Comparison experiments of neon and helium buffer gases cooling in trapped $^{199}Hg^{+}$ ions linear trap*

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(Received 17 January 2014; revised manuscript received 7 March 2014; published online 23 July 2014)

The influences of different buffer gas, neon and helium, on $^{199}Hg^{+}$ clock transition are compared in trapped $^{199}Hg^{+}$linear trap. By the technique of time domain's Ramsey separated oscillatory fields, the buffer gas pressure frequency shifts of $^{199}Hg^{+}$ clock transition are measured to be $(df/dP_{Ne})(1/f) = 1.8 \times 10^{-8}$ Torr^{-1} for neon and $(df/dP_{He})(1/f) = 9.1 \times 10^{-8}$ Torr^{-1} for helium. Meanwhile, the line-width of $^{199}Hg^{+}$ clock transition spectrum with the buffer gas neon is narrower than that with helium at the same pressure. These experimental results show that neon is a more suitable buffer gas than helium in $^{199}Hg^{+}$ ions microwave frequency standards because of the $^{199}Hg^{+}$ clock transition is less sensitive to neon variations and the better cooling effect of neon. The optimum operating pressure for neon is found to be about 1.0×10^{-5} Torr in our linear ion trap system.

Keywords: ion cooling, ion trapping, line shape widths and shifts, elastic collisions

PACS: 37.10.Rs, 37.10.Ty, 32.70.Jz, 34.50.Cx **DOI:** 10.1088/1674-1056/23/9/093702

1. Introduction

The ion trapping techniques are widely used in frequency standards due to the long interaction time between the ions and the external radiation fields. In the past decades, $^{137}Ba^{+}$, $^{9}Be^{+}$, $^{113}Cd^{+}$, $^{171}Yb^{+}$, and $^{199}Hg^{+}$ have been investigated by different research groups.[1–6] However, the features of $^{199}Hg^{+}$ ions microwave standards of small volume, good reliability, and environmental adaptation make it particularly attractive for space applications.[7–9]

One crucial technology of $^{199}Hg^{+}$ ions microwave frequency standards is the use of buffer gas to reduce the kinetic energy of $^{199}Hg^{+}$ ions. In the linear trap, because of the space charge repulsion forces pushing ions away from the nodal line of the trap, ions will be RF heated. Buffer gas molecules can cool the ions from about 3000 K of the ion cloud temperature down to near the room temperature by elastic collisions.[10,11] Although reducing the kinetic energy of ions, the presence of a buffer gas introduces a collision shift of the clock transition[12–14] which is one of the leading limitations to the long term stability of $^{199}Hg^{+}$ ions frequency standards.[15] Therefore, the choice of the buffer gas should be aimed to achieve the largest signal with the least pressure collision shift.

Traditionally, neutral helium as buffer gas is used to cool ions. However, neon is a more effective buffer gas candidate in theory. Firstly, collision rate of $^{199}Hg^{+}$ with neon molecules is larger than that of $^{199}Hg^{+}$ with helium molecules.[16–18] Secondly, the fractional energy loss per collision of $^{199}Hg^{+}$ with neon molecules, 20/199, is also larger than that of $^{199}Hg^{+}$ with helium, 4/199.[16,19] To find out which one of neon and helium is more suitable for a buffer gas in $^{199}Hg^{+}$ ions microwave frequency standard, we study the sensitivities of $^{199}Hg^{+}$ clock transition to neon and helium pressure changes and compare the cooling effect of neon with that of helium experimentally.

The frequency shift of $^{199}Hg^{+}$ clock transition with helium pressure was observed in Ref. [16]. The measurement was made by comparing the frequency of the $^{199}Hg^{+}$ standard to a cesium beam frequency standard stable to 10^{-13}. The neon pressure shift measurement in $^{199}Hg^{+}$ microwave frequency standards was reported in Ref. [12]. In frequency standards, the use of the technology of time domain's Ramsey separated oscillatory fields can obtain very narrow line width of the hyperfine clock transition spectrum.[20,21] In a hyperbolic Paul trap, the Ramsey fringes on the ground-state hyperfine transition of $^{199}Hg^{+}$ ions have been realized in our lab.[22] The full width at half maximum (FWHM) of central Ramsey fringes is about 260 mHz with a 2-s free precession period. Recently, we also observe the Ramsey fringes of $^{199}Hg^{+}$ clock transition in a quadrupole linear trap. Based on this technique, we carry out the measurements of frequency shifts of the 40.5-GHz clock transition under a series of neon pressures and he-

*Project supported by the National Natural Science Foundation of China (Grant No. 11074248).

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lium pressures.

The paper is structured as follows. The next section introduces the experimental system of the trapped $^{199}Hg^{+}$ ions microwave frequency standards. In Section 3, we report the buffer gas pressure shifts of the 40.5-GHz clock transition and the results of the FWHMs of $^{199}Hg^{+}$ hyperfine transition spectrum under a series of neon pressures and helium pressures. Some discussion and conclusions are made in Section 4.

2. Experimental setup

A schematic of the ion trap region is presented in Fig. 1. The trap consists of four stainless steel rods evenly spaced on a circle with a radius of 10 mm. The diameter of the rod electrodes is 4 mm. The length of each end-cap electrode is 20 mm and the two end-cap electrodes are 60-mm apart. A trap drive of approximately 600 V at 1 MHz is applied to a pair of opposing rods with the other pair being connected to the ground. Axial confinement is achieved by the dc voltage. Neutral ^{199}Hg is introduced into the vacuum by heating HgO powder. An electron pulse emitted from a heated LaB_6 cathode bombards neutral ^{199}Hg to create$^{199}Hg^{+}$. The trap is situated within an ultrahigh vacuum chamber. A pressure lower than 9×10^{-8} Pa in the chamber is measured from a vacuum gauge. A pair of Helmholtz coils is used to supply an adjustable homogeneous magnetic field parallel to the trap axis. The trap region is surrounded by two layers of magnetic shields in order to prevent ambient field disturbances from influencing the hyperfine transition of $^{199}Hg^{+}$.

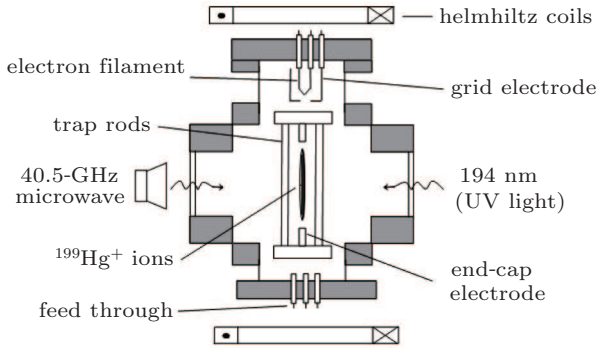


Fig. 1. The top view of quadrupole linear ion trap in our lab.

Neon is introduced through a precision leak valve (MDC-MLV). Different neon pressures are reached via a variable leak valve at a constant pump speed. The purity of neon is up to 99.9995%. Unlike neon, the buffer gas helium is taken into the vacuum chamber through a quartz leak consisting of a thin quartz tube wrapped with tungsten wire. With the quartz leak heated, the helium can enter the chamber through the quartz tube by diffusion. Different helium pressures can be obtained by changing the temperature of the quartz leak. The buffer gas pressure is measured with a quadrupole mass spectrometer (SRS-RGA300). In our experimental system, the pressure of helium is up to 1.0×10^{-5} Torr, while the pressure of neon is up to 3.0×10^{-5} Torr. The other residual gases such as water, nitrogen, oxygen can be reduced to a sufficiently small level about 10^{-10} Torr after baking the system to a high temperature for a long period of time.

We optically pump the $^{199}Hg^{+}$ ions with a $^{202}Hg^{+}$ lamp. This is because that the transition in $^{202}Hg^{+}$ which is 194.2 nm is fortuitously resonant with the transition from the upper ground hyperfine state ($6^2S_{1/2}$, $F=1$) in $^{199}Hg^{+}$ to the first excited state $6^2P_{1/2}$ shown in Fig. 2. The ions decay from the $6^2P_{1/2}$ state to both hyperfine states ($6^2S_{1/2}$, $F=0$ or $6^2S_{1/2}$, $F=1$), but the effect of continued illumination with 194.2 nm populates the lower hyperfine state ($6^2S_{1/2}$, $F=0$) in $^{199}Hg^{+}$ at the expense of the upper. This is called state selection. Applying resonance radiation at the hyperfine frequency 40.5 GHz restores the population of the upper state ($6^2S_{1/2}$, $F=1$).

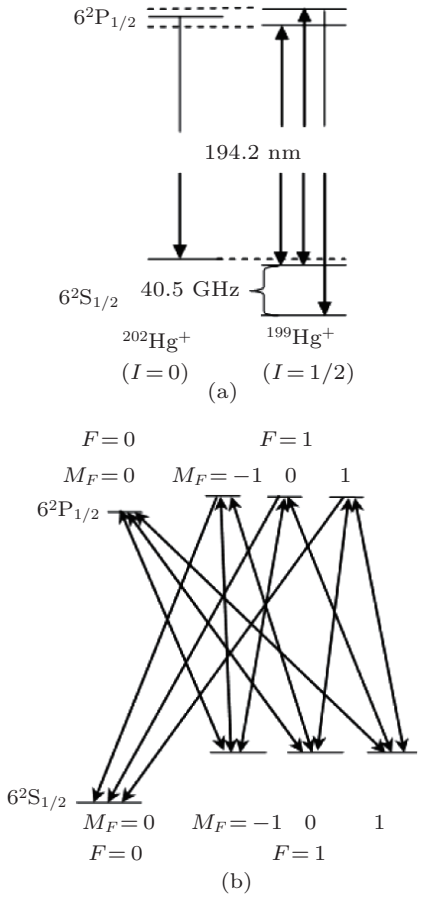


Fig. 2. (a) Energy levels of $^{202}Hg^{+}$ and $^{199}Hg^{+}$ showing the hyperfine splitting and isotope shifts. (b) Optical pumping transition in $^{199}Hg^{+}$ leading to preferential population of the $F=0$ sublevel.

The microwave radiation at 40.5 GHz to excite the hyperfine transition is produced by a high-precision microwave source (Agilent E8057D) with resolution up to 1 mHz. The

microwave radiation is introduced into the trap region by the gold plated horn at the direction perpendicular to the axis of the trap, as shown in Fig. 3. The ion motion in this direction is small compared with the microwave wavelength, thus the ions are Lamb–Dicke confined and the first-order Doppler shift is suppressed.[23] A spherical mirror and a cylindrical mirror are used to focus the UV light from $^{202}Hg^{+}$ lamp to trap region. The fluorescence light is detected by a solar blind photomultiplier tube (PMT) at the vertical direction of the UV state selection/interrogation light. A UV interference filter (bandwidth about 20 nm and peak transmission 10% at 200 nm) and UV grade quartz lenses are used in the detection path in order to suppress the spurious light with a wavelength different from 194.2 nm generated mostly from the $^{202}Hg^{+}$ lamp.

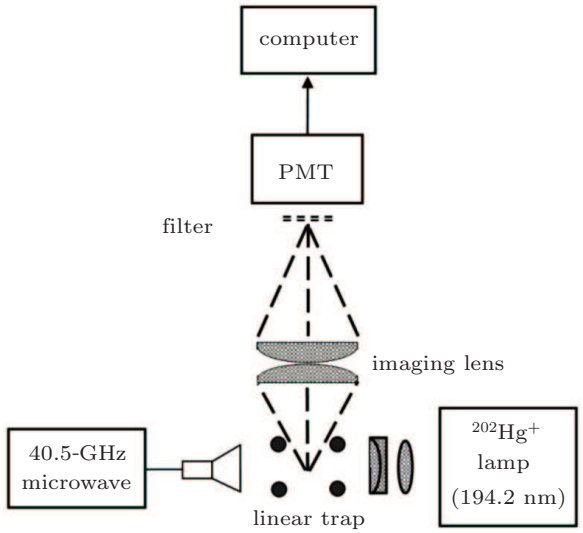


Fig. 3. Schematic representation for optical pumping and detection of fluorescent radiation from the trapped ions.

3. Experimental measurement

3.1. Measurement of the frequency shift of $^{199}Hg^{+}$ clock transition

In order to observe the frequency shift of the clock transition with buffer gas pressure, we carry out the experiment of the Ramsey fringes on the ground-state hyperfine transition of $^{199}Hg^{+}$ ions based on the technique of the time domain's Ramsey separated oscillatory fields. Representative microwave resonance Ramsey fringes of $^{199}Hg^{+}$ clock transition are shown in Fig. 4. The sweep width is 3.2 Hz and the frequency interval between points is 10 mHz. Two coherent 0.3-s microwave pulses separated by a 2-s free precession period are used to drive the hyperfine transition. The detail of experimental sequential operation was introduced in Ref. [22]. The Helmholtz coils are set to be 8 mA to create about 0.2 Gauss homogeneous magnetic fields in the region of the trap that can avoid loss of the Ramsey signal and the optimum power of microwave radiation is −55 dBm. The red curve is obtained by a least squares fit which is in accordance with the expected theoretical equation shown in Ref. [24]. The center frequency of hyperfine transition of $^{199}Hg^{+}$ is 40507348001.60 Hz with a neon pressure of 1.0×10^{-5} Torr at 8-mA current of the Helmholtz coils.

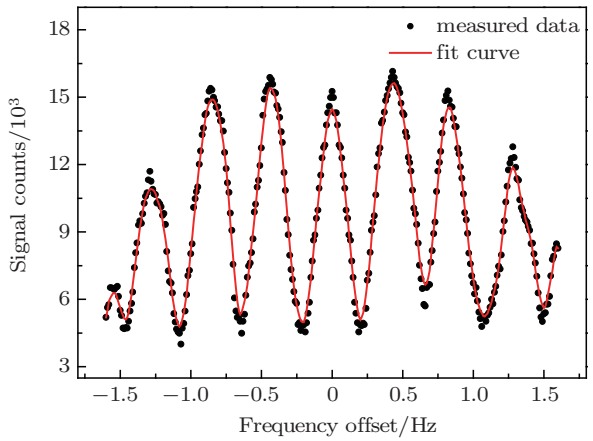


Fig. 4. (color online) Ramsey fringe of $^{199}Hg^{+}$ clock transition with 0.3-s microwave pulses separated by a 2-s free precession period.

Figure 5 represents the frequency variations of $^{199}Hg^{+}$ clock transition with the buffer gas pressures. The ion number is held constant to the utmost. The frequency shifts of the 40.5-GHz clock transition are $(df/dP_{Ne})(1/f) = 1.8\times10^{-8}$ Torr^{-1} for neon and $(df/dP_{He})(1/f) = 9.1\times10^{-8}$ Torr^{-1} for helium. The frequency shift with neon is 5 times less sensitive to the pressure changes than helium.

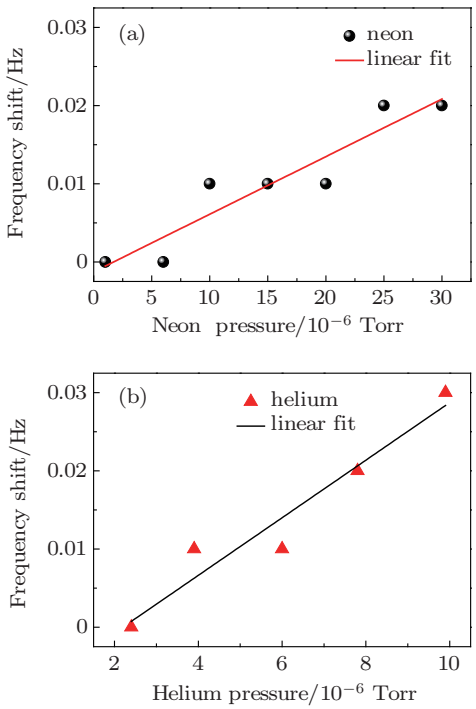


Fig. 5. (color online) Observed frequency shifts of $^{199}Hg^{+}$ clock transition as the changes of (a) neon pressure and (b) helium pressure.

3.2. Measurement of the fluorescence spectrum of $^{199}Hg^{+}$ clock transition

By sweeping the 40.5-GHz microwave frequency through the resonant frequency, we obtain the fluorescence spectrum of $^{199}Hg^{+}$ clock transition (see Fig. 6). The range of scanning microwave frequency is over ± 2.0 Hz around $f_0 =$ 40507348001.60 Hz. The power of microwave radiation is -55 dBm. We fit the experimental data by Voigt fitting. The Doppler broadened line is the Gaussian profile and the collision broadened line is the Lorentz profile.[25] The total broadened line shape is the Voigt profile which is convolution of the Gaussian profile and the Lorentz profile.[26] The signal counts are 18400 and the FWHM of the profile is 0.86 Hz with 1.0×10^{-5} Torr of neon pressure.

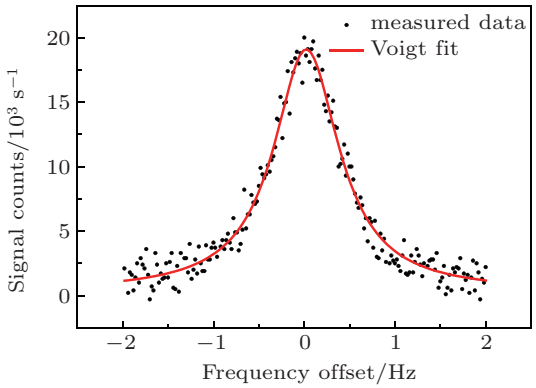


Fig. 6. (color online) The fluorescence spectrum of $^{199}Hg^{+}$ clock transition with 1.0×10^{-5} Torr of neon.

As we know, when the kinetic energy of the ion surpasses the trapping potential, the ion will not be stored in the trap. In Fig. 7, the signal counts increase with the increase of buffer gas pressure, which shows ions are cooled effectively by the buffer gas. At lower buffer gas pressures, we trap a smaller ion cloud, with a higher equilibrium temperature. At higher buffer gas pressures, we trap a bigger ion cloud, with a lower equilibrium temperature.

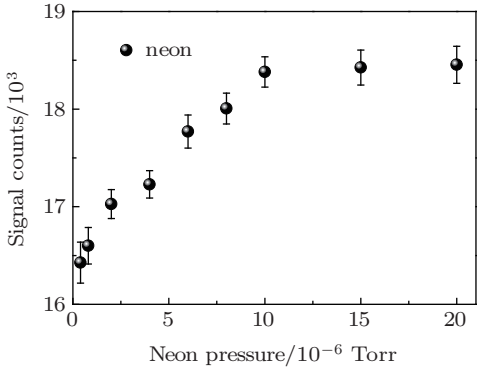


Fig. 7. The varying of the signal size of $^{199}Hg^{+}$ clock transition with respect to neon pressure. The signal size is proportional to the total number of ions stored.

Figure 8 compares the line-widths of $^{199}Hg^{+}$clock transition spectrum using neon as the buffer gas with those using helium as buffer gas. With neon, the FWHM of the line profile is decreasing with the increase of neon pressure when the pressure is smaller than 1.0×10^{-5} Torr. This is interpreted to the average kinetic energy of ions reduction and the second-order Doppler effect suppression. At pressure larger than 1.0×10^{-5} Torr, the ion cloud is in thermal equilibrium with neon molecules, but further increase in gas pressure leads to the collision between $^{199}Hg^{+}$ ions and neon molecules intensively. As a result, the FWHM of profile increases due to the broadened collision. With helium, at a pressure smaller than 6.0×10^{-6} Torr, the FWHM of clock transition is decreasing with the increase of helium pressure. Because the FWHM of profile with 8.0×10^{-6} Torr of helium is wider than that with 6.0×10^{-6} Torr, the line-width is inclined to broaden due to collision effect as helium pressure exceeds 8.0×10^{-6} Torr. At the same pressure, the line-width of the clock transition spectrum with neon is narrower than that with helium. This indicates that collisional cooling of neon is more effective than that of helium. The narrowest FWHM with neon is about 0.86 Hz while that with helium is about 1.19 Hz.

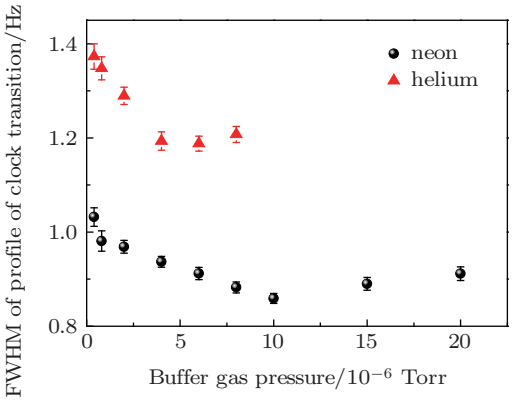


Fig. 8. (color online) The FWHM of profile of $^{199}Hg^{+}$ clock transition varying with neon pressure (black) and helium pressure (red).

4. Discussion

The buffer gas pressure shift of $^{199}Hg^{+}$ clock transition is 1.8×10^{-8} Torr^{-1} for neon and 9.1×10^{-8} Torr^{-1} for helium. Presently, the pressure can be stabilized to the level 1×10^{-7} Torr in our experimental system. Hence, the frequency offset of $^{199}Hg^{+}$ clock transition to neon pressure variations is estimated to be 2×10^{-15} and to helium pressure variations is 9×10^{-15}. An improvement in the long-term stability of $^{199}Hg^{+}$ ions microwave standards can be obtained by using neon as buffer gas.[27] The line-width of the clock transition spectrum using neon as buffer gas is narrower than that using helium. The quality factor Q ($Q = f_0/\Delta f$) with neon is larger than that with helium. Q is inversely proportional to the short-term performance of frequency standards. Therefore, the use of neon is also in favor of improving the short-term stability of $^{199}Hg^{+}$ ions microwave standards.[27] From the above, neon

is a better choice as a buffer gas than helium in $^{199}Hg^{+}$ microwave frequency standards.

The frequency shifts of the 40.5-GHz clock transition which we obtained, $(df/dP_{Ne})(1/f) = 1.8 \times 10^{-8}$ Torr^{-1} for neon and $(df/dP_{He})(1/f) = 9.1 \times 10^{-8}$ Torr^{-1} for helium, are larger than those reported in Ref. [12], $(df/dP_{Ne})(1/f) = 8.5 \times 10^{-9}$ Torr^{-1} for neon and $(df/dP_{He})(1/f) = 2.1 \times 10^{-8}$ Torr^{-1} for helium. The reasons for this difference may be that the test range of the buffer gas pressure in our experiments is mainly in the level of 10^{-6} Torr at which the frequency increases quickly[16] and the differences between the purity of buffer gas we used and that used in Ref. [12]. The error bars are given in Figs. 7 and 8 considering the drift of 194.2-nm pumping light power and the statistical errors during the spectra fitting.

5. Conclusion

In summary, we experimentally studied the influences of different buffer gas, neon and helium, on $^{199}Hg^{+}$ clock transition by observing the buffer gas pressure shifts of the clock transition and the variations of line-width of the clock transition spectrum under a series of pressures in trapped $^{199}Hg^{+}$ linear trap. Neon is a better choice as a buffer gas than helium due to the help to improve the long-term and short-term stabilities of $^{199}Hg^{+}$ ions microwave frequency standards. The fractional frequency offset of $^{199}Hg^{+}$ clock transition to neon pressure variations is estimated to be 2×10^{-15} and to helium pressure variations is 9×10^{-15}. The optimum operating pressure for neon is found to be about 1.0×10^{-5} Torr to maximize the clock resonance signal size and minimize line-width of the profile of $^{199}Hg^{+}$ clock transition in our linear ion trap system.

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