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To cite this version:

B. Dunlap, G. Shenoy. MEASUREMENT OF SMALL RESONANCE EFFECTS IN 237Np DEBYE TEMPERATURE OF Am METAL. Journal de Physique Colloques, 1976, 37 (C6), pp.C6-55-C6-58. <10.1051/jphyscol:1976613>. <jpa-00216644>

HAL Id: jpa-00216644
https://hal.archives-ouvertes.fr/jpa-00216644
Submitted on 1 Jan 1976

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MEASUREMENT OF SMALL RESONANCE EFFECTS IN $^{237}$Np DEBYE TEMPERATURE OF Am METAL (*)

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Résumé. — La résonance de $^{237}$Np a été mesurée à l'aide de la technique d'intégration en courant. On a obtenu des spectres avec des taux de comptage d'environ 5 MHz, ce qui permet d'accumuler des données d'excellente qualité en quelques minutes. La variation en fonction de la température de la fraction résonnante de Np dans Am métal a été mesurée à l'aide de cette technique. Dans la gamme $T = 20$ K à 100 K, on obtient une température de Debye de 125 K. Au-dessous de 20 K, la fraction résonnante décroît lorsque la température diminue. Cette anomalie est discutée en invoquant la dynamique des réseaux et l'effet de défauts de recul.

Abstract. — Measurements of the $^{237}$Np resonance have been performed using the current integration technique. Spectra have been obtained with count-rates of approximately 5 MHz, allowing high quality data to be accumulated in only a few minutes. Using this technique, the temperature dependence of the resonance fraction of Np in Am metal has been measured. In the range $T = 20$ K to 100 K, the data are described by a Debye temperature of 125 K. Below 20 K, the resonance fraction decreases with decreasing temperature. This anomaly is discussed in terms of lattice dynamics and recoil damage effects.

1. Introduction. — The 60 keV resonance in $^{237}$Np has provided a very useful tool for the measurement of electronic and magnetic properties in actinide materials. This is so because of the favorable nuclear parameters for this Mössbauer transition. In the past, the measurements have been limited to temperatures roughly below liquid nitrogen temperatures because of the small resonance effect at higher temperatures. Calculations show that reasonable effects should be obtainable to much higher temperatures than that of liquid nitrogen if natural width resonance lines could be observed. However, for reasons which still remain somewhat obscure, one always observes linewidths which are approximately thirty times the natural width, with a commensurate reduction in resonance amplitude. This, combined with the usual limitations on count rates to $\sim 100$ kHz imposed by pulse handling electronics, greatly diminishes the usable temperature range of this resonance. Thus far no systematic attempt has been made to improve either the instrumentation or the source material to utilize the resonance more properly. In this paper we discuss the use of the integrating counting method in $^{237}$Np spectroscopy. The technique is then used to measure the temperature dependence of the resonance fraction for Np in a common source material, $\alpha$-Am metal. An anomaly in the resonance fraction occurring below 20 K is presented and discussed.

(*) Work performed under the auspices of the U. S. Energy Research and Development Administration.

2. Experimental procedures and results. — In the integrating counting, or current integration technique, the usual pulse amplifier and single-channel electronics are omitted [1, 2]. Instead one devises a way to measure the photo-current from a scintillator directly, this being proportional to the total $\gamma$-ray flux onto the detector. Ideally, this is best applied to cases where only a single $\gamma$-ray is present. In practice, however, it may be useful in cases where this is not so since it may be possible to achieve enough increase in count rate to more than compensate the decrease in the resonance effect caused by including other $\gamma$-rays indiscriminately [1]. In the case of $^{237}$Np, the situation is near ideal. In the commonly used $\gamma$-ray source (the $\alpha$ decay of $^{241}$Am), only a single $\gamma$-ray is present, that being the 60 keV line of interest. There are some $x$-rays at $\sim 100$ keV emitted from the source as well as from the absorber, but these can be kept to under 10 % of the total intensity.

The system utilized here is essentially identical to that described by Viegers and Trooster [1]. The anode of the photomultiplier is connected to an integrating amplifier, which converts the current to an analog signal. This in turn is digitized using a voltage to frequency converter (Hewlett-Packard 2212A). Pulses are thus produced with a frequency proportional to the $\gamma$-ray count rate, which are fed into a multi-channel analyzer in the conventional time-mode configuration. With our source of $^{241}$Am in the form of $\alpha$-Am metal, we obtain the equivalent of a $\gamma$-ray count rate of approximately 5 MHz under experimental conditions.
Comparing the spectra obtained with those from a conventional pulse-handling system, we see none of the distortions in line shape discussed by Kankeleit when the current method is used at much higher count rates [2]. In figure 1, we show a spectrum taken with this system where the Am metal source has been held at 4.2 K but the NpO₂ absorber is at room temperature. Data were accumulated for about 15 hours. This is the first room temperature observation of this resonance.

Using the current integration method, we have measured the temperature dependence of the emission line from $^{237}$Np in an α-Am metal source. For this sample (approximately 8 years old), the impurity level is about 1.5 % $^{241}$Np resulting from the Am decay. In these experiments, an absorber of NpO₂ (200 mg/cm²) was held at 4.2 K, immersed in a He bath. The Am metal source was held in an exchange gas system so that the temperature could be varied over a wide range (from 4.2 K to ~100 K) without affecting the absorber temperature. The Am-metal source was pressed into an aluminum holder and subsequently mounted firmly in a brass container. Temperatures were measured at the surface of the brass container with both carbon and platinum resistance thermometers. At the lowest temperature, excellent spectra could be obtained in 1/2 hour. All data were analyzed by least-squares fitting to a single Lorentzian [3]. The line-width was essentially constant (see discussion below) independent of source temperature, at 4.2 ± 0.2 mm/s and the isomer shift at 8.2 ± 0.1 mm/s. Relative resonance intensity, $f(T)$, as a function of temperature is shown by the closed circles in figure 2 up to ~100 K. At higher temperatures, the resonance intensity became too small to be accurately measured.

Above 20 K, $f(T)$ shows an ordinary monotonic increase as the temperature decreases. However, below 20 K the resonance intensity decreases, falling by about 30 % between 20 K and 3 K. The primary suspicion in such a large anomaly arises from the self-heating in the sample due to α-decay in the material. If thermal contact to the thermometer is poor then the temperature in the source may be substantially different from that of the thermometer. However, this should show up as a hysteresis in the $f(T)$ curve. We have taken a large number of spectra in various heating and cooling cycles, as well holding at a fixed temperature for many hours, and find the temperature dependence does not depend on any of these conditions.

3. Discussion. — In figure 2, we have shown a number of plots of the temperature dependence of the recoil-free fraction as calculated from a Debye model. Experimentally, we have only relative and not absolute measurements. The calculated curves have been plotted in such a way as to get best agreement between experimental and theoretical curves for the slope at the highest temperatures. In this way one establishes a Debye temperature $\theta_D = 125 \pm 5$ K which provides a good description of the data in the temperature range from 20 to 100 K.

The unusual temperature dependence below 20 K may arise from several sources. However, a detailed evaluation of the possibilities is difficult at present, primarily because of a lack of other physical measurements on Am metal. This should become better in the relatively near future because of the recent availability of $^{243}$Am metal, which is substantially less radioactive than the $^{241}$Am in common supply. For the present, we will present a number of alternative explanations, some or all of which may be applicable.

1) The observed anomaly may be an indication of a soft mode transition at low temperatures in the host material, viz., Am metal. It is difficult to justify this without additional physical measurements, since such
phenomena have not been seen in any of the lighter actinide metals, which have been studied much more extensively [4].

2) The phonon spectrum of Am metal may be poorly described by a Debye model. Such a result is indicated by recent specific heat measurements on $^{241}$Am metal [5]. Although there is some uncertainty in that data due to an inability to clearly separate the electronic and lattice contributions to the specific heat, nonetheless there appears to be a large change in the apparent Debye temperature which varies from about 100 K at $T = 10$ K to 260 K at $T = 150$ K, remaining constant for higher temperatures. The present data would correspond to a variation in $\theta_D$ from 80 K at $T = 3$ K to 125 K at $T = 20$ K, remaining constant above this to $T \sim 100$ K, with the data being inadequate to investigate higher temperatures.

3) The anomaly may be a result of radiation damage occurring from the $\alpha$-decay at low temperatures [6]. From the standpoint of the present experiment, the primary effect of the $\alpha$-decay is to impart about 100 keV recoil energy to the neptunium nucleus emitting the 60 keV resonance gamma-ray. There are low temperature self-recoils which create damage, primarily vacancies and interstitials in the host Am metal. The primary recoils will eventually come to a stop in a time much shorter than the lifetime of the resonant nuclear state (60 ns) but at low temperatures considerable damage will be frozen in around the neptunium atom [7]. However, around 10-30 K, interstitial migration will occur and eventually interstitials will annihilate at higher temperatures. Thus the probability of forming a neptunium-interstitial complex following the recoil is highest at 4.2 K; their number will decrease as the temperature is increased beyond 30 K.

A number of experiments have now been performed on other metallurgically simple systems where it has been established that impurity-interstitial complexes are formed around 10-30 K, and that they have low vibrational frequency modes [8-11]. If we assume this to be the case for the neptunium-interstitial complex formed due to recoil we can qualitatively explain the variation of $f(T)$. As the temperature is increased above 4.2 K, the number of interstitials formed around a recoil atom decreases because of their ability to migrate and annihilate with nearby vacancies. Thus one obtains an increasingly large number of Np atoms with more nearly perfect environments, such sites giving a larger resonance fraction than the impurity-interstitial complex. Eventually this trend is turned around by the decreasing number of interstitial vacancy pairs available and by the normal decrease in $f(T)$ with temperature. These ideas are very much in agreement with those of Vogl, et al. [8], although one should clearly distinguish the self-recoil damage experiments, of the type reported here, from those in which the damage is produced at some distant place in the material and subsequently is migrated to the Mössbauer atom by isochronal annealing. It should also be noted that some calculations indicate that the annealing may not occur within the time scale of 63 ns established by the nuclear life-time [12].

A number of measurements have been made on self-irradiation damage in lighter actinide metals, primarily Pu and U [6]. Isochronal annealing studies of damage produced changes in resistivity and lattice constant show that the first stage annealing (that arising from interstitial migration and close-pair recombination) is generally completed by 80 K. However, such data does not show the sub-stages generally seen in simpler metals [8], so details of the migration process or the geometric configuration of impurity-defect complex are difficult to obtain. In addition, such experiments have not been done for Am metal. However, measurement of the temperature dependence of the specific heat following self-irradiation in Am at low temperatures shows large oscillations which has been interpreted as being due to the release of stored energy following defect annealing [5]. The first dip does in fact occur at about 80 K, and the authors indicate that some annealing may occur at temperatures as low as 10 K.

In view of the available data on Am metal, it appears likely that the observed temperature dependence of $f(T)$ is determined by either items 2) or 3) above, or by a combination of both. However, even in the absence of a detailed understanding of the phenomenon, it is clear that consideration of such an effect should be of interest both from its intrinsic value as well as from the practicality of carrying out the Np resonance measurements. Thus, it is clear that in instances when measured effects are small, it is a noticeable advantage to maintain the source at 20 K. It is also clear that Am metal, while having many advantages from the standpoint of availability, is not the ideal host since $\theta_D$ is rather low. If a source were obtained with somewhat better phonon characteristics, then the ability to obtain the resonance at higher temperatures even with a warm source would become feasible. Thus, there should now be considerable interest in pursuing the lattice-dynamical aspects of source materials for the $^{237}$Np resonance and the self-recoil damage studies.

References

[3] In the present configuration, the NpO$_2$ absorber is in a magnetically ordered state. However, this is known to cause a line-broadening which is only a perturbation on the already severely broadened line (DUNLAP, B. D.,
Very good fits to the data are obtained with a single Lorentzian.


[13] De Waard, H., Phys. Scr. 11 (1956) 156. We would like to thank G. Vogl for bringing this work to our attention.