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Invited Paper

High-resolution microwave spectroscopy on trapped ion clouds

G. Werth

Institut für Physik, Universität Mainz, D-55099 Mainz, Germany
(Fax: +49-6131/395169)

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Abstract. Ion traps are particular usefully devices for precision spectroscopy on ionic ground states in the microwave domain. Although ultimate precision is achieved only with laser-cooled single ions, in many cases the precision obtained using large uncooled clouds of ions is sufficient for many requirements in atomic physics. The stronger signal in this case makes possible experiments on forbidden transitions or on systems with complex spectra and many substates. Recent examples of laser-microwave double resonance spectra on Pb^+ and Eu^+ are presented along with attempts to laser-cool a large ion cloud in order to reduce uncertainties from the second-order Doppler effect.

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Ion storage by electromagnetic fields has been an important tool for high-precision experiments for more than a decade. Paul traps use time varying electric fields for confinement and Penning traps use a combination of static electric and magnetic fields. Both devices are characterized by collision free storage of a sample that is isolated in space and essentially free of uncontrollable perturbations such as collisions with walls or background gas molecules or perturbing external fields. These nearly ideal conditions allow extremely long observation times of the sample. In addition, the increased detection efficiency of scattered photons allows the observation of a single isolated ion which can be driven into the trap center where electric fields are zero [1]. The potential of this method has been demonstrated in a series of high-precision measurements, e.g., of hyperfine transition frequencies in alkali-earth elements, using Paul traps [2]. Such experiments are the basis for proposals of novel frequency standards, whose realizations are actively persued at present and have already achieved a level which is comparable to the hydrogen maser and the cesium clock, the most stable and accurate frequency sources to date [3]. In Penning traps, a large magnetic field is required to provide confinement. This leads, of course, to large frequency shifts in hyperfine transitions, the device has been used, however, for precise determination of electronic and nuclear g-factors [4].

The ultimate precision, as required for frequency standard applications, can be obtained only with a single trapped ion that is cooled to extremely low temperatures in the μK range. This virtually suppresses all influences on the spectrum caused by the ion motion. For many applications in atomic spectroscopy, however, such high precision is not at all required, since atomic physics calculations do not reach the same level of accuracy. In such cases large ion clouds may be more advantageous than single particles since they offer much higher signal-to-noise ratios. The ions then, of course, will move in regions of high electric fields and Doppler effects may limit the spectral resolution. Fortunately, for hyperfine measurements in the radiofrequency or microwave domain this happens only in second order, since even for large ion clouds of several mm diameter the ion oscillation amplitude generally is smaller than the wavelength of the radiation. In this case, as shown by Dicke [5], the first-order Doppler effect shows up in sidebands at the ion oscillation frequencies, symmetrically around an unshifted and unbroadened central carrier. Second-order Doppler shifts, which limit the precision in these cases, are of the order of 10^{-11} and are usually negligible for calculable effects in atomic physics.

Large ion clouds are particular useful when experiments are performed on ions whose level scheme differs from the rather simple structure of the alkaline-earth ions. One main difference arises if the ion has no strong electric dipole allowed transitions in the range of available lasers. The use of weak electric quadrupole or magnetic dipole transitions in optical experiments requires many ions to obtain a sufficient number of scattered photons. Similar problems occur if the electronic and nuclear angular momenta are large, leading to a large number of hyperfine or Zeeman sublevels. Then many stored ions are necessary to insure sufficient population in each of the sublevels. The maximum number of ions

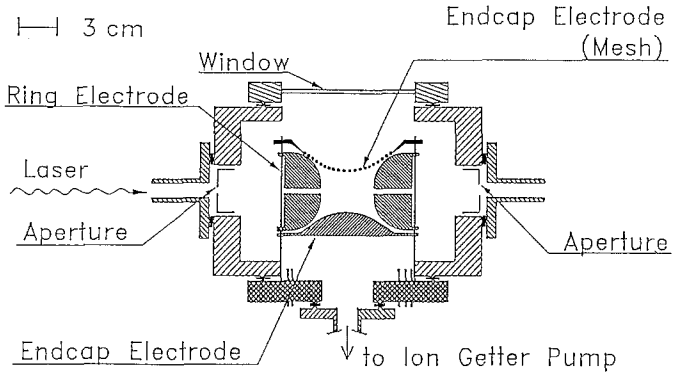

Fig. 1. Paul ion trap for laser and microwave spectroscopy

in a cloud is given by the equilibrium between the confining forces of the electric trapping field and the Coulomb repulsion between the ions. The highest densities for trap potential minima in the energy range of 10 eV are about 10^6 ions/cm^3. In thermal equilibrium the cloud assumes a Gaussian shape whose half width is about 1/10 of the trap diameter [6].

In this article I will present recent results obtained in such cases just described, where departure from the alkali-like level scheme of previously considered ions lead to substantial experimental difficulties. As will be shown below, the precision of such experiments does not match that of the alkaline-earth ions, but is sufficient for general atomic physics purposes and may indicate the possibility of precision spectroscopy on complex systems which may be of interest to answer specific questions.

The specific properties of the ion motion is of little interest in these experiments and the trap acts as a mere container for the particles. Therefore I will not describe the traps in detail. In all cases discussed below we have used a Paul trap with hyperbolic electrodes (Fig. 1), whose time-dependent quadrupole potential $\varphi(t)$ is given by

$$\varphi(r, z, t) = -\frac{m\Omega^2}{16}(a_z - 2q_z \cos \Omega t)(r^2 - 2z^2),$$

with $r^2 = x^2 + y^2$ and

$$a_z = -\frac{8eU}{mr_0^2\Omega^2} = -2a_r, \quad q_z = \frac{4eV}{mr_0^2\Omega^2} = -2q_r$$

as dimensionless stability parameters. Here r_0 is the radius of the traps ring electrode, $z_0 = r_0/\sqrt{2}$ the distance of the traps endcaps, U, V are the applied dc and ac voltages, respectively, and $\Omega/2\pi$ is the trap's driving frequency. Stable operation of the trap is given for certain values of a_z and q_z, derived from the solution of the equation of motion and extensively discussed in the literature [7]. In our actual experiment we use traps of 4 cm and 2 cm diameter, driving frequencies between 0.5 MHz and 3 MHz, dc voltages of a few volts and rf amplitudes of a few hundred volts. The stability parameters usually are close to the optimum value $a_z = -0.03$ and $q_z = 0.5$ [8].

1 The role of collisions in stored-ion spectroscopy

Ultrahigh-vacuum conditions, as required for ideal trap conditions, would lead to rather fast ion loss from the trap in the case of large ion clouds: ion-ion collisions change the phase of the ion motion with respect to the oscillating driving field. This leads to energy pick-up from the electric trapping field. In our cases we generally found exponential ion losses from the trap with a time constant of about 10 min when we operate a 4 cm diameter trap near the optimum trapping point. Such ion loss can be prevented by the use of buffer gases. Collisions with neutral background atoms damp the ions oscillation amplitude and leads to longer storage times. According to Major and Dehmelt [9], cooling occurs if the mass of the atom is smaller than that of the stored ion. In practice we find it necessary to have a mass ratio of at least 1:4 between the background gas atom and the ion in order to have effective cooling. We generally use He, H_2 or N_2 as buffer gas atoms. At pressures of about $10^{-5}-10^{-6}$ mbar we then achieved storage times of more than a day, and several weeks in the case of very heavy ions like Pb^+. The ion's temperature determined from the Doppler width of optical absorption lines is reduced by about one order of magnitude from an initial temperature in the range of 2.5×10^4 K | 10 |.

Besides cooling of the ion cloud, ion-atom collisions may also be beneficial if the ion has long-living metastable states. Here population trapping may occur, which substantially decreases the population of the ion's ground state. In the cases discussed below, the fluorescence from the ion cloud after laser excitation from a ground level is used for monitoring. To achieve a good signal-to-noise ratio, it is essential to bring the ions back to the ground state if they are trapped in the metastable states. If the number of metastable sublevels is small and the wavelength is in a suitable range, one may use a laser to repump the ions into a fast decaying state. This has been applied succesfully on ions like Ba^+, Ca^+, Sr^+ and Yb^+. However, in cases when the ion has many metastable states, laser repumping is not feasible and quenching by ion-atom collision must be used to reduce the effective lifetime of the ion's metastable states. Collisional quenching cross sections have been determined for several ion-atom combinations [11]. Typical orders of magnitude are 10^{-18} cm^2 for nobel gases and 10^{-16} cm^2 for molecules as neutral collision partners. Using pressures in the 10^{-6} mbar range, the effective metastable lifetime can be reduced from many seconds to a few tens of milliseconds [12].

The range of buffer gas pressure is limited, since collisions may also lead to a destruction of polarization or any deviation from thermal equilibrium population in ground state sublevels as required for microwave spectroscopy on these states. Fortunately, the relaxation rates in case of hyperfine or Zeeman sublevels is small in the pressure range around 10^{-6} mbar, particularly if noble gases are used as buffer gases. We have recently measured relaxation rates between $^{207}Pb^+$ ground state hyperfine levels for various gases and found cross sections of 10^{-14} cm^2 for molecules and 10^{-16} cm^2 for noble gas

atoms [13]. Similarly we have determined relaxation time constants for Zeeman sublevels in Ba^+ [4]. The observed time constants of many seconds are very well suited for very high resolution microwave spectroscopy.

2 Double resonance on forbidden transitions: $^{207}Pb^+$

The concept of double resonance spectroscopy on atomic ground levels has been applied succesfully in many cases in neutrals and in ions. By the proper choice of the polarisation of an incident resonant light beam a specific ground state sublevel can be selectively excited. This optical pumping scheme creates a non-thermal population of the ground state, which can be changed by a microwave field if it excites a transition to a different ground state sublevel. The second resonance can be monitored by the change in fluorescence intensity or polarization from the irradiated sample. Alternatively a ground state sublevel can be selectively excited by the proper choice of the wavelength of the light beam if the sublevel spacing is larger than the Doppler width of the optical transition and the spectral width of the light beam.

Fortunately this latter method can be applied in many cases in trapped ion spectroscopy. Although the temperature of uncooled ion clouds is in the range between 10^3 and 10^4 K, leading to Doppler widths of 1–2 GHz, very often the ground state hyperfine splitting exceeds this value and the hyperfine structure can be sufficiently resolved. Figure 2 shows an example in Pb^+, where even the excited level hyperfine structure is partially resolved. If we tune the laser to one of the hyperfine components, its intensity will go down by optical pumping, in the absence of relaxation, to zero. A microwave field which couples the depleted state resonantly to any other level will increase the population of the pumped state. The resonance thus can be observed by an increased fluorescence intensity.

In case of $^{207}Pb^+$, the only excitation from the $6P_{1/2}$ ground state to an excited level in the visible range is a fine structure transition to the $6P_{3/2}$ level at 710 nm. The fluorescence from this state occurs at the same wavelength and is very weak as evident from the 47 ms lifetime of this state. Since observation of the fluorescence is impossible in the presence of the exciting laser beam, we used a pulsed scheme where we detected the fluorescence in a time interval of the order of the lifetime when the exciting laser was blocked [14]. At low resolution we observe a spectrum consisting of the hyperfine transition and sidebands at the ion oscillation frequency, as predicted by the Dicke effect [5]. When we increase the resolution, we see Zeeman sidebands in the small magnetic field, arising from $\Delta m_F = 0, \pm 1$ transitions. The central resonance between the $F=1, m_F=0$ and $F=0, m_F=0$ sublevels is independent of magnetic field fluctuations and inhomogeneities to first order. Its linewidth is determined only by the microwave power and resolution (Fig. 3). In our experiments we obtained a minimum linewidth of 0.5 Hz in the 12.6 GHz transition frequency (Fig. 4) that was limited by the resolution and stability of our microwave generator. The ultimate resolution which could have been achieved is obtained when we plot the experimentally observed linewidth as a function of the microwave field amplitude. The extrapolation to zero

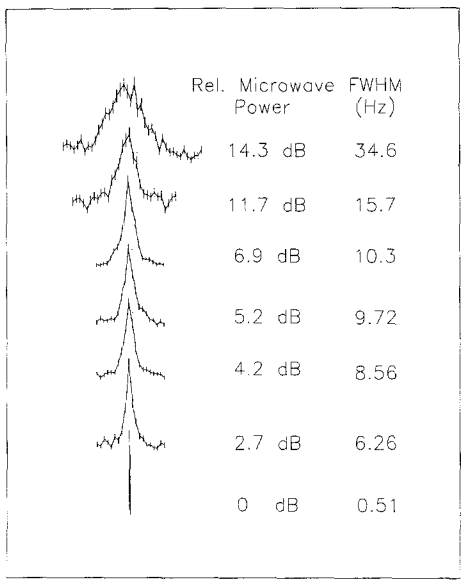

Fig. 3. $F=1, m_F=0$–$F=0, m_F=0$ hyperfine Zeeman transition of the $^{207}Pb^+$ ground state at different microwave powers

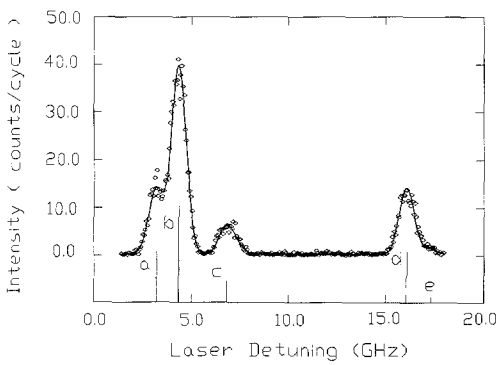

Fig. 2. Laser-induced fluorescence spectrum of the $6P_{1/2}-6P_{3/2}$ $M1$ transition of $^{207}Pb^+$ at 710 nm. Lines *a*, *b*, *d*, *e* are hyperfine components of $^{207}Pb^+$, line *c* is from a contamination by $^{208}Pb^+$. The stored ions are cooled by He buffer gas at 10^{-5} mbar

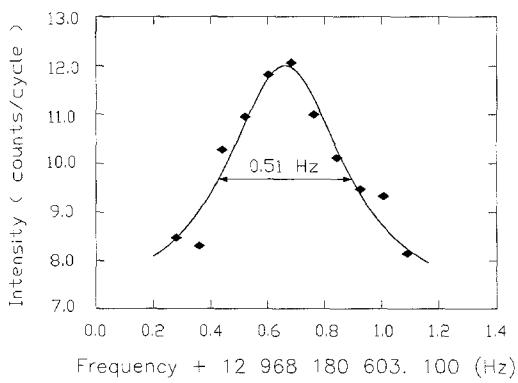

Fig. 4. Narrowest obtained hyperfine transition in $^{207}Pb^+$

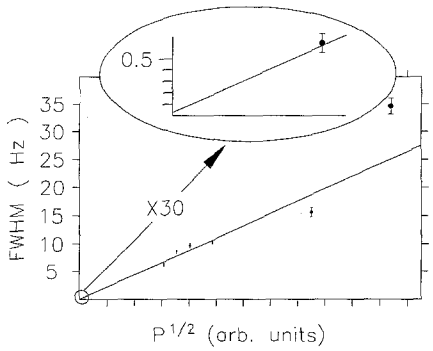

Fig. 5. Full linewidth of the microwave-induced hyperfine transitions in Pb^+ vs the square root of the power. The intersection at $P=0$ gives the minimal linewidth from relaxation by ion-ion or ion-molecule collisions

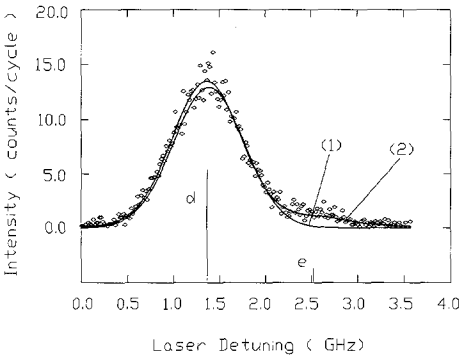

Fig. 6. Resolution of a small $E2$ admixture to the $6P_{1/2}-6P_{3/2}$ $M1$ transition. The rate of detected fluorescence photons in this component is about $1\ s^{-1}$. To emphasize the difference, labels (*1*) and (*2*) refer to fits by one or two Gaussians. *d* and *e* have the same meaning as in Fig. 2

amplitude gives the limit imposed by relaxations from ion-atom or ion-ion collisions. In our example (Fig. 5) we obtained $\Delta\ \nu_{min}=25$ mHz. The number of fluorescence counts is of the order of 10–50 s^{-1} from a sample of 10^5 ions at exciting laser powers of 50 mW and an overall fluorescence detection efficiency of 10^{-3}. To illustrate the sensitivity of this method we observed a small electric quadrupole admixture to the magnetic dipole amplitude in the $6P_{1/2}-6P_{3/2}$ optical transition, which leads to a $P_{3/2}$, $F=2$, $m_F=2-P_{1/2}$, $F=0$, $m_F=0$ component in the optical spectrum. The number of photons in this component is about $1\ s^{-1}$ (Fig. 6) [15].

3 Double resonance in complex atomic ions: Eu^+

Coupling of high nuclear spins and electronic angular momentum leads to many hyperfine sublevels, which requires the use of many ions to obtain a sufficient population in each of the sublevels. The stable isotopes of Eu^+ with masses 151 and 153 are the first examples of a complex system where double resonance in ion traps has been applied. The nuclear spin of each isotope is 5/2 and the total angular momentum is $J=4$ for the ground state. Hyperfine sublevels, which range from $F=13/2$ to $F=3/2$, are split in addition into many Zeeman sublevels in a residual magnetic field at the trap's position. Since the total angular momenta F are half integer, no substate $m_F=0$ exists and all transitions are subject to magnetic field shifts and possibly broadenings in first order. Careful shielding of the B-field is required to obtain high spectral resolution, which may be somewhat difficult if the ion cloud occupies a volume of the order of 1 cm^3 and if the B-field is inhomogeneous. The optical spectrum of a natural mixture of Eu^+ isotopes [16] (Fig. 7) shows, that the resolution is just good enough to identify the different hyperfine components in the $^9S_4-{}^9P_5$ electric dipole allowed transition at 382 nm. The ions here are moderately cooled by He buffer gas at 10^{-5} mbar. The excited $^9P_5$ level decays into three of several ^9D metastable states which have an estimated radiative lifetime of many seconds. The collisions with background atoms reduce this lifetime to about 100 ms. By optical pumping each of the hyperfine ground levels can become selectively depleted. Microwave-induced transitions to neighbouring hyperfine levels are observed by the change in fluorescence intensity from the excited $^9P_5$ level. To avoid the laser straylight background, we used the decay of this state into the $^9D_6$ level at 650 nm. This $^9D_6$ level contains 1 out of 10 decay photons. Shielding the magnetic stray field to values below 200 mG by 3 pairs of Helmholtz coils allows the resolution of the Zeeman sublevels. Figure 8 shows an example of the $\Delta m_F=0$ part of the $F=13/2-F=11/2$ hyperfine transition of $^{151}Eu^+$. We obtained linewidths between 0.2 kHz and 1 kHz for all possible hyperfine transitions in both isotopes. This high resolution requires that the interpretation of the spectrum in terms of hyperfine coupling constants not only takes magnetic dipole and electric quadrupole interactions into account, characterized by the coupling constants A and B, respectively, but also higher

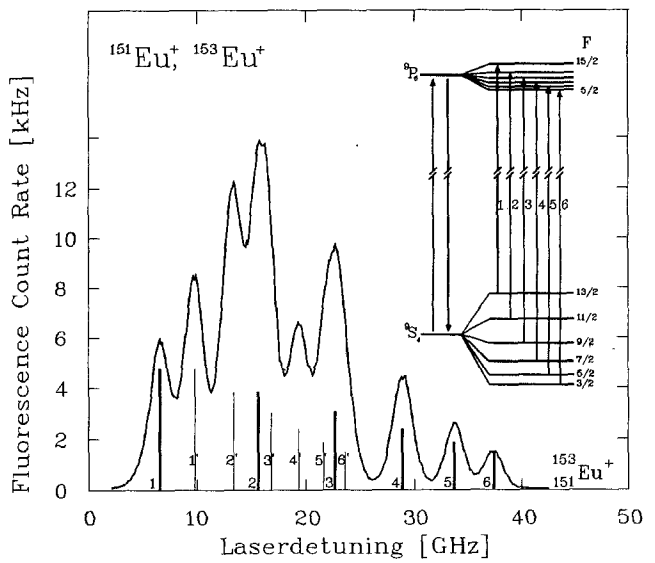

Fig. 7. Laser-induced fluorescence spectrum of a natural mixture of Eu^+ isotopes at 382 nm

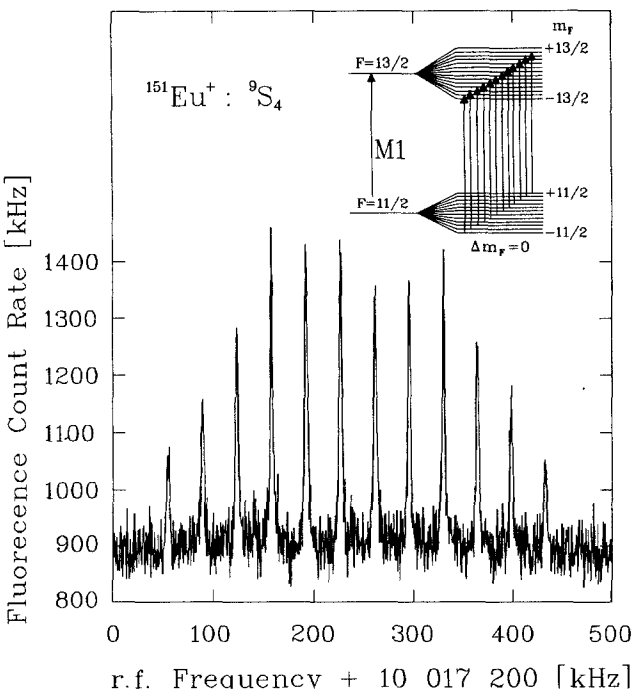

Fig. 8. $\Delta m_F = 0$ part of the $F = 13/2$–$F = 11/2$ ground state hyperfine transition in $^{151}Eu^+$. Similar spectra have been obtained for all other hyperfine transition in both odd stable isotopes of Eu^+

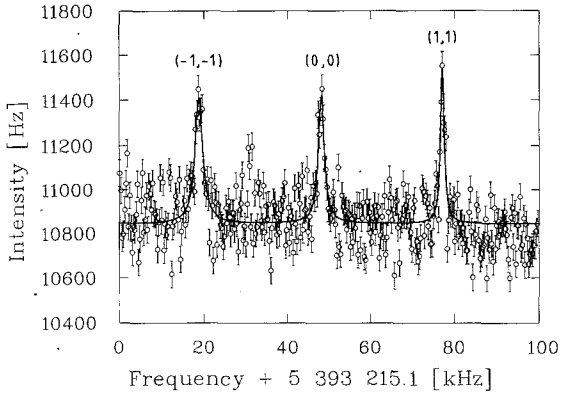

Fig. 9. Part of the $F = 9$–$F' = 8$ hyperfine – Zeeman spectrum in the ground state of $^{150}Eu^+$, obtained by laser-microwave double resonance, showing the 3 innermost lines. The labeling is $m_F \rightarrow m_{F'}$

order terms of the multipole expansion of nuclear moments. Equally important is the use of higher order perturbation theory to account for the influence of different excited states on the position of the ground state hyperfine levels. In our case we considered magnetic octupole interaction (C) and electric hexadecapole interactions (D) as additional higher-order terms. The perturbation theory included $M1$ and $E2$ interactions of the groundstate mainly with the $^7S_3$ excited state, but also with other $J = 3$ states and $^7D_7$ and $^7F_5$ levels.

We obtained a set of interaction constants which contains very high precision A- and B-factors as well as C- and D-constants with small error bars. This result demonstrates the feasibility of determining higher order nuclear moments in trapped ion spectroscopy (Table 1).

Making use of the high sensitivity of the ion trapping technique we are extending our measurements to unstable isotopes of Eu^+. Recently, we obtained first results on $^{150}Eu^+$, produced by high energy reactions at ISOLDE/CERN with a half life time of 36 years. This isotope has a nuclear spin of 5, leading to hyperfine levels of $F = 9$–$F = 1$. Using a total sample of 10^{13} atom collected at CERN, we observed optical and microwave spectra in the same manner as described above for stable isotopes. Figure 9 gives an example of a part of the $F = 9$–$F = 8$ microwave transition. At a total frequency of 5.3 GHz we presently achieved a resolution of 1.5 kHz, which is of the same order as with stable isotopes. The complex and time-consuming perturbation calculation to obtain the precise hyperfine coupling constants is under way.

This example shows that it is possible to extend precision hyperfine measurements to unstable isotopes. We estimate, that at least 10^{11} particles and lifetimes longer than one week will be the minimum requirement for other isotopes if they are to be investigated off-line from the production area. Results from those experiments on a chain of isotopes would allow the systematic comparison of nuclear shape influence on the hyperfine transitions. Together with values of the nuclear magnetic moments, which also may be obtained from trapped ion spectroscopy, values of the hyperfine anomaly

$$^1\Delta^2 = \frac{A_1}{A_2}\frac{\mu_2}{\mu_1} - 1$$

could be obtained. Subcripts 1,2 refer to two different isotopes, $A_{1,2}$ are their magnetic dipole interaction constants and $\mu_{1,2}$ their nuclear magnetic moments. Δ describes the deviation of the nuclear magnetization of an

Table 1. The hfs coupling constants for the magnetic dipole (A), electric quadrupole (B), magnetic octupole (C) and electric hexadecapole (D) interaction in the ground state $4f^7(^8S)\,6s$; $^9S_4$ of the two stabile isotopes $^{151}Eu^+$ and $^{153}Eu^+$

Hfs constant	$^{151}Eu^+$ [Hz]	$^{153}Eu^+$ [Hz]	$\frac{^{151}Eu^+}{^{153}Eu^+}$
A	1540297394 (13)	684565993 (9)	2.2500349 (6)
B	−660862(231)	−1752868(84)	0.37702 (19)
C	26 (23)	3 (7)	9 (28)
D	−6 (5)	−5 (2)	1.2 (1.2)

extended nucleus from that of a point-like nucleus (Bohr-Weisskopf effect).

4 Laser cooling of large ion clouds

The finite knowledge of the temperature represents in some cases a limitation to the precision of microwave spectroscopy on a stored ion cloud. Buffer gas collisions reduces the temperature from an inital value in the range of a few times 10^4 K to a value somewhat above room temperature but at the risk of inducing relaxation collisions which limit the spectroscopic resolution. The equilibrium temperature depends on the ion number and detailed characteristics of the trap. The ions near the boundary of the cloud experience strong electric fields and energy pickup leads to strong rf heating. If the number of ions is small they move near the center of the trap where electric fields are small. This has led to a number of experiments for laser cooling of small ion clouds, where the ions are irradiated by laser light tuned to the low frequency side of a strong optical transition [17]. With single ions, temperatures below 1 K have been obtained and for several ions crystallisation phenomena have been observed [18]. Larger ion clouds are much more difficult to cool because of the stronger rf heating. Furthermore, the ion cloud diameter may be larger than the laser beam diameter and the hot ions at the clouds boundary experience the cooling force only for a fraction of the time.

We have recently observed an indication of cooling of an ion cloud containing about 10^4 particles. The experiment was performed on Ca^+ ions confined in a 2 cm diameter trap. They can be excited by a $4S_{1/2}-4P_{1/2}$ resonance transition at 393 nm. A second laser at 866 nm is needed to repump the ions from a metastable $3D_{3/2}$ state into which the $4P_{1/2}$ level partially decays. When we tuned the 393 nm laser to the low-energy side of the Doppler-broadened transition we observed an increase of the fluorescence intensity with time as a result of the shrinking ion cloud diameter (Fig. 10). The time constant is of the order of 20 min indicating a small cooling force since the ions move outside the laser beam for most of their time. When we use similar times to sweep across a resonance, starting from the low energy side, we observe a narrowing of the Doppler linewidth from 1.5 GHz for fast sweep and essentially no cooling to about 400 kHz for slow sweep (Fig. 11). When the laser excites the ions at the high frequency side of the resonance, heating occurs. The increased ion oscillation amplitude leads to a decrease in fluorescence intensity, since the ions now spent again most of their time outside the laser beam. They are, however, not lost from the trap as it is the case for small ion numbers in miniature traps. A repetition of the cooling process with the same ion cloud yields identical results. The temperature of the ion cloud is about 100 K as determined by a Gaussian fit of the low frequency part of the resonance curve. The dependence of the final temperature on the ion number, the laser power, the detuning and the operating conditions of the trap remains to be investigated. In any case the temperature has been reduced by more than one order of magnitude, which substantially reduces the uncertainty from the second order Doppler shift in high-resolution hyperfine spectroscopy.

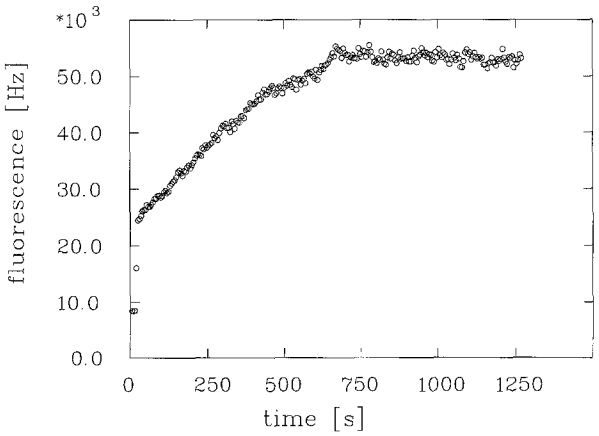

Fig. 10. Increase of fluorescence with time from a Ca^+ cloud stored in a Paul trap containing about 10^4 particles, when a laser is tuned to the low frequency side of the $4S_{1/2}-4P_{1/2}$ resonance line at 393 nm. The detuning was 400 MHz and the laser output power 10 mW. A second laser tuned to the $3D_{3/2}-4P_{1/2}$ transition was needed to repump the ions from the metastable $3D_{3/2}$ state. Note the time constant of 20 min

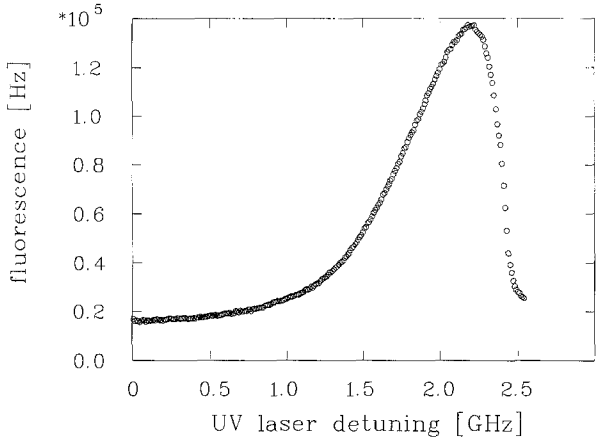

Fig. 11. Cooling of a cloud of 10^4 Ca^+ ions in a Paul trap. The cooling laser is swept at a speed of 0.5 MHz/s. At the high frequency side of the resonance curve, heating occurs and the ion cloud expands, leading to a decrease in fluorescence

5 Conclusion

The use of large stored ion clouds in high resolution microwave spectroscopy using optical pumping for state preparation is necessary if either the ion has only very weak optical transitions in the range of available laser radiation or if the energy level structure is complex and exhibits a large number of substates. Experiments on Pb^+ and Eu^+ ions serve as first examples for those ions. In the first case a spectral resolution of 2.5×10^{10} has been obtained, although the detected fluorescence count

rate was as low as a few photons per second. The limitation was given simply by the resolution of the microwave oscillator. The resolution on Eu^+ was about 10^8, which could be improved by more careful shielding of the stray magnetic field in the trap, but which is high enough to determine magnetic dipole and electric quadrupole hyperfine coupling constants with high precision. The sensitivity of the optical detection allows the extension of the experiments to unstable isotopes where only minute quantities are available. This may yield new information on the distribution of magnetization over the nuclear volume by comparison of many isotopes of the same element.

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