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Internal pair conversion in heavy nuclei

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Positrons and electrons from the internal pair conversion (IPC) decay of states populated in near-barrier collisions of very heavy ions have been studied using the APEX spectrometer. Using inelastic ^{206}Pb$+^{206}$Pb scattering at 5.90 MeV/nucleon, the gamma and IPC branches of the well-known $E1$ decay of the first $J^{\pi}=3^{-}$ state were kinematically reconstructed and the IPC-to-gamma branching ratio was determined to be $\beta=(4.0\pm0.7)\times10^{-4}$. We have also performed experiments with actinide beams or targets; ^{238}U$+^{181}$Ta, ^{208}Pb$+^{238}$U, ^{208}Pb$+^{232}$Th, and ^{238}U$+^{232}$Th, which resulted in the observation of new gamma-ray transitions from high-lying states in ^{232}Th and ^{238}U. None of these were populated with sufficient strength such that IPC could produce statistically significantly peaks in positron-electron sum energy spectra. [S0556-2813(97)50106-3]

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The emission of positrons and electrons from collisions of very heavy ions has been a subject of considerable interest for more than two decades. The overall yields and the shapes of the spectra are, by now, quite well understood and are found to vary smoothly with energy and collision parameters [1]. In dramatic contrast, the reported observations of sharp lines in laboratory-frame positron [2] and positron-electron [3–5] sum-energy spectra have been puzzling, both from a theoretical and experimental point of view. The only well-known physical mechanism for producing sharp sum-energy $e^{+}e^{-}$ lines is pair conversion of a transition between two well-defined nuclear states in a source at rest; the conversion can be in the field of the emitting nucleus (internal pair conversion, IPC) or in the field of another nucleus (external pair conversion). Although a physical origin for the lines now appears unlikely [6–9], it is still of interest to determine whether any manifestation of pair conversion of discrete transitions in the colliding nuclei could be evident in the sum-energy spectra, even when kinematically corrected.

In this paper we report on the results of three separate measurements of gamma rays and positron-electron pairs made using the APEX spectrometer [10] and beams from the Argonne ATLAS superconducting accelerator [11]. A preliminary version of these results has previously appeared in a conference proceedings [12]. First, we have examined collisions of ^{206}Pb$+^{206}$Pb at 5.90 MeV/nucleon. This study was aimed at populating a single state, the $J^{\pi}=3^{-}$ level in ^{206}Pb at 2648 keV, in order to measure the IPC branching ratio of the well-known 1844 keV $E1$ $J^{\pi}=3^{-}\rightarrow 2^{+}$ transition. This measurement allowed us to confirm the response of our spectrometer to a known signal and also to demonstrate the ability to correct for the severe kinematic broadening due to the moving source. Second, we have conducted a study of inelastic excitation of ^{232}Th and ^{238}U with a ^{208}Pb beam at 5.80 MeV/nucleon using large-volume, high resolution, intrinsic germanium detectors to search for gamma-ray transitions in actinide nuclei which could have IPC branches in the energy range of interest ($E_{\mathrm{pair}}\approx 1.5$–2.0 MeV). Finally, we have collected a large data set for $e^{+}e^{-}$ pairs produced in ^{238}U$+^{232}$Th collisions at 5.90 MeV/nucleon [6] to ascertain whether IPC from such high-lying states can be observed in the reaction where sharp sum energy lines were previously reported [3–5]. No such signal is evident.

In the ^{206}Pb$+^{206}$Pb study, a 1 mg/cm^{2} target of 99.76% enrichment was bombarded with a beam of 1 pna intensity for 40 h. Gamma rays were measured, in this case, with a single 70% intrinsic germanium (Ge) detector operated in coincidence with heavy ion detectors. The Ge detector was mounted 82 cm from the target to provide good angle definition of the detected gamma rays. Twenty-six gamma-ray transitions deexciting 14 states in ^{206}Pb were identified in the Doppler corrected spectrum gated by scattered ions corresponding to near elastic ^{206}Pb$+^{206}$Pb collisions (Fig. 1). The characteristic shape of the lines in the reconstructed spectrum arises from the analysis procedure. In a symmetric or near symmetric heavy ion collision, it is not possible to determine which ion was excited and thus emitted the gamma ray. The analysis must therefore be performed twice, assuming in turn emission from each ion. This process doubles the total number of events, but correctly gives the number of events in the discrete transitions. The miscorrected events from discrete transitions appear as the bell-shaped background under each sharp line. These are particularly clearly visible near 800 keV in Fig. 1. The correctly reconstructed lines have a width of 9 keV at 1 MeV. This width is dominated by contributions from multiple scattering of the heavy ions and the ϕ angle segmentation of the heavy ion detectors. The decays from the first $J^{\pi}=2^{+}$ and 3^{-} states in ^{206}Pb dominate the spectrum. Using known matrix elements [13] the yields of most transitions were reproduced to better than 10% by calculations with the multiple Coulomb excitation code Walküre [14]. The ^{206}Pb $J^{\pi}=3^{-}$ state was populated with a measured cross section of 61±4 mb for ions scattered in the range 20°–68°. By extrapolation, assuming pure Coulomb excitation, this yield corresponds to a total population cross section of 71±5 mb (predicted 68 mb). Gamma rays from one- and two-neutron transfer reactions were also observed, but only at the 1% level compared to Coulomb excitation of the first excited state in ^{206}Pb.

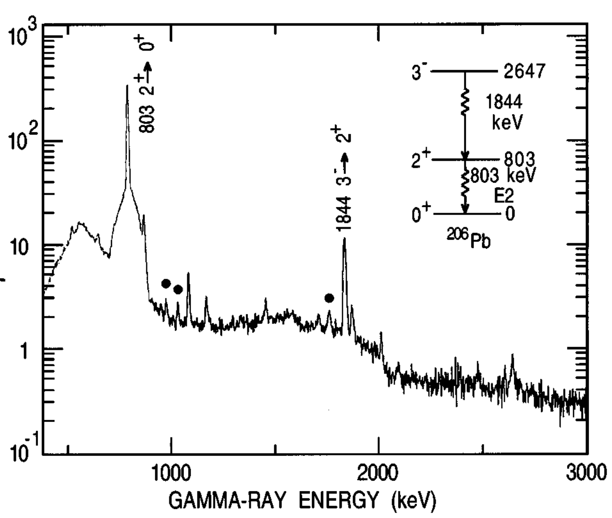

FIG. 1. Spectra from the ^{206}Pb$+^{206}$Pb reaction at 5.90 MeV/nucleon showing the Doppler reconstructed γ-ray spectrum. Transitions associated with the ^{206}Pb(^{206}Pb,^{205}Pb)^{207}Pb reaction are marked (●) and occur at a level of 1% of the ^{206}Pb($2^{+}\rightarrow 0^{+}$) Coulomb excitation. All other peaks arise from the decay of states in ^{206}Pb.

As the gamma-ray data were accumulated, positrons and positron-electron coincidences were also measured using the APEX spectrometer. The laboratory sum-energy spectrum of positron-electron pairs coincident with two heavy ions is shown in Fig. 2(a). Kinematic broadening, due to emission from ^{206}Pb ions, which have velocities up to $\beta=0.11$, results in a pair sum-energy peak for the 1844 keV transition with an expected width of 130 keV, which is barely visible in the

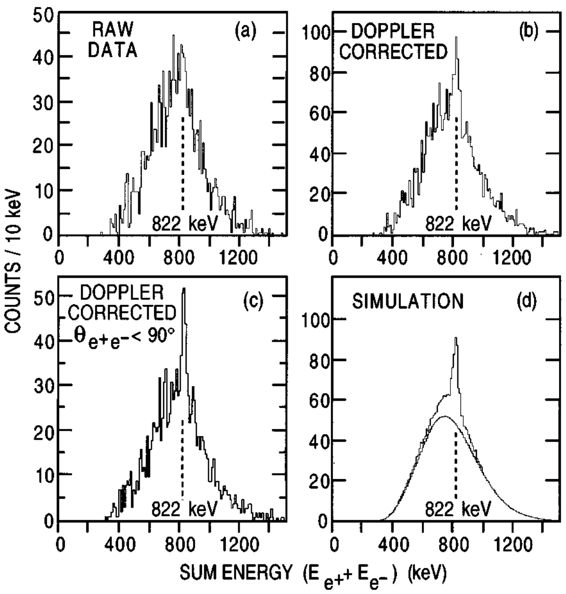

FIG. 2. Sum-energy spectra from ^{206}Pb$+^{206}$Pb at 5.90 MeV/nucleon. (a) The raw data, (b) after Doppler correction for emission from each of the scattered ions, (c) with a further selection on small positron-electron opening angles, and (d) a Monte Carlo simulation of (b).

raw sum-energy spectrum. Correction of the broadening requires knowledge of the angles and velocities of the scattered heavy ions, the positron, and the electron. Such a reconstruction of the events in Fig. 2(a) results in the spectrum shown in Fig. 2(b). A peak at 822 keV, corresponding to the pair branch of the ^{206}Pb 1844 keV transition is now clearly apparent. The width of the peak in the reconstructed spectrum is 38±3 keV. As was the case for the gamma rays, the total number of events in the corrected spectrum is double that in the raw data. Further selection of events, by choosing small opening angles between positron and electron [Fig. 2(c)] enhances the peak-to-background ratio by a factor of 2 and follows the expectations of IPC and Coulomb excitation theory. The contribution to these data from external pair production by 1844 keV gamma rays in the lead target was calculated to contribute less than 0.5% of the IPC decay. The intensity and shape of the 822 keV pair line reflects a convolution of the properties of IPC (its branching ratio, energy sharing between the positron and electron, and the relative angular distribution) with the response of the spectrometer. A Monte Carlo simulation of the 1844 keV $E1$ IPC decay from moving ^{206}Pb nuclei with the intensity and impact parameter dependence deduced from the γ-ray data and the theoretical IPC-to-gamma branch and correlations [15] has been carried out. Many detailed features of the decay, including the opening angle distribution, the forward-backward intensity, the energy sharing, the overall yield, and the observed width are all reproduced to within the statistical precision of the measurement. The efficiency of the spectrometer for the 1844 keV $E1$ IPC was determined from the simulation to be 0.33±0.04%. In addition to the many effects included in simulating radioactive source data [10], this simulation was also sensitive to the electronic timing resolution of the silicon detectors, as this is an important component of the precision of the angle reconstruction. A measured value of 3 ns, averaged over the operational detectors and lepton energies in the acceptance, was used. The peak-to-background ratio in the positron-electron sum-energy spectrum is not as good as that of the gamma-ray spectrum, as, in addition to the poorer energy resolution, there is an additional background arising from pairs originating from other production mechanisms. As the correlated peak IPC events (122±19 counts) constituted only 8% of the total number of events, this smooth continuum background can be effectively simulated [6] by selecting positrons and electrons from separate events (‘‘event mixing’’) and analyzing them in the normal fashion. This event-mixed background is shown with the expected IPC signal added in Fig. 2(d). With the simulated response, this procedure allows us to extract an IPC-to-gamma branching ratio of $\beta=(4.0\pm0.7)\times10^{-4}$ in comparison with the theoretical value [15] of 4.17×10^{-4}.

Next, we address the issue of which states in actinide nuclei may be strongly populated in low-energy heavy-ion collisions and whether they could produce peaks in the pair spectra similar to that studied in the ^{206}Pb experiment. We have searched for discrete γ-ray transitions between 1 and 4 MeV in ^{238}U and ^{232}Th in studies of ^{208}Pb+^{232}Th and ^{208}Pb+^{238}U at 5.80 MeV/nucleon, ^{238}U+^{181}Ta at 5.95 MeV/nucleon, and ^{238}U+^{232}Th at 5.90 MeV/nucleon. The ^{238}U targets were 300 μg/cm^2 UF$_4$ and the ^{232}Th and ^{181}Ta foils were 700 μg/cm^2 rolled metal. In these measurements, up to five intrinsic germanium detectors were used, each with relative efficiencies between 25% and 70%. A summary of these results is given in Fig. 3 which displays gamma ray spectra which have been Doppler reconstructed assuming emission from the actinide nucleus which, in the asymmetric cases, could be separated from the reaction partner. In the ^{238}U+^{232}Th system [Fig. 3(c)], where this is not possible, the spectrum is a composite of gamma rays from both nuclei. At the lower gamma-ray energies the ground state rotational bands are clearly visible. In order to enhance the acquisition of data for gamma rays of the highest energies, an electronic scaledown was used for the low-energy portion of the spectrum (usually by a factor of 20), which gives rise to the reduced yield in the spectrum below 600 keV [Fig. 3(a,d)] (1 MeV [Fig. 3(b,c)]).

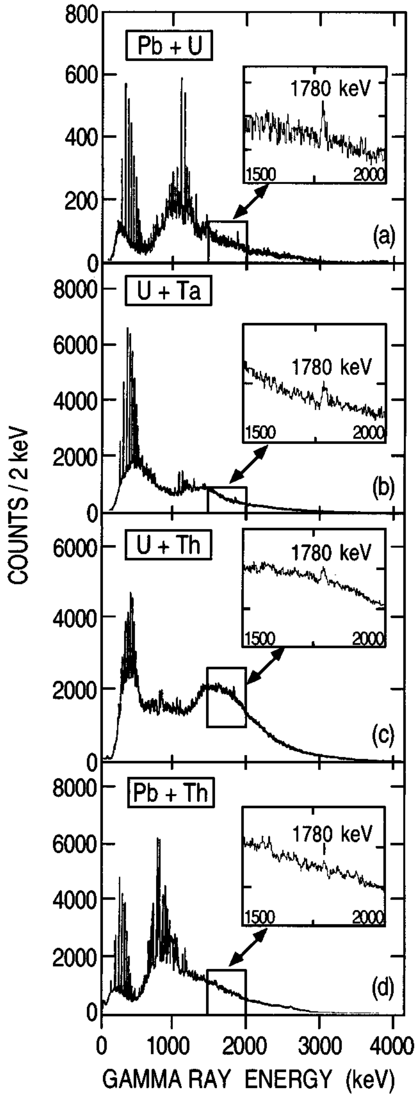

FIG. 3. Gamma-ray spectra Doppler reconstructed from heavy-ion collisions of actinide nuclei. (a) Uraniumlike events from ^{208}Pb on ^{238}U at 5.80 MeV/nucleon, (b) uraniumlike events from ^{238}U on ^{181}Ta at 5.95 MeV/nucleon, (c) uranium- and thoriumlike events from ^{238}U on ^{232}Th at 5.95 MeV/nucleon, and (d) thoriumlike events from ^{208}Pb on ^{232}Th at 5.80 MeV/nucleon. All the uraniumlike spectra show a prominent transition at 1780±2 keV.

In all cases, strong transitions were observed in the 1–2

MeV region. Of particular prominence, and interest in the present context, is a line at 1780±2 keV in ^{238}U which stands out above the continuum background. This may correspond to a line previously reported at 1764±5 keV in ^{238}U+^{181}Ta scattering [16]. This peak is shown in the insets in Fig. 3. The total population cross section for this transition varies between 27±3 and 34±4 mb, and follows the expectations for Coulomb excitation, both for impact parameter dependence and variation with beam and target. A transition at 1782 keV from a $J^{\pi}=2^{+}$ state has been reported in inelastic alpha particle scattering of ^{238}U by McGowan and Milner [17], but we calculate that their reported $E2$ matrix element would lead to a peak eight times smaller than observed in our data. Recently, a fluorescence experiment [18] has also populated a state at 1782 keV in ^{238}U, which was reported to be a dipole excitation. Both of these previous studies find strong decays to the ground state and to the first excited state. We observe only a single line, presumably the ground state transition. Thus, the structure of the states leading to the 1780 keV gamma rays observed in our measurement is unclear. One possibility is that the peak we see from ^{238}U may actually arise from several transitions of near equal energy crossing between the ground state band and a band with the same moment of inertia built on the 1782 keV bandhead. The measured width of the 1780 keV line in our data is almost a factor of 2 larger than that expected from the reconstruction procedure and also changes with heavy-ion scattering angle in a manner which would support this conjecture. Confirmation of this hypothesis would require several new and different measurements. The ^{232}Th spectrum, Fig. 3(d), shows similar intensity in the 1700–1900 keV region, but distributed in a series of smaller peaks.

We now turn to the question of the observation of sharp peaks in spectra collected following reactions of actinide nuclei. In laboratory spectra, without Doppler correction, sharp peaks are not expected, either for gamma rays or positron-electron pairs emitted from these swiftly moving ions due to the broad range of angles and recoil velocities associated with inelastic scattering as is evident from the ^{206}Pb+^{206}Pb data. The fraction of positron-electron events which are emitted with a combination of angles and energies so as to have canceling Doppler shifts is sufficiently small (3×10^{-3}) that sharp peaks from these events would not be visible in this type of experiment even if it were somehow possible to select this type of event. Inspection of the uncorrected γ-ray spectrum for sharp lines shows that less than 5×10^{-3} of the ions stop in the target.

Even in kinematically corrected spectra, IPC branches from the discrete states populated in collisions of actinide nuclei lie at the edge of our sensitivity. Compared to the ^{206}Pb +^{206}Pb experiment, the ^{238}U +^{232}Th discrete gamma-ray photopeak to background is much worse. In the case of ^{206}Pb+^{206}Pb collisions we observe the 1844 keV γ-ray transition with a peak-to-background ratio of 4:1 (Fig. 1) whereas in the case of ^{238}U+^{232}Th the 1780 keV transition has a peak-to-background ratio of 1:6 [Fig. 3(c)]. Both peak and background are comprised of gamma rays which can create pairs, therefore the degradation of peak to background in the gamma spectrum of a factor $\sim$24 should translate directly to the IPC component of the pair spectrum. In the pair spectrum, the peak-to-background ratio is actually far worse than this, due both to poorer energy resolution and to the large additional contribution from the combination of dynamic positrons and δ electrons which give rise to an additional continuum background. Between the cases of ^{206}Pb+^{206}Pb and ^{238}U+^{232}Th, the contribution of positrons induced by the dynamics of the collision process rises by a factor of 8 causing a similar rise in the contribution from uncorrelated pairs. Figure 4(a) shows the kinematically corrected positron-electron spectrum from ^{238}U +^{232}Th collisions, and Fig. 4(b) shows those events selected according to the same criterion which produced the cleanest line in the ^{206}Pb experiment, that is, those with small positron-electron opening angles. Converting the measured gamma yield into the expected pair yield by assuming the 1780 keV transition to be an $E1$ transition (the case with the largest pairs branch), indicates an expected peak with 80 counts with a width of 40 keV. This corresponds to a signal at the 1σ level in our data. A recent calculation of $M1$ transitions [15] indicates that the positron-electron angular distribution is more isotropic than for $E1$ decays, but that the conversion coefficient is slightly lower. Overall these effects reduce our sensitivity to $M1$ IPC pairs by about 30%, as compared to an $E1$ transition. All higher multipoles have IPC-to-gamma branches which are smaller than for $E1$ decays, so would be even more difficult to observe.

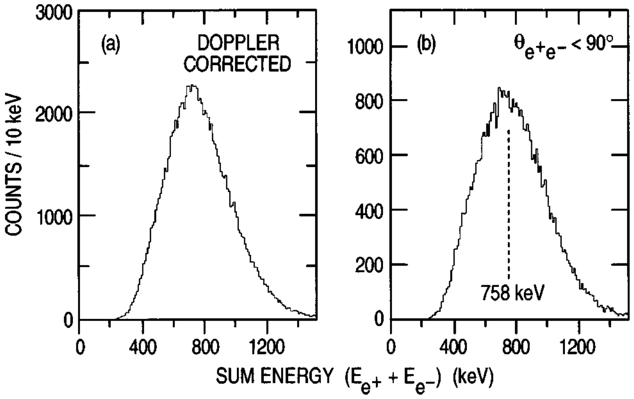

FIG. 4. Doppler reconstructed sum positron-electron spectrum from ^{238}U+^{232}Th at 5.95 MeV/nucleon. (a) Shows the total data and (b) those events with a positron-electron opening angle of less than 90°, a selection which enhances $E1$ IPC.

Finally, we should briefly mention the possibility of the population of excited states which do not have a gamma decay, but still produce pairs (e.g., excited spin 0 states). In this case, it is more difficult to reach definitive conclusions as there is less to measure in experiment. It is clear that the absence of an electromagnetic decay matrix element would also tend to suppress the Coulomb excitation of such states. Further, strong multistep electromagnetic excitation, most probably through the first excited state, would also result in observable deexcitation gamma decays through the same path, which we should see. We have performed a series of model dependent calculations based on the known systematic properties of actinide nuclei in order to estimate the population of spin zero states at about 1.8 MeV in ^{238}U by Coulomb multistep excitation. We find the population of these hypothetical states to be less than a few millibarns. As the decay of spin zero states in very heavy nuclei is dominated by internal conversion (IC), with an IC-to-IPC branching ratio of more than 400:1 [19], the pairs cross section could never be more than 1 μb/sr averaged over the solid angle of our heavy-ion counters. Without kinematic correction, a transition of this intensity emitted from a fast-moving ion could never result in an observable peak. Even with kinematic correction, this level of cross section is at the limit of our sensitivity.

In conclusion, we have used the APEX spectrometer to study IPC from fast moving heavy ions in collisions near the Coulomb barrier. In the case of ^{206}Pb$+$^{206}Pb, the IPC branch of an $E1$ transition was reconstructed and the branching ratio measured. New high lying states were found in ^{232}Th and ^{238}U, but were not populated with sufficient cross section to result in statistically significant peaks in the kinematically corrected positron-electron spectra, and not at all in the laboratory spectra. We therefore conclude that the previously reported sharp sum-energy peaks do not arise from any nuclear source. More precise tests of IPC theory in high-Z nuclei, especially of magnetic transitions, appear to be both desirable and possible, although light ion reactions or source measurements may prove more promising to reach an interesting level of precision, as they avoid the high positron and electron backgrounds encountered in heavy-ion collisions.

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