Correlated Two-Photon Lines from 6-MeV/nucleon U + Th, U + U, and Th + Th Collisions

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We find that narrow lines at 1043 and 1062 keV in the summed-energy, 180°-correlated two-photon spectrum from ≈ 6 -MeV/nucleon U+Th collisions can be produced by cascades from high-spin states (32⁺) in ²³⁸U. We see no evidence for narrow two-photon lines in U+Th, Th+Th, and U+U collisions corresponding to the electron-positron lines seen by others: In the summed-energy region between 1.2 and 2.0 MeV, we set an upper limit for the cross section of 6×10^{-31} cm² averaged over a target thickness of 1 mg/cm².

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Results with the EPOS spectrometer at Gesellschaft für Schwerionenforschung, Darmstadt (GSI) have revealed correlated equal-energy electron-positron lines at 615, 750, and 810 keV sum-energy in U+Th collisions between 5.8 and 5.9 MeV/nucleon, and in U+Ta collisions between 5.9 and 6.4 MeV/nucleon.¹ The Orangespectrometer group has investigated the momentum correlation of coincident e^+e^- pairs in U+U and U+Pb collisions at 5.9 MeV/nucleon and found evidence for 180° correlation for lines at 810 keV and possibly at 630 keV, but not at 750 keV.^{2,3} It has been hypothesized that these results might be explained by the formation and decay of a new light neutral system which has two or three different states of excitation near 1640, 1772, and 1832 keV.⁴⁻⁸ It has been noted that these states might also decay into two or three photons, depending on their spin and parity.⁹

Recently we have reported the observation of a narrow line (width 3.4 keV) at 1062 keV in the summedenergy, 180° -correlated two-photon spectrum from 5.95-MeV/nucleon U+Th collisions.¹⁰ This could be interpreted as the two-photon decay of yet another state of the hypothesized neutral system moving with the centerof-mass (c.m.) velocity of the collision partners.

We have extended our measurements to the collision system U+U and Th+Th and have considerably improved the statistics for U+Th. We find that the narrow-peak structure at 1062 keV in U+Th can be explained by a cascade of Coulomb-excited transitions starting at high spin (32^+) in ²³⁸U projectiles. The summed-energy line attains a narrow width because Coulomb excitation of the very-highest-spin states requires nearly head-on collisions which result in much lower average velocities than one would naively expect. Nevertheless, the velocities are high enough to wash out any structure in the singles spectrum. No structure corresponding to the GSI e^+e^- peaks is visible in our data.

The present experimental arrangement is similar to the one described in Ref. 10, but we used eighteen Ge detectors instead of fourteen. The Lawrence Berkeley Laboratory SuperHILAC delivered typical beam currents of 75-nA U^{42+} and Th³⁷⁺. In order to eliminate rapid target deterioration in these intense beams, five targets each were mounted on a rotating target wheel. The wheel was synchronized with the beam such that each 4.5-ms-long beam pulse would sweep over a 4×20mm² target area.¹¹ We used 1-mg/cm² targets of rolled thorium and evaporated metallic uranium.¹² The U targets were protected from oxidation by a $20-\mu g/cm^2$ carbon overcoating on each side. Target wheels were changed every 36 h. Examination of the irradiated targets with a microscope and an α -particle gauge revealed some degradation of homogeneity and thickness in the U targets.¹³

During a total of seven weeks of beam time, we investigated the collision systems U on U at 5.95 MeV/ nucleon (65 mC integrated beam charge), Th on Th at 5.75 MeV/nucleon (26 mC), Th on Th at 5.93 MeV/nucleon (30 mC), U on Th at 5.95 MeV/nucleon (63 mC), U on Th at 5.85 MeV/nucleon (12 mC), and U on Th at 6.05 MeV/nucleon (26 mC). The beam energy was checked with a phase probe, and with a crystal detector placed both before and after the target. The typical beam energy spread was 1%. The typical duty cycle of the accelerator was 15%, giving instantaneous counting rates in the individual Ge detectors of $\approx 3 \times 10^4$ s⁻¹. All coincidences between any two or more detectors were recorded in event mode. A sorting program selected coincidences between any forward detector and its 174°-correlated partner as well as the mirror partner of the latter, reflected in the plane containing the beam. In the discussion, the former coincidences are called correlated and the latter uncorrelated.

The kinematics of a two-photon decay of a moving system is explained in Ref. 10. If E_1 and E_2 are the laboratory photon energies of two 180°-correlated equalenergy γ rays emitted by the system, the expected counts lie in a wedge-shaped window in the (E_1, E_2) plane. A projection of this wedge onto the diagonal of the (E_1, E_2) plane produces the summed-energy spectrum. Coincident equal-energy photons that are exactly 180° correlated in the c.m. system of the emitter would show up as a narrow line (FWHM \approx 3 keV) in this spectrum, because the first-order Doppler shifts cancel exactly event by event. Only a small second-order Doppler effect and the detector resolution would contribute to the linewidth. For photons that are not exactly 180° correlated, the Doppler shifts in the opposing detectors would cancel only partially because of the 22° total opening angle of the individual detectors, and the summed-energy line would be broadened.

The correlated summed-energy spectra for the collision systems U+Th, U+U, and Th+Th are shown in Figs. 1(a)-1(c), respectively. Our new U+Th data are statistically consistent with the previous data, and we show the sum of all data in Fig. 1 to improve statistics.

At energies less than 1 MeV, the U+Th spectrum in Fig. 1(a) shows a series of broad peaks with a width of about 10 keV. Each of these summed-energy lines can be accounted for by cascades from Coulomb-excited high-spin states (14⁺, ..., 28⁺) in 238 U and 232 Th. The energies agree closely with those found in the literature. 14,15 (Some of the corresponding broad single lines can be identified in our single-photon spectra below 500 keV.) A narrow (3-keV) line at 1022 keV is due to the two-photon annihilation of positrons produced in the collision. The 3.4-keV-wide line at 1062 keV seen in our last experiment is reproduced, but with an increased area of ≈ 400 counts, roughly consistent with the increase in integrated beam charge. It has a statistical significance of 9 standard deviations above background. An additional narrow line at 1043 keV with an area of 250 counts has a statistical significance of only 5.5 standard deviations. None of these narrow lines show up in the uncorrelated spectra. There is also no trace of any corresponding component line in the single-photon spectrum.

The U+U spectrum in Fig. 1(b) shows a series of broad peaks at energies < 1 MeV. At higher energies, no narrow line, other than that from positron annihilation, is obvious in this spectrum. Instead we see two



FIG. 1. 180° -correlated summed-energy spectra for the collision systems (a) U+Th, (b) U+U, and (c) Th+Th.

broad structures around 1060 and 1040 keV. In the Th+Th spectrum in Fig. 1(c), broad Coulomb-excited lines again dominate the low-energy part. Again, no narrow structure other than from positron annihilation is obvious, but there appears to be a broad structure around 1055 keV.

The statistics of our data are good enough to sort the counts in the U+Th 1062-keV line on the difference energy $E_1 - E_2$ for each detector angle separately. The corresponding spectra for 43° and 65° detectors are shown in Figs. 2(a) and 2(b), respectively. Superimposed are two Monte Carlo simulations. One simulation is for a distribution expected for the two-photon decay of a neutral object moving slowly with respect to the c.m. of the heavy-ion collision, as described in Ref. 10. The widths and peak positions of the experimental distributions are close to the simulations, but the experiment shows an additional hump for negative energy differences that cannot be explained by a two-photon decay. This hump was not visible in last year's data because of the poorer statistics.

For the other simulation, the Rochester Coulombexcitation code¹⁶ was used to calculate the excitation probabilities for ²³⁸U and ²³²Th states with spin J > 20, using E2 matrix elements derived using the rigidspheroidal-rotor model and the known B(E2) values for the low-spin states. We then used these probabilities and the level energies from Refs. 14 and 15 in a relativistic Monte Carlo simulation of our collision systems and ex-



FIG. 2. Difference-energy distribution of the number of counts in the 1062-keV peak in Fig. 1(a). Superimposed are Monte Carlo simulations of a two-photon decay (dashed line), and of a cascade of deexcitation transitions following Coulomb excitation (solid line). (a) 43° detectors; (b) 65° detectors.

perimental conditions. The energies, shapes, and relative intensities of the cascade sum-energy lines below 1 MeV are reproduced for all three of our systems, if we correct the transition energies for the highest-spin states by a few keV. As an example, in Fig. 3(a) we show the shape of the 950-keV line in U+Th, which is created by a coincidence of the transitions $26^+ \rightarrow 24^+$ and $24^+ \rightarrow 22^+$ in ²³⁸U with energies of 484 and 466 keV. Both experiment and simulation produce lines of typically 10keV width.

No measurements for transitions from states higher than 30^+ in ²³⁸U have been published up to now. Assuming a reasonable energy of 543 keV for the 32^+ $\rightarrow 30^+$ transition in ²³⁸U in coincidence with the 519keV, $30^+ \rightarrow 28^+$, U line would yield a summed-energy line at 1062 keV. Figure 3(b) shows that for such a line the simulation for U projectiles on a Th target predicts a narrow line of 3.4-keV width, in excellent agreement with experiment. The other narrow line in the U+Th spectrum, at 1043 keV, is then due to a cascade in the ²³⁸U projectile, also starting at the 32^+ level, giving coincidence of 543-keV, $32^+ \rightarrow 30^+$ and 500-keV, $28^+ \rightarrow 26^+$ transitions.

The narrow width of the 1062-keV line in U+Th is a consequence of the kinematic conditions necessary for its excitation. From the Coulomb-excitation code one finds that U projectiles have to be scattered into laboratory angles between 55° and 80° to excite the 32^+ level, with resulting velocities of v/c=0.065 to 0.02. At these ve-



FIG. 3. Summed-energy line shape of 238 U cascade lines from experimental U+Th collisions (solid line) and simulation (dashed line). (a) $26^+ \rightarrow 24^+ \rightarrow 22^+$ at 950 keV; (b) $32^+ \rightarrow 30^+ \rightarrow 28^+$ at 1062 keV.

locities, the overall Doppler effects are large enough to wash out any line structure in a single-photon spectrum or in a sum-energy spectrum that is not correlated close to 180° . For opposing detectors, however, the velocities are small enough to produce a 3.4-keV sum-energy linewidth. (The angular correlation of two cascading quadrupole transitions favors 180° emission, but only by 20%.)

In the 1062-keV line, the emitted photons are not of equal energy, but their energy difference is just correct for the counts to appear in our wedge cuts originally chosen to pass equal-energy photons from a faster-moving source. Figures 2(a) and 2(b) show the simulated $E_1 - E_2$ distributions, at detector angles of 43° and 65°, respectively. Since either photon can go into the E_1 (or E_2) detector, there is a double-humped distribution. The experimental double-hump structure is reproduced, although not exactly, perhaps because of systematic errors in the background subtraction.

For the symmetric U+U system, the 1062-keV cascade can be emitted by target or projectile ions. For the close collisions necessary for excitation of the 32^+ level, the target ions are recoiling rapidly, producing a broad sum-energy line superimposed on the narrow projectile line. The simulation predicts an average linewidth of 10 keV, which corresponds approximately to the broad structure in the experimental U+U spectrum in Fig. 1(b). A similar effect occurs in the Th+Th spectrum.

The absence of a narrow 1062-keV line in U+U and



FIG. 4. Summed-energy spectrum obtained by summing data for all collision systems. Error bars correspond to 3 standard deviations. The energies corresponding to the GSI e^+e^- lines are marked by arrows.

Th+Th, as well as the double-hump structure of the $E_1 - E_2$ distribution of the 1062-keV line in U+Th, convinces us that this line cannot be due to the two-body decay of a neutral object produced in these collisions. The question then arises whether there is any indication for a two-photon decay at energies corresponding to the GSI e^+e^- lines. In Fig. 4, we show a summed-energy spectrum that was obtained by summing all data for all the collision systems used. The energies of the e^+e^- lines are marked by arrows. In the energy range above 1062 keV we do not see any statistically significant narrow structure in any of our collision systems. Correcting for absorption in the Pb absorber, for the solid angle, and for the efficiency of the detectors, we arrive at an upper limit (1 standard deviation) for the yield of any 4-keV-wide correlated two-photon line of $2 \times 10^{-12} \gamma \gamma$ decays per incident nucleus in a 1-mg/cm²-thick target, or an average cross section of 6×10^{-31} cm². This has to be compared with the yield of the e^+e^- lines in similar reactions. For the singles positron lines the EPOS group quotes a cross section of 10 μ b/sr.¹ They also state that the coincidence e^+e^- lines account for the full singles cross section. The Orange group obtains a value of 14 μ b/sr for the singles positron lines in U+U,² but their coincidence yield seems to be smaller.³ Assuming isotropic $e^+e^$ emission and integrating over the heavy-ion solid angle, we derive from these numbers a total production cross section for the e^+e^- lines of approximately 2×10^{-28} cm² for our systems. If there is only one $\gamma\gamma$ branch and if the full target thickness contributes, we can set an upper limit for the ratio R of the $\gamma\gamma$ decay branch to the e^+e^- decay branch of the presumed neutral particle of 3×10^{-3} . This can be compared to the theoretical absolute limits, $1 \times 10^{-6} < R < 6 \times 10^{5}$ for a scalar particle and $1.6 \times 10^{-5} < R < 7 \times 10^{4}$ for a pseudoscalar particle. 17

In conclusion, we have shown that narrow lines in the summed-energy spectrum can be produced by deexcitation cascades after Coulomb excitation of high-spin states. We have set limits on the $\gamma\gamma/e^+e^-$ branching ratio of a presumed neutral particle that exclude a range of 8 orders of magnitude out of 10 possible for a pseudoscalar and 9 out of 12 for a scalar particle.

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