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Author
Norman, E.B.

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Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

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E.B. Norman

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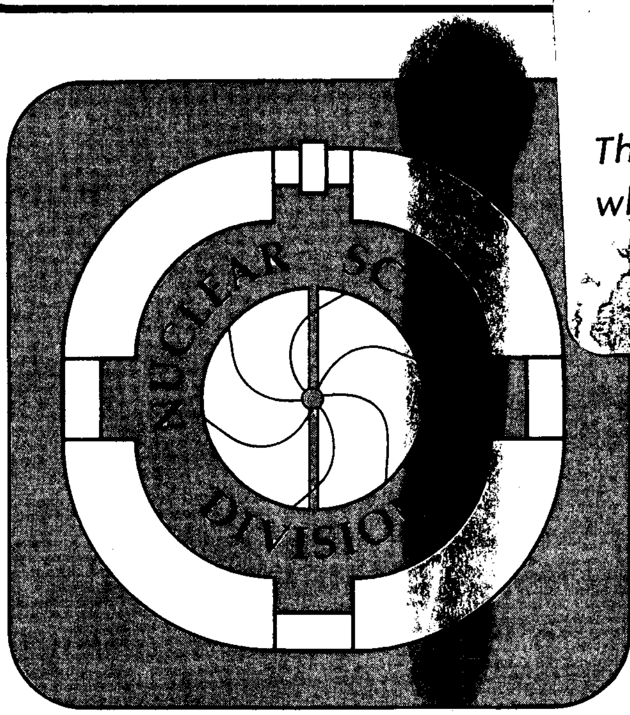

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Improved Limits on Double Beta Decay Half-Lives of ^{50}Cr, ^{64}Zn, ^{92}Mo, and ^{96}Ru*

Eric B. Norman

Nuclear Science Division

Lawrence Berkeley Laboratory

University of California

Berkeley, CA 94720

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Improved Limits on Double Beta Decay Half-Lives of ^{50}Cr, ^{64}Zn, ^{92}Mo, and ^{96}Ru

Eric B. Norman
Nuclear Science Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

Abstract

Searches have been made for the β^+/electron-capture type double beta decays of ^{50}Cr, ^{64}Zn, ^{92}Mo, and ^{96}Ru and for the double β^+ decay of ^{96}Ru . The sought-for signature of such decays was the emission of coincident back-to-back 511-keV annihilation gamma rays from the sample under study. No positive evidence of any of these decays was observed. However, from these experiments, improved lower limits on the half-lives of these nuclei against such decays have been established.

While there has been much experimental effort devoted to searches for double β^- decay, comparatively little work has been done on double β^+, β^+/electron-capture, and double electron-capture decay[1-4]. To date, there are no conclusive laboratory observations of any double beta decay mode. However, because of the possible implications for lepton-number conservation and for the mass of the neutrino, it is important to search for evidence of these processes with the greatest sensitivity currently possible. In a previous study[5], searches were made for the β^+/EC and double EC decays of ^{58}Ni and ^{106}Cd and for the $\beta^+\beta^+$ decay of ^{106}Cd. In the present work, a similar technique has been employed to search for the β^+/EC decays of ^{50}Cr, ^{64}Zn, ^{92}Mo, and ^{96}Ru, and for the $\beta^+\beta^+$ decay of ^{96}Ru. The possible decay schemes of these nuclei are shown in Fig. 1. All energies, spins, and parities used in the present work are taken from Ref. 6. For each of these nuclei, a ground state $\longrightarrow$ ground state β^+/EC decay is possible. For ^{96}Ru, β^+/EC decays to excited states below 1.698-MeV excitation energy in ^{96}Mo are also energetically allowed, as is a ground state $\longrightarrow$ ground state $\beta^+\beta^+$ decay.

If a β^+/EC or a $\beta^+\beta^+$ decay occurred within a thick sample of material, almost all of the positrons would stop and annihilate within the sample. Subsequently either two or four strongly correlated coincident 511-keV annihilation gamma rays would be emitted. The comparatively low-energy x rays that would also be emitted in the process of β^+/EC decay would be severely attenuated in the sample. To detect the gamma-ray signatures of β^+/EC and/or $\beta^+\beta^+$ decay, two 110 cm^3 high-purity Ge detectors were used. The sample under study was sandwiched directly between the front faces of the two detectors. This assembly was shielded by approximately 10 cm of lead on all sides. The experimental apparatus was located in a counting room of the 88-inch Cyclotron building at Lawrence Berkeley Laboratory. An event of possible interest was defined to be one for which there were coincident signals in the two detectors and in which 511 keV was deposited in one of them. For such events the energy signals of the two detectors were summed together. A signature of ground state ⟶ ground state β^+/EC decay would thus be a summed-energy line at 1022 keV, and the signal from β^+/EC decay to a state at an excitation energy E_x in the daughter nucleus would be a line at (1022 + E_x) keV. Data were accumulated in 2048 channels using a multichannel analyzer and were recorded on magnetic tape for subsequent off-line analysis.

All samples studied were natural isotopic composition[6] metals of at least 99% purity obtained from Alpha Products. The Cr (4.35% ^{50}Cr) and Ru (5.5% ^{96}Ru) samples consisted of 148 g and 50 g, respectively, of metallic powder contained in 5.5 cm x 5.5 cm x 1.4 cm plastic boxes. The Zn sample (48.6% ^{64}Zn) was made up of a stack of eight 5 cm x 5 cm x 0.16 cm plates. The Mo sample (14.8% ^{92}Mo) was a 5cm x 5 cm x 1.27 cm plate. "Blank" samples of high-purity Al and Cu of approximately the same size and shape as the other samples were used for background measurements. Energy and efficiency calibrations were performed in the experimental geometry using a calibrated ^{22}Na source obtained from Isotope Products Laboratories. The summed energy resolution for 1022 keV was approximately 4.5 keV FWHM. The 1022 photopeak detection efficiency varied from sample to sample due to differences in the gamma-ray attenuation within the samples. When the effects of this gamma-ray self-absorption and of the finite size of the samples are included, the 1022-keV photopeak efficiency was found to range from (2.1 - 4.1) x 10^{-3} .

The Cr, Zn, Mo, and Ru samples were counted for totals of 163, 161, 147, and 178 hours, respectively. The Al and Cu blanks were similarly counted for 138 and 108 hours, respectively. Spectra obtained from the Zn sample and from the Al blank are shown in Fig. 2. The spectra observed from all samples were very similar in overall shape and in the total number of counts obtained per unit time. In all spectra the only well-defined peaks are those at 1022 and 1461 keV. The 1461-keV line arises from the decay of the well-known long-lived isotope ^{40}K which is present in the building materials.

The 1022-keV counting rates observed from all of the samples studied are listed in Table I. These rates are much larger than those expected from cosmic-ray interactions. To determine the origin of this line, several tests were performed. The lead shielding was removed, and a ^{22}Na positron source was placed about 3 cm directly above the normal sample position. The source was collimated so that the positrons could not strike the top surfaces of the detectors. Data were accumulated both with and without a sample between the detectors. As was expected for such low-energy positrons, fewer 1022-keV events were observed when a sample was present than were seen without one. When similar tests were performed with a ^{56}Co gamma-ray source (which was surrounded with sufficient material to stop the positrons), just the opposite effect was seen; more 1022-keV events were detected with the sample present than without it. The sources were then removed, and counting was done both with and without samples present. Again, more 1022-keV events were seen with a sample than without one. Finally, the shielding was replaced, all sources were removed, and data were taken for a period of 52.4 hours with no sample present. As can be seen from Table I, fewer 1022-keV events were seen in this "empty" run than in any of the runs for which a sample was present.

These observations can be accounted for by the following mechanism. Although the flux of high-energy gamma rays reaching the sample volume is greatly reduced by the lead shielding, some still do so. These gamma-rays can then pair produce in the sample itself. The resulting positrons annihilate within the sample to produce the observed coincident 511-keV gamma rays. Possible sources of these high-energy gamma-rays include the decays of uranium and thorium daughter isotopes contained in building materials, and also the radiation produced by the nearby 88-inch Cyclotron. In fact, it was noticed that the 1022-keV counting rate increased substantially whenever high neutron fluxes were produced by the cyclotron beam. Data taken during these periods were discarded.

The counting rates in the 1022-keV peak observed from the Cr, Zn, Mo, and Ru samples were (within the statistical uncertainties) equal to those seen from the Al and Cu blanks. The net 1022-keV counting rate possibly attributable to β^+/EC decay was obtained by subtracting from the rate observed with each sample the average rate observed from the Al and Cu blanks. These net rates, together with the resulting 1-σ uncertainties, were then used to set upper limits on the net numbers of counts in each peak. From this analysis, 68% confidence level lower limits have been established on the half-lives of ^{50}Cr, ^{64}Zn, ^{92}Mo, and ^{96}Ru against ground state $\longrightarrow$ ground state β^+/EC decay. Searches were also made for ^{96}Ru β^+/EC decays to excited states in ^{96}Mo. No evidence of a peak was found at any of the expected positions. Calibrations performed with the ^{22}Na source demonstrated that with the system used in the present work, the probability for observing a three-fold coincidence is quite small. Thus, only very loose limits were obtained by searching for these higher energy peaks. However, these tests showed that the absence of a 1022-keV peak can also be used to establish limits on the half-lives of ^{96}Ru against β^+/EC decay to excited states in Mo and on the ground state $\longrightarrow$ ground state $\beta^+\beta^+$ decay half-life. These results are summarized in Table II.

There have been few previous searches for such decays. Fremlin and Walters[1] used photographic emulsions to record tracks of beta-particles emitted in the decays of a large number of nuclei, including ^{50}Cr, ^{64}Zn, and ^{92}Mo. The sensitivity of their experiment depended critically on the then unknown double beta decay energies. Using current values for these quantities[6], the limits established by Fremlin and

Walters are approximately 3.5×10^{14} yr, 1.9×10^{15} yr, and 5.6×10^{14} yr, for the β^+/electron capture decay half-lives of ^{50}Cr , ^{64}Zn , and ^{92}Mo , respectively. Berthelot et al.[2] used a proportional counter to search for the characteristic x rays expected to be emitted in the double electron-capture decay of ^{64}Zn and established a lower limit on the half-life for this process of 8×10^{15} yr. Again before the proper decay energies were established, Winter[3] used a cloud chamber to search for positron pairs from the $\beta^+\beta^+$ decay of ^{92}Mo . Since it is now known[6] that this decay mode is not energetically allowed, the limit obtained by Winter is meaningless. More recently Bellotti et al.[4] have used a single large volume Ge(Li) detector to search for gamma rays emitted following the double electron-capture decays of ^{92}Mo to excited states in ^{92}Zr . No evidence of such decays were observed and 90% confidence limits of $(3\text{-}6) \times 10^{18}$ yr were established for these modes. There have been no previous searches for any of the double beta decay modes of ^{96}Ru .

While the limits established in the present study are still substantially below the most recent theoretical estimates[7,8] for both the 0ν and 2ν β^+/EC and $\beta^+\beta^+$ decay half-lives of ^{92}Mo and ^{96}Ru , they represent improvements of factors of 500 - 1200 over those obtained in previous similar investigations. I am not aware of any published estimates for the β^+/EC decay half-lives of ^{50}Cr or ^{64}Zn . Increased sensitivity to these double beta decay processes will require higher detection efficiencies and lower background rates. The latter improvement could be obtained with the use of an active shield. For example, an annular NaI detector surrounding the Ge detectors could reduce the background rate by a factor of 2 - 3 .

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Figure Captions

1. Possible decay schemes of (a) ^{50}Cr , (b) ^{64}Zn , (c) ^{92}Mo , and (d) ^{96}Ru . Q_{KK} is the ground state $\longrightarrow$ ground state double electron-capture decay energy.

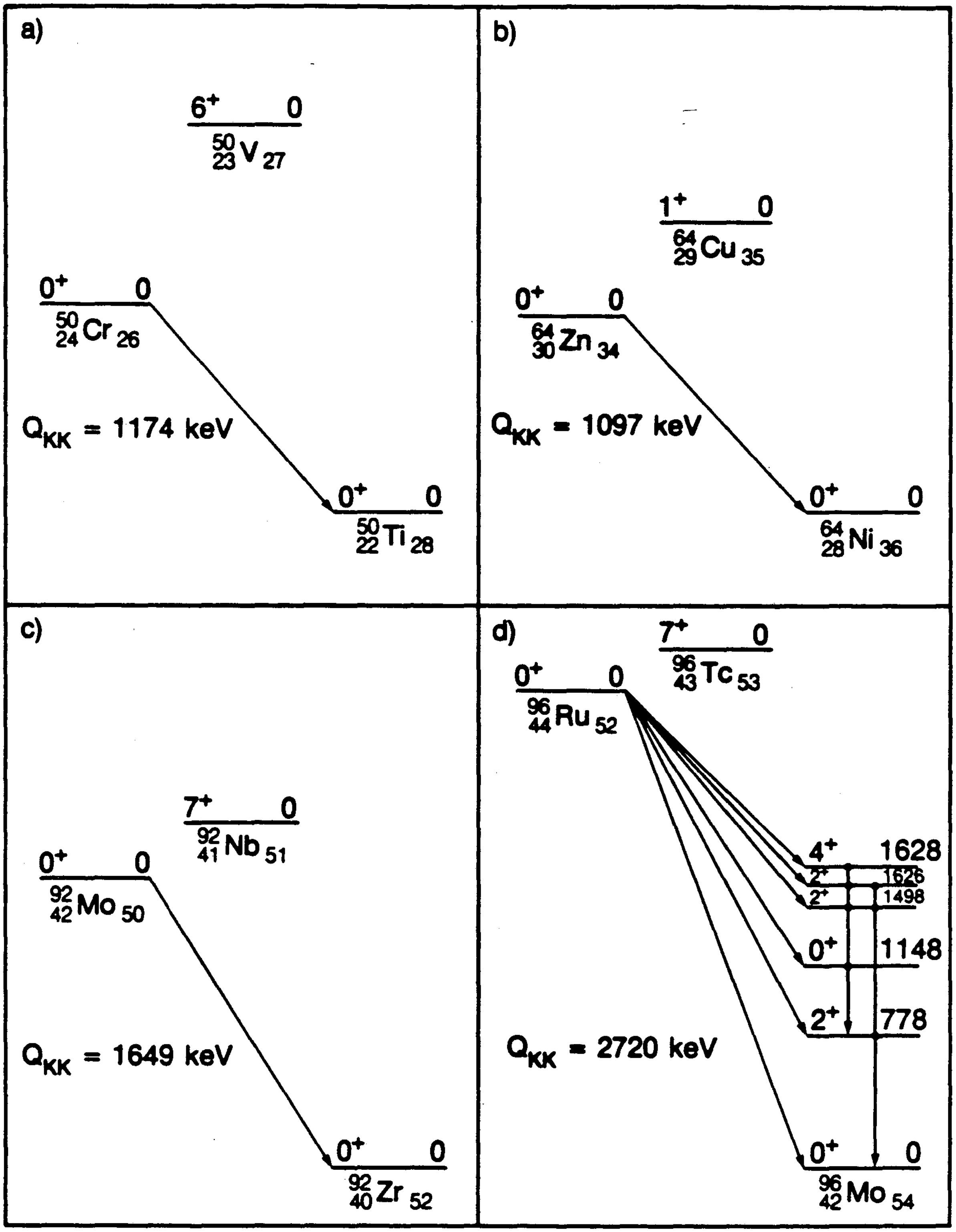
2. Summed energy gamma-ray spectra observed in (a) 72.7 hr of counting the Zn sample and (b) 69.3 hr of counting the Al "blank".

TABLE I. Observed 1022-keV photopeak counting rates.

Sample	N_{γ}(1022 kev)/10^5 sec.
Cr	7.6 ± 2.0
Zn	8.0 ± 1.9
Mo	11.7 ± 2.1
Ru	7.4 ± 1.9
Al	8.7 ± 2.0
Cu	9.8 ± 2.3
"Empty"	5.2 ± 3.8

TABLE II. Lower limits on the half-lives of ^{50}Cr, ^{64}Zn, ^{92}Mo, and ^{96}Ru established in the present study.

Decay	Mode	Final State J^{π}, E_x(keV)	$t_{1/2}$ (years)
$^{50}Cr \rightarrow {}^{50}Ti$	β^+/EC	0^+, 0	1.8×10^{17}
$^{64}Zn \rightarrow {}^{64}Ni$	β^+/EC	0^+, 0	2.3×10^{18}
$^{92}Mo \rightarrow {}^{92}Zr$	β^+/EC	0^+, 0	3.0×10^{17}
$^{96}Ru \rightarrow {}^{96}Mo$	β^+/EC	0^+, 0	6.7×10^{16}
"	"	2^+, 778	6.0×10^{16}
"	"	0^+, 1148	4.5×10^{16}
"	"	2^+, 1498	5.5×10^{16}
"	"	2^+, 1626	5.3×10^{16}
"	"	4^+, 1628	5.3×10^{16}
"	$\beta^+\beta^+$	0^+, 0	3.1×10^{16}



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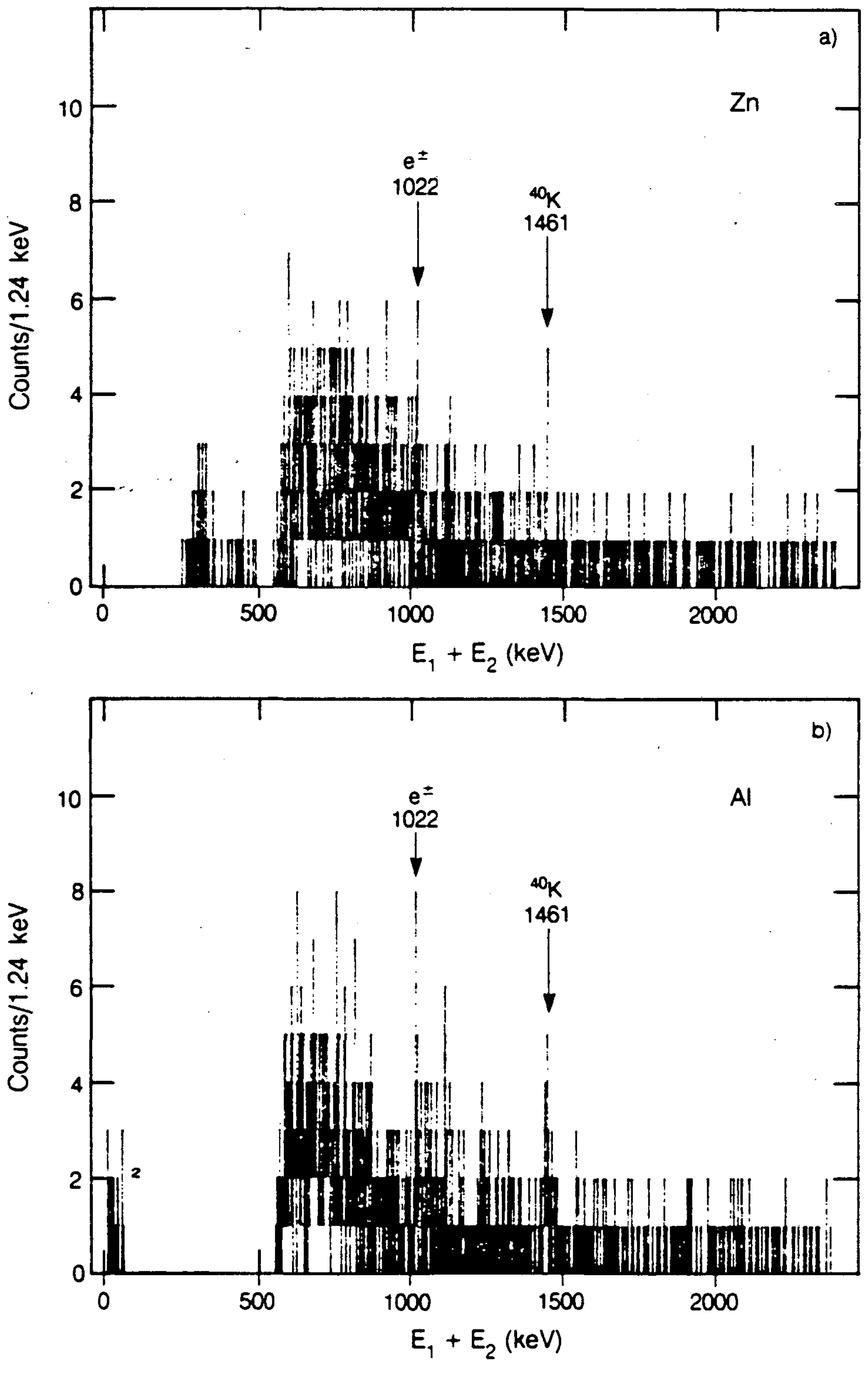

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LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720