PHYSICAL REVIEW LETTERS

VOLUME 34 | 27 JANUARY 1975 | NUMBER 4

Spectroscopy of α and K- and L-Electron Continua and L-Electron Pickup in ^{210}Po α Decay*

H. J. Fischbeck†
Chemistry Division, Argonne National Laboratory, Argonne, Illinois 60439, and University of Oklahoma, Norman, Oklahoma 73069

and

M. S. Freedman
Chemistry Division, Argonne National Laboratory, Argonne, Illinois 60439
(Received 19 September 1974; revised manuscript received 1 December 1974)

The continuous low-energy electron distributions emitted from K and L atomic shells in a small fraction of ^{210}Po α decays have been measured from 6 to 100 keV. The corresponding complementary α satellite continua have been measured. Electron and α shapes agree well, but disagree with all extant predictions. K and L emission probabilities were determined. About 65% of the L (but very few of the K) electrons ejected are captured into bound states of the α particle.

In α decay, deep core electrons may be ejected from the atom, with very small probability. The continuous energy distribution of the electrons emitted from the i shell is expected to be concentrated near zero energy. The spectrum of the associated α, with which the electron shares a fixed total energy $E_0 - B_i$ (B_i the i-shell binding energy) is then a complementary mirror-image satellite distribution below the main α peak (energy E_0), with its upper threshold limit displaced B_i below E_0. The α decays of ^{210}Po [1,2] and ^{238}Pu [2] have been observed in surface-barrier spectrometers, in coincidence with K or L x rays.

Several attempts[3-8] to calculate the probability of ejection for the K, L, and M shells in ^{210}Po decay gave poor agreement[7] with the measured intensities of associated x rays which signal a core-vacancy creation. In a recent adaptation of the binary-encounter approximation (BEA) with zero impact parameter to this problem Hansen[9] obtained fair agreement for K, L, and M ejection, but gave no prediction of spectrum shape. All theories treat the ejection as arising from direct Coulomb interaction between a relatively slow α particle and a fast core electron but take no account of quasimolecular energy-level shifts as the α emerges. For K, L, or M electrons, the α passage is essentially adiabatic, so electron shakeoff due to the relatively slow reduction of the central field should play a minor role.

We have made the first reasonably precise measurements of the K and L ejected-electron spectra in α decay, covering essentially the full intensity of these emissions, from 6 to 100 keV. Electrons from a fractional-monolayer 40-μCi source of $^{210}_{84}$Po formed by retarded ion-beam deposition on a graphite film were observed over 60 days in an iron-free double-lens magnetic spectrometer with an efficiency-calibrated NaI scintillation detector, in coincidence with Pb K or L x rays detected in a Ge(Li) spectrometer behind the source. Experimental details will be published later.

The measured momentum spectra, transformed to energy spectra, are given in Fig. 1. In the energy range 6.2–10.4 keV, the K coincidence

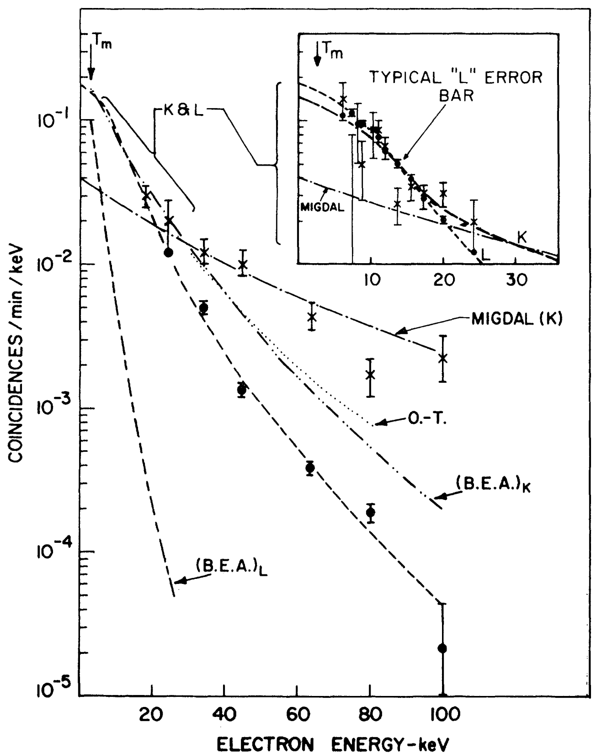

FIG. 1. Energy spectrum of ejected electrons associated with α decay of ^{210}Po. × and • denote electrons in coincidence with K and L x rays, respectively. K-shell ordinate values have been multiplied by 40. Insert shows that K and L spectrum shapes merge below ~20 keV. Short-dashed curve, drawn to guide the eye, extrapolated below 6 keV along locus which gives close fit to observed spectrum shape of L-ejection-associated *α satellite* (see text and Fig. 2). Dotted curve shows K data of Ovechkin and Tsenter (Ref. 10), matched to ours at 30 keV. Curve from Migdal theory (-•-) for K-electron ejection is matched to our data at 35 keV, and high-energy ($\gg T_m$) asymptotic forms of binary encounter approximation for K and L ejection are matched to our results at T_m ($\equiv 2.9$ keV).

rates were determined using only the $K\beta$ x rays [multiplied by $(K\alpha+K\beta)/K\beta=4.5$], to suppress coincidences between L Auger-electron lines in this range and $K\alpha$ x rays which precede them. A best fit through the K points below 20 keV supports the conclusion that K and L ejection shapes are quite similar in this range. This similarity in the region including the bulk of the intensity is predicted in general binary-encounter theory[11] *below* the energy $T_m = 4mE_1/M$, the maximum energy transferable in a collision between an impinging particle, mass M, energy E_1, and a free electron. However, for ^{210}Po α's, $T_m = 2.9$ keV, and *above* T_m, BEA calculations show a rapid divergence of the spectra of individual shells toward an asymptotic dependence[9,11] proportional to $(B_i+E_i)^{-9}$. Thus the observed agreement of K and L shapes to $\sim 7T_m$ disagrees with BEA theory, as applied to ionic bombardment of atoms, in the low-energy region. The high-energy BEA asympotic forms shown in Fig. 1 also grossly disagree with the K and L shapes. These spectral measurements are the first tests of BEA predictions for individually identified shells.

Figure 1 also shows the Migdal prediction for the K shell, normalized to the data point at 35 keV. Above 35 keV the agreement is fairly satisfactory, but clearly not below, although Migdal's prediction[3] for the K emission probability, 2.5×10^{-6} per α decay, is in fair agreement with our and others' data, as is Hansen's[9] value (see Table I.) He gave no L-electron-spectrum shape prediction.

We have also measured the K- and L-ejection-associated α continuum satellites in ^{210}Po, and the L-associated α satellite of the ground-state-feeding α group in ^{238}Pu decay. These were observed from similar ion-beam–deposited thin sources, in a high-resolution magnetic α spectrometer with a multielement focal-plane detector, in coincidence with K or L x rays in a NaI(Tl) spectrometer behind the source. An unfortunate accident disabled the spectrometer and prevented the accumulation of enough data to define the K continuum shape with satisfactory statistical accuracy; it also degraded the sensitivity of three detector elements on the low-energy tail of the L-associated satellite in ^{210}Po.

Figure 2 shows the ^{210}Po α-K and α-L continua, and the (chance coincidence) α_0 line. At the left is the *L-electron* energy spectrum from Fig. 1 on a linear intensity scale. We assumed that this measured shape is that of each of the L-subshell components. By locating the zero energy edge of the mirror images of these electron components at the indicated L-subshell α-continuum thresholds, and by letting both the relative subshell intensities and the form of the extrapolation of the L-electron shape below 6 keV (i.e., channel 2.4) be free parameters, the fit of the image-sum convolution with the α_0 line shape to the α-L continuum was tested. The exhibited best fit for L_1:L_2:L_3 = 0.35:0.35:0.30 is only slightly better than for that predicted by Hansen,[9] L_1:L_2:L_3 = 0.389:0.128:0.483. However, the quality of the fitting was fairly sensitive to the form of the extrapolation to zero of the L-electron energy spectrum, and ruled out any upturn or downturn which

TABLE I. Experimental and theoretical K- and L-electron emission probabilities per α decay of ^{210}Po.

Reference	Experiment	$P_K(\times10^6)$	$P_K(\text{theo-}\times10^6)$	Reference	Experiment	$P_L(\times10^4)$	$P_L(\text{theo-}\times10^4)$
This work	a) K X ray singles	2.03±.13[a]	Ref. 3 2.5	This work	d) L X ray singles	8.2±0.5	Ref. 3 1.13
	b) Elect.-KX coin.	2.5±0.7	Ref. 9 2.02		e) Elec.-LX coin.	3.05±0.46[b]	Ref. 9 5.9
	c) α-KX coin.	2.6±0.5			f) α-LX coin.	2.83±0.45[b]	
	b)/a)	1.28±0.32			e)/d)	0.37±0.06	
	c)/a)	1.23±.26			f)/d)	0.33±0.06	
Ref. 1	α-KX coin.	2.0±0.5		Ref. 1	α-LX coin.	8.6±2.2[c]	
Ref. 2	α-KX coin.	1.65±0.16		Ref. 14	L X singles	7.0±1.6[c]	
Ref. 10	K X singles	1.4±0.4[a]		Ref. 15	L X singles	8.0±1.2[c]	
Ref. 12	K X singles	2.1±0.4[a]		Ref. 16	L X singles	11.0[b]	
Ref. 13	K X singles	1.4±0.4[a]		Ref. 17	L X singles	3.8±.3[b]	
Ref. 14	K X singles	1.9±0.4[a]		Weighted average[b]		8.1±0.5[d]	
Weighted average		1.88±0.15[d]					

[a]Corrected for recent values of the intensity of the α branching to the 803-keV level in ^{206}Pb, 1.07×10^{-5}; for the K-conversion coefficient of the 803-keV $E2$ transition, 8.5×10^{-3}; and for the K-shell fluorescence yield, $\omega_K=0.968$.

[b]P_L values e) and f) from this work excluded from average. Ref. 16 value assigned arbitrary error of ±50%. Ref. 17 excluded from average.

[c]L x-ray singles or LX-α coincidence rates per α decay corrected by mean L-shell fluorescence yield, $\omega_L=0.37$.

[d]External error $=[\sum_i^n(X_i-\overline{X})^2/n(n-1)]^{1/2}$.

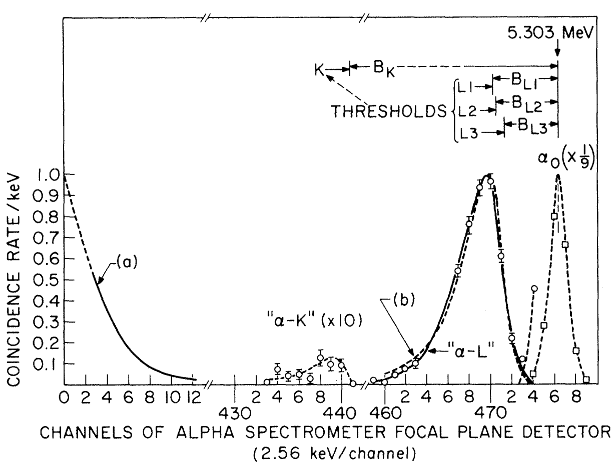

FIG. 2. α-satellite continuous energy spectra associated with ejection of K and L electrons by the ground-state (α_0) decay of ^{210}Po. α-K and α-L continua recorded in coincidence with K and L x rays of Pb. α_0 counts arise from chance coincidences. K and L binding energy displacements from α_0 shown. Curve a near zero energy is L-electron coincidence spectrum from Fig. 1; dashed part is extrapolated. Curve b is composite of three mirror images of curve a with leading edges at L_1, L_2, and L_3, and with relative intensities 0.35, 0.35, and 0.30, respectively, convoluted with α_0 line shape. Detector channels 464–466 failed during experiment. Absolute channel numbers of α continua are arbitrary with respect to channels for electron spectrum, a.

would change the area under the L spectrum from that of Fig. 1 by more than ±5%. Thus for the L spectrum the α and electron spectra supplement each other in determining low- (< 6 keV) and high-energy regions of the shape, respectively. To provide the convenience of an analytic form for the low end of the spectrum, we note that the extrapolation form thus selected happens to match the BEA K-shell (high-energy) asymptotic dependence, proportional to $(B_K+E_L)^{-9}$. A similar extrapolation is used for the K shape to integrate it. The excellence of the fit at the high-energy side of the α-L continuum indicates that the threshold displacements agree with the ($Z-2$; i.e., Pb) subshell binding energies to ±1 keV. For the spoiled α-K experiment the limit is ±2 keV.

In Table I are listed the K and L ejection probabilities per α decay of ^{210}Po, each derived from three measurements: the K and L x-ray singles intensities in the Ge(Li) spectrometer, the K- and L-electron coincidence spectrum integrals, and the K and L α-satellite coincidence spectrum integrals. The values are corrected for fluorescence yields and all apparatus parameters. The table also gives all published values[12-16] and Hansen's[9] and Migdal's[3] predictions. Our K- and L-x-ray singles data and our K-electron coincidence and $K-\alpha$-satellite coincidence data are all in satisfactory accord with other data and with Hansen's values. However, both the L-electron and $L-\alpha$-satellite coincidence values show only about 35% of the number of coincidences calculated from the observed L x-ray intensity even though they were measured simultaneously with the K coincidence (and K and L singles) measurements in both α and electron spectrometers. No apparatus defect such as coincidence inefficiency (measured to few percent accuracy) accounts for these nearly matching disparities in the L-e and L-α cases.

We believe this strongly suggests that in ~65% of the $\alpha-L$-electron-ejection interactions in ^{210}Po the electron is captured into a He^{++} bound state (~50% for ^{238}Pu). In such events neither the He^{+} nor the electron would be detectable in either of the magnetic spectrometers ($B\rho$ for He^{+} is beyond our spectrometer range), though the solid-state α detectors of Refs. 1 and 2 would respond indistinguishably to He^{+} or He^{++} and thus give P_L values consistent with L-x-ray singles data.

Halpern and Law[18] proposed K charge exchange into bound states of fully stripped projectiles to account for anomalously high x-ray yields ($\gg \propto Z^2$) in light ion (C^{+6}, N^{+7}, O^{+8}, and F^{+9}) bombardment of argon by Macdonald, Winters, and Brown.[19] They use a modified Brinkman-Kramers (BK) formulation of the K charge-exchange cross section, which depends sensitively and reasonantly on $\sim(v/u_K)^8(1+v/u_K)^{-20}$, with v the projectile velocity and u_K the initial bound-state K-orbital electron velocity. For ^{210}Po this σ_{BK} is only 1.1×10^{-30} cm^2 for K electrons, compared to our estimate of σ for K ejection of 10^{-26} cm^2, given by $P_K\times\sigma_{geom}$ for the K shell. This implies negligible K charge transfer to the α, as we observe. Extending this argument to the L-shell case indicates a probably unfavorable prediction for copious L charge exchange, unless the BK L-shell formula shows an even steeper dependence on v/u_L.

The spectrum shape disparities and the strong L-electron "pickup" suggests that theory must treat the continuously changing composite α-Pb level structure, and the desirability of ejected-electron spectrometry in current vacancy creation via ion bombardment studies.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

†Summer staff participant from University of Oklahoma.

[1]J. P. Briand, P. Chevallier, A. Johnson, J. P. Rozet, M. Tavernier, and A. Touati, Phys. Rev. Lett. 33, 266 (1974).

[2]M. Rapaport, F. Asaro, and I. Perlman, private communication.

[3]A. Migdal, J. Phys. (U.S.S.R.) 4, 423 (1941).

[4]J. S. Levinger, Phys. Rev. 90, 11 (1953).

[5]H. M. Schwartz, Phys. Rev. 100, 135 (1955).

[6]F. Grard, C. R. Acad. Sci. 243, 777 (1956).

[7]W. Rubinson, Phys. Rev. 130, 2011 (1963).

[8]G. Ciocchetti and A. Molinari, Nuovo Cimento 40, 69 (1965).

[9]J. S. Hansen, Phys. Rev. A 9, 40 (1974), and private communication.

[10]V. V. Ovechkin and E. M. Tsenter, At. Energ. 2, 282 (1957) [Sov. J. At. Energy 2, 344 (1957)].

[11]F. Folkmann, C. Gaarde, T. Huus, and K. Kemp, Nucl. Instrum. Methods 116, 487 (1974).

[12]W. C. Barber and R. H. Helm, Phys. Rev. 86, 275 (1952).

[13]M. A. Grace, R. A. Allen, D. West, and H. Halban, Proc. Phys. Soc., London, Sect. A 64, 493 (1951).

[14]M. Riou, J. Phys. Radium 16, 583 (1955).

[15]W. Rubinson and W. Bernstein, Phys. Rev. 86, 545 (1952).

[16]I. Curie and F. Joliot, J. Phys. Radium 2, 20 (1931).

[17]R. D. Scott, J. Phys. A: Proc. Phys. Soc., London 7, 1171 (1974).

[18]A. M. Halpern and J. Law, Phys. Rev. Lett. 31, 4 (1973).

[19]J. R. Macdonald, L. M. Winters, M. D. Brown, L. D. Ellsworth, T. Chiao, and E. W. Pettus, Phys. Rev. Lett. 30, 251 (1973).