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Investigation of bremsstrahlung emission in α-decay of heavy nuclei

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Abstract

Bremsstrahlung emission during α-decay of the ^{226}Ra and ^{214}Po nuclei was observed for the first time. The photon energy spectrum was analysed in the framework of a classical electrodynamic model in order to investigate the dynamics of the α-decay. The theoretical model predictions are compared with the experimental data of other authors obtained by also observing the bremsstrahlung emission in the low energy nucleus–nucleus scattering.

There were several reasons for us to investigate the emission of bremsstrahlung in the α-decay of heavy nuclei. Firstly, our recent investigations of the emission of high energy bremsstrahlung in spontaneous fission gave us the possibility to observe interference phenomena in the yield of photons due to the spatial distribution of the fission fragments [1]. A comparison between the data and the results of theoretical calculations in the framework of a classical electrodynamic model showed that the best agreement between theory and experiment was obtained when a finite kinetic energy of the fragments before scission was assumed. Secondly, a few years ago we began to investigate the phenomenon of atomic K-shell ionization in α-decay and we measured the probabilities for the creation of a K-shell vacancy during the α-decay of some polonium isotopes [2]. A discrepancy was observed between the predictions of the semiclassical theory of inner shell ionization in ion-atomic collisions [3] and the existing experimental data [2,4]. The reason for this phenomenon may be connected with the shake-off effect of atomic electrons when the α-particle is tunneling through the Coulomb barrier of the nucleus [5].

We therefore expect that it is possible to obtain information on the α-decay dynamics and, especially, on the formation of the α-particle near the nuclear surface [6,7] as well as on the tunneling process [8] by observing bremsstrahlung emission in coincidence with the α-particle.

^{226}Ra, whose decay scheme is known from the literature (see for example Ref. [9]), was chosen as the source of α-particles. This source was obtained by evaporating ^{226}Ra onto a 1.5 cm diameter Ni foil and it was put into a vacuum chamber for the measurement. The activity of the source was $\sim 10^4$ α-particles/s. These particles were detected with a silicon surface-barrier detector with a ~ 20 keV energy resolution at an α-particle energy of ~ 5.3 MeV. The experiment was conducted at the Laboratory of Nuclear Reactions

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of the Institute of Nuclear Physics of Moscow State University.

The energy spectrum of the α-particles from the ^{226}Ra source is given in Fig. 1a. The bremsstrahlung quanta were recorded with a NaI(Tl) (22 mm diameter and 10 mm thick) scintillator with a ~5 keV energy resolution at a γ-ray energy of about 60 keV. The scintillator efficiency was determined using the standard γ-sources ^{241}Am (E_γ=59.6 keV) and ^{57}Co (E_γ=122 and 136 keV) and also from the yield of the 186 keV γ-line from ^{226}Ra. Since we used the same NaI(Tl)-detector as in our previous experiments with

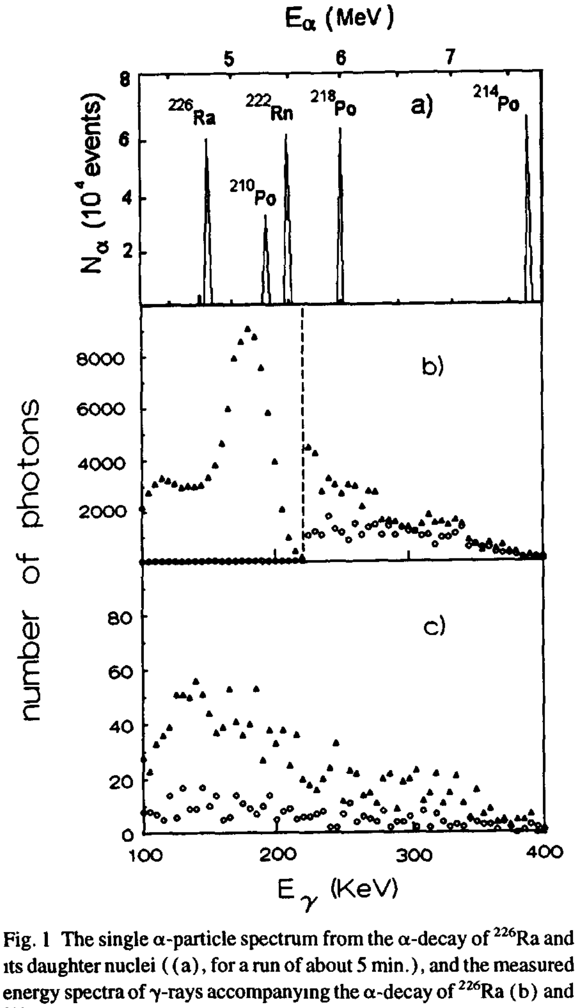

Fig. 1 The single α-particle spectrum from the α-decay of ^{226}Ra and its daughter nuclei ((a), for a run of about 5 min.), and the measured energy spectra of γ-rays accompanying the α-decay of ^{226}Ra (b) and ^{214}Po (c) for true coincidences (full triangles) and random events (open circles), for one run of 100 h The experimental data on the right side of (b) are multiplied by a factor of 50.

bremsstrahlung emission in nuclear reactions (see for example Ref. [10]), the response function for higher photon energies up to 500 keV was determined from the measured energy spectra of the bremsstrahlung arising from the scattering of 1.6 MeV protons on a thin carbon target and of 2.5–2.7 MeV protons on an oxygen target. We found that the experimental values are well described when one uses for the efficiency the following expression:

$$\varepsilon(E_\gamma)=A(E_\gamma)\{1-B(E_\gamma)\exp[-\mu(E_\gamma)d]\}, \qquad (1)$$

where E_γ is the photon energy, $\mu(E_\gamma)$ is the total γ-ray absorption coefficient of NaI and d is the detector thickness, while $A(E_\gamma)$ and $B(E_\gamma)$ are fitting parameters.

An angle of 90° was chosen between the γ-detector and the detector of the α-particles since, if the dipole contribution is the dominant one, we expect the maximum of the bremsstrahlung yield to occur at this angle. In fact, if we consider the ratio between the bremsstrahlung yields at angles of 90° and 15° (it is impossible to arrive at zero angle with respect to the α-emission because of the finite size of the detectors), we experimentally find a value of the order of 10^{-1}. The theoretical estimate in the framework of the classical electrodynamics model gives the same order of magnitude. The distance from the ^{226}Ra source to the NaI detector was 3 cm and it was 1 cm for the α-particle detector. It was possible to observe bremsstrahlung emission in the presence of a high background if the events, responsible for the photon emission, were selected by α–γ coincidences with a subsequent energy analysis of the α-particles and the bremsstrahlung photons. The aim of this analysis was to extract the coincidence events from the region of the (E_γ, E_α)-plane near the total energy conservation line, taking the detector energy resolutions (20 keV for alphas and 5 keV for photons) into account. The time resolution of the fast α–γ coincidences was 10 ns. At the first stage of the investigation we concentrated on two α-particle groups, one at the minimum observed kinetic energy (4.777 MeV coming from ^{226}Ra) and the other one at the maximum energy (7.695 MeV from ^{214}Po). For these groups of α-particles the energy spectra of the bremsstrahlung radiation are not distorted by electrons from conversion transitions up to 800 keV (the energy of the first excited level in the daughter nucleus for ^{214}Po) and from 186 keV (the energy of the first excited level for ^{226}Ra) up to the 4.777 MeV energy limit (kinetic energy of the α-particle). The other groups of α-particles with energies included in the 4.77 and 7.695 MeV range are not considered in our study because they may be influenced by distortion effects. The total time of the measurements was approximately 800 h.

In the same figure we present the measured energy spectra of γ-rays accompanying the α-decay of ^{226}Ra (Fig. 1b) and ^{214}Po (Fig. 1c) when a 10 ns window was selected around the prompt peak in the TAC spectrum (for real coincidences, full triangles) and with the same 10 ns window shifted by 200 ns with respect to the above-mentioned peak (for random events, open circles). For the case of the α-decay of ^{226}Ra we observe in the γ-ray spectrum a strong peak due to the 186 keV γ-ray emission from the first excited state of ^{222}Ra (see Ref. [9]). Therefore we restricted the bremsstrahlung spectrum to energies starting above 220 keV for the case of Ra and we removed in our analysis the events which are correlated with the ^{226}Ra α-decay to the first excited ^{222}Rn state. We estimate the influence of the 186 keV peak on the low energy part of the bremsstrahlung spectrum to be not larger than 20%. In the case of Po the energy interval of the bremsstrahlung spectrum was considered up to about 400 keV in order to exclude the contribution coming from the α-decay to the first excited state of the ^{210}Pb daughter nucleus whose energy is about 800 keV [9]. Considering that in our case the bremsstrahlung contributions are limited to the 140–400 keV region and taking the energy shift of the α-particles due to photon emission into account, we have no other γ-ray contributions due to the decay of the first excited state of the daughter nuclei since these γ-ray energies are higher than 800 keV.

In order to show the E_γ–E_γ event distribution along the total energy conservation line $E_\gamma + E_\alpha$ = constant related to a group of α-decay (where constant = $E_{\alpha,\max}$ when no photon is emitted) we present an appropriate two-dimensional map of the γ–α coincidences. Fig. 2 shows the two-dimensional E_γ–E_α even distribution of true + random coincidences (Fig. 2a) – when the 10 ns window is set around the prompt peak of the TAC – and random ones only (Fig. 2b) – when the 10 ns window was shifted by about 200 ns – for five groups of α-particle decay along the chain from ^{226}Ra to ^{214}Po. The two peaks in the coincidences of α-particles from ^{226}Ra correspond to the formation of the first excited state of the ^{222}Rn daughter nucleus and sequential decay

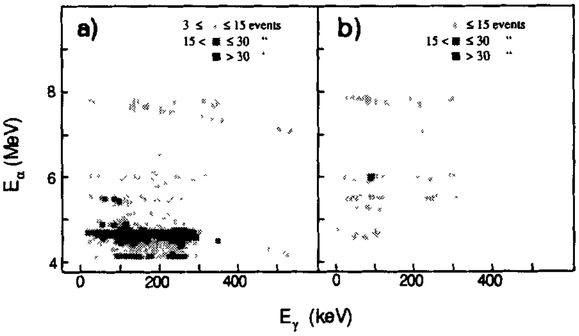

Fig. 2 Two-dimensional plot of E_γ–E_α coincidences for (a) true + random coincidences, (b) random coincidences, obtained during a run of 100 h with a 10 ns time window around the prompt peak of the TAC and one shifted by 200 ns, respectively. The concentration of events corresponding to the different α-particle groups emitted in coincidence with γ-rays can be observed.

with emission of the 186 keV γ-ray. The relatively feeble but clearly observed inclination of the lines in the true + random spectrum near the high intensity horizontal lines corresponding to the fifth, fourth and first α-groups (Fig. 2a) – with the slope supported by the $E_\gamma + E_\alpha$ conservation law – and the absence of these lines in the random spectra (Fig. 2b) is the evidence of bremsstrahlung emission accompanying the α-decays.

To check our method we estimated the probability of α-decay from the ground state of ^{226}Ra to the first excited state of ^{222}Rn accompanying the 186 keV γ-ray emission. This probability is equal to 0.034 ± 0.002 and it is in good agreement with the data of other authors [9]. The bremsstrahlung spectra were averaged over the photon energy in a 25 keV interval. The energy spectra of the bremsstrahlung radiation in coincidence with the α-particles from ^{226}Ra and ^{214}Po are shown in Fig. 3, after having selected the events along the energy conservation line of the first and fifth α-groups respectively. It will be noted that the bremsstrahlung emission probability depends on the energy of the α-particle and the yield is bigger for the higher energy α-particles (compare the experimental data of Fig. 3a with the ones in Fig. 3b for each E_γ value). Moreover, the energy dependence of the bremsstrahlung radiation is smooth and follows the $\mathrm{d}^2P/\mathrm{d}E_\gamma \mathrm{d}\Omega_\gamma \propto 1/E_\gamma$ law.

In our calculations we used the classical bremsstrahlung theory [11] for the two extreme cases of the sudden acceleration model (SA) and the Coulomb

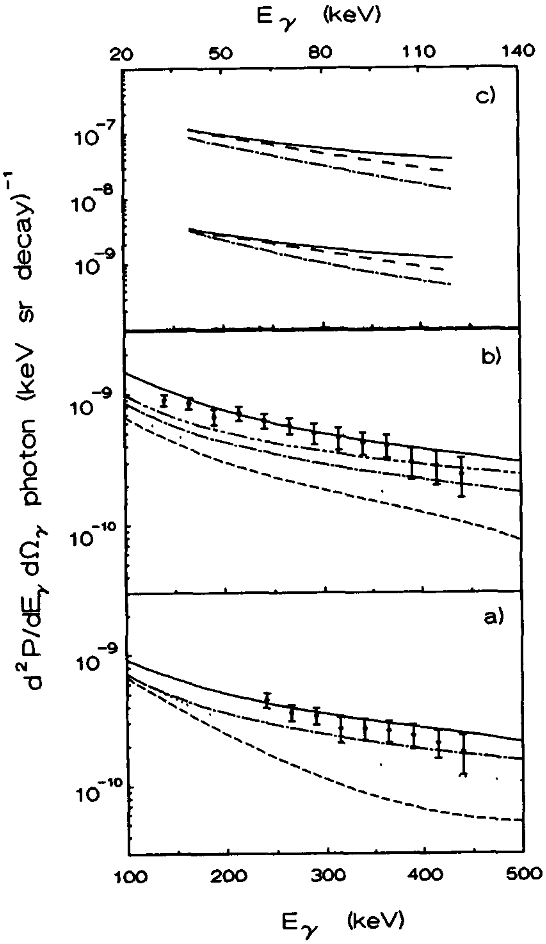

Fig. 3 The bremsstrahlung emission probability versus E_γ accompanying the α-decay of ^{226}Ra (a) and ^{214}Po (b). Full points are the experimental data Dashed lines are the CA-model calculations (when the intial velocities of the two bodies are zero), while the full lines represent those of the SA model (when the α-particle and the residual nucleus are instantaneously produced at their final kinetic energies) Dotted, dash-dotted and dash-double-dotted lines represent the calculations obtained by the CA model when one uses an initial kinetic energy of the α-particles of 1, 3 and 5 MeV, respectively In (c) the bremsstrahlung emission probabilities in the ^{35}Cl + ^{118}Sn (upper part of this figure) and ^{37}Cl + ^{112}Sn (lower part) scattering at $E_{lab}(^{35}Cl) = 44$ MeV and $E_{lab}(^{37}Cl) = 44\ 5$ MeV respectively, are shown The dash-dotted line represents the calculations obtained by Gaukler et al [13] for the two above-mentioned reactions Dashed and full lines represent our CA- and SA-model calculations (performed for the same interactions) obtained for the same angles used by Gaukler in his experiment and considering the same angular intervals subtended by the γ- and particle detectors

acceleration model (CA), respectively. The SA model suggests that the α-particle and the residual nucleus in the decay process are instantaneously produced at their final kinetic energies, while in the CA model the two bodies are accelerated in the Coulomb field assuming that the initial velocities are zero. For all cases the total vector potential of the electromagnetic field is represented as a sum of the vector potentials of the two fragments (α-particle and remaining nucleus):

$$A_{\text{total}}(\omega) = A_1(\omega) + A_2(\omega)\ , \tag{2}$$

where

$$A_i(\omega) = \left(\frac{e^2}{8\pi^2 c}\right)^{1/2} \int_{-\infty}^{+\infty} \mathrm{d}t\, Z_i$$

$$\times \exp\{\mathrm{i}[\omega t - \boldsymbol{k}\boldsymbol{r}_i(t)]\} \frac{\boldsymbol{n}\times[(\boldsymbol{n}-\boldsymbol{\beta}_i)\times\dot{\boldsymbol{\beta}}_i]}{(1-\boldsymbol{\beta}_i\boldsymbol{n})^2} \tag{3a}$$

in the case of the CA model, or

$$A_i(\omega) = \left(\frac{e^2}{8\pi^2 c}\right)^{1/2} \int_{-\infty}^{+\infty} \mathrm{d}t\, Z_i \frac{\boldsymbol{\beta}_i(t)}{1-\boldsymbol{n}\boldsymbol{\beta}_i}$$

$$\times \exp\{\mathrm{i}[\omega t - \boldsymbol{k}\boldsymbol{r}_i(t)]\} \tag{3b}$$

in the case of the SA model, and Z_i, $\boldsymbol{r}_i$, $\boldsymbol{\beta}_i$, $\dot{\boldsymbol{\beta}}_i$, being the charge, position, velocity and acceleration of the ith fragment (where $\boldsymbol{\beta}_i = \dot{\boldsymbol{r}}_i/c$); $\boldsymbol{k}$, $E_\gamma = h\omega$, R_0 being the wave vector, energy of the photon and the observation distance: and $\boldsymbol{n} = \boldsymbol{k}/k$ being the unit vector.

We chose to use formulae (3a) and (3b), that represent the complete contribution of the vector potential, instead of the usual approximation related to the multipole expansion [12]. The resulting differential number spectrum per unit energy and unit solid angle for the emission of the γ-rays is

$$\frac{\mathrm{d}^2N}{\mathrm{d}E_\gamma \mathrm{d}\Omega_\gamma} = \frac{2}{\hbar E_\gamma} \left|[A_1(\omega) + A_2(\omega)]\times\boldsymbol{n}\right|^2\ , \tag{3c}$$

considering the sum of the two contributions.

The comparison of the results of the SA- and CA-model calculations with the experimental data are shown in Fig. 3a for ^{226}Ra and in Fig. 3b for ^{214}Po. The CA-model calculations underestimate the experimental data in both cases. The results for the SA-model calculations were obtained when an initial kinetic energy equal to the final value is assumed. In this case the agreement between the theoretical calculations and the experimental data is better for both ^{226}Ra and ^{214}Po since the calculations obtained with an initial kinetic energy of the α-particle of 1 and 3 (for Figs. 3a and 3b) and 5 MeV (for Fig. 3b), respectively underpredict the bremsstrahlung emission.

The theoretical calculations in the framework of the SA model give us for the probability of bremsstrahlung emission

$$\frac{d^2P}{dE_\gamma d\Omega_\gamma} = 1.08\times10^{-7}E_\gamma^{-1} \text{ photon (keV sr decay)}^{-1} \text{ for } ^{226}\text{Ra}, \quad (4a)$$

and

$$\frac{d^2P}{dE_\gamma d\Omega_\gamma} = 1.50\times10^{-7}E_\gamma^{-1} \text{ photon (keV sr decay)}^{-1} \text{ for } ^{214}\text{Po}. \quad (4b)$$

The same calculations in the framework of the CA model yielded probabilities of $0.293\times10^{-7}\ E_\gamma^{-1}$ and $0.556\times10^{-7}\ E_\gamma^{-1}$ photon (keV sr decay)$^{-1}$, respectively, for ^{226}Ra and ^{214}Po.

As a next step we compare the results obtained from α-decay with those found by nucleus–nucleus scattering in order to check which model better describes the particular interaction dynamics. We therefore consider, for example, the ^{35}Cl + ^{118}Sn and ^{37}Cl + ^{112}Sn reactions [13] respectively at E_{lab} (^{35}Cl) = 44 MeV and E_{lab} (^{37}Cl) = 44.5 MeV.

In Fig. 3c we show the calculation obtained by Gaukler et al. [13] (dash-dotted line) for the cases of ^{35}Cl (44 MeV) + ^{118}Sn and ^{37}Cl (44.5 MeV) + ^{112}Sn scattering using the quantum mechanical multipole expansion approximation. In the same figure we also show our calculation (obtained for the same condition as Gaukler's experiment) of the bremsstrahlung emission obtained by formulae (3a), (3b) and (3c) for the above-mentioned interactions, in the frameworks of the CA and SA models. It must be mentioned that in Ref. [13] the authors averaged the angular distribution of the bremsstrahlung according to the experimental situation and presented their data in units of [photons 4π/(keV sr)]. As one can see, our calculations (in particular those obtained by using the CA model) agree with those shown by Gaukler et al. in their paper, where they concluded that, by using the computer code of Reinhardt et al. [12] for the nucleus–nucleus bremsstrahlung emission up to the quadrupole radiation amplitude, their calculations agreed with the experimental data that show a strong isotope effect. We also observe that the predictions of the SA and CA models agree better with each other than in the case of α-decay. This reflects the fact that in the case of Cl + Sn scattering the ratio of the kinetic energy E_P of the particle to the height of the Coulomb barrier V_C is higher than in the case of α-decay and, therefore, the CA model has a more realistic base (E_p/V_C is equal to 0.4 for Cl + Sn scattering and it is equal to 0.2 for α-decay of ^{226}Ra and 0.3 for ^{214}Po).

From all this follows that the appropriate determination of the kinetic energy of the α-particles at the time of the bremsstrahlung emission is important in order to understand the dynamics of this process. By comparing the bremsstrahlung emission in the α-decay with that in nucleus–nucleus scattering, we confirm that:

(i) the CA-model calculations underestimate the data of the bremsstrahlung yields, as in the case of bremsstrahlung emission in spontaneous fission [1], while in the present case the SA-model calculations with an initial kinetic energy of the α-particles close to the final kinetic energy seem to agree better with the experimental data obtained by observing the bremsstrahlung accompanying the ^{226}Ra and ^{214}Po α-decays;

(ii) the bremsstrahlung emission in nucleus–nucleus elastic scattering at energies lower than the Coulomb barrier is in agreement with CA-model calculations.

We believe that these results may be related to the different mechanism of photon emission in the cases (i) and (ii) or in other words due to the additional increase of the photon emission probability when the α-particles pass through the Coulomb barrier and in the external region near the barrier. Therefore, the bremsstrahlung emission accompanying the α-decay may be a useful tool in the investigations of α-decay dynamics where the model with instantaneously initial kinetic energy appears to describe the decay process well in the present cases of the ^{226}Ra and ^{214}Po nuclei.

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