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Laser cooling of trapped ytterbium ions using a four-level optical-excitation scheme

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We demonstrate laser cooling of $^{172}Yb^+$ ions in an rf trap using an alternative optical-excitation scheme. The metastable $4f^{14}5d\,^2D_{3/2}$ state of Yb^+, to which ions decay if the resonance (cooling) transition is driven, is depleted by driving an optical transition at a wavelength of 935 nm, which is much more attractive than the 2.438 μm radiation used previously. This couples the metastable state to a fourth energy level from which the ions decay rapidly to the ground state. Fluorescence line shapes of the cooling transition indicate that small numbers of Yb^+ ions were cooled in this way to kinetic temperatures below 1 K.

Ions stored in radio-frequency (rf) quadrupole traps can be laser cooled and confined to perturbation-free regions at the trap center. This offers unique possibilities for ultrahigh-resolution spectroscopy, studies of single quantum systems, and precision frequency metrology [1]. Cooling to subkelvin temperatures typically requires scattering of 10^4–10^5 photons per ion, during time intervals which are short relative to the time constants of heating processes such as collisions. Laser cooling of ions stored in rf traps has been demonstrated in a number of species [2–5] including Yb^+ [6]. In all these cases, the cooling radiation drives $^2S_{1/2}$-$^2P_{1/2}$ resonance transitions, permitting fast cooling rates. For laser cooling of Ba^+, Sr^+, and Yb^+, metastable $^2D_{3/2}$ levels need to be depleted by a second radiation source in order to avoid disruption of the cooling cycle due to population trapping in the metastable level. Conventionally, the second laser field is resonant with the $^2D_{3/2}$-$^2P_{1/2}$ transitions [2,5,6]. In Yb^+, this transition is at 2.438 μm.

In this paper, we report laser cooling of $^{172}Yb^+$ ions in an rf quadrupole trap using an alternative metastable-level depletion scheme. Population trapping in the $4f^{14}5d\,^2D_{3/2}$ level of Yb^+ is avoided by coupling this level to a fourth energy level which rapidly decays to the ground state. For laser cooling of Yb^+, the four-level optical-excitation scheme is particularly attractive because the required laser wavelength of 935 nm can now be generated more easily than the 2.438 μm used for the three-level scheme. This should be advantageous for future high-resolution spectroscopic studies of trapped Yb^+, which is a promising candidate for new optical-wavelength and frequency standards [7,8]. Another advantage of the four-level cooling scheme is that coherent population trapping in a superposition of the $^2S_{1/2}$ and $^2D_{3/2}$ levels does not occur because the driven optical transitions are not coupled. Coherent population trapping can lead to "coherence nulls" in the fluorescence line shapes [9] which could lead to reduced cooling efficiency.

Figure 1 shows a partial term scheme of Yb^+. A transition allowed by configuration interaction mixing at a vacuum wavelength of 935.2 nm connects the $4f^{14}5d\,^2D_{3/2}$ and the $4f^{13}5d6s\,^3D[3/2]_{1/2}$ levels [10]. From the upper level, the dominant decay is to the ground state, with a theoretically estimated lifetime of 17 ns [11]. An experimental value for this lifetime is 42(3) ns [12]. The corresponding decay rate is fast compared to the rate at which the $^2D_{3/2}$ level can be populated by spontaneous

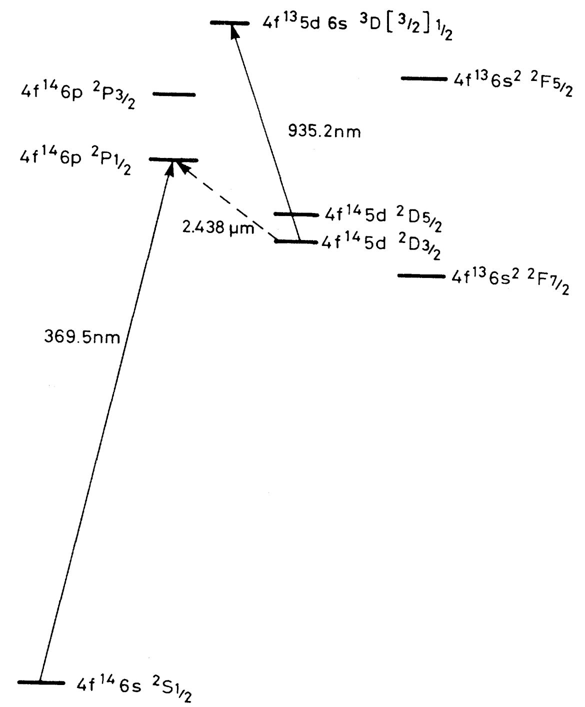

FIG. 1. Partial-term scheme of Yb^+, showing the transitions of interest. The solid lines indicate the transitions driven in this experiment. The dashed line indicates the transition previously used to depopulate the $^2D_{3/2}$ metastable level.

decay from the $^2P_{1/2}$ level. Thus one can expect that driving the 935-nm transition will allow almost complete depopulation of the $^2D_{3/2}$ level, even if the resonance transition is saturated.

Our experimental arrangement was similar to that of Klein *et al.* [6] for the initial demonstration of laser cooling of trapped Yb^+ in an rf quadrupole trap of size $r_0=\sqrt{2}z_0=5.0$ mm. The difference-frequency-mixing apparatus used for generation of the 2.438-μm radiation in the three-level excitation scheme was replaced by a Schwartz Electro-Optics cw Ti:sapphire laser tuned to the 935-nm $4f^{14}5d\,^2D_{3/2}$–$4f^{13}5d6s\,^3D[3/2]_{1/2}$ transition. The vacuum wavelength of the transition was measured to be 935.184(1) nm, using a Burleigh WA-20 wavemeter. The Ti:sapphire laser was operated in a standing-wave configuration, and included a birefringent filter and an uncoated 1-mm-thick étalon for tuning. Typically 2–3 adjacent cavity modes of the Ti:sapphire laser oscillated. In order to obtain a spectral power-density distribution averaged over a range of ≤ 1 GHz, the resonator length was dithered at a rate of 3.2 kHz to give a frequency modulation of greater than the cavity-free spectral range. Up to 9 mW of the 935-nm (ir) radiation was focused to a beam diameter of 2 mm and overlapped with the counterpropagating 369.5-nm resonance (UV) cooling radiation. The UV power was 0.15 mW in a beam diameter of 300 μm at trap center.

All experimental observations described in the following were conducted without buffer gas at a measured background gas pressure of less than 5×10^{-8} Pa. Fluorescence from the trapped ions was imaged by a lens (magnification factor of 1.8) onto a 100-μm aperture in front of the photomultiplier. Both large ion clouds, or a few ions, could be loaded into the trap [6,13].

The UV fluorescence count rates as a function of UV laser frequency and with 935-nm radiation present are shown in Figs. 2(a) and 2(b) for a large ion cloud and a few ions [3,6,14], respectively. Before each frequency scan, the UV cooling frequency was held at a fixed frequency $\sim$200 MHz below the line center, and the ir frequency optimized for maximum UV fluorescence yield. The data of Fig. 2 correspond to scans of increasing UV frequency centered on the initial cooling position. Scan times were 13 and 33 sec for Figs. 2(a) and 2(b), respectively. The decay of fluorescence observed at the beginning of each scan corresponds to the shift of UV frequency from the initial cooling position to the low-frequency point at the start of the scan.

The effect of the ir 935-nm laser power on the UV fluorescence can be seen in Fig. 3 for a large cloud at a constant UV detuning of -500 MHz. Similar saturation effects were observed for small numbers of ions. Fluorescence enhancement factors were such that blocking the 935-nm light reduced signal levels to the background noise level. This strong enhancement and saturation of observed fluorescence for ir power densities of a few mW mm^{-2} clearly shows that driving the $4f^{14}5d\,^2D_{3/2}$–

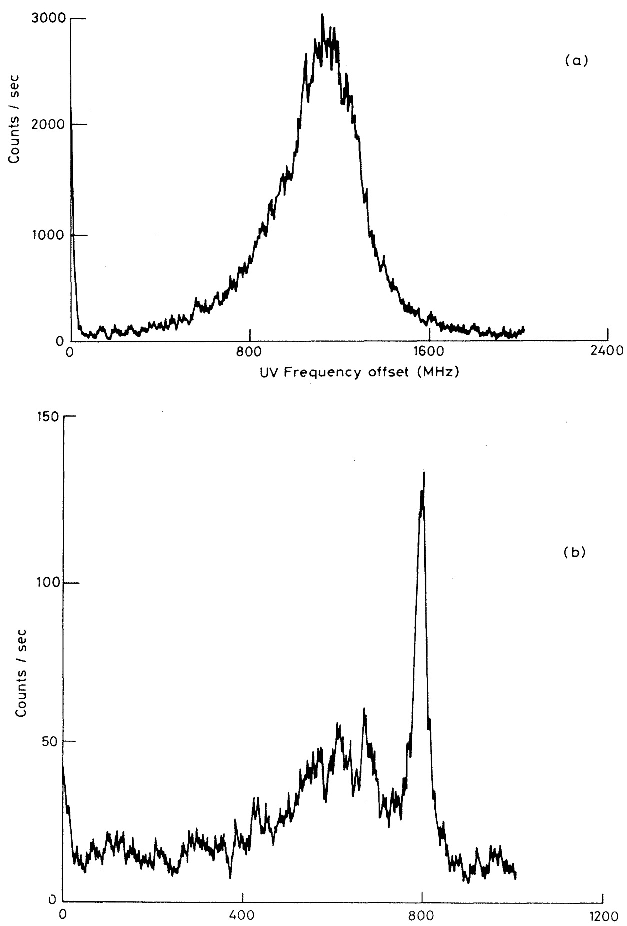

FIG. 2. Fluorescence signal as a function of the UV laser frequency for (a) a large cloud of ions and (b) a few ions.

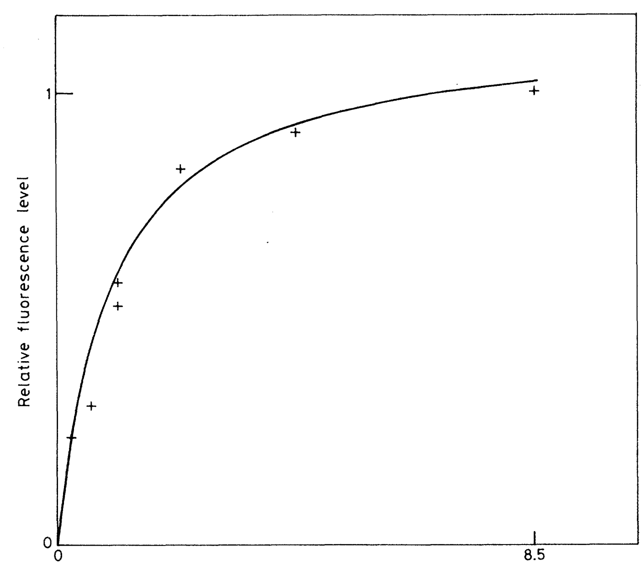

FIG. 3. Fluorescence signal from the trap as a function of 935 nm Ti:sapphire laser power. The fluorescence level is normalized to that at 8.5 mW. The solid curve is a fit derived from a simple four-level rate equation analysis (Ref. 13).

$4f^{13}5d6s\ ^3D[3/2]_{1/2}$ transition efficiently depletes the metastable $^2D_{3/2}$ level more quickly than it is populated.

The UV fluorescence line shape shown in Fig. 2(a) for a large ion cloud has a full width at half maximum (FWHM) of ≈ 400 MHz, and some asymmetry. If we neglect this asymmetry and assume the linewidth to be dominated by Doppler broadening, we can deduce an ion kinematic temperature of 80 K. For large ion clouds, this represents significant cooling from initial loading temperatures of typically 2000 K [14]. The 80-K temperature is expected to be an equilibrium temperature between laser cooling and rf heating of the ion cloud. The small amount of line-shape asymmetry may well result from changes in the cloud temperature as the frequency is scanned, with heating close to and above the line center, and some associated loss of fluorescence.

The fluorescence line shape shown in Fig. 2(b) for a few ions is distinctly asymmetric. It comprises a strong fluorescence peak of ≈ 30-MHz FWHM, but with some broad weak structure to the low-frequency side. This weak structure exhibited variations in shape and height on different scans with unchanged experimental parameters. Such variations may be due to frequency and intensity fluctuations of the Ti:sapphire laser. The strong peak is expected to have a number of contributions to its linewidth and shape. These include the natural width of the resonance transition (≈ 23-MHz FWHM), power broadening, residual Doppler broadening, the photometer time constant, and a rapidly decreasing fluorescence rate above the line center due to ion heating. In addition, the fluorescence may start to decrease slightly below the line center due to rf heating becoming dominant over the laser cooling. The following analysis is based on the premise that the fluorescence does not decrease until the line center is reached. If one assumes that the observed ≈ 30-MHz width corresponds to a 30-MHz half width at half maximum in order to account for the high-frequency ion heating signal loss [3], the line can be approximated by a Voigt profile with an estimated FWHM of 60 MHz. If we assume that the Lorentzian contribution is simply due to the natural width, this leads to a Gaussian contribution of 48 MHz. The corresponding upper limit for the temperature of the cooled ions is 1.2 K. Removing instead an estimated power-broadened Lorentzian [13] of 30 MHz leads to a reduced temperature of 0.9 K.

The low-frequency structure exhibited by Fig. 2(b) might be accounted for by a dynamic cooling description whereby the cooling rate changes during the low-frequency part of the scan. One possible development of this description is to suggest the onset of a phase transition from a small ion cloud into a "crystalline" or ordered state. This low-frequency shape of Fig. 2(b) may be compared with previous line-shape data exhibiting phase transitions, from about five rf trapped Mg^+ ions [15]. Such phase transitions are expected for cooled ion temperatures well below 1 K [16]. It should be stressed, however, that further work is needed to verify that this observed behavior is due to phase transitions, for example, by two-dimensional high-resolution imaging of the ions in order to observe spatial structures.

Further diagnostic studies of the Yb^+ four-level excitation scheme, in addition to the high-resolution camera studies of the cloud, are needed to more fully assess its capability and efficiency for Yb^+ cooling. In particular, the use of a single-mode Ti:sapphire laser with controlled tuning would be of significant advantage. In this case, the cooled 935-nm line shape could be readily observed. This transition has an estimated natural width of less than 10 MHz. The UV cooling radiation could be maintained at a constant frequency offset while the ir is scanned. Under these conditions, a narrower and more symmetric line shape for the cooled ions is expected, which could be analyzed more readily and precisely to determine the ion temperature.

In conclusion, $^{172}Yb^+$ ions in an rf trap have been laser cooled using an alternative optical-excitation scheme for depleting the metastable $4f^{14}5d\ ^2D_{3/2}$ level. In contrast to the usual three-level excitation scheme, the method described here relies on coupling the metastable level to a fourth energy level which rapidly decays to the ground state. The optical transition depleting the metastable level has a wavelength of 935 nm, which is in the emission range of Ti:sapphire lasers, Nd-doped fiber lasers [17], and also of recently available laser diodes [18]. Typical power-density levels of some mW mm^{-2} were found to be sufficient to deplete the metastable $^2D_{3/2}$ level. The optical cooling of a Yb^+ cloud of a few ions using 935-nm radiation has led to the observation of a much narrower feature on the resonance line shape. This suggests that a kinetic temperature of less than 1 K has been reached.

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