Kurzfassung

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Titel der Dissertation:	Growth and characterization of (doped) calcium fluoride crystals for the nuclear spectroscopy of Th-229

Thorium-229 provides the unique feature of a nuclear transition with an exceptionally low energy. The isomer, i.e. the excited state of the transition, is predicted to be only a few eV above the ground state, making typical atomic physics tools like lasers applicable for nuclear spectroscopy. The currently most accepted value for the isomer energy is $7.8 \pm 0.5\,\mathrm{eV}$, determined indirectly by gamma spectroscopy. This corresponds to wavelengths in the vacuum-ultraviolet, but a direct excitation or detection of the isomeric transition is still pending. The isomer lifetime strongly depends on the charge state and chemical environment of the ^{229}Th atom/ion, estimates for an isolated nucleus are on the order of a few 1000 s, providing a natural line width in the millihertz range. Possible applications are a nuclear optical clock with unprecedented stability, an optical gamma laser or a sensitive system for probing variations of fundamental constants.

The first ionization energy of the thorium atom is lower than the predicted transition energy, requiring to work with ions. Our experimental approach consists in embedding Th^{4+} into the ionic crystal lattice of calcium fluoride where it replaces a Ca^{2+} ion. CaF_2 provides a simple crystal structure and still transparent in the UV region down to 125 nm. It allows doping with a large numbers of thorium atoms ($> 10^{14}$) compared to ion traps, thereby facilitating a spectroscopic search for the nuclear transition. Numerical models have been applied to calculate the chemical environment around the thorium dopant site and how the band gap of calcium fluoride is affected by thorium doping. It is conjectured that the transparency window is only slightly reduced, validating the general approach.

In this thesis, a furnace for the growth of small-volume and highly-doped CaF_2 crystals was developed. The vertical gradient freeze method, in which a temperature gradient is established along the vertical axis of the crucible, was applied for crystal growth. The raw material was first chemically purified, removing oxygen contaminants, and then molten. Seeding presented the most delicate part of the growth process. The CaF_2 powder had to be molten without completely melting the seed crystal, which provided the orientation for the grown crystal. By precise temperature control in the furnace, the solid/liquid interface was slowly shifted upwards, thereby growing the crystal in the vertical direction.

High doping concentrations in the $0.1-1\%$ range were demonstrated for Th:CaF$_2$. The absolute concentration and the distribution of the dopant in the crystals have been determined by neutron activation analysis and gamma spectroscopy.

It was shown by UV spectroscopy that the thorium-doped CaF$_2$ crystals remain sufficiently transparent in the UV region, making nuclear excitation by lasers or synchrontrons possible, as well as the detection of gamma photons emitted during deexcitation of the ^{229}Th isomer. These photons have to be distinguished from the radio- and photoluminescence background of the doped crystals.