

## Brief Reports

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### Stored-ion collisional relaxation to equilibrium

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The rate of energy transfer between the radial and axial degrees of freedom of protons with measured temperature and number stored in a radio-frequency quadrupole ion trap is quantified. The results are discussed in terms of the theory of charged-particle collisional relaxation to thermal equilibrium.

#### I. INTRODUCTION

When stored in ion traps under ultrahigh-vacuum conditions, clouds of confined ions may relax to thermal equilibrium via ion-ion collisions. Ion-ion collisional relaxation,<sup>1</sup> or collisional cooling, are topics of active interest due to advances in laser cooling<sup>2</sup> of small numbers of stored ions, to possible "crystallization" of cold stored ion clouds,<sup>3</sup> to ongoing studies of one-component plasmas,<sup>4</sup> and to requirements for collisional cooling of ions or antiprotons injected into ion traps with keV energies.<sup>5</sup> Recent theoretical developments clarify the approach to thermal equilibrium via such collisions. Discussed here are measurements of the rate of relaxation between the radial and axial degrees of freedom of protons having a calibrated temperature, stored in a radio-frequency quadrupole ion trap. This relaxation is identified as arising from ion-ion collisions through its dependence on ion temperature. Numerical estimates of the time constants are in rather good accord with experimental results.

#### II. APPARATUS AND TECHNIQUE

The apparatus and ion measurement techniques are only briefly recalled here, since they have previously been described.<sup>6</sup> Protons produced by electron impact dissociative ionization of residual H<sub>2</sub> near 10<sup>-11</sup> Torr were identified and stored in a rf trap with dimensions 3.2 mm = 2z<sub>0</sub> = 2<sup>1/2</sup>r<sub>0</sub>. Axial and radial well depths were equalized near 10.4 V by applying both dc and rf voltages, leading to axial and radial oscillation frequencies of ω<sub>z</sub>/2π = 4.4 MHz and ω<sub>r</sub>/2π = 3.1 MHz, respectively. The ion axial motion was coupled to a parallel tuned circuit basically consisting of an inductor connected across the capacitance between the trap end caps. The ran-

domly varying potential differences across this tank circuit produced by thermal fluctuations, and by currents induced at resonance by the stored protons, were amplified and monitored. These tuned-circuit "temperature" signals were filtered and recorded using a chart recorder, an oscilloscope, or a multichannel analyzer, as appropriate.

About n = 1.5 × 10<sup>3</sup> hot ions initially produced in the trap were observed to cool from 1.2 × 10<sup>4</sup> K to 4.3 × 10<sup>3</sup> K ≡ T<sub>ic</sub> in ≈ 50 s via dissipation of random motional energy by Joule heating of the tuned circuit.<sup>6</sup> The limiting temperature represents a balance between the cooling and "rf heating" of the stored ion cloud. The ion number was estimated from coherent excitation signals produced by the ions when swept through resonance with an excited tuned circuit driven by an external oscillator. These measurements were subsequently confirmed within a factor of 2 using the temperature signals produced by the ions when the temperature scale was calibrated.

The ion temperature scale was obtained by two methods which were found to agree.<sup>6</sup> The quasiequilibrium relation between the monitored tuned-circuit temperature T<sub>i</sub> relative to equilibrium T<sub>0</sub>, and the ion temperature T<sub>i</sub>, is given to a good approximation by (T<sub>i</sub> - T<sub>0</sub>) = η(T<sub>i</sub> - T<sub>0</sub>). Here η = 3nt<sub>i0</sub>/t<sub>it</sub>, where t<sub>i0</sub> = Q/ω<sub>z</sub> = 6.5 μs, and t<sub>it</sub> = 13 s. When the tuned circuit was excited by white noise to a measured temperature T<sub>ih</sub> for a time long compared with t<sub>it</sub>, the ions came to temperature equilibrium at T<sub>ih</sub>. The signal corresponding to this temperature was observed when the noise excitation was removed. It was found that (T<sub>ih</sub> - T<sub>ic</sub>) = (T<sub>ih</sub> - T<sub>0</sub>), thus calibrating the ion temperature scale in terms of the tuned-circuit temperature scale, which was separately calibrated using electron current shot noise. The equality of ion storage times for uncooled ions, and for ions heated with white noise to

the same temperature, provided an independent temperature calibration.

In the above analysis, the plausible assumption is made that the ions have relaxed to a thermal equilibrium characterized by temperature  $T$  during each stage of the measurement process. Since the long ion storage times demonstrate that ion-atom collisions are rare, the exchange of energy between degrees of freedom of the motion must be accomplished either by ion-ion collisions or through some mechanism such as coupling between the orthogonal oscillations via departures from harmonicity of the confining potentials. Some anharmonic coupling was certainly present in this trap, but energy transfer is extremely slow for nondegenerate oscillation frequencies, and the measurements discussed below demonstrate that ion-ion collisions were the dominant relaxation mechanism.

### III. MEASUREMENTS AND ANALYSIS

The radial or axial degrees of freedom of the ion secular oscillations were separately excited parametrically by pulses applied to the trap ring electrode at frequencies  $2\omega_r$  or  $2\omega_z$ , respectively. The axial ion temperature was monitored with the tuned circuit. Figure 1 (top) shows the result when the  $2\omega_r$  excitation was applied, using an 80-ms pulse. The axial temperature increased to a maximum well after the pulse was over, and then slowly decreased with time due to ion cooling. Eventually the ions cooled to their original temperature. When excited at  $2\omega_z$ , as in Fig. 1 (bottom), the axial temperature increased immediately, then dropped to an intermediate equilibrium and slowly cooled. The time constants for the initial ion temperature increase or decrease depended on the amplitude of the parametric pulses. The response time of the measurement system was set at 80 msec for the Fig. 1 data.

In Fig. 1 (bottom) the ion axial temperature signal initially decreases to about one-third of its initial value. This is interpreted as a transfer of the excitation energy from the axial to the two radial degrees of freedom, until temperature equilibrium is reached at  $(T_{\text{eq}} - T_0) = (T'_z - T_0)/3$ . When the two degrees of freedom of the  $r$  motion are simultaneously excited by the parametric pulse, presumably  $(T_{\text{eq}} - T_0) = 2(T'_r - T_0)/3$ .  $T_0$  denotes the initial equilibrium temperature,  $T_{\text{eq}}$  is the new equilibrium temperature following excitation transfer, and  $T'_z$  and  $T'_r$  denote the temperature equivalents of the mean energy of the corresponding degrees of freedom immediately following excitation.

Assuming collisional energy transfer between ions with charge  $Q = qe$ , the Spitzer "self-collision time"<sup>1</sup>  $t_c = 11.4 A^{1/2} T^{3/2} / \rho q^4 \ln \Lambda$  is the characteristic time constant for relaxation to thermal equilibrium of a single-component charged particle gas, following a small perturbation. In this experiment,  $A$  is the ion mass in amu,  $T$  is the temperature in K,  $\rho$  is the ion density in  $\text{cm}^{-3}$ , and  $\ln \Lambda$  is a shielding parameter. The simulations by Kho<sup>7</sup> show that the mean ion energy is closely approached in about one time constant, and that the high-

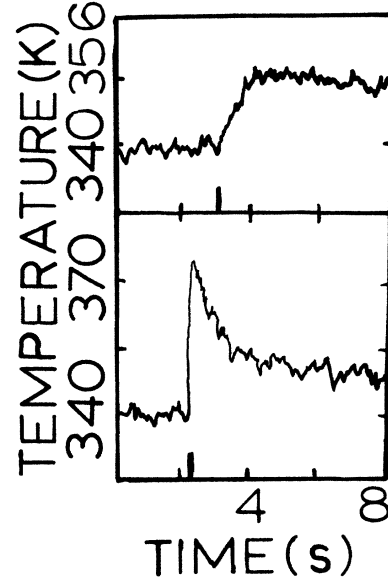


FIG. 1. Response of stored ions excited parametrically in the radial (top) and axial (bottom) degrees of freedom. The ion temperatures were proportional to the temperatures of the tuned circuit, which monitored the axial ion motion at resonance. The ions were excited by 80-ms pulses at times indicated by short heavy vertical lines in the figures.

energy tail of the distribution is filled in several time constants. These calculations replace an earlier numerical simulation,<sup>8</sup> which indicated that much longer times were necessary. The temperature dependence of  $t_c$  shows that equilibrium is more rapidly approached at lower temperatures, a consequence of the energy dependence of the Coulomb interaction.

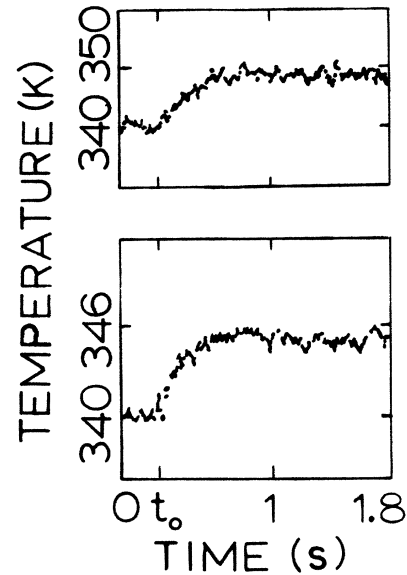


FIG. 2. The response of the axial ion motion to parametric excitation of the radial degrees of freedom by 10-ms pulses at time  $t_0$ . The pulse amplitude used in the bottom measurement was half that used in the top measurement.

This prediction was tested using pulsed parametric excitation of the ion radial motion starting at equilibrium. Measurements were performed using a 10-msec pulse with amplitude  $V$ , followed by similar measurements using a pulse with amplitude  $V/2$ . The lower mean energy of the ions following the weaker excitation resulted in more rapid relaxation, as shown in Fig. 2, top and bottom. This result is interpreted as a confirmation of the hypotheses of ion-ion collisional relaxation.

Using the measured values of ion number and temperature presented earlier, the density of the ions  $\rho = n/2\pi r^2 z = 2 \times 10^6/\text{cm}^3$  at the higher final temperature, and  $2.8 \times 10^6/\text{cm}^3$  at the lower final temperature. Here  $r^2 = 2kT/m\omega_z^2$  and  $\bar{z} = (kT/m\omega_z^2)^{1/2}$ . Assuming that the shielding distance in the theory corresponds to the trap dimensions,  $\ln\Lambda \approx 20$ . By calculation  $t_c = 240$  ms for  $T_{\text{eq}} = 9 \times 10^3$  K, while  $t_c = 120$  ms for  $T_{\text{eq}} = 7 \times 10^3$  K, where these ion temperatures are the values corresponding to the measurements in Fig. 2. These estimates are within 25% of the measured results for the relaxation time constant  $t_r$  for the data shown in Fig. 2;  $t_r(\text{top}) \approx 0.3$  s, and  $t_r(\text{bottom}) \approx 0.14$  s.

#### IV. CONCLUSION

The relaxation to thermal equilibrium of protons stored in a radio-frequency ion trap has been studied and found to be consistent with the mechanism of ion-ion collisions. The Spitzer self-collision time constant  $t_c$  for such collisions scales as  $m^{1/2}/q^4$ , generalizing the results to ions with different mass  $m$  and charge  $q$ . Previously published reports of similar relaxation observed in Penning ion traps<sup>9,10</sup> are consistent with the conclusions reported here, but are less precise in temperature calibration, and lack temperature dependence studies. It appears that if either the ion density or the ion temperature can be accurately obtained, relaxation measurements can be used to estimate the other unknown.

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