Laser Excitation of the Th-229 Nucleus in Th:CaF_2 Crystals

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The Th-229 nucleus is known for its unique low-energy isomeric state with an excitation energy of about 8.3 eV that places this nuclear transition in the vacuum-ultraviolet (VUV) spectral range and makes it accessible for experiments with tabletop laser systems and the tools of precision optical frequency metrology[1]. Several proposals have been made based on these properties, including a nuclear optical clock of very high accuracy and strong sensitivity in tests of fundamental physics. Reflecting the inherent robustness of nuclear transitions to external fields and chemical environments, even a solid-state version of such a clock has been proposed, based on Th-229 doped into a VUV-transparent crystal.

We report the first resonant laser excitation of the Th-229 nuclear resonance in Th:CaF_2 crystals grown at TU Wien[2] with a Th density up to $5\times10^{18}/cm^3$, using a dedicated tabletop VUV laser system developed at PTB[3], providing a spectral photon flux of more than 2×10^4 photons/(s Hz). Th:CaF_2 single crystals were grown using the Vertical Gradient Freeze method[2]. The VUV source[3] is based on four-wave mixing in Xe. It consists of two CW Ti:Sa ring lasers with two pulsed dye amplification stages, a third harmonic generation unit, a Xe gas cell, and an ultra-high vacuum VUV beam line. It operates at a repetition rate of 30 Hz, with a pulse duration of 10 ns and pulse energy of about 10 μJ. The measured spectral linewidth of the VUV source is below 10 GHz.

While scanning over the 148.3 to 149.1 nm wavelength range, the previous ±1σ-uncertainty range of the isomer transition[4], we observe a clearly detectable VUV fluorescence peak. A scan around the same wavelength with a higher doped crystal shows a higher signal with the same wavelength center position. The fluorescence lifetime in the crystal is 630(15) s, corresponding to an isomer half-life of 1740(50) s for a nucleus isolated in vacuum. The strength of the fluorescence signal is consistent with an estimate based on the laser spectral power density and the strength of the nuclear magnetic dipole transition, corresponding to the measured isomer half-life. To confirm that the laser-induced VUV fluorescence signal is due to the excitation of the Th-229 isomeric state and not related to color centers or crystal defects, we perform a ±1 THz scan around the resonance frequency using a Th-232 doped crystal, yielding no fluorescence signal above the detector dark noise level, and confirming that the observed signal is isotope specific. These results set the perspective for Th-229 nuclear laser spectroscopy towards optical nuclear clocks.

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