

## Positron diffusion and fluctuations around positrons in liquid metals

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We have introduced effective Lagrangian with spontaneously broken density (the hedgehog-like fluctuation) and the massive internal gauge fields. It is suggested that the temperature dependence of the positron diffusion length in the liquid In and Bi metals is described with the restoration of the spontaneously broken density around the positrons due to increasing of temperature.

Key words: positron diffusion, liquid metals, amorphous, fluctuation, self-trapping

### 1. INTRODUCTION

The study of liquid and amorphous structures is of significance for scientific and technological reasons. However, our knowledge of the liquid and amorphous structures is much less than our knowledge of the crystalline structures. This is due to the fewer techniques available to study liquids and amorphous materials. The dynamical properties of charged particles in liquids are of importance for understanding chemical reaction in fluids.

Since fluctuation from the equilibrium medium is preferable in the liquid phase, localization of charged particles such as positrons is highly probable [1,2]. Free energy density functional theories [3,4] provide self-trapping as a solution of the charged particle in a given host fluid. It is well known that positrons are strongly sensitive to vacancies and other open-volume defects in materials [5]. Because of the irregular arrangement of ions, a liquid can have regions of low ion density similar to vacancies or voids in solid materials. Thus positron can be an ideal probe for the structure and dynamics of these regions in the liquid phase. The sensitivity of positrons to changes caused by melting has already been reported by angular correlation [6-8] and lifetime [9,10] studies. Gramsh et al. [11,12] have measured temperature dependence of diffusivity of positrons in Ga, Bi, In, Na, Sn, and Pb in both solid and liquid states by means of a slow positron beam. They observed quite different behaviors of the diffusion length,  $L_+$ , of positrons in solids and liquids. On melting,  $L_+$  decreased remarkably, while it increased with temperature in the liquid phase. These results suggest strongly that a different mechanism is involved in the interaction of positrons with the liquid. Seeger [13] has proposed that the interaction of the positron in a liquid state is via two polaronic states, with large effective masses that depend on the materials.

Kanazawa [14,15] has presented a qualitative explanation for the increase with temperature of the positron diffusion length in liquid metals. In this model, as the temperature increases, the effective mass of the positron decreases due to the restoration of the spontaneously broken density around the positron. In this study, we will compare the positron diffusion length of In metal, which has vacancy trapping effect of positrons in solid state, with that of Bi metal, which has

no positron-trapping effect into vacancies in solid phase, from the view point of the theoretical formula [15].

### 2. GAUGE THEORY OF DISORDERED MATERIALS AND A MODEL SYSTEM

Liquids or amorphous substances are characterized by spatial fluctuations of the internal parameters (the local configuration, the curvature, etc.) of the constituting elements (matter field). Also, these substances are organized in space (they fill space with short-range order) so that a local transformation of an element should necessarily affect its immediate neighbors through the connection or gauge field  $A_\mu$ . Gauge invariant is the symmetry of disorder. Liquids and glasses are disordered substances, and can be represented as a fiber bundle [16], because a gauge field is, geometrically, a connection in a fiber bundle. A fiber bundle consists of a base space, a total space, and a map that projects every point of the total space onto a point in the base space. The set of all points in the total space that are mapped onto the same point in the base space is called the fiber. The connection, or 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 or the fiber bundle. In liquids and glasses, this connection (gauge field),  $A_\mu$ , is a natural and physical one. If internal parameters (matter field)  $\rho(x, y, z, u)$  are given everywhere in a continuum (the Euclidean space  $R^3$ ), this defines a mapping between  $R^3$  (base space) and manifold  $M$  of states of the internal parameter (the fiber of bundle).

$$\rho(x, y, z, u); \quad R^3 \rightarrow M$$

There is growing evidence that the short-range order of metallic liquids and glasses is predominantly icosahedral [17]. Long-range correlations in the orientations of icosahedral packing units appear in large "amorphon" cluster models of structure in metallic liquids and glasses [18].

We shall introduce icosahedral order in three-dimensional liquids. Due to the impossibility of tiling flat space with icosahedral in the base space in three dimensions, one looks for ideal translation on the surface  $S^3$  of a sphere in the total space in four dimensions [19,20]. The symmetries of the sphere  $S^3$

form the rotation group SO(4).

Extending this theoretical formula, Kanazawa has introduced the effective Lagrangian in the gauge-invariant formula with spontaneous symmetry breaking for two dimensional and three dimensional metallic liquid and glasses, and has discussed the origin of the boson peak, the melting, the glass transition, and the viscosity of the supercooled metallic liquids [21-26].

Now, 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 gauge fields  $A_\mu$  of SO(4) symmetry of  $S^3$  [19,20]. 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 curvature. We adopt the parameter  $\rho(t, r, u) \equiv \rho^a$  ( $a = 1, 2, 3, 4$ ), which is similar to that in the Sachdev and Nelson model [27]. The SO(4) quadruplet fields  $A_\mu$  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  $(0, 0, 0, v_4)$ .  $\psi$  is the wavefunction of a positron. It is known that Lagrangian formation is more transparent in order to treat the quantized Yang-Mills fields [30]. Now, we can introduce the approximate Lagrangian as follows,

$$\begin{aligned} L = & \psi^\dagger (i\partial_0 - g_2 T^a A_0^a) \psi - \frac{1}{2m} \psi^\dagger (i\nabla - g_2 T^a A_{\mu \neq 0}^a)^2 \psi \\ & - \frac{1}{4} (\partial_\nu A_\mu^a - \partial_\mu A_\nu^a + g_1 e_{abc} A_\mu^b A_\nu^c)^2 \\ & + \frac{1}{2} (\partial_\mu \rho^\beta - g e_{\beta\alpha\gamma} A_\mu^\alpha \rho^\gamma)^2 + c_3^2 \rho^a \rho^a - \lambda_3 (\rho^a \rho^a)^2 \end{aligned} \quad (1)$$

The first and second terms describe the gauge-invariant interaction between the gauge fields  $A_\mu$  and  $\psi$  Fermi field of the charged particle through covariant derivatives. The third term describes the standard Lagrangian of the Yang-Mills gauge fields  $A_\mu$ . The fourth term describes the gauge-invariant interaction between the gauge fields  $A_\mu$  and the Bose fields  $\rho^a$  through covariant derivatives. The fifth and sixth terms describe the effective potential for the Higgs effect. Then, we set the symmetry breaking as follows,

$$\rho^a \rightarrow (0, 0, 0, v_4) + (\rho^1, \rho^2, \rho^3, \rho^4).$$

Thus, we can introduce the effective Lagrange density,

$$\begin{aligned} L = & \psi^\dagger (i\partial_0 - g_2 T^a A_0^a) \psi - \frac{1}{2m} \psi^\dagger (i\nabla - g_2 T^a A_{\mu \neq 0}^a)^2 \psi \\ & - \frac{1}{4} (\partial_\nu A_\mu^a - \partial_\mu A_\nu^a + g_1 e_{abc} A_\mu^b A_\nu^c)^2 \\ & + \frac{1}{2} (\partial_\mu \rho^\beta - e_{\beta\alpha\gamma} A_\mu^\alpha \rho^\gamma)^2 \\ & + \frac{m_1^2}{2} [(A_\mu^1)^2 + (A_\mu^2)^2 + (A_\mu^3)^2] \\ & + m_1 (A_\mu^1 \partial_\mu \rho^2 - A_\mu^2 \partial_\mu \rho^1) + m_1 (A_\mu^2 \partial_\mu \rho^3 - A_\mu^3 \partial_\mu \rho^2) \end{aligned}$$

$$\begin{aligned} & + m_1 (A_\mu^3 \partial_\mu \rho^1 - A_\mu^1 \partial_\mu \rho^3) \\ & + g_1 m_1 \{ \rho^4 [(A_\mu^1)^2 + (A_\mu^2)^2 + (A_\mu^3)^2] \\ & - A_\mu^4 (\rho^1 A_\mu^1 + \rho^2 A_\mu^2 + \rho^3 A_\mu^3) \} \\ & - \frac{m_2^2}{2} (\rho^4)^2 - \frac{m_2 g}{2m_1} \rho^4 (\rho^a)^2 - \frac{m_2^2 g^2}{8m_1^2} (\rho^a \rho^a)^2 \end{aligned} \quad (2)$$

Where  $m_1$  is  $v_4 \cdot g$  and  $m_2$  is  $2 \cdot (2\lambda_3)^{1/2} v_4$ . The effective Lagrange density,  $L_{\text{eff}}$ , represents three massive vector fields  $A_\mu^1, A_\mu^2, A_\mu^3$ , and one massless vector field  $A_\mu^4$ . The generation function  $Z[J]$  for Green's functions is shown as follows;

$$\begin{aligned} Z[J] \sim & \int DADBD\rho Dc D\bar{c} D\psi^\dagger D\psi \times \\ & \exp i \int d^4x (L_{\text{eff}} + L_{GF+FP} + J \cdot \Phi) \\ L_{GF+FP} = & B^a \partial^\mu A_\mu^a + \frac{1}{2} \alpha B^a B^a + i c^a \partial^\mu D_\mu c^a \end{aligned} \quad (3)$$

where  $B^a$  and  $c^a$  are the Nakanishi-Lautrup (NL) fields and Faddeev-Popov fictitious fields, respectively,

$$\begin{aligned} J \cdot \Phi \equiv & J^{a\mu} A_\mu^a + J_B^a B^a + J_\rho \cdot \rho^a + \bar{\eta} \psi + \eta \psi^\dagger \\ & + \bar{J}_c c^a + J_c \bar{c}^a \end{aligned} \quad (4)$$

BRS-quartet [28,29] in the present theoretical system are  $(\rho^1, B^1, c^1, \bar{c}^1)$ ,  $(\rho^2, B^2, c^2, \bar{c}^2)$ ,  $(\rho^3, B^3, c^3, \bar{c}^3)$ ,  $(A_{L,\mu}^4, B^4, c^4, \bar{c}^4)$

where  $A_{L,\mu}^4$  is the longitudinal component of  $A_\mu^4$ . So we need these fields for the unitarity condition, although these fields are unobservable and fictitious ones. The masses of massive gauge fields  $A_\mu^1, A_\mu^2$ , and  $A_\mu^3$  are created through the Higgs mechanism through introducing the hedgehog-like clusters (solitons) induced with a charged particles such as positrons on the fluid host. Thus, the massive gauge fields  $A_\mu^1, A_\mu^2$ , and  $A_\mu^3$  are localized around the hedgehog-like cluster, where a positron is trapped. It is known that the effective masses of positron in liquid Ga and Bi become of the order of the proton mass [13]. It is suggested that the spontaneously broken density (the hedgehog-like fluctuation) and the massive gauge fields, which are induced with the positron, contribute strongly to the remarkable increase in the positron effective mass in the fluid host. Thus, the positron, and the spontaneously broken density (the hedgehog-like fluctuation) and the massive gauge fields, which are induced with the positron in liquid metals, might be thought of as a kind of complex particle.

The thermodynamic potential is derived from the partition function. That is,  $\Omega(T, v_4) \equiv -T \ln Z/V$ . In the mean field and high temperature approximations, the thermodynamic potential can be introduced as follows,

$$\begin{aligned} \Omega(T, v_4) = & \lambda_3 v_4^4 + [(\frac{\lambda_3}{3} + \frac{g^2}{8}) T^2 - c_3^2] v_4^2 \\ & - \frac{8}{90} \pi^2 T^4 - (\frac{c_3^2}{6} T^2 + \frac{g^2 \cdot c_3^2}{16\lambda_3} T^2) \end{aligned} \quad (5)$$

From Eq. (5) it is seen that the thermodynamic potential is a function of  $v_4$  and temperature  $T$ . As temperature increases, the minimum of the thermodynamic potential shifts to smaller values of  $v_4$ , and the minimum becomes less deep. The location of the minimum is

$$v_{4,\min} = \frac{c_3^2}{2\lambda_3} - \left(\frac{1}{6} + \frac{g^2}{16\lambda_3}\right)T^2$$

Although in three spatial dimensions we cannot determine quantitatively the mass  $M_{\text{eff}}$  of the positron in the field host in the present theory, it is assumed that the effective mass  $M_{\text{eff}}$  depends approximately linearly on the sum of the effective mass  $m_2$  of the excitation  $\rho^4$  and the massive gauge fields  $A_\mu^1$ ,  $A_\mu^2$ , and  $A_\mu^3$  which are localized around a positron in the fluid host. We can evaluate the effective masses  $m_1$  and  $m_2$  from Eq. (2).

$$m_1 = \left[ \frac{g_1^2 c_3^2}{2\lambda_3} - \left( \frac{g^2}{6} + \frac{g^4}{16\lambda_3} \right) T^2 \right]^{\frac{1}{2}}$$

$$m_2 = \left[ 4c_3^2 - \left( \frac{4}{3}\lambda_3 + \frac{g^2}{2} \right) T^2 \right]^{\frac{1}{2}}$$

In order to simplify the discussion, we shall consider only the mass of  $\rho^4$ . The diffusivity  $D_+$  of positron is introduced approximately from the Einstein relation,

$$D_+ = \frac{2 k_B T}{3 M_{\text{eff}}} \tau \propto \frac{k_B T \cdot \tau}{\left[ 4c_3^2 - \left( \frac{4}{3}\lambda_3 + \frac{g^2}{2} \right) T^2 \right]^{\frac{1}{2}}} \quad (6)$$

where  $\tau$  is the relaxation time, for example, the phonon relaxation time is  $\tau_{ph} \sim T^{-3/2}$ .

Restoration of the spontaneously broken density reduces the effective mass  $M_{\text{eff}}$  as temperature increases. From Eq. (6), it is seen that the diffusivity  $D_+$  increases with temperature.

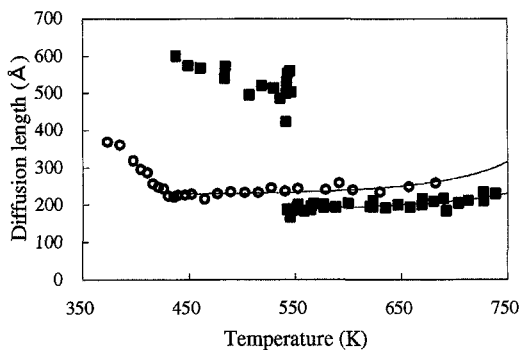


Fig. 1 Positron diffusion length versus temperature in the solid and liquid In and Bi metals. The open circles and solid squares are data points [12] in In and Bi metals, respectively.

We have analyzed the positron diffusion length data [12] in In and Bi metals. Figure 1 shows positron diffusion length versus temperature in the solid and liquid In and Bi metals. Open circles and solid squares correspond to data points of In and Bi metals, respectively. The solid lines show the fitting ones by using Eq. (6). It is suggested that the temperature dependence of the diffusion length for both liquid In and Bi metals is described well with the restoration of the spontaneously broken density due to temperature. The interesting point is that the diffusion length drops discontinuously at the melting temperature in Bi metal and on the other hand the diffusion length in In metal decreases gradually from  $\sim 300\text{K}$  to the melting temperature. In the case of solid In metal, more positrons are trapped into thermal vacancies as temperature increases. Thus the diffusion length in solid

In metal decreases gradually from  $\sim 300\text{K}$  to the melting temperature. This suggests that the spontaneously broken density state in the liquid phase of In metal near the melting point seems to be vacancy-like.

### 3. CONCLUSION

We have analyzed the positron diffusion length data of the liquid In and Bi metals, taking into account the restoration of the spontaneously broken density around the positron due to increasing of temperature. Then we have compared the positron diffusion length of In metal with that of Bi metal.

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