Evidence for the Exchange Effect Down to Very Low Energy in the Beta Decays of ^{63}Ni and ^{241}Pu

Martin Loidl, Cindy Le Bret, Matias Rodrigues, Xavier Mougeot

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Evidence for the Exchange Effect Down to Very Low Energy in the Beta Decays of ^{63}Ni and ^{241}Pu

M. Loidl · C. Le-Bret · M. Rodrigues · X. Mougeot

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Abstract Metallic magnetic calorimeters have been used for several years for the determination of the shapes of beta spectra. Experimental spectra of the beta decays of ^{63}Ni and ^{241}Pu exhibit, at very low energy, strong deviations from standard theoretical calculations including corrections for screening, radiative, and nuclear size effects. These discrepancies can be attributed to the atomic exchange effect. In particular the spectrum of ^{63}Ni measured with an electroplated ^{63}Ni source is in excellent agreement with theory including the exchange effect. This underlines the quality of the experimental spectrum as well as the correctness of the calculation of the exchange effect.

Keywords Metallic magnetic calorimeters · Beta spectrometry · Exchange effect

1 Introduction

The LNHB is developing metallic magnetic calorimeters (MMCs) for beta spectrometry. Several years ago, the spectrum of ^{241}Pu was measured [1]. A discrepancy between the experimental and a theoretical spectrum raised the question whether this discrepancy reflects a distortion of the experimental spectrum whose reason would need to be revealed, or is due to an insufficiency of the calculation of the theoretical spectrum: ^{241}Pu decays via a first forbidden, non-unique transition, and the spectra of forbidden, non-unique transitions are not straightforward to calculate. Consequently, the decision was taken to validate the potential of MMCs for beta spectrometry by measuring the spectrum of an allowed transition that can be calculated more reliably for comparison with the experimental spectrum. The pure beta emitter ^{63}Ni was chosen; its half life is 98.7 (24) years and the endpoint energy 66.980 (15) keV. This low energy beta emitter

M. Loidl (✉) · C. Le-Bret · M. Rodrigues · X. Mougeot
Laboratoire National Henri Becquerel, CEA, LIST, 91191 Gif-sur-Yvette, France
e-mail: martin.loidl@cea.fr

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was chosen also because in this energy range the potential energy loss due to escape of Bremsstrahlung photons from the detector can be safely neglected, as has been confirmed by Monte Carlo simulation. This makes the validation of the experimental method more reliable because no correction for this energy loss needs to be applied. The aim of this development is the determination of the shape factors particularly for beta emitters decaying via forbidden transitions, whose spectra are difficult to calculate and often experimentally not well known, and up to endpoint energies around 1 MeV.

The first measurements of the spectrum of ^{63}Ni were carried out using sources made by drying a small drop of $NiCl_2$ solution and enclosed in gold absorbers. The spectra resulting from several of these sources were all discrepant from one another, but more importantly they showed, in particular below ∼15 keV, a strong deviation from a standard theoretical calculation including corrections for screening, radiative, and nuclear size effects [2]. We found that this deviation is largely reduced when the atomic exchange effect is included in the calculation of the theoretical spectrum.

2 Exchange Effect

The exchange effect is one of several atomic effects that can influence the energy distribution and the decay rate of beta emissions: (a) the screening effect corresponds to the screening of the beta electron from the nuclear charge by the atomic electrons. (b) The change of total atomic binding energy is shared between the beta particle and the neutrino. (c) The difference between initial and final state atomic wave functions (nonorthogonality or "imperfect overlap") can give rise to shakeup and shakeoff, excitations of atomic electrons shifting the endpoint energy. (d) The exchange effect, finally, corresponds to the creation of the beta electron into a bound orbital of the daughter atom, accompanied by the simultaneous emission of a bound electron from the same orbital into the continuum. This exchange between beta electron and an atomic electron is possible because of the non-orthogonality of initial bound-electron states and final continuum state. Its probability, i. e. the magnitude of the exchange effect, depends firstly on the value of the bound electron wave functions in the nuclear region where the beta decay occurs, and secondly on the overlap integral between initial state bound-electron wave functions and final state continuum wave function.

After initial and partially contradictory treatments of the exchange effect [3,4], Harston and Pyper have conducted a more complete calculation for several nuclides [5], including ^{241}Pu. It predicts an energy-dependent enhancement of the beta emission probability that is very small in the higher energy part of the spectrum and increases towards low energies. Consequently, also the beta decay rate is enhanced and the mean energy of the beta emissions reduced.

Independently of ref. [5] Harston has also calculated the exchange effect for the decay of ^{63}Ni for energies greater than 5 keV and in 1 keV steps. The results are available in tabulated form [6]. Since the energy thresholds of the MMCs used in the present work are much lower, the calculation of the exchange effect was implemented in the code BetaShape developed at LNHB for the calculation of the shapes of beta spectra [7]. In the energy range below a few keV the magnitude of the exchange effect varies very strongly with energy, so the calculation was performed in steps of 100 eV.

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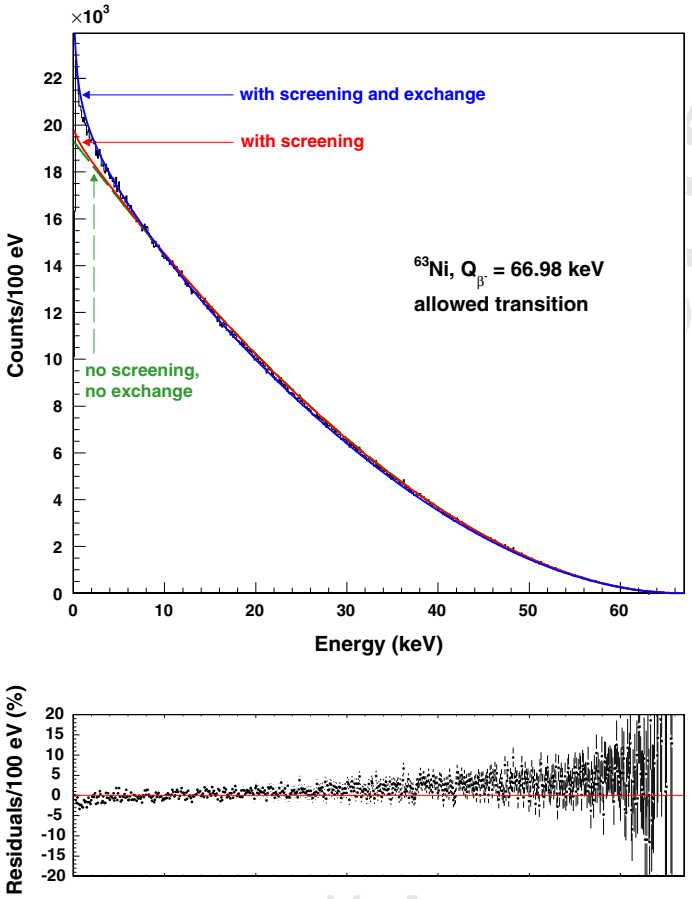

Fig. 1 Spectrum of an electroplated ^{63}Ni source enclosed in the gold absorber of an MMC ("rugged" *solid line*), compared with theoretical spectra without any correction for atomic effects, with screening correction and with screening and exchange effects. The *lower part* of the figure shows the residuals between experiment and the full theory including screening and exchange effects (Color figure online)

Experimental evidence for the exchange effect was reported only twice up to now, both times based on cryogenic detector measurements: qualitatively in [8] for the decay of ^{106}Pd (endpoint energy 37.5 keV), measured with a Sn microcalorimeter read out by an NTD Ge thermistor, and quantitatively in [9] for the decay of ^{63}Ni, measured in a thoroughly conducted and analyzed experiment with an InSb calorimeter read out by an STJ array. Both of these experiments had relatively high energy thresholds, ∼5 keV ([8]), respectively 8 keV ([9]).

3 The Spectrum of ^{63}Ni

As mentioned in the introduction, the spectra measured with sources consisting of a nickel salt are not reproducible from one source to another. The most likely explanation

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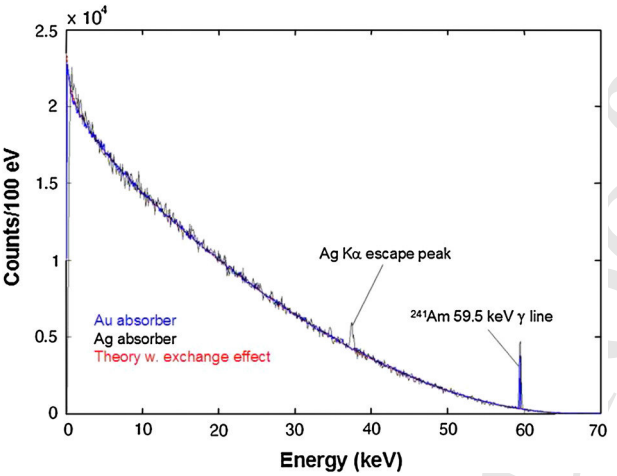

Fig. 2 Spectrum of an electroplated ^{63}Ni source enclosed in the silver absorber of an MMC, together with the spectrum obtained with the gold absorber and the theoretical spectrum including the exchange effect (Color figure online)

for this appears to be that part of the beta energy deposited in the $NiCl_2$ forming the source goes into the creation of metastable states like electron-hole pairs whose energy is not transformed to heat on a relevant time scale [10], so that not the entire beta energy is measured. Such a mechanism can in principle not exist in a metal. The next series of ^{63}Ni sources was made by electroplating on gold and silver foils, resulting in metallic nickel deposits. One source on a gold foil was sandwiched between two 16 μm thick gold foils forming the MMC absorber, and one source on a silver foil between two 16 μm thick silver foils. Each sandwich was diffusion welded thus enclosing the source and ensuring a good thermal contact within the absorber. Monte Carlo simulations were carried out to check that all beta particles are fully stopped in the absorbers and that the energy loss due to escape of Bremsstrahlung photons is negligible. The linearity of the detectors was checked using the gamma and X-rays of a combination of ^{55}Fe, ^{109}Cd and ^{241}Am sources. Within the energy range 5.9–88 keV covered by these sources, the maximum deviation of the measured positions of the lines from a linear fit is 0.1 %, and no trend of the residuals with energy is observed.

Figure 1 shows the spectrum measured with the gold absorber. It is in excellent overall agreement with the theoretical spectrum taking account of the exchange effect, whereas the theoretical spectrum ignoring the exchange effect is clearly incorrect. The calculation of the exchange effect in the code BetaShape is confirmed down to very low energy: the energy threshold in this measurement is 200 eV. There remains a small systematic deviation, revealed in the plot of the residuals. This is probably due to the calculation of the bound and continuum wave functions with analytical expressions using effective nuclear charges. A full Hartree-Fock calculation [11] of the electron wave functions should yield more exact results but is much less straightforward. The asymmetry of the residuals (−2 to 0 % in a small energy range; 0 to +5 % in a wider energy range) comes from the fact that a weighed mean squares fit was made and at

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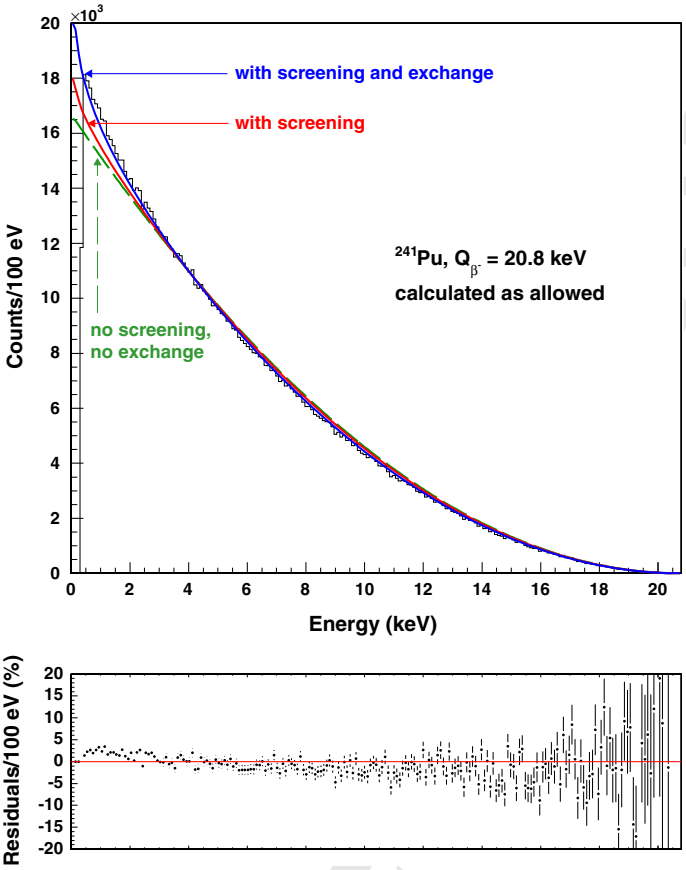

Fig. 3 Spectrum of ^{241}Pu measured with an MMC (*thin solid line*, *histogram*), compared with theoretical spectra calculated without any correction for atomic effects, with screening correction and with corrections for screening and exchange effect (Color figure online)

low energies statistics is higher. The residuals for the spectrum calculated only with the screening correction are at least as large at medium to high energies (with opposite sign) and up to + 20 % at low energies, confirming that the fit without exchange effect is much worse.

Figure 2 shows the spectrum measured with the silver absorber superimposed on the spectrum from the gold absorber. Although the noise level in the measurement with the silver absorber was higher and the statistics lower, the resulting spectrum confirms the spectrum shape obtained with the gold absorber.

4 The Spectrum of ^{241}Pu Revisited

After the strong evidence for the importance of the exchange effect in the case of ^{63}Ni the question arose to what extent the discrepancy between the experimental

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and theoretical spectra of ^{241}Pu published in [1] may be due to the omission of the exchange effect in the calculation of the theoretical spectrum. In its present version the code BetaShape can calculate only allowed and unique forbidden transitions, whereas ^{241}Pu decays via a first forbidden non-unique transition. It can be shown, however, that in the case of ^{241}Pu the approximation by an allowed transition should introduce only very small errors [7]. In Fig. 3 theoretical spectra of ^{241}Pu calculated without any correction for atomic effects, with screening correction and with corrections for screening and exchange effect are compared with the experimental spectrum presented in [1]. It is evident that the agreement between experimental and theoretical spectra is much better when the exchange effect is included. The remaining discrepancy is most likely due to the use of a dried source, added to the fact that the electron wave functions were calculated using effective nuclear charges.

5 Conclusions

The beta spectrum of ^{63}Ni was measured with electroplated sources enclosed in the absorbers of metallic magnetic calorimeters. The experimental spectra were compared with theoretical spectra calculated with and without the exchange effect. The good agreement between experiment and theory including the exchange effect confirms that the approximations made in the calculation of the exchange effect lead to errors of at most a few percent, even in the very low energy range between 200 eV and 5 keV, where such calculations have been performed for the first time. The theoretical spectrum ignoring the exchange effect is far off the experimental spectra. The enhancement of the spectrum by the exchange effect is as high as 23 % at 200 eV, and the mean energy of the beta emissions is lowered by 2.3 %. It was found that the previously measured spectrum of ^{241}Pu is also much better reproduced when the exchange effect is included in the theoretical spectrum.

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References

1. M. Loidl, M. Rodrigues, B. Censier, S. Kowalski, X. Mougeot, P. Cassette, T. Branger, D. Lacour, Appl. Radiat. Isot. **68**, 1454 (2010)
2. C. Le-Bret, M. Loidl, M. Rodrigues, X. Mougeot, J. Bouchard, J. Low Temp. Phys. **167**, 985 (2012)
3. J.N. Bahcall, Phys. Rev. **129**, 2683 (1963)
4. W.C. Haxton, Phys. Rev. Lett. **55**, 807 (1985)
5. M.R. Harston, N.C. Pyper, Phys. Rev. A **45**, 6282 (1992)
6. L. Angrave, PhD thesis, Magdalen College, Oxford, UK, 2000
7. X. Mougeot, M.-M. Bé, C. Bisch, M. Loidl, Phys. Rev. A **86**, 042506 (2012)
8. M.M. Lowry, D. Deptuck, I.C. Girit, J. Low Temp. Phys. **93**, 239 (1993)
9. L.C. Angrave, N.E. Booth, R.J. Gaitskell, G.L. Salmon, M.R. Harston, Phys. Rev. Lett. **80**, 1610 (1998)
10. D. McCammon, M. Juda, J. Zhang, R.L. Kelley, S.H. Moseley, A.E. Szymkowiak, IEEE Trans. Nucl. Sci. **33**, 236 (1985)
11. C.F. Fischer, *The Hartree–Fock Method for Atoms* (Wiley, New York, 1977)

Journal: 10909 Article No.: 1190 TYPESET DISK LE CP Disp.:2014/5/6 Pages: 6 Layout: Small-X