

NEGATIVE IONS IN LIQUID XENON

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The structure and mobility of negative ions of oxygen in liquid Xe is investigated. It is shown that the strong exchange interaction of the outer, weakly bound electron of the negative ion with the surrounding liquid leads to a partial compensation of the electrostriction effect, and it prevents the formation of a solid cluster around the negative ion. A simple perturbative model describing the structure of the negative ion in the liquid matrix is developed. The mobility of O_2^- ions in liquid Xe on the saturation line is estimated. The reasons for the difference in mobility of negative and positive ions are discussed.

1. INTRODUCTION

In liquefied rare gases, the structure and properties of admixed ions are considered, as a rule, in the context of Atkins' model [1]. This model takes into consideration the effect of electrostriction, and it predicts the formation of a solid cluster around the ion. The cluster then determines the transport properties of such an ion in the condensed phase. The consequences of this theory are in good agreement with experiments on the mobility of positive ions. However, Atkins' theory does not depend on the sign of the ionic charge, and it predicts the same effects for positive and negative ions.

Recently, experimental data on the mobility of certain negative and positive ions in liquid xenon became available which indicate a difference in the transport properties of positive and negative ions. It was found that negative ions exhibit higher mobility than positive ions of comparable size [2, 3].

The difference in transport properties of positive and negative ions is, above all, due to the fact that the outer electron of the negative ion is localized in a spatial region with a characteristic size appreciably greater than that of the inner electron shells of the parent atom or molecule of the ion. When a negative ion is located in a dense gas or liquid it perturbs the surrounding medium. The character of the perturbation depends on the character of interaction of the outer electron with the medium. One important factor that determines the qualitative difference in the perturbation near positive and negative ions is the repulsive exchange interaction of the weakly bound outer electron of the negative ion with the electron shells of the surrounding atoms or molecules.

The structure of negative ions in dense gases and liquids with atoms of low polarizability (He and Ne) were investigated recently [4, 5]. It was shown that the competition between the short-range exchange repulsion and the long-range polarization attraction of the outer electron with the atoms of the medium leads to the formation of a microcavity around the negative ion,

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which is surrounded by a denser layer of atoms. This effect largely determines the negative ion mobility. Similar complexes, in which a light quantum particle is localized inside the liquid density fluctuation and kept stationary, were first suggested and investigated by Ferrell [6] to explain the anomalously long life-time of positronium in liquid helium due to creation of a cavity («bubble») around the positronium. Bubbles in nonpolar liquids can also be created around free electrons, excited atoms, and positive and negative ions. Detailed discussions of this problem can be found in the special issue «Ions and Atom in Superfluid Helium», *Zeitschrift für Physik B, Cond. Matter* **98**, No 3 (1995).

A difference in the mobilities of positive and negative ions was also observed in liquefied rare gases with high polarizability. For example, in liquid xenon, negative ions of O_2 and SF_6 exhibit mobility several times that of positive ions [2]. As a result of the high polarizability of atoms of the medium, no microcavity is created in this case, but the strong exchange interaction leads to partial cancellation of the electrostriction and it prevents the formation of a solid cluster around the negative ion. The determination of the structure of the negative ion becomes complex because of the more complicated nature of the interaction of the outer electron of the ion with the liquid.

In this paper, we propose a simplified model of the ionic structure that considers the influence of the weakly bound electron of the negative ion on the liquid matrix. From these results, we estimate the mobility of O_2^- in liquid Xe by means of a modified Stokes equation.

2. SELF-CONSISTENT FIELD MODEL OF A NEGATIVE ION IN A NONPOLAR LIQUID

We consider a lone electron bound by the polarization potential to the molecule that formed the negative ion. The asymptotic form of the electron wave function, $\psi(r)$, in the polarization potential of the molecules is very well known. At large distances r , the wave function varies as $\psi(r) \propto r^{-1} \exp(-r/\lambda)$. The characteristic size λ of the spatial region where the electron is localized is determined by its ground state energy ϵ in the negative ion [7], $\lambda = \hbar/\sqrt{2m\epsilon}$.

In the following, we consider weakly bound electrons for which λ is appreciably greater than the size of the parent molecule. The weakly bound electron spends a significant time far from the molecule. It moves with an average kinetic energy $\epsilon_k \propto \hbar^2/2m\lambda^2$. The motion of such an electron has many features in common with the motion of a quasifree electron of the same energy. In rarefied gases this interaction results in scattering of the electron by isolated molecules. The scattering potential consists of a short-range repulsive part due to exchange interaction and an attractive long-range polarization part.

In a liquid, the situation is significantly more complicated. Here, the long-range part of the polarization potential is determined by the atomic polarization potentials of the atoms around the ion. In this case the potential of a scatterer can be represented by a superposition of the electron-atom interaction potential and the screened polarization potentials of all other atoms around the ion. The total scattering potential of an electron is described as a «muffin-tin» potential [8].

It is worthwhile to note that the potential decays faster than $1/r^4$ as a result of the screened polarization. This leads to a reduction in the characteristic size of the interaction region. In rare gases, in the supercritical region at liquid densities and on the saturation line in liquids, the elastic electron-atom cross-section depends weakly on electron energy and scattering angle. There is essentially no Ramsauer-Townsend effect, and the scattering cross-section of a slow electron is determined by an effective scattering length $L_{eff}(N)$.

The electron effective scattering length is a function of the liquid density N . It can be determined from experimental data on the quasifree electron mobility [9]. In the low-density limit, the effective scattering potential is transformed into the potential of an isolated atom, and $L_{eff}(N)$ becomes the electron scattering length of an isolated atom

$$L_a = \sqrt{\frac{\alpha}{a_0}} \operatorname{ctg} \sqrt{\frac{\alpha}{a_0 R_a^2}},$$

where α is the atomic polarizability, a_0 is the Bohr radius, and R_a denotes the effective radius of the short-range hard core of the electron-atom interaction potential [10]. With increasing liquid density, the relative role of attraction in the scattering process decreases as a result of polarization screening. This leads to a decrease in scattering length, and even to a sign reversal at sufficiently high densities.

In the optical approximation [7, 8], the energy of the bottom of the electron conduction band is $V_0 = 2\pi\hbar^2 N L_{eff}(N)/m + U_p(N)$, where $U_p = -\alpha e^2 F_L N/2\sigma$ is the shift in electron energy as a result of the screened polarization interaction. The Lorentz screening factor [8] $F_L(N) = [1 + 8\pi\alpha N/3]^{-1}$ takes into account the weakening of the polarization field of a particular atom by its interaction with the induced dipoles of all other atoms of the medium; σ is the radius of the first peak of the pairwise correlation function of the liquid.

We model the motion of the weakly bound electron of a negative ion in a nonpolar liquid as the motion of the electron in the field of point scatterers with an effective potential $V_{eff} = 2\pi\hbar^2 L_{eff}(N)\delta(\mathbf{r} - \mathbf{R}_i)/m$, where $\delta(\mathbf{r})$ is the Riemann delta function, \mathbf{r} and \mathbf{R}_i are the coordinates of the electron and scattering center, respectively. We also assume that the conditions for the optical model are satisfied, i.e.,

$$L_{eff}(N) \ll \lambda, \quad N L_{eff}^3(N) \ll 1. \quad (1)$$

The Schrödinger equation for the wave function of a weakly bound electron in the liquid acquires the form

$$-\frac{\hbar^2}{2m} \Delta\psi(\mathbf{r}) + U(\mathbf{r})\psi(\mathbf{r}) + \sum_i \frac{2\pi\hbar^2}{m} L_{eff}(N)\delta(\mathbf{r} - \mathbf{R}_i)\psi(\mathbf{r}) = [E - U_p(N)] \psi(\mathbf{r}), \quad (2)$$

where E is the electron ground state energy of the negative ion in the liquid and $U(\mathbf{r})$ is the potential of the molecule on which the electron is localized. The shift of the electron energy $U_p(N)$ was taken into account in Eq. (2). The electron energy is then

$$\varepsilon = E - U_p(N).$$

We assume that the effect of the medium on ε is small,

$$\varepsilon \gg \Delta\varepsilon = \left\langle \psi(\mathbf{r}) \left| \sum_j \frac{2\pi\hbar^2}{m} L_{eff}(N)\delta(\mathbf{r} - \mathbf{R}_j) \right| \psi(\mathbf{r}) \right\rangle. \quad (3)$$

First-order perturbation theory supports this notion [7]. Integrating Eq. (3) over the electron coordinate and averaging over the atomic coordinates, we obtain the following relationship between the de Broglie wavelength of the electron λ , the electron effective scattering length in the medium $L_{eff}(N)$, and the number density N of the liquid:

$$\frac{\lambda}{L_{eff}(N)} \gg 4\pi\lambda^3 N \exp\left(-\frac{2\sigma}{\lambda}\right). \quad (4)$$

This places an additional constraint on the electron binding energy of the ion. In fact, when $\lambda \gg \sigma$, the right-hand side of Eq. (4) is proportional to λ^3 , but its left-hand side is linear in λ . At very large λ , the number of atoms of the medium $N\lambda^3$ in the vicinity of the electron orbit can be large. In this case, the interaction of the electron with the medium, as determined by its scattering length, cannot be considered weak. The contribution of the medium to the electron binding energy is significant, and inequality Eq. (4) is violated. In conjunction with Eq. (1), Eq. (4) determines the range of applicability of the current approximation.

We now turn to a discussion of the free energy functional $\Delta F\{N(\mathbf{r}), \psi(\mathbf{r})\}$ of the negative ion-liquid system. The presence of the negative ion in the liquid leads to a change in the local structure of the liquid near the ion. In other words, the perturbation of the liquid affects the electron spectrum of the negative ion. The structure of the complex is determined by the minimum free energy, which can be represented as a sum of two terms,

$$\Delta F\{N(\mathbf{r}), \psi(\mathbf{r})\} = \Delta E\{N(\mathbf{r}), \psi(\mathbf{r})\} + \Delta F_l\{N(\mathbf{r})\}.$$

The first represents the change in electron ground-state energy due to the presence of the liquid. An accurate calculation of ΔE is complicated, and requires a self-consistent calculation of the local liquid density $N(\mathbf{r})$ and the electron wave function $\psi(\mathbf{r})$. If the conditions of the perturbation theory are satisfied, however, the problem can be simplified significantly, and ΔE can be written

$$\Delta E\{\psi(\mathbf{r}), N(\mathbf{r})\} \simeq \left\langle \psi(\mathbf{r}) \left| \sum_j \frac{2\pi\hbar^2}{m} L_{eff}(N)\delta(\mathbf{r} - \mathbf{R}_j) \right| \psi(\mathbf{r}) \right\rangle + U_p\{N(\mathbf{r})\}. \quad (5)$$

The second term in the free energy functional $\Delta F_l\{N(\mathbf{r})\}$ represents the change in free energy of the liquid as a result of a change in its local density. The relation between the optimum density profile $N(\mathbf{r})$ and the electron wave function $\psi(\mathbf{r})$ is determined by minimizing the free energy, $\delta\Delta F\{N(\mathbf{r}), \psi(\mathbf{r})\}/\delta N(\mathbf{r}) = 0$. A knowledge of $N\{\psi(\mathbf{r})\}$ enables one to determine the potential energy of electron interaction with atoms of medium. This leads to the nonlinear Schrödinger equation. Its solution, which can be obtained numerically, is usually very complicated, and we consider only the simplest case of small changes in the local density $|N(\mathbf{r}) - N|/N \ll 1$.

In this case, the relation between $N(\mathbf{r})$ and $\psi(\mathbf{r})$ becomes very simple:

$$N(\mathbf{r})/N = \exp\left[-\gamma\tilde{V}(\mathbf{r})/Ms^2\right], \quad (6)$$

where M is the atomic mass, s is the velocity of sound in the liquid, and $\gamma = C_p(T)/C_V(T)$ is the ratio of specific heats of the liquid. The function $\tilde{V}(\mathbf{r}) = \tilde{V}_1(\mathbf{r}) + \tilde{V}_2(\mathbf{r})$ represents the effective interaction potential between the electron and liquid. In first-order perturbation theory, the first term has the form

$$\tilde{V}_1(\mathbf{r}) = \left\langle \psi(\mathbf{r}') \left| \frac{2\pi\hbar^2}{m} L_{eff}(N)\delta(\mathbf{r} - \mathbf{r}') \right| \psi(\mathbf{r}') \right\rangle = \frac{2\pi\hbar^2}{m} L_{eff}(N) |\psi(\mathbf{r})|^2. \quad (7)$$

This takes electron scattering by the effective potential into consideration. The second term in $\tilde{V}(\mathbf{r})$ takes into account the fact that when the electron is near a scattering center, it interacts with the screening potentials of the other atoms of the liquid. These contribute to $U_p(\mathbf{r})$, but they do not participate in the scattering process:

$$\tilde{V}_2(\mathbf{r}) = \langle \psi(\mathbf{r}') | V_p(\mathbf{r} - \mathbf{r}') g(\mathbf{r} - \mathbf{r}') F_l(N) | \psi(\mathbf{r}') \rangle. \quad (8)$$

Here, $g(r - r')$ is the pairwise correlation function of the atoms comprising the liquid. In liquids made up of atoms with small polarizability, where the atomic polarization potentials are essentially unscreened, $\tilde{V}_2(r)$ is negligible in comparison with $\tilde{V}_1(r)$. Note that in the limit $r \gg \sigma$, $\tilde{V}_2(r)$ takes the natural form of the screening polarization interaction: $\tilde{V}_2(r) = -\alpha e^2 F_1(N)/2r^4$.

3. RESULTS AND DISCUSSION

The present model was used to investigate the structure of the negative ion of the oxygen molecule, O_2^- , in liquid Xe on the saturation line, with the aim of comparing calculated and experimental results [2]. Interest in the O_2^- ion stems from the presence of this ion as an admixture in essentially all experiments on electron and ion mobilities in liquefied rare gases. Furthermore, experimental data on the mobility of O_2^- in dense gases and liquids are available. The binding energy of the electron in O_2^- in vacuum (electron affinity) is $\varepsilon \simeq 0.46$ eV, which corresponds to $\lambda \simeq 5.5a_0$. In the atomic density range considered here, the electron-atom scattering cross-section depends weakly on the electron energy, and there is no Ramsauer-Townsend effect [11]. Therefore, we assume for simplicity that the cross-section is independent of energy, and that it is determined by the effective scattering length $L_{eff}(N)$. The applicability condition for perturbation theory is then satisfied for O_2^- in liquid Xe. The effect of the medium on the weakly bound electron of the negative ion O_2^- at the triple point results in a correction to the electron ground-state energy of $\Delta\varepsilon \simeq 0.05$ eV, which is significantly less than the electron affinity of O_2 in vacuum.

Liquid density profiles were calculated at the triple point of Xe, at $T = 161.4$ K, $N = 1.36 \cdot 10^{22} \text{ cm}^{-3}$, and on the saturation curve at $T = 200$ K, $N = 1.23 \cdot 10^{22} \text{ cm}^{-3}$. For these conditions we adopted effective electron scattering lengths $L_{eff}(N = 1.36 \cdot 10^{22} \text{ cm}^{-3}) = 0.6a_0$ and $L_{eff}(N = 1.23 \cdot 10^{22} \text{ cm}^{-3}) = 0.3a_0$. Results of the calculations are shown in Fig. 1. In the same figure, density profiles calculated by means of Atkins' model are shown. Far from the

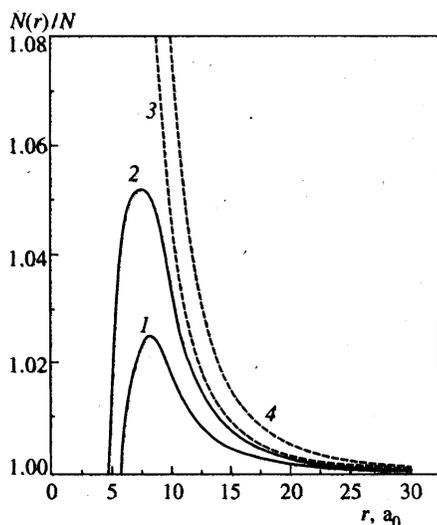


Fig. 1. Profiles of local density as a function of the distance to the center of the O_2^- ion in Xe on the saturation curve. The solid curves were calculated in accordance with (6) for different temperatures: 1) 161.4, 2) 200 K. The dashed curves were calculated in accordance with Atkins' model for the same temperatures: 3) 161.4, 4) 200 K

ion, the long-range polarization part of the interaction potential plays the main role. Closer to the center of the ion, the exponentially increasing term (see Eq. (7)), which takes the exchange interaction into consideration, becomes dominant. In liquid Xe, in spite of its high polarizability $\alpha = 27.11$ a.u., the exchange interaction is so strong that as the liquid density increases, $L_{eff}(N)$ reverses sign from negative to positive at $N^* \simeq 1.1 \cdot 10^{22} \text{ cm}^{-3}$. The increasing importance of $\bar{V}_1(r)$ results in a slower rise in the local density near the center of the ion. The density profiles $N(r)$, shown in Fig. 1 for small distances of the order of the radius of the first coordination sphere, cannot be correct, since the applicability of the present model would be violated. But we speculate that an accurate calculation would not change the qualitative picture, because the probability of finding an electron in this region is low.

As can be seen from Fig. 1, the assumption of small changes in liquid density near the ion is entirely justified (note the expanded scale of the ordinate). In contrast, the Atkins' model predicts a large increase in liquid density near the ion. As the result of the strong exchange interaction, the changes are so small that the creation of a solid-like cluster around a negative ion is not observed. We believe that this is the main reason for the experimentally measured [2] mobility difference between the O_2^- ion and positive ions. Measurements of the mobility of an O_2^+ ion are not available, but the mobility of the positive ions of *n*-pentane and tetramethylsilane in liquid Xe were found to be almost three times less than the O_2^- mobility.

For a simple estimate of the negative ion mobility in a liquid, we assume that during the motion of the ion in a weak electric field, a viscous flow of the liquid occurs that is determined by its local density near the ion. It is then possible to model the motion of both negative and positive ions as the motion of spheres with effective radii R_{eff}^\pm that differ for the two kind of ions. From the Stokes formula, the mobility is

$$\mu = \frac{e}{cR_{eff}^\pm \eta}, \quad (9)$$

where η is the viscosity of the undisturbed liquid and the numerical constant $c = 4\pi$ for a bubble or 6π for a cluster [12]. It follows from Eq. (9) that the product $\mu\eta$ is independent of the kinetic characteristics of the liquid. It is determined solely by the structure of the resulting ionic complex.

In Fig. 2 the experimental dependence of $\mu\eta$ on the temperature of liquid Xe along the saturation curve is shown. It is obvious that besides the quantitative differences in $\mu\eta$ of the positive and negative ions, there is also a qualitative difference between them. This is exhibited by the differing slopes of $\mu(T)\eta(T) = \text{const}$ for the two types of ions, and it demonstrates the differing dependencies of the structures of the positive and negative ions on liquid density and temperature.

Ionic motion in a liquid results in energy dissipation. The liquid layer with maximum viscosity is attached directly to the surface of the ionic sphere. This layer contributes most actively to dissipation. With this assumption, it is possible to estimate the effective radii of the charged complexes,

$$R_{eff}^\pm(N) \simeq R_c^\pm(N)(\eta_m/\eta), \quad (10)$$

where $R_c^\pm(N)$ are the radii of positive and negative ion complexes, respectively; η_m are the maximum viscosities near the ion, and η is, as before, the viscosity of the bulk liquid. Expanding η_m in Taylor series up to the second order in the relative variation in liquid density, we can write

$$\eta_m \simeq \eta(N, T) [1 + \xi_T(N_m - N)], \quad (11)$$

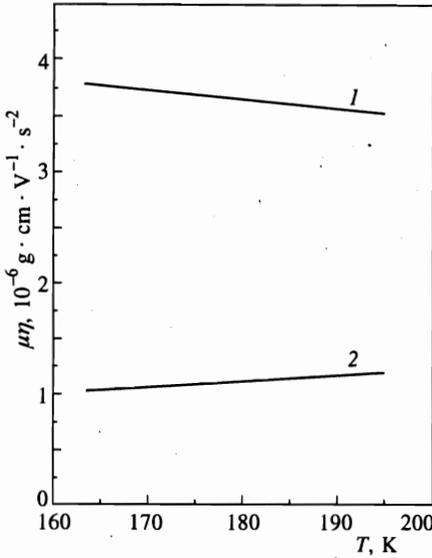


Fig. 2. Experimental results [2] for $\mu(T)\eta(T)$: 1) O_2^- ion; 2) positive ion of *n*-pentane in liquid Xe on the saturation curve

where N_m denotes the maximum density near the ion. The effective ionic radii are then

$$R_{eff}^\pm(N) = R_c^\pm(N)(1 + \xi_T \Delta N), \quad \Delta N = N_m - N. \tag{12}$$

We now analyze the situations for negative and positive ions separately. In case of the negative ion, it is evident from the calculations (Fig. 1) that beyond the maximum, ΔN decreases with N . In other words, the characteristic size of the localization region of the weakly bound electron λ determines the size of the negative ion. For a simple estimate, R_c^- can be set to λ and can be considered independent of the liquid density. Then, according to Eq. (12), $R_{eff}^-(N)$ is an increasing function of ΔN , and consequently a decreasing function of the liquid density N , which is consistent with the experimental data shown in Fig. 2.

We next consider the mobility of the negative ion, using the present model for the region of enhanced density and pressure near the ion. This region results in a local increase in shear viscosity. As the viscosity of the liquid $\eta(N, T)$ depends on density and temperature, $\eta(r)$ can be evaluated as a function of distance from the center of the ion.

As can be seen from Fig. 1, the local density increase is small. Moreover, the viscosity of the liquid Xe under these conditions is approximately a linear function of the liquid pressure. The local viscosity as a function of distance from the center of the ion can be written as $\eta(r) = \eta_\infty [1 + (R_\eta/r)^4]$, where η_∞ is the viscosity of the unperturbed liquid and $R_\eta = [(\partial\eta/\partial P)_T (\alpha F_L(N) e^2 N / 2\eta_\infty)]^{1/4}$ is the characteristic distance. R_η depends on properties of the liquid, and does not depend on any properties of the ion.

Previous results [13] were used to calculate negative ion mobility. The effective ion radius $R_{eff} = R_c^- \varphi$, where φ is a dimensionless number that includes corrections to the Stokes formula. For a bubble (perfect slip boundary conditions) of radius R_c^- and $y_0 = (R_\eta/R_c^-)^4 > 1$, we use the approximate expression [13]

$$\varphi(y_0) \cong \frac{8\sqrt{2}}{2\pi} y_0^{1/4} \left[1 - \frac{8\sqrt{2}}{3\pi} y_0^{-3/4} \right]. \tag{13}$$

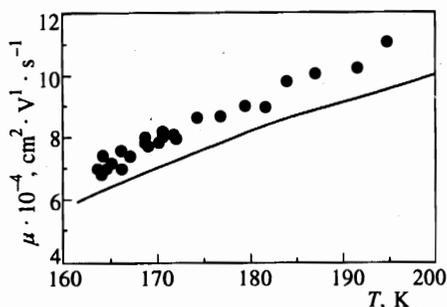


Fig. 3. Mobility of the O_2^- ion in liquid Xe on the saturation curve. The curve is calculated in accordance with the Stokes formula, and the points are the experimental results [2]

The estimates of R_η were made at $T = 170$ K and $T = 200$ K. The results were very similar for the two points. Based on the present approach for these two cases, $R_\eta \approx 8.6a_0$ and $\varphi \approx 1.56$. Our calculation of negative ion mobility by means of the modified Stokes formula (9), (13) is presented in Fig. 3. We obtain satisfactory agreement with experiment [2], although we consider these results to be preliminary.

For a positive ion, $R_c^+(N)$ denotes the radius of the cluster created from the surrounding atoms as a result of electrostriction. $R_c^+(N)$ is a sharp function of the liquid density, and it can be determined using the modified Atkins' model [2]. Near the triple point, the cluster size is determined by the surface tension of the liquid–solid interface, and can be written

$$[R_c^+(N)]^3 \approx \frac{\alpha e^2}{16\pi\epsilon_s\epsilon_l\sigma_{sl}^t(N_s - N)^3}. \tag{14}$$

Here α is the polarizability of the Xe atom, ϵ_s and ϵ_l are permittivity of solid and liquid Xe, respectively, N_s is the density of solid Xe, and σ_{sl}^t is the surface tension of the liquid–solid interface at the triple point. Substituting (14) into (12), we have $R_{eff}^+ = (\zeta_1/\Delta N) + \zeta_2$, where ζ_1 and ζ_2 are weak functions of N .

Thus, the effective radius of a positive ion is a decreasing function of ΔN , and consequently it is an increasing function of the liquid density N . We obtain the same condition on the saturation curve far from the triple point, where it is possible to neglect the effect of surface tension on cluster properties. In this case, the cluster radius is given by

$$[R_c^+(N)]^4 = \frac{\alpha e^2 N}{8\pi\epsilon_s\epsilon_l(P_s - P)} \propto \frac{\alpha e^2 N \gamma}{8\pi\epsilon_s\epsilon_l s^2 \Delta N}. \tag{15}$$

Substitution of Eq. (14) into Eq. (12) results in an increasing function of N at small ΔN . We obtain $R_{eff}^+ = \zeta_3(N/\Delta N)^{1/4} + \zeta_4 N^{1/4} \Delta N^{3/4}$, where ζ_3 and ζ_4 are also weak functions of N . From these qualitative considerations it can be seen that the increase in R_{eff}^+ with N is related to the increase in the cluster radius R_c^+ despite a simultaneous decrease in the distortion of the local liquid density ΔN near the cluster. This result is in good agreement with the experimental data [2] shown in Fig. 2.

The radius of the negative ion depends weakly on liquid density, and is mainly determined by the inner state of the ion. As result, the distortion of the liquid density near the negative ion is the principal factor determining $R_{eff}^-(N)$. With increasing liquid density, the role of the exchange interaction of the weakly bound electron with atoms is augmented, which results in an increase in $L(N)$ and a larger decrease in ΔN near the ion. Therefore, the present model enables us to interpret qualitative differences in the structure and mobility of negative and positive ions.

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