YU ISSN 0015—3206
FZKAAA 20 (1) 91 (1988)

SEARCH FOR DOUBLE-PHOTON DECAY OF THE ^{109}Ag METASTABLE STATE AT 88 keV

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Received 11 January 1988

UDC 539.16

Original scientific paper

Double-photon decay of the metastable state of ^{109}Ag at 88 keV was studied. In the process photon pairs which share continuously the transition energy are emitted. Radioactive ^{109}Cd was used as the generator of silver atoms in the excited state. A pair of germanium detectors in a 130° geometry and a fast-slow coincidence system with a 128 × 512 × 512 three-parameter pulse height analyzer were used in the measurements. Energy calibration was based on coincidence events of Kα and Kβ X-rays of silver with continuum radiation and on accidental coincidences of the 88 keV gamma rays. The sum spectrum shows no peak at the position where the events due to double-photon decay were expected. The analysis of data yielded an upper limit of the relative transition probability $w_{\gamma\gamma}/w_{\gamma} < 6\cdot10^{-7}$. The theoretical estimates derived from the theory of Grechukhin are $12 \cdot 10^{-7}$ for the square-well potential and $5.6 \cdot 10^{-7}$ for the spherical oscillator potential.

1. Introduction

Ordinary modes of decay of excited nuclear states below the nucleon emission threshold are the well known gamma ray emission, internal electron conversion, and internal pair creation processes. Sachs$^{1)}$ considered double-photon emission in an attempt to explain nuclear isomerism assuming that the two lowest states of the nucleus both have zero angular momentum. It was an extension to the nuclear decay of the earlier pioneering work of Goeppert-Mayer2), who developed the theory of double-photon emission to explain the decay of the hydrogen (and hydrogen like ion) 2s metastable state. Since then several theoretical investigations of double-photon decay were made. The most elaborated theories are given in the papers of Eichler and Jacob3) and of Grechukhin4).

Many measurements were made in attempts to observe double-photon decay in several nuclei, but mostly only upper limits of transition probabilities were obtained. A relatively recent very successful investigation of the double-gamma decay of the first excited O^+ states in ^{90}Zr and ^{40}Ca was made by Schirmer et al. 5).

An earlier attempt to observe double-photon emission in the decay of the metastable first excited state of ^{109}Ag was made by Knauf and Sommer6). They used two NaI(Tl) detectors and observed the two-dimensional field of coincidence events by means of an oscilloscope display. From the data they derived an upper limit of the relative transition probability of double-photon decay of the 88 keV metastable state $w_{\gamma\gamma}/w_{\gamma} \leq 1.9 \cdot 10^{-5}$. The high energy and time resolution of germanium detectors and improved recording and analysing techniques were considered to allow an essentially improved sensitivity of detection of double-decay events. Therefore a remeasurement of the decay was undertaken.

2. *Apparatus and measurements*

Electron capture process in ^{109}Cd (Fig. 1) populates the 88 keV metastable state of ^{109}Ag in 100% of decays7). In the central energy region of the double-photon line few events were expected due to other processes because the metastable state is relatively long lived.

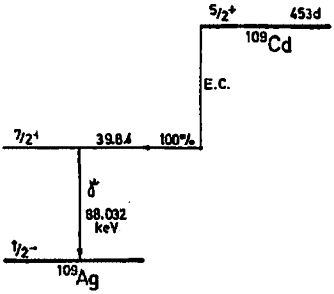

Fig. 1. Decay of ^{109}Cd.

^{109}Cd in a 1 N HCl solution was obtained from New England Nuclear (Boston, Mass., USA). A drop of the solution was placed onto a polyethylene sheet. A small piece of pure cellulose paper, cut to a size of approximately 0.4 mm × × 1.2 mm was placed into the drop and the drop was dried. The small piece of radioactive paper was placed between two foils of polyethylene (about 0.06mm thick) and carefully positioned at the edge of a shield. Two high-purity germanium detectors (supplied by ORTEC, Oak Ridge, Tenn., USA) of nominal sizes 200 mm^2 × × 7 mm thick (Fig. 2) were placed at either side in a 130° geometry. In front of each detector an iron absorber 0.18 mm thick and an aluminium absorber 0.35 mm thick was placed.

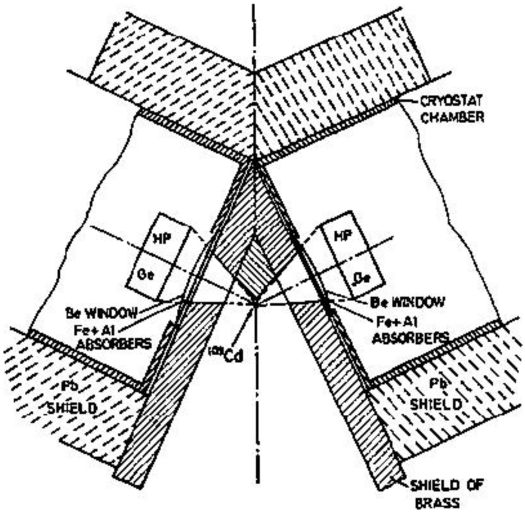

Fig. 2. The experimental setup. The angle between the center-lines of the detectors was 130 degrees.

The pulses from the detectors were fed into a fast-slow coincidence system and a three-parameter 128 × 512 × 512 channel analyzer (Fig. 3). For each coincidence event ($2\tau = 250$ ns) a record was made on paper tape of the time difference (time channel, k_0) and of the amplitudes of pulses from the two detectors (energy channels, k_1 and k_2). Nominal time constants of the amplifiers were 4 μs. The recorded data were analyzed off-line in a IBM 4341 computer and in personal computers.

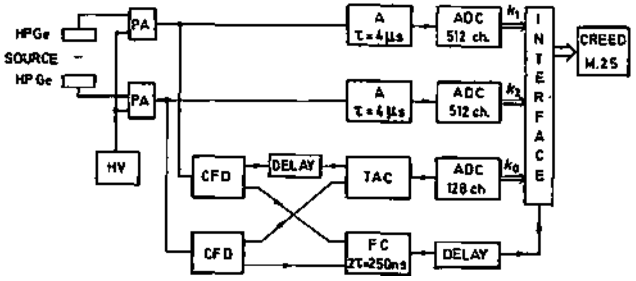

Fig. 3. The electronic system. Notation: HP Ge — high purity germanium detector, HV — high voltage stabilizer, PA — preamplifier, A — amplifier, CFD — constant fraction discriminator, TAC — time to amplitude converter, FC — fast coincidence, ADC — analog to digital converter, CREED — paper tape puncher.

The initial strength of the source was about 32 kBq. The shield between the detectors was made of brass and lead. The gain in either energy branch corresponded to 174 eV per channel. Before the start and after the end of each measurement the system was checked for the peak positions and for counting rates in the singles spectra, and calibrated with a high precision mercury-relay generator.

Sixteen records of coincidence data were made. Each record was transfered into computer files. Total projections of data onto the time (k_0), energy in detector 1 (k_1), and energy in detector 2 (k_2) axes were made separately for each record. The projection spectra were analyzed and the peak positions were compared. Of the sixteen measurements two were rejected because of shifts in the peak positions. The total collection time of data in the fourteen accepted records was 2198 hours.

3. *Analysis of data*

All data in the fourteen records were analyzed as one set. The basis of the energy calibration were the lines due to the coincidences of germanium Kα and Kβ X-rays with the internal bremsstrahlung radiation emitted in the electron capture process, and the peaks due to the accidental coincidences of the 88 keV gamma rays. Since the data from the complete set were used, the peak positions and the peak widths were determined in a consistent and reliable manner. From the data the numbers of in-coincidence events as functions of k_1 and k_2 channel numbers were made. Accurate determination of peak positions in the spectra were made by non-linear least-squares fits using over-channel-width integrated Gaussian functions and taking into account the structure of the peaks. The analysis yielded three calibration points on the k_1 axis and three calibration points on the k_2 axis at energies $E_{1,2} = 22.162$, 24.930, and 88.032 keV. Weighted least-squares fits of straight lines were applied to obtain the energy vs. channel calibration. The accuracy of the calibration in the region of the expected data due to double-photon decay is better than 0.1 channel.

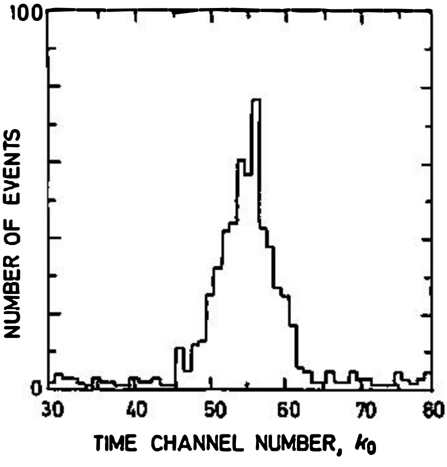

Fig. 4. Time spectrum in the region of the expected double-photon decay events. The peak is due to double bremsstrahlung in the electron capture decay of ^{109}Cd.

The time spectrum of the section of the $k_1 - k_2$ field in the central region where the data due to the double-photon decay were expected shows a peak (Fig. 4). It seems to be mainly due to the double bremsstrahlung in electron capture decay of ^{109}Cd. The peak was analized to determine the interval of time (k_0) channels of a width about 2 standard deviations which was chosen as the in-coincidence time interval.

4. *Results and discussion*

From the table of in-coincidence numbers of counts displayed versus k_1 and k_2 the sum-energy spectrum shown in Fig. 5 was obtained. The limits of summation were set a little above the lines due to the Kβ X-rays of silver. Therefore, in the analysis a range of energy from 27 to 61 keV was included. A least-squares fit to the data using a simple superposition of an energy independent background and of a Gaussian function with a fixed value of the centroid and width (which were derived from the energy calibration procedures), of a variable amplitude, yielded an upper limit of the number of counts due to the double-gamma decay

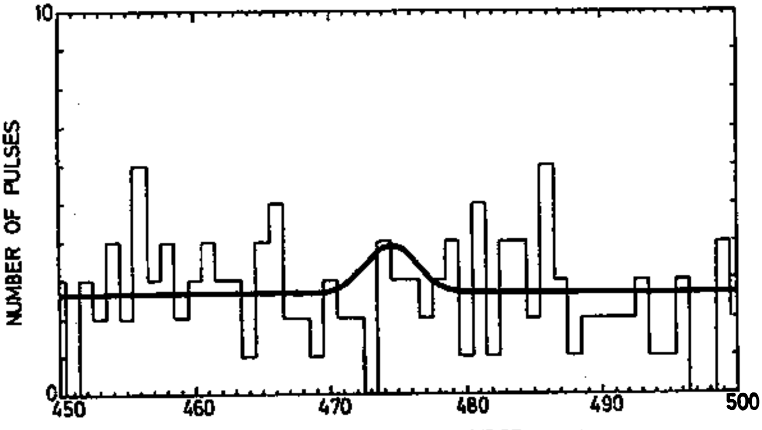

Fig. 5. Sum-spectrum in the region of double photon emission. The curve indicates the expected line intensity assuming the upper limit at the 95% confidence level of 6.6 counts.

$$n_{\gamma\gamma} < 6.6 \text{ counts.} \tag{1}$$

at the 95% confidence level.

The total number of 88 keV gamma rays emitted from the source in the 2198 hours of measurement was $2.96 \cdot 10^{10}$. The solid angle of either detector was about 0.43 sr.

The total relative probability of double-photon decay can be calculated from the expression

$$\frac{w_{\gamma\gamma}}{w_\gamma} = \int_0^{E_0} \int_{4\pi} \int_{4\pi} \frac{1}{w_\gamma} \frac{\mathrm{d}w_{\gamma\gamma}}{\mathrm{d}E\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2}\,\mathrm{d}E\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2. \tag{2}$$

The analysis was made assuming the double-photon decay of the 88 keV metastable state of ^{109}Ag to be of E1E2 or E2E1 type. The direct transition is of E3 type. That implies that the transition probability is expected to have the following energy and angular dependence

$$\frac{\mathrm{d}w_{\gamma\gamma}}{\mathrm{d}E\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2} = \text{const.}\,(1 + a_2 \cos^2 \Theta)\,[E^3 (E_0 - E)^5 + E^5 (E_0 - E)^3] \tag{3}$$

where $E_0 = 88.032$ keV, E and $E_0 - E$ are the energies of the photons, and $a_2 =$ $= -3/29$. The expected number of events in an energy interval from E_1 to E_2 can be calculated from the expression

$$n_{\gamma\gamma} = 2 \int_{E_1}^{E_2} \int_{\Omega_1} \int_{\Omega_2} n_\gamma \frac{1}{w_\gamma} \frac{\mathrm{d}w_{\gamma\gamma}}{\mathrm{d}E\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2} \exp\left[-\frac{C(E)}{\cos\Theta_1} - \frac{C(E_0 - E)}{\cos\Theta_2}\right] \varepsilon_1 (E) \times$$

$$\times\, \varepsilon_2 (E_0 - E)\, \varepsilon_c\, \mathrm{d}E\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2 \tag{4}$$

where n_γ is the total number of 88 keV gamma rays emitted from the source in the measurement, Ω_1 and Ω_2 are the solid angles of the detectors, $\varepsilon_1 (E)$ and $\varepsilon_2 (E_0 - E)$ are the peak efficiencies of the detectors and ε_c is the coincidence efficiency of the system. The exponential factors take into account the absorption of photons of energy E and $E_0 - E$ in their passage into detector 1 and 2, respectively.

The integral in Eq. (4) was evaluated numerically. The peak efficiencies $\varepsilon_{1,2} (E)$ of about 0.95 were determined from the singles spectra. Since the time interval of analyzed data corresponded approximately to two standard deviations, the coincidence efficiency was taken to be 0.95, too. From the above data, taking into account the upper limit of $n_{\gamma\gamma}$, Eq. (1), one obtains the following upper limit of double-photon decay of the 88 keV metastable state in ^{109}Ag

$$w_{\gamma\gamma}/w_\gamma \leq 6 \cdot 10^{-7}.$$

In the calculation of the corresponding theoretical value from the theory of Grechukhin[8,9] it was assumed that the single-photon E3 transition from the $7/2^+$ state at 88.032 keV to the $1/2^-$ ground state was competing only with the E1E2 double-photon transitions via the $5/2^-$ intermediate state of ^{109}Ag at 415.1 keV. Up to about 700 keV excitation no other $5/2^-$ or $3/2^+$ state is known[7]. The contributions from the $5/2^-$ and $3/2^+$ states at higher energies and from states of other angular momenta and parities are expected to be small and were neglected.

The theoretical result

$$w_{\gamma\gamma}/w_{\gamma} = 5.6 \cdot 10^{-7}$$

for the relative transition probability of double-photon decay in ^{109}Ag was obtained from Grechukhin's formula for $7/2^{+} \rightarrow 1/2^{-}$ transition channel as given in the Table 1[9] taking $a_s = 0$ to exclude the contribution from the $3/2^{+}$ state. The value of $f\,(2{,}6,\,10;\,a_t)$ with $a_t = 0.212$ was taken from the graph in Fig. 5[9]. The effective proton charges were taken to be N/A (62/109) for the E1 transition and 1 for the E2 and E3 transitions.

In the evaluation of the nuclear matrix elements Grechukhin used the single-particle model with a spherically symmetric harmonic oscillator potential. If the square-well potential is assumed and Weisskopf's estimates[10] are applied, the result for the relative transition probability of double photon decay of the 88 keV state in ^{109}Ag is

$$w_{\gamma\gamma}/w_{\gamma} = 1.2 \cdot 10^{-6}.$$

Acknowledgements

This work was in part financially supported by the Scientific council of the Republic Croatia and in part by the US National Science Foundation (project PN-734).

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TRAŽENJE DVOJNOG FOTONSKOG RASPADA METASTABILNOG STANJA ^{109}Ag NA 88 keV

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UDK 539.16

Originalni znanstveni rad

Proučavan je dvojni fotonski raspad metastabilnog stanja ^{109}Ag na 88 keV. U tom procesu emitiraju se dva fotona koji kontinuirano dijele prijelaznu energiju. Radioaktivni ^{109}Cd služio je kao generator atoma srebra u uzbuđenom stanju. Par germanijskih detektora u razmještaju na 130 stupnjeva i brzo-spori sudesni sustav s troparamctarskim 128 × 512 × 512 analizatorom impulsnih amplituda primijenjeni su u mjerenju. Energijska kalibracija zasnovana je na sudesnim događajima Kα i Kβ X-zračenja srebra s kočnim zračenjem i na slučajnim sudesnima gama zračenja 88 keV. Zbrojni spektar ne pokazuje vrh na mjestu gdje su očekivani podaci od dvojnog fotonskog raspada. Analiza podataka dala je gornju granicu relativne prijelazne vjerojatnosti od $w_{\gamma\gamma}/w_{\gamma} < 6 \cdot 10^{-7}$. Teorijske vrijednosti izvedene na osnovi Grečuhinove teorije su $12 \cdot 10^{-7}$ za pravokutnu potencijalnu jamu, a $5{,}6 \cdot 10^{-7}$ za sferni harmonički potencijal.