Determination of Translation Temperature of N₂⁺ Ions Trapped in 22-pole Trap

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Abstract. A laser induced reaction technique (LIR) was used to determine kinetic temperature of N₂⁺ ions trapped in a 22-pole RF trap. Described are the measurements of translation temperature of N₂⁺ ions stored in the trap cooled down to 10 K. As the monitor reaction we used endothermic charge transfer reaction N₂⁺ + Ar → Ar⁺ + N₂. Rate coefficients for ground state of N₂⁺ and for states excited with laser are different. The kinetic energy of N₂⁺ ions was determined from Doppler broadening of measured spectral lines in Meinel band (X²Σ⁺, v'' = 0 ← A²Π₂, v' = 2). The experiments show cooling of trapped N₂⁺ ions in collisions with helium buffer gas. Parameters of the apparatus, further improvements of ion trapping and cooling and calibrations are discussed.

Introduction

Experiments with the trapped ions at temperatures in range 10 – 300 K are important for laboratory studies of processes in the interstellar space, namely for study of ion-molecule reactions taking place in cold molecular clouds, e.g. reactions leading to formation of hydrocarbons [Wannier et al., 1999], reactions leading to formation of nitrogen containing molecules (molecular nitrogen was recently detected in dense molecular clouds [Knauth et al., 2004]) etc. Many ion-molecule reactions are very sensitive to the temperature and also to the kinetic energy and/or internal excitation of reacting ions and neutrals. Such reactions can be studied by using ion trap techniques, where ions are trapped and cooled e.g. in multipole RF trap and neutral reactant is flowing along the trap (e.g. as a supersonic beam) or it just fills volume of the trap together with buffer gas (usually He). Cooling and thermalization of the trapped ions is the key problem in such experiments.

In the present experiments the thermalization of ions trapped inside the cooled 22-pole trap is a result of dissipation of energy in collisions with helium atoms (buffer gas) and heating in collisions in presence of the RF field of the multipole trap [Asvany et al., 2009]. Ion heating occurs as a result of acceleration of ions by electric field interrupted by collisions with neutrals. After injection of ions to the trap these two processes (cooling and heating) can establish steady state which can be characterized by certain temperature. The temperature of trapped ions can differ from the temperature of neutrals and from the temperature of 22-pole trap and needs to be measured. The laser induced reaction technique (LIR) is excellent method for such measurements.

In our experiments we are using the LIR technique to measure the kinetic energy and internal excitation of N₂⁺ ions trapped in 22-pole. The suitable reaction for such study is endothermic charge transfer reaction of N₂⁺ with Ar. Previous studies of N₂⁺ + Ar → Ar⁺ + N₂ LIR reaction were made for gas temperatures higher than 100 K [Schlemmer et al., 1999], for our experiments it’s important to extend temperature range down to 10 K. This will allow study of ion-molecule reactions within whole range of temperatures relevant to astrochemistry.

The spectroscopic LIR method is the very sensitive instrument for the kinetic temperature determination from Doppler profile of absorption lines. To study e.g. rotational temperature, the population of several rotational states has to be measured. The charge transfer reaction N₂⁺ + Ar → Ar⁺ + N₂ is slow for ground state ions and fast for excited ions, so change in Ar⁺ ions number density can be used as a probe of internal excitation of N₂⁺ ions. A diode laser with tunable wavelength was used to excite N₂⁺ ions and to obtain an absorption spectrum.

Experimental

The central part of the experimental setup is the 22-pole ion trap (Figure 1) [Gerlich et al., 1992]. The trap can be cooled down to 10 K with the helium closed cycle refrigerator (cold head). Ions are produced in the storage ion source, where N₂ is ionized by an electron impact, and then the ions are guided through the quadrupole mass filter to the 22-pole trap [Gerlich et al., 1994]. For temperatures of the cold head higher than 50 K, the 22-pole trap can be filled in with buffer gas (He) and argon.

Injected N₂⁺ ions are cooled in collisions with helium buffer gas. Helium fills all volume of the 22-pole trap and it is cooled down directly with the cold head. After certain time the ions are extracted from the trap, mass-selected and sent to the scintillation Daly-type charged particles detector [Daly, 1960].
Figure 1. The basic scheme of the 22-pole ion trap apparatus with gas injection along the axis of the system (temperature range 10 – 300 K, pressure range 10^{-7} – 10^{-3} Pa, trapping time up to 100 ms): 22-pole trap (22PT) cooled with helium cryogenic system (cold head), the Storage Ion Source and the quadrupole (QP) ion guide, which leads to the 22-pole trap, system for formation of the neutral supersonic beam (with the piezo-valve) and the ion detector system with Daly-detector and the QP mass filter. After passing the 22-pole and the detector section, the neutral beam continues to the beam catcher (BC) chamber, where it is pumped by the turbo pump (TP).

For excitation of Meinel band \((X^2\Sigma_g^+, v'' = 0 \leftarrow A^2\Pi_u^+, v' = 2)\) of \(\text{N}_2^+\) ions a 5 mW infrared laser diode with adjustable wavelength is used. The wavelength is monitored by the wavemeter and Fabry–Pérot interferometer. The excited \(\text{N}_2^+\) ions then react with Ar neutrals forming \(\text{Ar}^+\).

For temperatures of the 22-pole trap lower than 50 K, we installed the neutral beam of Ar atoms as it is described in [Luca et al., 2002]. The beam is formed by passing through the piezo-valve and the skimmer. The temperature of the 22-pole trap is lower than the temperature of condensation of argon, so flow of argon gas should be minimal to prevent covering of the apparatus with argon condensate, but concentration of Ar atoms needs to be sufficiently high. The piezo-valve makes sequence of short pulses of gas synchronized with injection and extraction of the ions from the trap. The skimmer forms the beam that fits to the diameter of the trap inlet aperture (Figure 2).

Measurement

Temperatures of the cold head and 22-pole trap are measured directly by the temperature monitor (silicon diodes are used as temperature sensors). Pressure of the buffer gas in chambers is measured by Bayard-Alpert ionization gauges. The ionization gauge is calibrated with the spinning rotor gas friction vacuum gauge.

Figure 2. The principal scheme of pulse injection of the gas into the 22-pole trap. The system includes the piezo-valve (allows to adjust timing and to change intensity of the neutral beam), the skimmer (forms the supersonic molecular beam with diameter proper to the inlet aperture of the 22-pole trap), the system of differential pumping.
The concentration of argon in the beam in 22-pole trap is estimated from the flow of argon to the beam catcher chamber. This flow is calculated by measuring the change of pressures in the chamber ($\Delta p_{BC}$) with and without beam. The beam can be blocked for this measurement by mechanical shutter. This procedure allows to measure peak/background ratio of Ar concentration in beam passing through the trap. For partial pressure of argon in beam in the 22-pole we can write simple formula:

$$[\text{Ar}] = \frac{\Delta p_{BC}}{k_B T_{BC} S_{beam}},$$

where $S_{BC} = 170$ l/s is pumping speed of vacuum pump (Leybold Pfeiffer TMU 200 MP), $S_{beam} = A \sqrt{3RT_{beam}/M}$ is “injection speed” of argon beam, $T_{BC}$ – temperature of the gas in the beam catcher, $T_{beam}$ – temperature of injected gas, $M$ – atomic mass of injected gas, $A$ – cross section of inlet aperture of the 22-pole trap. The measured time profile of argon pulse is shown in Figure 3. Concentration of the gas was calculated from the measured time-relation of counts of Ar$^+$ ions, the ions counts to gas concentration ratio was determined in the continuous injection mode.

Measured counts of Ar$^+$ ions correlated to data from Fabry–Pèrot interferometer, give absorption spectra of N$_2^+$ ions (Figure 4). Absorption lines of the (X$^2\Sigma^+_u^+, v'' = 0 \leftarrow A^2\Pi_u^+, v' = 2$) transition were measured for different temperatures of the trap ($T_{22PT}$). For $T_{22PT} < 85$ K a fine structure in the spectrum was obtained (Figure 4. Right panel: observed rotational terms of the components of doublet states ($K'' = 2, J'' = 2.5$) and ($K'' = 2, J'' = 1.5$)). Spectra measured with the LIR technique corresponds to previous results [Douglas, 1953].

**Figure 3.** Time related concentration of argon in pulsed beam (frequency of piezo-valve 100 Hz ($T_{duty} = 1/100$ Hz = 1 ms), and Ar partial pressure in beam catcher $\Delta p_{BC} = 3.0 \times 10^{-7}$ Pa).

**Figure 4.** Measured spectral lines of N$_2^+$ (X$^2\Sigma^+_u^+, v'' = 0 \leftarrow A^2\Pi_u^+, v' = 2$) rotational transitions. Ar$^+$ ions detection in LIR process was used for measurements of spectra. The trapping time 200 ms was used in the experiment. The used concentrations are: [He] = $6.7 \times 10^{10}$ cm$^{-3}$, [Ar] = $4.2 \times 10^9$ cm$^{-3}$. **Left panel:** temperature of trap $T_{22PT} = 85$ K, temperature of ions obtained from Doppler profile of spectral lines $T_i = 115 \pm 10$ K. **Right panel:** temperature of trap $T_{22PT} = 10$ K, measured temperature of ions $T_i = 21 \pm 5$ K.
Translational temperature of N$_2^+$ ions was determined from the measured Doppler broadening of spectral lines. Change of frequency is a result of Doppler effect: $\omega = \omega_0 (1 + v/c)$. Intensity of absorption is proportional to number of particles, so assuming Maxwell velocity distribution Doppler profile of spectral line is:

$$I(\omega) = I_0 \exp \left( -\frac{mc^2(\omega - \omega_0)^2}{2KT\omega_0^2} \right),$$

where $T$ is a temperature of particles, $m$ – their mass and $\omega_0$ is the frequency of spectral line. Temperature of ions can be calculated from broadening of spectral lines as:

$$T = \frac{1}{2 \ln 2 k} \left( \frac{\Delta \omega c}{2 \omega_0} \right)^2,$$

where $\Delta \omega$ is the full width at half maximum (FWHM) of spectral line, for details see [Fox, 2006]. The translational temperatures of ions obtained from measured data and the standard error of measurements are written in Figure 4. For measurements were used laser diodes L780P010 and HL7851G with linewidth up to 30MHz [Zhang et al., 1998] which is five times lower than the FWHM of N$_2^+$ absorption lines at 10 K. Therefore the measured FWHM of N$_2^+$ absorption lines and the translational temperature can be higher than the real temperature as a result of convolution of laser and absorption line profiles, absolute deviation of temperature increases with temperature, but relative decrease.

**Conclusion**

The translational temperature of N$_2^+$ ions confined in 22-pole trap cooled down to 10 K was measured by laser induced reaction method. As a monitor reaction we have used N$_2^+$ + Ar ← N$_2$ + Ar$. \Sigma$, The stored ions were excited by diode laser ($^2\Pi_u^\text{v' = 0} ← ^2\Sigma_g^\text{v' = 2}$) and produced Ar$^+$ ions were detected.

It was shown that application of pulse injection of reactant gas directly into the trap, allows expanding the temperature range bellow the temperature of condensation of used reactant gas (Ar).

A difference of measured translational temperature ($T_0 = 21 \pm 5$ K) of confined ions (N$_2^+$) and temperature of the 22-pole ion trap ($T_{22PT} = 10$ K) is consequence balance between cooling and heating of ions including radio frequency heating in multipole trap filled with buffer gas. This result points to the necessity of additional experiments.

Further studies to clarify heating and cooling processes in a RF 22-pole ion trap and improve diagnostics are in progress.

**Acknowledgments.** Special thanks to prof. Dieter Gerlich, Faculty of Mathematics and Physics, Prague, Dipl. Phys. Thomas Mehner, scientific employee of TU-Chemnitz, Faculty of Natural Science, for the first-stage of preparation of experiment. This work is a part of the research plan MSM 0021620834 financed by the Ministry of Education of the Czech Republic and partly was supported by GACR (202/07/0495, 202/09/0642, 205/09/1183, 202/08/H057) and GAUK (86908, 25709, 54010).

**References**


