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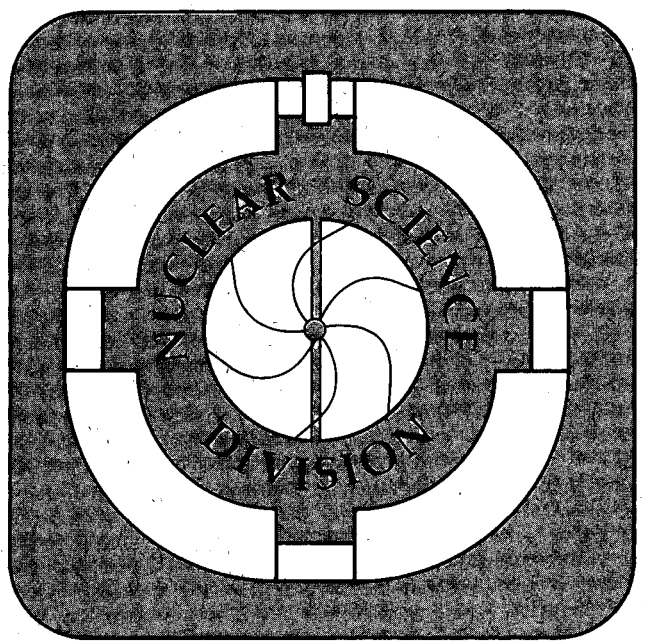

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^{40}K: Young or Old, the Decay Rate is the Same

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Over the years, many authors have pointed out that the exponential nature of the radioactive decay law is only an approximation.[1-17] The work of Khalfin and others on the time evolution of unstable quantum states showed that the decay rate should approach zero as $t \rightarrow 0$. Thus, there must exist a region between $t = 0$ and the known exponential domain where the decay rate is non-exponential. The existence of such a non-exponential regime was suggested as a possible explanation for the null results of proton-decay experiments and as a possible influence on the rates for double-beta decay transitions.[18-23] Obviously, if nuclear decay rates varied with time then the usefulness of radioactive dating techniques would be severely compromised. In spite of the considerable theoretical effort on this subject, we are aware of only one previous experimental search for non-exponential behavior at short times.[24] In the present work, we compared the decay rate of freshly prepared ^{40}K with that of ^{40}K that is $\geq 4.5 \times 10^9$ years old. We find that to within the experimental uncertainty of ±11%, the decay rates are the same. This indicates that the exponential nature of the decay law is valid at least down to time-scales on the order of $10^{-10}\ t_{1/2}$.

In 1988, Gopych and Zalyubovskii[25] suggested that an interesting test of the nature of the decay law at short times would be to measure the lifetime of freshly prepared ^{40}K. The decay scheme of ^{40}K is shown in Figure 1 (Ref. 26). Due its very long half life of 1.28×10^9 years, it should be possible to probe the nature of the decay law at times on the order of $10^{-10}\ t_{1/2}$. Since the ^{40}K naturally occurring on Earth is at least 4.5×10^9 years old, it is therefore comfortably in the known exponential decay regime. We decided to perform such a test by comparing the decay rates of "young" and "old" samples of ^{40}K. In order to do this, we made use of the fact that the natural isotopic composition of ^{39}K is 93.26%, while that of ^{40}K is only 0.0117% (Ref. 26). Thus it is possible by neutron irradiating a sample of potassium to produce a substantial amount of "young" ^{40}K via the $^{39}K(n,\gamma)$ reaction ($\sigma = 2.1$ barns) [Ref. 26]. If one then measures the 1461-keV γ-ray

emission rates and the ^{40}K isotopic abundances in the irradiated and unirradiated samples, one can compare the decay rates of "young" and "old" ^{40}K.

The emission rate of 1461-keV γ rays, R, from an unirradiated (u) naturally occurring potassium sample is

$$R_u = {}^oN_u \, \lambda^o \, B \qquad (1),$$

while that from a neutron-irradiated (i) potassium sample is

$$R_i = ({}^oN_i \, \lambda^o + {}^yN_i \, \lambda^y) B \qquad (2),$$

where ${}^oN$ is the number of "old" ^{40}K nuclei naturally occurring in the sample, ${}^yN$ is the number of "young" ^{40}K nuclei produced by the neutron irradiation, λ^o is the decay constant of "old" ^{40}K, λ^y is the decay constant of "young" ^{40}K, and B is the branching ratio for the 1461-keV gamma ray. Thus, the ratio of the decay constants for "young" and "old" ^{40}K is

$$\lambda^y/\lambda^o = (R_i / R_u - {}^oN_i / {}^oN_u) \, {}^oN_u / {}^yN_i \qquad (3).$$

The number of ^{40}K nuclei, N, present in a sample of a compound containing 1 potassium atom per molecule is

$$N = (m/MW) \, N_A \, [^{40}K] \qquad (4),$$

where, m is the mass of the sample, MW is the molecular weight of the compound, N_A is Avagadro's number, and $[^{40}K]$ is the isotopic abundance of ^{40}K in the sample. Thus, in terms of the experimentally measurable quantities,

$$\lambda^y/\lambda^o = (R_i / R_u - m_i / m_u) (m_u / m_i) / ([^{40}K_i] / [^{40}K_u] - 1) \qquad (5).$$

Note that in deriving these equations, we have ignored the small loss of ^{40}K in the irradiated sample caused by the $^{40}K(n,\gamma)^{41}K$ reaction. As is discussed below, under the conditions of the present experiment this effect is negligible.

To compare the decay rates of "young" and "old" ^{40}K, a 3.1-gram sample of 99.999% pure K_2CO_3 (obtained from Noah Technologies) was irradiated for one week in a flux of approximately 8×10^{13} thermal neutrons/cm^2-sec at the University of Missouri Research Reactor Facility. After allowing short-lived activities to decay away for three days, the sample was returned to Lawrence Berkeley Laboratory for analysis. Initial γ-ray counting revealed the presence of a number of radio-impurities that would interfere with the measurement of the 1461-keV γ-ray emission rate. As a result, we decided to chemically purify the potassium by first dissolving it in H_2O, then adding 6M HCl plus a small amount of concentrated HNO_3, and then loading it onto a column containing AG1-X8 anion exchange resin that had been previously treated with 10M HCl. The column was washed with additional 6M HCl and seven fractions were collected. γ-ray counting of each fraction showed that ^{42}K ($t_{1/2}$ = 12.4 hours), produced by the $^{41}K(n,\gamma)$ reaction, was present essentially in only the third and fourth fractions. These two fractions were then boiled to dryness and produced a 2.2916-gram sample of KCl which was used as the "irradiated" sample in the subsequent γ-ray and mass spectrometer analyses. A similar amount of unirradiated K_2CO_3 was converted into KCl by dissolving it in H_2O, adding concentrated HCl, and then boiling it to dryness. This produced an "unirradiated" KCl sample of 2.5332 grams that was analyzed in the same manner as the "irradiated" sample. Each KCl sample was placed inside a small glass vial for counting.

Gamma-ray counting was performed with a 100-cm^3 high-purity germanium detector. In order to stop the intense β's from the decay of ^{86}Rb present in the "irradiated" sample, a 1.27-cm thick block of plastic was placed between the sample position and the detector. 10 cm of lead was placed all around the sample and detector to reduce room background. Data were then accumulated from the "irradiated" sample, from the "unirradiated" sample, and from an empty glass vial. Four separate spectra, totaling 105.6 hours of counting, were collected from the

"irradiated" sample. Three spectra from the "unirradiated" sample, totaling 102.1 hours of counting, and three background spectra, totaling 70.2 hours, were also collected. The ^{40}K γ-ray counting rates observed in all of the sample runs and in all of the background runs were constant within their respective statistical accuracies. All of the measurements of the "irradiated" sample were made within one month following the irradiation. To obtain the net 1461-keV γ-ray emission rates from the two KCl samples, the total background spectrum was appropriately normalized and subtracted from the spectrum obtained from each KCl sample.

Figure 2 illustrates the relevant portions of the background-subtracted spectra observed from the two samples. The 1461-keV line from the decay of ^{40}K is clearly seen in both spectra. However, despite our efforts to purify the "irradiated" potassium, the line at 1408 keV indicates the presence of ^{152}Eu in this sample. This isotope must have been produced by the $^{151}Eu(n,\gamma)$ reaction on a small europium impurity in the original K_2CO_3 material. In addition to the strong line at 1408 keV, the decay of ^{152}Eu also produces a line at 1458 keV that can been as a small shoulder on the low-energy side of the 1461-keV peak in the spectrum from the "irradiated" sample. In order to obtain the net ^{40}K 1461-keV peak area, we took the measured area of the 1408-keV peak, scaled it by the ratio of the well-known intensities of these ^{152}Eu γ-ray lines[27] and the relative detector efficiencies at 1408 and 1458 keV, and subtracted the result from the area of the "1461-keV" doublet. The resulting observed ^{40}K 1461-keV counting rates (per 10^5 seconds) from the "irradiated" sample, the "unirradiated" sample, and the room background were 4949 ± 80, 3975 ± 42, and 1808 ± 42, respectively.

Thermal ionization mass spectrometry measurements were performed on both the "irradiated" and "unirradiated" potassium samples. Each of the two KCl samples was split into two aliquots and dissolved in 5N HNO_3. Each of the four resulting solutions was analyzed in triplicate on a VG Sector 54 single collector mass spectrometer. Examples of the measured mass spectra are shown in Figure 3. To assure reasonable counting statistics on $^{40}K^+$, a relatively large $^{39}K^+$ ion beam of 10^{-10} amp was used. Data were collected by magnetic field switching with 5 seconds of beam integration at each peak and background setting. For each analysis, 100 ratios of

. $[^{39}K]/[^{40}K]$ and $[^{41}K]/[^{40}K]$ were measured. In our experiment, the neutron irradiation converted less than 1 part in 10^4 of the existing ^{39}K into ^{40}K and less than 0.4% of the "old" ^{40}K into ^{41}K. Thus, these two abundance ratios provide a measure of the ^{40}K isotopic abundances in the "irradiated" and "unirradiated" samples. The measured abundance ratios were corrected for instrumental discrimination by normalizing the $[^{39}K]/[^{40}K]$ ratio to a value $[^{41}K]/[^{39}K]$ = 0.072168. Because of the small size of the $^{40}K^+$ beam in comparison to that of $^{39}K^+$, in each magnetic field scan of the three isotopes, three succesive measurements of $^{40}K^+$ were made to determine the effects of residual amplifier current from the $^{39}K^+$ measurement. Only the data from the third $^{40}K^+$ measurement, where the effects of residual current are less than 0.5% of the ^{40}K abundance, were used in the subsequent analysis. From this data, the isotopic abundance ratios of $[^{39}K]/[^{40}K]$ for the "irradiated" and "unirradiated" potassium samples were determined to be 5029±19 and 8063±23, respectively, or $[^{40}K_i]/[^{40}K_u]$= 1.603±0.008.

Inserting the measured values of the sample masses, the ^{40}K 1461-keV gamma-ray counting rates, and the ^{40}K isotopic abundances into equation (5) yields our result for

$$\lambda^y/\lambda^o = 0.999 \pm 0.106 \qquad (6).$$

We find that, to within the accuracy of our measurement, the decay rates of "young" and "old" ^{40}K are the same. This indicates that the exponential nature of the radioactive decay law is valid at least down to time scales on the order of $10^{-10}\ t_{1/2}$. Thus, the time domain currently being explored in studies of two-neutrino double-beta decay[28,29] has now been experimentally verified to be within the exponential regime. The relevant interval for proton decay, however, remains to be tested. From our results, we can set a limit on a possible linear variation of the ^{40}K decay rate with "age" as being $< 2.5\times10^{-9}$ %/year. The basis upon which the $^{40}K/^{40}Ar$ dating scheme rests is, therefore, secure. The present work represents the most stringent test of the exponential decay law at early times performed to date.

References

1. Khalfin, L. A. *Zh. Eksp. Teor. Fiz.* **33**, 1371-1382 (1957) [*Sov. Phys. JETP* **6**, 1053-1063 (1958)].
2. Matthews, P. T. & Salam, A. *Phys. Rev.* **115**, 1079-1084 (1959).
3. Winter, R. G. *Phys. Rev.* **123**, 1503-1507 (1961).
4. Goldberger M. L. & Watson, K. M. *Phys. Rev.* **136**, B1472-1480 (1964).
5. Khalfin, L. A. *Pis'ma Zh. Eksp. Teor. Fiz.* **8**, 106-109 (1968) [*JETP Lett.* **8**, 65-68 (1968)].
6. Ekstein, H. & Siegert, A. J. F. *Ann. Phys. (N.Y.)* **68**, 509-520 (1971).
7. Shirokov, M. I. *Yad. Fiz.* **21**, 674-687 (1975) [*Sov. J. Nucl. Phys.* **21**, 347-353 (1975)].
8. Knight, P. L. *Phys. Lett.* **56A**, 11-13 (1976).
9. Dolejsi, J. & Exner, P. *Czech. J. Phys. B* **27**, 855-864 (1977).
10. Chiu, C. B., Sudarshan, E. C. G. & Misra, B. *Phys. Rev. D* **16**, 520-529 (1977).
11. Arthur, M. D. *et al. Nuovo Cimento* **63B**, 565-587 (1981).
12. Margvelashvili, P. I. & Baker, A. *Phys. Lett.* **131B**, 390-392 (1983).
13. Robinson, E. J. *Phys. Rev. Lett.* **52**, 2309-2312 (1984).
14. Mittleman, M. H. & Tip, A. *J. Phys. B* **17**, 571-576 (1984).
15. Robinson, E. J. *Phys. Rev. A* **33**, 1461-1463 (1986).
16. Robinson, E. J. *Phys. Rev. Lett.* **57**, 1281-1283 (1986).
17. Greenland, P. T. *Nature* **335**, 298 (1988).
18. Khalfin, L. A. *Phys. Lett.* **112B**, 223-226 (1982).
19. Chiu, C. B., Misra, B. & Sudarshan, E. C. G. *Phys. Lett.* **117B**, 34-40 (1982).
20. Fleming, G. N. *Phys. Lett.* **125B**, 287-290 (1983).
21. Lane, A. M. *Phys. Lett.* **99A**, 359-360 (1983).
22. Miglietta, F. & Rimini, A. *Phys. Lett.* **139B**, 353-356 (1984).
23. Grotz, K. & Klapdor, H. V. *Phys. Rev. C* **30**, 2098-2100 (1984).
24. Norman, E. B., Gazes, S. B., Crane, S. G. & Bennett, D. A. *Phys. Rev. Lett.* **60**, 2246-2249(1988).

25. Gopych, P. M. & Zalyubovskii, I. I. *Sov. J. Part. Nucl.* **19**, 338-357 (1988).

26. Lederer, C. M. & Shirley, V. S. *Table of Isotopes* (Wiley, New York, 1978), 7th ed.

27. Browne, E. & Firestone, R. B. *Table of Radioactive Isotopes* (Wiley, New York, 1986).

28. Elliott, S. R., Hahn, A. A. & Moe, M. K. *Phys. Rev. Lett.* **59**, 2020-2023 (1987).

29. Ejiri, H. *et. al. Phys. Lett.* **258B**, 17-23 (1991).

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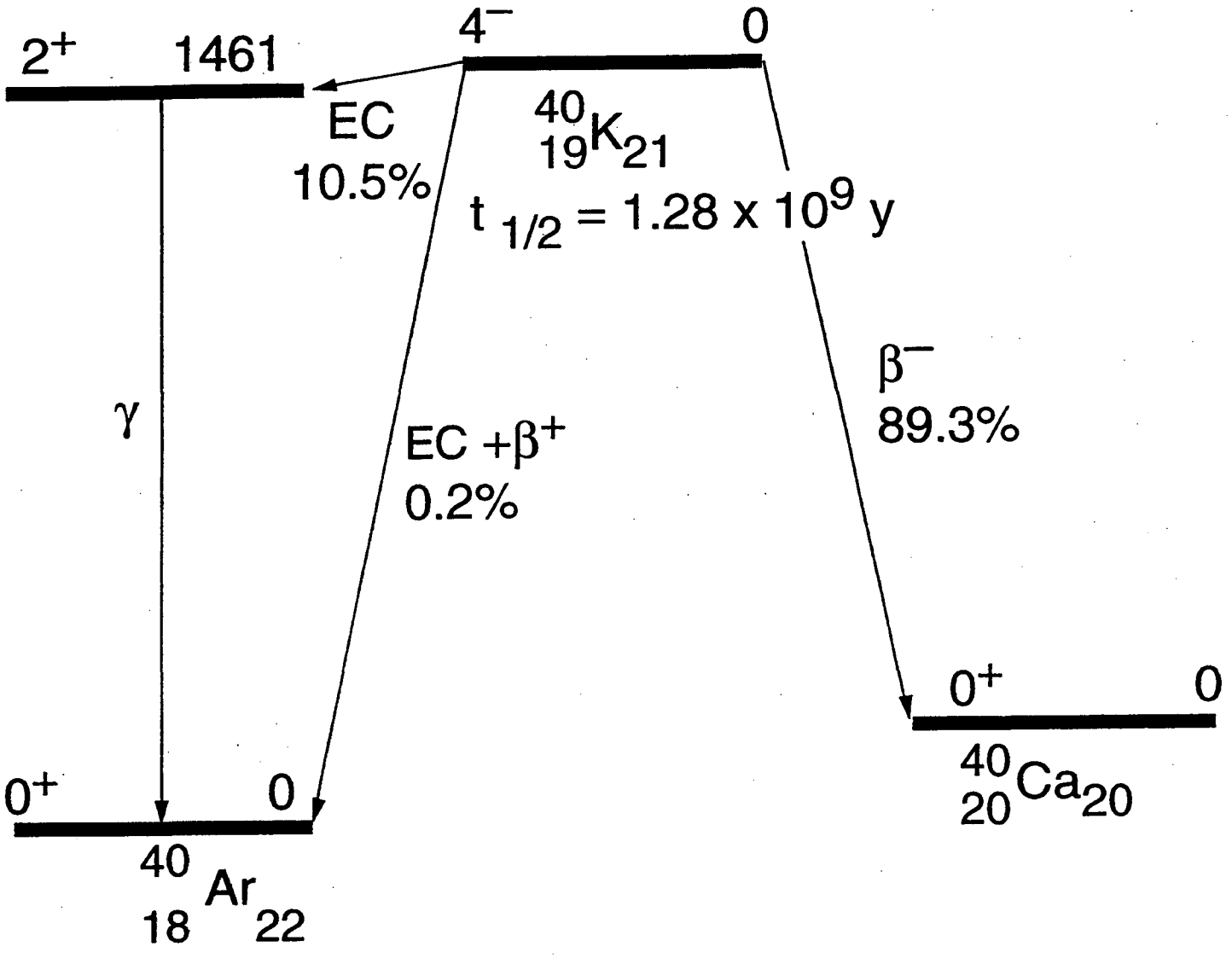
Figure Captions

Fig. 1. Decay scheme of ^{40}K. Energies are given in keV.

Fig. 2. Background-corrected gamma-ray spectra observed from the irradiated and unirradiated KCl samples. These spectra have been corrected for the different sample masses and counting times. Energies are given in keV.

Fig. 3. Mass spectra observed from (a) irradiated and (b) unirradiated samples of KCl. The vertical scales are the same in parts (a) and (b). Note, however, that in order to put the $^{39,40,41}K$ peaks all on the same plot, the vertical scale was changed in between mass 39 and 40, and again between mass 40 and 41.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1461
4−
0
EC
10.5%
40
19K21
t 1/2 = 1.28 x 10^9 y
γ
EC +β+
0.2%
β−
89.3%
0+
0
40
20Ca20
0+
0
40
18 Ar22

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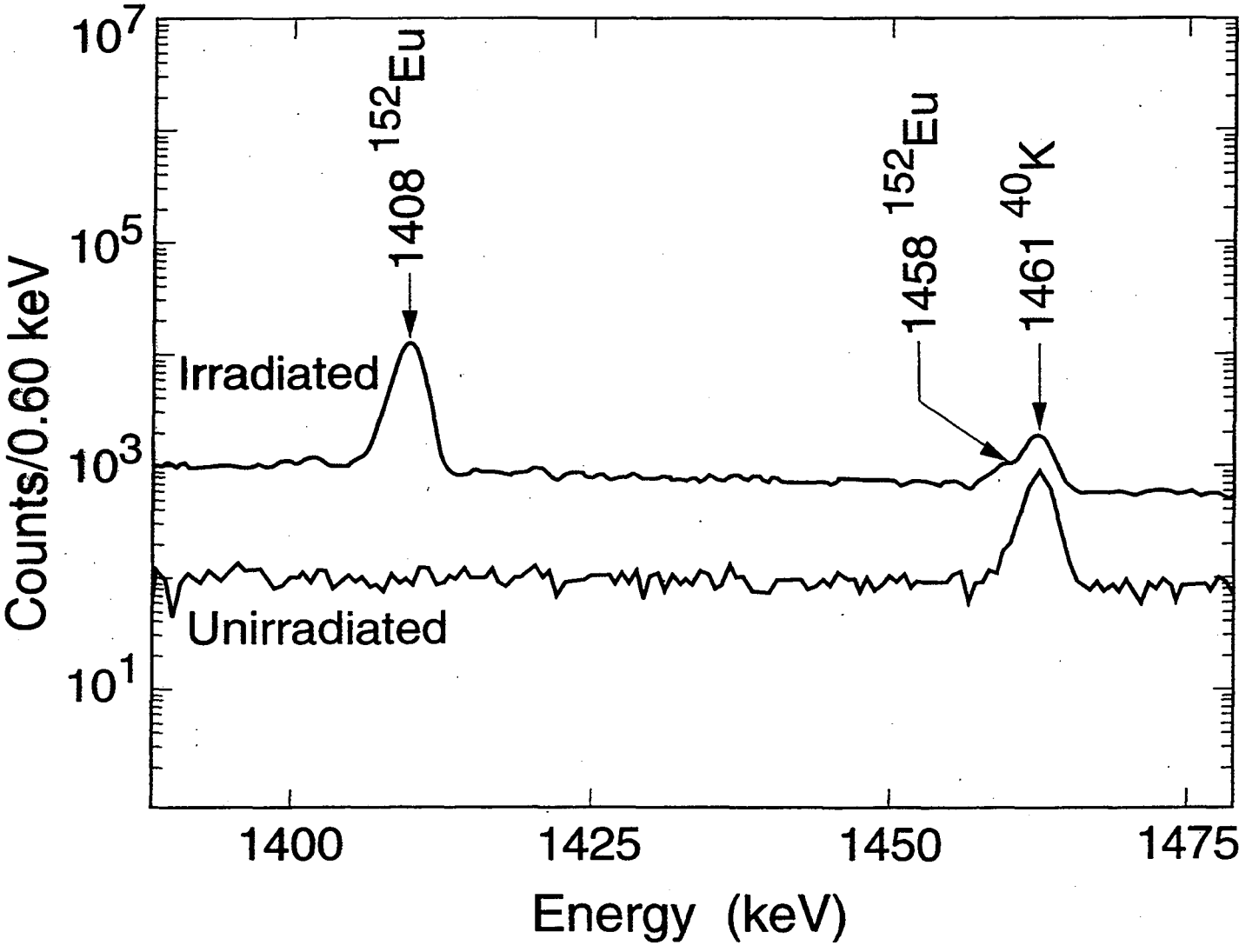
10^7
10^5
10^3
10^1
Counts/0.60 keV
^{152}Eu
1408
^{152}Eu
1458
^{40}K
1461
Irradiated
Unirradiated
1400
1425
1450
1475
Energy (keV)

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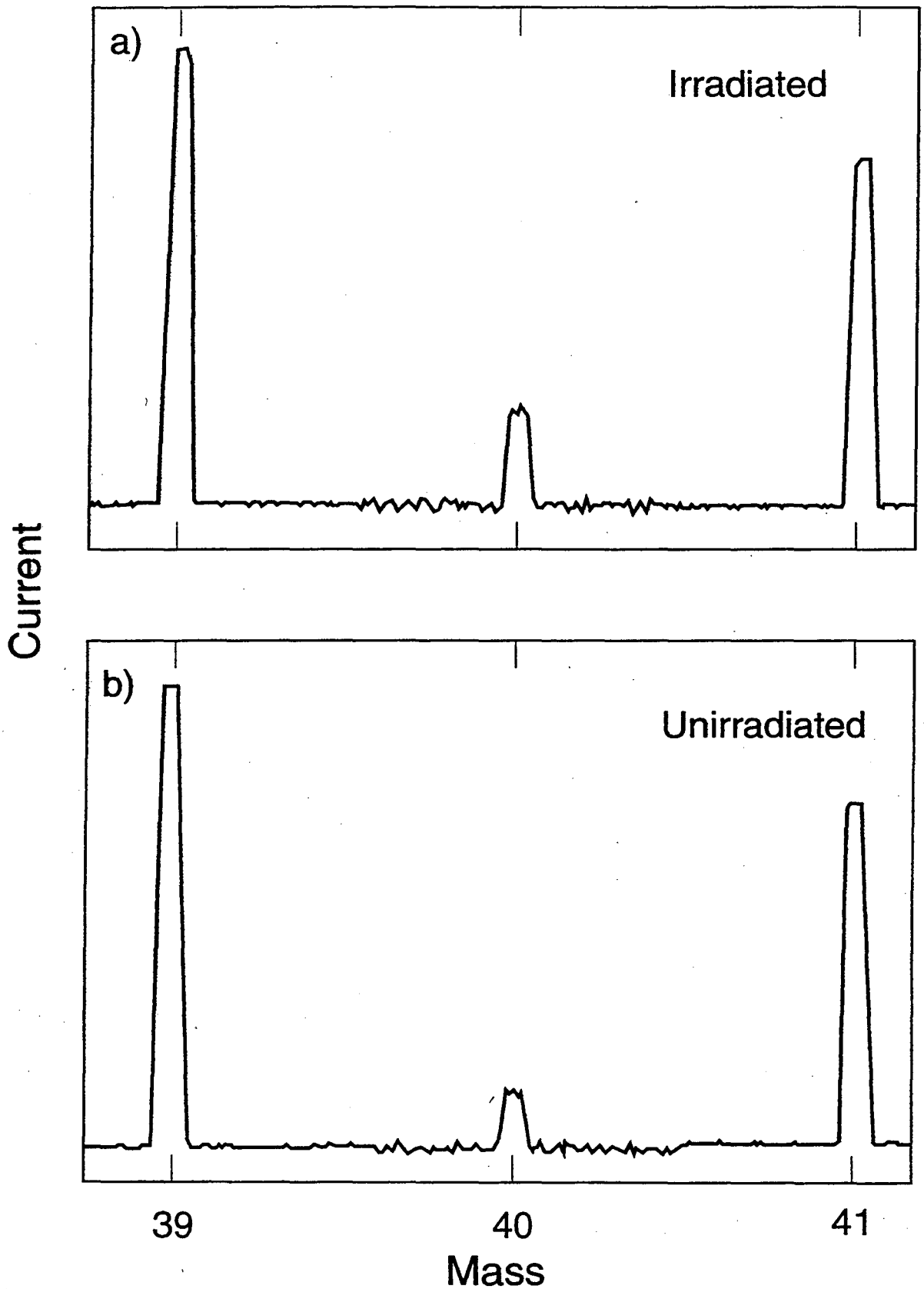

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