Bremsstrahlung in α Decay of ^{210}Po: Do α Particles Emit Photons in Tunneling?

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Emission probability of bremsstrahlung in the α decay of ^{210}Po was measured in α-γ coincidence measurements with Si and Ge detectors. It was found that the bremsstrahlung yields are much smaller than those predicted by a Coulomb acceleration model, in which γ rays are emitted during acceleration outside the barrier. This suggests that the radiation amplitude in the barrier cannot be neglected, and the discussion based on a quasiclassical approach is given. [S0031-9007(97)03609-0]

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Bremsstrahlung is a continuous-energy electromagnetic radiation which accompanies any interaction of charged particles, and its strength depends strongly on the acceleration. Thus, continuum γ ray emissions have been widely investigated to obtain detailed information on dynamics in various nuclear reactions. One can expect bremsstrahlung emission in a nuclear decay as well, since it is usually accomplished with separation and acceleration of charged particles in a very short time period. Except for bremsstrahlung in β decay, however, no observation of those associated with the decay of a nucleus has been reported until quite recently. Although a possibility of the bremsstrahlung associated with spontaneous fission of ^{252}Cf was reported by Kasagi *et al.* [1], the subsequent experiment performed by Luke *et al.* [2] gave null results and showed that the upper limit of the bremsstrahlung is much smaller than reported in Ref. [1].

The bremsstrahlung associated with α decay is also of interest, since an α particle emitting a photon must pass through a large Coulomb barrier; it is the so-called tunneling effect which can only be understood by the wave nature of the particle. In the classical picture, the α particle is accelerated outside the Coulomb barrier and, there, can emit the bremsstrahlung photon. Nevertheless, a theoretical consideration [3] of the radiation of the quasiclassical tunneling motion suggested appreciable radiation in the barrier region in the α decay. It is, therefore, of particular interest to ask whether the photon can be emitted only during acceleration in the Coulomb field or also in tunneling. Recently, D'Arrigo *et al.* [4] reported bremsstrahlung associated with α decay of ^{226}Ra and ^{214}Po. They obtained the emission probability for $300 < E_\gamma < 450$ keV to be about 4×10^{-10} and 3×10^{-10} keV^{-1} decay^{-1} sr^{-1} for ^{226}Ra and ^{214}Po, respectively. These values are much larger than those predicted with a Coulomb acceleration model in which α particles are accelerated outside the Coulomb barrier, but are very close to those predicted with a sudden acceleration model in which the α particle is instantaneously accelerated at its final kinetic energy. This is rather unexpected since a naive interpretation of the results requires that most of the photons should be emitted in the barrier.

We have also been investigating bremsstrahlung emission associated with the α decay of ^{210}Po and ^{244}Cm, using Ge detectors for γ-ray detection. Our results, however, are at least 1 order of magnitude smaller than that of D'Arrigo *et al.* [4]. Moreover, they are smaller than the prediction of the Coulomb acceleration model. In this Letter, we present the result of ^{210}Po, for which the measurements have been performed for more than 250 days, and discuss the bremsstrahlung in tunneling motion.

Measurements of γ rays in coincidence with α particles were performed in two phases with different sources. For the first phase, a ^{210}Po source of 3.47 kBq was used for the measurements of about 120 d. The measurements of the second phase were carried out subsequently with a source of 11.7 kBq for about 150 d. Both sources were commercially obtained from the Amersham Buchler Co. The size of the weaker source is 3 mm diameter and the other is 10 mm diameter, and they are mounted on a thin nickel circular plate. The detector used to measure α particles was a Si surface barrier detector of a transmission type (150 μm depletion layer and 200 mm^2 in area). The source and the detector were placed in a small vacuum chamber, and the distance between them is only 4.5 mm. For the first phase measurements, an n-type Ge detector with a thin Be window was used for γ-ray detection, and it was placed outside the chamber at about 20 mm from the source. For the second phase, two detectors of the same type were used to increase the statistics: the added one was placed in the opposite side of the chamber. The size of the Ge crystal is 43 mm in diameter and 55 mm in thickness. The Ge detector was placed in such a way that its center is set at 25° with respect to the center of the Si detector. The solid angles subtended by the detectors were quite large, so that the effect of the α-γ angular correlation was drastically reduced.

Efficiency calibration of the Ge detector was made with a mixed calibration source (Amersham, QCD.1). Yields of γ-ray peaks for $88 < E_\gamma < 1333$ keV were measured in the same arrangement, so that the absolute detection efficiency including the effect of absorption of various materials between the source and the Ge detector was obtained. Energy resolution of γ rays was 2.0 keV for 1.33 MeV. Energy calibration for α particles was made by using α-ray peaks observed in the coincidence measurements with the ^{244}Cm source as well as ^{210}Po. Energy resolution of α particles was 70 keV, for the first phase, and it was improved in the second phase to be 40 keV.

Signals from each preamplifier were fed into a timing filter amplifier (TFA) as well as a spectroscopy amplifier (SA). The timing information was obtained from a constant fraction discriminator following the TFA. A time-to-amplitude converter (TAC) was used to obtain the time spectrum between the α-particle and γ-ray events. Outputs of two SAs and a TAC were fed into a CAMAC analog-to-digital converter. Data were accumulated in event-by-event mode and written on a disk for off-line analyses. Singles spectra and scatter plots of the two-dimensional spectra (E_α vs E_γ) were displayed during the run to monitor the measurement. Yields of α particles were accumulated with a scaler during the coincidence measurements. The number of observed α decays was 3.29×10^9 for the first phase and 1.1×10^{10} for the second phase.

Figure 1(a) shows an example of α-γ TAC spectra. Because of real coincidences of α particles with the 803 keV decay γ rays, a prominent peak is observed in the time spectrum; the time resolution was about 15 ns. Prompt coincidence events were selected by setting a gate indicated with solid vertical lines on the time spectrum (prompt gate). Chance events were also selected by setting other gates (chance gate) as shown with dashed lines. Examples of two-dimensional scatter plots of E_α vs E_γ are also shown in Fig. 1: Figs. 1(b) and 1(c) are the plots for the prompt and the chance gates, respectively. Broad horizontal bands correspond to discrete α-particle decays, and sharp vertical lines correspond to the full energy peak of γ-ray decays. In the prompt spectrum [Fig. 1(b)], events due to the α decay to the 803 keV state and due to the x-ray emission associated with the α decay are seen in addition to the chance events. Bremsstrahlung events associated with the α decay to the ground state are clearly seen as a diagonal line satisfying the condition of $E_\gamma + E_\alpha = E_\alpha^{g.s.}$, especially for the low energy region ($E_\gamma < 300$ keV). Considering the energy resolution of the Si detector, we draw two parallel solid lines in the spectra, between which the bremsstrahlung are expected. The events between the parallel solid lines were projected onto the E_γ axis and are shown in Figs. 2(a) and 2(b), respectively, for the prompt and the chance gates. In Fig. 2(a), discrete transitions are observed in addition to continuously distributed events; they are the 803 keV γ ray and K_α and K_β x rays. The observed continuum events were not abundant. Therefore, they were binned with a rather wide width in order to get necessary statistics; $\Delta E_\gamma = 30$ keV for $100 < E_\gamma < 220$ keV and $\Delta E_\gamma = 120$ keV for 220 keV $< E_\gamma$. Since the events above 220 keV seem to contain unknown backgrounds, we estimated them by counting the events above and below the parallel lines in Fig. 1(b) and subtracted them. The events above 580 keV were excluded, because they are mainly due to the Compton events of the 803 keV γ rays. The chance coincidence events were also subtracted for each bin to deduce the subtracted yield Y_0. The corrections for the detection efficiency and for the angular correlation were made as follows. For the detection efficiency of the Ge detector, the average value of the efficiency $\langle \varepsilon_d \rangle$ was employed for each bin. The correction for the α-γ angular correlation was made with an assumption of pure dipole radiation. Thus, the corrected yield Y_c for 4π emission was obtained as $Y_c = (Y_0/\langle \varepsilon_d \rangle)/[1 + Q_2^{\mathrm{Si}} Q_2^{\mathrm{Ge}} A_2 P_2(\cos 25°)]$. Here, P_2 is the second order term of the Legendre polynomial, and A_2 is the α-γ angular correlation coefficient; $A_2 = -1.0$ for pure dipole radiation. Q_2^{Si} and Q_2^{Ge} are the geometrical attenuation factors of the Si and Ge detector. The photon emission probability per unit solid angle was finally deduced by dividing $Y_c/4\pi$ by the total number of α particles detected with the Si detector. The result is shown in Fig. 3. Associated errors show only statistical ones, and the horizontal bars indicate the bin width. As can be seen in Fig. 3, the deduced bremsstrahlung emission probability is

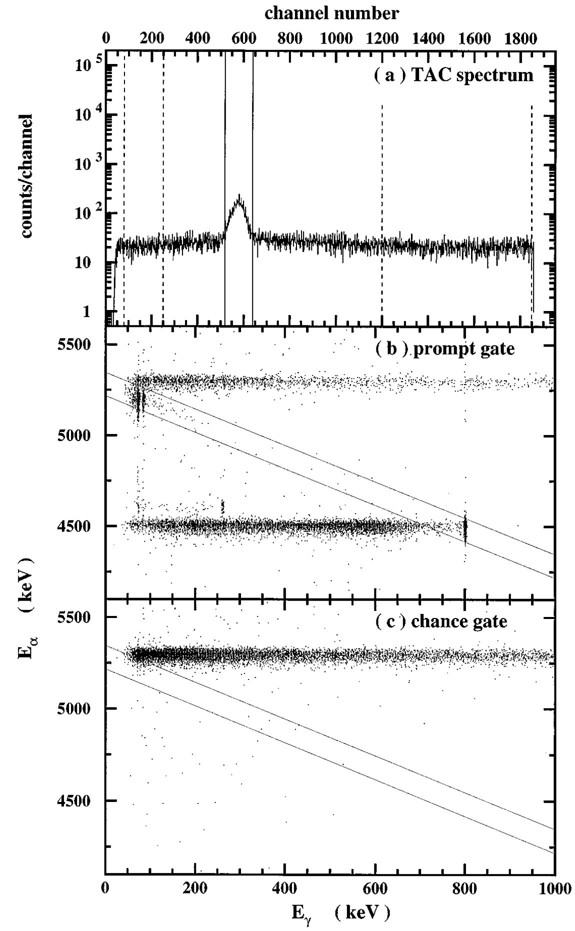

FIG. 1. (a) TAC spectrum of α-γ coincidence measurements in α decay of ^{210}Po. The prompt gate is indicated with solid lines and the chance gate with dashed lines. (b) Two-dimensional scatter plots of E_α vs E_γ for the prompt gate. (c) Same as (b) but for the chance gate.

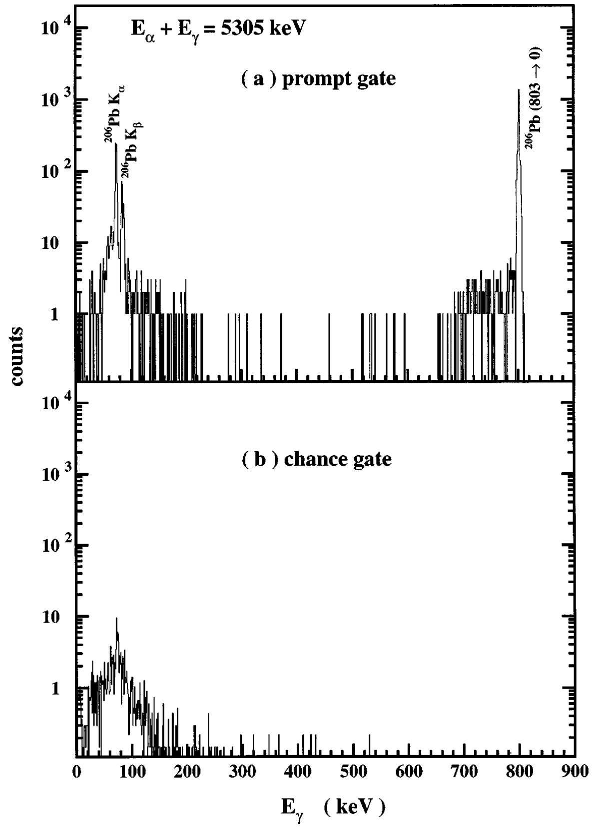

FIG. 2. Projected γ-ray spectra of the events between two parallel lines in Fig. 1; (a) for prompt gate and (b) for chance gate.

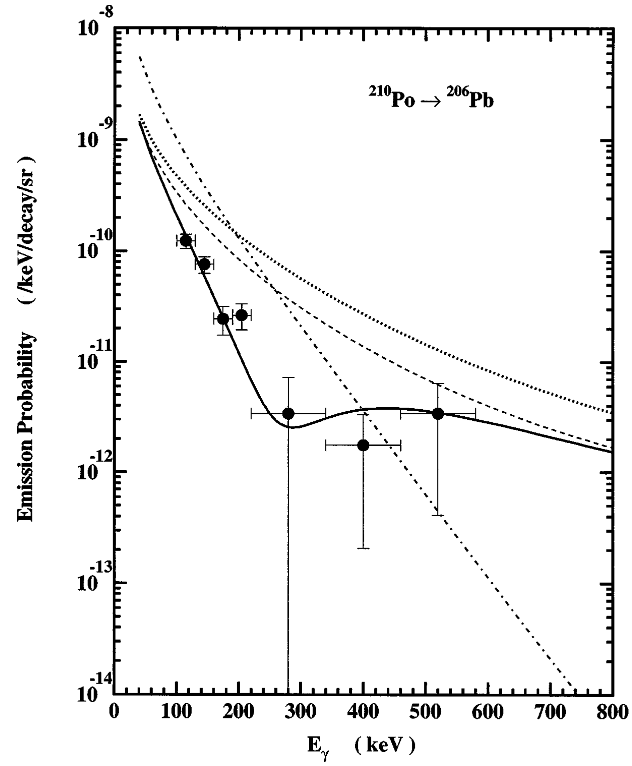

FIG. 3. Emission probability of bremsstrahlung photons in α decay of ^{210}Po. Result of the classical calculations is plotted with the dotted line for the simple Coulomb acceleration, and with the dashed line for the strict Coulomb acceleration. The result of the full quasiclassical calculation is indicated with the thick solid line. The dot-dashed line corresponds to the situation in which an α particle terminates its motion at the outer edge of the Coulomb barrier.

quite small, less than 10^{-11} keV^{-1} decay^{-1} sr^{-1} for $E_\gamma \sim 200$ keV; almost 2 orders of magnitude smaller than reported in Ref. [4]. It decreases monotonically as the photon energy increases up to $E_\gamma \sim 400$ keV.

In order to discuss the validity of the present analyses, nuclear transitions following the α decay of ^{244}Cm were analyzed. The deduced α-decay branching ratios to the 294, 597, 861, and 900 keV states in ^{240}Pu are $(3.42 \pm 0.09) \times 10^{-5}$, $(4.2 \pm 0.9) \times 10^{-7}$, $(1.42 \pm 0.16) \times 10^{-6}$, and $(4.9 \pm 0.8) \times 10^{-7}$, respectively. They agreed very well with those compiled in Ref. [5]. The γ-decay branching ratios from the same states were also compared with those of the compilation; they again agreed with each other within 10%. It should be noted that the examined transitions include an E1 transition from $J^\pi = 1^-$ to $J^\pi = 0^+$, which gives the same angular correlation as the dipole bremsstrahlung radiation. Slightly larger discrepancies were found for the transitions in ^{206}Pb. The deduced α-decay branching ratio to the 803 keV state of ^{206}Pb is $(8.83 \pm 0.47) \times 10^{-6}$, which is slightly smaller than reported in Ref. [6]. However, the K-electron ejection probability was deduced to be $(2.28 \pm 0.20) \times 10^{-6}$, which is larger than reported in Ref. [7]. Thus, we consider that this discrepancy in the ^{210}Po decay is not caused by a systematic error in the present measurement, and the uncertainty of the emission probability is estimated to be less than 10% except for statistical errors which are already associated with the data in Fig. 3.

We performed calculations based on the classical radiation theory [8] for an accelerated charged particle. In the nonrelativistic limit, $\beta \ll 1$, the intensity of the emitted radiation can be expressed as

$$\frac{d^2N(E_\gamma)}{dE_\gamma\, d\Omega} = \frac{\alpha}{4\pi^2 E_\gamma} \cdot \left| \int_{-\infty}^{+\infty} dt \sum_{i=1}^{2} [\vec{n} \times (\vec{n} \times \dot{\vec{\beta}}_i)] \cdot z_i \exp(-i\omega[t - \vec{n} \cdot \vec{r}_i/c)] \right|^2, \quad (1)$$

where α is the fine structure constant, the index i refers to two point charges (α particle and daughter nuclei), $\vec{n}$ is the unit vector along the radiation emission axis, z_i is the atomic number, and $\dot{\vec{\beta}}_i$ is the acceleration of the charge divided by the light velocity. The photon emission probability of the Coulomb acceleration can be obtained by calculating Eq. (1) numerically. The integration in Eq. (1) requires the time history of the two charged particles. We assumed that the acceleration begins ($t = 0$) from the classical turning point. In the tunnel, where the classical motion is forbidden, we simply assumed $\dot{\vec{\beta}}(t) = 0$ (for $t < 0$). After the acceleration started ($t > 0$), $\vec{r}(t)$ and $\dot{\vec{\beta}}(t)$

were determined by solving the differential equation of the motion for the two particles under the Coulomb force. The result is shown with the dotted line in Fig. 3. Clearly the calculation overestimates the data by 1 order of magnitude. This large discrepancy is a surprise, because the bremsstrahlung emissions in the Coulomb scattering, i.e., the low energy data of the $^{12}C(p, p\gamma)^{12}C$ reaction [9], are well explained by the same calculations. As Luke *et al.* discussed in Ref. [2], a simple integration for the Fourier transform in Eq. (1) may give an upper limit of the emission probability, because of the lack of a constraint of the energy conservation. Thus, we calculated the time integration so as to satisfy the condition, $\hbar\omega < Q_\alpha - V_c(r)$, at any instant of the acceleration period, where Q_α is the Q value of the α decay and $V_c(r)$ is the Coulomb energy at relative distance r [10]. The result is shown with the dashed line in Fig. 3. As expected, the emission probability is substantially suppressed for higher energies. Nevertheless, it still overpredicts the experiment. Thus, we conclude that the observed bremsstrahlung in the α decay cannot be explained by the Coulomb acceleration calculations.

The fact that the Coulomb acceleration model can explain the bremsstrahlung emissions in the Coulomb scattering but fails to explain those in the α decay suggests that the radiation during the tunneling process is not negligible. The situation will be fully understood with a quantum mechanical treatment, in which a photon creation amplitude is calculated for a transition from a decaying state to the continuum states. However, an accurate calculation is difficult because of the nonconverging nature of the wave function of the decaying state, and a new technique to treat the decaying state is being developed by Kato *et al.* [11]. Here, we discuss the situation with a quasiclassical approach to the bremsstrahlung in tunneling [3], although the proposed method is not completely proved to be a good approximation of the quantum mechanical calculation for the radiation of the charged particle.

Following Dyakonov and Gornyi [3], we assume that the tunneling motion can be treated as a classical motion by using the complex time and the photon emission amplitude is changed by multiplying the additional factor, $\exp(i\omega\Delta t)$, where Δt corresponds to the time difference in the complex time. In a practical manner, we define $t = 0$ at the classical turning point $r_{\rm ctp}$ and $t = -i\tau$ at the separation point $r_s = 1.2 \times (4^{1/3} + A_d^{1/3})$, where A_d is the mass number of the daughter nucleus (τ is the "bounce" time for tunneling). The integral in Eq. (1) is performed from $t = -i\tau$ to 0 along the imaginary axis, which corresponds to the tunneling motion from r_s to $r_{\rm ctp}$, and the integral from $t = 0$ to $+\infty$ along the real axis, corresponding to the Coulomb acceleration from $r_{\rm ctp}$. No radiation was assumed from $t = -\infty - i\tau$ to $-i\tau$. The time integral along the imaginary axis is, then, multiplied by $\exp(-\omega\tau)$ and the product is used to calculate the emission probability. The strict classical calculation was applied for the Coulomb acceleration. The result is plotted with the thick solid line in Fig. 3. As can be seen, the photon emission probability is well reproduced for $E_\gamma < 300$ keV. Moreover, the trend at the higher energy region is also reproduced. We also show the photon emission probability for an artificial situation in which the tunneling particle terminates its motion at the outer edge of the Coulomb barrier, with the dot-dashed line in Fig. 3. It is shown that photons can be emitted more in the barrier than the outside, for γ-ray energies below 250 keV. Thus, in this quasiclassical calculation, the suppression of the emission probability for $E_\gamma < 400$ keV can be understood as a consequence of destructive interference of the radiation amplitudes between the inside and the outside of the barrier.

In summary, we have measured bremsstrahlung emission in the α decay of ^{210}Po. The obtained bremsstrahlung yields are much smaller than those predicted by a Coulomb acceleration model. This strongly suggests that the radiation amplitude in the barrier cannot be neglected. The data seem to be well reproduced by the recipe of Dyakonov and Gornyi [3], although the quasiclassical treatment of the radiation in the barrier is not firmly justified. It is highly desirable to develop the quantum mechanical treatment for the decaying state, as well as to discuss the quasiclassical treatment for the radiation of the tunneling particle.

As mentioned earlier, the present result is not consistent with that for ^{226}Ra and ^{214}Po reported by D'Arrigo *et al.* [4]. The emission probabilities presently obtained are at least 1 order of magnitude smaller than their values. It is almost inconceivable that the emission probability of the bremsstrahlung changes drastically with a small change of the decaying nucleus. Thus, at present, we have no possible explanation for the different result.

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