EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

BETA-DELAYED PROTON EMISSION FROM ^{97}Cd AND ^{99}Cd

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ABSTRACT

Two new beta-delayed proton precursors, ^{97,99}Cd, have been identified at the ISOLDE on-line isotope separator. The proton branching ratio for ^{99}Cd is $\left(1.7\,^{+1.1}_{-0.5}\right) \times 10^{-3}$. The ^{99}Cd proton spectrum is discussed in terms of the statistical model with Porter-Thomas fluctuations. The half-life of ^{99}Cd was determined to be (16 ± 3) sec. A search for ^{98}Cd was also performed and it was found to be a pure beta emitter with a probable half-life of ∿ 8 sec.

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RADIOACTIVITY $^{97-99}Cd$ [from Sn(p,3p,Xn), chem. mass separation]. Measured β-delayed protons, E_p, $I(E_p)$, β^+-rays; deduced $T_{1/2}$ and P_p. Natural target, Si surface-barrier detectors, position-sensitive Si detector, plastic scintillator.

1. INTRODUCTION

Beta-delayed emission of charged particles (protons and alpha particles) is known to manifest itself in different ways for light[1] and heavy[2] nuclei. In light nuclei the particles are emitted from well-separated levels so that individual transitions are clearly identified, while for heavy nuclei the relevant region of excitation energy has much too high a level density to make observations of the single transitions possible with the present techniques. Therefore the beta-delayed particle spectra of heavy nuclides are characterized by a bell-shaped envelope several MeV wide.

This does not imply that fine structure is absent in the spectra of medium mass and heavy nuclides -- as we shall demonstrate by an example in this work. It means that the fine structure cannot simply be attributed to single strong transitions, but rather it emerges as the result of fluctuations in the nuclear transition probabilities, whose distribution is known to follow the Porter-Thomas law[3]. This fine structure, which is found in beta-delayed neutron emission as well, has until now only been observed for protons emitted from nuclides in the vicinity of A = 115 [e.g. ^{115}Xe [4] and ^{111}Te [5]]. Since information on level densities can be extracted[2] from the intensity fluctuations in delayed particle spectra, it is of special interest to look for new precursors in this mass region.

In this work we present the identification of two new neutron-deficient isotopes of Cd, which are beta-delayed proton precursors. The heavier of these, ^{99}Cd, is the only known $T_Z = +3/2$ precursor, while the lighter one, ^{97}Cd, is the heaviest member so far observed of the even Z, $T_Z = +1/2$ series from which five (tentatively six) proton precursors have been reported earlier[1,6]. On a part of the chart of nuclides, we show in fig. 1 the two new precursors together with the known ones from the $T_Z = +1/2$ series as well as the "island" of precursors around A ∿ 115.

2. EXPERIMENTAL TECHNIQUES

The neutron-deficient cadmium isotopes studied in the present work were produced in spallation reactions induced by bombarding a 115 g/cm^2 target of natural tin with 1.6 μA of 600 MeV protons from the CERN Synchro-cyclotron. The cadmium atoms were selectively evaporated from the molten tin, which was held at a temperature of ∿ 1200°C. They were continuously transferred by diffusion to a plasma ion source and subsequently separated into the constituent atomic masses by the ISOLDE electromagnetic isotope separator. The yield of ^{99}Cd was measured to be ∿ 4.5 × 10^2 atoms/sec, while that of ^{97}Cd was estimated to be ∿ 1 atom/sec on the basis of the proton branching ratio obtained from statistical model calculations[2)].

For the half-life determinations, two detection systems were used. In the first, the activity was collected on an aluminized mylar foil, and after a preset time transferred to a slot cut into a cylindrical plastic scintillator of dimensions 4 cm diameter × 3.5 cm length with a detection solid angle close to 4π sr. The positon signals obtained with this detector were recorded on a multichannel analyser that was operated in either multiscaling or multispectrum mode. To obtain half-lives of the Cd isotopes, different parts of the sequential energy spectra were analysed in order to obtain the best possible parent/(daughter + background) activity ratio.

Another system, which was used to measure the half-life for delayed proton emission from the ^{99}Cd source, consisted of a tape transport system by means of which the activity was moved with a preselected velocity along the sensitive area of a 45 mm × 7 mm × 340 μm position-sensitive surface-barrier (SB) detector. The detection solid angle for this system was 25% of 4π sr. The energy and position resolutions (FWHM) were 42 keV and 0.65 mm, respectively. By covering the detector with a thin brass plate, in which a row of slits -- 0.15 mm wide and 2.0 mm apart -- had been cut, the energy and position calibrations were determined from a single measurement of a standard alpha source (^{239}Pu, ^{241}Am, and ^{244}Cm combined).

For the beta-delayed particle spectroscopy the mass-separated ion beam was intercepted by a 20 μg/cm^2 carbon foil, behind which a counter system detected subsequent particle emission from the collected activity. During the experiment two detector set-ups were used. One was a SB counter telescope from which the summed energy signals, together with the particle identification signals, were stored two-dimensionally. The solid angle and energy resolution of this system were 4.8% of 4π sr and 41 keV, respectively. The other counter system consisted of a single high-resolution (FWHM = 17 keV) 100 μm, 150 mm^2 SB detector. The energy calibration of each detection system was performed with the alpha combination source and a pulse generator.

3. EXPERIMENTAL RESULTS

The experimental information obtained for the new cadmium isotopes is summarized as follows:

^{99}Cd

The half-life of ^{99}Cd was measured with the beta counter as well as with the position-sensitive detector. From the successive beta-spectra obtained in a multi-spectrum experiment we deduced $T_{\frac{1}{2}}$ = 15 ± 4 sec. In the analysis of the data from the position-sensitive detector, the position spectrum corresponding to proton energy signals was converted to a decay curve which is shown as the inset in fig. 2a. The result of this measurement is $T_{\frac{1}{2}}$ = 16 ± 3 sec. The two methods yield consistent half-lives, thus confirming the common source of both activities. The adopted half-life for ^{99}Cd is 16 ± 3 sec.

The proton spectra recorded with the detector telescope and the single SB detector are shown in fig. 2. In spite of the pronounced fine structure the spectra still show the few MeV wide bell-shaped envelope characterizing delayed particle spectra in the medium mass region. This gross structure is well understood in terms of the statistical model[2], while the fine structure is explained in terms of fluctuations in the transition probabilities. This will be treated in detail in the next section.

Following a recording of the particle spectrum with the telescope, the carbon collector foil was removed and the total number of collected atoms was determined via absolute counting of the residual 4.7 h ^{99m}Rh activity[7]. From this measurement a proton branching ratio of $\left(1.7^{+1.1}_{-0.5}\right) \times 10^{-3}$ was deduced for ^{99}Cd.

In addition to the proton spectrum, a few signals from alpha particles in the energy range 5-8 MeV were recorded. Beta-delayed alpha-particle emission has previously been observed[2)] from five precursors in this mass region. Four of these (all Cs isotopes) have predicted[8)] $Q_{EC}-B_{\alpha}$ values 2-8 MeV higher than that of ^{99}Cd, while the last one (^{76}Rb) has a value that is very nearly the same. The low alpha branching ratio measured[2)] for ^{76}Rb ($\sim 4 \times 10^{-9}$) and the difference in atomic number, which should result in a much higher Coulomb barrier for the Cd isotopes, do not make it very probable that these alpha particles are emitted from high-lying levels of ^{99}Ag populated in the beta-decay of ^{99}Cd. Yet an upper limit of $\sim 10^{-6}$ can be obtained for the absolute alpha branching ratio by assuming this assignment. However, it must be emphasized that no final conclusions concerning ^{99}Cd as a delayed alpha precursor can be drawn from the present results.

^{97}Cd

This new nuclide was identified unambiguously through its emission of beta-delayed protons observed in the energy range from $\sim$ 1-5 MeV. The production rate was too low to permit a determination of the half-life or the proton branching ratio.

In order to exclude a contamination from the much more abundant ^{99}Cd as the source of the proton activity seen at the position characteristic of mass-97, a one-hour measurement with the detector telescope at the mass-98 position was performed. No protons were observed during this period thus confirming the assignment of ^{97}Cd as the source of the protons.

Another conclusion that can be drawn from this measurement is that ^{98}Cd is a "pure" beta-emitter. The beta-activity at the mass-98 position was measured with the scintillator, and its decay was found to be complex. The shortest-lived component, with $t_{\frac{1}{2}} \sim 8$ sec, can presumably be attributed to ^{98}Cd.

4. DISCUSSION

Cadmium is one of the very few elements -- if not the only one -- for which one can expect to be able to study delayed particle precursors on both sides of a closed neutron shell. The most conspicuous effect of crossing the N = 50 neutron shell is found in the predicted P_α/P_p ratios for ^{97}Cd and ^{99}Cd. Because of the great difference in the Q_α values of the beta-decay daughters, ^{97}Ag (which has N = 50) and ^{99}Ag, the value of P_α/P_p is several orders of magnitude smaller for ^{97}Cd than for ^{99}Cd, in striking contrast to the less pronounced variation observed[9] among the Cs isotopes.

Using the statistical model[2,10] we have calculated the proton and alpha branching ratios for ^{99}Cd. In these calculations the spin and parity of ^{99}Cd were assumed to be $5/2^+$, in agreement with all known[11] values for odd nuclei with N = 51, and Q-values and particle separation energies were taken from Liran and Zeldes[12]. If the effects of Porter-Thomas fluctuations are ignored, the following results are obtained:

$$P_p = 2.2 \times 10^{-3}, \quad P_\alpha = 4.0 \times 10^{-9} .$$

The predicted alpha branching ratio is seen to be very small. The total gamma decay width is, however, so small over the entire proton energy spectrum that the depopulation of the particle-emitting states proceeds mainly via the proton and alpha channels. In such a case an important correction to the branching ratios due to the Porter-Thomas fluctuations of the transition probabilities has to be considered[2]. The mean value of the branching ratio

$$Z = \frac{\Gamma_\alpha}{\Gamma_\alpha + \Gamma_p} \tag{1}$$

actually becomes

$$\langle Z \rangle = \frac{\langle \Gamma_\alpha \rangle^{\frac{1}{2}}}{\langle \Gamma_\alpha \rangle^{\frac{1}{2}} + \langle \Gamma_p \rangle^{\frac{1}{2}}} . \tag{2}$$

Since $\Gamma_\alpha \ll \Gamma_p$ over the full spectrum, a considerable enhancement of the weak alpha branch is caused by the fluctuations. In this actual case the increase of P_α could be about three orders of magnitude, i.e. the alpha branch could be of the same order as the limit set in the present experiment.

The same calculation for ^{97}Cd, in which the ground state was assumed to have spin and parity $9/2^+$ (a neutron hole in the $1g_{9/2}$ shell), resulted in a proton branching ratio of 4%. This value was used in the estimate of the yield of ^{97}Cd in the present experiment.

The two new delayed-particle precursors reported here are remarkable in that they are nearer the closed N = Z = 50 shells than any other known precursor (see fig. 1). If one compares the delayed proton spectra from the three isotopes ^{119}Ba, ^{115}Xe, and ^{111}Te [which may all be found in a recent review[2)]] with that of ^{99}Cd, one notes a developing fine structure as the closed shells are approached. This effect is due to the change in the density of states ρ_i in the particle-emitting nucleus. The decrease in the level density with decreasing distance to the closed shells is demonstrated in fig. 3, which shows ρ_i, calculated from the formulae of Gilbert and Cameron[13,14)], as a function of excitation energy for these four proton emitters.

It is interesting to compare the ^{99}Cd proton spectrum with the spectrum of beta-delayed neutrons[15)] from ^{135}Sb, since, in both cases, the daughters after beta decay, are close to a double closed shell (N = Z = 50 and N = 82, Z = 50, respectively), which as mentioned implies low level densities. Ignoring the difference in the general shape at low energies, which is caused by different neutron and proton penetrabilities, the spectra resemble one another closely, each with a few strong "peaks". Such peaks are the result of Porter-Thomas fluctuations in the transition probabilities, and have recently[16)] been shown to be compatible with a purely statistical picture of the nucleus. For heavy precursors, the experimental line width is always large compared to the average spacing between states. An effect of this is illustrated in fig. 2 where ^{99}Cd proton spectra, measured with two different detection systems (FWHM = 41 keV and 17 keV, respectively) are compared. The high resolution spectrum shows clear substructure in the main peaks. Undoubtedly a further improvement in resolution would reveal even more.

The fine structure observed in delayed-particle spectra may be used to extract level densities in nuclei far from beta stability. The variance of the spectrum is related to the detector resolution W and the level spacing D_i, according to the expression[2)]:

$$\mathrm{Var}\, I = (2 \ln 2)^{\frac{1}{2}} \pi^{-\frac{1}{2}} W^{-1} \sum_f \sum_i \left[I^{if}\right]^2 D_i \alpha^{if} , \qquad (3)$$

where I^{if} is the particle intensity for each individual transition from an emitter state i to a final state f. The constant α^{if} is the normalized variance for the intensity of each such transition in the particle spectrum. In the cadmium cases the proton width dominates the total level widths in most of the spectrum, and thus the variance is due only to Porter-Thomas fluctuations in the beta intensity; under these conditions $\alpha^{if} = 2$. Through a comparison of expression (3) with the experimental variance, where W is known, a measure of the parameter D_i is obtained.

The variance for ^{99}Cd was obtained from an autocorrelation function analysis of the type described in ref. 2. The experimental spectrum g(x) was smoothed to yield $g_s(x)$, and a new spectrum

$$d(x) = g(x)/g_s(x) \qquad (4)$$

was formed. A new smoothing function, instead of the Gaussian one used earlier[2], was adopted here. Its general mathematical form is

$$S_\sigma^{(2n)}(x) = \sum_{k=0}^{n} \frac{(-1)^k \sigma^{2k}}{(2k)!!} h_\sigma^{(2k)}(x) \qquad (5)$$

where $h_\sigma^{(2k)}(x)$ is the $(2k)^{th}$ derivative of a Gaussian function with standard deviation σ. The higher-order functions of type (5) have the advantage of retaining the gross structure of the spectrum undistorted. The smoothed spectrum shown in fig. 4 together with the experimental one, was obtained using a folding function with n = 1. The autocorrelation function ψ(x) for d(x) was then derived and is shown as an inset in fig. 4 with a fitted theoretical curve for the same function[2]. The resulting variance, which corresponds to a 2.5 MeV average proton energy, is $\mathrm{Var}(I_p/\langle I_p \rangle) = 0.53$.

We may now proceed to calculate the variance from expression (3). The level spacing D_i was adjusted by variation of the level density parameter a (refs 13 and 14) until agreement was reached with the experimental variance. In this way we obtained a = 8.5 for ^{99}Ag, which is close to the recommended value[14] of 7.76 but indicates a 50% higher level density than predicted.

The level density is known[17] for two other silver isotopes, namely ^{108}Ag and ^{110}Ag. The a parameters for those and for ^{99}Ag are given in table 1. In all cases we find values close to but higher than the Gilbert and Cameron[13,14] values.

The statistics so far obtained for the proton spectrum of ^{97}Cd preclude any similar analysis. Hopefully this limitation can eventually be rectified since the nuclide belongs to the most extensive "family" of precursors known outside of the light nuclei, namely the $T_Z = +\frac{1}{2}$ family; from this, six (see fig. 1) delayed proton precursors, including ^{97}Cd, have been positively identified, and a systematic study of their behaviour in terms of a statistical-model analysis has begun[18]. Because of its position so near closed shells, ^{97}Cd should provide a critical test of whether a set of consistent parameters, capable of describing diverse but connected nuclear conditions, may successfully be obtained.

Acknowledgements

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Table 1

a parameters in the Gilbert-Cameron level density formula

Isotope	a parameters	
	Predicted a)	Experimental
^{99}Ag	7.76	8.5 b)
^{108}Ag	15.09	16.54 c)
^{110}Ag	16.23	18.24 c)

a) Ref. 14.

b) This work.

c) The a parameter was determined to reproduce the level spacing given in ref. 17.

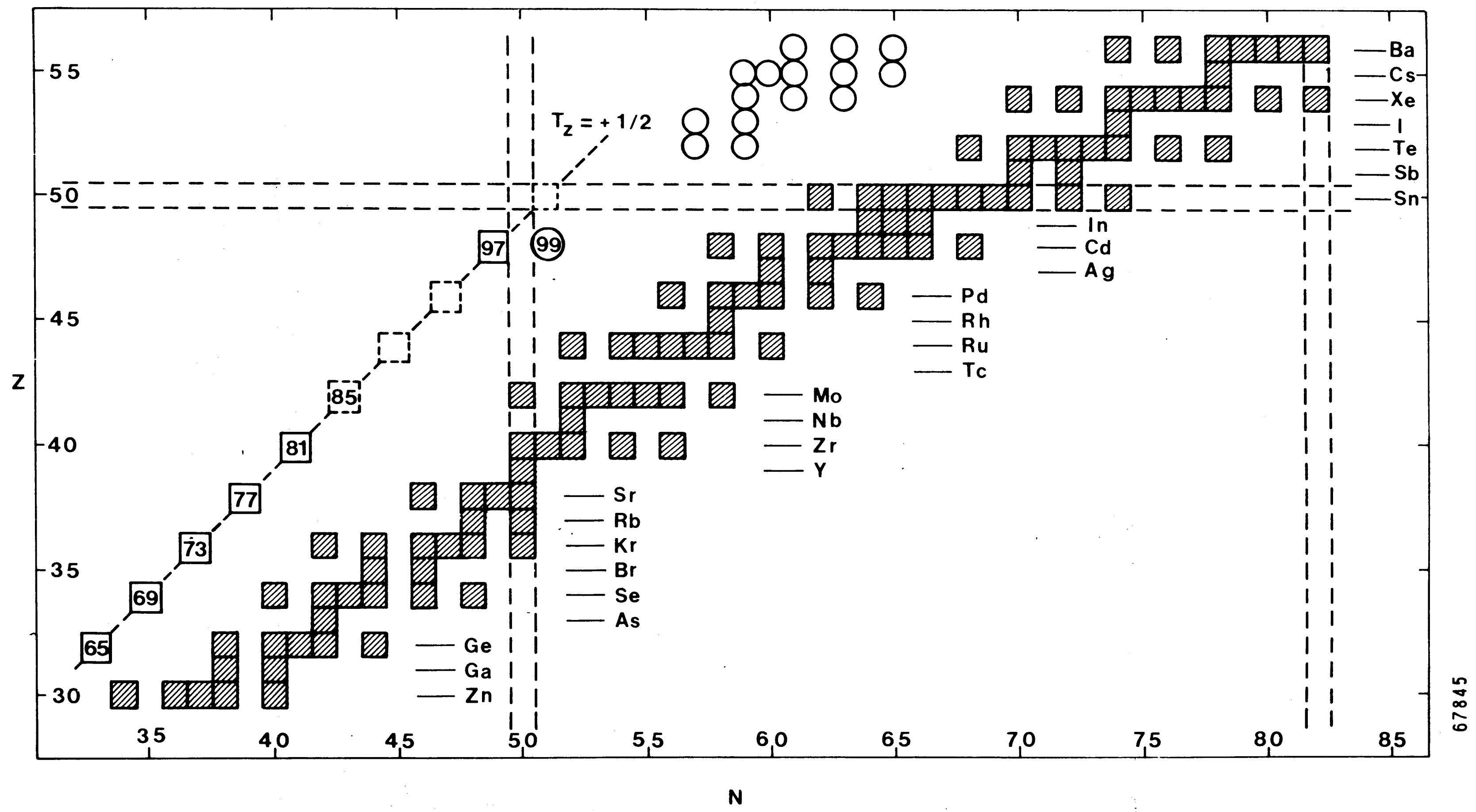
Figure captions

Fig. 1 : Part of the chart of nuclides showing beta-delayed proton precursors in the medium mass region. The precursors are divided into the $T_Z = +\frac{1}{2}$ series (□) and $T_Z > 1$ precursors (○). The hatched squares show the stable nuclides in this region.

Fig. 2 : Beta-delayed proton spectra from ^{99}Cd obtained with a) a SB counter telescope consisting of a 20 μm, 50 mm^2 ΔE detector and a 700 μm, 150 mm^2 E detector (FWHM = 41 keV), and b) a single 100 μm, 150 mm^2 SB detector (FWHM = 17 keV). The inset shows the decay curve obtained for the protons with the position-sensitive detection system described in the text.

Fig. 3 : Density (ρ) and average distance (D) of $I = 5/2$ levels (of one parity) at excitation energies around the proton-emitting regions of four known medium-mass proton emitters. The lines are fully drawn in the regions where proton emission is energetically allowed. The densities have been calculated from the formula of Gilbert and Cameron (refs. 13 and 14).

Fig. 4 : The measured energy spectrum of ^{99}Cd and a smoothed spectrum obtained with a folding function of second order (see the text for details). The autocorrelation function $\Psi(\tau)$ for a spectrum formed as the quotient of the two spectra (eq. 2) is shown in the inset. The solid curve shows a theoretical fit to the autocorrelation function.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85
97
99
Ba
Cs
Xe
I
Te
Sb
Sn
In
Cd
Ag
Pd
Rh
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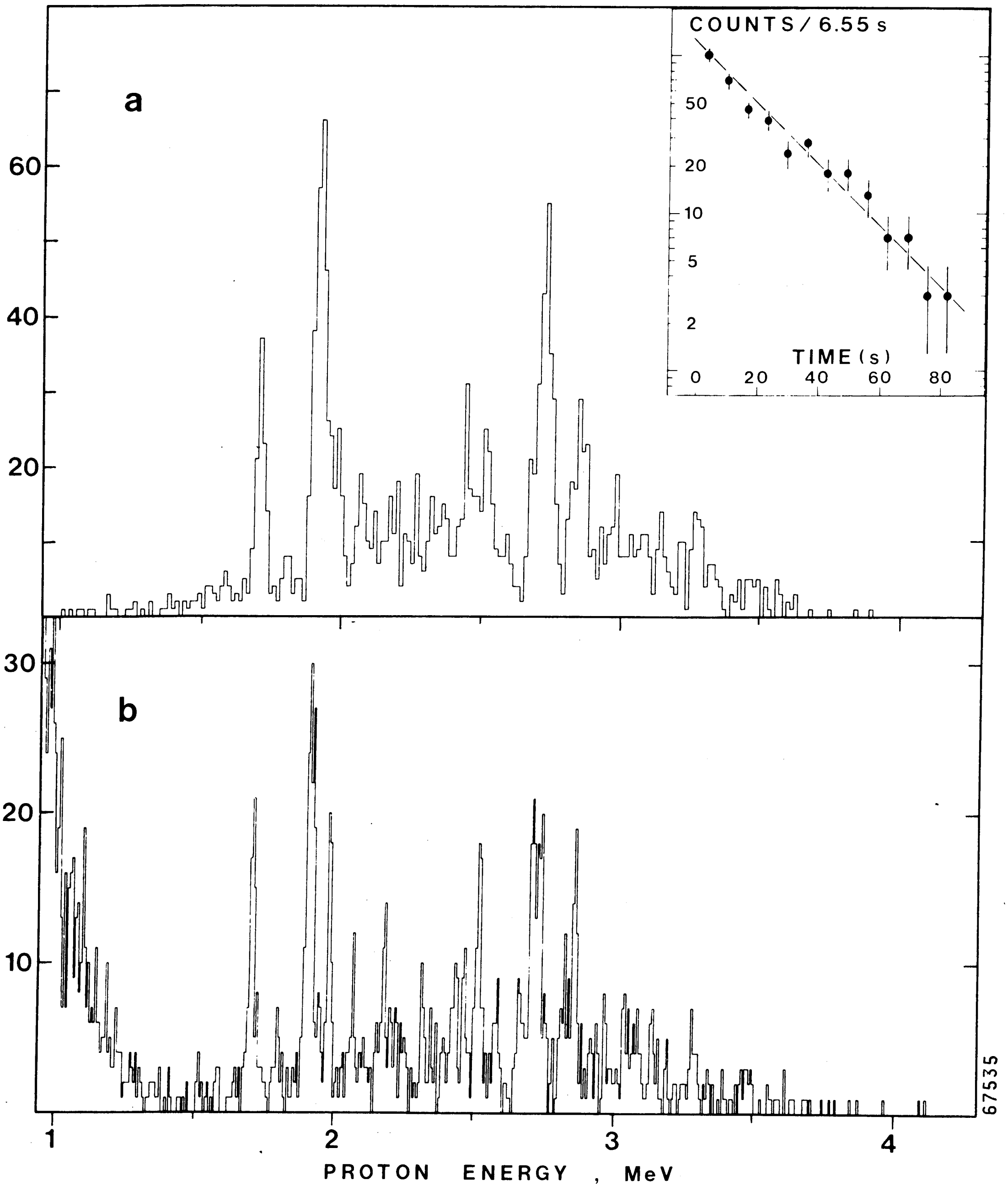
Fig. 1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TIME (s)
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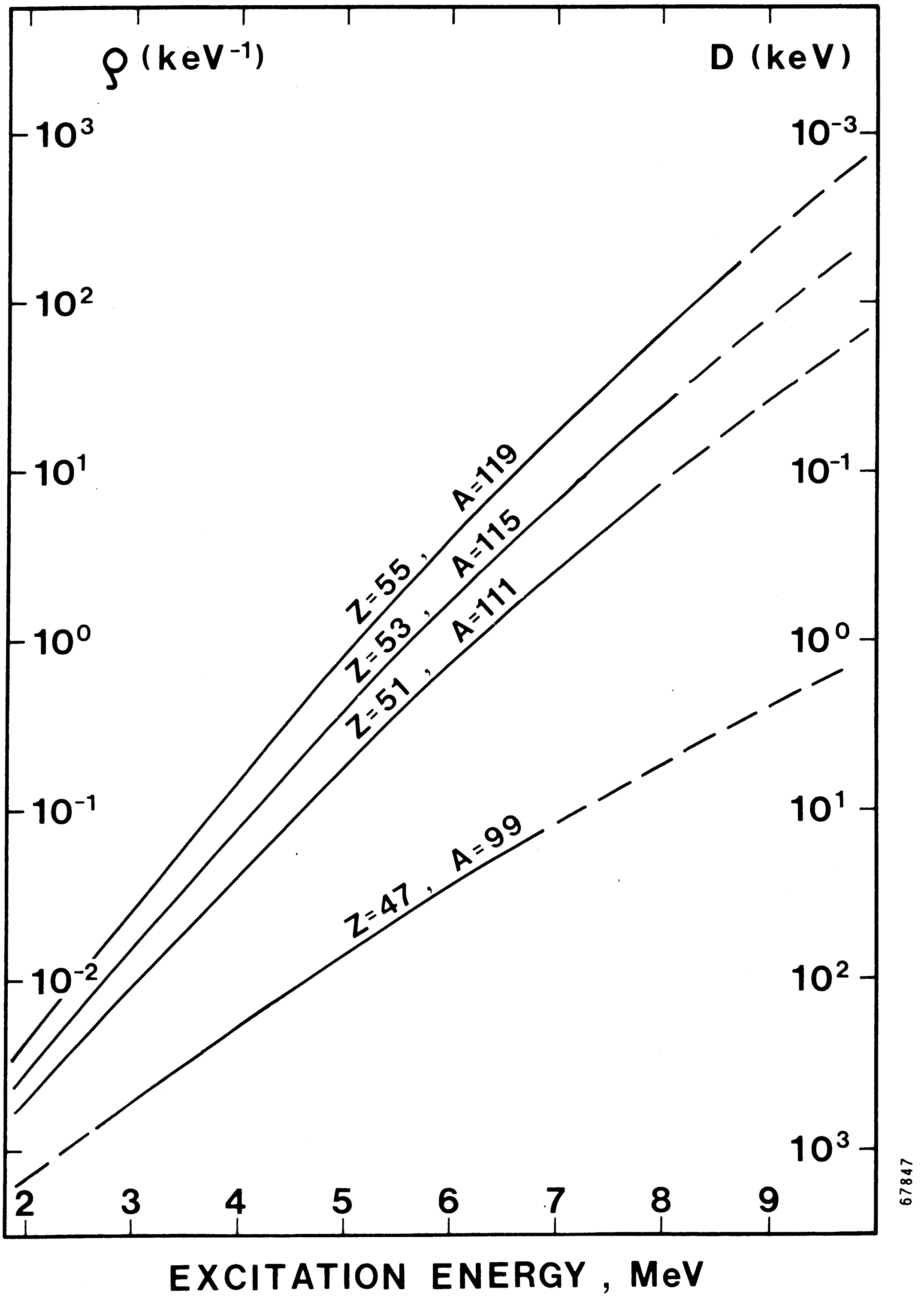
67535
1
2
3
4
PROTON ENERGY , MeV

Fig. 2



ρ (keV⁻¹)
D (keV)
10³
10²
10¹
10⁰
10⁻¹
10⁻²
10⁻³
10⁻¹
10⁰
10¹
10²
10³
Z=55 , A=119
Z=53 , A=115
Z=51 , A=111
Z=47 , A=99
2
3
4
5
6
7
8
9
EXCITATION ENERGY , MeV
67847

Fig. 3

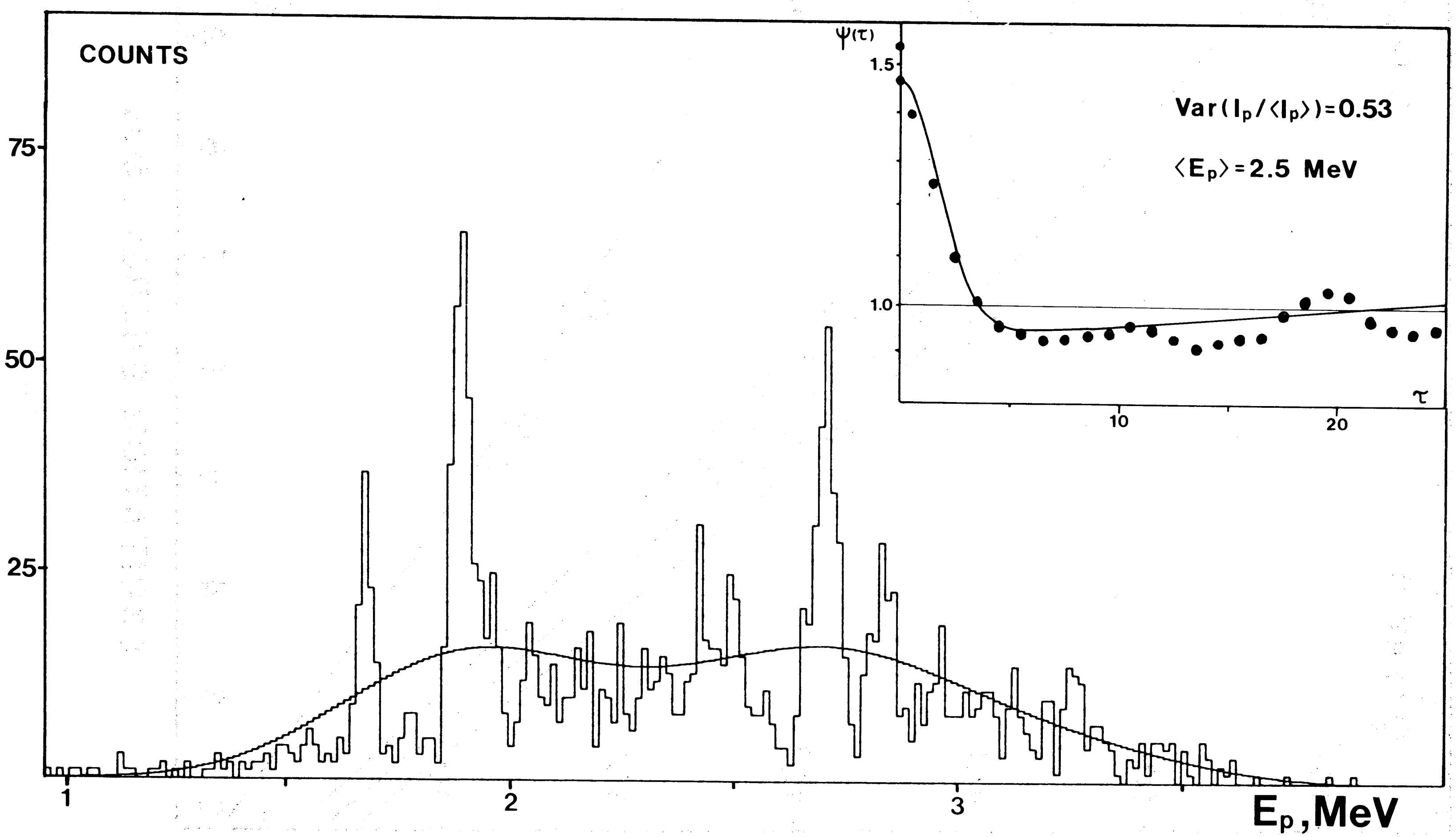


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Fig. 4