Nuclear laser spectroscopy
of the $3.5\,\mathrm{eV}$ transition in Th-229

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Abstract. – We propose high-resolution laser spectroscopy of the $3.5\,\mathrm{eV}$ nuclear transition in Th-229 in isolated atoms. Laser excitation of the nucleus can be detected efficiently in a double-resonance method by probing the hyperfine structure of a transition in the electron shell. It is shown that for a suitably chosen electronic level, the frequency of the nuclear transition is independent of external magnetic fields to first order and of electric fields to second order. This makes Th-229 a possible reference for an optical clock of very high accuracy. The nuclear-electronic double-resonance method can be conveniently applied to a laser-cooled ion of $^{229}\mathrm{Th}^{3+}$ in a radiofrequency trap. Further applications of nuclear laser spectroscopy are discussed.

High-precision γ-spectroscopy has revealed the existence of an isomeric state of the Th-229 nucleus at the unusually low excitation energy of $3.5 \pm 1.0\,\mathrm{eV}$ above the ground state [1]. The ground state of Th-229 undergoes α-decay with a half-life of 7880 years [2] and the half-life of the isomer for α-decay is estimated to be about 2–4 times smaller [3]. Both states are linked by a magnetic-dipole transition (cf. fig. 1) and the radiative lifetime of the excited state is estimated to be of the order of 2–4 hours [4, 5]. The energy of this nuclear excitation is comparable to typical excitation energies of outer electrons in the atomic shell. The expected transition wavelength of 350^{+150}_{-75} nm lies in the range where tunable laser sources are available so that the methods of high-resolution laser spectroscopy can be applied to this nuclear system. This observation has motivated a number of theoretical studies in a new borderland between atomic and nuclear physics [4–8]. In this letter we want to point out some possibilities for precision laser spectroscopy of such a low-energy nuclear excitation. Unlike the case of Mössbauer spectroscopy, where solid samples are studied, we will mainly focus on isolated atoms and show that such a transition might be used as the basis for a highly accurate optical clock.

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$\frac{3}{2}^{+}$ [631] ^{229m}Th isomer

$\Delta E = 3.5 \pm 1$ eV
M1 transition
$\tau = 10^4$ s

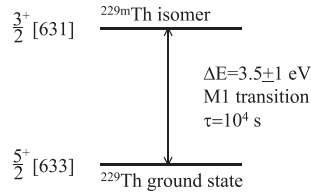$\frac{5}{2}^{+}$ [633] ^{229}Th ground state

Fig. 1 – Lowest nuclear energy levels of Th-229 with their Nilsson state classification.

Laser excitation of an isomer could be detected in a very efficient way by performing laser spectroscopy of the electron shell (cf. fig. 2): Imagine that the nucleus is in its ground state and that a laser is tuned to a closed two-level electric-dipole transition of the shell. The atom will then continuously scatter resonance fluorescence photons. If a second laser source now excites the nucleus to the isomeric state, the nuclear moments and nuclear spin will change and consequently the hyperfine splittings and total angular momenta of the energy levels in the shell. After the nuclear transition, the laser that was exciting the closed two-level system of the shell will thus be out of resonance and the fluorescence intensity will drop. For the individual atom the repeated excitation and decay of the isomer will lead to a sequence of periods of high and low fluorescence intensity. This double-resonance method for nuclear and electronic excitations is similar to Dehmelt's "electron shelving" scheme for the detection of the excitation of metastable states in the shell of a single trapped ion [9, 10]. A third laser could be used, that is tuned in resonance with a closed two-level system of the shell of the isomer. In this case the atom will continuously scatter photons, but at either one of two different wavelengths, indicating the state of the nucleus. In addition to the change of the hyperfine splitting, the change of the total angular momentum can also be exploited for the detection of the nuclear excitation via a change of the polarization of fluorescence photons. In order to avoid shifts of the nuclear transition frequency due to the optical excitation of the electron shell, the laser fields driving the nuclear and electronic transitions can be applied alternately. Similar cyclic excitation and interogation schemes are commonly employed in high-resolution laser spectroscopy of trapped ions [10].

As is well known, the level structure of the atomic electron shell is modified by the hyperfine
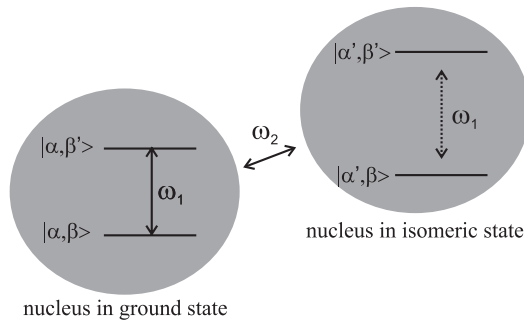

Fig. 2 – Simplified schematic of the nuclear-electronic double-resonance method: one laser at frequency ω_1 drives a two-level system of the shell of the nuclear ground state, but is out of resonance after a second laser at frequency ω_2 has excited the nucleus to the isomeric state (α labels the nuclear, β the electronic excitation). The nuclear transition is detected as a change in the fluorescence intensity of photons at ω_1.

interaction (HFI) between the electrons and the nuclear electric and magnetic moments. In nuclear spectroscopy of an isolated atom, HFI affects the nuclear spectra in a very similar way. The system can be described by a Hamiltonian that consists of four distinct parts: $H = H_{\mathrm{n}} + H_{\mathrm{e}} + H_{\mathrm{HFI}} + H_{\mathrm{ext}}$. Here H_{n} and H_{e} denote the nuclear and electronic interactions, respectively. H_{e} also includes the Coulomb interaction with the point charge ($\propto 1/r$) part of the nuclear potential. H_{HFI} describes the coupling between electron shell and nucleus caused by the internal nuclear structure. We assume that H_{HFI} is a small perturbation of the nuclear and electronic interactions H_{n} and H_{e}. H_{ext} describes the interaction with external fields and will be considered as a small perturbation of H_{HFI}. For the following we will assume that the differences between the nuclear transition frequency and all involved electronic transition frequencies are sufficiently large that electron bridge processes [6,7] which couple nuclear and electronic excitations are not significantly resonantly enhanced. In the case of resonance, one would expect that the lifetime of the isomeric state is much smaller than the unperturbed radiative lifetime.

One main contribution to H_{HFI} is the electric-monopole interaction between the nuclear charge distribution and the electronic charge density at the position of the nucleus. Changes in the latter produce the so-called "isomer shifts" or "chemical shifts" of nuclear transitions that are well known in Mössbauer spectroscopy. These frequency changes are typically less than 1 GHz and must also be expected to occur in an isolated atom when the electronic state or the charge state of the ion is changed. Another important contribution to H_{HFI} is the coupling between the nuclear magnetic-dipole moment and the magnetic field produced by the valence electrons. In the 3.5 eV transition of Th-229 the nuclear spin changes by $\hbar$ and the magnetic moment by about 0.5 nuclear magnetons [4]. For a $7s$ electronic state this will lead to a change of the hyperfine splitting of several gigahertz, which can easily be detected in high-resolution laser spectroscopy. This shows the practical feasibility of the double-resonance method proposed above.

With an estimated lifetime of the isomeric state of thorium of 2–4 hours, the natural linewidth of this nuclear transition will be of the order of 10 µHz and the quality factor of the resonance can approach 10^{20}. In addition to the narrow linewidth, the nuclear resonance also shows an extraordinary insensitivity of its center frequency to external perturbations, as we will show in the following. This makes the thorium nuclear resonance an interesting candidate for the reference of an optical clock that will be highly immune to systematic frequency shifts. Broadening and shifts of a resonance associated with the motion of the atom can be effectively eliminated using techniques of trapping and cooling. Especially single laser-cooled ions in RF traps provide nearly ideal conditions [9,10]. In this case systematic line shifts may still arise from the coupling to external electric and magnetic fields, including static fields, electromagnetic waves and collisions. Typical shifts in trapped-ion experiments are discussed in refs. [10,11].

For a bare nucleus only the —usually small— nuclear moments would couple to external fields, but in the atomic system we have to take HFI into account. In an LS coupling scheme [12] the eigenstates of the coupled electronic and nuclear system are characterised by sets of quantum numbers $|\alpha, I; \beta, L, S, J; F, m_F\rangle$, where I denotes the nuclear spin, L, S, J the orbital, spin and total electronic angular momenta, F and m_F the total atomic angular momentum and its orientation. α and β label the involved nuclear and electronic configurations. In the nuclear transition, the nuclear and total-angular-momentum quantum numbers (α, I, F, m_F) can change, while the purely electronic quantum numbers (β, L, S, J) remain constant. As far as external fields are concerned, the nuclear transition frequency is essentially independent of all mechanisms that produce level shifts depending only on the electronic quantum numbers (β, L, S, J), because these do not change and consequently the upper and the lower state of the transition are affected in the same way. This applies to the
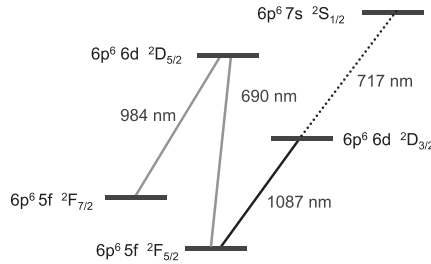
Fig. 3 – Lowest electronic energy levels of Th^{3+}. From the ground state both a closed two-level system and a closed three-level Λ system are available for laser cooling and fluorescence detection. If the excitation of the nuclear transition is performed with the electron in the metastable $^2S_{1/2}$ level, most systematic frequency shifts will be eliminated.

scalar part of the quadratic Stark effect, which typically is the dominant mechanism for the shift of electronic transition frequencies due to static electric fields, electromagnetic radiation, and collisions. The observed nuclear transition frequency is however shifted by the so-called hyperfine Stark shift, which depends on F and m_F. This effect is calculated in a higher order of perturbation theory. So far it has been considered only in the context of precision measurements of ground-state hyperfine splitting frequencies [13,14]. In the optical frequency range, a relative magnitude of 10^{-19} is expected for the hyperfine Stark shift caused by the $\approx 10\,\text{V/cm}$ room temperature black-body radiation field. In order to avoid the influence of the linear Zeeman effect, an electronic state has to be chosen such that F is an integer. In this case a Zeeman component $m_F = 0 \to 0$ is available, that shows only a small quadratic Zeeman effect around zero magnetic field (typically below $1\,\text{kHz}$ at $0.1\,\text{mT}$). Further field-dependent shifts may arise from the tensor part of the quadratic Stark effect and from the quadrupole interaction between the atomic-quadrupole-moment and electric field gradients. Both these shifts can be expressed as a product of J-dependent and F-dependent terms and vanish if either $J < 1$ or $F < 1$ [11,13]. Combining the requirements of vanishing linear Zeeman effect, quadratic Stark effect and quadrupole shift, the optimal electronic states for the interrogation of the nuclear transition are $^2S_{1/2}$ or $^2P_{1/2}$ levels in the case of a half-integer nuclear spin (as in Th-229), and $^1S_0$ or $^3P_0$ states if the nuclear spin is integer. Since one is primarily concerned with the nuclear transition, the charge state of a positive ion of the isotope in question can be selected to yield the appropriate electronic level.

Although the concepts outlined here are applicable to nuclear spectroscopy of isolated atomic systems in general, Th-229 is presently the only envisionable candidate for an experimental realization, because of its unusually low excitation energy. The next known low-energy nuclear transition is the $76\,\text{eV}$ electric octupole resonance of U-235. This isomer has already been excited in a laser-produced plasma [15]. The optical spectra of the actinides are characterised by a multitude of lines, and finding a closed two-level scheme of the shell for the nuclear-electronic double-resonance method may not always be simple. Again, the charge state of the positive ion can be selected to yield a suitable spectrum, in extremum by going to a hydrogen-like ion [16]. In the case of thorium, the triply charged ion is well suited: its level scheme is characterised by a single valence electron. While the transition wavelengths of Th^{3+} are known quite accurately [17], precise data on radiative lifetimes or hyperfine splittings are presently not available. The lowest electronic energy levels of Th^{3+} are shown in fig. 3. For laser cooling and fluorescence detection, a closed two-level system $^2F_{5/2} \to {}^2D_{3/2}$ can be excited from the ground state at a wavelength of $1087\,\text{nm}$. Alternatively, a closed

three-level Λ system $^2F_{5/2} \to {}^2D_{5/2} \to {}^2F_{7/2}$ is available which requires two laser wavelengths at 690 nm and 984 nm for continuous excitation. In this case, fluorescence could be observed at 690 nm where single-photon detection can be conveniently achieved with low background. All required laser wavelengths can be generated with semiconductor diode lasers. No electric-dipole transitions originate from the electronic ground state of Th^{3+} in the range of 1.8–15 eV so that electronic bridge processes are not expected to play an important role for the decay of the isomeric state. The $5f\,^2F_{5/2}$ ground state of Th^{3+} does not fulfill the condition $J < 1$ for elimination of the tensor Stark effect and the quadrupole shift, but a metastable $7s\,^2S_{1/2}$ state is also available. The radiative decay of this state is electric-dipole forbidden and its lifetime can be estimated to be of the order of 1 s.

Before attempting laser excitation of the Th-229 nucleus, the transition wavelength has to be determined with higher precision. Experiments that have tried to spectrally resolve the emission of visible or ultraviolet photons from the decay of the Th-229 isomer in the U-233 decay chain have so far been unsuccessful because of the high background of radioactivity-induced luminescence [18, 19]. Two groups have tried to measure the lifetime of the Th-229 isomer from a thorium sample immediately after its extraction from a U-233 source, but the results were not conclusive [20, 21].

Finally, let us mention some new applications afforded by laser excitation of the Th-229 nuclear resonance: An optical frequency standard based on a nuclear transition would constitute a high-precision oscillator whose frequency is predominantly determined by the strong interaction. This would be an interesting complement to present atomic frequency standards, which fall in one of two classes: Those that make use of atomic gross- or fine-structure transitions (*e.g.*, the single-ion optical frequency standards [10]) are determined by the electromagnetic interaction. The frequencies of those that use hyperfine transitions (*e.g.*, the caesium clock) or molecular vibrations are mainly determined by the electromagnetic interaction but also have contributions from the strong interaction via nuclear moments and masses. The comparison over time of the Th-229 transition frequency with these standards opens a new possibility for a laboratory search for temporal variations of the electromagnetic and strong-coupling constants [22, 23], respectively of their ratio, as they are predicted by some theories of grand unification.

Laser excitation can be used to address electronic and nuclear excitations independently, but both degrees of freedom are also coupled by the HFI or via electronic bridge processes [6,7]. The coherent evolution of the electronic state under the influence of a laser field will depend on the state of the nucleus and vice versa. This can be used for studies of quantum logic or entanglement, where the insensitivity of the nuclear transition to external perturbations will lead to low decoherence rates.

Laser spectroscopy of the Th-229 nuclear excitation may also be performed in a transparent solid like a crystal of ThO$_2$ or ThF$_4$ or a glass doped with thorium. Because of the high atom number, direct observation of resonance fluorescence on the weak isomer transition will be possible: a sample with 10^{10} nuclei would present a negligible radioactivity of about 1 Bq, but would scatter up to 10^6 photons per second under saturating excitation. At the high density of the solid state, strong effects of near-field coupling, superradiance or radiation trapping can be expected. Because of the isomer shift the transition frequency would depend on temperature (up to $\delta\nu/\nu\delta T \approx 10^{-10}\,\mathrm{K}^{-1}$), but nevertheless present an optical frequency reference with excellent short-term stability. The small frequency shifts produced in the nucleus by changes in the conditions of its surrounding might be used in studies of material properties, as is commonly done in Mössbauer spectroscopy.

In conclusion, we have presented a method for the detection of laser excitation of a low-energy nuclear transition in a small number of atoms or even in a single isolated atom. Such

a resonance —like that of a trapped and laser-cooled ^{229}Th^{3+} ion— is highly immune to systematic frequency shifts and might form the reference in an optical clock of very high accuracy. Apart from this application, nuclear laser spectroscopy would open a promising new field at the borderline between nuclear and atomic physics, will stimulate new applications in quantum optics, and allow improved tests of fundamental physics.

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