# Search for high energy $\gamma$ rays from the spontaneous fission of <sup>252</sup>Cf

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(Received 26 December 1990)

We have searched for high energy  $\gamma$  rays emitted from the spontaneous fission of <sup>252</sup>Cf. This search was carried out by two methods: the measurement of high energy  $\gamma$  rays in coincidence with fission fragments, and the measurement of high energy  $\gamma$  rays in coincidence with any other  $\gamma$  ray. The first of these two methods allowed us to establish a direct correlation between high energy  $\gamma$  rays and fission fragments. For the fission-fragment/ $\gamma$ -ray measurement, we were able to set an upper limit at the 95% confidence level of  $1.8 \times 10^{-6}$  photon per fission for the integrated yield of  $\gamma$  rays with an energy greater than 30 MeV. Additionally, we have established an upper limit at the 95% confidence level of  $1.0 \times 10^{-5}$  photon per fission for the integrated yield of photons with energy greater than 30 MeV from the  $\gamma$ - $\gamma$  measurement. These results are shown to be consistent with a classical model estimate of the upper limit of  $\gamma$  rays produced by a nucleus-nucleus bremsstrahlung process arising from the Coulomb acceleration of the fission fragments.

# I. INTRODUCTION

In the last several years there has been a renewed interest in the spontaneous fission of <sup>252</sup>Cf. The large amount of energy available in the fissioning system ( $\approx$ 185 MeV) has motivated numerous experiments looking for the emission of high energy  $\gamma$  rays [1], neutral pions [2-4], and charged pions [5]. In an amazing series of experiments [5, 6] Ion and his collaborators have reported searching for various radioactivities from the spontaneous fission of <sup>252</sup>Cf. They looked for  $\pi^{\pm}$ ,  $\mu^{\pm}$ , and have even suggested the possible existence of natural strange radiation in the form of the spontaneous emission of  $\Lambda^0$  ( $m_{\Lambda^0}$ = 1116 MeV). They have set an upper limit between 1  $\times$  10<sup>-8</sup> and 9  $\times$  10<sup>-8</sup> for the total branching ratios for the emission of  $\pi^{\pm}$  and  $\mu^{\pm}$ ,  $\Gamma_{\pi^{\pm},\mu^{\pm}}/\Gamma_{\text{fiss}}$ . Beene *et al.* [2] and Cerruti *et al.* [3] reported looking for neutral pions rather than charged pions. Both groups searched for back-to-back emission of high energy  $\gamma$  rays as evidence for the emission of  $\pi^0$  ( $\pi^0 \rightarrow \gamma \gamma$ , decay fraction = 98.8% ). Cerruti et al. established a branching ratio of  $\Gamma_{\pi^0}/\Gamma_{\text{fiss}} < 5.0 \times 10^{-9}$ . Beene *et al.* established an upper limit for the production of  $\pi^0$  from the spontaneous fission of  ${}^{252}$ Cf of  $1.0 \times 10^{-9}$  at the 90% confidence level and  $1.5 \times 10^{-9}$  at the 95% confidence level. More recently Stanislaus et al. [4] have established an upper limit for  $\Gamma_{\pi^0}/\Gamma_{\rm fiss} < 3.3 \times 10^{-10}$  at the 90% confidence level for the production of  $\pi^0$ 's. They have also suggested that is the practical limit for the upper limit for conventional  $\gamma$ ray detection (i.e., scintillation counters) because of the large cosmic ray background. Stanislaus et al. [4] also mentioned a proposed experiment at LAMPF by Morris and Knudson to look for  $\pi^0$ 's in the  $\pi^0$  spectrometer, but to date no data have been published.

Kasagi *et al.* [1] have reported the emission of very high energy  $\gamma$  rays from the spontaneous fission of <sup>252</sup>Cf. In their measurements  $\gamma$  rays with energies up to 160 MeV were observed. The reported photon spectrum was very flat for  $E_{\gamma} > 30$  MeV, and had a shape very similar to the cosmic-ray background spectrum. Their measurements of high energy  $\gamma$  rays were performed using a seven-element BaF<sub>2</sub> array. The high energy  $\gamma$  rays in the BaF<sub>2</sub> were detected in coincidence with  $\gamma$  rays in a NE213 liquid scintillator detector in an attempt to establish a correlation between the high energy  $\gamma$  rays and the fragments from the fission of <sup>252</sup>Cf. This approach was based on the fact that several prompt  $\gamma$  rays are emitted during each fission and, therefore, a coincidence between one of these  $\gamma$  rays and a high energy  $\gamma$  ray should reflect a correlation of high energy photons with the fission fragments.

The possibility of observing high energy  $\gamma$  rays during the fission of  $^{252}$ Cf was intriguing because the observation of  $\gamma$  rays with energies on the order of a hundred MeV was so unexpected. An experimental program was undertaken at the University of Washington to confirm the measurements of Kasagi *et al.* and, if a null result were obtained, to establish an upper limit for the production of high energy  $\gamma$  rays from the spontaneous fission of  $^{252}$ Cf.

### **II. EXPERIMENTAL DETAILS**

The present search involved two separate techniques; the first was the detection of high energy  $\gamma$  rays in direct coincidence with fission fragments, and the second involved the detection of high energy  $\gamma$  rays in coincidence with any prompt  $\gamma$  ray. The second method was used to reproduce, as closely as possible, the measurement made by Kasagi *et al.* Since the fission-fragment/ $\gamma$ -ray coincidence method allowed for the direct correlation of high energy photon and fission fragment emission, as well as photon energy with the energy of the fission fragments, it was the more desirable of the two methods. The fissionfragment/ $\gamma$ -ray trigger was expected to be much cleaner than the  $\gamma$ - $\gamma$  trigger, in that the fission fragment coincidence could not be triggered on background radiation, such as cosmic rays, as easily as the  $\gamma$ - $\gamma$  trigger. The measurement of both the fission-fragment and  $\gamma$ -ray energies also allowed a check for conservation of energy.

The fission-fragment/ $\gamma$ -ray coincidence measurement required the acquisition of a 50  $\mu$ Ci "fission-foil" <sup>252</sup>Cf source [7]. The source had isotopically pure <sup>252</sup>Cf electrodeposited on a platinum disk from which it was possible to detect the fragments from the spontaneous fission of <sup>252</sup>Cf on one side of the source. The experimental setup is shown in Fig. 1. The <sup>252</sup>Cf source was placed in the center of a scattering chamber at a distance of 1.2 cm from a silicon surface barrier detector. Several of these detectors were used in the course of the measurement: the detectors lasted two to three days before they became severely radiation damaged. The silicon detectors were collimated to yield a half angle of between 22° and 38°, depending on the size of the detector. With this collimation and source activity, a 2-8 kHz counting rate for the fission fragments was achieved. Data were collected in the fission-fragment/ $\gamma$ -ray coincidence mode for 240 hours, during which time a total of  $2.8 \times 10^9$  fissions were observed. The source was 50 cm away from a 25.4  $cm \times 38.1$  cm NaI detector which was surrounded by a passive lead shield and an active anticoincidence shield to minimize the cosmic-ray background. The solid-state detector was placed at an angle of 120° with respect to the axis of the NaI detector. Since the fission detector defines the fission axis of the detected fragments, this means that dipole radiation emitted from the fissioning system would come out primarily perpendicular to the fission axis (see Fig. 1). The 120° angle was chosen to maximize, as much as possible, the detection of higher multipole radiation, particularly the quadrupole radiation [8]. In addition the time of flight between events in the fission-fragment detector and in the NaI detector



FIG. 1. Experimental setup used for the present measurements. The fission axis (solid line) is defined in the fissionfragment/ $\gamma$ -ray measurement by the line between the fission source and the fission detector. The angular distributions for the dipole and quadrupole radiation [8] produced in a sudden approximation are also shown. The angular distribution from the dipole radiation has been multiplied by a factor of 5. The difference in relative intensities was calculated at a  $\gamma$ -ray energy of 40 MeV, using the most probable charges and masses of the fission fragments [16] to determine the effective dipole and quadrupole charges. The angular acceptance of the NaI detector is shown by the dash-dotted lines.

was recorded. The gate in the time-difference spectrum was narrow enough to allow the discrimination against neutrons of 25 MeV or less. The inability to discriminate against higher energy neutrons was mostly due to the degradation of the timing characteristics of the fission detectors from radiation damage.

The  $\gamma$ - $\gamma$  coincidence measurement was performed by placing a NE213 liquid scintillator 25.4 cm from the <sup>252</sup>Cf source; this geometry gave  $\approx 20\%$  geometric efficiency for the detection of the prompt  $\gamma$  rays. Time-of-flight and pulse-shape discrimination were used to separate neutrons from photons in the liquid scintillator. This discrimination made it possible to separate photons from neutrons of energy less than 44 MeV. Data were collected in this  $\gamma$ - $\gamma$  coincidence mode for 172 hours, which corresponds to  $7.8 \times 10^8$  detected fissions. The liquid scintillator was placed collinear with the axis of the NaI detector. The angular correlations between the fission fragments and the high energy  $\gamma$  rays were not as well defined in this case, since the prompt  $\gamma$  rays are emitted roughly isotropically from the fissioning system [9]. In both measurements the discrimination against high energy neutrons was not important since no neutron events were observed at energies greater than  $\approx 10-15$  MeV.

The NaI detector was calibrated up to a  $\gamma$ -ray energy of 30 MeV using low energy sources and  $\gamma$  rays from <sup>11</sup>B( $p, \gamma$ ) at E<sub>p</sub> = 7.25 and 14.3 MeV. The calibration was also checked against the cosmic-ray muon peak in the NaI spectra; the peak corresponds to an energy of  $\approx 123$  MeV for the detector geometry used in this work. The calibration reproduced the energy of the cosmic ray muon peak to better than one percent.

The absolute-efficiency-solid-angle product,  $\epsilon d\Omega$ , of the NaI detector was determined by measuring the resonance strength for the reaction  ${}^{12}C(p,\gamma_0){}^{13}N$  at a proton energy of 14.25 MeV and then comparing this value to the measurement of the absolute resonance yield given by Marrs *et al.* [10]. This measurement allowed the determination of the absolute efficiency for  $E_{\gamma} = 15$  MeV. Based upon a Monte Carlo simulation of the detector response [11], the total efficiency of the detector is nearly constant as a function of energy, while the "accepted" efficiency, defined by those events which are not in coincidence with the plastic shield, falls off exponentially above 15 MeV. The exponential slope was obtained by measuring the accept-to-total ratio for gamma rays from  ${}^{11}B(p,\gamma_{0,1})$  at various incident proton energies.

At the photon energies of interest,  $E_{\gamma} > 30$  MeV, cosmic-ray showers were expected to be the major source of background in the NaI detector. At these energies the measured cosmic-ray rejection efficiency is greater than 99%. The measured rate of cosmic-ray events which are not rejected by the anticoincidence shield is  $\approx 0.3$  event/MeV h. The coincidence resolving time between the NaI detector and the fission detectors leads to an estimated accidental cosmic-ray rate of  $\approx 6 \times 10^{-9}$  event/MeV sec for the fission-fragment coincidence measurement and an accidental rate of  $\approx 8 \times 10^{-10}$ event/MeV sec for the  $\gamma$ - $\gamma$  measurement, which in either case, corresponds to less than one count in the energy bin,  $30 < E_{\gamma} < 200$  MeV, for the duration of the present experiment.

A more serious cosmic-ray-induced background than the problem of accidental coincidences described above is that of pileup in which a cosmic-ray induced event overlaps a time coincidence between a low energy photon in the NaI and either one of the fission detectors. These pileup events can occur in the acceptance window period for processing "good" events. The expected rates of pileup events are approximately  $1 \times 10^{-8}$  and  $7 \times$  $10^{-8}$  event/MeV sec, for the  $\gamma$ - $\gamma$  and fission-fragment coincidence modes, respectively. These rates correspond to the expectation of one pileup event in the  $\gamma$ - $\gamma$  mode and seven pileup events with  $E_{\gamma} = 30-200$  MeV in the fissionfragment mode during the course of the experiment. The factors that influence the pileup rate are the NaI energy signal integration time, the total cosmic-ray event rate in the NaI and the NaI-fission coincidence rate. In order to identify pileup events the energy signal from the plastic anticoincidence shield was integrated for the same time period as the NaI signal, to obtain a correlation between plastic pulse height and NaI pulse height. Cosmic-ray pileup events are characterized by a large plastic signal correlated with a large NaI signal. With this type of analysis, one pileup event was seen in the  $\gamma$ - $\gamma$  mode and no events were seen in the fission-fragment/ $\gamma$ -ray mode.

## **III. EXPERIMENTAL RESULTS**

The solid histogram in Fig. 2 shows the  $\gamma$ -ray spectrum obtained from the fission of  $^{252}$ Cf in the fission-fragment/ $\gamma$ -ray coincidence mode. Photons having energy greater than 30 MeV were not observed in this measurement. The result from the fission-fragment/ $\gamma$ -ray measurement is very different from the result reported by Kasagi *et al.*, who claimed to see a considerable number of photons with energy greater than 30 MeV. In the fission-fragment/ $\gamma$ -ray coincidence mode there were no

pileup events seen in which there was a large plastic signal in coincidence with a large NaI signal. This indicates that the events were background free. In the  $\gamma$ - $\gamma$  mode, two events were seen in the energy region between 30 and 160 MeV, one event at 36 MeV, and one event at 103 MeV. The two events did not have large plastic signals, and therefore could be regarded as "real" events. An additional candidate event, observed at 192 MeV, is not included in Fig. 2. A  $\gamma$  ray of this energy seems rather unlikely to arise from the fission of <sup>252</sup>Cf, since there is only  $\approx 185$  MeV available in the fissioning system. The present measured yields were converted into angle-integrated cross sections by assuming isotropic photon emission from the radiation source. If the angular distribution were as strongly peaked into the angular acceptance of the NaI detector as indicated by our calculations discussed below, the high energy photon emission probability measured in the fission-fragment/ $\gamma$ -ray mode would be even *lower* by a factor of roughly 2-3 than is indicated in Fig. 2 and subsequent figures.

The solid curve in Fig. 3 shows the upper limit for the production of  $\gamma$  rays at the 95% confidence level for the fission-fragment/ $\gamma$ -ray coincidence data. The upper limit was calculated assuming a constant  $\gamma$ -ray emission probability as a function of energy from 30 to 200 MeV and was based on the observation that there were no counts in the energy range  $30 < E_{\gamma} < 200$  MeV. The upward slope in the upper limit corresponds to the efficiency of the NaI exponentially decreasing with increasing energy. Numerically this limit may be expressed as  $6.3 \times 10^{-12} \epsilon_{\text{NaI}}(E_{\gamma})$  (photon/fission MeV), where  $\epsilon_{\text{NaI}}(E_{\gamma})$  is the absolute efficiency of the NaI detector as a function of the  $\gamma$ -ray energy. The upper limit of the  $\gamma$ - $\gamma$  measurement is  $3.6 \times 10^{-11} \epsilon_{\text{NaI}}(E_{\gamma})$  (photon/fission MeV) at the 95% con-



FIG. 2. Measured  $\gamma$ -ray spectra for photon emission from the spontaneous fission of <sup>252</sup>Cf. The solid histogram is the spectrum in the fission-fragment/ $\gamma$ -ray coincidence mode. The dashed histogram is the spectrum for the  $\gamma$ - $\gamma$  coincidence measurement.



FIG. 3. Upper limits set for the high energy  $\gamma$ -ray production from the spontaneous fission of <sup>252</sup>Cf from the present work compared to measurements of Kasagi *et al.* The solid line corresponds to the upper limit set by the present fission-fragment/ $\gamma$ -ray coincidence measurement at the 95% confidence level. The dashed line is the upper limit set by the present  $\gamma$ - $\gamma$  coincidence measurement at the 95% confidence level. The dash-dotted line is the measured level of photon production reported by Kasagi *et al.* [1].

fidence level. The energy-integrated upper limits at the 95% confidence level for photon energy greater than 30 MeV are  $1.8 \times 10^{-6}$  and  $1.0 \times 10^{-5}$  photon/fission for the fission fragment/ $\gamma$ -ray and  $\gamma$ - $\gamma$  coincidence measurements, respectively. These upper limits are very conservative since they assume that the probabilities for the emission of any  $\gamma$  ray with energy greater than 30 MeV are equal. At the very least the photon emission probability should be inversely proportional to the  $\gamma$ -ray energy, leading to a lower upper limit.

The present experiment was not able to reproduce the high energy photon emission rate from  $^{252}$ Cf at the level reported by Kasagi *et al.* [1] in either the fission- $\gamma$  or  $\gamma$ - $\gamma$  modes. (Since the publication of their report, Kasagi *et al.* have communicated to us that their upper limit might be an order of magnitude below what they reported in Ref. [1].) Additionally the upper limit determined from the present measurement at  $E_{\gamma} = 40$  MeV is two orders of magnitude below the photon production reported in Ref. [1].

On the other hand, the medium energy,  $8 < E_{\gamma} < 20$  MeV, photon spectrum obtained in the fission-fragment/ $\gamma$ -ray measurement is in good agreement with the data of Kasagi *et al.* and of Dietrich *et al.* [12], as shown in Fig. 4. In comparing the medium energy photon data to that of Kasagi *et al.* and Dietrich *et al.* it was assumed that the emission of these photons was isotropic. This assumption was based on the work of Hoffman [9], who showed that the anisotropy in the emission of prompt  $\gamma$  rays (0.25 MeV  $< E_{\gamma} < 1.25$  MeV) in the fission process is on the order of 10–15 %.

The upper limit for the emission of high energy photons established by the fission-fragment/ $\gamma$ -ray coincidence mode measurement is clearly lower than the measurement of Kasagi *et al.* [1] and is a more direct measurement of the fission-fragment correlation with the emitted photons. The sensitivity of the present experiment was greatly enhanced by the efficient rejection of



FIG. 4. Medium energy  $\gamma$ -ray spectra from the spontaneous fission of <sup>252</sup>Cf. The rounded boxes are the data from Ref. [11] and the crosses are from Ref. [1]. The open diamonds are from the present measurement in the fission-fragment/ $\gamma$ ray coincidence mode.

the cosmic-ray background. This high cosmic-ray rejection efficiency was due to use of the active anticoincidence shield as opposed to the subtraction of an average cosmicray background as reported in Ref. [1]. In the experiment of Ref. [1] photon spectra were obtained with and without a fission source, and the corrected photon spectrum was obtained by subtracting a normalized background spectrum without the fission source from the spectrum obtained with the fission source.

### **IV. BREMSSTRAHLUNG CALCULATIONS**

In addition to establishing an upper limit for the emission of high energy photons from the spontaneous fission of <sup>252</sup>Cf, calculations were performed to predict the estimated yield of these photons. These calculations were motivated by the calculated values in Ref. [1] which were qualitatively consistent with their observations, but inconsistent with the present upper limits. Kasagi et al. [1] point out that the emission of high energy photons from the spontaneous fission of <sup>252</sup>Cf might be one of the only vehicles to study nucleus-nucleus bremsstrahlung, since most nuclear bremsstrahlung from the collision of heavy ions seems to arise from a nucleon-nucleon mechanism [13]. A detailed microscopic calculation of the bremsstrahlung produced in the spontaneous fission of <sup>252</sup>Cf would be a very difficult undertaking, and it is not clear whether the added complexity would shed any light on the problem. Since the language and methods of classical mechanics have been used in the past to deal with the fission process, it is reasonable to think that a simple model could be constructed to predict the radiation process via bremsstrahlung arising from the Coulomb acceleration of the two fission fragments from a scissionlike configuration to infinity.

Following Jackson [14], the radiation intensity from the interaction of charged particles is

$$\frac{d^2 I}{d\omega d\Omega} = 2 | \mathbf{A}(\omega) |^2, \tag{1}$$

where  $A(\omega)$  is the vector potential in frequency space

$$\mathbf{A}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} dt \quad \mathbf{A}(t) e^{-i\omega t}, \qquad (2)$$

and A(t) is the vector potential in time.

When two charged particles are involved in the radiation process the intensity of the emitted radiation is the coherent sum of the vector potentials of the two charged particles

$$\frac{d^2 I}{d\omega d\Omega} = 2 | \mathbf{A}_1(\omega) + \mathbf{A}_2(\omega) |^2, \qquad (3)$$

the vector potentials are

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$$\mathbf{A}_{i}(\omega) = \left(\frac{1}{\sqrt{2\pi}}\right) \left(\frac{c}{4\pi}\right)^{\frac{1}{2}} \left(\frac{1}{c}\right) \int_{-\infty}^{\infty} dt \quad \left[\frac{\hat{\mathbf{n}} \times \left[(\hat{\mathbf{n}} - \boldsymbol{\beta}_{i}) \times \dot{\boldsymbol{\beta}}_{i}\right]}{(1 - \boldsymbol{\beta}_{i} \cdot \hat{\mathbf{n}})^{2}}\right] q_{i} e^{-\imath \omega \left[t - \hat{\mathbf{n}} \cdot \mathbf{r}_{i}(t)/c\right]}.$$
(4)

The index *i* refers to each of the point charges  $q_i = z_i e$ ,  $t - \hat{\mathbf{n}} \cdot \mathbf{r}_i(t)/c$  represents retarded time,  $\hat{\mathbf{n}}$  is the unit vector along the radiation emission axis, and  $\beta$  and  $\dot{\beta}$  are the relative velocity and acceleration of the charges, respectively. In the nonrelativistic limit,  $\beta \ll 1$ , the intensity of the emitted radiation may be simplified to

$$\frac{d^2 I}{d\omega d\Omega} = \frac{1}{4\pi^2 c} \left| \int_{-\infty}^{\infty} dt \quad \sum_{i=1}^{2} [\hat{\mathbf{n}} \times (\hat{\mathbf{n}} \times \dot{\boldsymbol{\beta}}_i)] \, q_i e^{-i\omega[t - \hat{\mathbf{n}} \cdot \mathbf{r}_i(t)/c]} \right|^2.$$
(5)

The motion of the two fission fragments is confined to one-dimensional motion along the fission axis. The accelerations of the particles are simply  $\dot{\beta}_1 = \ddot{\mathbf{x}} \mu/cm_1$  and  $\dot{\beta}_2 = -\ddot{\mathbf{x}} \mu/cm_2$  where  $\ddot{\mathbf{x}}$  is the relative acceleration  $\ddot{\mathbf{x}}_1 - \ddot{\mathbf{x}}_2$ . The scalar products in the arguments of the exponentials are

$$\mathbf{\hat{n}} \cdot \mathbf{r}_1(t)/c = \mathbf{\hat{n}} \cdot \mathbf{x} \frac{\mu}{m_1 c}$$

 $\mathbf{and}$ 

$$\mathbf{\hat{n}}\cdot\mathbf{r}_{2}(t)/c=-\mathbf{\hat{n}}\cdot\mathbf{x}rac{\mu}{m_{2}c}$$

where  $\mathbf{x}$  is the relative position  $\mathbf{x}_1 - \mathbf{x}_2$ ; these scalar product terms correspond to the time retardation factor. With a little bit of algebra, the intensity of the emitted radiation becomes

$$\frac{d^2I}{d\omega d\Omega} = \frac{\mu^2}{4\pi^2 c} \left| \int_{-\infty}^{\infty} dt \quad \frac{1}{c} [\hat{\mathbf{n}} \times \ddot{\mathbf{x}}] e^{-i\omega t} \left( \frac{q_1}{m_1} e^{i(\omega/c)(\mu/m_1)\hat{\mathbf{n}}\cdot\mathbf{x}} - \frac{q_2}{m_2} e^{-i(\omega/c)(\mu/m_2)\hat{\mathbf{n}}\cdot\mathbf{x}} \right) \right|^2. \tag{6}$$

The intensity of emitted radiation can be converted to a differential number spectrum per unit energy by dividing by  $\hbar^2 \omega$  [15]. The resulting differential number spectrum per unit energy per unit solid angle for the emission of the  $\gamma$  rays is

$$\frac{d^2 N}{dE_{\gamma} d\Omega_{\gamma}} = \frac{\mu^2}{4\pi^2 (\hbar c) c^2} \frac{e^2}{E_{\gamma}} \left| \int_{-\infty}^{\infty} dt \quad \left[ \hat{\mathbf{n}} \times \ddot{\mathbf{x}} \right] e^{-i\omega t} \left( \frac{z_1}{m_1} e^{i(\omega/c)(\mu/m_1)\hat{\mathbf{n}} \cdot \mathbf{x}} - \frac{z_2}{m_2} e^{-i(\omega/c)(\mu/m_2)\hat{\mathbf{n}} \cdot \mathbf{x}} \right) \right|^2.$$
(7)

The above expression gives the exact energy spectrum, in the nonrelativistic limit, of the bremsstrahlung produced from acceleration of the two pointlike charges. In the case of the spontaneous fission of  $^{252}$ Cf, the asymptotic value of  $\beta_{rel}$  is about 0.04, so the nonrelativistic limit is adequate. Applying the formulation to the problem of bremsstrahlung produced by the fission fragments from spontaneous fission of  $^{252}$ Cf requires the determination of the time history of the two fission fragments. The task can be made simpler by assuming that only the time history of the fission fragments from a scissionlike configuration to infinite separation is relevant. The actual acceleration begins before scission and is much more gradual, so the present assumption leads to a lower limit for the acceleration and the radiation produced. The description of the motion of the fragments is determined by solving the differential equation for the two particles under the influence of a repulsive Coulomb potential

$$\frac{1}{2}\mu\dot{r}^2 + \frac{k}{r} = E,$$
(8)

where  $\mu$  is the reduced mass, k is  $z_1 z_2 e^2$ , r is the relative position between the two particles,  $\dot{r}$  is the relative velocity, and E is the energy of the system and is a constant taken to be the asymptotic relative energy of the two fission fragments. This differential equation can be solved by standard methods to yield a time, t(r), which is a function of the relative position of the fission fragments

$$t(r) = \left\{ \sqrt{\frac{\mu}{2E}} \left[ \sqrt{r^2 - \frac{kr}{E}} + \frac{k}{2E} \ln\left( \left( r - \frac{k}{2E} \right) + \sqrt{r^2 - \frac{kr}{E}} \right) \right] \right\}_{r_{\min}}^r.$$
(9)

The expression for t(r) can then be substituted into Eq. (7) to calculate the differential number spectra for the emitted radiation for photons of particular energies. If the long-wavelength limit were valid, one could expand the time retardation term in Eq. (7) to obtain a multipole expansion of the bremsstrahlung which is produced from the Coulomb acceleration of the fission fragments. Unfortunately the long-wavelength limit is not valid in

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the case of the Coulomb acceleration of the two fission fragments, since the time scale for the Coulomb acceleration is very long.

The solid line in Fig. 5 shows the upper limit calculated for the bremsstrahlung production using Eq. (7). The integral was done numerically in position space by using the expression for t(r) found in Eq. (9) and the relationship between position and time in Eq. (8). The calculation is an upper limit since conservation of energy is not taken into account. The dash-dotted line in Fig. 5 is the result of an approximate but more believable calculation for the bremsstrahlung, where a reduction in the emission rate of the radiation has been estimated by including a fragment-fragment barrier penetration effect at each point along the trajectory. The penetrability for each point along the trajectory is

$$P(r) = \frac{1}{1 + e^{2S(r)}},\tag{10}$$

where

$$S(r) = \int_{r}^{r_{\rm CTP}} ds \, \frac{\sqrt{2\,\mu}}{\hbar} \sqrt{[V_{\rm Coul}(s) - T(s)]}. \tag{11}$$

The lower limit r is the position at each point along the trajectory where a particular part of the radiation amplitude is generated and  $r_{\text{CTP}}$  is the classical turning point.  $V_{\text{Coul}}(s)$  is the potential energy at each point along the



FIG. 5. The solid line is the calculated upper limit for high energy photon emission at 120° with respect to the fission axis arising from a simple Coulomb acceleration model, in which the penetration through the fission fragment barrier is not taken into account. The dash-dotted line shows the results of a calculation in which the radiation is produced by Coulomb acceleration, but a barrier penetration effect has been taken into account, to reflect energy conservation. The long-dashed line corresponds to a sudden acceleration calculation of the bremsstrahlung radiation, in which the fission fragments achieve their asymptotic velocity instantaneously. The short-dashed line is the upper limit obtained from the present measurement in the fission-fragment/ $\gamma$ -ray coincidence mode and the dotted line is the measured level of  $\gamma$ -ray yield reported in Ref. [1].

trajectory and T(s) is the corresponding kinetic energy. The inclusion of barrier penetration in the calculation reflects an attempt to include conservation of energy in the calculation. This effect may be thought of as follows. If a high energy photon is produced in the acceleration of the fission fragments away from each other, this  $\gamma$ -ray production has to occur early in the time history of the fission fragments where the acceleration is the greatest. The production of such a  $\gamma$  ray will take away energy that would normally be converted to the kinetic energy of the fission fragments, thereby causing the fission fragments to enter a classically forbidden region. To get out of this region the fission fragments must "tunnel" through the barrier, resulting in a reduced probability for creating high energy photons.

The present calculations were performed for a distribution of the most probable masses and charges arising from the fission of  $^{252}$ Cf. These charge and mass distributions of the fission fragments were taken from the compilation of Wahl [16]. The calculation of the bremsstrahlung was done at an angle of 120° with respect to the fission axis only, to approximate the experimental situation. In converting to an angle-integrated yield isotropic emission was assumed [17]. A more quantitative comparison of the calculation to the experiment would require that the angular dependence be treated more rigorously, by folding the angular acceptance of the NaI detector into the calculation, as well as the angular acceptance of the fission detector.

The results of these dynamical calculations are shown in Fig. 5. If one neglects energy conservation as is done in the Coulomb acceleration calculation indicated by the full curve, one overestimates the yield for  $E_{\gamma} < 80$  MeV. The calculation incorporating an approximate correction due to energy conservation (dash-dotted curve) gives a yield well below the present experimental upper limit, suggesting that several orders of magnitude more fission events than collected in the present work would be needed to observe the process at this level. The long-dashed line in Fig. 5 shows the results of a calculation performed by assuming an impulse approximation for the acceleration of the fission fragments. In this calculation the fission fragments are assumed to achieve their asymptotic velocities instantaneously. This approximation is certainly incorrect but could be regarded as the "ultimate" upper limit for bremsstrahlung production.

The present calculations are quantitatively and qualitatively different from the calculations presented in Ref. [1]. Neither the shape of the energy spectra nor the absolute magnitudes of their calculated bremsstrahlung yields were reproduced in the present work. From the present calculations it is not clear how the earlier calculations (Ref. [1]) could yield similar shapes for both the sudden and Coulomb acceleration models.

### **V. CONCLUSIONS**

Upper limits for the photon emission rate from the spontaneous fission of  $^{252}$ Cf were measured. The result of these measurements indicates no evidence for the emis-

sion of high energy  $\gamma$  rays at the level of  $10^{-9}$  to  $10^{-8}$  photon/MeV fission in the range of  $E_{\gamma} > 30$  MeV, as was reported in Ref. [1]. After the completion of this work a report from Pokotilovskij *et al.* was discovered [18], which yielded an upper limit an order of magnitude below the measurement of Ref. [1].

In addition, the emission probability of high energy gamma rays through a bremsstrahlung process was calculated. The most reasonable of these calculations indicates that the expected level for the observation of energetic  $\gamma$  rays is several orders of magnitude below the sensitivity achieved in the present and previous work.

### ACKNOWLEDGMENTS

We wish to thank Brian McLain, John Behr, and Kurt Snover for their stimulating discussions and helpful suggestions. We would also like to thank Tom Brown for his helpful suggestions regarding the manuscript. This work was supported by the U.S. Department of Energy Grant No. DE-FG06-90ER40537.

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