

## ***E2 and E3 Transitions from Quadrupole-Octupole Coupled States in $^{144}\text{Nd}$***

S. J. Robinson,<sup>1,2</sup> J. Jolie,<sup>2,3</sup> H. G. Börner,<sup>2</sup> P. Schillebeeckx,<sup>2,\*</sup> S. Ulbig,<sup>4,†</sup> and K. P. Lieb<sup>4</sup>

<sup>1</sup>Physics Dept. Tennessee Technological University, Cookeville, Tennessee 38505

<sup>2</sup>Institut Laue-Langevin, BP 156, 38042 Grenoble Cedex 9, France

<sup>3</sup>University of Fribourg, CH-1700, Fribourg, Switzerland

<sup>4</sup>II. Physikalisches Institut, Universität Göttingen, D-37073 Göttingen, Germany

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Lifetime measurements of the  $3_1^-$ ,  $5_1^-$ , and  $1_1^-$  states in  $^{144}\text{Nd}$  show that the absolute  $E2$  and  $E3$  transition rates from the  $5_1^-$  and  $1_1^-$  states are consistent with their structure being formed by the coupling of the lowest quadrupole ( $2_1^+$ ) and octupole ( $3_1^-$ ) excitations. A level at 2205 keV has been identified as having  $J^\pi = 4^-$  and may be another member of this quadrupole-octupole coupled quintuplet. The energy spacing of the quintuplet can be explained by anharmonicities in the quadrupole-octupole interaction and the influence of the  $\nu^2(2f_{7/2}, 1i_{13/2})$  configuration.

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In vibrational nuclei a quintuplet of negative parity states ( $1^-$  to  $5^-$ ) can be produced by the coupling of the single phonon quadrupole and octupole excitations. In the weak coupling limit these states should all lie around an energy given by the sum of the single-phonon energies [ $E(2_1^+) + E(3_1^-)$ ] and their depopulating transitions should show a simple characteristic pattern. In particular,  $E2$  transitions to the  $3_1^-$  level would involve the destruction of a single quadrupole phonon and should be of the same strength as the  $2_1^+ \rightarrow 0_1^+$  transition. Similarly,  $E3$  transitions from the quadrupole-octupole coupled (QOC) quintuplet states to the  $2_1^+$  level should be the same strength as the  $3_1^- \rightarrow 0_1^+$  transition. Up to now, the identification of such QOC states has relied primarily on level energy systematics and the observation of enhanced  $B(E1; 1_1^- \rightarrow 0_1^+)$  values [ $\approx 10^{-3}$  Weisskopf units (W.u.)], such as those identified by Metzger [1]. More recently, enhanced  $E1$  transitions have also been observed in  $^{143}\text{Nd}$  [2] and between excited states in  $^{144}\text{Sm}$  [3]. However, such evidence is less direct, since it requires the introduction of two-body terms in the  $E1$  transition operator [4].

The nucleus  $^{144}\text{Nd}$  has long been a testing ground for the description of collective vibrations in spherical nuclei. The collective nature of the lowest quadrupole and octupole states, which would be used to build the QOC quintuplet, can be inferred from the strength of their transitions to the ground state, of  $B(E2; 2_1^+ \rightarrow 0_1^+) \approx 25$  W.u. [5] and  $B(E3; 3_1^- \rightarrow 0_1^+) \approx 34$  W.u. [6], respectively. Further, it is already known that  $1^-$  (2186 keV) and  $5^-$  (2093 keV) states exist at an energy very close to  $E(2_1^+) + E(3_1^-)$  (697 keV + 1511 keV). These states have been interpreted as members of the QOC quintuplet [1, 7, 8], despite the lack of any measured  $E2$  or  $E3$  transition rates and the lack of identification of the supposed  $2^-$ ,  $3^-$ , and  $4^-$  members of the QOC quintuplet. However, recent studies [6, 8] of the  $5_1^-$  state in  $^{144}\text{Nd}$  have shown it to have an enhanced ground-state transition of  $B(E5; 5_1^- \rightarrow 0_1^+) \approx 11$  W.u. This has been interpreted as evidence for a dominant ( $\geq 50\%$ ) neutron

two-quasiparticle component in the wave function of this state [9].

To gain further insight into the structure of these states, detailed studies of levels and transitions in  $^{144}\text{Nd}$  have been carried out at the Institut Laue-Langevin (ILL) in Grenoble, France, using the thermal neutron capture reaction. Gamma rays were studied using the GAMS bent crystal spectrometers ( $E\gamma < 1500$  keV), a HPGe detector ( $E\gamma < 4$  MeV) and the pair formation spectrometer PN4 ( $E\gamma > 2$  MeV). Conversion electrons were studied using the BILL magnetic spectrometer. The details of these studies will appear in a future publication [10] and only the results relevant to the QOC structure are quoted in this Letter. One important result is the assignment of negative parity to a state at 2205 keV (already assigned  $J = 4$  by Behar Grabowski, and Raman [7]) based on conversion coefficient measurements. These measurements show that the 890.0 keV transition to the  $4_1^+$  level has  $E1$  character ( $\alpha_{\text{expt}} = 1.2 \pm 0.2$ ,  $\alpha_{E1} = 1.0$ ,  $\alpha_{M1} = 3.9$ ,  $\alpha_{E2} = 2.5$ ) and that the 693.0 keV transition to the  $3_1^-$  level has  $M1(+E2)$  character ( $\alpha_{\text{expt}} = 6.4 \pm 1.0$ ,  $\alpha_{E1} = 1.7$ ,  $\alpha_{M1} = 7.1$ ,  $\alpha_{E2} = 4.3$ ). Thus, this state would seem to be another member of the QOC quintuplet.

To obtain absolute transition strengths, the lifetimes (or limits thereon) of eleven excited states in  $^{144}\text{Nd}$ , including the  $3_1^-$ ,  $5_1^-$ , and  $1_1^-$  states, were measured using the gamma-ray induced Doppler broadening (GRID) technique [11]. The GRID technique employs the ultrahigh resolution of the GAMS4 double-flat crystal spectrometer [12] to observe the Doppler broadening of gamma rays which depopulate an excited state. This broadening is induced by the recoil imparted to the nucleus by the emission of gamma rays which feed this intermediate state, following thermal neutron capture. A knowledge of the recoil distribution and the slowing down process enables the lifetime of the intermediate state to be extracted as a fitting parameter of the Doppler broadened line profile [11].

For the determination of lifetimes in  $^{144}\text{Nd}$ , a 500 mg sample of separated  $^{143}\text{Nd}$  (91%), in the form of  $\text{Nd}_2\text{O}_3$ ,

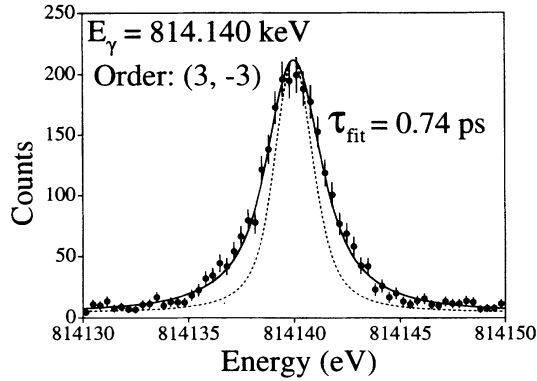


FIG. 1. Line profile of the 814.1 keV transition in  $^{144}\text{Nd}$  measured in third order of diffraction. The experimental data, shown as points with error bars, represent the sum of three individual scans. The solid line represents a best fit Doppler broadened profile to this data of  $\tau = 0.74$  ps. The dashed line represents the instrumental response function.

was placed in the H6/H7 through tube of the ILL reactor, in a neutron flux of  $5 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ . Line profiles of the intense 696.5 and 618.1 keV transitions were measured in a nondispersive geometry [11] to determine the response function of the GAMS4 spectrometer. The thermal Doppler broadening [11] was determined from measurements of the 696.5 keV ( $2_1^+ \rightarrow 0_1^+$ ) transition, which has a known lifetime of  $\tau = 4.5$  ps [5].

In the case of  $^{144}\text{Nd}$ , multibranch cascade feeding is dominant for most states and so the initial recoil distribution was simulated using a statistical model. This recoil simulation technique has already been successfully applied to a number of heavy nuclei, though absolute upper and lower limits can also be set using extreme feeding

scenarios [11]. In this work both techniques have been used. For the statistical simulations the constant temperature Fermi-gas formalism [13, 14] was used to calculate the cascade feeding, using the computer code GALENA [15]. Lifetimes were extracted using the computer code GRIDDLE [16] and employing the mean free path approximation [11] to model the slowing down process.

Figure 1 shows the Doppler broadened profile of the 814.1 keV transition, measured in third order of diffraction, and Table I gives the lifetimes extracted for the  $3_1^-$ ,  $5_1^-$ , and  $1_1^-$  states. [The lifetime extracted for the previously measured 2074 keV level ( $2^+$ ) is also included for comparison.] The results in Table I show that the calculated statistical feeding produces lifetimes which are consistent with previously published results for both the 2074 and 2186 keV levels. Therefore, the  $\tau_{\text{stat}}$  values were adopted in the calculation of absolute transition rates, except for the 2186 keV level, for which an average value of  $\tau = 18 \pm 4$  fs was adopted.

Table II gives the absolute rates (in W.u.) for transitions depopulating the  $2_1^+$ ,  $3_1^-$ ,  $5_1^-$ , and  $1_1^-$  levels. It is evident, despite the large errors in some cases, that the strength of both the  $1_1^- \rightarrow 3_1^-$  and the  $5_1^- \rightarrow 3_1^-$   $E2$  transitions is about the same size as the  $2_1^+ \rightarrow 0_1^+$  transition and, further, the strength of the  $5_1^- \rightarrow 2_1^+$   $E3$  transition is about the same size as the  $3_1^- \rightarrow 0_1^+$  transition. These transitions then follow the simple pattern outlined above for QOC states and this new evidence strongly supports the proposed QOC structure of these states. In addition, the absolute strengths of  $E1$  transitions from the  $3_1^-$ ,  $5_1^-$ , and  $1_1^-$  states are of the same strength as those  $E1$  transitions used to identify QOC states in  $^{143}\text{Nd}$  [2] and only slightly weaker than those observed in the  $N = 82$  nucleus  $^{144}\text{Sm}$  [3, 4].

TABLE I. Lifetimes of negative parity states in  $^{144}\text{Nd}$ . The result for the previously measured 2073.8 keV ( $2^+$ ) level is also given. The figures in parentheses give the uncertainties in the least significant digits for each result.

$E_x$ (keV)	$J^\pi$	$E_\gamma$ (keV)	$\tau_{\text{extreme}}$ (ps) <sup>a</sup>	$\tau_{\text{stat}}$ (ps) <sup>b</sup>	$\tau_{\text{lit.}}$ (ps)
1510.7	$3^-$	196.1 and 814.1	$0.56 < \tau < 1.21$	$0.81 (+11, -9)$	
2073.8	$2^+$	1376.3	$0.01 < \tau < 0.24$	$0.10 (+3, -2)$	$0.06 (\pm 3)^c$
2093.1	$5^-$	301.8 and 778.6	$0.5 < \tau < 3.4$	$1.2 (+11, -4)$	
2185.7	$1^-$	2185.8	$0.00003 < \tau < 0.24$	$0.03 (+4, -2)$	$> 0.016^d$ $0.014 < \tau < 0.034^e$ $0.015 (\pm 2)^c$ $0.031 (\pm 5)^f$

<sup>a</sup>Upper and lower limits determined using the extreme feeding scenarios described in Ref. [11].

<sup>b</sup>Values extracted using simulated cascade feeding, as described in the text.

<sup>c</sup>Ref. [17].

<sup>d</sup>Ref. [18].

<sup>e</sup>Ref. [19].

<sup>f</sup>Ref. [20].

TABLE II. Absolute transition rates (in W.u.) for negative parity states in  $^{144}\text{Nd}$ . The  $B(E2; 2_1^- \rightarrow 0_1^+)$  value is also given for comparison. The figures in parentheses give the uncertainties in the least significant digits.

Initial state	Final state	Transition energy (keV)	Multipolarity	$B(EL)$ (W.u.)
$2_1^+$	$0_1^+$	696.5	$E2$	$24.4(3)^a$
$3_1^-$	$0_1^+$	1510.7	$E3$	$33.9(17)^b$
	$2_1^+$	814.1	$E1$	$1.0(1) \times 10^{-3}$
	$4_1^+$	196.1	$E1$	$1.3(1) \times 10^{-3}$
$5_1^-$	$0_1^+$	2093.1	$E5$	$11(1)^b$
	$2_1^+$	1396.6	$E3$	$34(+16, -22)^c$
	$4_1^+$	778.6	$E1$	$5(2) \times 10^{-4}$
	$3_1^-$	582.5	$E2$	$20(+12, -10)$
	$6_1^+$	301.8	$E1$	$8(4) \times 10^{-4}$
$1_1^-$	$0_1^+$	2185.7	$E1$	$1.1(2) \times 10^{-3}$
	$2_1^+$	1489.2	$E1$	$2.4(5) \times 10^{-3}$
	$3_1^-$	675.1	$E2$	$20(5)^d$
	$2_2^+$	624.6	$E1$	$1.0(3) \times 10^{-4d}$
$(3\bar{Q}0c)$	$0_1^+$	2779	$E3$	$7.3(7)^b$

<sup>a</sup>Ref. [5].

<sup>b</sup>Ref. [6].

<sup>c</sup>Calculated using the branching ratio from Ref. [21].

<sup>d</sup>Calculated using the branching ratio from Ref. [7].

However, there are problems with a pure QOC interpretation. First, with negative parity now confirmed for the 2205 keV level, the directional correlation measurements of Behar, Grabowski, and Raman [7] indicate a dominant  $M1$  component ( $>97\%$ ) in the  $4_1^- \rightarrow 3_1^-$  transition, instead of the expected  $E2$  transition. Another problem is the lack of observed  $2^-$  and  $3^-$  states in this energy region. The next identified candidate for such a negative parity state is the  $3^-$  level at 2605 keV [5], and three other  $3^-$  levels have recently been identified below 3 MeV [6] (see Fig. 2). No candidates for  $2^-$  levels have ever been reported.

One explanation for this level spacing could be that anharmonicities in the quadrupole-octupole interaction have displaced the QOC  $2^-$  and  $3^-$  states to higher excitation energies. The calculations of Vogel and Kocbach for  $^{144}\text{Nd}$  [22] show this effect, to some degree. Their calculated level scheme is compared with experiment in Fig. 2 and shows a  $1^-$  state at the correct energy, but places the  $5^-$  and  $4^-$  states about 400 keV too high. However, their calculations for the ratios of transition rates from these QOC states are in good agreement with experiment, as shown in Table III. Definite identification of the QOC  $3^-$  level poses a problem, since its strength may be fragmented among the four  $3^-$  levels between 2.6 and 3.0 MeV. However the calculations of Vogel and Kocbach [22] indicate that the QOC  $3^-$  level should have a somewhat enhanced  $E3$  transition to the ground state and only one of these four levels shows this characteristic. This is the 2779 keV level, which has

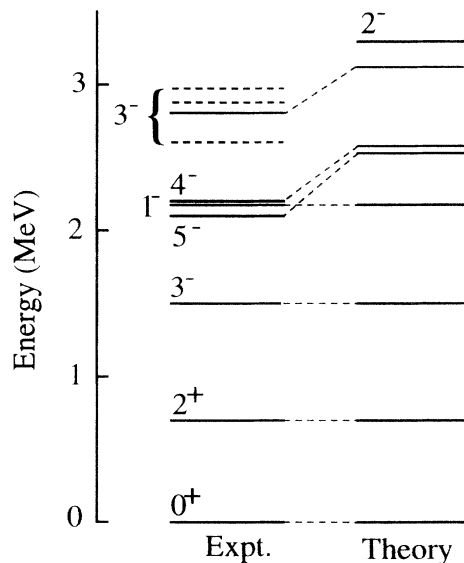


FIG. 2. Experimental and theoretical (from Ref. [22]) level schemes for the lowest negative parity states in  $^{144}\text{Nd}$ .

$B(E3; 3^- \rightarrow 0_1^+) = 7.3$  W.u. [6], and we can tentatively identify this level with the QOC  $3^-$  state (see Table II).

Another explanation of the structure of the  $5^-$  state, proposed by Cottle *et al.* [9], is that it contains a significant ( $\geq 50\%$ ) contribution from the  $\nu^2(2f_{7/1}1i_{13/2})_{5-}$  configuration. Such quasiparticle influence would not be surprising, since particle-core coupling model calculations [23, 24] have shown that the wave functions of the low lying positive parity states contain significant two-quasiparticle components. This explanation would presumably apply to the  $4^-$  level also, but cannot explain the presence of the  $1^-$  level, since this neutron configuration can only produce states with  $3^- \leq J^\pi \leq 10^-$ . In addition, this structure alone cannot account for the large  $B(E2; 5_1^- \rightarrow 3_1^-)$  and  $B(E3; 5_1^- \rightarrow 2_1^+)$  values reported in this work.

It is probable that the correct interpretation of these states involves both the QOC and quasiparticle configurations. If we consider the lower error bounds on the  $E2$  and  $E3$  decays from the  $5_1^-$  state to indicate the minimum possible QOC contribution to this state, we find that a QOC component of  $\geq 40\%$  is consistent with the data. This is not inconsistent with the  $\geq 50\%$  quasiparticle contribution inferred by Cottle *et al.* [9] and, in fact, between them these two results serve to characterize this state rather well. Such a structure can also provide an explanation for the apparent lowering in energy of the QOC  $3^-$ ,  $4^-$ , and  $5^-$  states, as compared to the calculations of Vogel and Kocbach [22]. In their random phase approximation (RPA) calculation of the structure of the quadrupole and octupole phonons, the only coupling of the  $\nu^2(2f_{7/2}, 1i_{13/2})$  configuration to play a significant role will be the contribution of the  $J^\pi = 3^-$  configuration to the octupole phonon. [Indeed, the influence of the  $1i_{13/2}$  neutron orbital can be seen in the large  $l = 6$

TABLE III. Ratios of  $E2$  and  $E3$  transition rates from negative parity states in  $^{144}\text{Nd}$ . The theoretical values come from the work of Vogel and Kocbach [22].

	$\frac{B(E2; 5_1^- \rightarrow 3_1^-)}{B(E2; 2_1^+ \rightarrow 0_1^+)}$	$\frac{B(E2; 1_1^- \rightarrow 3_1^-)}{B(E2; 2_1^+ \rightarrow 0_1^+)}$	$\frac{B(E3; 5_1^- \rightarrow 2_1^+)}{B(E3; 3_1^- \rightarrow 0_1^+)}$	$\frac{B(E3; (3_{\text{QOC}}^-) \rightarrow 0_1^+)}{B(E3; 3_1^- \rightarrow 0_1^+)}$
Theory	0.84	1.07	0.8	0.08
Experiment	0.8(4)	0.8(3)	1.0 (+5, -6)	0.22(2)

transfer observed in the  $^{143}\text{Nd}(d, p)$  channel leading to the  $3_1^-$  state in  $^{144}\text{Nd}$  [25]. If, as suggested above, the  $4^-$  and  $5^-$  quasiparticle configurations mix to a large degree with the relevant QOC quintuplet states, the resulting states will be lowered considerably, with respect to the unperturbed QOC energies. The effect on the QOC  $3^-$  state would be less, since much of the  $\nu^2(2f_{7/2}, 1i_{13/2})_3$ -configuration strength is already contained in the  $3_1^-$  state.

Some measure of the influence of this neutron configuration can be obtained from the results of the particle-core coupling calculations of Copnell *et al.* [23]. Although this model does not include the anharmonicities which give rise to the desired splitting, its results do show that the effect of this neutron configuration is to lower the energy of the  $4_1^-$ ,  $5_1^-$ , and  $3_2^-$  states by several hundred keV, relative to the unaffected  $1_1^-$  and  $2_1^-$  states. If these effects were superimposed on the calculated level spectrum of Vogel and Kocbach [22] a situation very similar to that found experimentally would result. It is also interesting to note that the  $E2$  decay from the  $1_1^-$  level (for which no quasiparticle component is possible) places a stricter bound on the QOC contribution to its wave function ( $\geq 60\%$ ) and that this level lies at almost exactly the energy calculated by Vogel and Kocbach [22].

In conclusion, we have measured lifetimes for the  $3_1^-$ ,  $5_1^-$ , and  $1_1^-$  states in  $^{144}\text{Nd}$  and have shown that the  $E2$  and  $E3$  transition rates for the  $5_1^-$  and  $1_1^-$  states are consistent with a structure formed by the coupling of the single quadrupole and single octupole excitations. In addition,  $E1$  transition strengths from the  $3_1^-$ ,  $5_1^-$ , and  $1_1^-$  levels are consistent with those used to identify possible QOC states in neighboring nuclei. A  $4^-$  level has been identified at 2205 keV and may be another member of the QOC quintuplet. The  $E2$  and  $E3$  transition properties of these states are in agreement with the calculations of Vogel and Kocbach [22], but are also compatible with a significant contribution from the  $\nu^2(2f_{7/2}, 1i_{13/2})$  configuration, as suggested by Cottle *et al.* [9]. One serious problem with this interpretation concerns the dominant  $M1$  character of the  $4_1^- \rightarrow 3_1^-$  transition. Indeed if the  $E2$  component of this transition were of the same strength as that predicted by Vogel and Kocbach [22], this would translate to an absolute  $M1$  strength of several  $\mu_N^2$ , clearly beyond the recommended upper limit for  $M1$  transitions [26]. It is essential, therefore, that this mixing ratio be confirmed and that the lifetime of the  $4_1^-$  level be determined. Also important to the QOC picture is the further study of the known  $3^-$  levels and the location of the "missing"  $2_1^-$  level.

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\*Present address: Euratom, Ispra, Italy.

†Present address: Redaktion der Physikalischen Blätter, Pappelallee 3, D-69469 Weinheim, Germany.

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