Molecular dynamics simulation of gaseous ion-motion in electrostatic fields

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A molecular dynamics (MD) method has been developed for the simulation of the motion of ions in neutral gases under the influence of homogeneous electrostatic fields. The method treats the translational motion of the ions and gas molecules classically and thus requires as input the ion–molecule interaction potentials. The continuous dissipation of a part of the ion-energy to a thermal bath is accomplished through the introduction of ''iconical interactions'' between ions and images of the neutrals created and stored in the memory of a computer during ion–atom encounters. The steady ion motion is then simulated by usual equilibrium MD methods using periodic boundary conditions. The resulting ion mobilities, effective temperatures, and third other velocity-distribution moments, expressed as skewness parameters, compare well with available results derived from the moment solution of Boltzmann equation and Monte Carlo simulations using the same interaction potentials in the cases of K^+ in He and Ar, as well as of Ba^+ in Ar. The additional reproduction of experimental data provides an independent test for the accuracy of the employed model potentials. Although the method has been applied to atomic systems it is easily extendable to the more complex molecular systems but at the expense of computer time.

I. INTRODUCTION

The motion of ions in neutral gases under the influence of homogeneous electrostatic fields has been studied extensively over the last decades, due primarily to the advent of drift tube mass spectrometers which enabled the production and accurate characterization of steady ion currents in buffer gases.[1] Thus, many ion–gas combinations have been studied experimentally and the corresponding measured ion mobilities and diffusion coefficients have been compiled so far.[2–5] Further, the implementation of laser-induced fluorescence in drift tube experiments enabled the determination of mean ion-kinetic energies parallel, perpendicular, and at intermediate angles to the field as well as of third and fourth order moments of the ion velocity distribution,[6,7] though for selective systems only. In similar studies, the population distribution of rotational states and alignment of N_2^+ in drift tubes[8] has been probed. In addition, vibrational deactivation[9] and chemical rates of the drifting ions[1,10] have been measured, though with less accuracy.

The interpretation of the transport data has been achieved through the moment solution of Boltzmann-type kinetic equations but has been restricted mainly to atomic systems with the exception of Viehland and co-workers on Li^+ in N_2 and CO.[11] Recently, attempts are made for the application of the analytic theory to molecular ion systems.[12] Parallel to the analytic theory, Monte Carlo methods[13,14] have been developed which can also provide transport properties from cross sections. Such methods depend on stochastic models for the determination of free flight times of the ions.

An alternative approach to the simulation of the ion-motion independent of stochastic models and thus easily extendable to molecular systems is provided by molecular dynamics (MD). A new such procedure is presented in this work. Here we will primarily be concerned with atomic systems and the three first moments of the ion velocity distribution function, from which the ion-mobility, mean kinetic energies or effective temperatures and skewness parameters are determined. The method requires as input the external conditions that characterize the steady motion of the ions in drift tubes, that is the gas temperature T and the ratio of the field strength to the gas number density E/N, as well as the ion–molecule and molecule–molecule interaction potentials; though the interactions between the neutral molecules do not affect the results of the simulation significantly. Although the procedure can be extended straightforwardly to the case of molecular ions, results for such systems are deferred to a future application.

Normally, such a realistic reproduction of the ion-motion which follows the evolution of the system of ions and molecules through integration of Newton's equations would require the introduction of a thermal bath, in order for the gas temperature to remain constant throughout the simulation. This is so, because a part of the energy of the ions which is gained from the electric field is continuously dissipated to the gas through the ion–gas collisions.

Instead, to avoid complications related to continuous control of the state of the gas which would slow down the procedure, we prefer to maintain the gas at equilibrium by not allowing the gas molecules to feel the ions. What is crucial however, is for the ions to feel the forces of the gas molecules. Such a situation can been implemented in a computer through the introduction of ''iconical interaction,'' that is, interactions of ions with images of the neutral molecules which are created during collisions. These considerations are consistent with drift tube experiments where tenuous amounts of ions are introduced into the buffer gas, with very small relative density, thus allowing efficient transfer of ther-

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mal energy to a bath without disturbing the gas from equilibrium.

As prototype systems for testing the reproduction of ion-mobility we have chosen K^+ in He and in Ar because they both have been extensively studied experimentally, as well as through the analytic theory and by stochastic simulations using the same ion–atom interaction potentials. In addition to the reproduction of the mobility, the determination of the next two higher moments of the *v*-distribution, expressed as effective temperatures and skewness parameters, respectively, have been tested against results of analytic calculations and experimental data for Ba^+ in Ar.

The results of the present procedure can converge to any predetermined accuracy but improvements of convergence below a few percent require expense of large amounts of computer time. We find that a workable accuracy is within ±2.5% for the mobility and 5% for higher moments. These errors are comparable to convergence limits of the analytic theory (generally 0.2% and 2%, respectively, but in some cases as much as ~1% and ~5%), and of stochastic simulations (~1% and ~2%), as well as to experimental errors (2%–5% and ~5%), as to allow meaningful comparisons for consistency with theoretical calculations and measurements to be drawn. The overall consistency is found to be good and in line with experimental data, as it should be, since the interaction potential used in the calculations were determined primarily from inversion of mobility data.

We proceed by first presenting the developed molecular dynamics procedure and the conditions of its operation. Second, the results for the selected ion–atom test systems are presented and compared to theoretical results and experimental data. Finally, in the conclusions we comment on the possibility of the extension of the current procedure to more complex systems with internal degrees of freedom.

II. MOLECULAR DYNAMICS METHOD

We are interested in simulating drift tube experiments in which a small amount of ions is moving through a low density neutral gas under the influence of a homogeneous electrostatic field. The ions after being injected in the tube follow a short relaxing stage and finally attain steady motion during which the energy they acquire from the field is continuously dissipated, through the ion-neutral collisions, to the buffer gas at steady rate. The gas, in turn, efficiently transfers the heat to a thermal bath without being disturbed from equilibrium. This is so, because usually the ratio of the ion density to the neutral one is of order 10^{10}.[1] Since in computer experiments a reasonable value of this ratio is quite smaller, of the order of 10^3 to 10^4, it is impossible to mimic the real dissipation mechanism in a simulation and thus an effective method has to be introduced for this purpose. Although control mechanisms have been developed and used in nonequilibrium molecular dynamics simulations especially for dense fluids,[15] we do not need to invoke them here because the experimental conditions described above allow us to develop a simpler, more efficient and in addition realistic technique. First, the use of small amounts of ions in experiments allows us to ignore safely the Coulombic interactions between the ions and assume them independent during their steady motion. Second, the fast restoration of equilibrium Gaussian *v*-distribution of the gas indicates that the motion of the gas molecules could be simulated independently from the ions with an equilibrium MD method, provided the ions interact through an effective mechanism with them. For this reason we choose to simulate the gas without adding to the forces felt by the neutral molecules the forces due to the presence of the ions. However, since the neutral molecules will not be affected by the presence of ions, it is obvious that the ions should not feel the forces of *these molecules* either, otherwise the collisions would not be realistic. A way out from this contradiction is achieved by not allowing the ions to interact directly with the gas molecules, but only with images of them generated in the memory of the computer at the starting point of each ion–molecule encounter. The images evolve independently from other ions and gas molecules as long as the interaction takes place, and at the end of the collision they are erased from the memory of the computer. These interactions, named here ''iconical interactions,'' allow the efficient loss of excess energy from the system in a realistic way.

In support to the above considerations, we mention that similar assumptions have been utilized in the moment solution of Boltzmann-type kinetic equations. There, the interactions between the ions have been neglected and the *v*-distribution for the gas has been considered spherical and Gaussian. In addition, the theory itself is based on the assumptions that only two-body interactions take place and that consecutive collisions are uncorrelated. The latter restrictions for the ion–gas molecule collisions have also been introduced into our method indirectly through the iconical interactions which exclude multiple scattering and correlation between the ions and the gas. We should thus expect the present procedure to be consistent with the analytic theory, and since at the very low density experimental conditions the above considerations are reasonable, the present method should reproduce, in addition, the experimental transport data successfully.

To explain the technical role of the iconical interactions, we can say that at each instant the MD simulation of the gas provides initial conditions for the exact scattering of the ions. As designed, these interactions allow the use of usual MD methods for the integration of neutral and ion trajectories, such as the Verlet algorithm[16] with periodic boundary conditions, even though the ion motion has cylindrical symmetry.

Despite the above simplifications, two other problems have to be overcome. First, the low density of the gas requires very long computer runs, therefore a scaling with respect to the density should be looked for. Fortunately, the ion-transport properties at low gas densities depend on the ratio of the field strength to the gas density, E/N. Thus higher densities than the experimental ones can be employed in the simulation, provided the field strength is raised proportionally. In the present calculation we have increased the density by three orders of magnitude, in order to increase the speed of the computation by one order of magnitude.

The second problem is the runaway phenomenon[1] that occurs at high field strengths, where the decrease of scattering cross sections allows the ions to accelerate continuously

TABLE I. Potential parameters[a] for alkali ion–noble gas interactions, in atomic units (a_0=0.529 177 25 Å).

System	$\upsilon(e^2/a_0)$	$\varrho(a_0)$	$R_m(a_0)$	$a_d(a_0^3)$	$a_q(a_0^5)$	$a_0(a_0^7)$	$C_6(e^2a_0^5)$	$C_8(e^2a_0^5)$
K^+–He	0.2690	0.8589	5.49	1.3831	2.4434	10.614	5.835	72.05
K^+–Ar	1.202	0.9478	5.87	11.08	54.14	531.2	38.97	741.8

[a] The parameters are taken from Ref. 17.

and gain high energies. Such events, although rare, can occur during a simulation below the actual instability point. To exclude them from the statistics, in cases where the real runaway instability has not been reached, we have set a velocity-cutoff, after which the velocity of a runaway ion is reduced to the instantaneous drift velocity of the rest of the ions. This approach allows us to obtain results safely even at very high field strengths.

We have to mention also that the classical behavior of the systems is crucial for the applicability of a MD technique. However, since quantum effects are known to contribute to the transport properties only at low temperatures, below 40 K, and for species of low mass, such as H and He,[1] the range of application of the present technique should remain wide.

We turn now to testing the accuracy of the results of the developed molecular dynamics method.

III. TESTS OF THE METHOD

We choose to test our method by studying the reproduction of the three first moments of the ion *v*-distribution against calculated and experimentally measured data. Specifically, the first moment provides the ion-mobility defined through $K=v_d/E$, where the drift velocity, $v_d=\langle v_z\rangle$, is the mean velocity in the direction of the field. The second moments provide mean kinetic energies parallel and perpendicular to the field, expressed here as effective temperatures, $T_i=(m/2k_B)(v_i-\langle v_i\rangle)^2$, where the subscript indicates the direction of the motion parallel or perpendicular to the field and k_B is Boltzmann constant. Finally, the third moment is expressed as skewness parameter,

$$\delta=[(v_z-\langle v_z\rangle)^3]^{1/3}/(\langle v_z^2\rangle-\langle v_z\rangle^2)^{1/2}, \tag{1}$$

defined only in the direction of the field, because the parameters perpendicular to the field vanish due to the cylindrical symmetry of the ion velocity distribution.

We test the results of the method against ion mobilities of K^+ in He and Ar, calculated through the moment solution of Boltzmann equation and by Monte Carlo stochastic simulation, as well as, against ion mobilities, effective temperatures and skewness parameters for Ba^+ in Ar calculated through the analytic theory. All these quantities have also been measured experimentally and relevant comparisons against experimental data essentially provide new tests for the employed interaction potentials.

We have simulated all systems with 500 ions in 108 neutral molecules using the Verlet algorithm for the integration of the equations of motion, since the simplicity of the system does not necessitate the use of a more elaborate and thus slower algorithm. The length of the runs was from 10^5 to 2×10^5 steps of 10^{-14} s in order to achieve convergence within a few percent in mobility and effective temperatures. In the case of the skewness parameter twice as many steps were required to achieve the same consistency. The overall accuracy of the results was established by using different initial conditions, by doubling the length of the simulation, and by increasing the number of ions and neutral molecules. Better accuracies can in principle be achieved by extending these operational variables.

The neutrals were first simulated over 50 000 steps with microcanonical conditions starting from a fcc configuration and then were set free to evolve in parallel with the ions without control of their temperature, though their state was continuously monitored throughout the simulation. We observed that for a few hundred of thousands of iterations the gas remained at the preestablished equilibrium. Finally, the ions initially were randomly positioned in the unit cell with Gaussian *v*-distribution of the gas temperature. They have to evolve for roughly 30 000 steps in order to relax to a steady state and thus averages for the ion properties were collected after 40 000 steps. The velocity-cutoff was set at 20 000 m/s after experimentation with simulations at the experimental high field limit under the provision that only a few ions had to cross the velocity limit during the whole duration of the simulation.

A. K^+ in He and Ar

The transport properties of K^+ in He and Ar has been calculated in the pasts through the analytic theory[17] and MC simulation[18,19] using in both cases the KMV model potentials[17] among others. In order to study the consistency of our MD results on ion-mobilities with the results of these methods, we employ the same ion–atom interaction potentials for the simulations. The model potential consists of two parts, one representing effectively exchange interactions and the other Coulombic interactions,

$$V(R)=V_{\rm ex}(R)+V_c(R). \tag{2}$$

The Coulombic part of the potential, in turn, is described by a damped truncated perturbation expansion for the induction and dispersion energies,

$$V_c(R)=[V_{\rm ind}(R)+V_{\rm dis}(R)]h(R), \tag{3}$$

where, in atomic units,

$$V_{\rm ind}(R)=e^2a_d/2R^4+e^2a_q/2R^6+e^2a_o/2R^8,$$

$$V_{\rm dis}(R)=C_6/R^6+C_8/R^8,$$

and

TABLE II. Mobility and effective temperatures for K^+ in He at 300 K calculated at various field strengths by molecular dynamics simulation.

E/N(Td)	v_d(m/s)	K_0(cm^2/V s)	$T_\parallel$(K)	$T_\perp$(K)
10	600	22.3	381	314
15	892	22.1	450	355
30	1716	21.3	869	552
60	3002	18.6	2008	1086
80	3690	17.2	2813	1500
100	4306	16.0	3388	2057
120	4840	15.0	4520	2529
150	5624	14.0	5706	3515
200	6563	12.2	8651	4527
400	8719	8.11	26450	19340

$$h(R)=\begin{cases}\exp(1.28R_m/R-1)^2 & \text{for } R<1.28R_m\\ 1 \quad \text{for } R>1.28R_m, & \end{cases}$$

where R_m is the position of the potential minimum, C_i are dispersion coefficients, and a_n are n-pole polarizabilities of the atoms (d=dipole, q=quadrupole, and o=octopole). Finally, the exchange energy is determined by two unitless empirical parameters υ and ϱ through $V_{\text{ex}}(R)=\upsilon V^*_{\text{ex}}(R/\varrho)$, where V^*_{ex} is a universal function of form $V^*_{\text{ex}}(R)=Ae^{-\alpha R}-Be^{-\beta R}$, with $A=146.98$, $B=70.198$, $\alpha=1.5024$, and $\beta=1.4041$. All model parameters are presented in Table I.

For the simulation of the noble gases at 300 K we have employed Lennard-Jones (12-6) potentials because they are simpler and faster in the computation than other more elaborate available ones. The quality of the calculations, however, is not affected by this simplification, because at low densities the gases behave ideally and thus their state is not sensitive to small variations in the interaction potential. The employed potential parameters for the He gas are $\epsilon=10.22$ K and $\sigma=2.556$ Å,[20] and for the Ar gas are $\epsilon=119.7$ K and $\sigma=3.405$ Å.[21] The resulting standard mobilities, that is mobilities reduced at standard gas conditions through $K_0=K(273.15\ \text{K}/T)(P/101325\ \text{Pa})$, at various field strengths represented as E/N in Td, ($1\ \text{Td}=10^{-21}\ \text{V m}^2$), are reported in Tables II and III. In addition, we report the resulting effective temperatures parallel and perpendicular to field for future comparisons. The attained convergence in the mobility, (±2.5%), at almost all the range of field strengths is good enough to allow safe conclusions for consistency to be drawn. Indeed, as can be inferred from Figs. 1 and 2, the three approaches coincide with each other and all together with the experimental mobilities.[2,5,22] The latter comparisons, actually, provide a test for the accuracy of the ion–atom interaction potentials.

We mention also, that at very low field strengths the results did not converge to the prescribed accuracy, due to large ion-velocity fluctuations relative to the drift velocity which tends to zero at vanishing field strengths. Although improved accuracy can be achieved by extending the length of the simulation, the low quality of the results can be compensated by the fact that, in this case the mobility is almost independent from the field.

TABLE III. Mobility and effective temperatures for K^+ in Ar at 300 K at various field strengths calculated by molecular dynamics simulation.

E/N(Td)	v_d(m/s)	K_0(cm^2/V s)	$T_\parallel$(K)	$T_\perp$(K)
15	104	2.58	335	312
30	223	2.76	422	364
60	465	2.89	841	589
100	849	3.16	2026	1269
150	1311	3.25	4146	2419
200	1673	3.11	6030	3853
300	2287	2.84	10188	6793
400	2748	2.55	14199	9933
500	3278	2.44	22060	15236
600	3579	2.22	29910	18804

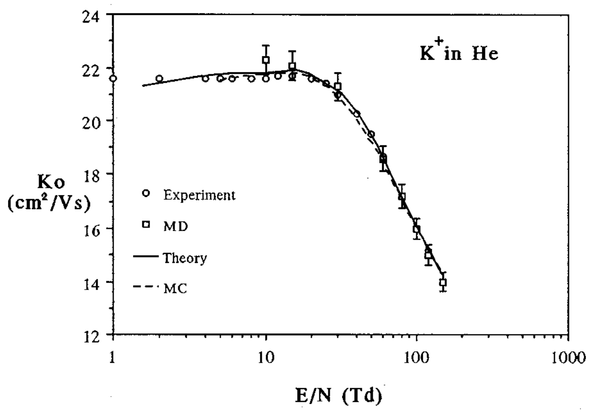

FIG. 1. Standard mobility of K^+ in He at 300 K. Circles are experimental data from Ref. 2, squares with 2.5% error bars are present MD results, the solid line has been calculated through kinetic theory, Ref. 17, and the dotted line represents Monte Carlo simulations, Ref. 18.

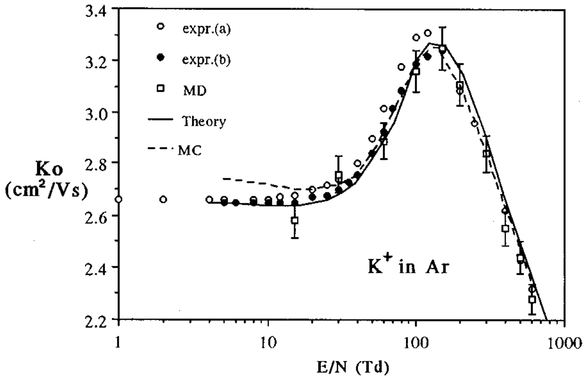

FIG. 2. Standard mobility of K^+ in Ar at 300 K. Circles represent experimental data from Ref. 2, (open circles), and from Refs. 5 and 22, (filled circles), squares with 2.5% error bars are present MD results, the solid line has been calculated through kinetic theory, Ref. 17, and the dotted line has been determined from Monte Carlo simulations, Ref. 19.

TABLE IV. Transport properties for Ba^+ in Ar at 305 K calculated at various field strengths by molecular dynamics simulation using a (7.5, 6, 4) model potential proposed by Viehland and Hampt.[a]

E/N(Td)	v_d(m/s)	K_0(cm^2/V s)	$T_\parallel$(K)	$T_\perp$(K)	δ
20	98	1.82	343	315	0.422
40	204	1.90	404	348	0.451
60	313	1.94	585	412	0.699
80	445	2.07	897	554	0.821
90	508	2.10	1054	611	0.810
120	765	2.37	2027	1011	0.871
130	826	2.36	2291	1101	0.876
150	987	2.45	2889	1452	0.837
170	1105	2.42	3413	1741	0.808
200	1290	2.40	4114	2245	0.805
300	1834	2.28	6842	3996	0.783
400	2233	2.08	9353	5538	0.783

[a] Reference 24.

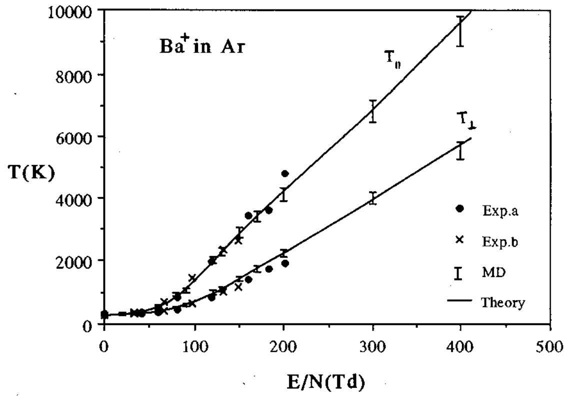

FIG. 4. Effective ion-temperatures parallel (upper curve) and perpendicular (lower curve) to the field for Ba^+ in Ar at 305 K. Circles and crosses are experimental data from Refs. 7(a) and 7(b), respectively, squares with 5% error bars are present MD results, and the solid line has been calculated through kinetic theory, Ref. 23.

B. Ba^+ in Ar

Data for testing the results of the simulation of Ba^+ in Ar consist of mobilities, effective temperatures parallel and perpendicular to the field, as well as of skewness parameters, which all have been calculated through the moment solution of the Boltzmann equation.[23] As an ion–atom interaction potential we employ the one used in the analytic calculations. This is a (n, 6, 4) potential, with $n=7.5$, $\gamma=0.7137$, $\epsilon=0.00351$ (e^2/a_0), and $R_m=6.5\ a_0$, which has been determined by Viehland and Hampt[24] under the provision to reproduce accurately the experimental transport data through the kinetic theory. Finally, for the atom–atom interactions we have employed the same Lennard-Jones potential which has been presented in the previous subsection.

The results for Ba^+ in Ar, summarized in Table IV, have converged within 2.5% for the mobility and within 5% for the effective temperatures and the skewness parameter. The comparisons against the analytic results and the experimental data,[7] which are presented in Figs. 3–5, indicate consistency between the two methods for all first three moments of the ion v-distribution and, in addition, problematic reproduction of experimental T's and δ at high fields. This inconsistency has been attributed in the past to possible interference of higher electronic states to the Ba^+–Ar interactions. Further tests with model potentials of more flexible functional form could strengthen this assertion.

IV. CONCLUSIONS

We have presented a novel molecular dynamics technique for the simulation of the gaseous ion transport under the influence of an external electric field. The main feature of this method is the dissipation mechanism, which has been introduced in order for the excess ion-energy acquired from the electric field to be dissipated as heat in the gas, realistically and efficiently for the computation. This is achieved through the iconical interactions between the ions and images of the neutrals, created and stored in the memory of a computer as long as the collisions take place. The buffer gas

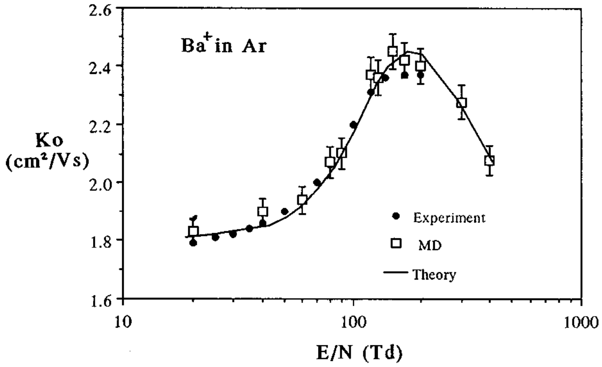

FIG. 3. Standard mobility of Ba^+ in Ar at 305 K. Circles are experimental data from Ref. 5, squares with 2.5% error bars are present MD results, and the solid line has been calculated through kinetic theory, Ref. 23.

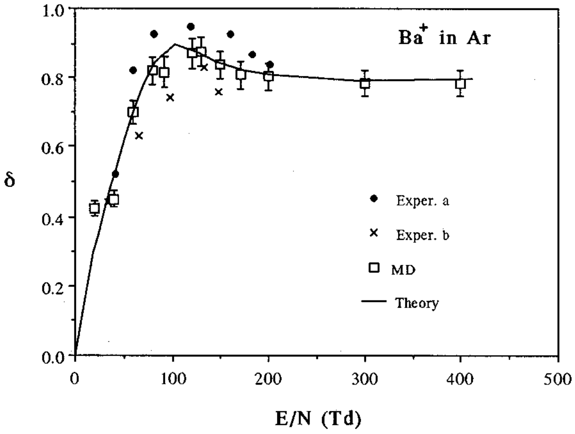

FIG. 5. Skewness parameters for Ba^+ in Ar at 305 K. Circles and crosses are experimental data from Refs. 7(a) and 7(b), respectively, squares with 5% error bars are present MD results, and the solid line has been calculated through kinetic theory, Ref. 23.

remains always in equilibrium by not allowing the neutral atoms to interact directly with the ions. The whole scheme preserves the periodic boundary conditions, despite the cylindrical symmetry of the ion-motion due to the presence of the electric field.

The comparisons of the calculated ion-mobilities, effective temperatures, and skewness parameters against results of the analytic theory and the Monte Carlo simulations using the same interaction potentials reveals the consistency of all approaches. In addition, the reproduction of the corresponding experimental data provides an independent test for the accuracy of the employed ion–atom interaction potentials.

The two simulation methods and the analytic theory, however, differ with respect to the accuracy they attain and the hardware they require for efficient computations. In general, the two simulation methods can become comparable in accuracy and both require the use of a supercomputer, although calculations based on the analytic theory can be performed more easily on workstations and can attain higher accuracies. The main advantage, however, of the simulations may lie in their easy extension to more complex systems with internal degrees of freedom, though with the expense of an even higher amount of computer time.

Here we have been concerned with the first three moments of the ion v-velocity distribution, however, the present method is easily extendable to allow for the calculation of other moments as well as of ion-autocorrelation functions and diffusion coefficients. In addition, the method can easily be extended to account for internal degrees of freedom, such as the rotational motion, of both ions and gas molecules. This is so because the iconical interactions can easily be added to the well developed equilibrium molecular dynamics methods that treat rotational motion of molecular species. Work in this direction is under way.

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