学位論文の要旨

Abstract of Thesis

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学位論文題目 Title of Thesis（学位論文題目が英語の場合は和訳を付記）

The X-ray induced quenching of the thorium-229 isomer states in a CaF_2 crystal host
（CaF_2 結晶ホスト中におけるトリウム 229 アイソマー状態の X 線誘起消光）

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The first excited energy level of the thorium-229 nucleus lies at approximately 8.356 eV and is referred to as *thorium isomeric state*, denoted as ^{229m}Th. This isomeric state is remarkably low in energy compared to typical nuclear excited states, which often involve gamma radiation in the keV to MeV range. In contrast, the photon emitted during the de-excitation of the ^{229m}Th isomer has a wavelength of about 148 nm, placing it in the vacuum ultraviolet (VUV) region of the electromagnetic spectrum. The lifetime of the isomer is very sensitive to the environment and it charge compensation. The ^{229m}Th lifetime has been measured to be 7 μs in neutral thorium atoms, ~10 and ~30 minutes in thorium ions doped in wide-bandgap crystal hosts and in ion trap, respectively.

The high transition energy (8.4 eV) and extremely narrow linewidth (10^{-3} Hz) of the thorium-229 isomeric state make it an ideal resonator, with a quality factor on the order of 10^{19}, for constructing a frequency standard. In other words, the transition between the ground state and the isomeric state of the ^{229}Th nucleus can serve as the basis for a nuclear clock. Moreover, given the current state-of-the-art tabletop laser technology, the ^{229}Th isomer is the only known nuclear excitation that can be directly accessed using such lasers. Consequently, thorium-229 is the sole viable candidate for realizing a practical nuclear clock.

There are currently two main approaches to construct such a nuclear clock: confining the $^{229}Th^{3+}$ ions in an ion trap or dope the $^{229}Th^{4+}$ in to a VUV transparent signal crystal host. The former scheme exhibits higher clock accuracy and the later one provides a large signal-to-noise ratio due to the large number of ^{229}Th interrogated simultaneously. The later scheme is called the solid-state nuclear clock, and it benefits from the needless of laser cooling and portable.

Recent studies have demonstrated direct laser excitation of thorium nuclei doped in single crystals of CaF_2 and $LiSrAlF_6$, marking a significant milestone toward the realization of a solid-state nuclear clock. However, several challenges remain before a high-performance solid-state nuclear clock can be achieved. One major obstacle is the exceptionally long lifetime of the isomeric state, which prolongs the interrogation cycle in clock operation (excitation and detection phases). Fortunately, our group has discovered that X-ray irradiation can accelerate the de-excitation process of the thorium isomer when embedded in a crystal host, such as CaF_2. We call this

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phenomenon as X-ray induced *isomer quenching*. For example, under nominal synchrotron X-ray irradiation, the isomeric state's decay rate can be enhanced by ten times. This observation offers a promising pathway for the artificial control of the isomeric state's dynamics in a solid-state environment, thereby advancing the feasibility of a future solid-state nuclear clock.

In this thesis, I will present the experiment setup we developed to realize the X-ray pumping of the thorium isomer state and detect the radiative decay signal of the thorium isomers. To study the temperature influence on the quenching process of the isomer state in CaF_2 crystal, I update the setup to enable the cooling of the crystal. With the updated setup, we conducted the experiments investigating X-ray-induced isomer quenching in the CaF_2 host, focusing on the effects of X-ray flux and temperature on the lifetime and yield of the isomer state. Our studies revealed a correlation between isomer production, isomer lifetime during irradiation, and post-irradiation afterglow of the target crystal across different temperatures, highlighting a strong relationship between isomer quenching and color-center dynamics. I developed a model to interpret the isomer quenching and the crystal's luminescence.

The experiment result indicate that higher X-ray flux and higher temperatures enhance the ^{229}Th isomer quenching in the CaF_2 crystal environment. However, when the X-ray source is turned off, isomer quenching ceases, regardless of the crystal's temperature. The quench lifetime (^{229m}Th lifetime during X-ray irradiation), isomer yield, and afterglow yield exhibit similar behavior when controlling the temperature, suggesting that isomer quenching is mediated by the internal conversion process in the crystal. From fitting the quench lifetime, we determine the activation energy of the electrons involved in isomer quenching to be 0.033(6) eV. The isomer quench and yield are primarily influenced by the Arrhenius factor associated with this activation energy, which corresponds to a characteristic temperature near 383(70) K. Variations around the main trend are attributed to the temperature-dependent trapping of electrons by the crystal, which affects the number of electrons captured by the intermediate state thus influencing the isomer quenching rate.

Our findings not only provide new insight into the unresolved mechanism behind the quenching of the ^{229}Th isomer in a crystal host, but also open a novel avenue for exploring the interplay between excited dopant nuclei and surrounding matrix electrons−an intrinsically intriguing topic with broad appeal to the scientific community, including nuclear physicists and condensed matter researchers. I believe that the experimental efforts presented in this thesis will make a meaningful contribution to the advancement of thorium-based solid-state nuclear clock research.