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## Optical spectroscopy in ion traps

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**Abstract.** Spectroscopic experiments in static or dynamic ion traps have produced results of extremely high precision. They have been, however, performed mainly on systems with simple electronic level schemes as available in singly charged ions of the earth-alkaline elements. In this contribution the most complex system investigated so far,  $Eu^+$ , is used as example to discuss the possibility of spectroscopy on superheavy elements in traps.

**PACS.** 32.80. Pj Optical cooling of atoms; trapping – 33.80. Ps Optical cooling of molecules; trapping – 39.30.+w Spectroscopic techniques

#### **1** Introduction

Excitation energies, lifetimes of excited states, fine- and hyperfine-separations, and electronic q factors provide information on the atomic structure. Different experimental techniques like laser absorption spectroscopy on atomic species in cells, collinear beam laser spectroscopy, resonance ionisation spectroscopy, or microwave spectroscopy have been employed and a wealth of data have been collected in the past. Ion traps entered this field since about 20 years and have been shown to be extremely useful instruments, particularly when high precision is required. They have been applied to the investigation of simple systems such as alkali-like atoms as well as ions with more complex electronic level structure. Based on the experience with such systems one may estimate whether they may also contribute to the investigation of atomic properties of superheavy elements.

In this contribution I will briefly review some features of ion traps as far as they are of relevance for optical spectroscopy. Examples of spectroscopic experiments will be presented and conclusions for the application on superheavy elements are drawn.

#### 2 Features of ion traps

Two types of traps for charged particles are in use for spectroscopy [1,2]: Paul traps which use radiofrequency electric fields for dynamical 3-dimensional confinement, and Penning traps using static voltages for particle trapping in one (axial) direction and a strong magnetic field for confinement in the radial plane. Conventional traps use hyperbolic shaped electrodes to create a potential in the



Fig. 1. Linear Paul trap (left) and open endcap cylindrical Penning trap (right). A radio-frequency voltage  $V \cos(\omega t)$  applied to the rods of the linear Paul trap with opposite rods electrically connected confines charged particles in the radial direction, a positive *d.c.* voltage at the end segments serves for axial trapping. In the Penning trap the end cap electrodes are at positive potential with respect to the central ring electrode. Additional guard electrodes between the ring and the end caps serve for partially compensation of deviations from the ideal quadrupole potential near the trap center. Ion excitation can be performed by *r.f.* fields applied to segments of the electrodes.

space between the electrodes which depends on the square of the coordinates. This leads to linear forces and the motion of a single charged particle inside such a potential can be described analytically. Often other electrode configurations are used such as cylindrical Penning traps [3,4] or linear Paul traps [5,6] (Fig. 1). They have the advantage that they are easy to manufacture and to align, and in addition they allow optical access into the inner region without further modification. Sizes of such traps range from below a mm to several cm in diameter. The fact that the forces are no longer linear is of no importance as long as they serve just as container for the charged particles and no detailed knowledge of their motion is required.

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The depth of the confining potential in Penning traps is equal to the static potential difference between the trap center and the endcap electrodes, in Paul traps it depends mainly on the frequency and the amplitude of the r.f. trapping field, typically at about 1 MHz and several 100 V, respectively. Potential depth between 1 and 50 eV are easily obtained. The total number of ions which can be confined in such a potential is limited by space charge. For the parameters discussed above a density of  $10^6$  cm<sup>-3</sup> is a typical number.

The average kinetic energy of stored ions depends on the trapping conditions. As a rule of thumb we may assume 1/10 of the potential depth when no cooling method is applied [7]. This is a fairly high value compared to the kinetic energy of atoms under normal conditions, corresponding to temperatures of several 10 000 K. It leads generally to large first order Doppler broadening and prevents accurate determinations of excitation wavelength. When, however, the ion motion is cooled, the Doppler width reduces to values below the natural linewidth and high accuracy is possible.

Ion cooling can be obtained in different ways: laser cooling [8,9] uses the recoil of absorbed photons from a laser tuned slightly below a resonant excitation energy. Very low ion temperatures below 1 mK have been obtained [10,11], the method, however, requires a repetitive excitation from the ground state to an excited level. For this we need a 2-level system which is available only in few atomic systems. Superheavy elements, in particular, have in general many long lived metastable states into which an excited state can decay. Then the ion is lost from the absorption-reemission cycle and no further cooling takes place. Cooling by Coulomb interaction with a simultaneously confined ion which can be laser cooled is a possible solution [12]. It will, however, not discussed further here since it is subject to other contributions at this conference [13].

An alternative way to reduce the trapped ions energy is resistive cooling [14]. Here a tank circuit connects two electrodes. When the ions oscillates in the trap at the same frequency as the resonance frequency of the circuit the image charges induced in the trap electrodes by the oscillating ions leads to a current through resonance impedance R of the circuit and the associated thermal energy is dissipated to the environment until the ions temperature matches that of the tank circuit. Although this has been used successfully in several cases [15, 16], problems may occur when applied to superheavy elements: the time constant for exponential energy loss is given by

$$\tau = \left(\frac{2z_0}{q}\right)^2 \left(\frac{m}{R}\right)$$

where  $2z_0$  is the distance between the endcaps, q and m the charge state and mass of the ion, respectively. When we assume a quality factor Q of the tank circuit of 100, a trap size of 1 cm, and an ion mass of 200 a.u. we arrive at a time constant of about 15 min. Obviously this can not be applied to short lived species. Moreover it requires a well defined ion oscillation frequency. This may be difficult



Fig. 2. Laser excitation of the  $6S_{1/2}-6P_{1/2}$  resonance line in stored Ba<sup>+</sup> ions. Left: under ultra-high vacuum conditions, right: background pressure  $10^{-5}$  mbar (N<sub>2</sub>). At UHV conditions most of the ion population is trapped in a long lived metastable state, collisions with background molecules quench the metastable state and the ions return to the ground state in a short time (from Ref. [18]).

to obtain when large clouds of ions are confined because of the changing space charge potential during the cooling process. For ions of superheavy elements the potential can be always considered very likely as sufficiently harmonic because of the small production rate and the corresponding low space charge density.

Cooling by collisions with neutral buffer gas molecules [17] is rather easy to obtain. Although not as effective as the other cooling methods mentioned above it has the additional advantage that collisions reduce the lifetime of metastable states into which a laser excited ion may decay. Thus it increases the number of ions in the ground state and consequently leads to higher signal strength when photon absorption is detected by reemitted fluorescence quanta. Figure 2 shows an example taken on  $Ba^+$  ions which has a very long lived metastable  $5D_{3/2}$ state into which the laser excited  $6\mathrm{P}_{1/2}$  state decays with 30% probability [18]. While at ultra-high vacuum conditions almost no fluorescence photons are detected in a laser sweep across the  $6S_{1/2}$ – $6P_{1/2}$  resonance, the signal becomes much larger when buffer gas is introduced into the system, since collisions reduce the effective lifetime of the metastable level from 80 s to several 10 ms.

Traps have the utmost sensitivity allowing investigations on single particles. This arises from the fact that after resonant optical excitation at an allowed electric dipole transition the excited state decays in a time of typically  $10^{-8}$  s. If this decay leads to the original electronic state the ion can be excited again leading finally to the emission of about  $10^8$  photons per second. Even if we consider finite solid detection angles, transmission losses and limited detector efficiencies we generally arrive at signals of about  $10^4-10^5$  detected photons per second — a rather strong signal from a single particle. The conditions of a



Fig. 3. Fluorescence spectra from a laser excited cloud of  $Eu^+$  ions confined in a Paul trap. The cloud of about  $10^5$  particles is a mixture of the stable isotopes  $^{151}Eu^+$  and  $^{153}Eu^+$ . The insert shows the hyperfine components of the levels involved in the transition (from Ref. [19]).

closed two level energy diagram as required for repetitive excitation, however, is fulfilled only in few cases. Particularly for superheavy elements one expects many low lying levels into which an excited state may decay. Then the ion is lost from the excitation-reemission cycle until it returns to the ground state. The fluorescence signal strength then is determined by the lifetime of the excited state. If this is of the order of ms, the signal strength is reduced by a factor of  $10^5$  compared to the 2-level case. The typical signal rate then would of the order of  $1 \text{ s}^{-1}$  per trapped ion. With a few ions in the trap the signal is comparable to the dark current of sensitive photon detectors and averaging for a few seconds would result in a detectable signal.

#### 3 Spectroscopy on complex ions

Almost exclusively spectroscopy on trapped ions has been performed on ions with simple electronic level structures. The only exception to date is the case of Eu<sup>+</sup> which may serve as an example for potential spectroscopy on superheavy elements.  $Eu^+$  has a  ${}^9S_4$  ground state. The stable isotopes of Eu (mass 151 and 153) have a nuclear spin of 5/2 giving rise to many hyperfine sublevels by coupling to the electronic angular momentum J = 4. The ground state can be excited to a  ${}^{9}P_{5}$  level at 382 nm by a laser entering the trap through a hole in one of the electrodes. The excited state decays back either to the ground state or into several long lived metastable  $^9\mathrm{D}$  states. Buffer gas collisions reduce the effective lifetime of these states to the ms range. Fluorescence is detected at a  ${}^{9}P_{5}-{}^{9}D$  transition at 664 nm perpendicular to the laser beam free of laser stray light. The fluorescence spectrum, taken in a Paul trap, shows several partly resolved hyperfine components (Fig. 3) [19]. The Doppler width of the lines is 3.7 GHz, corresponding to an ion temperature of 7000 K.



Fig. 4.  $\Delta m = 0$  manyfold of the microwave induced F = 11/2 - F' = 13/2 hyperfine transition in the <sup>9</sup>S<sub>4</sub> ground state of <sup>151</sup> Eu<sup>+</sup> (from Ref. [19]).

In spite of the low resolution in the optical spectrum high accuracy can be obtained in optical-microwave double resonance experiments: the laser can be tuned to one of the hyperfine components and depletes selectively the corresponding ground state hyperfine level by optical pumping. This results in a decrease of the fluorescence intensity. A microwave induced transition to one of the adjacent hyperfine level repopulates the depleted state. Resonance is detected by an increase in the fluorescence intensity. Figure 4 shows as an example the F = 11/2 - F' = 13/2hyperfine transition which is split by the ambient magnetic field of a few Gauß in Zeeman sublevels.

The line center of the transitions can be determined to a few 10 Hz corresponding to a fractional uncertainty of below  $10^{-8}$ . First order Doppler broadening does not show up in spite of the high ion temperature because of the Dicke effect: when the wavelength of the radiation is larger than the oscillation amplitude of the ions in the trap the Doppler effect results in well resolved sidebands of the spectrum at the ions oscillation frequencies. The Dicke criterion is easily fulfilled in the microwave domain without any cooling. Similar spectra as shown in Figure 4 have been obtained for all possible  $\Delta F = 1$ ,  $\Delta m = 0$ ,  $\pm 1$  transitions. From the transition frequencies the magnetic dipole and electric quadrupole constants A and Bhave been derived with uncertainties of  $10^{-8}$  and  $10^{-4}$ , respectively. Similar results have been obtained for several unstable long lived isotopes of Eu [20].

Similarly in a Penning trap with a superimposed strong magnetic field Zeeman transitions can be induced by optical-microwave double resonance [21]. Figure 5 shows an example of induced nuclear Zeeman transitions in <sup>151</sup>Eu<sup>+</sup> [22]. It has led to a determination of the nuclear magnetic moment with  $4 \times 10^{-4}$  uncertainty, one order of magnitude more precise than previously known.



Fig. 5. Induced nuclear Zeeman transition  $(m_J = 4, m_I = 3/2 - m'_I = 5/2)$  in trapped <sup>151</sup>Eu<sup>+</sup>at 6.55 GHz The fractional uncertainty of the line center is  $5.6 \times 10^{-8}$  (from Ref. [22]).

# 4 Prospects for spectroscopy on superheavy elements

The example of Eu demonstrates that even in cases where the electronic level structure is rather complex as expected in superheavy elements, precise information can be obtained which may serve for tests of atomic physics calculation. Since superheavies will not be available in large quantities the practical question remains how many particles are reasonably needed for experiments. As mentioned above even small numbers of trapped ions will lead to observable fluorescence signals. The number actually depends critically on the choice of background pressure conditions because ion-neutral collisions determine to which extent the bottleneck of long lived metastable states is removed. Sympathetic cooling of the ions under investigation with other laser cooled species would increase the spectral and spatial overlap between the trapped ions and the exciting laser beam and ultimately even single ion spectroscopy may be possible [13]. When the production of the elements is performed in short pulses the ions can be caught in flight by switching of the potentials with nearly 100% efficiency as demonstrated first [23] with the ISOLTRAP mass spectrometer now installed at ISOLDE/CERN and routinely used also at several other ion trap facilities. Continuous production and injection into the trap requires friction, most easily by collisions with buffer gas atoms. At background pressures of a few times  $10^{-6}$  of He, an efficiency exceeding 10% has been obtained [24].

An other way to bring ions into the trap is to collect them on a filament which then can be placed in an opening of one of the trap electrodes. Heating of the filament produces ions by surface ionisation when the work function of the material exceeds the ionisation potential of the element. This can be easily fulfilled for superheavy elements. As an example we may take experiments on  $^{131}$ Ba<sup>+</sup> (lifetime 11 d) where a sample of  $10^{13}$  atoms were collected on a Pt filament at the ISOLDE facility and transported to the laboratory in Mainz. After several days of ultra-high vacuum preparation and test measurements a trap could be filled about 100 times over a period of one month with  $10^5$  ions and measurements of very high precision with good signal/noise ratio, similar as in the case of stable isotopes, have been performed successfully [25].

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