CONF-810831--92
UCRL 85437
PREPRINT

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UCRL--85437

DE82 019102

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FROM PROMPT GAMMA DECAYS RESULTING FROM
NEUTRON CAPTURE

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July 20, 1981

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EXACT CALCULATIONS OF NUCLEAR-RECOIL ENERGIES FROM PROMPT γ DECAYS RESULTING FROM NEUTRON CAPTURE

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The results of an accurate determination of the recoil spectrum from (n, γ) reactions in molybdenum are presented in this paper. The recoil spectrum has been calculated from nuclear level structure data and measured branching ratios. Angular correlations between successive gammas have been accounted for using the standard theoretical techniques of Racah algebra and the density matrix formalism.

1. INTRODUCTION

A major objective in radiation damage research is the development of a basis for comparing one kind of irradiation with another. An important quantity obtained from such a basis would be the average number of displaced atoms produced when neutrons of a given energy collide with atoms in the solid.

Current models of defect production in metals are often based upon a modified Kinchin-Pease equation of the form:

$$\langle \nu \rangle = KE/2T_A; \quad \dot{E} \gg 2T_A \quad (1)$$

where $\langle \nu \rangle$ is the average number of Frenkel pairs, E is the damage energy, T_A is the effective displacement energy and K is an efficiency factor which has been found to be energy dependent.^{1,2}

Obtaining the value for K as a function of primary recoil energy has been the goal of many recent experiments.³⁻⁵ Such determinations, however, have required some method of calculating the recoil spectrum for various neutron spectra. Presently, the recoil spectra from collisions with high energy neutrons can be calculated from experimental cross sections and knowledge of reaction kinematics.⁶ For the case of thermal neutron collisions, on the other hand, accurate recoil spectra remain unknown. This makes it difficult to interpret low energy neutron damage data in terms of current models.

In this paper, the results from a more complete calculation of (n, γ) recoils in molybdenum will be discussed. The effects of γ - γ angular correlations will be accounted for, and the effects of the neglect of correlations on the earlier studies will be examined. Finally, a model will be developed to handle the occurrence of long-lived states in γ cascades.

2. PREVIOUS WORK

The first major efforts to calculate (n, γ)

recoil energies were made by Colman, et al.^{7,8} Colman assumed that the angular distribution of gammas resulting from neutron capture was isotropic. This allowed him to compute the average recoil energy of a nucleus emitting two or more gammas by simply summing the separate recoil energies arising from each gamma. This gave the average recoil energy as:

$$\langle E \rangle = (533/M) \sum_i P(E_i) E_i \quad (2)$$

where E_i is the gamma-ray energy, $P(E_i)$ is the intensity of gammas with energy E_i , M is the mass of the nucleus in a.m.u. and 533 is a factor which contains the necessary conversions so that $\langle E \rangle$ is in eV when E_i is in MeV.

Using the gamma-ray spectra tabulated by Groshev, et al.,⁹ Colman calculated the mean recoil energies for a number of metals. A few of these recoil energies are listed in Table I, along with values calculated by this author using more recent data on line and continuum gamma-ray yields. Because of the proximity of these mean energies to the displacement threshold, errors introduced by neglecting the angular correlations and level lifetimes could be critical in estimating radiation damage from thermal neutrons.

Aside from possible errors introduced by the neglect of angular correlations, a major shortcoming is inherent in all of these calculations; namely, that no information about the distribution of recoil energies about the mean can be

Element	$\langle E \rangle^7$	$\langle E \rangle^9$
Ag	136 eV	125 eV
Al	771 eV	801 eV
Cu	374 eV	425 eV
Mo	160 eV	189 eV
W	58 eV	61 eV

Table I: Average recoil energies calculated by Colman (ref. 6 and 7) and by this author using data from reference 9.

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obtained with this method. This is a significant problem for two reasons. First, because of the proximity of the mean recoil energy to the displacement threshold, a large spread in the distribution of recoil energies could greatly affect the measurements of K based upon the mean. Second, K changes rapidly with energy in this region of recoil energies, making knowledge of the distribution important in estimating defect production from thermal neutrons.

3. CALCULATIONAL MODEL

3.1 Background

In principle, it should be possible to calculate an exact recoil distribution from knowledge of all nuclear energy levels and their corresponding spins and parities. The momentum transferred to the nucleus in a thermal neutron capture event would be the sum of the momenta from the individual gammas in the cascade (neglecting the insignificant momentum transferred by the .025 eV neutron). Unfortunately, the high neutron binding energies in most metals lead to level densities near the capture state which cannot be fully resolved experimentally. Thus, only a small fraction (~15% in ^{95}Mo) of the primary gammas can be assigned to a level diagram.

This paper will consider the recoil spectrum of ^{95}Mo from neutron capture in ^{95}Mo using the information obtained from the nuclear data available on this isotope. Future work will be based upon a statistical nuclear model, which will aid in estimating how the results of this study should be modified to account for the unresolved gamma transitions.

3.2 Reaction Kinematics

The recoil energy of a nucleus after the emission of one or more gammas is given by:

$$E_R = |P|^2 / 2M \quad (3)$$

where $|P|$ is the magnitude of the momentum transferred to the nucleus from the gammas and M is the mass of the nucleus. From momentum conservation, P is given by

$$P = P_1 + P_2 + \dots + P_n$$

where P_n are the momenta of the individual gammas.

For the case of two gammas, $|P|^2$ has a simple form:

$$|P|^2 = |P_1|^2 + |P_2|^2 + 2|P_1||P_2|\cos\theta_{12} \quad (4)$$

where θ_{12} is the angle between the two gammas running from 0 to π . For the case of three gammas, the problem is no longer planar, and the azimuthal angle of the third gamma around the second must be accounted for. This gives the more complicated expression:

$$|P|^2 = |P_1|^2 \sin^2 \theta_{13} \cos^2 \phi_{123} + |P_2|^2 \sin^2 \theta_{13} \sin^2 \phi_{123} + |P_3|^2 \cos^2 \theta_{13} + 2|P_1||P_2|\cos\theta_{12} + 2|P_1||P_3|\cos\theta_{13}\cos\phi_{123} + 2|P_2||P_3|\cos\theta_{23}\sin\phi_{123}$$

$$+ |P_2|^2 \sin^2 \theta_{13} + |P_3|^2 \cos^2 \theta_{13} \sin^2 \phi_{123} + |P_1|^2 \sin^2 \theta_{23} \sin^2 \phi_{123} + |P_1|^2 \cos^2 \theta_{23} \cos^2 \phi_{123} + |P_1||P_2|\cos\theta_{12} + |P_1||P_3|\cos\theta_{13}\cos\phi_{123} + |P_2||P_3|\cos\theta_{23}\sin\phi_{123}$$

where θ_{12} is the angle between the second and third gamma and ϕ_{123} ($=\pi/2 - \theta_{12}$) is the rotation of the third gamma around the second.

Expressions for the cases when four or more gammas are emitted can also be obtained, though it is not necessary to express them here. What is important, though, is the realization that if the gammas are emitted isotropically, then the angular terms in Equations 4 and 5 vanish when averaged are calculated. The average recoil energies from Equations 4 and 5 then reduce to Coleman's expression (Equation 2).

Figure 1 shows a few of the nuclear levels for ^{95}Mo populated by thermal neutron capture. On the left side are the values of spin and parity assigned to the levels. On the right side are the level energies and measured lifetimes. When no value for the lifetime is given, the levels are assumed to have lifetimes of much less than 10^{-14} seconds.

The arrows represent observed gamma transitions. The numbers associated with these transitions are the intensities based upon 100 thermal neutron captures. It can be observed that for this isotope, only 14% of the total number of primary gammas have been assigned to this diagram.

3.3 Level Lifetimes

The probability for a transition from one nuclear energy level to another via the emission of a gamma-ray is given by

$$W_{fi} = (2\pi/M)\rho_f | \langle \psi_f | H' | \psi_i \rangle |^2$$

where W_{fi} is the transition probability per unit time, ρ_f is the density of final states, ψ_f and ψ_i are the wave functions describing the final and initial states and H' is the interaction Hamiltonian. This implies that the transition probability varies roughly as

$$W_{fi} \propto E^2 \epsilon^2$$

where E is the energy of the transition and ϵ is the quantum of angular momentum carried off by the gamma-ray.

The level lifetime is inversely related to the transition probability. This means that if any levels have measurable lifetimes, they most likely will be found in the first few excited states, where ρ_f and E are both small. Measured lifetimes in ^{95}Mo are given on the right side of figure 1.

Computer calculations of 600 eV displacement cascades^{1,2} have indicated that the cascade is completed within 5×10^{-11} seconds. Therefore, decays from levels with lifetimes greater than

this should be treated as separate events. However, since the maximum energy transferred to a ^{90}Mo nucleus during a transition from a long-lived state is much less than the displacement energy, it was decided not to include gamma transitions from these states in the recoil energy calculations.

3.4 Angular Correlations

The possible effects of gamma-ray angular correlation on the calculations of mean recoil energies have concerned many workers in radiation damage.^{7,14} It has been known for many years that gamma-ray emission is not isotropic.¹⁵ This is due to the tensor nature of the electromagnetic interaction Hamiltonian.

It is possible to derive the angular distribution function for two gammas as:⁶

$$W(B) = \sum_{LL' = \pm 1, \pm 2} B_K(J_1) R_K(LL' J_1 J_2) \times P_K(\cos \theta) \frac{J_1 \cdot J_2}{E E_L}$$

where α is the angle between the gammas, S_L and S_L' are the mixing ratios, and B_K and A_K are constants constructed of Clebsch-Gordan and Racah coefficients. The $P_K(\cos \alpha)$ terms in the above equation are the Legendre polynomials.

Symmetry requirements lead to non-zero values only for even terms in k . Therefore, we can write $W(\theta)$ in the more condensed form:

Figure 1 : Levels populated by primary gamma transitions in ^{16}Mo .
Energies are in MeV.

$$W(\theta) = \frac{1}{K} a_{2K} P_{2K}(\cos \theta) \quad (6)$$

where the above equation is normalized so that $a_0 = 1$. The important conclusion drawn from inspection of this equation is that the correlation function is symmetric about $\theta = 90^\circ$. Therefore, the average recoil energy as calculated using Equation 2 will not be affected by the angular correlation of the emitted gammas. The distribution of recoils about the mean could be greatly affected by gamma-ray angular correlation, however.

4. RESULTS OF RECOIL CALCULATIONS

Because of the statistical nature of gamma-ray transitions, it was decided that a Monte Carlo calculation would be the best method for determining the recoil spectra. The gamma cascades were chosen with a weighting determined from the measured branching ratios. The angular correlation function was determined from measurements when available and from Equation 6 when not. The mixing ratios in Equation 6 were estimated from nuclear systematics.

Figure 2 shows the probability distribution function of recoils calculated assuming isotropic gamma-ray emission and the absence of long-lived states. The average recoil energy, $\langle E \rangle$, calculated in this manner is $258 \pm .5$ eV. This value is much greater than that calculated from Colman's procedure where $\langle E \rangle = 169$ eV is obtained. The reason for this difference will be discussed in the conclusion. It is important to note, however, that the recoil energies are evenly distributed around the mean.

Figure 3 shows the effects of including both the level lifetimes and the angular correlations in the calculations. The solid lines show the effects of including level lifetimes, and the dashed lines show the effects of including both lifetimes and angular correlations. As expected, this has the effect of eliminating both the very low and the very high energy recoils. It also significantly skews the distribution to the high energy side, although the average recoil energy is lowered somewhat ($\langle E \rangle = 255 \pm 2$ eV).

The more important information is the integral damage energy, which is shown in Figure 4 for all three cases. In this Figure, the solid line represents the integral damage energy calculated assuming no angular correlations or level lifetimes. The dashed line represents the integral damage energy when these assumptions are removed. As can be seen, accounting for level lifetimes has the greatest effect on the recoil distribution function.

5. CONCLUSION

The exclusion of transitions having lifetimes greater than 5×10^{-13} seconds has the greatest effect on the shape of the recoil distribution function. The effect is due to the removal

Figure 2: Recoil distribution for ^{95}Mo (n, γ) ^{95}Mo calculated by neglecting angular correlation and possible long-lived states.

Figure 3: Recoil distribution calculated assuming long-lived states are present (solid line) and both long-lived states and angular correlations (dashed line).

of the last one or two MeV from the total energy of the cascade, which both reduces the fraction of high and low energy recoils and skews the distribution to higher energy.

Gamma-ray angular correlation has no effect on the mean recoil energies calculated using Colman's method (equation 2). Instead, angular correlation changes the distribution of recoil energies about the mean. The results of these calculations indicate that this redistribution is very small.

The average recoil energies obtained from the present calculations are much greater (~50%) than those obtained by Colman. Theory, however, indicates that the results of average recoil energy calculations using Colman's procedure should not differ from the averages calculated using nuclear models. The discrepancy between the different values can

Figure 4 : Integral damage energy calculated for the three cases mentioned in the text.

be explained by including gamma transitions from the unresolved levels (those levels between 3.5 and 9.1 MeV in ^{235}Pu). This can be demonstrated as follows.

The nuclear data (figure 1) for ^{235}Pu shows no primary cascades from the capture state to levels above 3.5 MeV. Transitions within this region must occur, however, in order to account for the large number of unassigned gammas. If two 3 MeV gammas are emitted during a transition to the lower energy states, an average of ~100 eV will be transferred to the nuclear recoil. If, however, only one 6 MeV gamma is emitted during a similar transition, an average of ~200 eV will be transferred. Therefore, multiple cascades within the unresolved levels will greatly reduce the average recoil energy.

Future work will use statistical nuclear models to evaluate the effects of unresolved transitions on the nuclear recoil energy distribution resulting from neutron captures. Only when this is done will it be possible to make quantitative predictions about thermal neutron damage in metals.

REFERENCES

- [1] Guinan, M. W. and Violet, C. E., in Symposium on Neutron Cross-Sections from 10 to 40 MeV, BNL-NCS-50681 (U.S. ERDA, BNL, 1977), 361.
- [2] Maury, F., Biget, M., Vajda, P., Lucasson, A. and Lucasson, P., Rad. Effects 38 (1978) p. 53.
- [3] Averback, R. S., Benedek, R. and Merkle, K. L., in International Conference on the Properties of Atomic Defects in Metals (Argonne, Illinois, 1976) p. 785.
- [4] Kirk, M. A. and Greenwood, L. R., J. Nuc. Mat. 80 (1979) p. 159.
- [5] Coltman, R. R., Klabunde, E. E. and Williams, J. M., Rates of Defect Production by Fission Neutrons in Metals at 4.7 K, to be published in J. Nuc. Mat.
- [6] Coltman, et al., J. of Applied Physics 33 (1962) p. 3509.
- [7] Coltman, R. R., Klabunde, C. E. and Redman, J. K., Phys. Rev. 156 (1967) p. 715.
- [8] Groshev, L. B., Demidov, A. M., Lutsenko, V. N. and Pelekhov, V. I., Atlas: Gamma-Ray Spectra from the Capture of Thermal Neutrons (The Chief Bureau of the Use of Atomic Energy, Council of Ministers, USSR, 1959).
- [9] Orphan, V. J., Rasmussen, N. G. and Harper, T. L., Line and Continuum Gamma-Ray Yields from Thermal-Neutron Capture in 75 Elements (Gulf General Atomic GA-10248, 1970).
- [10] Heck, D., et al, Nuclear Phys. A159 (1970) p. 49.
- [11] Barrette, J., et. al, Nuclear Phys. A172 (1971) p. 41.
- [12] Paradis, P., et al., Phys. Rev. C 14 (1976) p. 835.
- [13] Guinan, M. W. and Kinney, J. K., Molecular Dynamic Calculation of Energetic Displacement Cascades, to be published in this conference proceedings.
- [14] Robinson, M. T., in Robinson, M. T. and Young, F. W. (eds.), Fundamental Aspects of Radiation Damage in Metals, Conf-751006-P1 (U.S. ERDA, Gatlinburg, TN, 1975), p. 1.
- [15] Brady, E. L. and Deutsch, M., Phys. Rev. 78 (1950) p. 558.
- [16] Gill, R. D. Gamma-Ray Angular Correlations (Academic Press, London, 1975).

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