Resonant versus nonresonant nuclear excitation of ^{115}In by positron annihilation

D. B. Cassidy,[1,2,*] A. W. Hunt,[2] P. Asoka-Kumar,[3] B. V. Bhat,[4] T. E. Cowan,[3,†] R. H. Howell,[3] K. G. Lynn,[2,5] A. P. Mills, Jr.,[6] J. C. Palathingal,[7] and J. A. Golovchenko[1]

[1]*Department of Physics, Harvard University, Cambridge, Massachusetts 02138*

[2]*Center for Materials Research, Washington State University, Pullman, Washington 99164-2711*

[3]*Lawrence Livermore National Laboratory, Livermore, California 94551*

[4]*Sri JCBM College, Sringeri, Karnataka, India*

[5]*Department of Physics, Washington State University, Pullman, Washington 99164-2814*

[6]*Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974*

[7]*Department of Physics, University of Puerto Rico, Mayaguez, Puerto Rico 00680*

(Received 4 December 2000; published 1 October 2001)

We have measured the resonant cross section σ_n for nuclear excitation of ^{115}In via the radiationless annihilation of a positron with a *K*-shell electron using a monoenergetic positron beam and a thin In target. We find an upper limit on the resonant cross section $\sigma_n < 4.3\times10^{-26}\,\mathrm{cm}^2$ at a 99% confidence level, compared to the cross section $\sigma_\beta = 1.7\times10^{-25}\,\mathrm{cm}^2$ determined by two previous measurements of nuclear excitation of ^{115}In using the broad spectrum of positrons from the beta decay of ^{64}Cu. Together these results imply the existence of a hitherto unidentified nonresonant channel for nuclear excitation via energetic positrons.

DOI: 10.1103/PhysRevC.64.054603 PACS number(s): 25.30.−c, 34.85.+x, 78.70.Bj

An energetic positron may annihilate with an inner shell atomic electron into one, two, or possibly more photons. An additional radiationless annihilation channel was envisioned in 1951 by Present and Chen [1,2]: a single virtual photon created in the annihilation process may be absorbed by the nucleus of the atom, giving rise to nuclear excitation, if the incident positron energy meets the nuclear resonance criteria. Many measurements [3–10] as well as numerous calculations [11–18] have followed. The experiments to measure the cross section σ_n for radiationless (i.e., resonant) nuclear excitation via positron annihilation have relied on the irradiation of thick targets containing the nuclei of interest using the broad β^+ spectra from radioactive sources. The measurements have yielded unassailable evidence for nuclear excitation via positron annihilation, but the implied values of σ_n are clouded by thick target complications and are generally much larger than the theoretical consensus. We report here the first direct measurement of σ_n using a thin sample and a monoenergetic beam. Our result for σ_n is a factor of 4 less than the cross section determined via radioactive sources, which suggests the existence of a hitherto unidentified and presumably nonresonant (i.e., inelastic) channel for nuclear excitation via positron annihilation.

^{115}In was chosen as the target material because it has been the main focus of study in nuclear excitation by positron annihilation experiments [3,4,7,8,10]. This nucleus has also been extensively studied in nuclear resonance fluorescence experiments [19] on account of its very convenient level structure see (Fig. 1). The 4.49 h half-life of the 336 keV isomer makes it possible to subject the nuclei to intense irradiation and then measure the effects of the irradiation in a low background environment. Although the multipolarity of this state renders it very difficult to populate directly, there are a number of accessible excited states that decay to the isomer. The state we seek to excite via resonant positron annihilation in this work is the 1078 keV level.

When a positron undergoes single quantum annihilation with a *K*-shell electron a photon of energy T is created, where

$$T = E_{e^+} + 2m_0c^2 - B_K. \tag{1}$$

Here E_{e^+} is the positron beam energy, m_0 is the electron rest mass, and B_K is the *K*-shell electron binding energy (27.94 keV for indium). We neglect contributions from other elec-

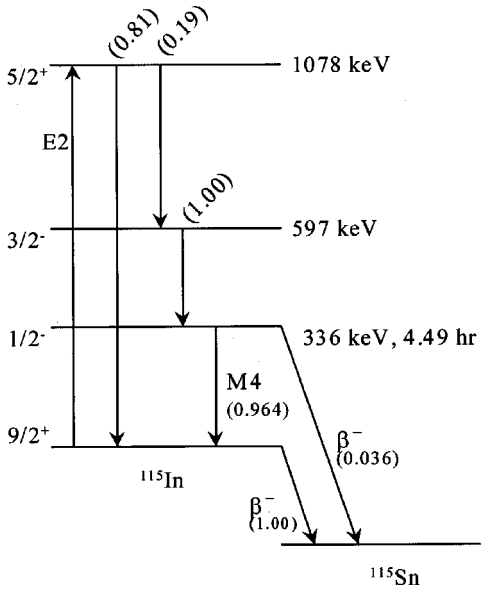

FIG. 1. A partial level diagram of ^{115}In, showing only the transitions investigated in this work. The numbers in parentheses indicate the branching ratios for the relevant transition.

*Email address: cassidy@physics.harvard.edu

†Present address: GA Photonics, 3550 General Atomic Ct., San Diego, CA 92121.

tron shells since they are reduced from the *K*-shell contribution by at least an order of magnitude [9]. Figure 1 shows a partial level scheme for ^{115}In, showing only the transitions relevant to this work. Once excited, the 1078 keV state will promptly decay to the 597 keV state (with a branching ratio of 19%) and thence fully to the isomer, which has a branching ratio to the ground state of 96.4% and decays via a highly converted $M4$ transition ($e/\gamma=0.89$). From Eq. (1), we see that a beam energy of 83.9 keV is required to excite this level by the resonant nuclear excitation process. Thus, following irradiation the sample may be removed to a low background environment where both conversion electrons (47%) and photons (53%) will be emitted as the isomer decays. Observation of this radiation constitutes, in the absence of competing processes, direct observation of nuclear excitation by positron annihilation.

The indium target foil used in our experiment was 5 μm thick with an 18-μm acrylic backing, which we were careful to align as the beam exit side. The foil was mounted in an Al holder that presented a 12.7 mm diameter exposed area. The manufacturer-specified purity of the foil was 99.8%. The target arrangement was mounted inside an accelerating section at the end of a magnetic slow positron beamline and could float at potentials up to 100 kV.

The positrons were created in a bremsstrahlung shower from the Lawrence Livermore National Laboratory 100-MeV electron linac [20]. Positrons created by pair production in the vicinity of a tungsten foil arrangement were moderated and electrostatically focused into a beam, which was then magnetically guided to the target region. Before irradiating the foil, the beam intensity in the target region was measured using a NaI(Tl) scintillation detector. An intensity $I=(5\pm1)\times10^{8}\ e^{+}\ \mathrm{s}^{-1}$ was measured, where the error assignment corresponds to two standard deviations or 95% confidence limit that includes the estimated uncertainties in the measurements of the solid angle and overall detector efficiency and estimated errors associated with corrections for scattered γ rays, pileup, and extrapolation to full beam intensity. During irradiation the beam was monitored for stability with the above detector as well as an integrating dosimeter placed near a constricted region of the transport system. The beam was found to be stable to within $\pm10\%$ during the 11 h irradiation.

Prior to the start of irradiation, a retractable microchannel plate (MCP) with a phosphor screen was extended into the beam path approximately 15 cm in front of the target to locate and focus the beam. The beam spot diameter measured at points of less than 25% of the central intensity was less than 1 cm. The target region layout is shown in Fig. 2. When the channel plate detector was retracted, the beam continued on to the indium and most of it passed through the thin foil. Using two highly collimated plastic scintillator detectors (labeled $D1$ and $D2$ in the figure) we were able to determine whether the beam was passing through the foil and annihilating on the end of the target chamber, or if it was annihilating on the Al sample holder. We optimized the beam location for maximum transmission by steering the beam first to hit the Al and then to pass through the In foil. All tests with

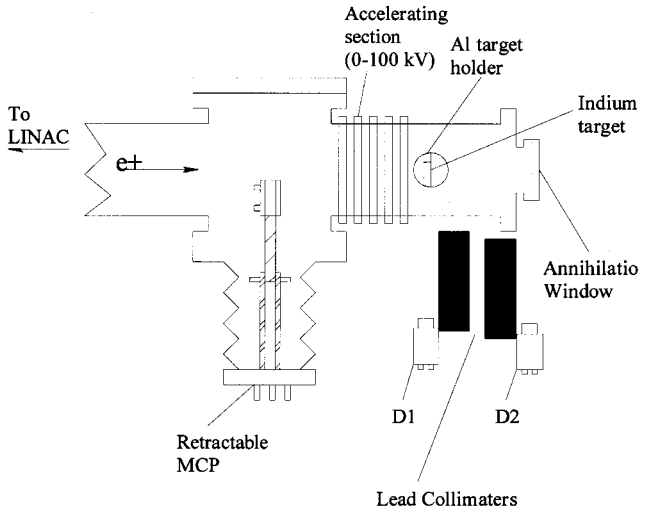

FIG. 2. Target area for irradiating the In foil with monoenergetic positrons. Detectors $D1$ and $D2$ have restricted fields of view due to the presence of the lead collimators. The retractable microchannel plate (MCP) is used to image the beam before it hits the target.

the beam were performed at 83 keV, just below the resonance energy so as not to complicate the cross-section determination.

The irradiation was performed for 11 h at a (calibrated) beam impact energy of 89 keV in a vacuum of $\sim5\times10^{-7}$ torr. This beam energy was chosen in accordance with the positron stopping power of the foil. Positrons were expected to lose $\sim$10 keV in passing through the foil [21], so by implanting the beam at 5 keV above the resonance energy the point of maximum ^{115}In activation was expected to be located close to the center of the In foil. After irradiation the foil was removed from the vacuum system and placed in close proximity to a well-shielded HPGe detector to detect the 336 keV photons from the decay of the isomeric state. The output from the detector was amplified and routed to a CAMAC controlled data acquisition system that downloaded and saved spectra from a memory buffer at regular intervals.

As a test of our system we photoactivated a thick indium sample in the linac bremsstrahlung shower. The ^{115}In decay fluorescence spectrum shown in Fig. 3(a) identifies the expected peak location at 336 keV, the expected peak line shape, and the detector resolution. Spectra from the positron-activated foil and from background are shown in Fig. 3(b). The data in Fig. 3(b) were taken for the first 8 h of the decay of the 336 keV state, or approximately 1.26 mean lives, in order to optimize the signal-to-noise ratio. The background spectrum run lasted 100.5 h and is normalized to the 8 h run. To arrive at an upper limit on the 336 keV peak signal we compute $\chi^2=\Sigma(d-b)^2/b$, where the sum is over the three channels around 336 keV; d is the measured positron-induced signal; and b is the expected number of counts [i.e., the average background of the signal (from 322–348 keV) plus a times the photoinduced signal from Fig. 3(a), normalized to unity when summed over the three peak channels]. We compute the likelihood function $L(a)=\exp\{-\chi^2(a)\}$ for positive values of the fitting parameter a and normalize it to unit area. From the value of a for which $\int_0^a L(a')da'$

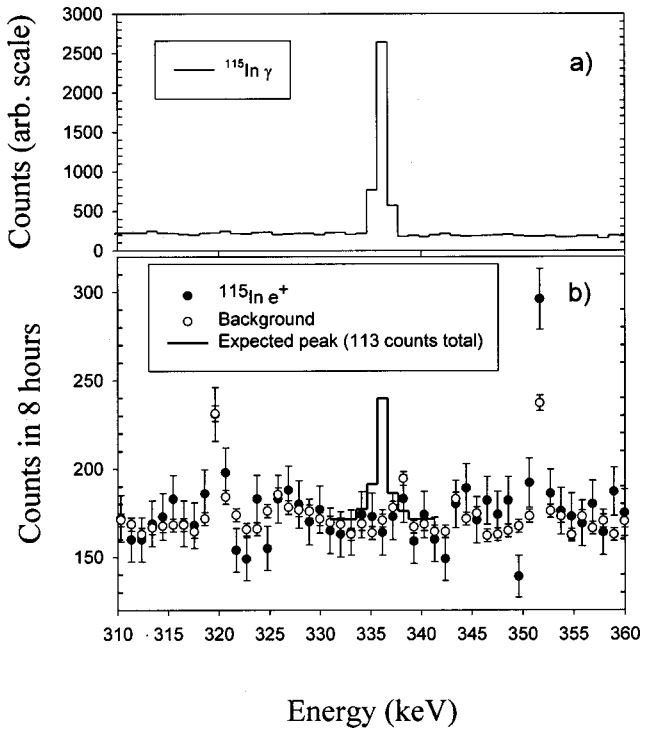

FIG. 3. (a) Photoactivated ^{115}In line shape. (b) Measured positron-activated signal and background. The lines at ~320 and 350 keV are due to radioactivity from the lead detector shielding.

$=0.990$, we find the upper limit on the amplitude of the peak signal over background in Fig. 3(b) is 28.6 at a 99% confidence level. We would expect 113 counts if the resonance cross section were $1.7\times10^{-25}\,\mathrm{cm}^2$ as determined by the positron source experiments [8,10] (see discussion below).

Since this experiment produced a null result it should be pointed out that the data presented here do not represent the only set collected. An earlier run, which also yielded no signal, alerted us to the fact that the expected count rate (see Fig. 3) might not be as large as expected. The data collected from this earlier run are not shown here for several reasons. The statistics present in that data set were not good due to a different experimental procedure that was adopted. In this case we attempted to measure both the emitted photons and conversion electrons from the decay of the isomer. This technique proved to be less efficient than optimizing for one type of signal or the other, and the experiment was considerably simplified by taking photon data only. There exist a number of other procedures that, ideally, we would liked to have undergone, such an additional runs at varying target thickness and beam energies, and perhaps even the investigation of other isotopes. However, both time and cost considerations prevented us.

The irradiation process creates isomeric states such that after an irradiation time $t_{\mathrm{irrad}}=11\,\mathrm{h}$ the total number of isomers present is

$$N_b(t_{\mathrm{irrad}})=\kappa\lambda_b[1-\exp\{-\lambda_b t_{\mathrm{irrad}}\}]/\lambda_b\,, \tag{2}$$

where we use $N_b(0)=0$ and the subscripts a and b refer to ground and excited states, respectively, and $\lambda_b=0.154\,\mathrm{h}^{-1}$ is the decay rate of the 336 keV state. The time between the end of the irradiation and the beginning of the counting was 9 min and is neglected. $\kappa=0.19$ is the branching ratio to the isomer from the 1078 state. The constant λ_a is the rate at which nuclei are excited and is given by

$$\lambda_a=I\sigma_n N_a(0), \tag{3}$$

where I is the positron beam intensity, σ_n is the resonance cross section, $N_a(0)$ is the number of available target atoms per unit area, and we use the fact that the typical rate at which any one nucleus becomes excited is negligible compared to λ_b.

Since we are assuming in our analysis that this is a resonant process, we must be cautious when defining the number of available target atoms, $N_a(0)$. We include as the thickness of our foil only the *effective thickness*. d_{eff}, which is simply that fraction of the foil through which the passing positrons are in the resonant energy range. This is clearly determined by the resonance width and the stopping power of the material. At the resonant positron energy of 83.9 keV the stopping power is $(dE/dx)_{\mathrm{res}}=18\,980\,\mathrm{keV\,cm}^{-1}$ [21]. As pointed out by Grechukhin and Soldatov [11], the resonance width is given by

$$\Gamma=\Gamma_n+\Gamma_k\approx\Gamma_k\,, \tag{4}$$

where the subscripts n and k refer to the nuclear and k levels, respectively. The nuclear width, $\Gamma_n=6\times10^{-4}\,\mathrm{eV}$ [22], is orders of magnitude smaller than the electron shell width $\Gamma_k=7.3\,\mathrm{eV}$ [23], which therefore dominates. The appropriate evaluation of d_{eff}, is then,

$$d_{\mathrm{eff}}=\Gamma_k/(dE/dx)_{\mathrm{res}}=3.85\times10^{-7}\,\mathrm{cm}^{-2}. \tag{5}$$

The effective number of target atoms presented to the beam is given by

$$N_a(0)=N_A f\rho d_{\mathrm{eff}}/A=1.41\times10^{16}\,\mathrm{cm}^{-2}. \tag{6}$$

Here $N_A=6.02\times10^{23}$ is Avogadro's number, $f=0.9572$ [24] is the natural abundance of ^{115}In, and $\rho=7.29\,\mathrm{g/cm}^3$ and $A=115$ are the density and atomic weight of indium, respectively. We obtain our experimental value for $N_b(t_{\mathrm{irrad}})$ by using the 99% confidence level upper limit of 28.6 detected isomeric decays. Since we have used data integrated to $t_{\mathrm{count}}=8\,\mathrm{h}$ we have only counted a fraction $1-\exp\{-\lambda_b t_{\mathrm{count}}\}=0.708$ of $N_b(t_{\mathrm{irrad}})$. The upper limit on the detected rate is corrected by dividing it by the product of $1-\exp\{-\lambda_b t_{\mathrm{count}}\}$, the measured detection efficiency, $\varepsilon=0.12\pm0.005$, the branching ratio of the isomer to ground state (0.95) and the photon fraction (0.53) to yield $N_b(t_{\mathrm{irrad}})<668$. The upper limit for the resonant cross section is then

$$\begin{aligned}\sigma_n&=N_b(t_{\mathrm{irrad}})\lambda_b\kappa^{-1}N_a(0)^{-1}F^{-1}(1-\eta)^{-1}\\&\quad\times[1-\exp\{-\lambda_b t_{\mathrm{irrad}}\}]^{-1}\\&<4.3\times10^{-26}\,\mathrm{cm}^2\ [99\%\ \text{confidence level}],\end{aligned} \tag{7}$$

where we are using a 3 standard deviation or 99.7% confidence level positron flux lower limit $F > 3.5 \times 10^8\ e^+\ s^{-1}$ in the evaluation, and $\eta = 0.15$ is the fraction of the positrons that are transmitted or backscattered without being moderated to the resonance energy [25].

We are aware of five published experimental determinations of the ''resonant'' cross section of nuclear excitation of ^{115}In [3,4,7,8,10] by positron annihilation with a *K*-shell electron. Two of the results [8,10] are reevaluations of earlier works [7,4] that we believe represent the most accurate determinations of the rate for positron-induced excitation of ^{115}In using the continuous spectra of positrons emitted from a radioactive source. The reanalysis in Ref. [10] involved correcting for the effective thickness of their target foil, defined as in Eq. (5). The work of Vishnevskii *et al.* [7] was reevaluated [8] by considering a more accurate representation of the source distribution in the target material, leading to an increased cross section. These authors recognized the need to consider the *K* hole in the resonance width but did not take account of the effective thickness of their target material. Since both experiments used stopping targets of a similar thickness, we may apply the analysis of Saigusa and Shimizu [10] to the data of Vishnevskii *et al.* [8]. The resulting deduced resonance cross sections from these two experiments are almost identical, both yielding $\sigma_\beta = 1.7 \times 10^{-25}\ \mathrm{cm}^2$, which is four times greater than our upper limit.

Various competing processes that might be present in the source-based experiments [8,10] do not seem to be responsible for this disparity. Barring an unusually large and unexpected error either in the present result or simultaneously in both source experiments [8,10], a remaining possibility is that the large energy distribution of the β^+ particles from a ^{64}Cu source ($E_{\max} \sim 650\ \mathrm{keV}$), allows some lower cross section inelastic processes to dominate in the radioactive source-based experiments. In these processes, the resonance constraint on the positron energy would be relaxed and the number of positrons that can take part in the excitation process would be greatly increased. The simple process of emitting a bremsstrahlung photon during the nuclear excitation [12] is too improbable to make any significant contribution to the rate [13].

Our measurement allows an unambiguous comparison with theoretical estimates of the resonant cross section for nuclear excitation. We make no comparison to the theoretical work of Present and Chen [1] and Watanabe, Mukoyama, and Shimizu [4] because they used the wrong nuclear transition ($E1$) and were based on a two-step approximation to the virtual annihilation. Of the remaining theories our upper limit is approximately consistent with the cross section calculated by Kolomietz and Fedotkin [16] and is consistent with the small cross section from the theory of Grechukhin and Soldatov [11]. In view of our result it is not surprising that there has been a persistent disparity between the experimental and theoretical results, since they are evidently not describing the same process.

Although we are unable to shed light on the details of the excitation mechanism, our upper limit on the resonant cross section implies that some sort of nonresonant process is occurring in the source-based experiments. Indeed, the combination of our result with these experiments is a compelling indication of the existence of some as yet unidentified process. In order to investigate this further it would be of interest to scan a more intense beam over several hundred keV above the resonant energy in both a thin and a thick target. However, the experiment to resolve the nuclear excitation mechanism must probably await the implementation of a positron beam with an intensity of at least $10^{10}\ e^+\ s^{-1}$.

We gratefully acknowledge helpful discussions with R. S. Raghavan and W. Stoeffl. This work was supported by the National Science Foundation under Grant Nos. DMR-9623610 and PHYS-9505886, and was also performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

[1] R. D. Present and S. C. Chen, Phys. Rev. **83**, 238 (1951).
[2] R. D. Present and S. C. Chen, Phys. Rev. **85**, 447 (1952).
[3] T. Mukoyama and S. Shimizu, Phys. Rev. C **5**, 95 (1972).
[4] Y. Watanabe, T. Mukoyama, and S. Shimizu, Phys. Rev. C **19**, 32 (1979).
[5] Y. Watanabe, T. Mukoyama, and R. Katano, Phys. Rev. C **23**, 658 (1981).
[6] Y. Watanabe, T. Mukoyama, and S. Shimizu, Phys. Rev. C **21**, 1753 (1979).
[7] I. N. Vishnevskii, V. A. Sheltonozhskii, V. P. Syvato, and V. V. Trishin, JETP Lett. **30**, 366 (1979).
[8] I. N. Vishnevskii, V. A. Sheltonozhskii, V. P. Syvato, V. V. Trishin, and A. I. Feoktistov, Sov. J. Nucl. Phys. **38**, 161 (1983).
[9] I. N. Vishnevskii, V. A. Sheltonozhskii, and V. M. Kolomietz, Sov. J. Nucl. Phys. **19**, 101 (1988).
[10] T. Saigusa and S. Shimizu, Hyperfine Interact. **89**, 445 (1994).
[11] D. P. Grechukhin and A. A. Soldatov, Sov. Phys. JETP **47**, 6 (1978).
[12] R. S. Raghavan and A. P. Mills, Jr., Phys. Rev. C **24**, 1814 (1981).
[13] A. Ljubicic, M. Kremar, K. Pisk, and B. A. Logan, Phys. Rev. C **30**, 2109 (1984).
[14] K. Pisk, M. Kremar, A. Ljubicic, and B. A. Logan, Phys. Rev. C **32**, 83 (1985).
[15] V. M. Kolomietz, J. Phys. G **16**, 615 (1990).
[16] V. M. Kolomietz and S. N. Fedotkin, Sov. J. Nucl. Phys. **52**, 272 (1990).
[17] R. Horvat, K. Pisk, and B. A. Logan, Phys. Lett. **214**, 490 (1988).
[18] I. S. Batkin and T. A. Churakova, Sov. J. Nucl. Phys. **35**, 161 (1982).
[19] P. Von Neumann-Cossel *et al.*, Phys. Lett. B **266**, 9 (1991).
[20] For example, R. H. Howell, in *Positron Beams and Their Ap-*

plications, edited by P. Coleman (World Scientific, Singapore, 2000), and references therein.
[21] *Stopping Powers for Electrons and Positrons*, International Commission on Radiation Units and Measurements (ICRU, Washington, 1984), p. 4.
[22] F. S. Dietrich *et al.*, Nucl. Phys. **66**, 230 (1965).
[23] W. Bambynek *et al.*, Rev. Mod. Phys. **44**, 716 (1972).
[24] *Table of Isotopes*, 7th ed., edited by V. S. Shirley (Wiley, New York, 1986).
[25] This is a cautious estimate based on the backscattering and energy loss profile as calculated from the POS-SPRITE program. K. A. Ritley, V. J. Ghosh, K. G. Lynn, M. McKeown, and D. O. Welch, Comput. Phys. Commun. **109**, 93 (1998).