Energy limitation for models to simulate the buffer gas cooling (postprint)

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Full Text

Preamble

Energy limitation for models to simulate the buffer gas cooling

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1. Introduction

Buffer gas cooling is widely applied at rare-isotope facilities today [1–4]. When combined with radiofrequency ion guides, this technique can improve continuous ion beams and provide cooled ion bunches with low emittance and small energy spread. The method is universally applicable to all elements from helium to the heaviest species [5] and is rather fast, enabling ions to be cooled within a single pass through the cooler device. It has become an essential component for successful high-precision Penning trap mass measurements.

To achieve high-performance ion beam cooling, detailed simulation of the buffer gas cooling process is critical. Three models are frequently used for such simulations: the viscous damping force (VDF) model, hard sphere collision (HSC) model, and realistic interaction potential (RIP) model. The VDF model is a macroscopic approach that provides a simple prescription for the time-averaged cooling force, while the other two are microscopic methods that track the cooling process by simulating individual collisions between ions and buffer gas molecules.

The buffer gas cooling effect has been simulated by many researchers. Lunney and Moore [6] developed a simulation package for a radiofrequency quadrupole rod system operated in helium buffer gas to cool ion beams and reduce emittance. Their results indicated that an emittance reduction of almost two orders of magnitude in all coordinates was possible with near 100% transmission, barring charge exchange and molecular formation. Kim [7] used both the HSC and RIP models to simulate ion cooling in a gas-filled ion guide and compared calculated temperatures of cooled ions with experimental data. Only calculations with realistic potentials could accurately simulate both cooling rates and reproduce experimentally obtained temperatures. Schwarz [8, 9] developed a Monte Carlo simulation package to calculate ion motion in the presence of arbitrary electric (and/or magnetic) fields and buffer gas. Mobility data K_0 as a function of drift velocity obtained from simulations with realistic scattering potentials showed good agreement with experimental data [10, 11], which was not the case for hard-sphere collision simulations.

We emphasize that all these simulations have been performed for ion energies below 100–200 eV and not beyond. The suitable energy (velocity) range (or energy limitation) for these models in buffer gas simulations remains unknown. In this paper, we present our results comparing stopping ranges of various ions in different buffer gases.

2.1. Hard-sphere collision (HSC) model

The HSC model is the simplest approach for handling the cooling force, treating collisions between ions and buffer gas particles as hard-sphere collisions. The ion travels through the neutral background gas, colliding with gas particles moving with randomized velocities according to the Maxwell-Boltzmann distribution, and scatters elastically. This approach has been used in numerous calculations and implemented in several simulation packages [7, 12].

2.2. Viscous damping force (VDF) model

Viscous damping arises from long-range induced-dipole interactions, whereby an ion experiences the effect of many gas atoms simultaneously rather than the very short-range force of a direct collision. Experimental ion mobility values at low energies enable more accurate simulation of viscous damping than hard-sphere collisions. The validity of the VDF model has been verified by the cooling time of ^{39}K in helium gas [6].

When an ion with charge q and mass m moves through gas, its motion is viscously damped, in principle down to the gas temperature itself. The time-averaged cooling effect of the buffer gas is often approximated by a viscous drag force. In simulation, the effect of buffer gas cooling can be implemented by adding a viscous damping term to the ion's acceleration, which can be expressed as [6]:

$$m\frac{d^2\mathbf{u}}{dt^2} = q\mathbf{E} - \frac{m}{K}\left(\frac{d\mathbf{u}}{dt} - \mathbf{v}_{th}\right)$$

where **u** represents the ion's position (x, y, z), K is the ion mobility, **v**_{th} is the thermal velocity of the ion, and K_{th} is its corresponding mobility. The last term prevents the unrealistic situation where ions would be cooled to zero temperature, as the statistical nature of collisions between ions and buffer gas molecules has not been taken into account.

The mobility K at a given pressure p and temperature T can be normalized from the "reduced" value K_0 by:

$$K = K_0 \frac{273.16\,\mathrm{K}}{T}\frac{p}{1013\,\mathrm{mbar}}$$

K_0 values are known for a wide variety of ions at low energies [10, 11, 13]. For the high-energy regime, they can be approximately calculated from:

$$K_0 \propto \frac{(m+M)^2}{N\sigma v}$$

where M is the mass of the gas particle, N is the gas density, and σ is the cross section for hard-sphere collision.

The complete curve can be easily fitted by the following formula:

$$K_0(v) = \frac{v+a}{b_0 + b_1(v+a) + b_2(v+a)^2}$$

where a, b_0, b_1, and b_2 are fitting parameters. As an example, the reduced mobility K_0 of $^{133}Cs^+$ ions in helium gas is shown in Fig. 1 [Figure 1: see original paper]. Data points with velocities lower than 10,000 m/s are taken from Ref. [13], and others are calculated from Eq. (3). The fitted parameters for different ion-molecule combinations are listed in Table 1 .

Table 1 : Fitted parameters for the VDF model for different ion/molecule combinations

Ion/Molecule	a ($×10^{4})	b{0}$ ($×10^{8})	b{1}$ ($×10^{4})	b_{2}$	
Ar^+/He	1.34	6.29	-8.21	0.395	
Cs^+/He	2.41	2.22	-1.82	0.395	
Hg^+/He	2.70	2.72	-1.92	0.395	
Cs^+/Ne	2.83	1.09	-5.12	0.395	
Cs^+/Ar	1.58	8.86	-3.05	0.395	

Note: K_0 values are in units of $cm^2\ V^{-1}\ s^{-1}$, and velocities are in m/s.

2.3. Realistic interaction potentials (RIP) model

Although the hard-sphere collision concept is useful for understanding the cooling mechanism, it ignores the long-range interaction between the ion and buffer gas particle that causes the gas to act as a viscous medium and produces a damping force. The RIP model considers this interaction in a "realistic" way.

The RIP model employs potentials V(r) (also called (n,6,4)-potentials) at distance r as the sum of one short-range repulsion term and two attractive terms [14]:

$$V(r) = \frac{B}{r^n} - \frac{C_6}{r^6} - \frac{C_4}{r^4}$$

The C_4/r^4 and C_6/r^6 terms quantify the attractive interaction of the ion's charge with the electric quadrupole and dipole moments, respectively.

Equation (5) can alternatively be written in dimensionless form as:

$$V(r) = \varepsilon \left[\frac{n}{n(3+\gamma) - 12(1+\gamma)} \left(\frac{r_m}{r}\right)^n - \frac{12(1+\gamma)}{n(3+\gamma) - 12(1+\gamma)} \left(\frac{r_m}{r}\right)^6 + \frac{4\gamma}{n(3+\gamma) - 12(1+\gamma)} \left(\frac{r_m}{r}\right)^4 \right]$$

where r_m and are the position and depth of the potential minimum, respectively, and γ is a dimensionless parameter measuring the strength of the r^{-6} term relative to the r^{-4} term.

The parameters in the potentials (Eq. (5) or (6)) can be derived by fitting the so-called collision integral—a theoretical expression for collision cross-sections—to experimental mobility data [14, 15].

The mobility K can be represented by:

$$K = \frac{3q}{16N} \sqrt{\frac{2\pi}{\mu k T_{\text{eff}}}} \frac{1}{\Omega(T_{\text{eff}})}$$

where q and m are the charge and mass of the ion, M is the mass of the gas particle, N is the number density of gas molecules, k is Boltzmann's constant, and = mM/(m+M) is the reduced mass. The effective temperature T_{eff} accounts for both the gas temperature T_{gas} and the drift velocity v_d of the ion:

$$k T_{\text{eff}} = k T_{\text{gas}} + \frac{1}{2} \mu v_d^2$$

The collision integral (first order) Ω(T) is a function of temperature:

$$\Omega(T) = \sqrt{\frac{kT}{2\pi\mu}} \int_0^\infty \int_0^\infty (1 - \cos\theta(b, E)) b \, db \, e^{-E/kT} \frac{dE}{kT}$$

The deflection angle (b;E) is calculated as a function of impact parameter b and relative collision energy E:

$$\theta(b; E) = \pi - 2b \int_{r_a}^\infty \frac{dr}{r^2 \sqrt{1 - b^2/r^2 - V(r)/E}}$$

where the distance of closest approach r_a is the outermost root of:

$$1 - \frac{b^2}{r_a^2} - \frac{V(r_a)}{E} = 0$$

By inserting Eq. (5) or (6) into Eq. (10) and the result into Eq. (7), one obtains a relationship between the sets of potential parameters (n,B,C_6,C_4) or (n,r_m, ,γ) and the collision integral.

Fitted parameters for the (n,6,4)-potentials for numerous ion-molecule combinations can be found in Ref. [9]. These potentials can also be compared with high-level *ab initio* calculations. Larry A. Viehland et al. [16–20] performed such calculations to investigate interactions between atomic cations and rare gas particles. They calculated potential energy curves and compared spectroscopic parameters and transport coefficients with experimental mobility data, finding excellent agreement. Figure 2 [Figure 2: see original paper] shows *ab initio* calculated potential energy curves for Cs^+/He and Hg^+/Ar, along with potentials calculated from Eq. (5) using parameters from Ref. [9]. In general, the two sets of potential curves are in good agreement, although the (n,6,4)-potentials vary more rapidly than the *ab initio* calculations at short ion-molecule separations. Among all combinations considered in this paper, the potential curves for the Hg^+/Ar combination show the best agreement.

3. Simulations

Since buffer gas cooling is typically used for ion manipulation in ion coolers and bunchers—such as those in ISOLTRAP [22], SHIPTRAP [23], JYFLTRAP [24, 25], LEBIT [4], CPT [26], LPT [27, 28], and others—both the electric (and/or magnetic) fields must be known accurately and the effect of the buffer gas must be modeled adequately. In our simulations, after defining the electrode layout, the ion-optics simulation package SIMION [29] was used to obtain electromagnetic forces acting on ions. However, for this study, we disabled all electrodes (i.e., set them to 0 V) so that ion motion would not be affected by their presence.

To simulate buffer gas cooling, we employed the three models mentioned above: VDF, HSC, and RIP. Within the SIMION framework, we developed user programs implementing the VDF and HSC models to simulate collisions between injected ions and buffer gas. For the RIP model, which is difficult to implement in SIMION, we used the "IonCool" code [8] developed by Stefan Schwarz instead. For convenience and comparison, "IonCool" was also used for HSC model simulations. After detailed testing, we found that HSC model results from both SIMION and "IonCool" were in good agreement, so we report only the SIMION results for the HSC model.

For all models, we simulated combinations of three ion species—a light ion ($^{40}Ar^+$), a medium ion ($^{133}Cs^+$), and a heavy ion ($^{201}Hg^+$)—and three buffer gases (He, Ne, and Ar) commonly used in coolers and bunchers. Each simulation tracked 1,000 ions. The ion's traveling distance in buffer gas was defined as the

stopping range when its kinetic energy decreased to approximately its thermal energy.

Based on the study by Major and Dehmelt [30], we report only results for combinations where the ion mass is much heavier than the buffer gas particle mass, since the cooling effect is our primary interest. In our calculations, parameters from Ref. [31], Ref. [9], and Table 1 were used for the HSC, RIP, and VDF models, respectively.

We employed the SRIM code [32] for comparison with experimental data. Using their extensive stopping power data, Helmut Paul and Andreas Schinner [33] compared these data to various stopping power tables and computer codes to estimate their reliability. They found that SRIM worked fairly well for all ions and energies. Therefore, we used SRIM to calculate stopping powers/ranges and treated these calculated values as "experimental" data.

4. Results and Analysis

Model differences are illustrated in Fig. 3 [Figure 3: see original paper], which compares calculated trajectories based on the VDF, HSC, and RIP models. The chamber is filled with helium buffer gas at 1 Pa pressure with no electric field present, and a single $^{40}Ar^{+}$ ion is injected along the Z-axis with 400 eV kinetic energy.

In VDF model simulations, the ^{40}Ar ion travels along the Z-axis with no divergence to the X/Y axes (radial direction) except at the very end of the trajectory, where it nearly stops. This contrasts sharply with HSC and RIP simulations, where ions initially travel along the Z-axis, then diverge radially, and at the end of their trajectories move randomly without stopping. This difference arises from whether ion-gas collisions are explicitly considered: they are not in the VDF simulation but are included in HSC and RIP simulations.

The corresponding kinetic energy of the ^{40}Ar ion versus elapsed time is shown in Fig. 4 [Figure 4: see original paper]. The HSC simulation yields the shortest cooling time among the three, while the RIP simulation gives the longest. Faster energy loss corresponds to shorter cooling time and smaller stopping range.

Stopping ranges for Ar^{+}, Cs^{+}, and Hg^{+} ions in He buffer gas obtained from simulations using the different models are shown in the upper panels of Fig. 5 [Figure 5: see original paper], with SRIM results included for comparison. The VDF and HSC models perform better than the RIP model when ion energy is less than approximately 5 eV/u, with stopping ranges agreeing more closely with SRIM results. In the low-energy regime, the RIP model underestimates stopping power (i.e., overestimates stopping range) for ions in He buffer gas. However, in the relatively high-energy regime, the RIP model works better than the others. Comparing only the VDF and HSC models, the VDF model is significantly superior, which can be explained by its use of experimental mobility values that are only available for low-energy ions.

Regarding suitable energy ranges, in He buffer gas the VDF model should be used for ion energies below approximately 5 eV/u, and the RIP model up to approximately 80 eV/u. Above this energy, none of the three models can be reliably used, as simulation errors become too large.

Figure 5 [Figure 5: see original paper] also shows stopping ranges for Cs^+ ions in Ne and Ar buffer gases from simulations. As the buffer gas becomes heavier, the upper energy limits for model applicability decrease. For the RIP model, this limit decreases from approximately 80 eV/u in He gas to approximately 50 eV/u in Ne gas and then to approximately 20 eV/u in Ar gas. For the VDF model, it decreases from approximately 5 eV/u in He gas to approximately 2 eV/u in Ar gas.

5. Conclusions

We have investigated the VDF, HSC, and RIP models for simulating ion behavior in buffer gas in detail. Our findings are:

- The VDF and HSC models perform better in the low-energy regime, while the RIP model is superior in the relatively high-energy regime.
- In He buffer gas, which is commonly used in coolers and bunchers, the VDF model should be used for ion energies below approximately 5 eV/u, and the RIP model up to approximately 80 eV/u. Above this energy, none of the three models can be reliably used.
- As the buffer gas becomes heavier, the upper energy limits for model applicability decrease. For Cs^+ ions, the RIP model's upper limit decreases from approximately 80 eV/u in He gas to approximately 50 eV/u in Ne gas and then to approximately 20 eV/u in Ar gas. For the VDF model, it decreases from approximately 5 eV/u in He gas to approximately 2 eV/u in Ar gas.

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Note: Figure translations are in progress. See original paper for figures.