Population Trapping in Excited Yb Ions

H. Lehmitz, J. Hattendorf-Ledwoch, R. Blatt,[a] and H. Harde
Fachbereich Elektrotechnik, Universität der Bundeswehr Hamburg, Holstenhofweg 85, D-2000 Hamburg 70, West Germany
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Resonant laser excitation of Yb ions confined in an rf ion trap is found to result in perfect population trapping in the highly metastable $^2F_{7/2}$ state. The lifetime of this level is measured to be greater than 8 d. Population and depopulation of this level under narrow- and broad-band laser excitation conditions is investigated and its potentiality for ultimate-resolution laser spectroscopy indicated.

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In recent years the ion-storage technique has disclosed a new dimension of fundamental and high-accuracy experiments. So with single trapped ions the phenomenon of quantum jumps was demonstrated,[1-3] and very recently optically cooled ions were observed to form a crystal structure in the trap potential.[4,5]

Ion traps are also promising tools for ultimate-resolution laser spectroscopy because the ions can be observed for long periods without perturbations and therefore transit broadening as well as collisional effects can largely be eliminated. Because of these properties, ion traps are widely discussed as new frequency standards in the microwave as well as in the optical region.[6-12] For these applications, exceptionally well-defined atomic transitions are of interest which in the microwave range can be found between hyperfine levels of the ground state and in the optical reigon between long living excited states and the ground state. A variety of ions, especially the group-II*A* -II*B*, and -III*A* elements, but also Li^+ and Yb^+ have been investigated in the trap, and excited-state lifetimes as great as several tens of seconds have been measured.[1,10,13-16]

In this paper we report the first observation of nearly perfect population trapping of Yb ions in the metastable $4f^{13}6s^2\,{}^2F_{7/2}$ state. This state is found to possess an extraordinary long lifetime for an excited atomic state of greater than 8 d. Population and depopulation of this level is investigated, and the feasibility to use the transition from the $^2F_{7/2}$ to the ground state as a reference for an optical frequency standard is discussed.

For our experiments we used an rf ion trap by which up to 10^7 ions can be stored (see, e.g., Ref. 17). The experimental setup is shown in Fig. 1. The ions are created by electron bombardment of atomic Yb and evaporated in an oven or by a wire covered with metallic Yb. The storage time of the ions in the trap is about 7 h. The pressure of background gas in the vacuum vessel can be reduced to less than 10^{-6} Pa, which is necessary to suppress collisional deactivation of excited-state ions. For well-aimed investigations of collisional effects, the vacuum chamber can be backfilled with rare gases to the order of 0.1 Pa. The ions are optically excited by frequency-doubled laser light from a cw Rhodamine 700 dye laser tuned to the D_1 resonance line of Yb^+ at $\lambda = 369.42$ nm. The infrared radiation behind the frequency doubler ($LiIO_3$ crystal) is blocked by a filter. For the excitation, laser power of 20 μW with a bandwidth of about 5 GHz is available. The laser can be pumped also synchronously and mode locked then delivering pulses of 10-ps duration with a bandwidth of 80 GHz and an average power of 1 mW and 369 nm. For time-resolved measurements, the laser beam can be blocked or released within a few ms by a shutter. Scattered fluorescence light is detected by a photomultiplier and observed perpendicular to the laser beam through one of the trap electrodes which is formed out of transparent mesh. The output signal of the photomultiplier is stored by a computer as a function of time.

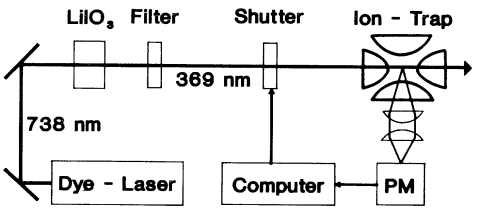

FIG. 1. Experimental arrangement.

When resonantly exciting the ions from the $6s\,{}^2S_{1/2}$ to the $6p\,{}^2P_{1/2}$ state with cw laser light we observe a rapidly decreasing fluorescence signal which goes down to zero level. A measurement which was performed with a laser power of 16 μW and which clearly reveals this behavior is shown in Fig. 2. The fluorescence decay time linearly depends on the laser power and in this case is found to be $T_f = 7.1$ s. In order to explain the vanishing fluorescence, at first one might speculate that some light-mechanical effect or photoionization could cause the ions to leave the trap potential. A further analysis, however, shows that the ions can still be found in the trap but were transferred to the highly metastable $4f^{13}6s^2\,{}^2F_{7/2}$ state. Therefore they are no longer available for an excitation from the ground state.

The population of the $^2F_{7/2}$ state was verified by selective excitation to higher-lying levels with wavelengths in

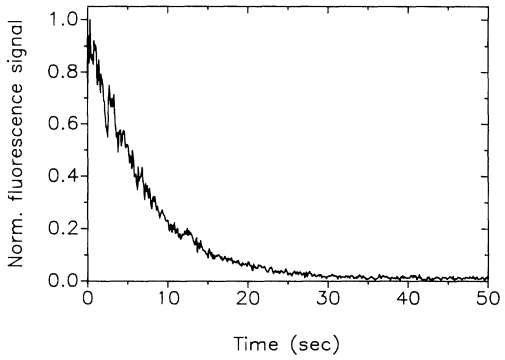

FIG. 2. Decay curve of laser-induced fluorescence radiation. The laser was tuned to the D_1 resonance line of Yb II and operating with about 5-GHz bandwidth at 16-μW power.

direct vicinity of the resonance transition (see Fig. 3). Tuning the laser to a wavelength of $\lambda = 369.11$ or 369.37 nm, after sufficient long illumination (usually a few seconds up to 1 min, depending on the laser power) all the ions can be transferred back to the ground state and a new excitation on the resonance line again results in the full fluorescence signal. While the first wavelength corresponds to a transition

$$^2F_{7/2} \rightarrow 4f^{13}(^2F^0{}_{7/2})5d^2(^3F^0)^3[\tfrac{11}{2}]^0{}_{11/2}$$

(for notation see Ref. 18), which is assumed to be a quadrupole transition, the second is assigned to a two-photon transition $^2F_{7/2} \rightarrow 4f^{13}(^2F^0{}_{5/2})5d6p(^1D_2)_{5/2}$.

Special attitude of the fluorescence is observed when the laser is synchronously pumped and mode locked, then operating with a spectral width which simultaneously covers the resonance line and the two-photon transition. Again the initial fluorescence rapidly decreases but then stabilizes at about half the initial signal. This indicates that now about half of the ions are left in the ground state and those which were excited on the resonance transition and passed to the $^2F_{7/2}$ state can again be pumped back to the ground state. In this way the deadlock of population trapping in the $^2F_{7/2}$ state is overcome and a stable fluorescence level is obtained when the effective population rate just balances the depletion rate. At first it seems surprising that such a high fluorescence level is maintained, although the induced transition probability on the two-photon transition is considerably smaller than the one on the resonance line. But this is attributed to a comparatively large number of excitation cycles on the resonance line which are necessary to transfer an ion to the $^2F_{7/2}$ state.

The population of this level can be explained by a cascading decay of the ions from the $^2P_{1/2}$ via the $4f^{14}$-$5d\,^2D_{3/2}$ state (see Fig. 3). The branching ratio Q_P from the $^2P_{1/2}$ to the $^2D_{3/2}$ or $^2S_{1/2}$ level is not accurately known and therefore was estimated with $\frac{1}{300}$.[19] From the $^2D_{3/2}$ level with a lifetime of 52 ms (Ref. 16), the

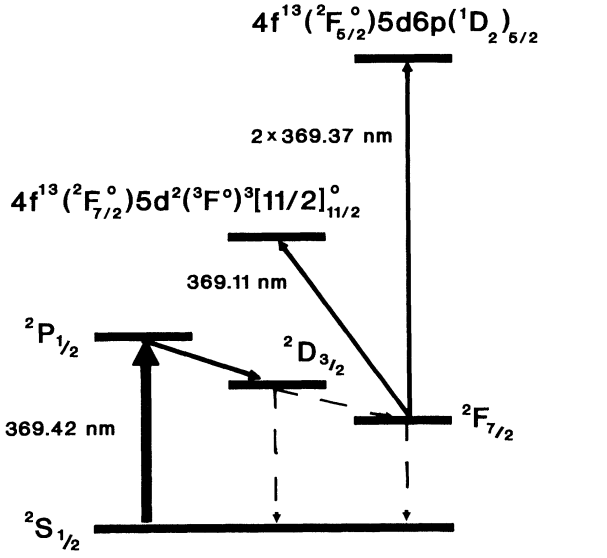

FIG. 3. Relevant energy levels and transitions of Yb^+.

ions then undergo a transition to the $^2F_{7/2}$ or the ground state.[20]

The corresponding branching ratio Q_D can be derived from the measured fluorescence decay time T_f of Fig. 2. But this still requires the knowledge of the laser excitation rate R_e on the resonance transition. This rate is determined experimentally by measuring the fluorescence decay which is caused by optical pumping between Zeeman levels with circularly polarized light in a small magnetic field. For the measurement of Fig. 2, an excitation rate for an individual ion of $R_e = 65$ Hz is evaluated in good agreement with the theoretically expected value. The quantitative relation between the branching ratio, fluorescence decay time, and excitation rate is found from the rate equations for the levels involved. Under our experimental conditions, where population changes of the $^2P_{1/2}$ and $^2D_{3/2}$ states can be assumed to be zero, these equations simplify considerably and then give the relation $T_f = (1+Q_P^{-1})(1+Q_D^{-1})R_e^{-1}$. The branching ratio Q_D for a transition from the $^2D_{3/2}$ to the $^2F_{7/2}$ or the ground state now can be calculated with the knowledge of the other branching ratio Q_P. For this purpose the estimated value $Q_P = \frac{1}{300}$ is inserted and together with $T_f = 7.1$ s and $R_e = 65$ Hz a branching ratio $Q_D = 1.9$ is found. Even though this can only be an approximate value, it shows that more than half of the ions in the $^2D_{3/2}$ state are decaying to the $^2F_{7/2}$ state. In previous publications a transition to this level and a trapping effect in this metastable state were apparently not notices. But it is of fundamental importance for future ultimate-resolution laser spectroscopy in Yb^+ or the realization of an Yb^+ frequency standard,[12] which in the microwave range might operate on the ground-state hyperfine transition of one of the odd isotopes,[17] or in the

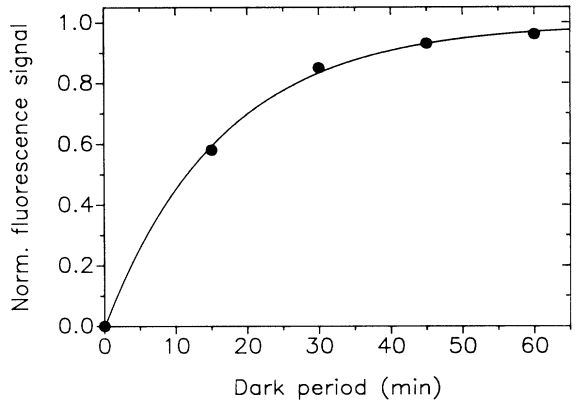

FIG. 4. Measurement of the relaxation time of the $^2F_{7/2}$ state in Yb^+ with atomic Yb vapor as background gas. The dots represent the measured fluorescence intensity indicating the return of ions to the ground state during the chopping-off period of the laser. The solid line is a least-squares fit of an exponential to the experimental data.

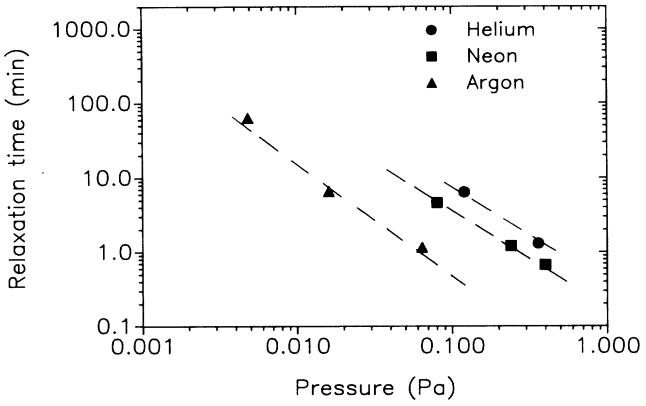

FIG. 5. Compilation of measured relaxation times of the $^2F_{7/2}$ state as a function of the buffer gas pressure for the rare gases Ar, Ne, and He.

optical region on one of the well-defined transitions from the $^2D_{5/2}$ or $^2D_{3/2}$ to the ground state.

The product $T_f R_e = 460$ represents the average number of excitation cycles on the resonance line that each ion experiences, before it ends in the $^2F_{7/2}$ state. For the case of broad-band laser excitation, as discussed before, this means that the simultaneous excitations on the resonance line and the two-photon transition also approximately differ by this factor. From this it further results that a transition on the weak two-photon line can be monitored with considerably increased sensitivity by detecting the laser-induced fluorescence radiation on the resonance line, quite similar to an amplification scheme as used for the detection of shelved optical electrons.[1]

The complete disappearance of the resonance signal even at very low pumping rates, as observed with narrow-band laser excitation, is an indication of an extremely long lifetime of the $^2F_{7/2}$ state. We have measured this lifetime by applying the following procedure: First, complete population inversion is built up and all ions are pumped to the $^2F_{7/2}$ state by resonant excitation. Then the laser is blocked for a definite dark period, and finally the repopulation of the ground state and hence the depopulation of the $^2F_{7/2}$ state is probed by a second excitation on the resonance line by monitoring the laser-induced fluorescence radiation.

An example for the measured reappearance of the fluorescence as a function of the dark period is shown in Fig. 4. The observed recovery time, i.e., the relaxation time of the ions strongly depends on collisional quenching. For the measurement of Fig. 4, atomic Yb vapor with a number density of approximately 10^6–10^7 cm^{-3} in the trap was acting as the background gas yielding a lifetime of 15 min. The influence of some rare gases on the dwell time of the ions in the metastable state is represented in Fig. 5. This compilation shows the rapid increase of the relaxation time with decreasing buffer gas pressure. But even at the higher pressures (about 0.4 Pa) still an unexpected long-time constant of about 1 min can be measured.

At ultrahigh-vacuum conditions ($\leq 10^{-6}$ Pa) without an additional background gas the lifetime by far exceeds the ion-storage time. After a dark period of 4 h, less than 2% of the ions, which are still in the trap, have undergone a transition into the ground state.[21] From this measurement the radiative lifetime of the $^2F_{7/2}$ state is estimated to be greater than 8 d. To our knowledge this is by far the longest lifetime which has been reported up to now for an optically excited atomic state. Preliminary numerical calculations even give a theoretical value of 650 d.[22]

From the measured lifetime value we can derive a natural linewidth for the transition $^2F_{7/2} \rightarrow {}^2S_{1/2}$ with a wavelength near $\lambda = 467$ nm of less than 0.3 μHz and a line Q of better than 10^{21}. For the future, therefore, this transition could serve as an excellent reference for an optical frequency standard. An induced transition on the extremely weak electric octupole line can again be detected via laser-induced fluorescence changes on the resonance line. The amplification factor of 460× can be considerably increased with the use of an additional laser, which is tuned to the transition $^2P_{1/2} \rightarrow {}^2D_{3/2}$ near $\lambda = 2.44$ μm. Thus, even experiments with single trapped Yb^+ ions are possible and optical cooling can be applied. This would result in an optical frequency standard of unprecedented performance.

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[a]Permanent address: Universität Hamburg, I. Institut für Experimentalphysik, Jungiusstrasse 9, D-2000 Hamburg 36, West Germany.

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[21]For a normalization of the signal and correction due to the limited ion-storage time, the number of ions in the trap after the dark period was measured by repopulating the ground state and monitoring the maximum fluorescence on the resonance line.

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