Low-Lying 1− States in Even-Even Nuclei*

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In a limited isotopic region, there have been found low-lying states in even-even nuclei of spin 1, odd parity. Other energy levels in the vicinity are the well characterized 0+, 2+, and 4+ states of the even-even nuclear type. The 1− states were identified through α-γ angular correlation measurements supported by internal conversion coefficient data.

I. INTRODUCTION

IT has become well established that for even-even nuclei in the region well above the closed shell of 82 protons and 126 neutrons there exist low-lying energy levels with spin and parity assignments 0+, 2+, 4+,$\cdots$.[1] Such a sequence of levels is described as a *rotational band* because the spins, parities, and energy spacing conform with the expectations of rotational states according to the Bohr-Mottelson[2] unified collective and individual particle nuclear model. In all cases examined by study of the alpha-decay process the fundamental state is the 0+ ground state, and the energies of the 2+ and 4+ states follow a regular trend in their level spacings: narrower toward heavier nuclei and wider toward lighter nuclei.[2–4] For Th^{228} decay, as an example, the 2+ state is at 84 kev and the 4+ state at 253 kev.[5] Consistent with the parities and spins of these even spin states is the finding that they are joined by cascading $E2$ transitions.

In a rather confined region (around $Z=90$, $N=136$) a low-lying excited state appears which is not a member of this rotational sequence. The level is apparently 1− and was first recognized in Th^{228} decay (excited states of Ra^{224}).[5] Here there was found a state between the 2+ and 4+ states which decayed to the 2+ and 0+ (ground state) by $E1$ transitions. The $E1$ character of the radiation allows only a 1− assignment and furthermore the odd spin-odd parity combination is a conservation requirement in the alpha decay of an even-even nucleus.

There are now five cases in which the 1− state has been recognized, lying either between the 4+ and 2+ states or just above the 4+ state. The present paper is concerned with α-γ angular correlation measurements on three of these, the results of which are consistent with the 1− assignment, and incidentally, show the difference in correlation with the α-γ sequence involving the 2+ states. The three alpha emitters examined were Th^{228}, Th^{226}, and U^{230} which give information on the excited states of Ra^{224}, Ra^{222}, and Th^{226}, respectively.

II. EXPERIMENTAL

The gamma side of the angular correlation apparatus consisted of a commercial Harshaw NaI (Tl activated) crystal ($1\frac{1}{2}$ in. diameter by 1 in. thick) mounted on a Dumont 6292 photomultiplier tube and placed from two to four inches away from the sample. The output from the photomultiplier tube was amplified and shaped and then fed into a gated 50-channel pulse height analyzer.

Two different types of alpha detectors were used depending upon whether or not it was necessary to distinguish alpha groups. In the case of Th^{228}, where the daughter Ra^{224} grows in relatively slowly, it was possible to use a ZnS screen fastened to the end of an RCA 5819 photomultiplier tube, the output of which was amplified, shaped, and then used as a gate for the gamma-ray detector as it fed into the 50-channel pulse height analyzer. For Th^{226} and U^{230}, however, where the daughters grow in rapidly, it was necessary to have sufficient resolution of alpha energies to be able to separate the five alpha emitters in the family and to measure gamma-ray coincidences with each alpha emitter. This was accomplished by using a thin NaI crystal as a rough spectrometer in the following manner: The newly cleaved crystal ($\frac{1}{4}$ in. square$\times\frac{1}{32}$ in. thick) was fastened (in a drybox) onto a Dumont 6292 photomultiplier tube. The assembly was then mounted in a vacuum chamber, and samples introduced by means of an air lock. Silica gel was present in the vacuum chamber to absorb moisture and a liquid nitrogen trap was placed between the pump and the chamber, but even so the resolution decreased from an optimum of 3 percent (full width at half maximum) to about 5 percent over a period of two weeks. The output from the photomultiplier tube was amplified, shaped and fed into a single channel pulse height analyzer. Pulses of a particular height were selected and used to gate the 50-channel pulse height analyzer. The resolving time of the coincidence unit described was of the order of ten microseconds.

An example of the alpha spectrum so obtained for the U^{230} series is shown in Fig. 1. Each of these peaks is comprised almost entirely of the ground state transi-

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[1] F. Asaro and I. Perlman, Phys. Rev. **91**, 763 (1953).
[2] A. Bohr and B. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953); A. Bohr, *Rotational State of Atomic Nuclei* (Ejnar Munksgaards, Copenhagen, 1954).
[3] F. Asaro and I. Perlman, Phys. Rev. **87**, 393 (1952).
[4] K. W. Ford, Phys. Rev. **90**, 29 (1953).
[5] Asaro, Stephens, and Perlman, Phys. Rev. **92**, 1495 (1953).

tion which in some cases is considerably removed from the low abundance-low energy group which is coincident with the gamma ray of interest. Nevertheless, with a knowledge of the gamma ray under consideration, the alpha energy gate could be set at the proper place to minimize conflicting coincidences.

The Th^{228} and the U^{230} family sources for the alpha-gamma coincidence measurements were prepared somewhat differently. The Th^{228} sample, separated from its decay products by a resin column technique,[5] was simply evaporated from aqueous solution as a $\frac{1}{4}$-in. diameter spot on a 6-mil thick aluminum plate. The low atomic number was employed for the backing since the gamma rays had to penetrate it to reach the detector.

The U^{230} source was one previously prepared for measuring its alpha spectrum.[6] The material had been vacuum sublimed onto a 2-mil platinum plate forming a band $\frac{1}{8}$ in.$\times$1 in. Because of the appreciable attenuation of the gamma rays in traversing the platinum plate, differences in attenuation due to differences in thickness were minimized by rotating the plate with the gamma-ray detector. Both of these sources were considerably larger than desirable for precise angular correlation measurements.

III. RESULTS

U^{230} family gamma-ray spectrum.—Previous work,[6] using both chemical methods and recoil techniques to separate members of the decay family, identified the following gamma rays in the series:

U^{230}	Th^{226}	Ra^{222}	Ra^{222} (or daughters)
70 kev	109	330	609
160	~130		
230	~190		
	240		

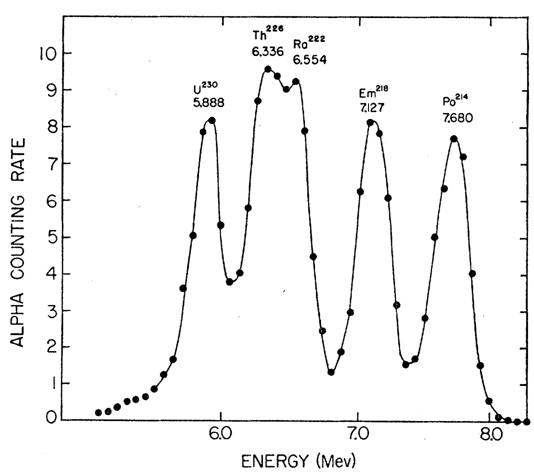

FIG. 1. Alpha-particle spectrum of U^{230} series taken with NaI crystal and pulse-height analyzer. (Ordinate scale is arbitrary.)

[6] Asaro, Slater, and Perlman (unpublished).

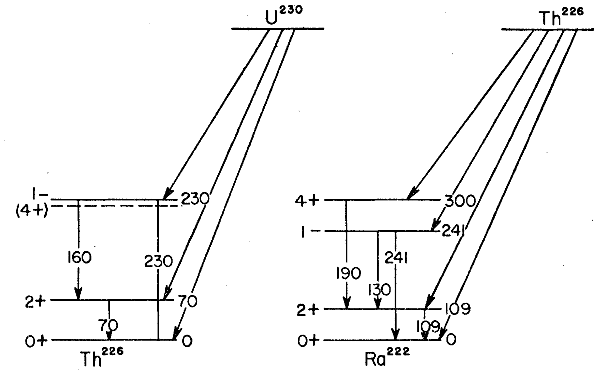

FIG. 2. Decay schemes of U^{230} and Th^{226}.

These assignments were confirmed with the apparatus described above by observing the gamma-ray spectrum as a function of the position of the gate set according to alpha-particle energy. For example, the 230-kev gamma ray of U^{230} decay and the 240-kev gamma ray of Th^{226} would not be resolved by the gamma-ray spectrometer operating as a single event instrument, but when the alpha-particle gate was set at about 5.66 Mev (U^{230} maximum energy minus 230 kev) the 230-kev gamma ray showed a maximum coincidence rate and when the gate was set at about 6.10 Mev (Th^{226} maximum energy minus 240 kev) the 240-kev gamma ray showed up. Since the 240-kev peak is four times as intense as the 230-kev peak, there was still some overlap when the gate was set at 5.66 Mev. It was estimated that roughly 15 percent of the 230-kev peak was due to 240-kev gamma rays. In addition to the above confirmation of assignments, the 609-kev gamma ray was proved to belong to Em^{218}, which assignment was suspected but not determinable from the earlier measurements.

The proposed decay schemes for U^{230} and Th^{226} are shown in Fig. 2. These are based on alpha and gamma spectra and gamma-gamma coincidence measurements made previously[6] and on the alpha-gamma angular correlations of the present study. It will be noted that the 230-kev state of U^{230} is shown as an unresolved doublet comprising the 1− and the 4+ states while in Th^{226} the 1− state is well separated from the others.

U^{230} series alpha-gamma angular correlations.—The angular correlations obtained by gating with the alpha particles of a particular energy and rotating the gamma ray detector, are shown in Figs. 3 and 4. In Fig. 3 the gate was set corresponding to 6.10 Mev which is the energy of Th^{226} α_{240},[7] and the gamma-ray peak at 240 kev shows a distribution with a minimum at 180° and maxima at 90° and 270°. The theoretical correlation function for the sequence $0+ \xrightarrow{\alpha} 1- \xrightarrow{\gamma} 0+$ is plotted as Fig. 5. It is seen that maxima and minima occur in the same positions, but the angular

[7] This nomenclature refers to the alpha group of Th^{226} which populates the 240-kev level of the daughter nucleus.

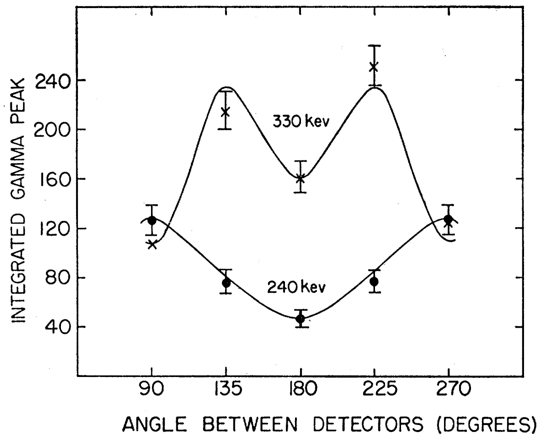

FIG. 3. α-γ angular correlations for 240-kev gamma ray of Th^{226} decay and 330-kev gamma ray of Ra^{222} decay.

anisotropy is not so pronounced for the experimental data. Two of the most important reasons for this are the large sample size and relatively high geometry, for which no corrections were made. Nevertheless this correlation may be taken as rather good support for the 1− assignment of the 240-kev state of Th^{226}, particularly since there are limitations in other possible assignments. From the alpha-particle spectrum of Th^{226} and the present coincidence measurements, it is fairly certain that the 240-kev gamma ray leads to the ground state of Ra^{222} and is therefore electric radiation. (The conservation of spin and parity demands that a gamma ray must be electric if it leads to the ground state of an even-even nucleus and arises from a level populated by alpha decay.) Assignment of $E3$ and higher multipolarities can be ruled out by consideration of lifetimes since in this case prompt coincidences were observed (few microseconds maximum delay) whereas the calculated lifetime for an $E3$ transition would be 10^6 fold longer. Therefore, we need consider only $E1$ or $E2$ assignments and it will be shown presently that the angular correlation expected if the radiation were $E2$ is quite different from that observed.

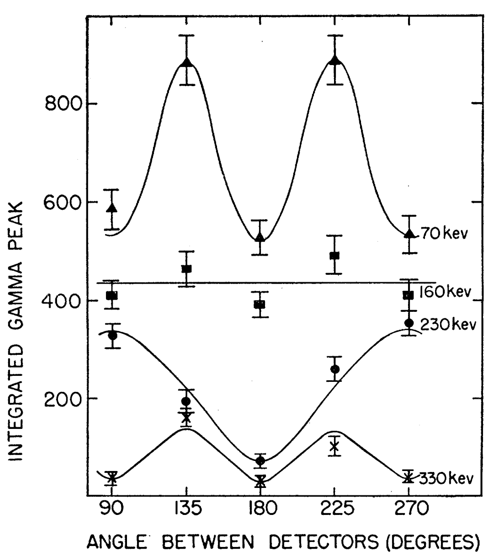

FIG. 4. α-γ angular correlations for the following gamma-ray transitions: 70, 160, and 230 kev from U^{230} decay; 330 kev from Ra^{222} decay.

As seen in Fig. 3, a peak at 330 kev with different correlation has appeared. This comes from coincidence with α_{330} of Ra^{222} (6.23 Mev) which, because of the poor resolution of the alpha peaks, overlaps considerably the gating energy (6.10 Mev). Because of this lack of discrimination and the fact that the 330-kev gamma ray is more intense than the 240, the coincidence intensity of the 330-kev peak is likewise more intense. The angular correlation of the 330-kev gamma ray is consistent with that for a $0+ \xrightarrow{\alpha} 2+ \xrightarrow{\gamma} 0+$ sequence as would be expected for an alpha transition to the first excited state of an even-even nucleus.

The data in Fig. 4 were taken in the same manner except the alpha gate was set at 5.66 Mev which corresponds to α_{230} of U^{230}. A different gamma spectrum appeared and included was the 230-kev gamma ray showing the $0+ \xrightarrow{\alpha} 1- \xrightarrow{\gamma} 0+$ correlation. The 160-kev peak involves the same energy level and its virtual absence of angular correlation is consistent with our supposition that the 230-kev level is a doublet of a 4+ and a 1− state and therefore the sequence measured is a mixture of $0+ \rightarrow 1- \rightarrow 2+$ and $0+ \rightarrow 4+ \rightarrow 2+$. Other evidence that the 230-kev level is actually a pair of close lying levels will not be reviewed here in detail. Briefly, part of the argument lies in the well-established systematic spacing of the 4+ state which, for U^{230} decay, should be at about 230 kev. Also, the abundance ratio of the 160-kev gamma ray to the 230-kev gamma ray appears to differ by a factor of two between the alpha decay of U^{230} and the beta decay of Ac^{226}.[8] If true, this can mean only that there are two levels populated to different degrees by the two modes of decay.

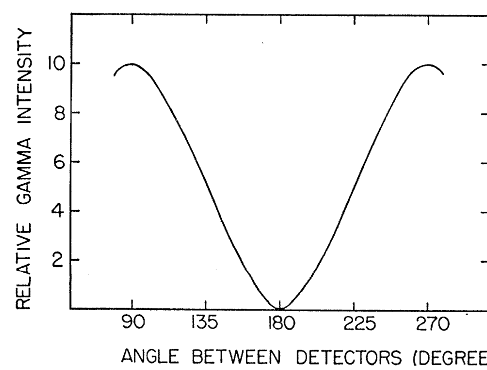

FIG. 5. Theoretical α-γ angular correlation for a $0+ \rightarrow 1- \rightarrow 0+$ sequence.

The abundant 70-kev transition of U^{230} decay shows

[8] J. Grover and G. T. Seaborg (unpublished).

up in Fig. 4 with the $0+ \rightarrow 2+ \rightarrow 0+$ correlation as expected, as does the 330-kev gamma ray of Ra^{222} decay appearing in low intensity.

Th^{228} α-γ angular correlations.—The decay scheme previously proposed[5] for Th^{228} is shown in Fig. 6. The 212-kev gamma ray was considered to be of principal interest and its angular correlation is shown in Fig. 7. The correlation is seen to be similar to that for the 230-kev transition of U^{230} and the 240-kev transition of Th^{226} indicating that all three involve the sequence $0+ \rightarrow^{\alpha} 1- \rightarrow^{\gamma} 0+$. The correlation for the 84-kev transition (not shown) behaved as expected for the type $0+ \rightarrow^{\alpha} 2+ \rightarrow^{\gamma} 0+$.

1− states in Th^{230} and U^{232} decay.—In addition to the three cases for which angular correlation studies were made in the present study, two others also apparently have the same type of decay scheme. In the decay of Th^{230} a 250-kev level has been recognized[9,10] which decays both to the ground state $(0+)$[11] and to the first even spin state $(2+)$.[12] The similarity of this decay

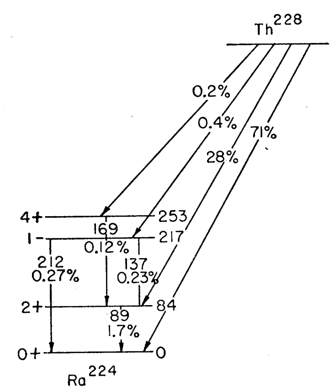

FIG. 6. Decay scheme for Th^{228}.

scheme and the proximity of this isotope to others having 1− states in the same energy range make it likely that this 250-kev level is a 1− state. In the decay of U^{232} a similar structure is seen from a level of 330 kev above the ground state.[13] Figure 8 shows a plot of the energy of these 1− states as a function of neutron number. A few other nuclei in this region are susceptible to this type of measurement and should be of interest in extending the picture based on these few cases.

IV. DISCUSSION

There is currently a considerable amount of speculation on the nature of the nuclear spectroscopic states in the heavy element region and this inevitably carries back to some nuclear model. It is not immediately obvious how the existence of low-lying 1− states fits as a predictable consequence into any of the pictures. A factor which must be considered in an explanation is that the appearance of these states at low enough energies to be discernible in the alpha-decay process apparently takes place only in a limited region. Although energy level density near the ground state becomes greater as one proceeds to heavier nuclei, the 1− state has apparently disappeared as indicated by careful studies of the alpha spectra of Pu^{238}[14] and Cm^{242}.[15] The failure to find 1− states for these heavier nuclei could stem from two reasons: Either the levels have become so high that the alpha population to these states is low beyond detection, or the levels are in a region in which the energy dependence is not the deterring factor but some selection process hinders the transitions. In this connection it should be mentioned that the population by alpha decay of the 1− states which have been observed are of the order of tenfold lower than would be expected from alpha-decay theory. Those transitions to the 4+ state in this region are similarly hindered.

If the 1− states are to be explained by some coupling of single-particle states, there is then implied close proximity of states of opposite parity. Other evidence that this is the case in the heavy element region may be taken from the identification of a number of low-lying

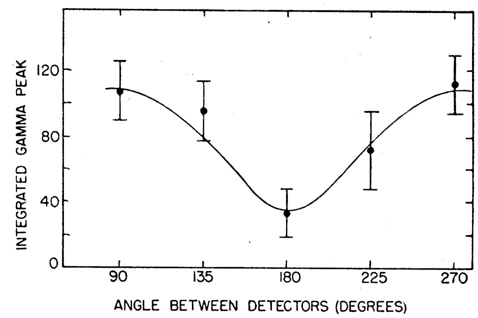

FIG. 7. α-γ angular correlation for 217-kev transition for Th^{228} decay.

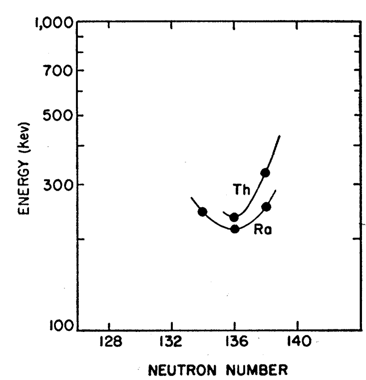

FIG. 8. Energy levels of 1− states in thorium and radium isotopes.

[9] Rosenblum, Valadares, Blandin-Vial, and Bernas, Compt. rend. **238**, 1496 (1954).
[10] G. Valladas and R. Bernas, Compt. rend. **236**, 2230 (1953).
[11] F. Rasetti and E. C. Booth, Phys. Rev. **91**, 315 (1953).
[12] Stephens, Asaro, and Perlman (unpublished).
[13] F. Asaro and I. Perlman (unpublished).
[14] F. Asaro and I. Perlman, Phys. Rev. **94**, 381 (1954).
[15] Asaro, Thompson, and Perlman, Phys. Rev. **92**, 694 (1953).

$E1$ transitions in odd-nucleon nuclei, as for example, in the decay of Am^{241} [16] and Am^{243} [17] where the states involved lie 60 and 75 kev respectively above the ground states.

Rasmussen[18] has discussed some of the implications of the Am^{241} decay scheme and his considerations may be applicable to the heavy element region in general. He pointed out that for protons in the 82–126 shell the only even parity orbital is $i_{13/2}$. The odd parity orbitals should have considerably lower j values and the only manner in which an $E1$ transition could arise is if j is not a good quantum number. His explanation is through the unified nuclear model involving strong coupling between the single particle and nuclear surface deformation which is a collective property. If strong coupling is assumed, the particle angular momentum vector j precesses rapidly around the symmetry axis of the spheroidal nucleus and the projection of j on the symmetry axis defines the spin. We then have a situation in which transitions between states of similar spin can involve large changes in the single particle wave function. Rasmussen suggests that such a situation may be responsible for the abnormally long lifetime for the 60-kev $E1$ transition. He also has found it necessary to employ this picture to explain features of β^- decay processes which would be anomalous on the basis of conventional selection rules.

The pertinence of this discussion to the problem at hand is that, at least in the heavy-element region, it may be necessary to look more deeply into the meaning of spectroscopic states than would appear just from spin assignments. Some of the implications of this idea have also been discussed in reference to the alpha-decay process.[18,19]

[16] Beling, Newton, and Rose, Phys. Rev. **87**, 670 (1952).
[17] F. Asaro and I. Perlman, Phys. Rev. **93**, 1423 (1954).
[18] J. O. Rasmussen, Jr., Arkiv Fysik **7**, 185 (1953).
[19] Perlman, Ghiorso, and Seaborg, Phys. Rev. **77**, 26 (1950).

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Capture-Positron Branching Ratios*

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Expressions are given for allowed and first-forbidden K and L capture probabilities, and rules are formulated for obtaining such expressions for capture of any order of forbiddenness from any orbit. The effect of screening on branching ratios is discussed, and a table for the rapid calculation of allowed branching ratios, including screening effects, is given. The results are then applied to the decays of Zn^{65}, Na^{22}, Sc^{44}, and V^{48}.

I. INTRODUCTION

In attempting to classify a negatron decay as to order of forbiddenness and to gain information as to the form of lepton-nucleon interaction, one generally uses as evidence the shape of the spectrum, the logft value of the decay, and the spin and parity changes involved, the last either measured or obtained from shell-model considerations.[1]

In the case of radioactive nuclei which emit positrons, however, there is another piece of evidence available, namely the branching ratio between orbital electron capture and positron emission. The branching ratio (b.r.) is defined by the relation

$$\text{b.r.}=\lambda_c/\lambda_+, \tag{1}$$

where λ_c and λ_+ are, respectively, the probabilities per unit time of electron capture and positron emission.

The quantity λ_+ is given by the well-known relation for an nth forbidden decay,

$$\lambda_+=\frac{1}{2\pi^3}\sum_{x,y} g_x g_y \tfrac{1}{2}(1+\delta_{xy}) \times\int_1^{W_0} pW(W_0-W)^2F_0(W,Z)C_{nxy}(W,Z)dW =\int_1^{W_0} N(W,Z)dW, \tag{2}$$

where the notation is the same as Greuling's[2] except that here we allow for the possibility of a linear combination of interactions—hence the presence of two indices, x and y. Greuling tabulates the C_{nxy} for pure interactions while Smith[3] and Pursey[4] give the interference terms which arise when more than one form of interaction is assumed to be present.

In calculating λ_+, numerical values for the Fermi

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† Operated for the U. S. Atomic Energy Commission by the General Electric Company.
[1] Mayer, Moskowski, and Nordheim, Revs. Modern Phys. **23**, 315 (1951); L. W. Nordheim, Revs. Modern Phys. **23**, 322 (1951).
[2] E. Greuling, Phys. Rev. **61**, 568 (1942).
[3] A. M. Smith, Phys. Rev. **82**, 955 (1951).
[4] D. L. Pursey, Phil. Mag. **42**, 1193 (1951). We adopt Pursey's notation rather than Smith's.