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EXCITATION OF THE LOW-LYING ISOMERIC LEVEL IN ^{229}Th

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Gamma-ray spectroscopy measurements have revealed the presence of an isomeric excited state in ^{229}Th only (3.5 ± 1.0) eV above the ground state, and recent measurements with a high resolution detector have resulted in the value of (7.6 ± 0.5) eV. The state is of interest due to the unique atomic-nuclear interactions which should govern its decay. The state is also of interest as excitation of the level allows the study of processes important in the development of gamma-ray lasers. Although a small number of experiments have been done in an attempt to observe the de-excitation of the isomeric level populated following alpha decay of ^{233}U, the results remain controversial and inconclusive, mainly due to ambiguities in samples used by various researchers. To date no attempts have been made to excite the low-lying level from the ground state of ^{229}Th, nor has there been a systematic study of ^{233}U in various sample forms with suitable uranium control isotopes to look for evidence of the decay of the low-lying level consistent with theoretical predictions. We present a comparison of some possible excitation techniques, including the resonant (Mössbauer effect) and non-resonant (broadband) methods, and propose some possible experiments.

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1. Introduction

Experimental gamma-ray spectroscopy measurements have revealed the presence of an isomeric excited state in ^{229}Th only a few eV above the ground state, with an initial estimate of (-1 ± 4) eV [1] Refined measurements and techniques yielded a better defined energy (3.5 ± 1.0) eV [2]. A reanalysis of the data in reference [2] resulted in a value of (5.5 ± 1.0) eV [3], and recent measurements with a high resolution detector have resulted in a value (7.6±0.5) eV [4], although the best accepted value is still taken to be 3.5 eV. These experimental measurements all rely on taking appropriate sums and differences of higher energy transitions in ^{229}Th to deduce the energy spacing between the ground state and the isomeric level. The history of these measurements, as shown in Fig. 1, is somewhat puzzling, with the most recent measurement more than 3 standard deviations above the previous [4], but at the very least it is established that the isomer is within 10 eV of the ground state. Observation of the actual decay of the isomeric level to the ground state has remained elusive.

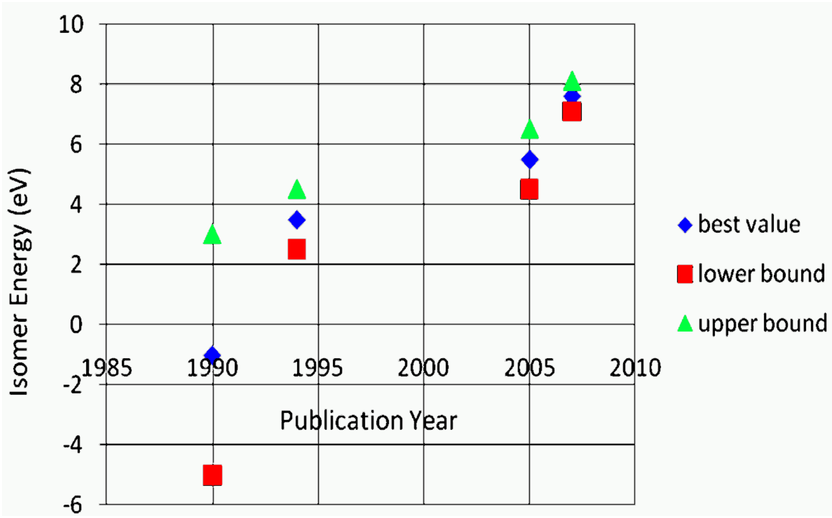

Fig. 1. Experimental determination of the energy separation of the 3/2[631] and 5/2[633] bandheads in ^{229}Th. *Data: 1990-Reich and Helmer [1]; 1994- Helmer and Reich [2]; 2005- Guimarães-Filho, Helene et. al. [3]; 2007-Beck et al. [4].*

The existence of such a low-lying nuclear level suggests a number of interesting applications, including new clock standards and tests of the standard model [5]. It has been established theoretically by Tkalya [6] that a gamma-ray laser based on the low-energy isomeric level is feasible. Such a device would require the excitation of a number of ^{229}Th nuclei to the isomeric level (perhaps using laser radiation), the creation of an inverse population by manipulation of the Zeeman sublevels of the ground state and isomeric state, and the recoilless emission and absorption of the radiation (the optical Mössbauer effect). A survey of a number of isomeric nuclear transitions with interesting level structure from the standpoint of gamma-ray lasers has been tabulated [7]. For ^{229}Th, the decay of the nuclear level is expected to be strongly dependent on the atomic configuration and hence on the chemical and physical form, opening up the possibility of using ^{229}Th as a probe for studying materials.

The low-lying state is the band-head of a 3/2[631] rotational band in ^{229}Th, and should decay to the 5/2[633] ground state by an M1 transition. A portion of the low-energy level structure in ^{229}Th is shown in Fig. 2. This extraordinarily low-lying level would have a number of possible decay channels [8 − 10], strongly dependent on the local environment of the thorium atom. Feynman diagrams for these decay channels are indicated in Fig. 3. In a neutral atom, direct gamma-ray emission is unlikely for such a low-energy gamma transition, and internal conversion is not possible since the ionization potential of the thorium atom (about 6 eV) exceeds the nuclear excitation energy (at least for a 3.5 eV energy). Consequently, a third-order process, known as the electronic bridge [8] should dominate the decay of the low-lying level. In this process, the nuclear de-excitation takes place through “discrete conversion” [9], in which an electronic transition in the thorium atom takes place, and photons with the remaining energy are emitted. The energies of the resulting emitted photons differ from the nuclear energy by the energy of the atomic transition in the atom. In a metal, non-radiative internal conversion channels with conduction electrons would dominate the decay [10].

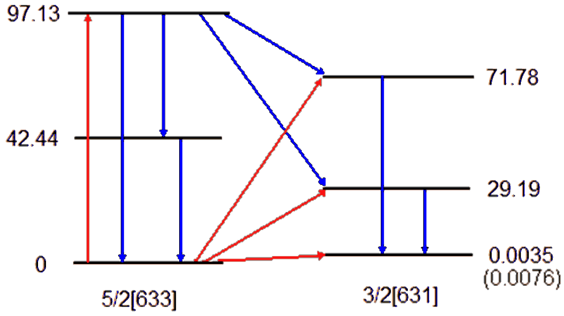

Fig. 2. Low energy nuclear structure of ^{229}Th, *indicating transitions observed from the* 97.13 keV *level following* α*-decay of* ^{233}U, *and possible excitation transitions from the ground state which populate the low-lying isomer. Energies are indicated in* keV.

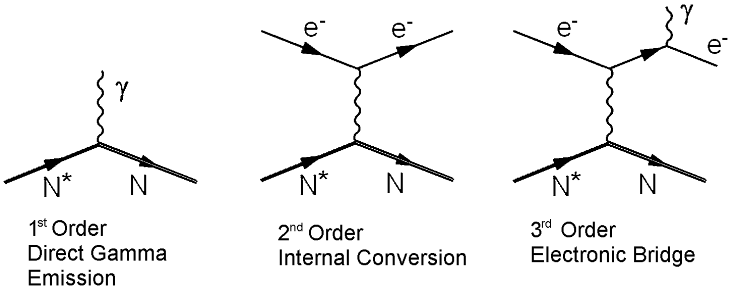

Fig. 3. Feynman diagrams for the decay process in nuclear de-excitation. For ^{229}Th *the 1st-order process dominates in wide band-gap materials like thorium dioxide, the 2nd-order process dominates for metals, and for a neutral* Th *atom the 3rd-order process dominates.*

In the wide band gap insulator thorium dioxide (ThO_2), direct gamma-ray emission in the optical range should dominate [10], as the band gap ($\sim$ 6 eV) exceeds the nuclear excitation energy. Internal conversion thus cannot occur for a 3.5 eV nuclear energy. For a nuclear energy in the range of 0.25 eV to 4.5 eV, Flambaum [5] calculates a theoretical lifetime of 10 min to 60 min in ThO_2. Beck et al. [4] suggest a halflife of about 5 hours for the newest estimate of 7.6 eV, with direct gamma-ray emission still dominating the internal conversion channel (estimated lifetime of 1000 hr). Another candidate sample form has ^{229}Th implanted into $LiCaAlF_6$, in which the Th^{4+} substitutes for the Ca^{2+}. The sample has an expected energy gap of about 10 eV [6], which would be more suitable if the 7.6 eV energy estimate proves accurate.

Based on the results of Refs. [8] and [10], the first experiment was attempted to observe optical emission from de-excitation of the low-lying level in ^{229}Th populated in the alpha decay of ^{233}U [11]. It has been suggested [10] that the electronic bridge mechanism would involve the $6d_{3/2} - 7s_{1/2}$ transition in the thorium atom with an energy separation of about 1.3 eV. Hence, based on a 3.5 eV isomer energy, one would expect optical photons emitted with energy 3.5 eV - 1.3 eV= 2.2 eV. These experiments involved observing weak optical emission from poorly-characterized solid samples of ^{233}U, which alpha decays into ^{229}Th, populating the low-lying state in a small fraction ($\sim 2\%$) of the alpha decays. Optical emission in the expected range was observed [11], as well as ultraviolet emission near 3.5 eV in a broad band of apparently discrete lines. Soon after, other researchers published the results of a similar experiment with better characterized liquid samples, including a ^{232}U control sample [12]. After correcting for the usual fluorescence of the solutions, they found a residual spectrum from the ^{233}U sample, which was similar to spectra of Ref. [11], The ^{232}U sample showed no such emission. This seems to indicate that an isotope-dependant effect is crucial to any future studies, and that the resulting emission agrees with theoretical considerations [9].

Other researchers have offered negative experimental results [13, 14], as well as explanations for the UV emission observed in Refs. [11] and [12]. At least part of the UV emission in the samples is due to excitation of nitrogen gas in air by alpha rays from the samples, in better agreement with the expected low intensity of UV emission due to the domination of the electronic bridge mechanism [15]. Although the exact physical makeup of the liquid samples in Ref. [12] is not known, it is unclear why the same spectrum was not observed in the ^{232}U sample. Theorists in Ref. [10] consider the negative results of Refs. [13] and [14] as evidence for the internal conversion of the level in the samples. All of these considerations are complicated by the dependence of the emission on the precise chemical and physical makeup of the samples, and to date no systematic study with well-characterized samples has been done.

We describe here some possible excitation mechanisms and experiments, involving both the interaction of the nucleus with intense optical photon sources both directly and indirectly via atomic electrons, as well as excitation mechanisms based on higher gamma-ray levels. Both resonant (e.g., Mössbauer effect) and non-resonant (broadband) mechanisms are discussed. The non-resonant excitation processes are relevant to experiments at both optical and X-ray energies at the Advanced Photon Source (APS) at the Argonne National Laboratory (ANL), the Duke Free Electron

Laser (FEL) Laboratory, the Jefferson Lab FEL Thomson X-ray Program, and the Idaho National Engineering Laboratory (now the Idaho National Laboratory) Advanced Test Reactor (ATR) Gamma Facility. Mössbauer effect (resonant) processes in the isomer energy range $3.5-7.6$ eV are described, and are especially interesting from the standpoint of gamma-ray laser development. Finally, some proposed experiments with ^{233}U sources of the type described in Refs. [11] and [12] are described, making use of more suitable sample forms. The ^{233}U sources also provide a source of recoillessly emitted gamma-rays for Mössbauer excitation.

2. *Excitation mechanisms*

No experiments to date have been performed to successfully excite the low-lying level from the ground state of ^{229}Th. Critical to any excitation method is the ^{229}Th sample itself. In Ref. [6], wide band-gap dielectrics are suggested as possible sample forms that are relevant to the excitation processes described here as well as gamma-ray laser applications. Excitation would involve irradiation of a sample of ^{229}Th in a suitable form with intense radiation, laser, synchrotron etc., looking for evidence of the decay of the isomer in the VUV and optical emission spectrum. As discussed above, theoretical work in Ref. [6] suggests that ThO_2 and ^{229}Th implanted into $LiCaAlF_6$ are good candidates for excitation studies. The dominant decay mode should be direct gamma-ray emission in the UV. In addition to ThO_2, other physical and chemical forms could be prepared to further investigate the influence of local environment on the nuclear properties of the level. In particular, a goal is to measure the lifetime of the level in ThO_2, for comparison with theory [6]. As with the ^{233}U experiments, identical control samples containing other isotopes of thorium would be required, to discount effects not specific to the ^{229}Th isotope. We tentatively suggest ^{228}Th as a candidate control isotope, but a detailed survey of candidates will be made to determine the best match.

2.A Broadband excitation

Direct photoexcitation of nuclei with broad source distributions may be thought of as a "brute force" method, since only photons in the very narrow linewidth Γ take part in the excitation. Even lasers have spectral widths much larger than the narrow natural linewidth of the isomeric level. Therefore, we refer to excitation processes that do not involve the Mössbauer effect (resonant absorption) as broadband. This also would include excitation sources such as the Jefferson Lab FEL Thomson X-ray source, with about 0.1% bandwidth, that is still much broader than the natural linewidth of the nuclear levels.

2.A.1. Optical energies

Direct excitation of the low-lying level in ^{229}Th has been suggested using intense UV sources [6]. The theoretical halflife of the level is estimated to be 15 s [6] with a corresponding linewidth $\Gamma_{\rm is} \approx 3 \times 10^{-17}$ eV in wide-band gap insulators. Other authors predict a halflife as long as 5 hours [4] with a linewidth $\Gamma_{\rm is} \approx 2.5 \times 10^{-20}$ eV. These linewidths are much smaller than typical optical linewidths in atomic transitions. Direct excitation may be difficult due to the extremely narrow line-width of the isomeric level, and would require intense photon sources. We propose using the intense, tunable UV laser at the Duke Free Electron Laser (FEL) Laboratory for this type of excitation. Such optical pumping of one nuclear level to another is unique in the nuclear physics world, and is of great interest in the development of gamma-ray laser technology (i.e. a coherent photon source based on nuclear levels), since such a process could be part of a three-level laser scheme [16]. Such a scheme would require a close level spacing at an excited state rather than at the ground state as in ^{229}Th, but the process of optical pumping would be similar, and may lead to techniques to identify such level structures in other isotopes. An optical gamma-ray laser based on the isomer transition itself has also been considered [6].

Processes have been studied theoretically in which the nuclear isomer is excited directly by optical photons, and the case in which the nucleus is excited indirectly via an inverse electronic bridge (EB) mechanism [17]. The inverse electronic bridge process has the advantage that the the nuclear energy need not be exactly matched, and instead one tunes the radiation source to levels in the thorium atom. It has been suggested [17] that a number of possible channels of a 3.5 eV isomer should exist, and in particular one should look for a resonance condition around a known E1-transition from the ground state to an intermediate level at 4.05 eV, or 306 nm, that has a large radiative width. Other suitable levels exist up to 5.7 eV. If the more recent energy determination of 7.6 eV in Ref. [4] is accurate, then it may be that other channels may occur as well, with increasing atomic level density.

2.A.2. High energy photoexcitation

In the decay of ^{233}U, the low-lying level is populated predominately by transitions from the 29.19 keV level in ^{229}Th. Figure 1 shows transitions starting from the 97.13 keV level following α-decay of ^{233}U, and potential excitation transitions from the ground state which populate the low-lying state. This suggests the possibility of populating the low-lying isomer indirectly by exciting the nucleus to the 29.19 keV level using intense X-rays [10]. Theoretical calculations for the case of ^{229}Th in ThO_2 are given in Ref. [10], in particular, for irradiation at the Advanced Photon Source (APS) at the Argonne National Laboratory (ANL). It is suggested that indirect excitation of the isomeric level can be achieved using synchrotron radiation by exciting the 29.19 keV level in ^{229}Th, and they estimate the partial linewidth for gamma-ray excitation to be $\Gamma_\gamma \approx 5.6 \times 10^{-9}$ eV for M1 radiation (the reduced transition probability is $B_{\rm Wu} \approx 0.011$ in Weisskopf units), and $\Gamma_\gamma \approx 0.92 \times 10^{-11}$ eV for E2 radiation ($B_{\rm Wu} \approx 6.5$) [10]. Following the excitation of the 29.19 keV level, the nucleus would then make a transition to the $3.5-7.6$ eV isomeric level. The synchrotron radiation would not damage the dielectric structure of ThO_2, which would, e.g., create luminescence centers that hamper optical measurements. The $0.0 \rightarrow 71.78$ keV transition could also populate the isomeric level, but with a much smaller probability [10]. We propose performing these types of experiments at APS, as well as similar experiments at the Jefferson Lab FEL Thomson X-ray Program, as an alternative approach which would take advantage of the production of intense, essentially monochromatic photon beams at X-ray energies (the facility is also capable of producing UV at the 1 kW level) [18]. The Jefferson Lab high average power (10 kW) FEL facility [18] has the capability of producing high flux, sub-picosecond X-rays, with intensities of the order of 10^{12} X-rays/sec/mm^2/mrad^2/(0.1% bandwidth).

Excitation of isomeric nuclear levels has been done in the past as part of studies of gamma-ray laser development [7] and studies utilizing intense bremsstralung radiation have been performed [19]. Examples of photo-excitation include well-known isomeric levels in ^{115}In, ^{113}In and ^{111}Cd, which have also been activated using intense gamma-rays from spent nuclear fuel rods at the Idaho National Engineering Laboratory (now the Idaho National Laboratory) at the Advanced Test Reactor (ATR) Gamma Facility, providing $\sim 10^6$ R/hr [20]. Such wide-band photon sources provide high intensity, but do not allow a detailed study of the high-energy "gateway levels" which are excited by the intense continuum sources to feed the isomeric level. These gateway levels correspond, for example, to the 29.19 keV level which populates the isomeric level in ^{229}Th in the excitation scheme in Ref. [10], as well as higher levels. The gateway levels may be deduced by varying the endpoint energy of the electron beam in the bremsstralung experiments. For example, in Ref. [19] gateway levels around 3 MeV were deduced for populating the 335 keV isomer in ^{115}In. The use of tunable narrow-band photon sources, such as the High Intensity Gamma Source at the Duke FEL, would help to identify these gateway levels and help to clarify the excitation processes. We propose a study of ^{115}In, ^{113}In and ^{111}Cd, as well as other suitable long-lived isomeric nuclei, in addition to ^{229}Th.

2.B. Resonant excitation

One of the necessary requirements of a gamma-ray laser is the occurrence of the Mössbauer effect, the resonant absorption of gamma radiation by nuclei. The resonant absorption of optical photons by atoms is quite common, but in nuclei the higher energy photon emission imparts considerable recoil momentum to the nuclei, thus diminishing slightly the energy of the emitted photon. Though slight, the energy loss of the photon can be orders of magnitude greater than the natural linewidth of absorption, thus eliminating resonance. To make matters worse, the absorbing nucleus must also recoil, compounding the loss of resonance. In the Mössbauer effect, the nuclei are locked in a high Debye temperature solid, and recoil of individual nuclei cannot occur if the recoil energy is insufficient to excite phonon modes in the solid. Instead, the entire crystal recoils, and does so with negligible energy, thus restoring resonance between the emitter and absorber. In Mössbauer excitation processes, the excitation radiation and the absorption cross section both have natural linewidths.

The nuclear gamma-ray cross-section is described by a Lorentzian distribution of energy, the so-called Breit-Wigner resonance, with a linewidth determined by the full width of the level, which may be written [21]

$$\sigma(E) = \sigma_0 \frac{(\Gamma/2)^2}{(E - E_0)^2 + (\Gamma/2)^2} \,. \tag{1}$$

Here Γ is the *total* linewidth of the level. The recoilless events in the Mössbauer effect occur with this cross section σ_{RL} where the subscript RL refers to recoilless events. The peak cross-section is determined from the nuclear parameters for the case of single transition (as in the 14.4 keV transition in ^{57}Fe), and may be expressed

$$\sigma_0 = 2\pi \left(\frac{\hbar c}{E_0}\right)^2 \left(\frac{2I_e + 1}{2I_g + 1}\right) \frac{1}{1+\alpha} \tag{2}$$

where the total width is traditionally written $\Gamma = \Gamma_\gamma(1 + \alpha)$, where Γ_γ is the radiative linewidth of the level and α is the internal conversion coefficient. The recoil-less fraction is given by a Debye-Waller (Lamb-Mössbauer) factor

$$f(T) = \exp\left[-\frac{6E_g}{k_B\,\theta_D}\left(\frac{1}{4} + \frac{T^2}{\theta_D^2}\int\limits_0^{\theta_D/T} \frac{x\mathrm{d}x}{\mathrm{e}^x - 1}\right)\right], \tag{3}$$

where θ_D is the Debye temperature of the solid, and the nuclear recoil energy is $E_g = E_\gamma^2/(2Mc^2)$. Emission events which occur with recoil have a fraction $1-f(T)$. One may define such a factor for both the source and the absorber, both of which are best cooled cryogenically for maximum recoilless fraction.

2.B.1. Optical Mössbauer effect at 7.6 eV

For the case of the optical Mössbauer effect in ^{229}Th, M is the ^{229}Th mass and $E_\gamma \approx 3.5 - 7.6$ eV is the energy of the optical photon emitted. The isomer recoil energy is very small ($E_g \approx 1.4 \times 10^{-10}$ eV) and so $f \approx 0$. However, the linewidth is extremely narrow, which means that inhomogeneous hyperfine effects may adversely affect the measured linewidth, at the expense of peak cross section. For the 7.6 eV isomer transition, the internal conversion coefficient in thorium dioxide is thought to be small ($\alpha \approx 0.1$ for a 1000 hour conversion half-life compared with a 5 hour half-life for gamma-ray emission [4]), and so direct gamma-ray emission should dominate. In principle, a ^{233}U source could be a source of the 3.5-7.6 eV photons. The observance of the Mössbauer effect for the isomer transition in ^{229}Th is crucial to the development of a gamma-ray laser based on the low energy level.

2.B.2. Mössbauer effect at 97.1 keV

As described above, the 7.6 eV level has been considered as a Mössbauer transition, a necessary requirement for a gamma-ray laser based on this low energy level. We consider here the possibility of utilizing another Mössbauer transition, an intra-band transition from the ground state to the 97.13 keV level in ^{229}Th. As indicated in Fiq. 1, inter-band transitions from the 97.13 keV level to the 71.78 keV and 29.19 keV levels populate the low-lying isomer. For the application of the Mössbauer effect, a suitable source of the radiation must also exist. In this excitation scheme, a ^{233}U source would be used as a source of 97.13 keV gamma-rays. This gamma ray is the second most intense observed in the decay of ^{233}U (the most intense is the 42.44 keV, which should also show the Mössbauer effect). Based on observed gamma-ray intensities observed in the alpha-decay of ^{233}U [22], we estimate the partial width for excitation of the 97.13 keV level to be $\Gamma_\gamma \approx 4 \times 10^{-8}$ eV (Coulomb excitation studies suggest the value $\Gamma_\gamma \approx 1 \times 10^{-7}$ eV [23]). The peak cross section in Eq. (1) is $\sigma_0 \approx 5 \times 10^{-21}$ cm^2. The recoil energy of the ^{229}Th nucleus is about 0.022 eV. Thorium dioxide has a relatively high Debye temperature $\theta_D = 397$ K [24] which makes it suitable for Mössbauer experiments. This gives a recoilless fraction $f \approx 0.38$, or 38% for ^{229}Th in ThO_2 at low temperatures ($T \ll \theta_D$), e.g. 4.2 K. At 77 K, the recoilless fraction is retained at $f \approx 0.37$, and room temperature it drops to about $f \approx 0.08$. Based on the transition intensities from the 97.13 keV level, we estimate the fraction of inter-band transitions that populate the isomer to be about 0.27. If the ^{229}Th nuclei are in a similarly suitable absorber, then the emission and absorption lines of the source and absorber overlap, leading to much higher cross-sections than obtained using broad-band sources, such as APS and Jlab. This process thus may offer the possibility of obtaining observable activity of the isomer with relatively low radiation intensities.

Since there are several transitions that occur from the 97.13 keV level, one can define an "effective" conversion coefficient such that $\Gamma = \Gamma_\gamma(1 + \alpha_{\rm eff})$ is defined similarly, and write more generally

$$\sigma_0 = 2\pi \left(\frac{\hbar c}{E_0}\right)^2 \left(\frac{2I_e + 1}{2I_g + 1}\right) \frac{\Gamma_\gamma}{\Gamma} \,. \tag{4}$$

Here the quantity $\alpha_{\rm eff}$ includes not only the internal conversion channel to the ground state, but also includes both gamma-ray and internal conversion channels for the three other transitions as seen in Fig. 1. For the 97.13 keV level, $\alpha_{\rm eff} \approx 84$. Since the total linewidth Γ is relatively large and the partial width for gamma absorption Γ_γ is relatively small compared with internal conversion channels, the peak cross section is smaller than cross sections for processes involving only a single transition.

The Mössbauer excitation scheme is of interest for several reasons. Like the direct UV excitation described above, the Mössbauer excitation is interesting from the standpoint of gamma-ray lasers. Any proposed gamma-ray laser scheme would presumably require the Mössbauer effect as part of the lasing process [16]. Also, this would be an important example of the use of the Mössbauer effect to populate a nuclear isomeric level (besides the Mössbauer level itself, of course). Finally, a Mössbauer excitation experiment would not require the large photon source facilities (APS, Duke FEL, Jlab FEL) which are needed for broadband excitation, although it would probably require cryogenically cooling the ^{233}U source and ^{229}Th absorber to obtain as high a recoilless fraction as possible. As with the other excitation methods, evidence of isomer population could be done with VUV optical detection methods.

Since the Mössbauer effect is temperature dependent (the recoilless fraction is given by the Debye-Waller factor), the excitation rate should similarly show temperature dependence, providing a unique way of correlating the results of optical measurements. Also, the Mössbauer effect already has a "built-in" way of affecting the resonance by Doppler shifting the source relative to the absorber. Due to the relatively short half-life of the 97.13 keV level (0.172 ns), the velocity line-width 2Γ for this resonance would be about 18 mm/s. This compares with 0.19 mm/s for the Mössbauer transition in ^{57}Fe, half-life 100 ns. The source and absorber would be virtually in resonance at zero Doppler velocity, and hyperfine interactions would probably not drastically affect the resonance condition. The source could then be Doppler-shifted to move off resonance, providing a method (along with temperature dependence) of confirming that the resulting optical measurements are correlated with the excitation process. Note that other types of excitation processes do not provide similar methods for correlating the optical measurements.

It is interesting to note that the Mössbauer effect would be expected to occur in old samples of ^{233}U in which ^{229}Th nuclei in the ground state had gradually collected. This Mössbauer self-absorption by these nuclei would occur with great efficiency (in a material with a high Debye temperature θ_D), and would increase the kind of temperature-dependent isomer activity that would be obtained in the Mössbauer effect experiments described above. Thus increases in the recoilless fraction at cryogenic temperatures would allow for studying this Mössbauer excitation process in the source. At low temperatures ($T \ll \theta_D$) the observed activity of the isomeric level should be highest, and at high temperatures ($T \gg \theta_D$) only the isomer activity associated with the alpha decay of ^{233}U should be observable. Since a Mössbauer excitation experiment would require just such a ^{233}U source (as a source of either the 7.6 eV radiation or the 97.13 keV radiation), such a setup would allow the measurement of vacuum ultraviolet (VUV) photons as well as the ^{229}Th target (absorber).

3. ^{233}U *experiments*

Experiments aimed at observing the decay of the isomeric level in ^{229}Th by measuring the emission from ^{233}U sources as attempted by [11, 12] may still present a way of revealing the nuclear properties of ^{229}Th, given the difficulties associated with excitation experiments with low cross sections. The interpretation of experimental work of ^{233}U decay experiments, as in Refs. [11 − 14], is difficult because each researcher used a different sample form, with little possibility for comparison. Theoretical analysis of this case is problematic for solid samples, as the daughter ^{229}Th atom may sit in a defect site in the Uranium material, depending on its composition. Also, except for reference [11], in which ^{233}U and ^{232}U sources were compared, experiments aimed at isolating isotopic effects have not been performed.

4. Conclusion

The exotic nuclear structure of ^{229}Th makes it a candidate for a number of important applications. Key to the development of these applications are techniques to excite the isomer and observe its decay to the ground state. We have considered a variety of photoexcitation techniques including resonant and non-resonant processes. We propose preparing a set of well-characterized samples in various physical and chemical forms (e.g., solids, solutions, perhaps even gases), including identical samples containing ^{234}U for comparison. ^{234}U seems to be a good control isotope as its half-life is on the order of that of ^{233}U, and hence one would expect the samples would have similar spurious effects due to fluorescence. The ^{232}U source in Ref. [11] had considerably higher fluorescence than the ^{233}U source in the liquid samples used, which may add some ambiguity to the interpretation of their results, and ^{234}U might be a better control source. Suitable ^{233}U sources would be required for the Mössbauer excitation experiments, described above, as a source of 97.13 keV gamma-rays. For this purpose the ^{233}U isotope must be put into a high Debye temperature solid to maximize the recoil-less fraction of emitted gamma-rays. The source must, of course, have low self-absorption of the 97.13 keV gamma-rays, and ^{233}U in thorium dioxide appears to be a suitable sample form for this purpose.

References

[1] C. W. Reich and R. G. Helmer, Phys. Rev. Lett. **64** (1990) 271.

[2] R. G. Helmer and C. W. Reich, Phys. Rev. C **49** (1994) 1845.

[3] Z. O. Guimarães-Filho and O. Helene, Phys. Rev. C **71** (2005) 044303.

[4] B. R. Beck, J. A. Becker, P. Beiersdorfer, G. V. Brown, K. J. Moody, J. B. Wilhelmy, F. S. Porter, C. A. Kilbourne and R. L. Kelly, Phys. Rev. Lett. **98** (2007) 142501.

[5] V. V. Flambaum, Phys. Rev. Lett. **97** (Sept. 2006) 092-502.

[6] E. V. Tkalya, Cornell University Library, arXiv:1011.0858v1 [nucl-th] 2 Nov 2010.

[7] E. D. Arthur and D.C. George, J. Quant. Spectrosc. Radiat. Transfer **40**, 6 (1988) 651.

[8] V. F. Strizhov and E. V. Tkalya, Zh. Eksp. Teor. Fiz. **99** (1991) 697 [Sov. Phys. JETP **72** (1991) 387].

[9] I. M. Band, Zh. Eksp. Teor. Fiz. **120**, No. 5 (2001).

[10] E. V. Tkalya, A. N. Zherikhin and V. I. Zhudov, Phys. Rev. C **61** (2000) 064308.

[11] G. M. Irwin and K. H. Kim, Phys. Rev. Lett. **79** (1997) 990.

[12] D. S. Richardson, D. M. Benton, D. E. Evans, J. A. R. Griffith and G. Tungate, Phys. Rev. Lett. **80** (1998) 3206.

[13] S. B. Utter, P. Beierdorfer, A. Barnes, R. W. Lougheed, J. R. Crespo Lopez-Urrutia, J. A. Becker, and M. S. Weiss, Phys. Rev. Lett. **82** (1999) 505.

[14] R. W. Shaw, J. P. Young, S. P. Cooper, and O. F. Webb, Phys. Rev. Lett. **82** (1999) 1109.

[15] G. M. Irwin and K. H. Kim, Phys. Rev Lett. **83** (2 Aug. 1999) 1073.

[16] G. C. Baldwin and R. V. Khokhlov, Physics Today, February 1975, p.32.

[17] E. V. Tkalya et. al, Physica Scripta **53** (1996) 296.

[18] G. Neil et al., *Status of the Jefferson Lab IR/UV High Average Power Light Source*, FEL 2002, Chicago, IL 8-13 Sept. 2002.

[19] P. von Neumann-Cosel, A.Richter, C. Spieler, W. Zeigler, J. J. Carroll, T. W. Spinor, D. G. Richmond, K, N. Taylor and C. B. Collins, Phys. Lett. B **266** (1991) 9.

[20] G. M. Irwin, *Photoexcitation of Nuclear Isomers in the ATR Gamma Facility*, EG&G Internal Technical Report ST-PHY-91-046, Idaho National Engineering Laboratory, Sept. 1991. This work was performed at the Idaho National Engineering Laboratory at the Advanced Test Reactor (ATR) Gamma Facility, 1991.

[21] Hans Frauenfelder, *The Mössbauer Effect*, W. A. Benjamin, Inc. New York, 1962.

[22] L. A. Kroger and C. W. Reich, Nucl. Phys. A **259** (1976) 29.

[23] C. E. Bemis, Jr., F. K. McGowan, J. L. C. Ford, Jr., W. T. Milner, R. L. Robinson and P. H. Stelson, Physica Scripta **38** (1988) 657.

[24] B. T. M. Willis, Proc. Roy. Soc. (London), Series A, Mathematics and Physical Sciences, Vol. **274**, No. 1356 (Jun. 25, 1963), pp. 134.

UZBUDA NISKOLEŽEĆEG IZOMERNOG STANJA U ^{229}Th

Spektroskopska mjerenja gama zračenja otkrila su izomerno uzbudno stanje u ^{229}Th samo (3.5 ± 1.0) eV povrh osnovnog stanja, a novija mjerenja s detektorom visokog razlučivanja dala su iznos od (7.6 ± 0.5) eV. Zanimanje za ovo stanje je zbog mogućeg jedinstvenog međudjelovanja atom-jezgra koje upravlja njegovim raspadom. To je stanje zanimljivo i zbog mogućnosti proučavanja procesa koji su važni za razvoj lasera gama zračenja. Iako je načinjen malen broj pokušaja da se opazi raspad tog izomernog stanja koje se popunjava alfa raspadom ^{233}U, ishodi su nesuglasni i nisu zaključni, najviše zbog neodređenosti u sastavu uzoraka koje su razni istraživači rabili. Do sada se nije pokušalo uzbuditi to niskoležeće stanje polazeći od osnovnog stanja ^{229}Th, niti je načinjeno sustavno proučavanje s uzorcima ^{233}U u različitim spojevima, i s odgovarajućim uranovim izotopima za provjere, da bi se dobili podaci o raspadu o tom niskoležećem stanju radi usporedbe s teorijskim predviđanjima. Opisujemo usporedbe nekih mogućih metoda za uzbudu, uključujući rezonantnu (Mössbauerov efekt) i nerezonantne (širokopojasne) metode, te predlažemo neke moguće eksperimente.