(\omega) - \epsilon(\omega = 0)}{4 \pi} \right\} \left[ D \times (E_{\text{exp}} \times E_0) + \nabla (E_0 \cdot E_{ext}) \right].$$

(6)
3. Conclusion
We have discussed positron and ion diffusions in liquids, by using the gauge-invariant effective Lagrange density with the spontaneously broken density and internal Yang-Mills gauge fields. In addition, the relation to the Hubbard-Onsager theory has been discussed.

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