Hyperfine Interact (2015) 236:87–94
DOI 10.1007/s10751-015-1188-y

Characterization of ion Coulomb crystals for fundamental sciences

Kunihiro Okada[1] · Masanari Ichikawa[2] · Michiharu Wada[1]

Published online: 13 May 2015

Abstract We performed classical molecular dynamics (MD) simulations in order to search the conditions for efficient sympathetic cooling of highly charged ions (HCIs) in a linear Paul trap. Small two-component ion Coulomb crystals consisting of laser-cooled ions and HCIs were characterized by the results of the MD simulations. We found that the spatial distribution is determined by not only the charge-to-mass ratio but also the space charge effect. Moreover, the simulation results suggest that the temperature of HCIs do not necessarily decrease with increasing the number of laser-cooled ions in the cases of linear ion crystals. We also determined the cooling limit of sympathetically cooled $^{165}Ho^{14+}$ ions in small linear ion Coulomb crystals. The present results show that sub-milli-Kelvin temperatures of at least 10 Ho^{14+} ions will be achieved by sympathetic cooling with a single laser-cooled Be^{+}.

Keywords Sympathetic cooling · Highly charged ion · Ion trap

1 Introduction

Sympathetically cooled molecular ions and highly charged ions (HCIs) are fascinating research objects for studying fundamental sciences, such as cold/ultracold ion chemistry

Proceedings of the 6th International Conference on Trapped Charged Particles and Fundamental Physics (TCP 2014), Takamatsu, Japan,1-5 December 2014

✉ Kunihiro Okada
okada-k@sophia.ac.jp

Michiharu Wada
mw@riken.go.jp

[1] Department of Physics, Sophia University, 7-1 Kioicho, Chiyoda, Tokyo 102-8554, Japan

[2] RIKEN Nishina Center for Accelerator-Based Science, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

[1, 2] and the study of possible time variations of fundamental constants via precise optical spectroscopy [3, 4]. Recently, cold highly charged ions were considered to be good candidates for probing the time variation of the fine structure constant α [5–7]. In performing these studies, the sympathetic cooling by ion Coulomb crystals in a linear Paul trap is a promising method for generating cold molecular ions or cold highly charged ions. Since ultra-high accuracy measurements of optical transitions are indispensable for studying possible time variation of α, for sympathetically cooled HCIs the so-called Lamb-Dicke criterion should be satisfied in a linear Paul trap. Thus, for such measurements, a linear configuration of ion crystals is much more suitable than large two-component Coulomb crystals with shell structures.

Recently precise laser spectroscopy of Ar^{13+} and Ir^{17+} ions by the sympathetic cooling technique was proposed to test quantum electrodynamics and to probe possible time variation of α [8, 9], and then a cryogenic linear Paul trap connecting with the EBIT facility was developed [10]. Trapping and cooling of externally injected Ar^{13+} into the cryogenic linear Paul trap was reported very recently [11, 12]. However, the direct observation of laser-induced fluorescence from crystallized HCIs has not been performed yet.

In this work, we performed molecular dynamics (MD) simulations in order to search the conditions for efficient sympathetic cooling of HCIs in a linear Paul trap. The characterization of small two-component linear ion Coulomb crystals consisting of HCIs and laser-cooled ions (LCIs) was performed using the simulation results. First we discuss the spatial distribution of sympathetically crystallized HCIs in a linear Paul trap. In particular the ion distribution of very highly charged $^{165}Ho^{66+}$ ions is discussed. Then the sympathetic cooling efficiency of HCIs via laser-cooled Be^{+} ions for small ion crystals is investigated in detail by changing the simulation conditions. Finally we investigate the cooling limit of sympathetically cooled HCIs, namely $^{165}Ho^{14+}$, which is a promising candidate for detecting possible time variation of the fine structure constant [13].

2 Molecular dynamics simulation

We performed molecular dynamics simulations to characterize two-component linear ion Coulomb crystals in a linear Paul trap. In the present simulations, all forces, i.e., static and time-dependent quadrupole electric fields, radiation-pressure forces, average recoil forces by spontaneous emissions, and ion-ion Coulomb repulsion forces are taken into account in Newton's equations of motion of all the trapped ions. The equations are numerically integrated by the fourth-order Runge-Kutta algorithm with a 2 ns time step. In order to reduce a computational time for obtaining the quasi-equilibrium states of Coulomb crystals, we introduced cold elastic collisions between laser-cooled ions (LCIs) and virtual very light atoms in the early stage of integration steps. The details of the method is described in [14]. After this process, only the radiation pressure forces are applied to LCIs. The sympathetic cooling of HCIs is achieved by only the Coulomb interactions with LCIs.

Since the positions of all ions at each integration step are recorded, the simulation image of crystallized ions like an observed laser-induced fluorescence image is produced by the density plot of existence probabilities of the ions as follows. First we divide an predetermined image area into many small cells and the content of each cell is set to 0. Then, at each integration step, we increment a cell counter by one if an ion exists in this cell. The spatial distribution of ions can be evaluated by the production image. We also obtain the

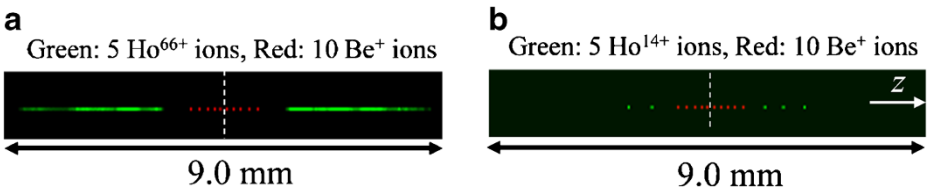

Fig. 1 Simulation images of two-component linear ion crystals consisting of 10 Be^+ and 5 Ho^{q+} ions: (a) $q = 66$, (b) $q = 14$. The vertical dotted line indicates the center of the trap along the z-axis. Simulation parameters: $f_{RF} = 10$ MHz, $V_{ac} = 30$ V, $V_z = 0.1$ V. The trapping parameters are $q(Ho^{66+}) = 0.123$, $q(Ho^{14+}) = 0.0262$, and $q(Be^+) = 0.0343$, respectively. The dimensions of the linear Paul trap are as follows: $r_0 = 2.18$ mm, $z_0 = 25$ mm, where r_0 and $2z_0$ are the closest distance from the trap axis to the rod electrode and the axial length of the ion trap, respectively. The geometrical factor (κ) along the trap axis was taken as 0.33. The intensity and the detuning of the cooling laser for calculations of radiation pressure forces were set to 3 mW/cm^2 and -10 MHz, respectively. The incidence angle of the cooling laser is 5 deg. with respect to the trap axis

micromotion energies of HCIs by averaging the kinetic energies of the trapped ions. The present simulations were performed by modifying the previously developed codes [14, 15].

3 Results and discussions

3.1 Spatial distribution of sympathetically cooled HCIs

In the case of singly charged ions, the ions with a higher charge-to-mass ratio (Q/m) are subjected to a stronger trapping force and consequently gather near the trap axis under the influence of the sympathetic cooling effect. However this is not the case for two-component Coulomb crystals including highly charged ions. Figure 1a shows a simulation image consisting of 10 Be^+ and 5 $^{165}Ho^{66+}$ ions at the quasi-equilibrium state. Although (Q/m) of Ho^{66+} is larger than that of Be^+, the HCIs are distributed in the outside of the LCIs. Since the Be^+ ions firstly occupy the positions around the minimum of the trapping potential by the cooling effect, the sympathetically cooled Ho^{66+} ions are pushed to the outside of the trap center by the space charge of the cold Be^+ ions.

It is to be noted that the size of the linear ion crystal is considerably larger than those of normal two-component linear ion crystals consisting of singly charged ions [14, 16]. This is also due to the strong Coulomb interactions between Ho^{66+} and Be^+ ions. Actually the axial size of a linear ion crystal becomes smaller as the charge state decreases, as shown in Fig. 1b. On the one hand, we need to increase the axial static voltage (V_z) applied along the trap axis in order to compress the axial distribution. However, the high V_z leads to breaking of the string shape of the ion crystal and then induces a high rf heating rate. In fact the Ho^{66+} ions are easily lost from the trap by increasing V_z from 0.1 to 1.0 V in the simulation conditions of Fig. 1a. It is also important to reduce asymmetric fields in the radial direction as much as possible. For the explanation of these points see also in Section 3.2.

Figure 2 shows the coolant ion-number dependence of the average kinetic energy of 6 Ho^{66+} ions. In these simulations, the average kinetic energy of the coolant Be^+ ions was maintained to be 23 mK by applying cold elastic collisions with virtual very light atoms. The reasons why this method works well to simulate the Coulomb crystals are described in the previous paper [14]. The temperatures indicated in the images in Fig. 2 show the average kinetic energies of 6 Ho^{66+} ions expressed by the unit of Kelvin. In the present simulation

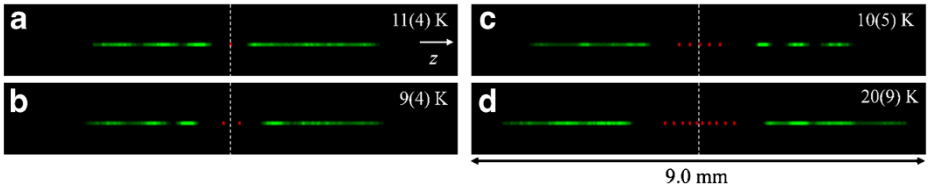

Fig. 2 Simulation images of two-component Coulomb crystals consisting of 6 Ho^{66+} ions (*green*) and the different numbers of Be^{+} ions (*red*), where the average kinetic energy of the coolant ions is maintained to be 23 mK by cold elastic collisions with virtual very light atoms [14]. The vertical dotted line indicates the center of the trap along the z-axis. The other simulation parameters are the same as in Fig. 1

conditions, the average kinetic energy of the Ho^{66+} ions is almost same within the standard deviations in the case that the number of the Be^{+} is up to 5. However, the average kinetic energy of Ho^{66+} increases as the number of Be^{+} ions increases, as shown in Fig. 2d.

This phenomenon is possibly explained as in the following. The space charge of the coolant ions decreases the trapping potential depth but creates a barrier around the trap center. As the number of the coolant increases, the radial deviations of either coolant or HCIs become larger under larger axial stress from the extended string, leading to larger rf heating rates especially in light of the off-axis component of the cooling laser force. Even if the deviation from the trap axis is small, the large rf heating effect on the HCIs is expected. Thus, the excess number of the coolant ions possibly leads to low sympathetic cooling efficiency.

Note that it might be possible to take other ion configurations in small linear ion crystals consisting of Ho^{66+} and Be^{+} ions as shown in Figs. 1 and 2. The configurations possibly depend on the initial conditions of the MD simulations [11, 16, 17]. Although the observed configurations are stable at least and are taking the local minimum of the energies under the present simulation conditions, it is not clear whether those are taking the minimum energy configurations or not. Moreover, it might be possible to take more stable configurations by collisions with residual gases [18] or by intensity fluctuations of lasers. The former effect may also change the charge state of HCIs. Since in the present simulations such the effects were not included, it needs further studies.

3.2 Cooling limit of sympathetically cooled Ho^{14+} with a single Be^{+}

It is interesting to know the cooling limit of Ho^{14+} by sympathetic cooling, since the Ho^{14+} is considered to be a good candidate to probe possible time variation of the fine structure constant via optical atomic clock [13]. Here we perform extensive MD simulations in order to search the conditions for efficient sympathetic cooling of Ho^{14+} in a linear Paul trap. As described in Section 3.1 the excess number of coolant ions leads to low sympathetic cooling efficiency. Therefore we first tested a single Be^{+} ion as a coolant. Figure 3a shows plots of the average kinetic energies of Ho^{14+} and a single laser-cooled Be^{+} as a function of time. The inset figure shows the position of each ion and the incident direction of the cooling laser. In this simulation, we applied the following calculation procedures.

In the period I, the cold elastic collisions between Be^{+} and virtual very light atoms with thermal energy of 7 mK were introduced [14] and the sympathetic cooling of Ho^{14+} ions is achieved by the Coulomb interaction with Be^{+}. This approach drastically reduces calculation steps in order to achieve the quasi-equilibrium state of the two-component Coulomb crystal. This collision cooling method is useful to reproduce the realistic Coulomb crystals,

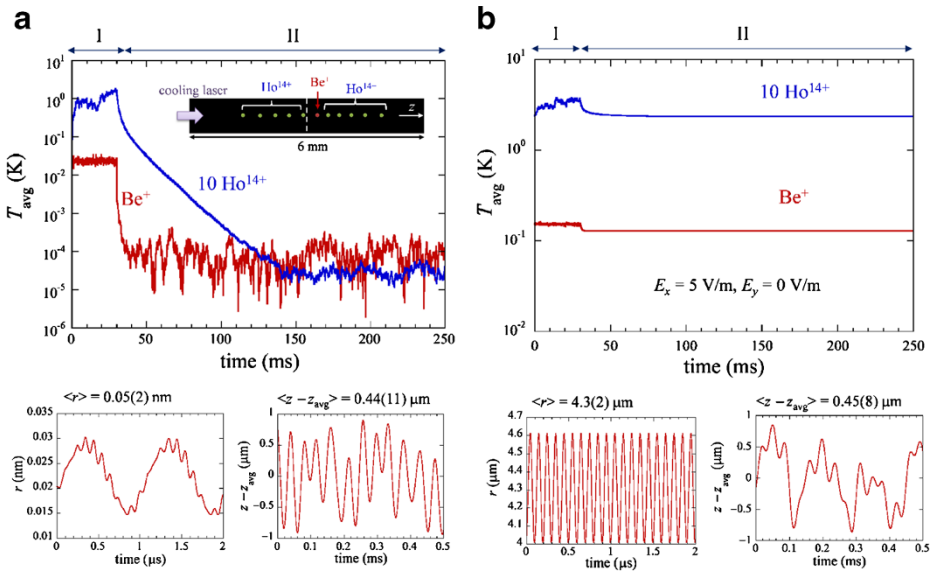

Fig. 3 Plots of average kinetic energies of Ho^{14+} and Be^{+} as a function of time under the ideal trapping fields (a) and under applying the asymmetric static filed of $E_x = 5$ V/m along the x axis (b). The inset figure shows the position of each ion and the incident direction of the cooling laser. In the period I, the cold elastic collisions between Be^{+} and virtual very light atoms with a thermal energy of 7 mK were introduced [14]. Then, in the period II, only the laser radiation pressure force was applied to Be^{+}. The lower figures show plots of the time variation of r and $z - z_{avg}$ of a certain Ho^{14+} ion, where z_{avg} is the average ion position in the z direction. We also show the average values of r and $z - z_{avg}$ at the quasi-equilibrium state of the linear ion crystal. The intensity and the detuning of the cooling laser were set to 1 mW/cm^2 and -10 MHz, respectively. The ion trap parameters: $f_{RF} = 10$ MHz, $V_{ac} = 80$ V, $V_z = 0.1$ V. The other simulation parameters are the same as in Fig. 1

where the cooling and heating effects are balancing each other due to occasional collisions with background gases [14]. However, it is not appropriate for the present purpose, i.e. searching for the sympathetic cooling limit of Ho^{14+}. Thus, in the period II, only the radiation pressure force was applied to Be^{+} .

We have successfully obtained the efficient cooling conditions of 10 Ho^{14} by a single laser-cooled Be^{+} under the ideal trapping fields. The average kinetic energies of Ho^{14+} and Be^{+} are 28(8) μK and 98(56) μK, respectively. The reason why the average kinetic energies appears to be lower than the Doppler cooling limit of Be^{+},435 μK, is that the residual kinetic energy is distributed to the individual ions. In actual, the sum of the individual ion energy is $3.8(1.4)\times10^2$ μK, which is consistent with the Doppler cooling limit of Be^{+}. The average radial position of Ho^{14+} is also obtained to be 0.05(2) nm. The position deviation of Ho^{14+} ions from the trap axis is supposed to be caused by the intentional cooling laser misalignment of 5 deg. It is noted that we can not obtain the actual time to achieve the cooling limit from the present simulations, since the purpose is the search for the cooling conditions and the cooling limit of Ho^{14+}.

In order to investigate the effect of imperfect radial electric fields caused by patch potentials or misalignment of the trap electrodes, we perform the MD simulation by applying a static electric field (E_x) in the x direction. Figure 3b is a plot of the average kinetic energy of 10 Ho^{14+} sympathetically cooled by a single Be^{+}. The simulation parameters are the same

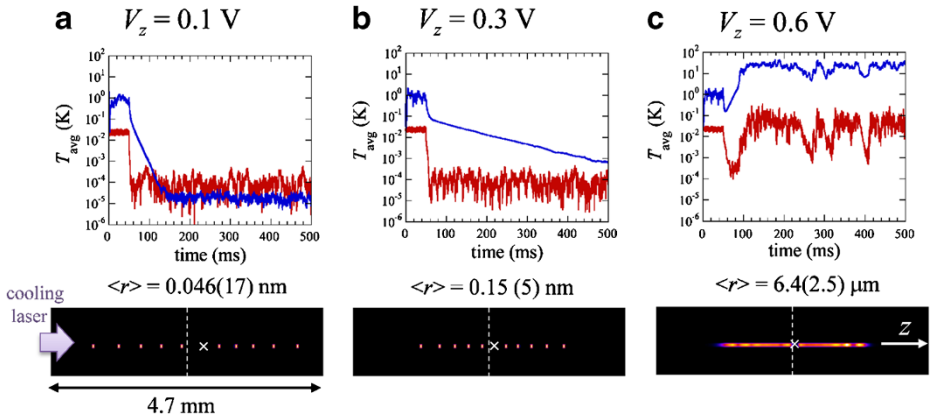

Fig. 4 Plots of average kinetic energies of 10 Ho^{14+} (*blue*) and a single Be^+ (*red*) as a function of time under the ideal trapping fields: (a) $V_z = 0.1$ V, (b) $V_z = 0.3$ V, (c) $V_z = 0.6$ V. The lower figures show the simulation images of Ho^{14+} ions corresponding to the above plot. The vertical dotted line indicates the center of the trap along the z-axis and single Be^+ ion is located at the cross point. $\langle r \rangle$ value indicates the average radial position of the Ho^{14+} ions. The other simulation parameters are the same as in Fig. 3 except for V_z and the intensity of the cooling laser (10 mW/cm^2)

as in Fig. 3a except for the applied asymmetric field of $E_x = 5$ V/m. We observe that a considerable increase of the average kinetic energy of both Be^+ and Ho^{14+}. Since the positions of the ions shift from the trap axis by the static electric field, the average kinetic energy of Ho^{14+} drastically increases by micromotions. At the quasi-equilibrium state, the average radial position and the kinetic energy of Ho^{14+} are 4.3(2) μm and 2.4 K, respectively.

Next we tested the axial voltage dependence of sympathetic cooling efficiency. As shown in Fig. 4, the sympathetic cooling efficiency decreases with increasing V_z. By applying a higher V_z the distance between adjacent ions becomes shorter and the radial positions of the ions easily deviate from the trap axis by ion-ion collisions. Then the rf heating occurs on the HCIs, as mentioned in the previous section. As a result, the final average-kinetic energy of Ho^{14+} drastically increases.

It may be worth mentioning about the influence of initial conditions in MD simulations. Figure 5 shows the reproduce figure of the observed arrangement of the ion Coulomb crystal consisting of 10 Ho^{14+} and a single Be^+. A colored circle indicates an approximated position of each ion. These arrangements were obtained by randomly changing the initial positions of the ions in the MD simulations. As expected the final kinetic energy of Ho^{14+} ions possibly depends on the ion arrangement. In this point there is room for further investigation.

Finally we tried to increase the number of cold Ho^{14+} ions by sympathetic cooling with a few Be^+ ions. One of the simulation results is shown in Fig. 6. In this example, we performed the MD simulation of 16 Ho^{14+} and 3 laser-cooled Be^+ ions. The final average kinetic energy of Ho^{14+} is sufficiently lower than 1 mK at the quasi-equilibrium state. However, we anticipate that it is difficult to increase further the number of cold Ho^{14+} ions by increasing the number of Be^+ ions, because the increase of the space charge caused by Ho^{14+} and Be^+ ions easily induces the rf heating of the ions. The solution to this problem is the use of a segmented linear Paul trap. That is, it is possible to increase the number of cold Ho^{14+} ions up to over 100 by connecting 10 segmented linear Paul trap in series. This idea can be realized by applying the similar method described in Ref. [19] to a linear Paul trap.

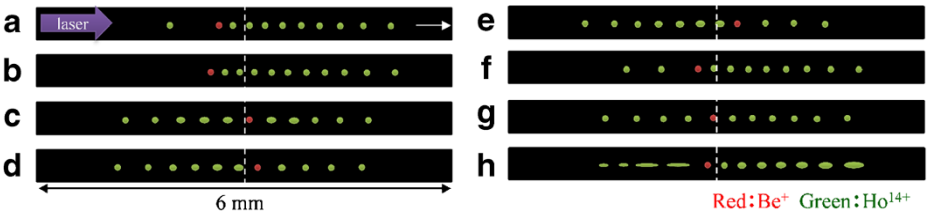

Fig. 5 Reproduce figures of the observed arrangements of the linear ion Coulomb crystal containing 10 Ho^{14+} (*green*) and a single Be^{+} (*red*). The colored circle indicates the approximated position of each ion. The vertical dotted line shows the center of the trap

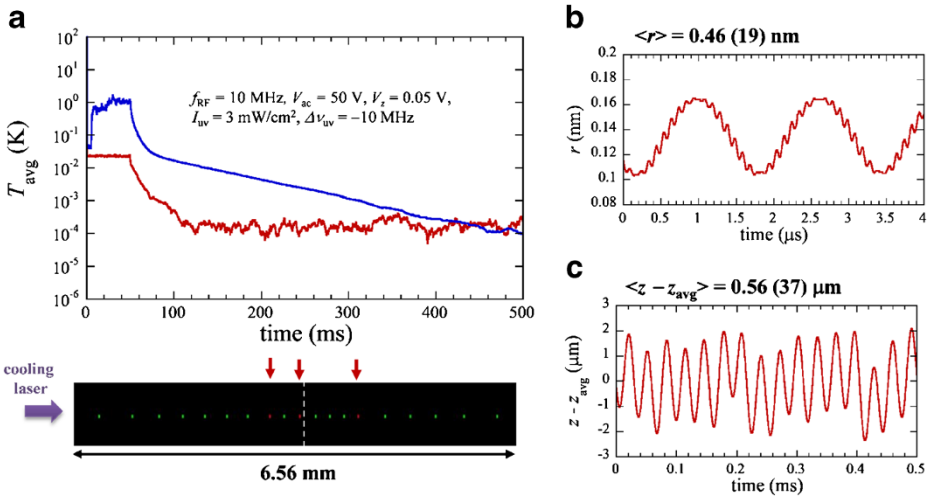

Fig. 6 Sympathetic cooling of 16 Ho^{14+} ions with 3 laser-cooled Be^{+} ions. (a) Plots of the average kinetic energies of 16 Ho^{14+} and Be^{+} ions. The final average kinetic energy of the Ho^{14+} ions is $1.3(8)\times10^{2}$ μK. The lower image shows the ion arrangement in the quasi-equilibrium state. The red arrows in the image indicate the positions of the Be^{+} ions. The simulation parameters are shown in the figure and the other parameters are the same as in Fig. 1. Figs (b) and (c) show example plots of the time variation of r and $z-z_{\mathrm{avg}}$ of a certain Ho^{14+} ion. We also show the average values of r and $z - z_{\mathrm{avg}}$ at the quasi-equilibrium state

4 Summary

In summary we have performed MD simulations to find out the conditions for efficient sympathetic cooling of HCIs in a linear Paul trap. Our extensive simulations show that the space charges of HCIs play important roles for the determinations of the axial distribution and the sympathetic cooling limit of the ions. Moreover, we found that a single laser cooled Be^{+} can generate sufficiently cold at least 10 Ho^{14+} ions in a linear Paul trap with the ideal trapping fields, and the number of cold HCIs can increase using the segmented linear Paul trap in series. Since cold HCIs produced by sympathetic cooling are promising candidates for probing possible time variation of the fine structure constant [5–7, 13], further simulation studies will be performed in future.

Acknowledgements We thank to H. Katori for helpful comments and suggestions. This work is financially supported in part by a Grant-in-Aid for Young Scientists (A) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT).

References

1. Okada, K., Suganuma, T., Furukawa, T., Takayanagi, T., Wada, M., Schuessler, H.A.: Phys. Rev. A **87**, 043427 (2013)
2. Hall, F.H.J., Aymar, M., Bouloufa-Maafa, N., Dulieu, O., Willitsch, S.: Phys. Rev. Lett. **107**, 243202 (2011)
3. Schiller, S., Korobov, V.: Phys. Rev. A 71, 032505 (2005)
4. Kajita, M., Gopakumar, G., Abe, M., Hada, M., Keller, M.: Phys. Rev. A 89, 032509 (2014)
5. Berengut, J.C., Dzuba, V.A., Flambaum, V.V.: Phys. Rev. Lett. 105, 120801 (2010)
6. Safronova, M.S., Dzuba, V.A., Flambaum, V.V., Safronova, U.I., Porsev, S.G., Kozlov, M.G.: Phys. Rev. Lett. **113**, 030801 (2014)
7. Safronova, M.S., Dzuba, V.A., Flambaum, V.V., Safronova, U.I., Porsev, S.G., Kozlov, M.G.: Phys. Rev. A **90**, 052509 (2014)
8. Mäckel, V., Klawitter, R., Brenner, G., Crspo López-Urrutia, J.R., Ullrich, J.: Phys. Rev. Lett. **107**, 143002 (2011)
9. Versolato, O.O.M., Schwarz, M., Windberger, A., Ullrich, J., Schmidt, P.O., Drewsen, M., Crspo López-Urrutia, J.R.: Hyperfine Interact. **214**, 189 (2013)
10. Schwarz, M., Versolato, O.O., Windberger, A., Brunner, F.R., Ballance, T., Eberle, S.N., Ullrich, J., Schimidt, P.O., Hansen, A.K., Gingell, A.D., Drewsen, M., Crespo López-Urrutia, J.R.: Rev. Sci. Instrum. **83**, 083115 (2012)
11. Schmöger, L., Versolato, O.O., Schwarz, M., Kohnen, M., Windberger, A., Piest, B., Feuchtenbeiner, S., Pedregosa-Gutierrez, J., Leopold, T., Micke, P., Hansen, A.K., Baumann, T.M., Drewsen, M., Ullrich, J., Schmidt, P.O., Crespo López-Urrutia, J.R.: Science **347**, 1233 (2015)
12. Versolato, O.O. et al.: To be appeared in Hyp. Int. of this volume
13. Dzuba, V.A., Flambaum, V.V., Katori, H.: Phys. Rev. A 91, 022119 (2015)
14. Okada, K., Wada, M., Takayanagi, T., Ohtani, S., Schuessler, H.A.: Phys. Rev. A **81**, 013420 (2010)
15. Okada, K., Yasuda, K., Takayanagi, T., Wada, M., Schuessler, H.A., Ohtani, S.: Phys. Rev. A **75**, 2007
16. Blythe, P., Roth, B., Fröhlich, U., Wenz, H., Schiller, S.: Phys. Rev. Lett. **95**, 183002 (2005)
17. Feldker, T., Pelzer, L., Stappel, M., Bachor, P., Steinborn, R., Kolbe, D., Walz, J., Schmidt-Kaler, F.: Appl. Phys. B **114**, 11 (2014)
18. Zhang, C.B., Offenberg, D., Roth, B., Wilson, M.A., Schiller, S.: Phys. Rev. A **76**, 012719 (2007)
19. Fujitaka, S., Wada M., et al.: Nucl. Instrum. M.th. B **126**, 386 (1997)