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# Modeling elastic momentum transfer cross-sections from mobility data

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**Abstract** – In this letter we present a new method to simply obtain the elastic momentum transfer cross-section which predicts a maximum of reduced mobility and its sensitivity to the temperature variation at low energies. We first determined the transport cross-section which resembles mobility data for similar closed-shell systems by using the Monte Carlo method. Second, we selected the most probable reactive processes and compiled cross-sections from experimental and theoretical data. At the end, an elastic momentum transfer cross-section is obtained by subtracting the compiled cross-sections from the momentum transfer cross-section, taking into account the effects of the angular scattering distributions. Finally, the cross-section set determined in such a way is used as an input in a final Monte Carlo code run, to calculate the flux and bulk reduced mobility for  $\text{Ne}^+ + \text{CF}_4$  which were discussed as functions of the reduced electric field  $E/N$  ( $N$  is the gas density) for the temperature  $T = 300$  K.

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**Introduction.** – Cold plasmas are often exploited in new technologies where they favorably offer non-intrusive production or modification of various substances [1]. The main characteristics of these plasmas are their high electron temperature and low gas temperature where the non-equilibrium behavior of a large number of species takes place [2]. The present computer resources allow studies of complex global models [3,4] describing the behavior of such plasmas by taking into account a very large number of particles. In such cases the knowledge of the ion-neutral reaction and transport parameters for ions resurges as interesting mostly in cases in which reactive processes take place [5–7]. In spite of great efforts and numerous results [8–15], the knowledge about ion-molecule reactions holding a prominent place in the descriptions of these plasmas, is far from satisfactory. The main reason for that are the poorly known ion scattering properties in various gases that must be guessed in order to obtain transport properties during modelling [12,16–18].

Quantum-mechanical calculation of the ion-neutral scattering cross-sections requires the knowledge of accurate potential energy surfaces which are known only to within limits of appropriate (usually very complex) theories [19], while experiments at low energies are sensitive to small stray fields and many other technical problems.

Thus, simple and effective methods based on known principles are highly valuable.

**Calculation of the cross-section.** – Due to the lack of experimental and theoretical mobility data Gatland *et al.* [20] found that all the experimental mobility curves can be unified into a single mobility curve by using a model interaction potential. Their interaction potential predicts a peak in reduced mobility ( $K_0$ ) and suggests generalized mobility curves for alkali ions in rare gases (closed-shell systems). The generalized mobility curves are defined as  $K_0$  normalized by a polarization limit (PL) value  $K_{\text{PL}}$  [6,20] as a function of the effective temperature normalized to the well depth of the potential minimum. Takebe [21] made a further step in the unification of mobility data. Apart from similar conclusions as in [20], *e.g.* that the ratio  $K_{\text{max}}/K_{\text{PL}}$  dominantly depends on the observed ion, he showed that transport of ions proceeds in the presence of clustering collisions and found out that the activation energy for the reversible process accounts for half of the potential well depth. Takebe's reduced mobilities in the  $T \rightarrow 0$  limit were slightly above the PL values. Takebe's approach is justified only for closed-shell systems but presents a good base for upgrading for more complex systems. By following a vast amount of data in [10] one

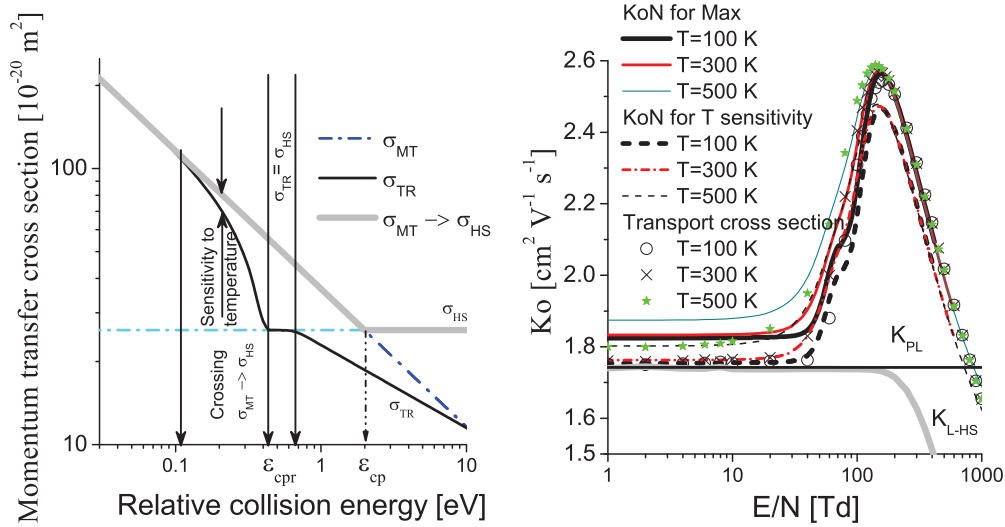


Fig. 1: (Colour online) (a) Transport cross-sections as a function of relative collision energy and (b) reduced mobility as a function of  $E/N$  where temperature  $T$  is used as a parameter.

may conclude that for a broad range of systems mobility behaves similarly to closed-shell systems, *i.e.*, where, apart from the association reaction, other reactions are negligible and the dominant effect on transport comes from the elastic momentum transfer cross-section. Since at low pressures the association cross-section is few percent of the elastic momentum transfer cross-section and is dominant at low collision energies, at  $T \rightarrow 0$ ,  $E \rightarrow 0$  affects the reduced mobility which goes to values slightly higher than the polarization limit. For the same reasons the association will not significantly affect the mobility peak. We then exploited a hard-sphere cross-section in order to define the maximum cross-section for reactive collisions.

#### Modelling the transport cross-section by mobility curve.

In order to model the transport cross-section one may exploit either numerical solutions of Boltzmann's kinetic equation or the stochastic Monte Carlo (MC) method [22, 23]. In this work we used the well-tested MC code [23].

Central information about the interaction is drawn from the differential cross-section with all the possible quantum-mechanical effects stored within, not exclusively from interaction potentials. Thus, in the MC modeling the momentum of the transfer cross-section  $\sigma_{\text{MT}}$  [22, 24–26] with the assumption of isotropic scattering in the center-of-mass frame is very often used. The idea behind is the need to properly account for ion momentum losses which also appear in balance equations [12] and are desired in modeling many technologically interesting cases. Ion mobility is exactly described with  $\sigma_{\text{MT}}$ , at low energies where the average distance at which the ion and the molecule collide is within the range of the ion-induced dipole potential  $\sim r^{-4}$ . The same potential for the reaction coefficient gives a constant value calculated from the known gas polarizability and reduced mass of ion and gas.

Average polarizabilities for most gaseous atoms/molecules are generally known [27] so simple approximations

based on them are useful for a wide range of systems. For  $r^{-4}$  the potential momentum transfer cross-section  $\sigma_{\text{MT}} = \sigma_{\text{DL}} = 1.105 \cdot \sigma_{\text{L}}$  has the same energy dependence ( $\sim \epsilon^{-0.5}$ ) as Langevin's cross-section  $\sigma_{\text{L}}$  [17] and a constant mobility (see solid line denoted by  $K_{\text{PL}}$  in fig. 1(b)) is obtained analytically by the so-called mean-free time theories such as, for example, the theory in [22]. In this case the obtained mobility is not a function of temperature and pressure [6, 26] and is taken as a limiting case for the polarization attraction between ion and neutral [17]. If for the same potential, Langevin's cross-section is used either partially or fully to describe the loss of particles, for example, the association reaction [5] reduced mobility increases. Generally inelastic collisions cause the decrease of mobility [28] while reactive collisions increase the mobility [6] which in general can be limited by the possibility of inverse reactions. Both reduced mobility and reaction rate coefficients are easily calculated by the MC code [24], where  $\sigma_{\text{MT}}$  is used as previously explained. The calculated values are precise (reproducible) to a few significant digits depending on the computing resources [4, 24].

A hard-sphere (HS) cross-section  $\sigma_{\text{HS}}$  [29, 30] is often used to represent ion transport [24, 30] at high collision energies (see fig. 1(a)) where actually the repulsive nature of collisions becomes dominant. Scattering in this case, described by a potential  $\sim r^{-n}$  ( $n \rightarrow \infty$ ), is isotropic in the center-of-mass frame [24] and is exactly represented by  $\sigma_{\text{MT}} = \sigma_{\text{HS}}$  in Monte Carlo codes. At the same time  $\sigma_{\text{HS}}$  is used as a starting point in theories of ion reactions, for example as a good approximation in cases in which processes of charge transfer dominate [29, 31] so it can be a potentially good approximation for more complex cases. If one crudely joins the above-mentioned approximations simply by using  $\sigma_{\text{MT}} = 1.105 \cdot \sigma_{\text{L}}$  below  $\epsilon_{\text{cp}}$  and  $\sigma_{\text{MT}} = \sigma_{\text{HS}}$  above  $\epsilon_{\text{cp}}$ , trying to describe ion mobility in a wider energy range (see thick solid line in fig. 1(a)) than the MC

Table 1: Characteristic values for mobility peaks with respect to polarization limit values in percent [10,33] and appropriate  $E/N$  values in Td ( $1 \text{ Td} = 10^{-17} \text{ Vcm}^2$ ).

		$^4\text{He}$	$^{20}\text{Ne}$	$^{40}\text{Ar}$	$^{84}\text{Kr}$	$^{131}\text{Xe}$	$^{222}\text{Rn}$
$^7\text{Li}^+$	$K_{\text{omax}}/K_{\text{opL}}(\%)$	71	59	62	66	65	–
	$E/N \text{ (Td)}$	48	50	117	112	144	–
$^{11}\text{Be}^+$	$K_{\text{omax}}/K_{\text{opL}}(\%)$	47	47	68	61	52	52
	$E/N \text{ (Td)}$	30	40	165	190	265	263
$^{23}\text{Na}^+$	$K_{\text{omax}}/K_{\text{opL}}(\%)$	57	42	46	47	50	56
	$E/N \text{ (Td)}$	34	53	151	156	156	174
$^{27}\text{Al}^+$	$K_{\text{omax}}/K_{\text{opL}}(\%)$	37	35	37	43	40	41
	$E/N \text{ (Td)}$	20	38	129	173	213	223
$^{39}\text{K}^+$	$K_{\text{omax}}/K_{\text{opL}}(\%)$	36	27	33	34	37	43
	$E/N \text{ (Td)}$	27	44	131	155	185	171
$^{40}\text{Ca}^+$	$K_{\text{omax}}/K_{\text{opL}}(\%)$	36	27	33	34	37	43
	$E/N \text{ (Td)}$	27	44	131	155	185	171

code [22,23] which properly takes into account thermal collisions [10], gives a reduced mobility in fig. 1(b) (solid line labelled by  $K_{\text{L-HS}}$ ).  $K_{\text{L-HS}}$  at  $T = 300 \text{ K}$  overlaps the PL curve and is a uniformly decreasing function at high average energies.

It is commonly accepted that the mobility peak represents the average energy where attractive intermolecular forces balance repulsive forces [24,32]. The height of the peak is best represented with respect to the polarization limit value, while the  $E/N$ 's scale can be well normalized by the well depth of the interaction potential [20,21].

In table 1 are shown the mobility peak values obtained from the newest theoretical data for the reduced ion mobility for closed-shell systems [10] which are the best fit to all the existing experimental data, all normalized at the polarization limit value ( $K_{\text{omax}}/K_{\text{PL}}$ ). It is evident that  $K_{\text{omax}}$  mainly depends on ion species in closed-shell systems, so one may pull a more general conclusion that  $K_{\text{omax}}/K_{\text{PL}}$  decreases with the increase of the ion atomic number in inert gases, and even in minor cases a small discrepancy exists from such average trend. With the increase of the atomic number  $K_{\text{omax}}/K_{\text{PL}}$  are at higher  $E/N$ 's since less collision energy is transferred from the laboratory system with lighter ions to the gas for the similar effect.

For our test case ( $^{20}\text{Ne}^+ + ^{88}\text{CF}_4$ ) we chose  $^{23}\text{Na}^+ + ^{84}\text{Kr}$  as nearest closed-shell system due to its similarity with the atomic number and polarizability value.

From table 1, one may see that the mobility peak for the  $\text{Na}^+$  ion is 46% relatively to the polarization limit data and is characteristic not only for Kr [21] but also for all closed-shell atoms. Thus, by analogy, the mobility peak for  $\text{Ne}^+ + \text{CF}_4$  should be very similar to the peak for  $\text{Na}^+ + \text{Kr}$  if one neglects the different reactivity of these two systems. For  $\text{Na}^+ + \text{Kr}$  both interaction potential and bulk of the experimental measurements for

the transport coefficients are available [33–36], so in the following we will refer to these data in order to model the mobility of  $\text{Ne}^+$  in  $\text{CF}_4$ . The theoretical data for  $K_{\text{O}}$  from [36] (see also [10]) give an excellent fit of the data in [33,34] at  $T = 300 \text{ K}$  so as to be used also for the description of the temperature variation of the reduced mobility. We selected data for temperatures  $T = 100 \text{ K}$ ,  $300 \text{ K}$  and  $500 \text{ K}$  and normalized them at the  $\text{Ne}^+ + \text{CF}_4$  polarization limit (solid lines denoted as “ $K_{\text{O}}N$  for Max” in fig. 1(b)).  $\text{Ne}^+$  has slightly lower atomic number than  $\text{Na}^+$  and so slightly higher  $K_{\text{omax}}/K_{\text{PL}}$  for which we used 49% at  $E/N = 155 \text{ Td}$ .

*Most probable processes in  $\text{Ne}^+ + \text{CF}_4$  scattering.* It is known [29,37] that the scattering probability for  $\text{Ne}^+$  on  $\text{CF}_4$  is largest for charge transfer that has been measured with a guided ion beam apparatus [29] ( $\text{CF}_3^+$ ,  $\text{CF}_2^+$  and  $\text{CF}^+$ ) for energies below  $50 \text{ eV}$  and where also the absence of  $\text{NeCF}_4^+$  and  $\text{CF}_4^+$  was notified. The cross-section extrapolation up to  $1000 \text{ eV}$  was done according to the emission measurements of Motohashi *et al.* [37] and data in [31]. The line spectra of excited atoms obtained in spectrometric measurements in  $\text{CF}_4$  indicate that the charge transfer reaction is by far the most dominant process in collisions with inert-gas ions, so other processes such as Ne excitation can be safely neglected. Having in mind measurements of the charge transfer cross-sections for  $\text{F}^+$  and  $\text{C}^+$  production at high energies [31] we also deduced these cross-sections by using similarities with data for other radical ions [29,31], from  $\text{He}^+ + \text{CF}_4$  scattering data [29], and for the emission cross-sections in [37]. Thresholds for the production of all radical ions were selected from the data for ionization energies for  $\text{CF}_4$  [29].

Since it is not known whether dissociative processes proceed from excited  $\text{CF}_4$  or a  $\text{NeCF}_4^+$  complex, we calculated the dissociation cross-sections by using basic gas phase enthalpies of formation for  $\text{CF}_4$  and products [29]. With the same input data we calculated thermodynamic thresholds and used them as cross-section thresholds.

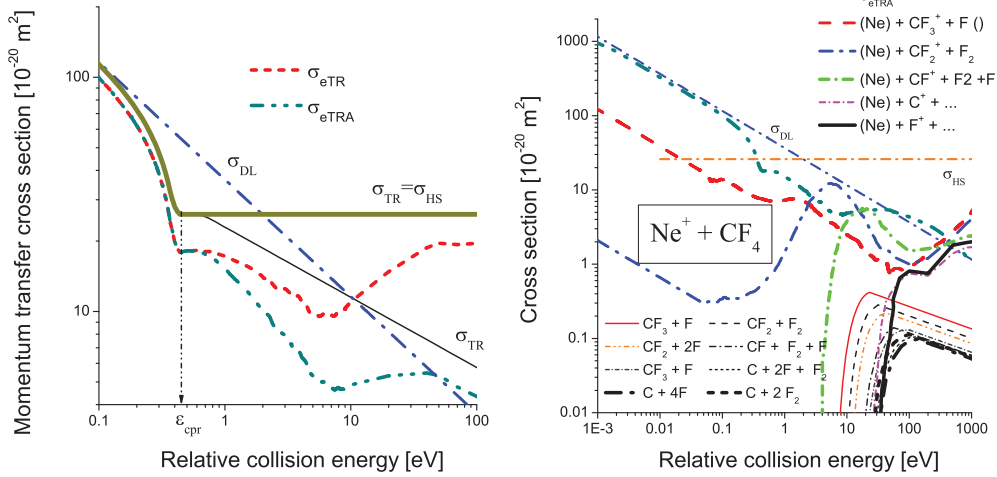


Fig. 2: (Colour online) (a) Momentum transfer cross-section and (b) cross-section set as a function of collision energy for  $\text{Ne}^+ + \text{CF}_4$ .

#### Modelling the elastic momentum transfer cross-section.

Since reactive collisions reduce the total number of trajectories, then  $\sigma_{\text{TR}}$  can be obtained by deducing all non-continuing trajectories represented by reactive cross-sections. The same is easily implemented in Monte Carlo codes by simply removing the particle followed from the simulation. This introduces non-conservativity in transport coefficients dividing them into flux and bulk coefficients [38]. Due to the uncertainties related to the reactive cross-sections it is possible that at high collision energies the sum of the reactive cross-sections be higher than  $\sigma_{\text{TR}}$ . With that in mind the momentum transfer cross-section is approximated by  $\sigma_{\text{TR}} = \sigma_{\text{HS}}$  for collision energies  $\varepsilon > \varepsilon_{\text{cp}}$  (see fig. 2(a)) in order to account for the possible reactions and if combined to a soft-sphere cross-section which is the one actually determined from the mobility of the closed-shell system to take into account scattering anisotropy. If now reactive cross-sections are subtracted, then the resulting cross-section  $\sigma_{\text{eTR}}$  represents all the elastic and inelastic losses. Now we may correct  $\sigma_{\text{eTR}}$  for more realistic angular dependences at high collision energies by applying the function  $A(\varepsilon)$  to obtain the cross-section  $\sigma_{\text{eTRA}} = A\sigma_{\text{eTR}}$  in fig. 2(a). If inelastic collisions can be neglected  $\sigma_{\text{eTRA}}$  represents the elastic momentum transfer cross-section  $\sigma_{\text{elm}} \sim \sigma_{\text{eTRA}}$  which can be easily treated in Monte Carlo codes. If inelastic scattering is included in  $\sigma_{\text{eTRA}}$  the momentum balance will still be holding providing a precise calculation of the ion drift velocity and consequently of the flux. For the case of the previously derived  $\sigma_{\text{eTRA}}$  and reactive processes selected for  $\text{Ne}^+ + \text{CF}_4$ , one may claim that the cross-section set for  $\text{Ne}^+ + \text{CF}_4$  is derived, which recovers the transport properties described by the given transport cross-section for a similar closed-shell system and set of reactive cross-sections (fig. 2(b)).

The mobility of high recombination energy ions such as  $\text{Ne}^+$  ions in  $\text{CF}_4$  is not measured up to now although the

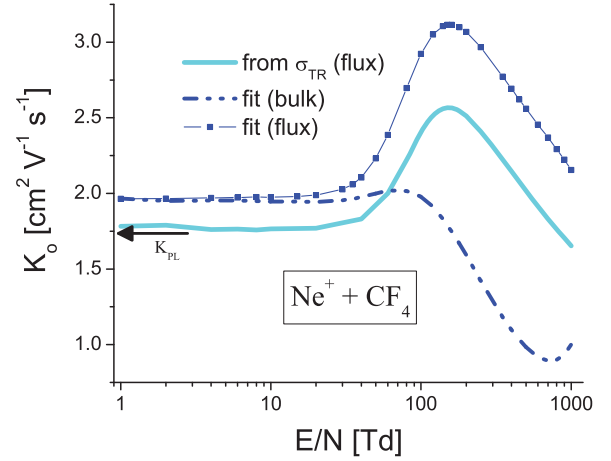


Fig. 3: (Colour online) Reduced mobility as a function  $E/N$  for  $\text{Ne}^+$  ions in  $\text{CF}_4$  gas for temperature  $T = 300 \text{ K}$ .

small reactivity at low  $E/N$  does not present a significant difficulty.

**Transport parameters.** – The reduced mobility for  $\text{Ne}^+$  ions in  $\text{CF}_4$  as a function of  $E/N$  ( $E$  is the electric-field strength,  $N$  the gas number density) compared with bulk and flux values is shown in fig. 3. The bulk drift velocity ( $W = d\langle x \rangle / dt$ ) is the reaction corrected flux drift velocity ( $w = \langle v \rangle$ ):  $W = w + S$ , where  $S$  is the term representing a measure of the effect of the reactions on the drift velocity. The difference between bulk and flux reduced mobility is a consequence of the energy-dependent reactions.

Very different values of flux and bulk reduced mobility are obtained (above about 20 Td), both with peaks of different height, as a consequence of reactive collisions. A large mobility peak appears for the flux component of the reduced mobility, while a much smaller peak appears in the bulk component of the reduced mobility. The flux



reduced mobility peaks at the same  $E/N$  values where the reduced mobility peak is obtained from the momentum transfer cross-section corresponding to the closed-shell system.

**Conclusion.** – We presented a simple and effective method to obtain an elastic momentum transfer cross-section for a modeled system from mobility data for a similar closed-shell system. The method can be described in a step-by-step manner as follows. A similar closed-shell system is used as a reference system with the absence of reactivity. For that system the elastic momentum transfer cross-section ( $\sigma_{\text{TR}}$ ) as a function of energy is deduced by using Monte Carlo simulations where possible clustering reactions and temperature-dependent mobility data are taken into account. The momentum transfer cross-section of the modeled system is then constructed from  $\sigma_{\text{TR}}$  by using its polarization limit and atomic mass. At the same time at high energies a hard-sphere cross-section is used in order to recover the reactivity of the modeled system by subtracting from it the reactive cross-sections. By taking into account the angular dependence of the reference system one finally arrives at an elastic momentum transfer cross-section as a function of the collision energy and at the assessment of the cross-section set.

The method is applied for the case of  $\text{Ne}^+$  scattering on  $\text{CF}_4$  for which the cross-section set is determined. By using the Monte Carlo technique in the final run we calculated transport parameters and discussed flux and bulk mobility as a function of  $E/N$ , which were not available up to now.

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