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Nuclear Instruments and Methods in Physics Research A 580 (2007) 18–21
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Monte Carlo calculations of drift velocities and diffusion coefficients of Ar^+ ions in helium

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Available online 21 May 2007

Abstract

Results are presented for the calculated drift velocities and diffusion coefficients for Ar^+ ions in helium at atmospheric pressure, temperature $T = 300$ K and for reduced electric fields E/N from about 1 Td up to about 150 Td, using Monte Carlo techniques. The drift velocities range from 5.94×10^3 to 559.0×10^3 cm s^{-1} for the Ar^+ ions in the ground state $^2P_{3/2}$ and from 5.85×10^3 to 545.0×10^3 cm s^{-1} for the Ar^+ ions in the metastable excited state $^2P_{1/2}$. These values are in good agreement (within about 5%) with the few experimental values available. The mobilities and diffusion coefficients for atomic Ar^+ ions in helium gas show no significant dependence on the spin state of the ion.

PACS: 51.20; 61.18.B; 51.10; 52.25.Fi; 29.40.C; 52.65

Keywords: Ion mobility; Diffusion coefficients; Monte Carlo simulation; Ion transport in gases

1. Introduction

The study of the drift of noble gas ions in gases is of great interest for a variety of fields (radiation detectors, plasma physics, etc.) and can be the subject of detailed calculations using Monte Carlo techniques provided the relevant cross-section data (either theoretical or experimental) are available. In the present work, we have applied the Monte Carlo method using a three-dimensional simulation model, based on the work for electrons of Dias et al. [1], to study the drift of Ar^+ ions in gaseous helium, for $p = 760$ Torr and $T = 300$ K, under the influence of a uniform electric field. In the simulations we use a set of integral and differential elastic collision cross-sections for Ar^+ ions with neutral He, previously calculated by the authors [2] using a modified Tang–Toennies ion–atom interaction potential and the JWKB approximation to calculate the phase shifts.

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doi:10.1016/j.nima.2007.05.097

2. Interaction potentials and collision cross-sections of Ar^+–He

The Ar^+ ion has five electrons in the outer p orbital and is subject to spin–orbit coupling. Therefore, the $^2P_{3/2}$ ground electronic state of the Ar^+ ion interacts with closed shell He atom in its $^1S_0$ ground state to produce $X\ ^2\Sigma_{1/2}$ and $A_1\ ^2\Pi_{3/2}$ molecular states, Fig. 1, of which the $X\ ^2\Sigma_{1/2}$ is the molecular ground state. The two molecular states have the same probability and the cross-sections for the interaction of the ions in the $^2P_{3/2}$ ground state with helium atoms are weighted as $\frac{1}{2}(X\ ^2\Sigma_{1/2}) + \frac{1}{2}(A_1\ ^2\Pi_{3/2})$. The first excited state $^2P_{1/2}$ of the Ar^+ ion has an energy of 0.1775 eV [3] and is metastable, so it is likely that it will be present when the Ar^+ ions are formed and may be involved in mobility measurements. From the interaction of the Ar^+ ion in its $^2P_{1/2}$ metastable excited state with the helium atom ($^1S_0$) results only the $A_2\ ^2\Pi_{1/2}$ state.

The elastic integral and differential collision cross-sections for Ar^+ ions with neutral He atoms used in this work were previously calculated [2] by the authors, at

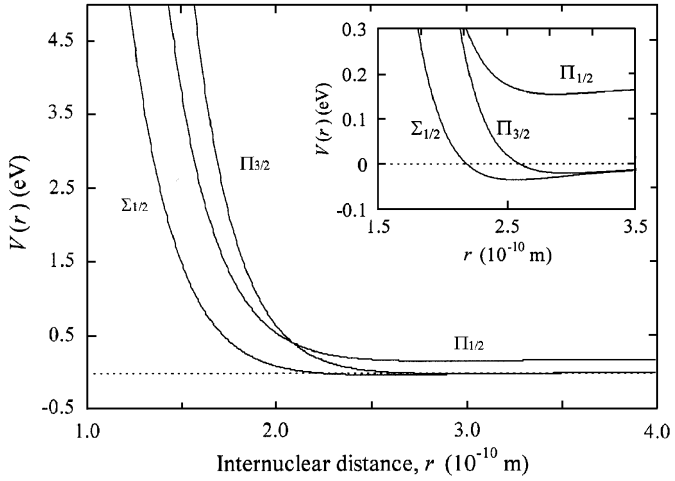

Fig. 1. Potential energy curves for $HeAr^+$ with spin–orbit coupling.

centre-of-mass energies in the 1 meV to 10 eV range. In this calculation was used a modified Tang–Toennies model [4] (based on universal damping functions for the dispersion coefficients) for the ion–atom potential energy curves for each of the three $HeAr^+$ quasimolecular states ($X\ ^2\Sigma_{1/2}$, $A_1\ ^2\Pi_{3/2}$ and $A_2\ ^2\Pi_{1/2}$):

$$V(r) = A\ \exp(-br) - B\ \exp(-br/2) - \sum_{n=2}^{3} f_{2n}(r) C_{2n} r^{-2n}$$

and the damping function, $f_{2n}(r)$, is given by

$$f_{2n}(r) = 1 - \exp(-br) \sum_{k=0}^{2n} \frac{(br)^k}{k!}.$$

This model can fit the available spectroscopic data or *ab initio* calculations, through the adjustable parameters A, B and b, using the well depth D_e, minimum position r_e and the zero crossing position σ of the interaction potential and also reproduce the correct long-range behavior of the interaction potential by incorporating the coefficients C_4 and C_6. The coefficient C_4 takes into account the ion-induced dipole interaction and the coefficient C_6 the van der Waals dispersion and ion-induced quadrupole interaction [5] and are calculated from dipole and quadrupole polarizabilities [5,6] of Ar^+ and He, and are found to be $C_4 = 1.4760 \times 10^{-40}$ eV m^4 and $C_6 = 4.6260 \times 10^{-60}$ eV m^6. The parameters for the potential energy curves for each state $X\ ^2\Sigma_{1/2}$, $A_1\ ^2\Pi_{3/2}$ and $A_2\ ^2\Pi_{1/2}$ are listed in Table 1.

The phase shifts, for the calculation of scattering amplitudes and subsequent elastic differential and integral cross-sections by the partial waves method, were calculated using the JWKB approximation [8].

3. The Monte Carlo method

Detailed three-dimensional simulation techniques using the Monte Carlo method, based on the work for electrons of Dias et al. [1], are used to study the drift of Ar^+ ions in gaseous helium under the influence of a uniform reduced electric field E/N. Our Monte Carlo method, as usual, consists in following a large number of individual ions in a gaseous medium under the influence of an applied uniform electric field, E. The ions are assumed to change direction as the result of binary and instantaneous atomic collisions; between collisions they move in free parabolic paths due to the electric field.

Table 1
Potential parameters for the three lowest states of $HeAr^+$

	State		
	$X\ ^2\Sigma_{1/2}$	$A_1\ ^2\Pi_{3/2}$	$A_2\ ^2\Pi_{1/2}$
D_e (ev)	0.0350[a]	0.0200[b]	0.0230[a]
r_e (10^{-10} m)	2.565[a]	2.963[b]	2.872[a]
σ (10^{-10} m)	2.187[a]	2.593[b]	2.4861[a]
A (eV)	845.962	5710.83	1900.350
B (eV)	2.381724	3.911398	3.113334
b (10^{+10} m^{-1})	4.036878	4.421191	4.077286

[a]Spectroscopic, from Ref. [4].
[b]Model, from Ref. [7].

As the Ar^+ ions collide with the helium gas atoms with centre-of-mass energy ε, we use the previously calculated integral cross-sections, $Q_S(\varepsilon)$, and the null collision method [9] to determine the time, t, for the free path between collisions. The velocity of the helium gas atom for each collision is randomly sampled from a Maxwellian distribution function of the temperature. The angular differential scattering cross-section, $I_S(\varepsilon, \Theta)$, will determine the direction of the ion after the collision in the center of mass reference frame, the polar scattering angles Θ are sampled between [0, π], using the cumulative probability function $P(\varepsilon, \Theta)$ for the scattering of an ion with energy ε:

$$r = P(\varepsilon, \Theta) = \frac{\int_0^{\Theta} I_s(\varepsilon, \Theta') \sin\ \Theta'\, d\Theta'}{\int_0^{\pi} I_s(\varepsilon, \Theta') \sin\ \Theta'\, d\Theta'}$$

where r is a random number with uniform distribution in [0, 1]. The azimuthal scattering angle φ is sampled from a uniform distribution in [0, 2π].

4. Results and discussion

The Monte Carlo simulation was performed, using the above cross-sections, to study the drift of Ar^+ ions in gaseous helium at atmospheric pressure, $p = 760$ Torr, and temperature $T = 300$ K, under the influence of a uniform reduced electric field E/N in the 1–150 Td range (N is the neutral gas number density and 1 Td $= 10^{-17}$ V cm^2), i.e. electric field intensities from about 244.7 to 36,700 V cm^{-1}.

The transport properties of ions drifting in a neutral gas in the presence of an electric field are described by the following transport parameters: the drift velocity v_d and the mobility $K = v_d/E$ as well as the longitudinal D_L and transverse D_T diffusion coefficients, parallel and perpendicular to the direction of the electric field, respectively.

For the Ar^+–He system studied, we performed separate Monte Carlo calculations for each of the two ion states ($^2P_{3/2}$ and $^2P_{1/2}$) of Ar^+.

A sample of 3000 ions is allowed to drift under the applied electric field E for a time long enough (i.e. well above the relaxation time, which is 2 or 3 ns) so that the mean ion energy reaches a constant value and the average values of the axial position z, square of the radial position r^2, and axial position quadratic deviation have a linear variation with time, keeping constant with time the following parameters:

$$v_d = \frac{d\bar{z}}{dt}, \quad D_T = \frac{1}{4}\frac{d\overline{r^2}}{dt}, \quad D_L = \frac{1}{2}\frac{d(\overline{z^2} - \bar{z}^2)}{dt}.$$

Monte Carlo simulation results for Ar^+ ions (in the ground state $^2P_{3/2}$ and metastable excited state $^2P_{1/2}$) drifting in He gas are given in Table 2.

The reduced mobility K_0, normalized to the standard gas number density $N_0 = 2.687 \times 10^{19}\,cm^{-3}$, is obtained from the ion drift velocity by the relation

$$K_0 = \frac{v_d}{N_0(E/N)}.$$

In Fig. 2 we present the calculated mean energy and the reduced mobility K_0 for the ground state $^2P_{3/2}$ and the metastable excited state $^2P_{1/2}$ of Ar^+ ions in helium, as a function of the reduced electric field E/N for a gas temperature of 300 K. Except for some deviation at very low values of E/N, our Monte Carlo simulation calculations yield results in a very good agreement with the available mobility experimental data [10] to within 5% for all values (Fig. 2). The minor discrepancy may be due to inaccuracies in the interaction potential used in the cross-section calculations.

We point out that, for krypton and xenon ions in helium, different drift velocities have been reported for the ions in the $^2P_{3/2}$ and $^2P_{1/2}$ states [11], which result from different interaction potentials for the $^2P_{3/2}$ and $^2P_{1/2}$ ion states, with helium. For Ar^+ ions, we do not observe significant differences in the drift velocities for the two states.

The simulation results for the ratio of the longitudinal and transverse diffusion coefficients to the mobility, D_L/K and D_T/K, for $Ar^+(^2P_{3/2})$ ions in helium are presented in Fig. 3. At low reduced electric fields E/N, as it was expected, the ion–gas diffusion occurs at the same rate in directions parallel and perpendicular to the electric field, i.e. $D_L/K \approx D_T/K$.

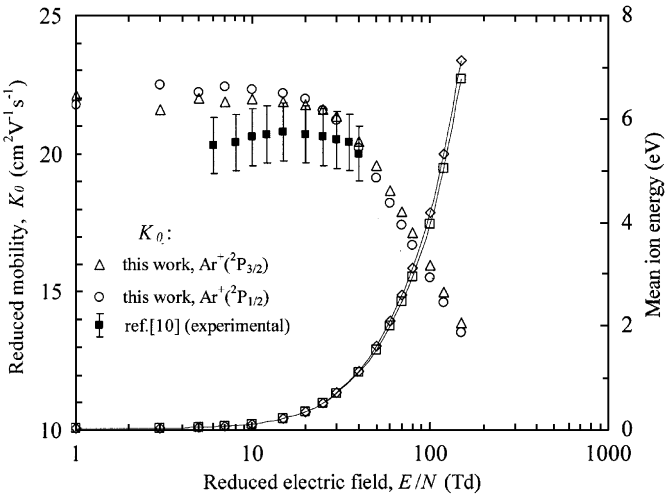

Fig. 2. Mean ion energy in eV and reduced mobility K_0 in $cm^2\,V^{-1}\,s^{-1}$ for Ar^+ ions in He as function of the reduced electric field E/N. Reduced mobility: this work, Monte Carlo simulation results for $T = 300$ K, (Δ) ground state ion $^2P_{3/2}$ and (○) metastable ion $^2P_{1/2}$; (■) experimental results [10] (error ± 5%). Mean ion energy: this work, (–◇–) $Ar^+(^2P_{3/2})$ and (–□–) $Ar^+(^2P_{1/2})$ ions in He.

Table 2
Monte Carlo simulation results of drift velocity v_d and diffusion coefficients D_L and D_T for the drift of atomic Ar^+ ions in the $^2P_{3/2}$ and $^2P_{1/2}$ state in He gaseous at 300 K and 760 Torr

E/N (Td)	$Ar^+(^2P_{3/2})$ ions in He			$Ar^+(^2P_{1/2})$ ions in He		
	v_d ($m\,s^{-1}$)	D_L ($cm^2\,s^{-1}$)	D_T ($cm^2\,s^{-1}$)	v_d ($m\,s^{-1}$)	D_L ($cm^2\,s^{-1}$)	D_T ($cm^2\,s^{-1}$)
1	59.45	0.614	0.620	58.51	0.614	0.617
3	173.94	0.611	0.618	181.40	0.642	0.614
5	295.60	0.655	0.605	298.32	0.665	0.645
7	411.31	0.698	0.646	421.82	0.696	0.628
10	590.18	0.754	0.720	600.13	0.811	0.679
15	881.39	0.995	0.788	894.62	1.022	0.776
20	1169.3	1.339	0.908	1180.0	1.197	0.930
25	1449.8	1.467	1.028	1448.6	1.378	1.023
30	1721.0	1.699	1.183	1710.0	1.576	1.229
40	2198.6	2.004	1.541	2169.2	1.830	1.447
50	2630.5	2.406	1.871	2571.0	2.140	1.820
60	3007.9	2.439	2.182	2937.0	2.342	1.966
70	3365.2	2.911	2.401	3275.2	2.550	2.293
80	3682.8	3.002	2.679	3587.6	2.743	2.519
100	4284.0	3.572	3.245	4167.4	3.343	3.087
120	4832.5	4.319	3.877	4701.0	3.918	3.489
150	5590.0	4.898	4.702	5450.0	4.758	4.175

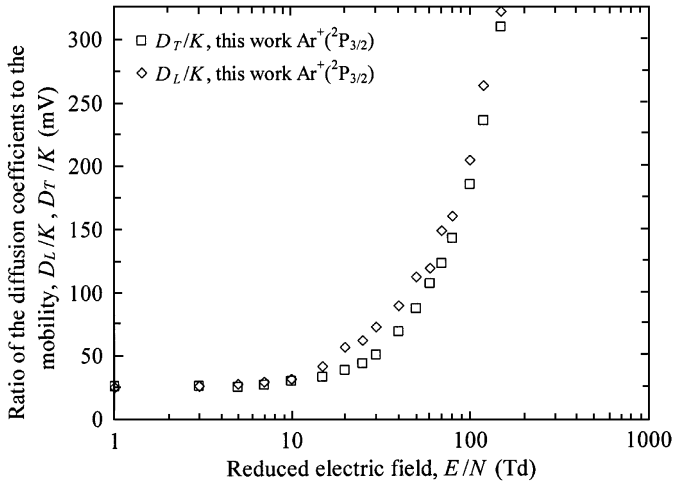

Fig. 3. Simulation result of the ratio of the longitudinal D_L/K (◇) and transverse D_T/K (□) diffusion coefficients to the mobility in mV, for $Ar^+(^2P_{3/2})$ ions in He at 300 K, as function of E/N.

Theoretically, as E/N tends to zero, both D_L/K and D_T/K approach the theoretical limit of 25.85 mV for $T = 300$ K as predicted by the Nernst–Townsend–Einstein relation [5].

Acknowledgments

Work supported by FEDER through Project POCTI/FP/FNU/50240/2003 from Fundação para a Ciência e para a Tecnologia, Portugal.

References

[1] T.H.V.T. Dias, F.P. Santos, A.D. Stauffer, C.A.N. Conde, Phys. Rev. A 48 (1993) 2887.
[2] J.A.S. Barata, C.A.N. Conde, Radiat. Phys. Chem. 71 (2004) 647.
[3] C. Yamada, H. Kanamori, E. Hirota, J. Chem. Phys. 83 (1985) 552.
[4] P.E. Siska, J. Chem. Phys. 85 (1986) 7497.
[5] E.A. Mason, E.W. McDaniel, Transport Properties of Ions in Gases, Wiley, New York, 1988.
[6] V.P. Shevelko, Atoms and their Spectroscopic Properties, Springer, Berlin, Heidelberg, 1997.
[7] D. Hausamann, H. Morgner, Mol. Phys. 54 (1985) 1085.
[8] C.J. Joachain, Quantum Collision Theory, third ed., North-Holland Publishing, Amsterdam, 1983.
[9] H.R. Skullerud, J. Phys. D 1 (1968) 1567.
[10] L.A. Viehland, E.A. Mason, At. Data Nucl. Data Tables 60 (1995) 37.
[11] B.K. Bluhm, S.W. North, D.H. Russell, J. Chem. Phys. 114 (2001) 1709.