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Alkali ions mobility in parent vapor

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Certain approximations of mobility coefficients for the metal atoms' ions in parent vapor are investigated, a comparison between experimental data was drawn, and Monte Carlo calculations are performed. In contrast to ions of noble gases, ions and atoms of alkali metals have a number of features that must be considered when building a model of ion-atom collisions. Approximations for mobility coefficients for the alkali ions in parent vapor are given, they are valid not only in the weak electric field limit, but also in strong fields, when ion heating enacts in external electric field. On the basis of the analysis of Monte Carlo calculations and comparing them with experimental data on the mobility of alkali metal ions in noble gases, we obtain approximation formulas for the mobility of alkali metal ions in parent vapors. The parameters found earlier for the approximation of the drift velocity of ions of noble gases are also given as a reference material. The parameters obtained in this work for the approximation of the mobility of alkali metal ions in parent vapors can be used to estimate the characteristics of a gas-discharge plasma.

Key words: gas discharge, ion mobility, alkali atom, Monte Carlo, ion-atom collision, discharge plasma.

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1. Introduction

At present experimental and theoretical data on kinetic coefficients were obtained mainly 30-50 years ago. That time was the period when gas discharge plasma was very popular, as there were some technological problems in microelectronics, medicinal applications, surface treatment issues, tasks of obtaining plasma light sources, etc. The detailed review of the vast amount of experimental and theoretical results is given in a number of books and reviews [1-5].

Modern technologies often use plasma processes, in which there are metal vapors in the core. They fall into the gas discharge zone or due to the spraying of the structural elements of the gas-discharge chamber, or they are intentionally introduced.

Often the presence of impurities arising, which appeared, for example, due to sputtering of the cathode as a result of ion bombardment, is not taken into consideration while analyzing the results of the experiments. It is well-known that sometimes even very small impurities (ppm) can

fundamentally change the discharge characteristics [6-11].

It is necessary to know the kinetic coefficients – mobility, diffusion, viscosity, etc. for the qualitative estimation, for modeling the processes in gas discharge plasma on the basis of multifluid gas flue models. This article is devoted to closing the gap in the part of the alkali ions mobility coefficient characteristics in parent vapor, as the available data are sketchy and differ greatly.

2. Ion mobility in a weak electric field

Let's describe different drift velocity approximations for the case of small field gradients, when the drift velocity is much smaller than the thermal atom velocity and therefore the anisotropy of the ion velocity distribution function can be neglected. A detailed and thorough review of the available experimental and computation data along with different approximations is given in the book [1] and review [2 – 5]. The ion drift velocity in the gas under the influence of constant and uniform electric field is determined by the expression.

$$u = KE, \quad (1)$$

where K – ion mobility coefficient, E – electric field strength. The mobility coefficient in this equation is a function of the density and temperature of the atoms, as well as the field strength.

In the case of small fields, the temperature of the atoms determines the average collision energy and the dominant type of collisions. Consequently, it is the temperature that determines the values of the coefficients of mobility and diffusion of ions in the limit of a weak field. Measurement of the dependence of the mobility of ions on the gas temperature allows us to determine the potential of the interaction of an ion with an atom [1, 12].

When an ion collides with an atom of its own gas, there is a probability of electron transition from atom to ion without changing the internal energy of the colliding particles. Such a process is called resonant charge exchange and usually refers to inelastic collisions, since it has a quantum mechanical nature [1, 12]. Collisions with resonant charge transfer dominate at not very low temperatures.

Then, the solution of the Boltzmann kinetic equation at low electric field strengths in the first approximation of the Chapman – Enskog leads to the following expression for the mobility of ions in its own gas [12]:

$$K_0 = \frac{3\sqrt{\pi}e}{16(m_i T)^{1/2} \sigma_{res} (3v_T) N_0} = \frac{1.342 \times 10^4 \text{ cm}^2}{\sqrt{m_i T} \sigma_{res} V \cdot s} \quad (2)$$

Here K_0 – mobility at standard gas density $N_0 = 2.686763 \times 10^{19}$ atoms in cm^3 , m_i expressed in proton mass units, atom temperature T – in Kelvin degrees, and the cross section for resonant charge exchange σ_{res} – in 10^{-16} cm^2 , thermal velocity of atoms $v_T = \sqrt{T/m_i}$. The numerical factor in the formula (2) $C_{C-E}^{(1)} = 3\sqrt{\pi}/16 \approx 0.332$ corresponds to the first approximation of Chapman – Enskog, in the second order approximation for it is obtained close value $C_{C-E}^{(2)} = 31\sqrt{\pi}/168 \approx 0.327$ which indicates the

rapid convergence of the expansion of the distribution function in a series of polynomials.

The probability of electron transfer from atom to ion (charge exchange) drops exponentially sharply with increasing distance between particles. Therefore, with good accuracy, we can assume that after the passage with the distance of the closest approach $r_{\min} > R_0$ the probability of electron transfer is negligible small, where the magnitude R_0 determines the reaction threshold. If an ion approaches an atom close enough, so that $r_{\min} < R_0$ then the electron orbits of the atom and ion overlap. In this case, the electron during the collision will make many transitions from atom to ion and after the collision will remain with equal probability in one of the colliding particles. The charge exchange cross section in this approximation is determined by the relation:

$$\sigma_{res} = \frac{1}{2} \pi R_0^2. \quad (3)$$

As shown by theoretical and experimental data, the resonance charge exchange cross section logarithmically weakly depends on the relative velocity $v/2$ of the colliding particles and can be approximated by the following relationship:

$$\sigma_{res}(\varepsilon_{12}) = \pi a^2 \ln^2(U_0 / \varepsilon_{12}) = \sigma_{res}(\varepsilon_0) [1 + a \ln(\varepsilon_0 / \varepsilon_{12})]^2, \quad (4)$$

or

$$\sigma_{res}(\varepsilon_{12}) = \sigma_{res}(\varepsilon_0) + B \ln(\varepsilon_0 / \varepsilon), \quad (5)$$

where $U_0, a, B, \sigma_{res}(\varepsilon_0)$ – positive approximation constants [1, 12]. At collision energies $\varepsilon_{12} = m v_{12}^2 / 2 < Ry$ cross section $\sigma_{res}(\varepsilon_{12})$ usually several times greater than the gas kinetic cross section (m is the mass of an ion, for the case of collisions in its own gas, equal to the mass of the atom).

At not very low collision energies, the cross section for resonant charge exchange weakly depends on the collision energy (relative velocity of the colliding particles); therefore, in derivaton (2), it was assumed that the mean free path does not depend on the collision energy. Due to the polarization of the atom in the ion field, an attraction arises between them. The diffusion cross section for scattering caused by the scattering of

ions due to the polarization of the atom in the ion field is [1, 12]:

$$\sigma = 1.105\sigma_{capture} = 2.21\pi\sqrt{\alpha_d e^2 / \mu v^2}. \quad (6)$$

Here $\sigma_{capture} = 2\pi\sqrt{\alpha_d e^2 / \mu v^2}$ – is the capture cross section on the spiral orbit in the polarization potential. Accordingly, the mobility of ions in the polarization limit is determined by the well-known expression [1, 2]:

$$K_0 = \frac{0.217e}{(m_i T)^{1/2} [R_0^2 (3v_T) + \alpha_d e^2 / 4TR_0^2 (\sqrt{6T / m_i})] N_0}, \quad (8)$$

The mobility of ions in their own gas, calculated by the formula (8), in the limit of low temperatures, does not differ much from the exact result. Having

$$K_0 = \frac{0.217e}{(m_i T)^{1/2} [R_0^2 (3v_T) + \alpha_d e^2 / 4TR_0^2 (\sqrt{6T / m_i}) + 0.16\sqrt{\alpha_d e^2 / T}] N_0}. \quad (9)$$

3. Ion mobility in a strong fields

When ion drift in a strong field, the drift velocity becomes comparable or even greater than the thermal velocity of the atoms; therefore, the anisotropy of the ion distribution function in velocities cannot be neglected. Note that the criterion of a strong field depends not only on the electric field strength, but also on the gas temperature. Generally speaking, at any field strength, the gas can be of such a temperature that the thermal velocity of the atoms will be less than the drift velocity. Therefore, in cryogenic discharges, even at field strengths of the order of 2 V / cm, which are typical for a gas discharge under reduced pressure, the field can be considered as a strong [13, 14].

Numerous experimental data show that the drift velocity of ions in their own gas is well described by the semi-empirical Frost formula [15], which takes

$$K_{pol} = 13.853(\alpha_d \mu)^{-1/2}, \quad (7)$$

where α_d – polarizability in cubic angstroms, μ – reduced mass in grams per mole.

To take into account the polarization interaction, the first Chapman – Enskog approximation for mobility in the absence of elastic scattering was corrected in [12] and the following expression was obtained:

corrected formula (8) so that in the limit of low temperatures it gives an exact result, the following expression for mobility was obtained in [12]:

into account the dependence of the drift velocity on the field strength:

$$u = a(1 + bE)^{-1/2} E. \quad (10)$$

This approximation for dependence of the drift velocity on the electric field has two parameters: a – mobility in the weak field limit, and $b = 1 / E_{heating}$. Magnitude $E_{heating}$ – this is the value of the electric field strength, in which, due to the heating of the ions, the mobility according to the Frost formula decreases by a factor of two. Both of these parameters depend only on the atomic temperature and density, but do not depend on the electric field intensity.

Let us rewrite Frost's formula (10) as a dependence of the drift velocity on the reduced electric field strength:

$$u = K_0 N_0 \left(1 + (E / N) / (E / N)_{heating} \right)^{-1/2} (E / N). \quad (11)$$

Here are two parameters $(E/N)_{heating}$ and K_0 depend only on temperature: $(E/N)_{heating}$ – is the value of the reduced electric field strength, at which, due to the heating of the ions, the mobility according to the Frost formula decreases by a factor of two.

4. Approximation of mobility in a weak fields

In the literature, the values of ion mobility K_0 are mainly given at room temperature 291~300K. There is also an insignificant amount of experimental data on the characteristics of the drift of helium and neon ions at cryogenic temperatures near the 77 K and 4.2 K points [3 – 5].

Experimental data on the mobility of lithium, sodium, and cesium ions in helium in the atomic temperature range from 20 to 500 K are given in review [4], and data on the mobility of alkali metal ions in inert gases at room temperature are given in book [12]. Experimental data on the mobility of alkali ions in their own vapors for potassium at room temperature and rubidium, cesium at atom temperatures from 620 to 660 K are given in book [12].

In [9], based on the analysis of Monte Carlo calculations of the kinetic characteristics of ion drift in its own gas for all noble gases, an approximation was chosen for the dependence of mobility in a weak field on the gas temperature:

$$K_0 = K_{pol} / (1 + T / \varepsilon_0)^{1/2}. \quad (10)$$

Here $K_{pol} \equiv K_0(0)$ is the value of polarization mobility at the standard gas density in the limits of weak field and low temperature. Fitting parameter ε_0 in (10) has the meaning of the characteristic collision energy, upon reaching which the polarization interaction no longer makes the main contribution to the collision frequency and elastic collisions and resonant charge transfer begin to play the role. Table 1 shows the approximation parameters (10) for all noble gases, found in [9]. Parameter $(E/N)_0$, given in this table has the meaning of the upper limit of the electric field strength, at which the effects of a strong field begin to play a role. This parameter is used in constructing an approximation of the dependence of mobility on field strength.

Table 1 – The value of the approximation parameters for the mobility of ions of noble gases in its own gas.

System	K_{pol} cm ² /c B	ε_0 , K	$(E/N)_0$, Td
He ⁺ in He	21.6	90	16
Ne ⁺ in Ne	6.8	210	34
Ar ⁺ in Ar	2.42	240	73
Kr ⁺ in Kr	1.36	330	106
Xe ⁺ in Xe	0.85	270	122
Hg ⁺ in Hg	0.61	58	63

In contrast to ions of noble gases, ions and atoms of alkali metals have a number of features that must be considered when building a model of ion-atom collisions. The polarizability of ions of noble gases is not very different from the polarizability of atoms with the exception of the hydrogen-like helium ion. Since polarizability is directly related to some effective atomic radius, it was assumed during the play of ion-atomic collisions of inert gases that the gas-kinetic radius of an ion (hard sphere) is equal to the atomic radius. But for alkali ions, the situation is completely different. The outer electron shell of a single-ionized

alkali metal atom is similar to the outer electron shell of the corresponding inert gas atom. But it is in a stronger field than the shell of an inert atom, so its effective size will be smaller. Measurements show that the polarizability of an alkali metal ion is about one and a half to two times less than the polarizability of the atom of the corresponding inert gas, therefore it is quite logical to assume that the radius of the ion of the alkali atom is equal to the radius of the corresponding atom of the noble gas (lithium ion radius is equal to radius of a helium atom, sodium ion radius is equal to radius of neon atom, etc.)

Table 2 – Polarizability of noble gases, alkali and metals and their ions, where α_d – polarizability in cubic angstroms [1]

	α_d		α_d		α_d		α_d
He	0.205	He ⁺	0.0417	Li	24.0	Li ⁺	0.0285
Ne	0.395	Ne ⁺	0.21	Na	24.2	Na ⁺	0.280
Ar	1.64			K	41.3	K ⁺	1.09
Kr	2.48			Rb	43.6	Rb ⁺	1.81
Xe	4.04			Cs	53.0	Cs ⁺	2.73

There are very few experimental data on the mobility of alkali metal ions in their own vapors, but there is a lot of data on their mobility in the weak field limit in inert gases. In the book [12],

experimental data on the mobility of alkali metal ions in inert gases at room temperature are given. In Tab. 3, these data are compared with the calculations performed in this work.

Table 3 – The ratio of the mobility of alkali metal ions in inert gases at room temperature to the polarization mobility calculated by the formula (7). The experimental data is the *exp* line, italicized and bold, the Monte Carlo calculations are the *MC* line.

<i>Ion</i>		<i>He</i>	<i>Ne</i>	<i>Ar</i>	<i>Kr</i>	<i>Xe</i>
<i>Li⁺</i>	<i>exp</i>	1.27	1.17	1.06	1.07	1.06
	<i>MC</i>	1.05	1.06	1.05	1.08	1.07
<i>Na⁺</i>	<i>exp</i>	1.38	1.24	1.07	1.06	1.09
	<i>MC</i>	1.04	1.04	1.07	1.08	1.07
<i>K⁺</i>	<i>exp</i>	1.35	1.28	1.08	1.10	1.07
	<i>MC</i>	1.01	1.05	1.06	1.05	1.06
<i>Rb⁺</i>	<i>exp</i>	1.29	1.26	1.08	1.08	1.07
	<i>MC</i>	0.94	1.04	1.07	1.06	1.06
<i>Cs⁺</i>	<i>exp</i>	1.19	1.18	1.07	1.08	1.07
	<i>MC</i>	0.92	1.02	1.08	1.08	1.04

Note that for all the experimental data given in the table there is a pronounced effect of exceeding the ion mobility as compared with the polarization mobility. This effect is discussed in detail in many papers, and is usually associated with the competition between the attractive and repulsive parts of the interaction potential of an ion with an atom. A more detailed study of this effect will be given in a separate paper.

5. Approximation of mobility in a strong fields

To approximate the mobility of ions in a strong field, we use the Frost formula in the form (9). Based on the analysis of a large number of Monte Carlo calculations, it was found that the value of the parameter $(E/N)_{heating}$ can be very well approximated by the dependence on the same

parameter ε_0 for that obtained for approximating the mobility in the weak field limit.

Let us find the dependence of the value of the parameter $b^{-1} = (E/N)_{heating}$ on gas temperature. From the analysis of drift velocity calculations using the Monte Carlo method, we assume that the approximation formula is:

$$1/b = (E/N)_0 (1 + 1.5T_{atom} / \varepsilon_0). \quad (11)$$

Here $(E/N)_0$ – is the value of the heating field at zero gas temperature. We introduce a new parameter

$$\langle \varepsilon \rangle_{pol} = \frac{1}{2} m [1.5 (E/N)_0 K_{pol} N_0 / N]^2 \quad (12)$$

this is the average energy of the directional motion of an ion in a field with a reduced intensity $1.5(E/N)_0$ at zero atomic temperature and polarization mobility of ions (3). With an accuracy of about 1%, the values of $\langle \varepsilon \rangle_{pol}$ and ε_0 coincide, i.e. the parameter $(E/N)_0$ is not independent, but can be determined from the relation $\langle \varepsilon \rangle_{pol} = \varepsilon_0$:

$$\left(\frac{E}{N}\right)_0 = \frac{(\varepsilon_0 / 2m)^{1/2}}{3K_{pol}N_0}. \quad (13)$$

Slightly more complex than (11), the approximation of the form

$$1/b = (E/N)_0 [(1 + (1.5T_{atom} / \varepsilon_0)^{0.8})^{1.2}] \quad (14)$$

gives a slightly better agreement with the results obtained from the analysis of the calculations of mobility by the Monte Carlo method. Tabl. 4 shows the values of the parameter ε_0 , the polarization mobility K_{pol} and cross section parameters for alkali metal ions.

Table 4 – Parameter ε_0 , the polarization mobility K_{pol} and cross section parameters for alkali metal ions

	ε_0, K	$K_{pol} \text{ cm}^2/\text{c B}$	$(E/N)_0, Td$	$r_{gas}, 10^{-8} \text{ cm}$	$\sigma_{rez}(\varepsilon) 10^{-16} \text{ cm}^2$	b
Li	130	1,52	205,08	1,05	230,0	0,11
Na	110	0,83	188,65	2,234	260,0	0,11
K	100	0,48	239,32	2,77	350,0	0,10
Rb	410	0,31	497,63	2,48	247,0	0,024
Cs	370	0,23	533,37	2,68	295,0	0,025

Thus, we have the following approximation for the drift velocity:

$$u = \frac{K_{pol}N_0}{(1 + T_{atom} / \varepsilon_0)^{1/2}} \left(1 + \frac{3K_{pol}N_0}{(\varepsilon_0 / 2m)^{1/2} (1 + 1.5T_{atom} / \varepsilon_0)} \frac{E}{N} \right)^{-1/2} \frac{E}{N}. \quad (15)$$

For the drift velocity, formula (15) gives values that almost coincide with the available experimental data [3-6].

6. Conclusion

The available experimental data are fragmented and cannot serve as a reliable basis for a comprehensive analysis of a wide variety of processes and phenomena in a gas-discharge plasma. Previously obtained approximations for the mobility of ions in parent vapors, given, for example, in the book [12], have a large error due to the fact that they are based on the assumption of the decisive influence of collisions with charge exchange.

In the present work, on the basis of the analysis of Monte Carlo calculations and comparing them with experimental data on the mobility of alkali metal ions in noble gases, we obtain approximation formulas for the mobility of alkali metal ions in

parent vapors. The parameters found earlier for the approximation of the drift velocity of ions of noble gases are also given as a reference material. The parameters obtained in this work for the approximation of the mobility of alkali metal ions in parent vapors can be used to estimate the characteristics of a gas-discharge plasma.

The original purpose of the calculations was to demonstrate the paramount importance of the effect of gas temperature on the heating of ions under conditions of a cryogenic discharge, namely, in experiments with dusty plasma [6, 7]. But the results of calculations are much more general in nature, and in addition to cryogenic discharge can be used in the analysis of processes in gas-discharge plasma in installations of ion and plasma-chemical etching, deposition, sputtering in magnetron installations, in which the gas temperature can significantly exceed room temperature.

Generally speaking, the drift of ions in a gas discharge has a very significant effect on the

characteristics of a gas discharge. For example, a small fraction of mercury vapors in argon leads to a change in the ionic composition, a significant decrease in the electron temperature and a change in the radiative plasma characteristics [10].

The obtained parameters of the approximation of the drift rate of alkali metal ions in parent vapors can be used to estimate the characteristics of a gas-

discharge plasma, when analyzing and planning experiments with dusty plasma under cryogenic discharge conditions, when considering discharge in a mixture of heavy and light gases [6, 7], when analyzing experiments with the scattering of ultracold ions into the surrounding gas. For the drift velocity, formula (10) gives values that almost coincide with the available experimental data [1–5, 16-22].

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