Angular correlation between ejected L electrons and α particles in ^{210}Po decay*

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The angular correlation between α particles and electrons emitted from the L atomic shell during ^{210}Po α decay has been measured over an angular range from 0° to 90° for electrons in the energy range from 5 to 20 keV. The angular distribution of the α particles in the forward hemisphere with respect to the direction of the electrons was found to be almost isotropic with no strong forward peaking. Implications of this result on the hypothesis of electron pickup by α particles are discussed.

I. INTRODUCTION

Inner-shell ionization as a consequence of the emission of an α particle by the nucleus has been the subject of renewed interest both theoretical and experimental in the last few years. The application[1] of the binary-encounter approximation (BEA) in an impact parameter representation seemed to have overcome the discrepancies between the experimentally measured K-, L-, and M-shell ionization probabilities in the α decay of ^{210}Po (the only suitable α emitter for study having only a single α group and no γ rays) and the older theoretical predictions[2] based on time-dependent perturbation theory. Experimental work progressed from simple measurement of the individual shell ionization probabilities in ^{210}Po to our spectroscopic measurement[3] of the energy distributions of the ejected K and L core electrons, and of the companion α "K" and "L" satellite continua. Calculations of the electron energy spectra by Watson[4] based on the BEA formulation of Hansen[1] agree only at low electron energy with the observations on the L, but not with the K, spectrum shape, while the prediction given by Migdal[2] for the K-shell spectrum based on time-dependent perturbation theory shows fair agreement only at high electron energy.

While measuring[3] the electron and α energy spectra in coincidence with K and L x rays with a magnetic electron and a magnetic α spectrometer, respectively, a strange phenomenon was observed. We found that the number of electrons in the spectrum in coincidence with L x rays of Pb was only about 35% of the number expected from the number of L vacancies produced by α-L-electron interactions, and the number of α's in coincidence with L x rays was also only about 35% of the expectation. The number of K and L vacancies produced was known from the singles K and L x-ray counts in the x-ray detectors in both the electron and alpha spectrometer coincidence arrangements, corrected, of course, for fluorescence yield. The (x-ray singles) measured K and L vacancy rates (P_K and P_L) were in all cases in good agreement with the averages of all earlier measurements, and also with more recent values. Both the electron K and L coincidence spectra were measured simultaneously, as also were the K- and L-x-ray associated α coincidence spectra. In contrast to the gross deficiencies found in the electron-L x-ray and α-L x-ray coincidence rates, the corresponding electron-K and α-K x-ray coincidence rates were consistent with the observed K x-ray singles rates. The coincidence efficiencies were measured in regular intervals during each experiment to be near unity to a few percent accuracy.

Since this lack of L-x-ray associated electrons and α's was observed in two completely different experimental arrangements, i.e., an α spectrometer and an electron spectrometer, we suggested an interpretation that is in sharp contradiction to current expectations, namely, that in 65% of the α-L electron interactions resulting in L vacancy creation in ^{210}Po decay, the electron is captured into a He^+ bound state. In such events neither the electron nor the He^+ would be detectable in either magnetic spectrometer ($B\rho$ for a 5.3-MeV He^+ was beyond the range of the α spectrometer used).

To explore this assumption we cooperated with Dyer[5] *et al.* in an experiment aimed at the direct observation of He^+ emitted from a Po source. In this experiment the ratio He^+/He^{++} ions emitted from a thin ^{210}Po source was measured by differential magnetic deflection, in coincidence with Pb L x rays. The fraction of the He^+ ions observed was very small, about 1%, consistent with the known charge-state equilibrium fraction for α's of 5.3 MeV, which would appear to rule out electron capture as an explanation of the abovementioned absence of e-L x-ray and α-L x-ray coincidences.

Unfortunately it was not possible to perform the He^+ search experiment with sources produced in the same way as those used for the α-L x-ray and electron-L x-ray spectrometry experiments. The influence of this difference is uncertain, but perhaps critical. In the latter case the sources were produced by retarded-ion-beam (200 eV) deposition of ^{210}Po onto thin Al or C foils, while the source for the former experiment was prepared by autodeposition of ^{210}Po from a $\frac{1}{2}N$ HCl solution onto Ag. From the known amount of Po present in the source, both source types correspond only to a fraction of a monolayer of ^{210}Po. However, the degree of Po agglomeration is not known for either type of source. If the source preparation method used in the He^+ search experiment forms highly agglomerated deposits, the postulated 65% of He^+ ions formed by the α-L electron capture interaction could have been efficiently converted to He^{++} ions by charge-exchange collisions with other Po atoms in the local cluster to yield the equilibrium He^+/He^{++} ratio. Thus if the ion-beam-deposited spectrometer sources were not similarly agglomerated, then the negative result of the He^+ search experiment would not be conclusive evidence against the L-electron capture hypothesis.

It had also seemed of interest (actually prior to our experiments together with Dyer and Burch[3]) to obtain experimental evidence which would provide information of a different character on the inner-shell ionization caused by α emission. If the interaction between the α particle and the L orbital electron has the strong close coupling character implied by a high relative electron capture probability to give 65% He^+, one might expect the remaining 35% of the knocked-out L-electrons to be emitted highly preferentially in the direction of the α particle. Such forward peaking, if observed, would, in the absence of an existing theoretical calculation of the angular correlation, provide suggestive support for the capture hypothesis, but it is not an obviously necessary condition.

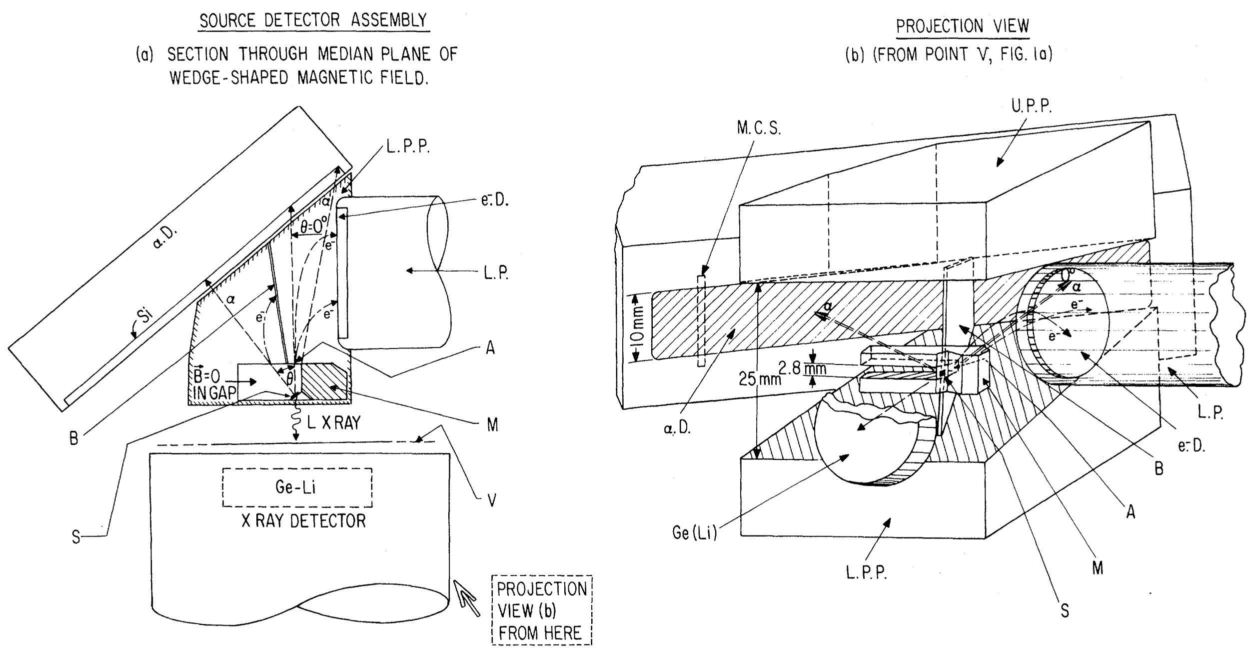
We therefore constructed an apparatus which allowed the simultaneous coincidence observation of α particles, electrons emitted in the 5–20 keV range, and L x rays of Pb. The angular distribution of the α particles in the forward hemisphere with respect to the direction of the electrons was observed by means of a position-sensitive α detector, to test this implication of a strong forward-peaked correlation of α-e_L in ^{210}Po decay.

II. EXPERIMENTAL SCHEME

The experimental arrangement is shown in Fig. 1. The forward direction ($\theta = 0$) is established by the (2.8×2.8 mm^2) aperture A in the magnetic shield source mount M, which defines the electron beam. The shield reduces the magnetic field in its 2.8-mm gap to a few percent of the nearby external field of about 150 Oe. Thus the trajectories of 5–20 keV electrons are very nearly linear within the shield, and their initial direction can be defined independently of their energy within 1° by aperture A, with a solid angle of 1.3% of 4π. This beam of electrons is aimed initially towards one end of the position-sensitive α detector and is then energy selected and steered toward the electron detector by a nondispersive wedge-shaped magnetic field with a 5:1 gradient. The thin brass baffle B stops electrons from reaching the e^- detector which have an initial angle of more than 11° with respect to the forward direction, but does not seriously interfere with the α particles emitted in the direction of the α detector. The shadow of the baffle B is observed in the α particle position spectrum as a dip in the continuous intensity distribution which can be used for position calibration.

The pole pieces of the adjustable magnetic-shunt-controlled permanent magnet which deflects the electrons were shaped so that electrons with $11° \geq \theta \geq -11°$ and energy in the range from about 5–20 keV would be deflected toward the detector; in this range it "focused" over 95% of the electrons passing aperture A onto the detector. This detector was a cleaved bare NaI (Tl) crystal 12 mm in diameter and 1 mm thick, coupled via a 24 cm long 2.54-cm-diam light pipe to a Quantacon photomultiplier tube with GaP first dynode coating. The energy spectrum of emitted L electrons drops rapidly with energy,[3] about 86% having less than 20 keV. The range of electrons selected was dictated by the lower limit for efficient detection of the electron detector and by practical limits on the range in $B\rho$ (2:1) of electron momenta that could be focused over a 22° range in initial angles onto a small detector area by the magnetic field. About 49% of all L electrons liberated by α particles are in the accepted kinetic energy range[3] from 5–20 keV. No α particles can quite reach the electron detector past the end of the slot in M.

The direction of emission of the α particles relative to the electrons was observed with α 1×5 cm^2 position-sensitive Si-Au surface barrier detector (Nuclear Diodes NTC – 1050 – 300), subtending 5.4% of 4π solid angle at the source. (The α-particle direction is insignificantly deviated by the few hundred oersteds magnetic field.) The detector provides two signals, one proportional to the α energy (resolution 80 keV) and the other proportional both to the distance between the α particle point of impact and the right-hand end of the detector and also to the α energy. After electronically dividing the position signal (P) by



α.D.=POSITION-SENSITIVE α DETECTOR
A= e^- ACCEPTANCE APERTURE
B= e^- BAFFLE (BRASS)
e^-D.=BARE Na I(TL) e^- DETEC. 12mm x 1mm
L.P.= LIGHT PIPE-LUCITE
L.L.P.= LOWER POLE PIECE
M=MAGNETIC SHIELD SOURCE MOUNT; YOKE SECTIONED

M.C.S.=2mm MYLAR CALIBRATION STRIP-(1 OF 5)
S=(1mm)^2 SOURCE ON 2mm x 7mm MYLAR STRIP
Si= RECESSED Si SURFACE (10 mm x 50mm)
U.P.P.=UPPER POLE PIECE
V= VACUUM WALL WITH Be WINDOW

FIG. 1. (a) Source-detector assembly; plan view of section through median plane of wedge-shaped magnetic field. Upper and lower pole pieces define wedge-shaped gap with 10-mm spacing along sensitive plane of the α detector and 25-mm separation at source. Inhomogeneous magnetic field "steers" electrons with $5 \leq E \leq 20$ keV onto NaI(Tl) scintillation detector. C-shaped soft-steel magnetic shield M defines field-free region within its 2.8-mm gap in which the source is mounted, such that initial electron trajectories are straight from source to electron acceptance aperture A ($2.8 \times 2.8 \text{mm}^2$) which defines direction $\theta = 0°$. Brass e^- baffle B bounds left edge of aperture. Magnetic shield M (7mm thick) is suspended within 25-mm pole gap. Source faces middle of α detector through slot in shield M. Magnet yoke, shunt, Alnico magnet, and vacuum chamber not shown. Two sample α trajectories and three sample e^- trajectories shown. (b) Projection view into pole gap from slightly above median plane from viewpoint V of (a). Only "functional" components shown. Face of bottom pole piece, lower inside face of slot in shield M, and Au-plated surface of α detector all shown hatched. Two α and two e^- trajectories shown.

the energy signal (E), the energy-independent position (P/E) signal was observed in a multichannel analyzer (MCA) gated by the triple coincidence ($\alpha - e_L - x_L$). A collimated beam of α's on a micromanipulator was used to establish the relation between channel and position.

The α detector was subject to severe radiation-damage degradation which required overnight annealing in a humid atmosphere at 70 °C in an oven every 48 h to restore its performance. The position resolution deteriorated from an initial 2 mm to ~3 mm under the intense α bombardment of 2×10^6/min. Two such detectors were alternated in the angular correlation apparatus. The continuous degradation in energy and position signal amplitudes required an *in situ* position calibration device that could be easily and frequently monitored. To this end, five 6.4-μm Mylar strips each 2 mm wide and 12 mm long were mounted at regular intervals close to the detector surface. α particles reaching the detector after passing through the Mylar are degraded about 1 MeV in energy. This degradation does not influence the position spectrum since each (degraded) position pulse is divided by its correspondingly degraded energy pulse. However, if the α-position spectrum is periodically briefly observed by gating the MCA only on full energy alpha events (in self-gated mode, not requiring electron or L x-ray gating signals), then each strip will appear on the α singles position spectrum as a hole in the intensity distribution which enables one to identify the known strip positions with channel numbers.

The α detector was mounted so that the α's emitted in the forward direction, i.e., through the aperture A at $\theta = 0°$, registered in channel 35, corresponding to a distance of 10 mm from the right end of the active detector area, Fig. 1, while α's emitted at 88° from forward registered in channel 150 corresponding to a distance of 49 mm from the same end of the active area.

The L x rays from Pb, the daughter product of ^{210}Po α decay, were observed with a 6.5-mm-thick Ge(Li) low energy photon spectrometer (ORTEC) which had an active area of 4.9 cm^2 and a resolution of 900 eV for 6.4-keV x rays. The axis of the detector disk was in the median plane of the magnetic field, which was the plane of symmetry for all three radiation detectors and for the source. The detector was mounted outside a thin Be vacuum window about 3 mm behind the source, and subtended a solid angle of about 12% of 4π. The x rays traversed 182 mg/cm^2 of Be absorber to reach the detector.

The assembly was housed in a diffusion-pumped brass chamber at a pressure of 5×10^{-5} Torr. At this pressure 10-keV electron mean-free-path lengths are about 100 times the trajectory lengths, and He$^+$ ions would experience only about 1% probability of a charge-stripping collision.

III. ELECTRONICS

The triple-coincidence circuit used two time-to-amplitude converters (TAC) which were both started by pulses from the x-ray detector (see Fig. 2). One TAC was stopped by the α-energy signal and the other was stopped by the electron signal. The base width of the $\alpha - x$ prompt coincidence time peak was 0.25 μsec and the base width of the e-x time peak was 1 μsec. The output of each TAC was fed into a fivefold slow coincidence circuit. The remaining three coincidence requirements came from the α energy discriminator, the electron signal amplitude discriminator and the x-ray energy discriminator. The α discriminator was set low enough to reject only noise. The electron discriminator was set to accept pulses above the upper end of the well defined one photoelectron peak amplitude to discriminate against single photoelectron pulses arising from the decay of long-lived trapping states[6,7] in NaI (Tl) that are prolifically populated by high-energy ionizing events, such as α's. With this discriminator level the detection efficiency for 5-keV electrons has been measured as ~60%, and rapidly rising with electron energy. The electron discrimination is set differentially so as also to reject large overload pulses caused by α's coming

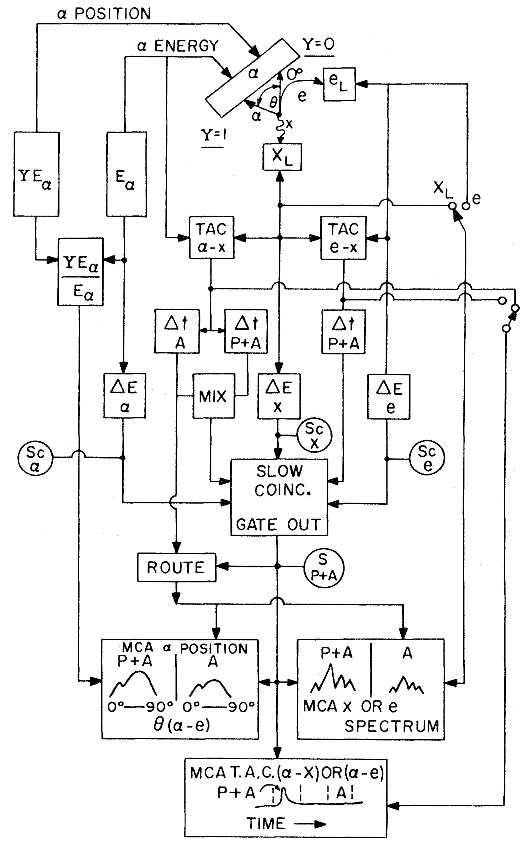


FIG. 2. Schematic block diagram of electronics. Amplifiers, fast pulse forming components, delays, inverters, bias supplies all omitted. Scalars shown as circles. $0 \le Y \le 1$ denotes α-particle point-of-impact fractional displacement along α-detector plane. ΔE, Δt denote single-channel analyzers, MCA denotes multichannel analyzers; P is prompt, A is accidental. Detector array at top; See Fig. 1. See text.

from some contamination on the NaI (Tl) which developed continuously during the experiment by the well-known vapor phase spreading of Po. The x-ray discriminator was set differentially to record only the energy region of Pb L x rays, ~8–17 keV.

The output of the slow-coincidence circuit was used to gate the MCA's which recorded the α-position spectrum, the x-ray or electron spectrum, and either TAC spectrum. Only in the $\alpha - x_L$ channel was there a significant number of chance events. These were recorded simultaneously with the true coincidence events by placing a single-channel analyzer (SCA) window "A" (of width equal to the SCA window width setting "P" which covered the region of prompt events) on the flat time spectrum above the region of prompt events. The chance events thus selected by the "A" SCA were routed into the second half of the analyzer memories for the MCA's recording the α position and the x or e energy spectra. These were then subtracted from the "$P+A$" events in the prompt-gated first-half memory sections to obtain true coincidences.

IV. CALIBRATION

The apparatus was tested by observing the angular correlation between α particles in the decay of ^{238}Pu and L conversion electrons from the 43-keV transition in the daughter product ^{234}U in coincidence with U L x rays. This decay is a fairly good standin for the ^{210}Po L-ejection decay; the α energy is 5.4 vs 5.3 MeV, the L_2 and L_3 conversion electrons of ~22 and 26 keV can be "focused" with fair efficiency in our setup, and the L_α x rays are 13.5 vs 10.5 keV. The 28% α branch to the 43-keV level and the large internal conversion coefficient gives large coincidence rates compared to the 8×10^{-4} L vacancies per alpha decay for ^{210}Po.

The results are shown in Fig. 3 together with the theoretical prediction for the $\alpha-e_L$ angular correlation for a $0^+-2^+-0^+$ transition calculated from Rose, Biedenharn, and Arfken[8];

$$W(\theta)=1+b_2(e)b_2(\alpha)A_2(\gamma)P_2(\cos\theta)+b_4(e)b_4(\alpha)A_4(\gamma)P_4(\cos\theta);$$

$b(p)$ is the particle parameter for particle p.

The curve marked M–F–exp. was obtained from a measurement of the $\alpha-\gamma$ (43 keV) angular correlation by Milton and Fraser[9] by multiplying

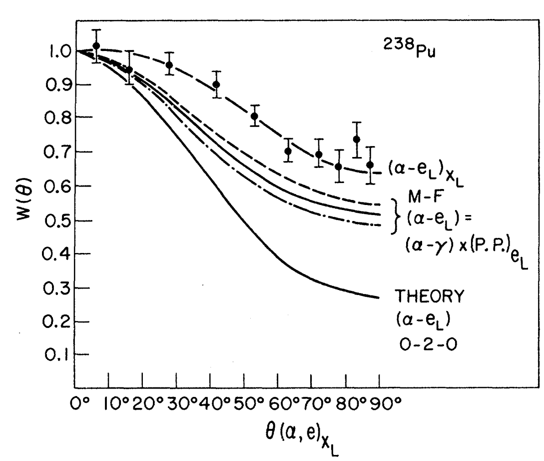


FIG. 3. Angular correlation α-L conversion electron of 43-keV transition in α decay of ^{238}Pu. Lower curve marked theory is prediction for a $0^+-2^+-0^+$ $\alpha-e_L$ correlation. Middle band is from Milton and Fraser's α-γ (43-keV) measurements, including experimental error, times particle parameters for e_L. Upper curve is present measurement in coincidence with $\alpha-e_L-x_L$.

their *experimental* values $g_2(\gamma)b_2(\alpha)A_2(\gamma)=0.330\pm0.037$ and $g_4(\gamma)b_4(\alpha)A_4(\gamma)=-1.145\pm0.065$ by the respective conversion electron particle parameters $b_2(e)=1.354$ and $b_4(e)=0.085$. [$g_i(\gamma)$ are γ correlation attenuation coefficients.] All curves are normalized to one at 0°. The α-e-x_L data have been corrected for the distortions arising from $\alpha-e$ angular correlation effects, as explained later in Sec. V. The corrections are small, and apply only in the angular range −10°–0°. No correction is needed for possible $\alpha-x_L$ correlation asymmetry since, as will be shown in the ^{210}Po case, this is mainly a rate-dependent effect, insignificant for the ^{238}Pu case.

As can be seen the *shapes* of the distributions are in satisfactory agreement. The differing attenuations in the experimental results (α-e_L as calculated from MF's data and α-e_L as measured) compared to theory can be attributed to nuclear quadrupole interactions with electric field gradients after the recoiled nucleus has relocated within the lattice of the source material. Since the lifetime of the 43-keV level in ^{234}U is 2.7×10^{-10} sec and the stopping time of the recoiling nucleus is $\sim4\times10^{-14}$ sec practically all of the L conversion electrons will come from U atoms dislocated into the source backing. The source preparation method used by Milton and Fraser for their $\alpha-\gamma$ angular correlation experiment was different from our method of depositing the ^{238}Pu sample. They deposited Pu from a HNO_3 solution onto 1-mg/cm^2 mica while our source was an invisible deposit of 1 μgm/cm^2 by retarded (300 eV) ion beam in an isotope separator onto 0.07-mm Al.

Our results have not been corrected for finite detector solid angle [we have estimated these to give only (2–3)% correction], while our data compared to the $\alpha-e_L$ correlation calculation based on Milton and Fraser's results is attenuated by a further 16%. We cannot say whether this discrepancy is due to more severe attenuation in our source due to quadrupole interaction or due to scattering of the low-energy electrons emitted from the deeply buried stopped recoils (about 10 μgm/cm^2 deep) in the source. Whichever it is, the Po $\alpha-e_L$ correlation would not be affected to nearly the same extent if the ^{210}Po and ^{238}Pu sources were similarly distributed since the electrons from the Pb L shell are emitted simultaneously with the α before the recoiling atom has moved into the backing or has precessed. This point must remain moot since the ^{210}Po source could not be prepared for the angular correlation experiment by the preferred method of ion beam deposition, but was made by autodeposition from solution. For such a source local agglomeration may enhance scattering effects.

In summary, the test measurement of ^{238}Pu indicates the capability of the method to yield angular correlations showing the expected shape, with perhaps some attenuations that may arise in the source. The result surely does not imply that the method grossly suppresses a strongly peaked correlation, whose search, in the L-electron ejection in ^{210}Po α decay, is the goal of the experiment.

V. ^{210}Po EXPERIMENTS

A. Source preparation

The Po source was prepared by autodeposition on Ag from a solution of Po in $\frac{1}{2}$ N HCL. The solution was made[10] by bringing to dryness 0.32 ml of a solution of Po $(NO_3)_2$ in dilute nitric acid containing 1 mCi of ^{210}Po, and taking it up in $\frac{1}{2}$ N HCL. Since the original solution contained a visible amount of inorganic residue (probably silica) which interfered with clean deposition, it was found necessary to filter the HCL solution through a micropore membrane. The HCL was ultrapure[10] and only specially leached glassware was used. A 1 mm^2 spot of 80-μgm/cm^2 Ag was vacuum evaporated onto one side of the end of a 1-mm-wide strip of 76-μm Mylar. The Ag spot was dipped into the ^{210}Po solution for 15 min, washed with distilled water and α counted, then dipped again for an additional 15 min and rinsed. The α activity measured in a low-geometry α counter was 4.14×10^7 dpm, corresponding to an average mass density of 0.5 μgm/cm^2. Inspection at 100-diam magnification showed a dark, smooth, slightly grainy surface. The 5.3-MeV α group observed with a surface barrier detector had a half width of 21.5 keV, matching the detector resolution. At 2% of peak height the width was 86 keV, broadening towards the low-energy side, indicating some source energy degradation. Less than 2% of the Po was on the backside of the Mylar.

The source spot was stuck onto a 2 mm wide $\times$7 mm long strip of Scotch tape stretched across the gap of a two-legged brass clip which was fastened to the magnetic shield M, Fig. 1, with the Ag spot facing the α detector midline through the slot in M. Thus α's are measured within $\pm 45°$ to the source normal, electrons at $-45°$ to the normal and the about 12-keV L x rays must penetrate the source support film and the Ag spot.

B. α-x_L angular correlation

Preliminary runs were made on double-coincidence angular correlations of α's with L x rays and with electrons.

The angular correlation between α particles which have created L vacancies and the successive L x rays is expected to be isotropic and can thus be used to explore any intrinsic assymetry of the apparatus. The observed α-position spectrum in coincidence with Pb L x rays, $N_{\alpha x}(\theta)$, Fig (4a), compared to the singles spectrum $N_\alpha(\theta)$, shows anisotropy at the ends of the angular range, Fig. Fig. 4(b), $N_{\alpha x}(\theta)/N_\alpha(\theta)$.

First we note that the α singles position spectrum has the strongly centrally peaked and bumpy shape seen in the curve in Fig. 4(a). This extremely nonuniform position spectrum for the isotropically emitted α particles is, surprisingly, about what is expected from the calculated variation of source-detector solid angle along the α detector-sensitive surface, including the effects of partial shadowing by the electron baffle B and the inner walls of the slot in the source mount M, and the end drop-off in detector sensitivity. This shape was seen with two different sources, ^{210}Po and ^{238}Pu. In the case of ^{238}Pu such features as the notch near 10° are more sharply defined; the ~1 mm position resolution eroded with the very

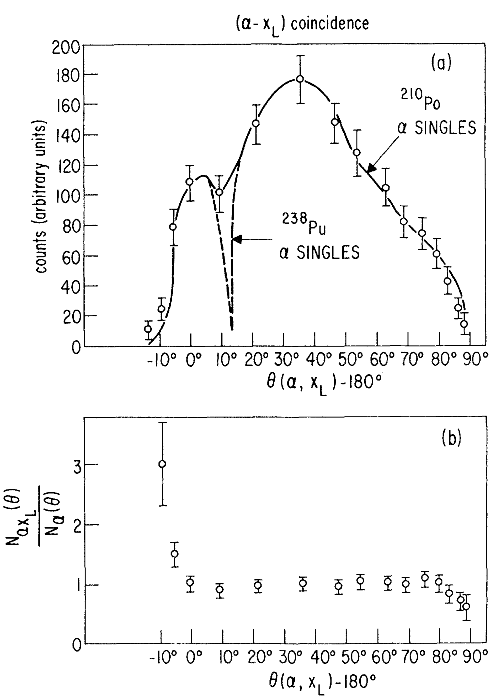


FIG. 4. (a) $\alpha - x_L$ coincidence counts as a function of $\alpha - x_L$ angle θ ($-180°$) in ^{210}Po decay. Solid curve is α singles position spectrum of ^{210}Po. Shallow "notch" near 12° shows poorer resolution of shadow of e^- baffle B at very high α count rates compared to dashed curve which is α singles of weaker ^{238}Pu source. (b) Angular correlation $\alpha - x_L$ of ^{210}Po.

high α rate of the ^{210}Po source, to ~2 mm. The (P/E) α position divider circuit incurred an ~30% dead time counting loss at this α rate.

Two causes could account for the the observed $\alpha - x_L$ anisotropy, Fig. 4(b). A shift of a few channels to smaller angles of the α-position spectrum observed in coincidence (i.e., when the MCA is gated by coincidence pulses) relative to the α-position spectrum when seen in singles (MCA not gated) could give the observed rise at $-10°$ and drop off near 90°. Such a shift (two channels) was observed at low count rate with the ^{238}Pu test source. In this case the position of the e^- baffle B could be identified very accurately within one channel. This was not possible with the ^{210}Po source which was 200 times more intense.

Some of the small excess $\alpha - x_L$ anistropy at low angles compared to the drop near 90° may also be due to low-angle scattering of α particles on the e^-baffle B, producing K x rays of Cu and Zn. The Cu and Zn K x rays are countable within the SCA window setting of 8–17.0 keV for the Pb L x rays. The geometry of the source was such that this would result in preferential observation of such events in the forward direction. The observation that when the $\alpha - x$ angular correlation was measured accepting a much wider range of photon energies the anisotropy at low angles increased considerably seems to support this explanation. The $\alpha - e - x_L$ coincidence results were corrected for the instrumental $\alpha - x_L$ asymmetry by dividing the triple coincidence rate by the double coincidence rate at each angle.

C. α-e^- angular correlation

A pronounced peak in the forward direction was observed for those α particles which were in coincidence with *all* electrons in the energy range 5–20 keV, Fig. 5. Since no x-ray coincidence was required any electron in this energy range liberated by an α particle from the decaying atom or any other was accepted. For reasons given below we do not believe this peak arises from $\alpha - e_L$ interaction; it is not the peak we seek.

Glancing angle collisions of the α's with the slot walls of the source mount M, in front of the source, and with the e^- baffle B will produce secondary electrons which are projected towards the e^- detector if formed near $\theta = 0°$, and the scattered α particle has a high probability of aiming at the α detector. Such glancing angle scattering is known to be highly probable. α sources plated onto stainless-steel disks exhibit an effective solid angle of 51.5% in a $2\pi\alpha$ counter, the excess 3% (of the 50% initially emitted into the steel surface) arising from small-angle cumula-

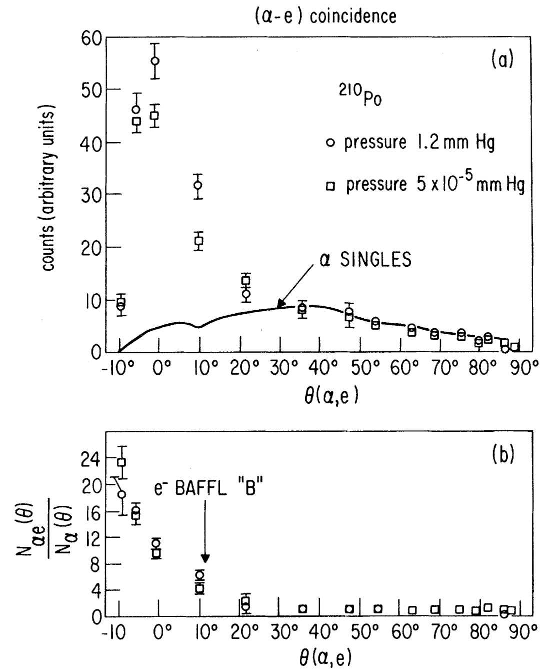


FIG. 5. (a) $(\alpha - e)$ coincidence counts as function of $\alpha - e$ angle θ in ^{210}Po decay. (b) $(\alpha - e)$ "angular correlation" as measured in ^{210}Po decay.

tive Moliere scattering back into the counter. This explanation for the forward peak was substantiated by observing the α *energy* spectrum in coincidence with the $\alpha - e$ coincidences near 0°, which we interpret as mainly scattered events. The energy of these α particles was found to be reduced by 3% (~150 keV) compared to the α's at high angles. This loss in energy can readily yield electrons (δ rays) in the energy range 5–20 keV which can be focused, and is, moreover, much greater than the loss in energy associated with L-electron ejection in the α-decaying atom, (~20 keV), so these latter α's do not produce the 0° peak in this experiment. We also noted that raising the pressure in the detector chamber to 1.2 Torr caused the peak to increase; see Fig. 5. In this case those electrons produced by α-gas molecule collisions which occur on the detector side of the e^- baffle B can also reach the detector. We also observed that the energy spectrum of electrons in coincidence with alphas at 0° showed a much more prominent low-energy component, as expected for δ-rays, than did the electrons singles spectrum, which contains a smaller δ-ray component.

α's which have interacted with an L electron in the decaying atom are equally subject to such low-angle scattering, with the electrons which result from the scattering enhancing the probability for coincidence detection to essentially the same degree as for normal (i.e., non-L-electron interacting) α's, owing to the low-electron-acceptance solid angle of the e^- aperture A, 1.3%. Therefore a correction for the observed $\alpha - e_s$ anisotropy, $W_{\alpha e_s}$, was applied to the $\alpha - e - x_L$ coincidence data, which examines only these α's.

The $\alpha - e$ coincidence rate at each angle can be expressed as

$$N_{\alpha e}(\theta) = N_0[\epsilon_\alpha(\theta)\epsilon_e P_L W_{\alpha e}(\theta) + \epsilon'_\alpha(\theta)\epsilon'_e P_s W_{\alpha e_s}(\theta)], \quad (1)$$

where P_L is the L-shell ionization probability per α decay and P_S is the probability for α-particle scattering with the emission of a δ ray. The unprimed α and electron detection efficiencies $\epsilon_\alpha(\theta)$ and ϵ_e refer to those particles which come from the α source and are not scattered. The primed efficiencies refer to the particles involved in scattering. It is convenient to define an angular distribution function for the scattered particles

$$W'_{\alpha e_s} = \frac{\epsilon'_\alpha(\theta)\epsilon'_e W_{\alpha e_s}(\theta)}{\epsilon_\alpha(\theta)\epsilon_e}$$

and to rewrite Eq. (1)

$$N_{\alpha e}(\theta) = N_0\epsilon_\alpha(\theta)\epsilon_e[P_L W_{\alpha e}(\theta) + P_s W'_{\alpha e_s}(\theta)]. \quad (2)$$

If $N_\alpha(\theta) = N_0\epsilon_\alpha(\theta)$ is the α singles rate at each angle (θ) one obtains

$$P_S W'_{\alpha e_s}(\theta) = \frac{1}{\epsilon_e}\frac{N_{\alpha e}(\theta)}{N_\alpha(\theta)} - P_L W_{\alpha e}(\theta) \approx \frac{1}{\epsilon_e}\frac{N_{\alpha e}(\theta)}{N_\alpha(\theta)}.$$

The approximation seems justified since $P_L = 8\times10^{-4}$ and $(1/\epsilon_e)[N_{\alpha e}(\theta)/N_\alpha(\theta)] = 4\times10^{-2}$ in the region of the forward peak. For angles $> 35^\circ$ we have $N_{\alpha e}(\theta)/N_\alpha(\theta) = \text{const.}$ which implies $W_{\alpha e}(\theta) \approx 1$ for this region. The factor $P_S W'_{\alpha e_s}$ is used to correct the observed $\alpha - e - x_L$ angular correlation to obtain the $W_{\alpha e_L}(\theta)$ angular correlation.

D. α-e_L-x_L angular correlation

The angular correlation function $W_{\alpha e_L}(\theta)$ between L electrons and α particles is given by

$$W_{\alpha e_L}(\theta) = C N_{e\alpha x}(\theta)/N_\alpha(\theta), \quad (3)$$

where

$$N_{e\alpha x}(\theta) = N_0\epsilon_e\epsilon_\alpha(\theta)\epsilon_x\omega_L P_L W_{\alpha e_L}(\theta)$$

is the triple $e_L - \alpha - x_L$ coincidence rate and $N_\alpha(\theta) = N_0\epsilon_\alpha(\theta)$ is the α singles rate at each angle. The constant $C^{-1} = \epsilon_e\epsilon_x W_L P_L$ is the product of detection efficiencies $\epsilon_e\epsilon_x$, the fluorescence yield $\omega_L = 0.37$, and the L-shell ionization probability per α decay $P_L = 8\times10^{-4}$. The actually observed triple coincidence rate is given by

$$N'_{e\alpha x}(\theta) = N_{e\alpha x}(\theta)W'_{\alpha x}[1 + P_s W'_{\alpha e_s}(\theta)] \quad (4)$$

and includes contributions from the instrumental $\alpha - x_L$ anisotropy $W'_{\alpha x}(\theta)$ and the δ-ray angular distribution $W'_{\alpha e_s}(\theta)$. The observed triple-coincidence rate was corrected by dividing by

$$W'_{\alpha x}(\theta)[1 + P_s W'_{\alpha e_s}(\theta)] = \frac{N_{\alpha x}(\theta)}{N_\alpha(\theta)}\left[1 + \frac{1}{\epsilon_e}\frac{N_{\alpha e}(\theta)}{N_\alpha(\theta)}\right],$$

where the correction term $P_S W'_{\alpha e_s}(\theta)$ for the contribution of δ rays from α scattering is only 17% at -9.5°, 12% at -5.8°, 3% at -2.7°, and negligible at all positive angles. Due to the small probability for L-electron ionization in α decay, 8×10^{-4}, and small efficiency for triple-coincidence detection, true coincidence rates were extremely low, about 4/h, despite the intense source and barely tolerable α singles rates.

The result of the $\alpha - e_L - x_L$ angular correlation measurement corrected for $\alpha - e_s$ and $\alpha - x_L$ assymetry is shown in Fig. 6(a). A total of 641 true triple coincidences over all angles was observed in 9147 min. The true-to-chance ratio was 6.5. The solid line represents the average of the α singles spectra, $N_\alpha(\theta)$, which were taken at regular intervals during the run in order to correct for drifts in the α detector.

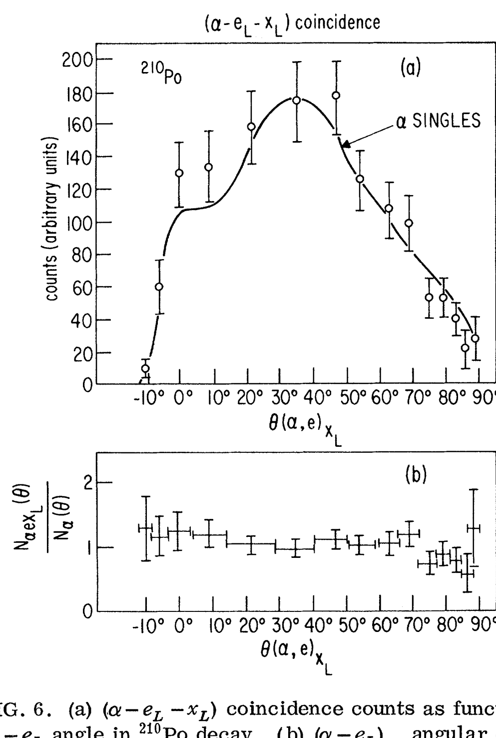


FIG. 6. (a) $(\alpha - e_L - x_L)$ coincidence counts as function of $\alpha - e_L$ angle in ^{210}Po decay. (b) $(\alpha - e_L)_{x_L}$ angular correlation in ^{210}Po decay.

The ratio of the coincidence rate divided by the singles rate at each angle is shown in Fig. 6(b). As can be seen the angular distribution is essentially isotropic in the range from 0°–90° at least within experimental error. A possible anisotropy of about 20% can, however, not be ruled out with certainty. No finite detector solid angle correction has been applied; such correction would have negligible effect on the correlation ($\Omega_e = 1.3\%$; $\Omega_\alpha \simeq 0.9\%$ per θ interval).

VI. DISCUSSION

A small anisotropy may have escaped detection in the present experiment due to possible electron scattering in the source. That such possibility exists is indicated by the attenuation of our ^{238}Pu $\alpha - e_L$ conversion electron correlation measurement, which was attenuated by ~16% compared to the expectation based on Milton and Fraser's[9] $\alpha - \gamma$ angular correlation measurements. As discussed in Sec. IV on the ^{238}Pu results, there are factors which might either diminish or increase such attenuation in the ^{210}Po case. The truly prompt electron ejection accompanying the α emission into the open hemisphere in ^{210}Po decay avoids augmented scattering effects that arise from electron emission from the deeply buried (~10 μg/cm^2) ^{234}U recoil after ^{238}Pu α emission. On the other hand, the scattering effects on electrons might be more severe because of possible local agglomeration in the autodeposited ^{210}Po source, and because the average L-electron energy is much lower, ~8 keV compared to ~24 keV. However, it is certain that an initially strongly forward peaked correlation could not have been wiped out by electron scattering in the actual ^{210}Po sample. The local sample thickness could not likely have exceeded about 10 μg/cm^2 effective ^{238}Pu source thickness that gave approximately the expected anisotropy.

The absence of a strong forward peak in the $\alpha - e_L$ angular correlation seems to indicate that there is no strong interaction between the α particle and the liberated L electron while leaving the atom. The result of this investigation together with the failure to find the predicted numbers of He$^+$ ions directly,[5] diminishes the likelihood for the validity of the hypothesis that a substantial fraction of L-electron capture into bound He$^+$ states takes place during α emission. On the other hand these results do not negate the capture idea, and some facts still suggest it.

At this point the phenomena of the missing coincidences in both the α and electron spectrometric measurements[3] in coincidence with L x rays still waits for an explanation. In the present experiment the $\alpha - x_L$ coincidence rate corresponded to a P_L value consistent with the current average value, 8.1×10^{-4}, namely $(7.9\pm0.7)\times10^{-4}$. (The δ-ray component in the $\alpha - e$ measurement and the unknown contribution to the coincidence rate by electrons ejected from the M and outer shells precludes obtaining a meaningful P_L value from $\alpha - e$ coincidences.) However, just as in the electron spectrometry experiment, where the coincidence requirement demands a free electron the present $\alpha - e_L - x_L$ coincidence rate was again very low, corresponding to a P_L value of $(3.2\pm0.8)\times10^{-4}$, only $(41\pm11)\%$ of the best value. This result is not inconsistent with the electron spectrometry experiment where only $(37\pm6)\%$ of the coincidences calculated from the observed L x-ray intensity were observed. The $\alpha - x_L$ P_L value is irrelevant to the question of possible L-electron capture, inasmuch as the alpha detector responds without discrimination between He$^+$ and He^{++}.

Essentially the same unhappy qualifications on the conclusions from this experiment as were discussed in the introduction about the He$^+$ search experiment also apply, namely, those arising from the autodeposition method of source preparation in both these experiments as contrasted to the retarded ion beam deposits used in the electron and alpha spectrometric measurements on ^{210}Po. If the autodeposited sources were agglomerated to the extent that the He$^+$ ions were converted to He^{++}, the electron transferred by collision would probably not be detectable. Even if it became a free electron its energy would be ~1.4 keV (equating electron and α speeds), far below our detection limit of 5 keV. Thus the events *initially* producing He$^+$ ions would not be detectable in $\alpha - e_L - x_L$ coincidence, so they could not affect the sought for correlation.

All these considerations make the inference of no e_L capture probable, but less than firm. They do not, however, affect the experimental fact of an *almost* isotropic angular correlation. Any theory of $\alpha - e_L$ interaction has to account for this. The only calculation we are aware of, that of Choi and Merzbacher,[11] is only partly relevant, and that calculation suggests a significant anisotropy, not surprisingly.

Angular correlations of electrons ejected from K and L shells with the direction of the incoming proton have been calculated[11] using time-dependent perturbation theory. The ratio of the number of electrons emitted in the forward direction to those emitted at 90° with respect to the incident particle may be considered a measure of forward peaking. For 4.2-MeV protons on Au this ratio varies from 2 to 5 for L electrons emitted with energies of 10 and 20 keV, respectively. As one expects, the electrons are more likely to be emitted in the projectile direction as the energy transfer increases.

As can be seen from Figs. 2 and 3 of Ref. 11 the angular distribution becomes much less forward peaked as the energy transfer decreases and the asymmetry depends strongly on the incident energy parameter $\eta_L = E_{inc}/[13.6(M/m)Z_L^2]$; M is the mass of α, m is the electron mass, and Z_L is the effective nuclear charge (screened) seen by an L electron. Thus one expects a smaller forward peak in the case of α particles incident on Pb, where $\eta_L = 0.007$ compared to $\eta_L = 0.03$ at an electron energy $W_L = 0.3$ (10 keV), shown in Fig. 2, Ref. 11, upper curve.

However, the calculation of Choi and Merzbacher for the case of target irradiation with a particle beam necessarily integrated over all impact parameters, and thus is not applicable to the case of α decay, for which the impact parameter is zero. Since energy transfer is larger at small impact parameters, forward peaking would also increase in a calculation particularized to α decay. From the strong forward peaking (2:1, 0°–90°) for the upper curve in Fig. 2 of Ref. 11, it seems very improbable that such a zero impact parameter (outgoing α wave only) calculation would not still indicate an anisotropy, in contradiction to the present result.

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[1]J. S. Hansen, Phys. Rev. A 9, 40 (1974).

[2]A. Midgal, J. Phys. (USSR) 4, 423 (1941); M. S. Freedman, Ann. Rev. Nucl. Sci. 24, 209 (1974).

[3]H. J. Fischbeck and M. S. Freedman, Phys. Rev. Lett. 34, 173 (1975).

[4]R. Watson, Phys. Rev. A 12, 2628 (1975).

[5]P. Dyer, D. Burch, H. J. Fischbeck, and M. S. Freedman, Phys. Rev. Lett. 36, 903 (1976).

[6]D. Beery, F. T. Porter, and M. S. Freedman, unpublished work on efficiency determination of NaI(Tl)-Quantacon photomultiplier detection of few keV electrons.

[7]F. T. Porter, M. S. Freedman, F. Wagner, Jr., and I. S. Sherman, Nucl. Instrum. Meth. 39, 35 (1966).

[8]M. E. Rose, L. C. Biedenharn, and G. B. Arfken, Phys. Rev. 85, 5 (1952).

[9]J. C. D. Milton and J. S. Fraser, Phys. Rev. 95, 628 (1954).

[10]We thank E. P. Horwitz for the ultrafiltration purification of the ^{210}Po solution. The micropore membrane had pore sizes of 0.04 μm. Ultrex HCl was used.

[11]B.-H. Choi and E. Merzbacher, Phys. Rev. A 1, 299 (1970).