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Level Lifetimes in ^{94}Zr from DSAM Measurements following Inelastic Neutron Scattering

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Abstract. The lifetime of the second 2^+ state in ^{94}Zr was redetermined by the Doppler-shift attenuation method following inelastic neutron scattering (DSAM-INS) from metallic Zr and ZrO_2 samples of natural isotopic abundance. The new value for the level lifetime differs significantly from the previously published value, with the new lifetime found to be roughly twice that value. A reanalysis of the original γ-ray data from the enriched $^{94}ZrO_2$ sample failed to expose the source of this discrepancy; however, powder X-ray diffraction and scanning electron microscopy performed on each scattering sample, including the enriched sample used previously, provide clues to an explanation and reveal the role of the chemical properties of the sample material in DSAM-INS lifetime determinations.

1 Introduction

Several years ago, our group performed ^{94}Zr$(n, n'\gamma)$ measurements with an enriched $^{94}ZrO_2$ scattering sample. Based on the observation that the 752-keV transition from the second 2^+ state at 1671 keV to the first excited state at 919 keV exhibits a large $B(M1; 2_2^+ \rightarrow 2_1^+)$, we identified the second 2^+ state as the lowest mixed-symmetry state [1, 2]. In addition, these measurements of ^{94}Zr revealed anomalous behavior not observed in any other nucleus; the $B(E2)$ value for the transition from the second excited 2^+ state to the ground state was found to be larger than that from the first 2^+ state to the ground state. This nucleus thus emerged as the lone example of an inversion of the $E2$ strengths for the lowest-lying 2^+ excitations. As questions have recently been raised about this anomaly, we carried out additional $(n, n'\gamma)$ measurements using metallic zirconium, zirconium (IV) oxide, and zirconium (IV) hydroxide samples of natural isotopic composition.

2 The Doppler-shift attenuation method following inelastic neutron scattering

The Doppler-shift attenuation method following inelastic neutron scattering (DSAM-INS) may be utilized to obtain level lifetimes in the femtosecond regime. By measuring the γ-ray energy as a function of the angle of detection with respect to the direction of the incident neutrons, the experimental

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attenuation factor, $F(\tau)$, which describes the slowing down process of the recoiling nucleus, can be determined. Equation 1 demonstrates this relationship, where $E_\gamma(\theta)$ is the γ-ray energy as a function of the angle of detection with respect to the direction of the incident neutrons, E_0 is the energy of the γ emitted by the nucleus at rest, $v_{c.m.}$ is the recoil velocity in the center-of-mass frame, and c is the speed of light.

$$E_\gamma(\theta) = E_0\Big[1 + F(\tau)\frac{v_{c.m.}}{c}cos\ \theta\Big], \tag{1}$$

The experimental attenuation factor can be compared with theoretical calculations of $F(\tau)$ as a function of lifetime to determine the level lifetime. An $F(\tau)$ value near unity corresponds to very short lifetimes (few fs), while $F(\tau)$ values near zero correspond to long lifetimes (> a few ps). A detailed description of the DSAM-INS may be found in Ref. [3].

3 Level lifetimes in the stable Zr isotopes by the Doppler-shift attenuation method

Angular distribution measurements were performed with the ^{nat}Zr$(n, n'\gamma)$ reaction on scattering samples of metallic zirconium and zirconium (IV) oxide, ZrO_2, of natural isotopic composition. Nearly monoenergetic neutrons were produced by the ^{3}H$(p, n)^3$He reaction with incident energies of 2.0, 2.3, 2.5, 2.8, and 3.5 MeV. The emitted γ rays were detected with a single HPGe detector and the Doppler-shift attenuation method was then used to extract lifetimes.

As the samples were of natural isotopic abundance, lifetimes were obtained for levels in all of the stable Zr nuclei, *i.e.*, A = 90, 91, 92, 94, and 96. This sample composition permitted comparisons with lifetimes reported in the literature, which employed non-DSAM techniques for ^{90,91,96}Zr [4–6]. Excellent agreement was demonstrated both between the results obtained with the metallic and oxide scattering samples and with literature values, and is considered a validation of the method. However, for the lifetimes measured for ^{94}Zr, a discrepancy is apparent compared with the published values from our prior measurements [1, 2]; the new values are approximately a factor of two longer than the previous ones. The lifetime of the 1671-keV 2_2^+ state was previously published to be 183^{+13}_{-12} fs, but is now quoted as a weighted average of the metal and oxide measurements (shown in Fig. 1), 368^{+27}_{-23} fs.

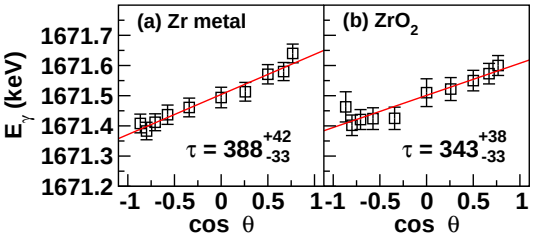

Figure 1. Measured γ-ray energies as a function of $cos\ \theta$ for the 1671-keV γ ray from the 1671-keV level in ^{94}Zr at E_n=2.0 MeV for (a) metallic Zr and (b) ZrO_2. The quoted lifetime, a weighted average of the two values shown, is 368^{+27}_{-23} fs.

4 Investigating the chemical properties of the scattering samples

The source of the discrepancy in the level lifetimes measured for ^{94}Zr was not revealed by a reanalysis of the prior data. Therefore, other possible causes required investigation, the first of which was the

scattering sample. Powder X-ray diffraction (XRD) and scanning electron microscopy (SEM) were utilized to investigate the chemical properties of both the enriched $^{94}ZrO_2$ and natural ZrO_2 material. While the XRD confirmed that both materials are monoclinic zirconium (IV) oxide, the spectrum for the enriched material indicated an amorphous component may be present as well. The scanning electron micrographs show that the natural material is of uniform crystalline composition, but the enriched oxide does indeed contain a large amorphous component in addition to a crystalline component.

5 Amorphous materials as scattering samples for DSAM-INS measurements

As it is normally assumed the scattering sample is composed of crystalline material in the DSAM analysis, we chose to reproduce the measurements using an amorphous Zr-containing compound, zirconium (IV) hydroxide, $Zr(OH)_4$. The sample was confirmed to be amorphous using XRD and SEM.

The lifetimes measured using $Zr(OH)_4$ differ significantly from those obtained using the natural metallic and oxide scattering samples. For example, the lifetime obtained for the 1671-keV 2_2^+ level using the hydroxide sample at E_n=2.0 MeV is 198^{+24}_{-19} fs, compared to the average of the metallic and oxide results of 368^{+27}_{-23} fs. It is obvious that the amorphous hydroxide sample results in systematically shorter lifetimes.

6 Lifetime as a function of particle size

The next logical step was to pursue the cause of this adverse effect on the measured lifetimes. One possible explanation is that amorphous materials are composed of small particles, which may allow the recoiling nucleus to scatter between multiple particles before coming to rest. This description would mean that the nucleus experiences essentially no slowing between particles and its velocity at the time of decay would be larger than that of a nucleus which experiences uniform slowing as would occur within crystalline material. A larger velocity would result in a larger attenuation factor, and a shorter lifetime.

To investigate this supposition, we examined ZrO_2 with various crystalline domain sizes. These samples were prepared by dissolving zirconium (IV) oxynitrate hydrate, $ZrO(NO_3)_2 \cdot nH_2O$, in deionized water and precipitating $Zr(OH)_4$ by adding concentrated ammonium hydroxide. The precipitate was then filtered, dried, and calcined (heated in static air) at various temperatures. The crystalline domain size of the product, ZrO_2, was controlled by the calcination temperature, *i.e.*, higher temperatures result in larger crystalline domains. Samples were prepared at four different temperatures, resulting in four different crystalline domain sizes as measured from XRD: 400°C, 600°C, 800°C, and 1000°C yielding domain sizes of 6.1 nm, 15.7 nm, 24.6 nm, and 36.8 nm, respectively. By using these materials as scattering samples for INS at E_n=2.5 MeV, level lifetimes were measured as a function of crystalline domain size. It was found that for a given level, the lifetime increases with increasing crystalline domain size as shown in Fig. 2. Note also that the accepted lifetime is that determined at E_n=2.0 MeV, where (slow) feeding is not a factor as it can be at higher energies.

7 Conclusions

The new DSAM-INS measurements of level lifetimes in ^{94}Zr show that the previous measurements [1, 2] were in error. The discrepancy is attributed to the chemical properties of the scattering sample material, which contained a large amorphous component. This premise is supported by measurements

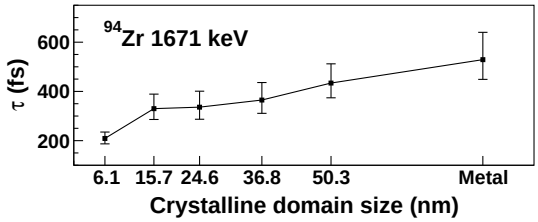

Figure 2. Lifetime as a function of particle size for the 1671-keV level in ^{94}Zr at E_n=2.5 MeV. The largest domain size corresponds to the commercial material; the value for the metallic sample is plotted at the far right.

utilizing a known amorphous material, which were also erroneous, and by measurements which employed materials containing various particle sizes, where the lifetime was found to increase with increasing particle size. Therefore, it is concluded that the chemical properties of the scattering sample, particularly the size of the particles, are of utmost importance for properly determining level lifetimes with DSAM-INS.

The aforementioned anomalous inversion of the $E2$ strength and the structure of ^{94}Zr must also be reconsidered. As the new lifetime for the 1671-keV 2_2^+ level was found to be approximately a factor of two longer than the published value [1, 2], the $B(E2)$ is approximately half. This nearly excludes the possibility of $B(E2; 2_2^+ \rightarrow 0_1^+)$ being greater than $B(E2; 2_1^+ \rightarrow 0_1^+)$, 3.9(3) W.u. vs. 4.9(11) W.u. [7, 8]. However, a strong $M1$ transition from the 1671-keV 2_2^+ level to the 919-keV 2_1^+ is still evident, 0.17 μ_n^2, thus this level remains a candidate for the lowest 2^+mixed-symmetry state, although alternative interpretations have been offered [8].

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