PHYSICAL REVIEW C VOLUME 54, NUMBER 5 NOVEMBER 1996

Internal bremsstrahlung spectrum of ^{139}Ce

M. M. Hindi and R. L. Kozub
Physics Department, Tennessee Technological University, Cookeville, Tennessee 38505
(Received 15 May 1996)

The internal bremsstrahlung spectrum which accompanies the electron capture decay of ^{139}Ce to the first excited state of ^{139}La has been measured in coincidence with the nuclear gamma ray which deexcites that state. The measured intensity above 42 keV is found to be (1.070 ± 0.024) times that predicted by the recent calculations of Surić *et al.* The Q_{EC} value is found to be 264.6 ± 2.0 keV. [S0556-2813(96)02411-9]

PACS number(s): 23.40.−s, 14.60.Pq, 27.60.+j

I. INTRODUCTION

Electron capture is a weak interaction process in which a nucleus decays by capturing an atomic electron and ejecting an electron neutrino. A small fraction of such decays proceed radiatively, with the neutrino, atom, and emitted photon sharing the decay energy statistically. The produced radiation, which has a continuous energy spectrum, is known as internal bremsstrahlung (IB) and the decay process is referred to as internal bremsstrahlung electron capture (IBEC). The study of IBEC can yield information on the weak interaction, nuclear parameters, and atomic wave functions; accurate isobaric atomic mass differences can also be deduced from the end-point energy of the IB spectrum. A thorough review of the theory and experimental status of IBEC up to 1977 is given by Bambynek *et al.* [1]. As pointed out by that review, while the main features of IBEC are generally understood, there is still a great need for experimental work to test the details of the theory. In particular, Bambynek *et al.* point out that precise measurements of normalized IB spectra are very much needed, as well as measurements of partial spectra that accompany the capture of electrons from specific atomic subshells.

Interest in IBEC decays was renewed after De Rújula's suggestion in 1981 [2] that the shape of the IB spectrum near the end point be used to search for neutrino mass. The sensitivity is obtained for decays with low Q values, where the IB spectrum is dominated by capture from p orbitals and could be strongly enhanced by the presence of $p \rightarrow s$ poles in the electron propagator, if a decay with the right energy is found. Riisager *et al.* [3] carried out detailed measurements on the IBEC decay of ^{193}Pt to test De Rújula's extensions of the Martin-Glauber theory [4,5]. Although the calculations reproduced the measured shapes of the IB spectra, the magnitudes were off by factors of 1.5–2. The upper limit which they quote on the mass of the electron neutrino is 500 eV, at the 90% confidence level. Springer, Bennett, and Baisden [6] measured the IBEC spectrum of ^{163}Ho and compared the data with a model similar to that used by Riisager *et al.* They quote an upper limit on the mass of the electron neutrino of 225 eV, at the 95% confidence level, and conclude that improvements of this result are severely limited by uncertainties in atomic interference effects.

The above limits on the mass of the electron neutrino are much higher than the corresponding limits of 5–10 eV [7] on the mass of the electron antineutrino and could certainly be improved upon, if, among other things, the uncertainties in the calculated IB spectra could be removed. Recently, Surić, Horvat, and Pisk [8] reformulated the relativistic theory of IBEC and developed a numerical code to calculate IBEC matrix elements. They included all relativistic and screening effects within an independent-particle approximation. They examined the case of ^{163}Ho and found a simpler structure of the total IBEC spectrum and a smaller interference suppression than predicted by the previous calculations [3,6]. Their calculation for ^{192}Pt, however, was still lower than the data by about a factor of 2.

To test Surić *et al.*'s reformulation of the IBEC theory, we have recently compared their calculation with the measured IB spectrum of ^{125}I above the 1s end point [9,10]. It was found that the calculation reproduces the shape and relative intensity of the partial IB spectra to within the experimental error of a few percent and the absolute intensity to within 15%. In [10], where we reported the measured intensity to be 0.86 ± 0.10 times the calculated intensity, the IB matrix elements were calculated in the field of the parent (^{125}I) atom. It is more appropriate to calculate the IB matrix element in the field of the *daughter* atom (^{125}Te); using these matrix elements we find that the measured intensity is 1.00 ± 0.12 times the calculated intensity, in even better agreement with Surić's calculation than reported before.

The ^{125}I data were taken in the course of a search for 17 keV neutrinos [9] and since the emphasis there was on the portion of the spectrum above the 1s binding energy, the low energy portion of the spectrum was attenuated severely, for practical purposes. Hence it was not possible to test the Surić calculation in the x-ray region. The ^{192}Pt and ^{163}Ho IB data, however, are near the x-ray region (and so will probably be future data from any experiment that seeks to lower the limit on the electron-neutrino mass) and therefore it is important to extend tests of the Surić calculation down to that region. In this paper we report a measurement of the absolute intensity of the IB spectrum which accompanies the EC decay of ^{139}Ce. The decay scheme of ^{139}Ce is shown in Fig. 1. It is an allowed decay which proceeds exclusively to the first excited state with an end-point energy of $\approx$99 keV. This energy is low enough that the IB is dominated by p capture. The nuclear γ ray that deexcites the first excited state has an energy (165.9 keV) which is higher than the IB end point; in a singles spectrum the low intensity IB would be overwhelmed by the Compton tail of that γ ray. Thus, to separate

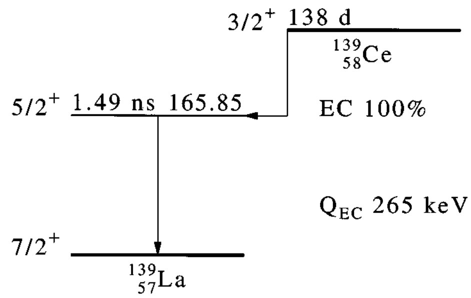

FIG. 1. Decay scheme of ^{139}Ce. The level energy is in keV.

the IB from the Compton tail of the nuclear γ ray, a coincidence measurement was performed in which the IB was detected in one detector and the γ ray, simultaneously, in another.

II. EXPERIMENTAL PROCEDURE

The IB data presented in this paper were obtained during the course of an experiment to measure the probability of double K-shell vacancy production in the electron capture decay of ^{139}Ce [11]. The essential features of the experiment will be reproduced here for completeness; further details can be found in Ref. [11].

Sources of approximate strengths 10–40 nCi were prepared by evaporating several drops of an active solution of $CeCl_3$ in 1 N HCl onto cellophane tape, and sealing the dried tape with a similar piece. These sources were sandwiched between two intrinsic Ge (Gamma-X) detectors placed face to face. One of the detectors had a 43.5 mm diameter, 45.3 mm depth, and a 3 mm crystal-to-endcap distance, and the other a 54.2 mm diameter, 39.2 mm depth, and 5 mm crystal-to-endcap distance. Both detectors had 0.5 mm Be windows; these were thick enough to stop Auger and conversion electrons associated with ^{139}Ce decay. The resolution of the detectors at the La $K\alpha$ line ($\approx$ 33 keV) was 715 eV.

The detectors were connected to amplifiers equipped with pileup rejection circuitry (ORTEC 572). A standard circuit was used to generate coincidence signals. The digitized energy and fast-timing signals from each detector were recorded event by event on magnetic tape for later analysis. Most of the recorded data consisted of coincidences only, with singles-plus-coincidence data recorded in short runs every few days. Typical singles counting rates were about a few hundred counts per s. The total counting time was approximately 90 days.

III. DATA ANALYSIS AND RESULTS

The data on the magnetic tapes were replayed to generate singles and prompt coincidence spectra. The energy gains of the two detectors were matched and stabilized in software and the spectra in coincidence with the 166-keV γ ray were subjected to a two-dimensional (2D) software gate in the energy vs risetime plane to reduce pileup, as described in Ref. [11]. Figure 2 shows a sample singles spectrum [Fig. 2(a)], and the total coincidence spectra gated on the $K\alpha$ x ray [Fig. 2(b)] and the 166-keV γ ray [Fig. 2(c)]. The $K\alpha$-(166-) gated coincidence spectrum is the sum of two spectra, one from each detector gated on the $K\alpha$ (166-keV γ) in the other detector.

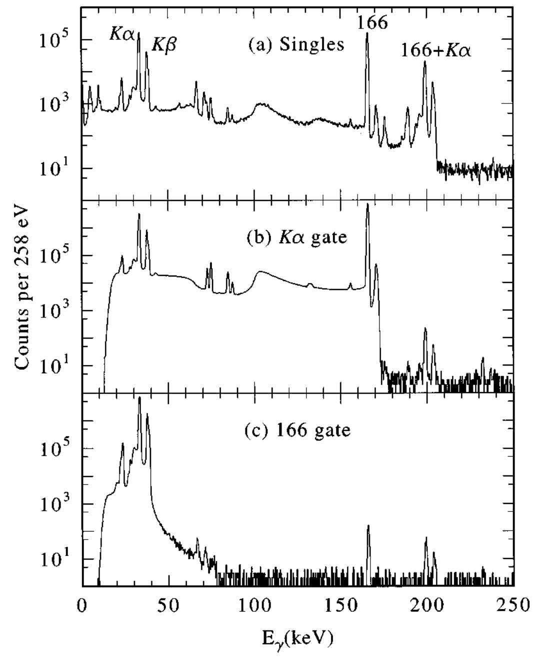

FIG. 2. A sample singles spectrum and total gated coincidence spectra from the EC decay of ^{139}Ce. Energies are in keV. The K x rays belong to La.

The total IB spectrum was obtained from the spectrum in coincidence with the 166-keV γ ray [Fig. 2(c)] after subtracting the contributions of the following: (1) residual pileup coincidences; these are mostly due to real coincidences between the 166-keV γ ray and a $K\alpha$ or a $K\beta$, followed by a random photon which adds to the $K\alpha$ or $K\beta$ signal, (2) accidental coincidences between the 166-keV γ ray and a random single photon in the other detector, and (3) true coincidences between background signals in the 166-keV gate region and the IB detector. The residual pileup spectrum is, to a very good approximation, a convolution of the coincidence spectrum with the singles spectrum. This is so because the combination of the electronic and software pileup rejection gives a resolving time of about 100 ns, which is short enough to make the combination of two pulses arriving within that time interval appear as the full energy sum. Nevertheless, we have employed a new Monte Carlo simulation procedure which improves on the shape of the calculated pileup spectrum, especially in the region of peak +peak sums. The shape of the amplifier signal was digitized and later approximated by a semi-Gaussian. A random pulse with a height distributed according to the coincidence spectrum is generated. Another random pulse with a height distributed according to the singles spectrum is generated within a uniformly distributed random time between 0 and 100 ns. The two pulses are added and the location of the maximum is found numerically. Repeating this procedure gives the desired pileup spectrum. The spectra thus obtained give a shape for the peak+peak sums which agrees very well with that of measured pileup spectra. The intensity of the pileup spectrum was obtained by normalizing the Monte Carlo simulated $K\alpha+K\alpha$ peak to the corresponding peak in the coincidence spectrum, after subtracting the contribution

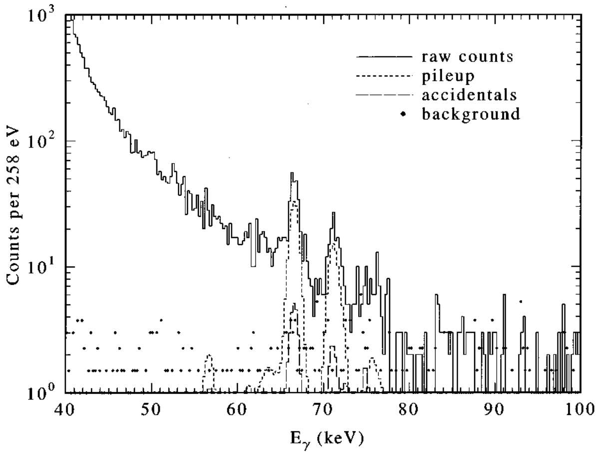

FIG. 3. The raw IB spectrum (solid line) and the normalized components which were subtracted from it to get the net spectrum: pileup (short-dashed line), accidental coincidences (long-dashed line), and background coincidences (solid circles).

of accidental coincidences to the latter (see below).

Accidental coincidences were subtracted by assuming the shape of the accidentals spectrum to be the same as that of the singles spectrum and normalizing this spectrum to the yield of accidental 166-keV–166-keV coincidences. The background coincidence spectrum was generated by setting a gate on a region just above the 166-keV line. Figure 3 shows the raw IB spectrum together with the normalized pileup, accidentals, and background spectra. Clearly, IB dominates from the region of the $3p \rightarrow 1s$ pole up to 65 keV, while the other components dominate beyond that energy.

In principle, the time resolution of the pileup rejection circuitry could depend on the relative sizes of the pulses. To ascertain that the Monte Carlo generated spectrum, which assumes the same pulse-pair resolution regardless of relative pulse size, adequately accounts for pileup in the 40–60 keV region, we compared the IB yield from a run using a weak source with that from a run using a stronger source. Figure 4 shows the ratio of the normalized IB yields in the 40–60 keV region, together with the ratio of accidental 166-keV–166-keV coincidences for the two runs. While the accidental coincidences for the ''high count rate'' run are a factor of 2.35 ± 0.29 higher than those for the low count rate run, the ratio of the yields in the 40–60 keV region are consistent with unity, thus confirming the adequacy of our pileup-subtraction procedure.

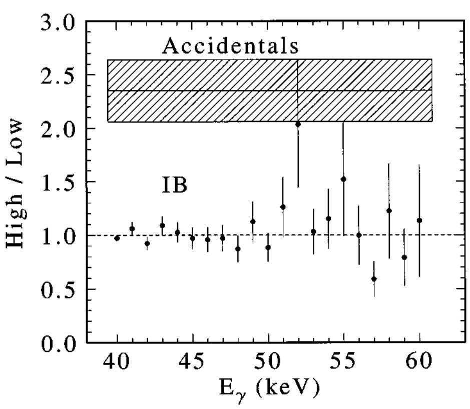

FIG. 4. Ratio of IB yield for the high count rate run to the low count rate run. The shaded block shows the 1σ range of the ratio of accidental 166-keV–166-keV coincidences for the two runs.

The IB yield in the region just above the pole is about 4–5 orders of magnitude smaller than the yield of the $K\alpha$ and $K\beta$ x rays, just below; a legitimate question, therefore, is the following: is it possible that a substantial portion of the yield just above the x-ray peaks is due to the response function of the detector, i.e., could an instrumental long term high energy tail account for a substantial portion of the counts observed above the x-ray peaks? It is very difficult to test the response function of the detector at that level in that energy region. All radioactive sources are accompanied by internal or external bremsstrahlung at a level comparable to that of ^{139}Ce. It is possible to verify that the yield in the IB region is due to higher energy γ rays, rather than the tail of lower energy x rays, by means of differential attenuation, but the time required for a statistically meaningful result would have been prohibitive. That the yield above the x ray peaks is primarily due to IB can nevertheless be inferred from Fig. 2(b), which shows that the yield above the 166-keV γ ray[1] in coincidence with $K\alpha$ [which has the same counts as the $K\alpha$ peak in Fig. 2(c)] is less than 10^{-6} of the 166-keV peak, and that yield, within statistics, can all be attributed to pileup and accidental coincidences. Thus any high energy tail (above the usual Gaussian tail) must be smaller than $\sim 10^{-7}$ of the peak height. The region of the detector sampled by the 166-keV γ ray includes as a subset that sampled by the x rays; hence one would not expect the average amount of charge collected per unit energy deposited in the detector (and the statistical variation thereof) to differ substantially between the 166-keV γ ray and the x rays. This is confirmed by the measured linearity of peak centroid and variance with γ-ray energy, from ~ 5 to 200 keV. Accordingly, the high energy tail, if present, is less than $\sim 1\%$ of the IB yield in the 40–50 keV energy region, and well within the statistical error throughout the investigated IB region.

The net IB yield, divided by the number of K captures and by the width of the energy bin (0.258 keV) is shown in Fig. 5. The number of K captures was deduced from the $K\beta$ yield [Fig. 2(b)] and using a fluorescence yield $\omega_K = 0.907$ [12] and a $K\beta$ fraction of 0.193 [13]. The normalized experimental spectrum was fit with a theoretical spectrum calculated using the code developed by Surić *et al.* [8], and convoluted with the response function of the detector. The response shape was parametrized as the sum of a Gaussian peak, an exponential tail on the low energy side, Ge $K\alpha$ and $K\beta$ x-ray escape peaks, and a flat tail. Both the exponential tail and the flat tail were convoluted with a Gaussian that had the same width as the main Gaussian peak. The shape parameters were interpolated as a function of photopeak energy from the shape parameters obtained for the ^{133}Ba x-ray, 53-keV and 81-keV lines. The variation of photopeak efficiency with energy was also obtained using the ^{133}Ba source, after correcting for summing effects. In calculating the theoretical IB yield the atomic binding energies (of

[1]The peak just above the 166-keV peak is due to true summing of the 166-keV γ ray with L x rays.

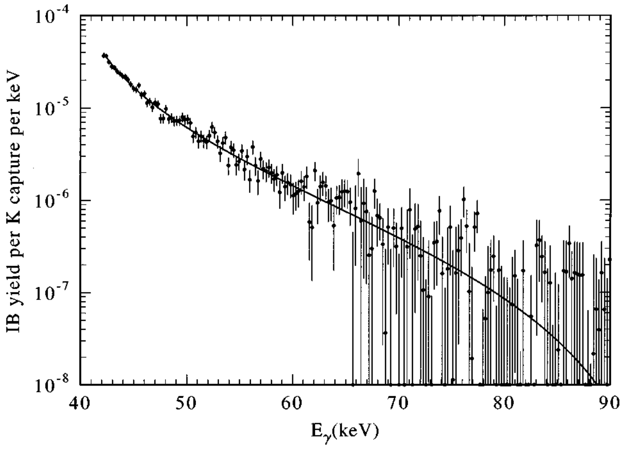

FIG. 5. Net IB spectrum of ^{139}Ce above the $K\beta$ energy. The solid line is a normalized fit using the Surić *et al.* calculation. The (multiplicative) normalization factor for the theoretical spectrum is 1.070 ± 0.024.

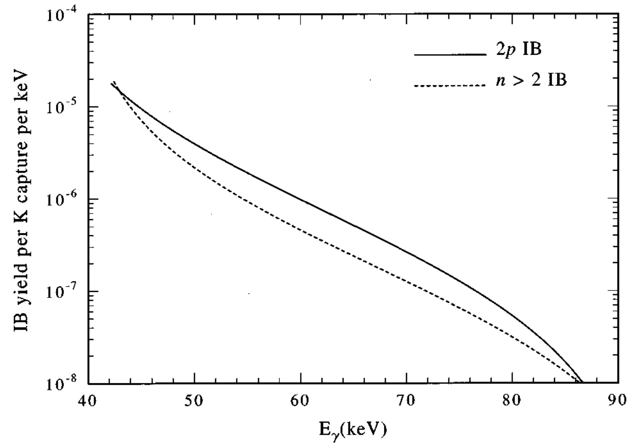

FIG. 6. A comparison between the $2p$ component (solid line) and the $n\geqslant3$ components (short-dashed line) of the theoretical IB spectrum. Note the different energy dependences in the 40–50 keV region.

La) which enter into the phase space factor were obtained from the tables of Larkins [14]. The wave functions which enter into the calculation of the energy-dependent IB matrix elements were obtained in the field of the daughter (La) atom, while the normalizing electronic wave function which determines the (non-radiative) K-capture rate was obtained in the field of the parent (Ce) atom.

Two parameters were allowed to vary freely in the fit: (1) a multiplicative normalization factor A, and (2) the end-point energy Q_0 of the transition. Figure 5 shows the best fit to data in the 42–91 keV energy range. The resulting χ^2 per degree of freedom is 192.9/190; the best fit parameters are $A=1.070\pm0.022\pm0.010$ and $Q_0=98.7\pm1.7\pm1.0$ keV, where the first error is statistical and the second systematic. The latter was estimated from the change in the fitted parameters with reasonable variations in background, accidentals and pileup subtractions, the relative photopeak efficiency, and the shape parameters used in convoluting the theoretical IB spectrum. The sensitivity of the fitted parameters to the energy interval selected for fitting was also explored: fits in which the high end was varied between 60 keV and 91 keV and the low end varied between 40 keV and 45 keV were conducted. The best fit values were all consistent with the numbers quoted above. In particular, a fit over the energy range 42–63 keV (which excludes the region where one might question the use of the χ^2 estimator because of the small number of counts per bin) gives $A=1.051\pm0.027$ and $Q_0=100.8\pm2.6$ keV, and a χ^2 per degree of freedom of 87.7/80.

In the above fits the relative contribution of IB from different shells was fixed (to the value given by the Surić *et al.* calculation [8]). Two independent attempts were made at extracting the relative contribution of $2p$ IB to the total IB spectrum from the data. In one attempt the contribution of IB from the $n=3$ shell was multiplied by a scaling parameter r which was allowed to vary freely in the fit. A fit in the region 42–91 keV gave $r=0.79\pm0.35$, $A=1.15\pm0.16$ and $Q_0=98.1\pm2.2$ keV. That is, this fit suggests that the relative contribution of the $n=3$ shell to the total IB spectrum is less than that given by the Surić calculation (or, equivalently, that the relative contribution of the $2p$ IB is larger than that given by the Surić calculation), though, within the indicated error, not inconsistent with it. As Fig. 6 shows, the sensitivity to the $n=3$ contribution comes about because of the different energy dependences of the $2p$ and $3p$ IB spectra (the latter being the dominant contributor to the $n\geqslant3$ spectrum) in the vicinity of the $3p\rightarrow1s$ pole; this pole contributes to the $3p$ spectrum, but not to the $2p$ spectrum.

Second, the ratio of $2p$ IB to the total IB was obtained by comparing the yield of IB in coincidence with the $166+L$ sum peak to that with the 166-keV γ peak. As Fig. 2(b) shows, the spectrum gated on $K\alpha$ shows coincidences not only with the 166-keV γ ray, but also with the 166-keV γ ray summed with the L x ray which sometimes depopulates the L hole left after the $K\alpha$ emission ($\omega_{L_{2,3}}=0.103$ [12]). A coincidence spectrum gated on $K\beta$ showed only the 166-keV peak, not the $166+L$ peak, thus confirming the origin of the latter. From the ratio the $166+L$ peak to the 166-keV peak (in coincidence with $K\alpha$) one obtains the absolute efficiency for L x-ray detection. Using that efficiency one can obtain the $2p$ contribution to the total IB spectrum by comparing the area of the $166+L$ peak to that of the 166-keV peak in a spectrum gated on the IB. For IB in the region 40.63–50.85 keV we find the $2p$ fraction to be 0.66 ± 0.16, whereas Surić's calculation predicts a ratio of 0.481 in that region. That is, the measured $2p$ fraction is 1.37 ± 0.33 times that predicted by Surić *et al.'s* calculation, or, equivalently, the measured fraction of $n\geqslant3$ IB is 0.66 ± 0.31 times that predicted by the calculation. This latter number is in agreement with that obtained from the spectral shape analysis, though the statistical error is large in both cases.

IV. DISCUSSION AND CONCLUSIONS

The measured end-point energy gives a Q_{EC} value of $264.6\pm1.7\pm1.0$ keV. This value is lower than the value 277 ± 9 keV, deduced from an unweighted average of many K-capture probability measurements [15], but is in agreement with the value 265 ± 5 keV given in the mass evaluations of Wapstra and Audi [16], which is based on a subset of the measured P_K's. While the Q_{EC} value obtained in this work has a much smaller error than the previous values, it should be pointed out that the current result presumes that the Surić calculation gives the correct shape of the IB spectrum in the fitted energy region. (The statistics in the vicinity of the end point are too poor to pin down Q_0 from a fit in a limited region near the end point.) Although there is no reason to suspect Surić's theoretical shape — indeed, the χ^2 for the fit is quite reasonable and the shape was found to agree very well with the data in the case of ^{125}I [9], which has a Z not too different from ^{139}Ce and an IBEC spectrum dominated by p capture, like ^{139}Ce—the current analysis does not preclude, on its own, that a small shape factor is being compensated by a slightly errant Q_0 value.

The measured normalization parameter of $A=1.070 \pm 0.024$ (where, for simplicity, we have combined the statistical and systematic errors in quadrature) suggests that Surić's calculation is able to reproduce (to within $\leqslant 10\%$) the absolute total intensity of IB in regions both far from the poles, as was verified by the ^{125}I measurement [10], as well as close to the pole, as demonstrated in this work. While the current work suggests that the Surić calculation does underestimate the intensity of the IB spectrum, it does so by only $7.0 \pm 2.4\%$, and not by the factor of 1.5–2 which is obtained for ^{193}Pt [3]. To confirm that the Surić calculation does not get substantially worse as Z increases we have conducted a recent experiment to measure the absolute intensity of the IB spectrum of ^{179}Ta [17]. Our preliminary value for the normalization parameter is 1.13 ± 0.02. While the underestimation seems to grow with Z, it is substantially less than the factor of 1.5–2 observed for ^{139}Pt. Thus, the large discrepancy observed for ^{139}Pt remains somewhat of a puzzle. Perhaps the treatment of the decay as an allowed decay, even though it is first-forbidden nonunique, is not valid. A confirmation of the measured intensity might be desirable.

The discrepancy of $7.0 \pm 2.4\%$ between the intensity predicted by the Surić calculation and the current measurement, while relatively small, is, nevertheless, statistically significant. The origin of this discrepancy is not obvious to us. Possible improvements to the Surić calculation which could be made within the context of the independent-particle approximation are (1) inclusion of exchange-overlap corrections and (2) taking into account the different Z dependences of the two time orders in the electron propagator, namely, the fact that radiation before capture occurs in the field of element Z, while radiation after capture takes place in the field of element $Z-1$ [18]. Both of these effects have been considered before by Persson and Koonin [18,19], but at much lower Z and higher Q_0, where the IB spectra are dominated by $1s$ and (to a lesser extent) $2s$ capture. At some stage true many-body effects in the atomic wave functions may have to be taken into account, as well, but these are obviously outside the scope of Surić's independent-particle approximation.

Two independent attempts were made at extracting the relative intensities of the $2p$ to $3p$ IB partial spectra; the results could not confirm the Surić calculation to a high degree of accuracy because of the small efficiency for detecting L x rays in one case, and insufficient statistics in the relevant part of the spectrum, in the other. The results in ^{125}I [9], however, suggest that the Surić calculation gives the correct relative intensities (to within a few percent) in this atomic number region.

ACKNOWLEDGMENTS

The assistance of Randy Bybee and Alan Helfer in collecting the data is gratefully acknowledged, as is the assistance of Dr. Paul Semmes in preparing the sources. The loan of electronic modules by Dr. Peter Parker of Yale University is deeply appreciated. The authors would also like to thank Dr. T. Surić and Dr. I. Žlimen for providing the code for calculating the IB matrix elements and for helpful discussions. This work was supported by the U.S. Department of Energy, Nuclear Physics Division, via Grant Nos. DE-FG05-87ER40314 and DE-FG02-96ER40955.

[1] W. Bambynek, H. Behrens, M. H. Chen, B. Crasemann, M. L. Fitzpatrick, K. W. D. Ledingham, H. Genz, M. Mutterer, and R. L. Intemann, Rev. Mod. Phys. **49**, 77 (1977).
[2] A. De Rújula, Nucl. Phys. **B188**, 414 (1981).
[3] K. Riisager, A. De Rújula, P. G. Hansen, B. Jonson, and H. L. Ravn, Phys. Scr. **31**, 321 (1985).
[4] R. J. Glauber and P. C. Martin, Phys. Rev. **104**, 158 (1956).
[5] P. C. Martin and R. J. Glauber, Phys. Rev. **109**, 1307 (1958).
[6] P. T. Springer, C. L. Bennett, and P. A. Baisden, Phys. Rev. A **35**, 679 (1987).
[7] L. Montanet *et al.*, Phys. Rev. D **50**, 1173 (1994).
[8] T. Surić, R. Horvat, and K. Pisk, Phys. Rev. C **47**, 47 (1993).
[9] M. M. Hindi, R. L. Kozub, and S. J. Robinson, Phys. Rev. C **49**, 3289 (1994).
[10] M. M. Hindi, R. L. Kozub, and S. J. Robinson, Phys. Rev. C **52**, 2780 (1995).
[11] M. M. Hindi and R. L. Kozub, Phys. Rev. C **43**, 461 (1991).
[12] M. O. Krause, J. Phys. Chem. Ref. Data **8**, 307 (1979).
[13] S. I. Salem, S. L. Panossian, and R. A. Krause, At. Data Nucl. Data Tables **14**, 91 (1974).
[14] F. B. Larkins, At. Data Nucl. Data Tables **20**, 313 (1977).
[15] T. W. Burrows, Nucl. Data Sheets **57**, 337 (1989).
[16] A. H. Wapstra and G. Audi, Nucl. Phys. **A432**, 1 (1985).
[17] M. M. Hindi *et al.*, to be submitted to Phys. Rev. C.
[18] B. I. Persson and S. E. Koonin, Phys. Rev. C **5**, 1443 (1972).
[19] S. E. Koonin and B. I. Persson, Phys. Rev. C **6**, 1713 (1972).