Multiply Charged Thorium Crystals for Nuclear Laser Spectroscopy

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We have produced laser-cooled crystals of $^{232}Th^{3+}$ in a linear rf Paul trap. This is the first time that a multiply charged ion has been laser cooled. Our work opens an avenue for excitation of the nuclear transition in a trapped, cold $^{229}Th^{3+}$ ion. Laser excitation of nuclear states would establish a new bridge between atomic and nuclear physics, with the promise of new levels of metrological precision.

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Coherent excitation of the electronic states of atoms and molecules with lasers is at the heart of modern spectroscopy and metrology. To extend these techniques to nuclear states would be a tremendous advance. However, the typical excitation energies for nuclear matter are in the keV to MeV energy range, where coherent radiation sources are lacking. In the remarkable case of the ^{229}Th nucleus, the energy splitting of the ground state doublet is only several eV, which may be within the reach of coherent table-top UV lasers. Next to ^{229}Th, the lowest known energy of a nuclear excited state is 76.8 eV for ^{235}U. This makes ^{229}Th unique in having a nuclear excitation energy commensurate with electronic and optical energy scales and provides intriguing possibilities for coherently manipulating nuclear matter with lasers [1]. The excited state of the nuclear doublet decays via photon emission, with a predicted lifetime of $\sim$5 hr [1]. The narrowness of the nuclear transition and the superb isolation of the nucleus from external fields and collisions could form the basis of an optical clock with exceptional precision [2]. Additionally, precision studies of this transition could provide a sensitive probe of the effects of the chemical environment and atomic electron charge states on nuclear decays [3]. Other intriguing possibilities include the use of this transition to search for variations in fundamental constants. Flambaum has suggested that this transition may be uniquely sensitive to relative variation of the strong interaction parameter and the fine structure constant [4], although this point requires further study [5–8].

The first suggestion that ^{229}Th had a uniquely low-lying excited state was made in 1976 by Kroger and Reich [9], based on results of gamma-ray spectroscopy of high-lying ^{229}Th nuclear states populated via alpha decay of ^{233}U. They determined that the ground state was a doublet, with a very low-lying energy $I^{\pi} = 3/2^{+}$ isomeric state, denoted ^{229m}Th, lying less than 100 eV above the $I^{\pi} = 5/2^{+}$ ground state, denoted ^{229g}Th. Subsequent measurements yielded a higher precision result of 3.5 ± 1 eV [10,11].

Several studies have confirmed the rotational band structure identified by Kroger and Reich; however, attempts to directly observe the emission of the gamma ray from the isomeric decay in the energy range of 3.5 eV have been unsuccessful, or at best inconclusive [12–14].

Last year, a new measurement of the energy splitting reported a value of 7.6 ± 0.5 eV [1]. This observation sheds new light on the negative results of previous searches [15,16], as the decay energy for this splitting corresponds to an emitted photon wavelength of 153–175 nm, which is outside the bandwidth of the detectors employed in the searches and outside the transmission window of the quartz cells used to contain the samples.

The trapping and cooling techniques of modern atomic physics provide for single atom, state-sensitive detection with essentially 100% efficiency. In this way, it is possible to detect even very weak transitions. For example, the electric octupole transition in Yb^{+} ($\tau = 6$ yr) has been observed using just a single ion [17].

In choosing a suitable atomic charge state for these studies, important factors include the convenience of the atomic level structure with respect to available closed transitions and lasers and the ionization energy. In principle any charge state should exhibit the isomer transition, including neutral Th, Th^{+}, and Th^{2+}. On the other hand, as pointed out by Peik and Tamm [2], Th^{3+} is a particularly attractive candidate because of the availability of closed transitions and the fact that its ionization potential (27 eV) is well above the isomeric transition energy. To excite and accurately measure the isomeric transition in a single trapped $^{229}Th^{3+}$ ion with optical radiation, several challenges will have to be addressed. The nuclear excitation energy is known only approximately, and the transition is very weak. The hyperfine structure of both the nuclear ground state and excited isomer state of $^{229}Th^{3+}$ is not known.

As a first step, we here report confining and laser cooling of $^{232}Th^{3+}$ in a linear rf Paul trap. Previously, sympathetic cooling has been employed to create crystals of multiply charged ions [18,19]. The ^{232}Th isotope is convenient for initial work due to its lack of hyperfine structure and low cost. Loading Th^{3+} into an ion trap and subsequent laser

cooling pose significant challenges. Thorium is a refractory metal with a high melting point (2050 K) and low vapor pressure, and hence producing an atomic source is not trivial [20,21]. Furthermore, generating multiply ionized species by electron impact is typically much less efficient than generating singly ionized species.

The general schematic of the apparatus and the relevant atomic energy levels are shown in Fig. 1. The trap consists of four stainless steel rods of diameter 9.5 mm whose centers are arranged on the vertices of a square with sides of length 12.4 mm. The trapping potential is created by a 3.1 MHz rf voltage (20–700 V_{0-pk}) applied to all four pins. Confinement in the axial direction is provided by dc voltages (10–600 V) applied to stainless steel end cap electrodes, each having a 5 mm hole coaxial with the trap axis drilled through for optical access and sample loading. The end caps are separated by 39 mm and are electrically insulated from the rf rods by alumina spacers. The large trap size was chosen to maximize the loading efficiency of Th^{3+}.

To create Th^{3+}, we employ laser ablation of a thorium target using the third harmonic of a pulsed YAG laser. The 5 ns laser pulses, each with energy of $\sim$200 μJ, are focused down to a 15(10) μm spot size on a thorium metal sample. Because of the conservative nature of the Paul trap, the potential of the end cap nearest the sample is pulsed from 100 V to ground and back in sync with each ablation pulse, allowing cumulative axial ion loading. Much of the slower moving Th^{+} and Th^{2+} components of the ablation plume are electrically blocked from entering the trapping volume by careful timing of the potential pulses. To remove any trapped pollutant ions, the well understood mass selective capabilities of rf ion traps are employed.

To increase loading efficiency and facilitate efficient laser cooling, we introduce 99.9999% pure helium buffer gas at pressures of $10^{-7} - 10^{-5}$ torr into the vacuum chamber. The gas collisionally cools the ablated Th^{3+} ions from tens of thousands of Kelvin to about room temperature, after which it is pumped out, allowing for laser cooling to take place. In this process, the vacuum environment returns to a pressure of $<2 \times 10^{-11}$ torr within 30 seconds of shutting off the helium source.

Th^{3+} offers closed atomic transitions at convenient optical wavelengths that lend themselves to fluorescence detection and laser cooling (see Fig. 1). There is an available 2-level system at 1087 nm between the $5^2F_{5/2}$ ground state and the $6^2D_{3/2}$ excited state. The lifetime of the $6^2D_{3/2}$ state was recently calculated by Safronova *et al.* to be 1.09 μs, corresponding to a linewidth of 145 kHz [22]. Additionally, there is a 3-level system involving the $6^2D_{5/2}$ state and the $5^2F_{5/2}$, $5^2F_{7/2}$ states. The lifetime of the $6^2D_{5/2}$ state was calculated to be 0.676 μs (234 kHz linewidth) with a 1:8 branching ratio to the $5^2F_{5/2}$, $5^2F_{7/2}$ ground states, respectively. The sub-MHz linewidths of the transitions should allow for direct Doppler cooling into the motional ground state, if a higher frequency trap is employed [23].

Initial laser cooling is accomplished by illuminating the ion cloud with 4 red detuned 1087 nm laser beams propagating in the same plane at 30° angles to the trap axis (see Fig. 1). The beams, derived from a diode laser, have a cross-section of 4 by 1 mm, and each have a power of 4 mW, producing an intensity $\sim$100$\times$ above saturation per beam. In order to laser cool the atoms from room temperature to the ultracold regime, the detuning of the 1087 nm light is swept from -200 MHz to -25 MHz over a period of $\sim$15 s. This slow scan is required due to the relative weakness of the cooling transition; it is $\sim$100-fold narrower than typical ion cooling transitions. At this point, the 690 nm and 984 nm beams, copropagating along the trap axis, aid in cooling to ion crystallization. Both fields are derived from diode lasers, with the 690 nm light on resonance and the 984 nm light detuned $\sim$1 MHz to the red of resonance. A low-noise CCD camera (Andor DU401) and a high numerical aperture collection lens ($NA = 0.32$) are used to detect fluorescent signals of the trapped ions. We have observed fluorescence on each of the transitions shown in Fig. 1. The 984 nm transition provides the largest signal due to the optimal combination of the decay rate and the CCD quantum efficiency.

Through optimization of the trapping parameters and stability, we have observed a wide variety of crystals and chains with single ion resolution. Some representative examples are shown in Fig. 2. For these images, the detected single atom signal is $\sim$20 counts/s, and the exposure time is 6 s. The secular frequencies of the trap are 10–50 kHz. The high charge state of the ions results in a large

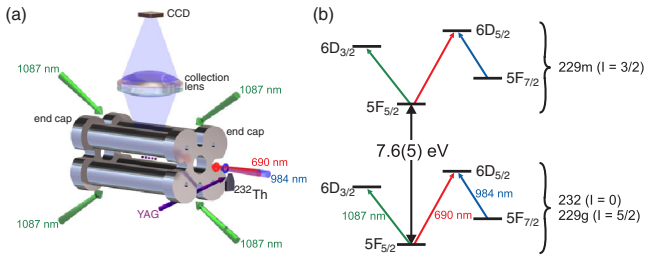

FIG. 1 (color online). (a) Schematic of linear rf Paul trap and ^{232}Th sample. (b) Lowest electronic energy levels of Th^{3+}.

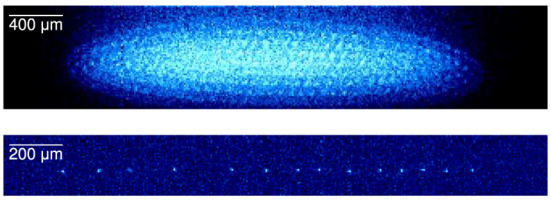

FIG. 2 (color online). 984 nm fluorescence images of trapped Th^{3+} samples. (Top) A Coulomb crystal of $\sim$1700 ions. (Bottom) A crystallized chain of 14 ions. The integration time for each image is 6 s.

ion separation; in the lower image, the spacing between ions in the one-dimensional chain is ~100 μm, while it is somewhat less in the larger crystal.

With ultracold samples, laser spectroscopy was performed on all three atomic transitions using a wave meter (High-Finesse WS7), with stated 1-σ absolute uncertainty of 20 MHz, as a reference. By scanning each of the three lasers over their respective transitions, narrow resonances were observed via fluorescence detection. A sample scan of the 984 nm laser is shown in Fig. 3. Measurements taken over the course of 2 months resulted in similar standard deviations of ~14 MHz for each data set. Before all measurements, the WS7 was calibrated against a diode laser stabilized to an ^{87}Rb transition at 780 nm via saturation spectroscopy. Additional systematic errors including ac Stark shifts from laser and trapping fields, probe laser back action, and Zeeman shifts were estimated to be less than 2 MHz, also less than the short term fluctuations of the wave meter. Because these errors are upper bound estimates and not statistical in nature, they were added linearly to the absolute wave meter uncertainty [24]. The transition frequencies were found from our measurements to be 275 606 583(26) MHz for the $5F_{5/2} \leftrightarrow 6D_{3/2}$ transition, 434 291 397(26) MHz for the $5F_{5/2} \leftrightarrow 6D_{5/2}$ transition, and 304 619 344(26) MHz for the $5F_{7/2} \leftrightarrow 6D_{5/2}$ transition. These results are a significant improvement on the best previous measurements (of ~500 MHz accuracy) [25]. The lifetime of a cold cloud typically exceeded several minutes, as shown in Fig. 4. The process limiting this lifetime is thought to be charge exchange collisions with residual buffer gas impurities. This can be circumvented by cooling the ions from the loading stage to crystallization entirely with lasers, using large initial detuning to compensate for the high ablation temperatures.

The ^{229}Th isotope is available in only very small (1 mg) quantities and is typically provided in nitrate form, $Th(NO_3)_4$, rather than elemental form. Therefore, it will be important to employ efficient processing techniques and to maximize the ionization yields and trapping efficiency. A large body of prior work on highly efficient trapping of trace amounts of atomic and molecular ions will serve as a guide in this process.

Observation of the isomeric state, $^{229m}Th^{3+}$ via fluorescence detection of the trapped ions will be facilitated by the fact that the ground and excited isomer states have different nuclear spins and hence distinct hyperfine structures. Triply ionized thorium belongs to the francium isoelectronic sequence. Whereas francium (atomic number $Z = 87$) has ground term level [Rn] $7s^2S_{1/2}$, for Th^{3+} ($Z = 90$) it is [Rn] $5f^2F_{5/2}$. This reflects the crossover between the $7s$ and $5f$ one-electron energies at around $Z = 90$ [22]. Calculations of the atomic structure of Th^{3+}, including the one-electron energies, require a relativistic treatment of correlated electrons. Quantitative calculations of energy levels, dipole moments, polarizabilities, hyperfine structure, etc., require use of sophisticated many-body perturbation theory, taking into account simultaneous excitations of the valence and several core electrons.

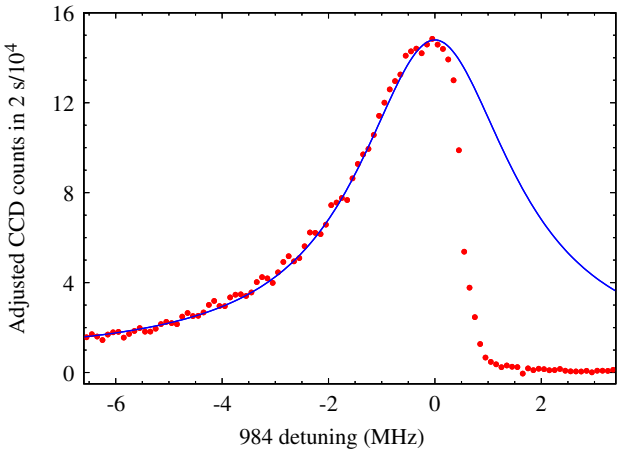

FIG. 3 (color online). Background subtracted ion fluorescence rate vs detuning of the 984 nm laser and Lorentzian fit to the red detuned region of the data. The fit gives a standard error of 50 kHz for the center frequency and FWHM of 3.51(8) MHz. The width is likely limited by a combination of laser linewidth (~1 MHz) and Zeeman and power broadening. Back action from the probe laser is evident as heating occurs on the blue side of resonance. To step the 984 field frequency, an acousto-optic modulator was used. Sinusoidal intensity oscillations with period ~460 kHz were present in the 984 nm beam, consistent with a $v/2l$ period expected from partial acoustic wave reflection inside the acousto-optic modulator crystal. Here, v is the acoustic wave velocity in the acousto-optic modulator crystal and l is the length of the crystal transverse to the acoustic wave k vector.

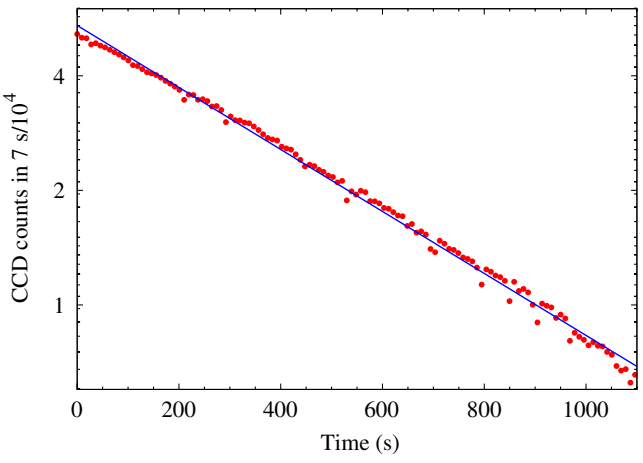

FIG. 4 (color online). Measured fluorescence signal from the 984 nm transition of a cold Th^{3+} cloud. The 984 nm field was detuned ~1 MHz below resonance, while the 690 nm field was on resonance to avoid optical pumping into the $5F_{5/2}$ state. The exponential fit gives a $1/e$ lifetime of 646(10) s. The relatively short ion lifetime is attributed to charge exchange collisions with residual buffer gas impurities.

Recently there has been significant progress in this direction [22,26].

The hyperfine structure of the isomeric state can be determined from knowledge of the hyperfine constants of the nuclear ground state, along with ratios of the magnetic dipole and electric quadrupole moments of the two nuclear states. This suggests a semiempirical strategy: measure the hyperfine structure of the $^{229g}Th^{3+}$ electronic energy levels experimentally and deduce the isomeric hyperfine structure by employing nuclear models to compute the ratios of the dipole and quadrupole moments. An experimental measurement of the $^{229g}Th^{3+}$ hyperfine structure would in itself represent an important advance, providing a stringent test of state-of-the-art theories of complex atoms and ions. Dykhne and Tkalya have estimated the magnetic dipole moment of $^{229m}Th^{3+}$ to be $-0.076\mu_N$ [27], while for $^{229g}Th^{3+}$ $\mu = 0.44\mu_N$ [28]. This work needs to be extended to determine the $^{229m}Th^{3+}$ nuclear quadrupole moment.

The isomer transition may be directly excited using light at 7.6 eV ($\lambda = 163$ nm) generated, for example, by frequency mixing in atomic vapors [29,30]. Alternatively, the isomer state could be populated using a two-photon electronic excitation through the $6D_{5/2}$ or $6D_{3/2}$ levels, or three-photon through the $7S_{1/2}$ level, assisted by nuclear-electron spin coupling (the so-called electron bridge). For example, in the former case, the 690 nm transition could be saturated, while a laser in the 200–230 nm wavelength range would excite the ion into the isomer state. Once the transition energy has been accurately determined, one could use a deep UV frequency comb for direct excitation [31]. The frequency comb is compatible with the ultranarrow linewidth frequency stabilization (1 Hz) required for metrological applications and has sufficient power to saturate the isomeric transition.

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