Abstract—We report the results of the experiment aimed at measuring the mobility of $O_2^-$ ions in dense argon gas in the temperature range $180 \, \text{K} \leq T \leq 300 \, \text{K}$. We show that an adequate theoretical description of the data is obtained by using the thermodynamic free volume (FV) model, originally developed to describe the electron bubble mobility in superfluid helium and successfully exploited for describing the $O_2^-$ mobility in near critical neon gas. The model goal is to thermodynamically predict the free space available for ion motion. By implementing the FV model with the Milikan–Cunningham (MC) slip correction factor, we can describe the ion mobility in the crossover region bridging the dilute gas kinetic regime to the high-density hydrodynamic regime of ion transport. These results confirm the validity of the model and the universality of some of its features.

Index Terms—Argon gas, free volume (FV) model, hydrodynamic regime, kinetic regime, oxygen ion mobility, slip correction factor.

I. INTRODUCTION

The detailed knowledge of the transport properties of ions in dense gas and liquid dielectrics is the cornerstone for understanding, designing, and harnessing processes in several natural as well as applicative fields. Control over biological processes, chemical synthesis protocols, electrical discharge equipment facilities, among many others requires that the behavior of ions drifting under the action of electrical fields is known and/or predictable in advance. Also, the behavior of high-energy detectors [1] and low-temperature plasma processes is affected by the transport behavior of ions [2]. Moreover, the profound comprehension of the physicochemical mechanisms of the processes occurring in the atmosphere, so important to ascertain several aspects of climate change, is based on the knowledge of the ion transport regimes [3]. In addition, last but not least, the investigation on how ions drift in a dense disordered dielectric medium can shed light on the fundamental ion–neutral interaction mechanisms and provides pieces of information on the ion–neutral interaction potentials [4], [5].

Typically, two different regimes for ion transport are considered. In dilute gases, the ion mobility $\mu$ is mainly determined by binary collisions with the host gas atoms, and the classical kinetic theory gives an accurate description of $\mu$ as a function of electric field and temperature provided that the ion–atom scattering cross section is known, and the appropriate collision integrals can be computed. Conversely, mobility data can be used to invert the equations of the classical kinetic theory to determine the interaction potential [6], [7], [8]. If the neutral–ion interaction can be modeled as a hard-sphere interaction with hard-core radius $R_0$, the density-normalized mobility $\mu N$ of the ions in thermal equilibrium with the gas, as is the case of the present experiment, is obtained as follows:

$$\mu N = \frac{3}{2\sigma} \left(\frac{\pi}{2m_e k_B T}\right)^{1/2}$$

in which $\sigma = \pi R_0^2$ is the scattering cross section, $N$ is the gas number density, $T$ is the temperature, $k_B$ is Boltzmann’s constant, and $m_e$ is the ion–neutral reduced mass.

At the other boundary of the thermodynamic state of the medium, ions drift in a liquid and their thermal mobility is described by the Stokes hydrodynamic formula

$$\mu N = \frac{eN}{6\pi \eta R}$$

in which $e$ is the charge of the (singly charged) ion, $\eta$ is the gas viscosity, and $R$ is the so-called hydrodynamic (or, effective) radius, which is assumed to be a constant for a given ion–neutral pair [9], [10].

However, this experiment, as several more similar ones [11], [12], [13], is carried out in a dense gas, for which a full-fledged theory for ion transport encompassing the crossover region between the dilute gas limit and the hydrodynamic regime is absent. Moreover, negative ions are not extensively investigated as the positive ones because of the difficulty of their production. Whereas positively charged ions are produced by direct ionization, the formation of a negative ion goes through several steps, which may not necessarily occur in the correct sequence. First, low-energy electrons must be captured by electronegative molecular impurities. Then, the vibrationally excited transient anions, thus, formed quickly decay by autodetachment, but a fraction of them can be
stabilized by collisions with a third body, typically a host gas atom that carries away the excess energy [14], [15]. Only this fraction of ions is sufficiently long-lived to allow the experimenters to measure their mobility. Only a small number of stable ions are made available, because this negative ion production mechanism is not efficient and strongly depends on the environment. The delicate balance of short-range repulsive exchange forces acting between the excess electron in the ion and the electronic clouds of the surrounding atoms, and the long-range polarization interaction leads to the birth of a state that cannot adiabatically be obtained by simply adding an ion to the medium. Therefore, negative ions are endowed with a structure more complicated than that of cations. The ion is localized in an empty cavity surrounded by a solvation shell produced by electrostriction. The properties of the structure depend on the atomic polarizability of the gas and on its thermodynamic state [16], [17], [18].

Over the years, we have been carrying out several experimental investigations on the mobility of O$_2^-$ ions in dense noble gases in wide temperature and density ranges [11], [12], [13], [19]. Oxygen is the most ubiquitous and important attaching species for evident reasons and is readily available as an impurity even in the best purified gas. Being the same ionic species in different gases, its behavior sheds light on gas specific features.

The choice to carry out measurements in a supercritical or near-critical gas is dictated by the fact that the gas density can be varied at will by changing pressure or temperature. By so doing, also the Knudsen number, $K_n = \ell (N, T)/R$, which is the ratio of the mean free path $\ell$ of the neutrals to the ion effective radius, can be varied in quite a range, thereby bridging the $K_n \ll 1$ hydrodynamic regime to the $K_n \geq 1$ kinetic regime.

As pointed out several times [11], [12], [13], [19], [20], [21], in the broad temperature and density ranges, we have explored, neither the kinetic nor the hydrodynamic description of the experimental data are adequate. However, a thermodynamic model (the FV model) has been recently developed that aims at computing the $N$ and $T$ dependence of the free volume (FV) available for the ion motion via an equation of state (EOS) [22], [23], [24]. The size of the FV, which is of spherical shape based on symmetry arguments, is assumed to be the effective ion radius to be inserted in the Stokes formula of the hydrodynamic regime. The extension of the model prediction to smaller densities to describe the crossover region toward the kinetic regime is obtained by adopting a suitably modified form of the Millikan–Cunningham slip correction factor [25], [26], [27].

Despite being originally developed to describe the mobility of electron bubbles in superfluid helium, the model has proven very effective to also describe the mobility of any kind of ions in several liquids and gases. We have successfully adopted this FV model to describe the mobility of O$_2^-$ ions in dense (supercritical and near-critical) neon gas [20], [21] in extremely broad $N$ and $T$ ranges.

Owing to the efficacy of the FV model and to test its validity for other systems, we have carried out the measurements of the O$_2^-$ mobility in argon gas in the temperature range $180 \, K \leq T \leq 300 \, K$. We report here the new data and the result of their analysis with the FV model.

II. EXPERIMENTAL DETAILS

The experiment has thoroughly been described elsewhere [11]. We briefly recall here its main features. We used the pulsed Townsend photograph injection technique. The experimental cell can be pressurized up to $\approx 8 \, \text{MPa}$, and its temperature regulated within $\pm 0.01 \, \text{K}$ in the range $25 \, K \leq T \leq 340 \, K$. O$_2^-$ is produced by resonant electron attachment to O$_2$ molecular impurities in a concentration of a few ppm [20]. Pressure readings are accurate within $\pm 2 \, \text{kPa}$. The gas density is computed by means of an accurate EOS [28]. A dc, high-voltage generator energizes the drift capacitor up to a maximum voltage of $\approx 3 \, \text{kV}$. The drift distance is $d \approx 1 \, \text{cm}$. The drift signal is passively integrated to improve the signal-to-noise ratio, and the drift time is obtained by the analysis of the signal waveforms [29]. The explored range of reduced electric field is limited to $E/N \leq 40 \, \text{mTd}$ ($1 \, \text{mTd} = 10^{-24} \, \text{V m}^2$). Thus, the ions always are in thermal equilibrium with the gas, and their mobility $\mu$ turns out to be field-independent even at the lowest $N$, as can be realized by inspecting Figs. 1 and 2.

We carried out measurements along several isotherms between $T \approx 180 \, K$ and $T \approx 300 \, K$. The explored density range spans from $N \approx 1 \times 10^{26} \, \text{m}^{-3}$ up to $N \approx 50 \times 10^{26} \, \text{m}^{-3}$, well in the crossover region from the kinetic to the hydrodynamic regime.
The FV model has successfully been adopted to rationalize both electron bubble [22], [24], [31] and positive ion mobility [32] in liquid helium, but also $O^+_2$ ion mobility in low-density helium [23] and in dense gaseous neon [20], [21]. The main reasons for the success of the FV model in different systems are that it computes in a thermodynamical way the effective ion radius, therefore accounting for the structure surrounding the ion itself, and that it exploits a modified form of the MC slip correction factor to bridge the two limiting transport regimes.

The cornerstone of the FV model is the concept of FV $V_s$, i.e., the volume available for the ionic motion through the medium. Ions are the solute species in the gas that acts as a solvent. The FV is given by $V_s = V - b$, in which $V$ is the macroscopic volume occupied by the gas and $b$ the covolume of its atoms. As the ion number produced in the experiment is extremely low (their concentration is $\leq 10^{-15}$), their covolume can safely be neglected.

The FV per particle $V_s = V_f/N$ is described by a van der Waals-like EOS

$$V_s = \frac{V_f}{N} = \frac{k_B T}{P + \Pi}. \quad (3)$$

$P \equiv P(N, T)$ is the hydrostatic pressure, which incorporates the attractive potential energy contributions among neutrals, and $\Pi$ is the internal pressure that accounts for the excess attractive potential energy contributions in the systems, i.e., those due by the long-range polarization interaction between ions and neutrals. The simplest analytical form for the internal pressure is

$$\Pi = \alpha N^2 \quad (4)$$

in which $\alpha$ is a constant that determines the density at which the compressibility is maximum and that has been proven to be system-independent according to the analysis of several ion–gas and ion–liquids systems [20], [23], [31], [32].

Owing to the spherical symmetry of the ion–neutral interaction, $V_s$ is assumed to have spherical shape, and its radius is considered to be the effective hydrodynamics radius $R$. As we expect that the size of the solvation shell depends on the ion–gas system compressibility, we must seek an analytic form relating $R$ to $V_s$ that accounts for this effect by enforcing agreement between the theoretical prediction and the experimental data.

To extend the range of applicability of the Stokes formula to crossover and to lower density regions, the pure hydrodynamic Stokes formula is modified by the introduction of the empirical MC slip correction factor $\phi(K_s, T)$, thereby leading to the following expression for the mobility:

$$\mu N = \frac{e N}{6\pi \eta R} (1 + \phi[K_s(N, T)]). \quad (5)$$

The behavior of $\phi$ is such that it must vanish at very small Knudsen number, i.e., at high density, where the Stokes formula adequately describes the mobility, and that it must give (1) for large Knudsen number, i.e., at low enough density.
The Knudsen number is evaluated by using the kinetic theory expression for the mean free path of the neutrals [33]

\[ \ell(N, T) = \frac{3\eta}{N\sqrt{8m_k T / \pi}} \]  

(6)

As the experiment is carried out at temperature quite higher than the critical one, we can use a version for the hydrodynamic radius with a much simpler temperature dependence than that used for neon gas close to the critical temperature [20], [21]. Thus, we assume

\[ \frac{R}{R_0} = 1 + \frac{(V_0 / V_s)^{\epsilon_1}}{1 + \gamma (V_0 / V_s)} \]  

(7)

in which the parameters \( R_0 \), i.e., the hard-sphere radius of the ion–medium interaction, \( \gamma \), \( \epsilon_1 \), and the FV of a suitable reference state, \( V_0 = \delta k_B T \), have all to be adjusted to give the best agreement with the experimental mobility data. We found \( R_0 = 0.555 \text{ nm} \), \( \epsilon_1 = 12 \), and \( \gamma = 2 \). Moreover, we found that \( \alpha = 0.8937 \text{ MPa nm}^2 \) and \( \delta = 0.15 \text{ m}^2 / \text{J} \). It is worth noting that the present values of \( \alpha \) and \( \delta \) are the same that give the best agreement in all investigated systems [20], [21], [22], [23], [31], [32] and also in several cation–liquids systems not yet published. Thus, the parameters \( \alpha \) and \( \delta \) appear to be universal. We believe that this universality arises as a consequence of adopting a van der Waals-like approach to the ion-gas thermodynamic description that leads to the manifestation of a sort of Law of Corresponding States.

Finally, the slip correction factor can be cast in the form

\[ \phi(N, T) = A (\frac{N_c}{N})^\epsilon \]  

(8)

in which \( N_c \approx 80.8 \times 10^{26} \text{ m}^{-3} \) is the critical density of argon, \( A \approx 1/4 \) and \( \epsilon \approx 1.1 \).

V. COMPARISON OF EXPERIMENTAL OUTCOME AND THEORETICAL PREDICTION

In Fig. 5, we compare the predictions of the FV model with the experimental data for \( T = 180 \text{ K} \) and \( T = 260 \text{ K} \). Similar results are obtained for all other investigated isotherms. We note that the values of the free parameters appearing in (4), (7), and (8) have been determined once for all by seeking agreement between the \( T = 180 \text{ K} \) experimental data and the model and retain their values through all other isotherms. As we can see, the agreement between the model and the experiment is satisfactory.

We note that the value of the parameter \( \epsilon \) is slightly larger than 1. This value would be problematic in the limit of zero density. However, the value \( \epsilon \approx 1.1 \) is obtained by a global fit of the ratio of the mean free path to the hydrodynamic radius over all the investigated density range. Actually, we found that \( \epsilon \) depends on \( N \) in such a way that \( \epsilon = 1 \) at low density and \( \epsilon \to 1.2 \) at high density. We believe that this behavior is an artifact of the choice of using the kinetic expression of the mean free path that involves the dynamic viscosity of the gas instead of using the simpler expression \( \ell = 1/N \sigma \), in which, unfortunately, the scattering cross section \( \sigma \) is not known. We further observe that in the real experiment, the true zero-density limit is not even closely approached. Actually, the lowest density we achieved is on the order of \( 1 \times 10^{26} \text{ m}^{-3} \), i.e., roughly four times the density of an ideal gas at a pressure of 1 atm and at a temperature of 0 °C.

The FV model can also predict that the effective hydrodynamic radius bridging the hydrodynamic- to the kinetic transport regime over the density crossover region is cast in the form

\[ R_{eff} = \frac{R}{1 + \phi} \]  

(9)

which has to be compared to its experimental determination \( R_{eff} = e / 6\pi n \eta \mu \). The comparison of the two determinations of the effective radius is shown in Fig. 6. We only report the results for \( T = 180 \text{ K} \), because its investigated density range is the broadest among all isotherms. Moreover, the resulting temperature dependence of \( R_{eff} \) is so weak that no further insight is gained by plotting the results for all remaining temperatures.

On the one hand, we note that the pure hydrodynamic radius \( R \) is almost constant throughout the whole investigated density range. This fact implies that the experiment has been carried out in the crossover density region but on the low-density side of it. Consequently, the pure Stokes formula would certainly fail at describing the mobility data.

On the other hand, we see that the agreement between the model prediction and the experimental determination of the
effective radius is quite good. We observe that $R_{\text{eff}}$ linearly depends on $N$ at very low density. This means that in this limit, $\mu N$ turns out to be independent of the density as required by the kinetic theory. As the density is increased, the effective radius increases with decreasing slope and tends to a saturation value, thereby approaching the pure hydrodynamic radius and, hence, the hydrodynamic regime.

We can conclude that in the explored temperature and density ranges, in particular, at temperatures quite higher than the critical one, the most important factor to get agreement with the experimental data is to implement the results of the FV model with a suitable form of the slip correction factor. The same conclusions apply to all investigated isotherms.

Although the model has been developed in such a way to extend the Stokes formula to relatively low densities, nonetheless, it also predicts quite correctly the temperature dependence of the zero-density limit of the density-normalized mobility, $(\mu N)_0$, defined as follows:

$$
(\mu N)_0 = \lim_{N \to 0} \mu N.
$$

We show in Fig. 7 the temperature dependence of the experimentally determined $(\mu N)_0$ and the prediction of the FV model. In spite that the accuracy of $(\mu N)_0$ is relatively poor, because it is obtained by extrapolating the available experimental data to $N \to 0$, nonetheless, it appears that the FV model gives a satisfactory description of the experimental outcome. We must stress the fact that, although we reached the lowest possible densities in the experiment, nonetheless, we are by far distant from the pure kinetic regime. Actually, in the limit $N \to 0$, $(\mu N)_0$ should be given by the so-called polarization (or, Langevin) limit [34]

$$
(\mu N)_0 = \frac{4.81 \times 10^{-4}}{\sqrt{m_r \alpha_s}} \approx 6.9 \times 10^{21} \text{ (Vms)}^{-1}
$$

in which $\alpha_s = 11.08 a_0^3$ is the atomic polarizability of argon, $a_0$ is the Bohr radius, and $m_r \approx 2.95 \times 10^{-25}$ kg is the $O_2^-$-Ar reduced mass. This picture once more confirms the efficacy of the FV model at treating the transition transport regime.

VI. Conclusion

In this article, we have reported on new measurements of the $O_2^-$ ion mobility in dense argon gas in quite extended temperature and density ranges and on their theoretical rationalization with the FV model. The measurements are carried out for densities that are too large for the pure kinetic theory to apply and too low for the hydrodynamic theory to be valid. The measurements, thus, explore a crossover region between the two limiting transport regimes.

We have shown, by means of MD simulations, that the structure of the fluid surrounding the ions does determine their transport behavior.

The FV model that treats the ions as a low concentration solute in a solvent is just aimed at computing the volume associated with each ion that is available for its motion by introducing a van der Waals-like EOS for the solute. The linear size of this volume is assumed to be the ion hydrodynamic radius to appear in the hydrodynamical Stokes formula.

The model, although relying on some adjustable parameters, nonetheless, shows some universal features. First, the hydrodynamic radius turns out to be a function of the FV. Moreover, the expressions for the internal pressure $\Pi$ and of the volume of a reference thermodynamical state $V_0$ turn out to be common to all investigated systems and are, thus, universal. We believe that this universality stems from a sort of Law of Corresponding States following the van der Waals-like approach. Finally, the model parameter, adjusted to fit one specific isotherm, keeps their values though all remaining isotherms.

To bridge the high- to the low-density regions, the Stokes formula for the mobility is supplemented with the introduction of a suitable slip correction factor in the wake of the work of Millikan and Cunningham. By so doing, the FV model quite well describes the mobility in dense argon gas in the crossover region. Similar good results were also obtained for the mobility of $O_2^-$ ions in dense neon gas, of electron bubbles in normal liquid, and superfluid helium and in several more systems, including cations in different liquids.

Owing to the efficacy of the model, we plan in the next future to apply it to the $O_2^-$ mobility in dense argon gas very near to the critical temperature where the large gas compressibility presents a harsh challenge for testing the versatility of the FV model.

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