Direct measurement of the K electron capture to positron emission ratio in the decay of ^{22}Na

1969 J. Phys. A: Gen. Phys. 2 392

(http://iopscience.iop.org/0022-3689/2/3/018)

J. PHYS. A (GEN. PHYS.), 1969, SER. 2, VOL. 2. PRINTED IN GREAT BRITAIN

Direct measurement of the K electron capture to positron emission ratio in the decay of ^{22}Na

M. F. McCANN† and K. M. SMITH
Department of Natural Philosophy, University of Glasgow
MS. received 9th December 1968, *in revised form 10th February* 1969

Abstract. The 0·87 kev K electron capture peak of ^{22}Na has been observed using an internal source, scintillation counter technique. Direct measurement of the K/β^+ ratio was therefore possible: the result obtained was $K/\beta^+ = 0{\cdot}105 \pm 0{\cdot}009$. A review is given of recent precise results of the total capture to positron ratio (e/β^+) in ^{22}Na and the interpretation of these experiments is discussed. In conclusion, it is shown that the correction for electron exchange and overlap effects to the K/β^+ ratio has a smaller effect than has been supposed for low atomic numbers and that it is impossible to deduce an estimate for the Fierz interference term b from K/β^+ or e/β^+ measurements on light isotopes.

1. Introduction

Measurements of electron capture to positron emission branching ratios have been of importance to beta decay theory chiefly because the ratio is sensitive to Fierz interference between the scalar and vector, or tensor and axial vector interactions responsible for the decay. Either the total capture to positron ratio (e/β^+) or the K capture to positron ratio (K/β^+) in allowed transitions can be used to find the Fierz term b from the expression

$$b = \frac{R - R_0}{R_0 + R\langle W^{-1}\rangle}. \tag{1}$$

Here R is the experimental ratio, R_0 is the calculated ratio with $b = 0$, that is, in the absence of Fierz interference and $\langle W^{-1}\rangle$ is the mean value of the inverse of the positron energy. We note that accurate knowledge of both R and R_0 is required to determine b.

The pure Gamow–Teller decay of ^{22}Na to the 1·274 MeV excited state of ^{22}Ne has been prominent in the literature concerning the subject of Fierz interference. By about 1960 the total capture to positron emission ratio had been measured by several workers (Sherr and Miller 1954, Kreger 1954, Allen *et al.* 1955, Konijn *et al.* 1958, Ramaswamy 1959). The weighted mean of their results gave e/β^+ equal to $0{\cdot}1130 \pm 0{\cdot}0035$ (cf. Leutz and Wenninger 1967), while the calculated ratio was e/β^+ equal to $0{\cdot}1135 \pm 0{\cdot}002$ (Sherr and Miller 1954). This evidence for a near-zero value of b was supported by capture to positron measurements on other isotopes and by work on the shapes of beta spectra and the polarization of electrons in beta decay (Wu 1965). These results lent support to the well-known $V-A$ theory of beta decay.

In recent years, however, three new precise measurements of e/β^+ in ^{22}Na have been reported; these are summarized in table 1 below.

Table 1. Recent results for e/β^+ in ^{22}Na and theoretical predictions

Experiment	Theory	Theory with exchange correction
$0{\cdot}1041 \pm 0{\cdot}0010$ (W)	$0{\cdot}1135 \pm 0{\cdot}002$ (S)	
$0{\cdot}1048 \pm 0{\cdot}0007$ (L)	$0{\cdot}1138 \pm 0{\cdot}0025$ (L)	$0{\cdot}100 \pm 0{\cdot}006$ (L)
$0{\cdot}1042 \pm 0{\cdot}0010$ (V)	$0{\cdot}1118 \pm 0{\cdot}002$ (V)	0·0974 (V)

S, Sherr and Miller 1954; W, Williams 1964; L, Leutz and Wenninger 1967; V, Vatai *et al.* 1968.

† Now at the Department of Natural Philosophy, University of Edinburgh.

It is seen that these new results are in rather poor agreement with the usual theory. Two explanations have been put forward as a source of the discrepancy: either the existence of a finite Fierz term b (Williams 1964) or a correction to the theoretical e/β^+ value for the effects of exchange and overlap (Leutz and Wenninger 1967, as proposed by Bahcall 1963). Further discussion of these effects is postponed to § 4 below.

In view of the interesting developments outlined above, it seemed to us that an attempt should be made to detect the electron capture decay directly. This mode of attacking the problem had not been tried previously, because of the low energy (about 0·87 kev) of the neon x-rays and Auger electrons emitted after capture; thus all previous measurements had relied on inferring the electron capture fraction from other measurable aspects of the decay scheme. A direct measurement, however, seemed possible with the NaI(Tl) internal source technique (Joshi *et al.* 1963). The improved high-quantum-efficiency low-noise photomultipliers becoming available had already allowed detection of electron capture peaks, from internal sources, at energies as low as 3 kev (McCann *et al.* 1967) and 2 kev (Schulz 1967). The measurement of the K/β^+ ratio was therefore undertaken. Since a coincidence technique was used, effects due to ground-state transitions, photomultiplier noise and other sources of background were avoided.

2. Experimental procedure

A small amount of ^{22}Na in the form of aqueous NaCl solution supplied by the Radiochemical Centre, Amersham, was incorporated into a NaI(Tl) crystal grown from the melt as described previously (Joshi *et al.* 1963). A small 4 mm × 3 mm × 0·5 mm slice of the

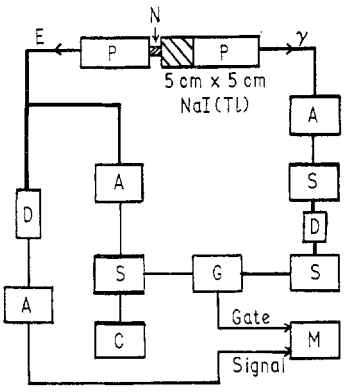

Figure 1. Block diagram of apparatus: A, amplifier; C, scaler; D, delay line; G, anticoincidence gate; M, multichannel analyser; N, source crystal with ^{22}Na; P, photomultiplier; S, single-channel analyser.

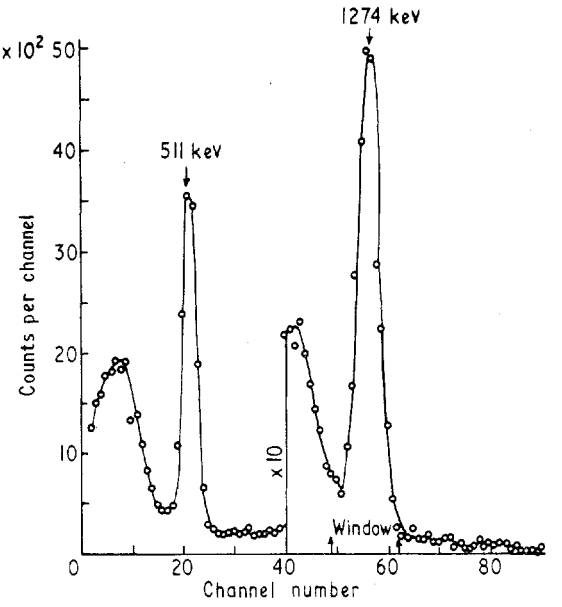

Figure 2. Gamma-ray spectrum in large NaI(Tl) scintillator.

grown crystal with an activity of approximately 300 disintegrations per second was immersed in liquid paraffin, covered with a 25 μm aluminized Mylar reflector and mounted on a low-noise D213 photomultiplier supplied by E.M.I. Ltd. As shown in the block diagram of figure 1, pulses from the multiplier amplified by a non-overloading NE5202 amplifier were analysed in a Laben 512-channel analyser when in coincidence with the 1·274 Mev ^{22}Ne

gamma rays. The latter were detected in a 4·4 cm diameter by 5 cm thick NaI(Tl) scintillator located close to the small radioactive crystal. The amplified pulse-height spectrum from the gamma-ray detector, shown in figure 2, was analysed by an I.D.L. 672A single-channel pulse-height analyser to produce coincidence gate pulses for the multi-channel analyser from input pulses in the energy 'window' indicated in figure 2.

Measurement of the random coincidence spectrum was effected by delaying the single-channel analyser output pulse by 14 μs using 1 kΩ delay cable. It was found that the 'random' coincidence spectrum measured in this way was higher than the real coincidence spectrum for very low energies (less than 400 ev), as shown in figure 3(*a*). This clearly unreal result was ascribed to the existence of afterpulses due to phosphorescence, which are associated with large energy deposition by positrons in the radioactive scintillator. Delayed emission from NaI(Tl) has also been observed by previous workers (Porter *et al.* 1966). Since the detection efficiency of the source crystal for positrons is effectively 100%, there will be a high-energy pulse (> 1 kev) in real coincidence with nearly every ^{22}Ne gamma ray detected. The afterpulses following these high-energy events will not, however, be detected since they lie in the deadtime following a real coincidence event; on the other hand, as soon as the coincident time relationship between the high-energy positron pulse and the gating gamma ray is destroyed, that is, in the random measurement situation, these pulses can contribute to the measured spectrum.

In order to demonstrate that the observed excess in the random coincidence spectrum was due to afterpulses following high-energy events in the radioactive crystal, an artificial deadtime was introduced after each large pulse. This was effected by using the high-energy positron pulse to operate an anticoincidence gate, thus blocking any gamma-ray gating pulses for a period of 450 μs after the high-energy event. The real and random coincidence spectra obtained with this deadtime in operation are shown in figure 3(*b*), which illustrates the relative reduction in the measured random rate, consistent with the existence of afterpulses. The real spectrum is almost unaffected by the inclusion of a deadtime apart from a slight overall reduction in intensity, which may be accounted for by the deadtime correction. By measuring the random spectrum in this way, therefore, the intensity of the random contribution could be obtained to a satisfactory degree of accuracy for the very low-energy region of the K peak where it is most important.

By virtue of the effectively 100% detection efficiency of the radioactive NaI(Tl) scintillator for positron events, the positron intensity was measured by the number of pulses in the peak shown in figure 3(*a*), which corresponds to limiting signals in the amplifier. The validity of this procedure was established by the consistent values obtained for the positron intensity in subsidiary measurements of the positron spectrum made with lower amplifier gain settings (see figure 4). Any correction to the measured positron intensity for escape from the surface of the source crystal is negligible, since a positron need only deposit about 2 kev in the crystal to be counted.

3. Results

As shown in figure 3(*a*) (no deadtime) and figure 3(*b*) (450 μs deadtime), the K peak can be fitted fairly well in the high-energy region by a Poisson distribution with a mean value $m = 3$, consistent with previous experience in this laboratory. The Poisson curve is not inconsistent with the observed shape at lower energies after subtraction of a random coincidence spectrum obtained as detailed above. The estimated uncertainty of 6% in determining the shape of the K peak below about 400 ev is the largest source of uncertainty in the measurement of the K capture rate.

Compton scattering of the 511 kev positron annihilation gamma rays in the source crystal could possibly have given rise to low-energy Compton electrons in the K peak region. These, however, would be displaced to higher energies by summing in the crystal with the parent positron events, so that any background in the K peak region due to Compton scattering was negligible.

The positron decay rate was obtained from the intensity of the saturation peak in figure 3(*a*). Since in the majority of cases each positron is accompanied by two 511 kev

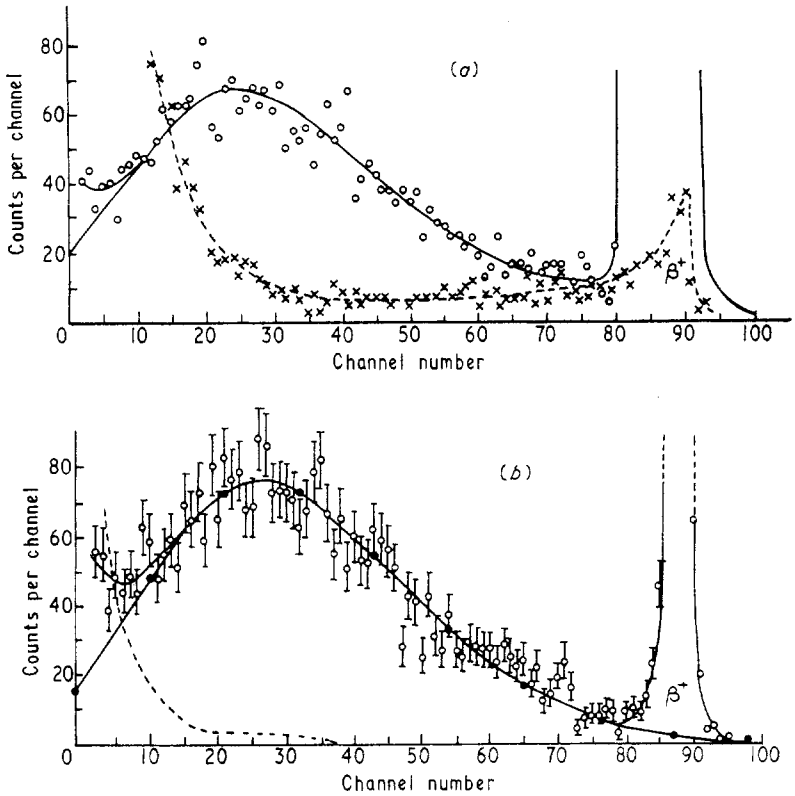

Figure 3. (*a*) K capture peak from ^{22}Na detected in small crystal with measured 'random' spectrum: full curve, coincidence spectrum; broken curve, 'random' spectrum. (*b*) K capture peak with 450 μs deadtime after every coincident positron pulse above about 5 kev: full curve, coincidence spectrum with Poisson fit; broken curve, 'random' spectrum.

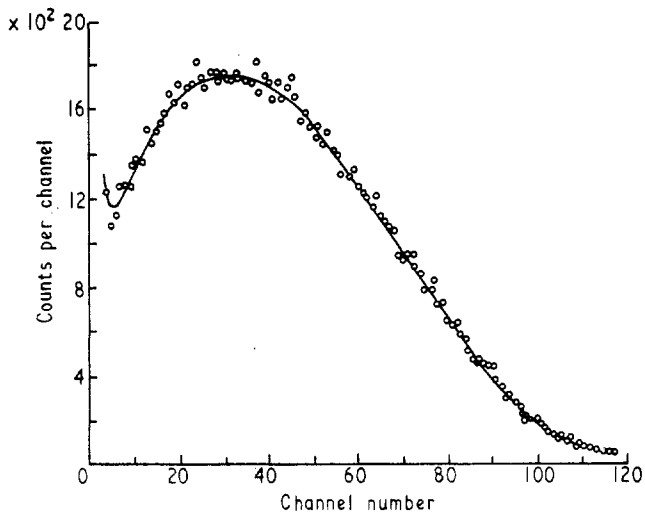

Figure 4. Positron spectrum from ^{22}Na in radioactive crystal.

annihilation gamma rays which are coincident with the 1·274 MeV nuclear gamma ray, there was a probability that at least one of the 511 keV quanta would sum with the 1·274 MeV gamma ray in the 5 cm crystal. Both gamma rays in this case would give their usual Compton plus photopeak distributions with the result that the pulses in the 1·274 MeV gating 'window' could in practice be augmented or depleted by sum events. Since the sum process is correlated only with a positron event, and not with a K capture event, there could have been a loss or gain of positron counts relative to K counts. This possibility was examined using tabulated cross sections for the two gamma rays and constructing a theoretical sum spectrum. This led to a small correction for loss of positrons of $(1 \pm 1)\%$ to the measured positron decay rate.

For the runs carried out with and without anticoincidence deadtime the mean value obtained for the K capture to positron ratio was $0{\cdot}105 \pm 0{\cdot}009$.

The error limits stated are made up as follows: extrapolation of low-energy end of K peak, $\pm 6\%$; statistical error on both K and positron counts, $\pm 2\%$; summing correction to positron count, $\square 1\%$.

4. Discussion

The calculated K/β^+ ratio for ^{22}Na is $0{\cdot}1058 \pm 0{\cdot}0015$ (Leutz and Wenninger 1967). It can be seen that our experimental result of $0{\cdot}105 \pm 0{\cdot}009$ is in agreement with this value.

It is interesting to examine the effects of possible Fierz interference and exchange corrections on the K/β^+ ratio. Reference to the precise total capture to positron ratios in table 1 will allow us to draw some conclusions here. The agreement among these experimental results themselves is excellent; we conclude, therefore, that a definite disagreement with theory exists, of magnitude about $(8 \pm 2)\%$. That the discrepancy is *not* due to exchange corrections (as postulated by Leutz and Wenninger) is pointed out by Murthy and Ramaswamy (1964) and by Williams (1968): it is shown that the total capture to positron ratio is virtually unaffected by exchange. Thus to explain the disagreement one must fall back on the possibility of Fierz interference and/or some other unconsidered effect, such as an error in the electron wave functions used to calculate the e/β^+ ratio (cf. Winter 1968). Whatever the source of the disagreement, we might expect it to have the same influence on the K/β^+ ratio as on the e/β^+ ratio. This would reduce the theoretical K/β^+ ratio to $0{\cdot}0973 \pm 0{\cdot}003$, which is still in agreement with our experimental figure.

It is important to notice that the K/β^+ ratio, in contrast to the e/β^+ ratio, is expected to be affected by exchange. Although no explicit calculation of the correction factor has been made below atomic number 14, an extrapolated value of $0{\cdot}85 \pm 0{\cdot}05$ for ^{22}Na (atomic number 11) was found by Leutz and Wenninger. Application of this correction would reduce the theoretical K/β^+ ratio to $0{\cdot}815 \pm 0{\cdot}007$, which is in poor agreement with our experimental result. It has been pointed out previously that values of exchange correction factors found by extrapolation of Bahcall's formula are invalid (Bahcall, quoted by Campbell *et al.* 1967; Vatai *et al.* 1968). Our result gives direct evidence of this. Although the magnitude of the exchange correction is not known theoretically, inspection of other K/β^+ results at low atomic numbers indicates that the correction must be close to unity. In the case of ^{19}Ne, for example, at atomic number 10, a recent experiment gives the result $K/\beta^+ = (9{\cdot}6 \pm 0{\cdot}3) \times 10^{-4}$, in excellent agreement with the usual theoretical value of $9{\cdot}5 \times 10^{-4}$ (Leiper, to be published).

In view of the discussion above, it is worth mentioning that measurements of K/β^+ or e/β^+ in light isotopes are quite useless from the point of view of determining unambiguously the existence or otherwise of Fierz interference; other methods must be relied on for this. Even without the complication of a Fierz term, i.e. even if b were proved to be zero, the situation in electron capture work at low atomic numbers is still quite confused. Precise measurements of e/β^+, K/β^+ and L/K ratios in light isotopes would aid in solving the remaining problems of the accuracy of wave functions and the magnitude of exchange corrections.

Acknowledgments

The authors would like to express their appreciation to Dr. G. M. Lewis for invaluable discussions, to Professor W. E. Burcham for kind comments and advice and to Professor

P. I. Dee for his interest in this work. Discussions with Dr. E. Vatai are also highly appreciated. One of us (M.F.McC.) wishes to acknowledge the support of a Science Research Council grant.

References

ALLEN, R. A., *et al.*, 1955, *Proc. Phys. Soc.* A, **68,** 681–5.
BAHCALL, J. N., 1963, *Phys. Rev.*, **129,** 2683–94; **131,** 1756–9; **132,** 362–7.
CAMPBELL, J. L., LEIPER, W., LEDINGHAM, K. W. D., and DREVER, R. W. P., 1967, *Nucl. Phys.* A, **96,** 279–87.
JOSHI, B. R., LEWIS, G. M., and SMITH, K. M., 1963, *Nucl. Instrum. Meth.*, **24,** 77–80.
KONIJN, J., VAN NOOIJEN, B., HAGEDOORN, H. L., and WAPSTRA, A. H., 1958, *Nucl. Phys.*, **9,** 296–305.
KREGER, W. E., 1954, *Phys. Rev.*, **96,** 1554–5.
LEUTZ, H., and WENNINGER, H., 1967, *Nucl. Phys.* A, **99,** 55–64.
MCCANN, M. F., LEWIS, G. M., and SMITH, K. M., 1967, *Nucl. Phys.* A, **98,** 577–84.
MURTHY, A. S. V., and RAMASWAMY, M. K., 1964, *Indian J. Pure Appl. Phys.*, **32,** 101.
PORTER, F. T., FREEDMAN, M. S., WAGNER, F., and SHERMAN, I. S., 1966, *Nucl. Instrum. Meth.*, **39,** 35–44.
RAMASWAMY, M. K., 1959, *Indian J. Phys.*, **33,** 285–94.
SCHULZ, G., 1967, *Nucl. Phys.* A, **101,** 177–86.
SHERR, R., and MILLER, R. H., 1954, *Phys. Rev.*, **93,** 1076–81.
VATAI, E., VARGA, D., and UCHRIN, J., 1968, *Nucl. Phys.* A, **116,** 637–42.
WILLIAMS, A., 1964, *Nucl. Phys.*, **52,** 324–32.
—— 1968, *Nucl. Phys.* A, **117,** 238.
WINTER, G., 1968, *Nucl. Phys.* A, **113,** 617–24.
WU, C. S., 1965, *Alpha-, Beta- and Gamma-ray Spectroscopy*, Ed. K. Siegbahn (Amsterdam: North Holland), pp. 1365–96, 1415–22.