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Fast $E1$ transitions and evidence for octupole-octupole and quadrupole-octupole excitations in ^{144}Sm

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The nucleus ^{144}Sm has been studied with the $(n,n'\gamma)$ reaction and lifetimes of many states have been extracted from the observed Doppler shifts of the deexciting γ rays. A large number of fast $E1$ transitions have been observed and have led to the identification of possible members of the octupole-octupole and the quadrupole-octupole multiplets. Substantial fragmentation of the two-phonon octupole strength is indicated.

The role of vibrational excitations in nuclei has been studied for many years, but our knowledge of these fundamental modes remains incomplete. In the quadrupole case, equally spaced, degenerate phonon multiplets are expected, and there are many examples in even-even nuclei near closed shells where the E_{4^+}/E_{2^+} ratio is near the harmonic value of two. However, the anticipated, closely spaced 0^+, 2^+, and 4^+ two-phonon triplet is seldom observed. Evidence for three-phonon quadrupole states was sparse until the recent report[1] of a complete quintet of levels in ^{118}Cd, but recent lifetime measurements[2] in this nucleus indicate that the anharmonicities are larger than originally thought. In closed-shell nuclei, the octupole vibrations often occur at relatively low excitation energies and compete successfully with the quadrupole mode.[3] Data on multiphonon states involving octupole excitations are rare.

In two heavy nuclei, ^{146}Gd and ^{208}Pb, the 3^- state actually lies lower than the quadrupole phonon and is the first excited state in each. These states decay with large, similar $E3$ transition probabilities[4] of 37 and 34 Weisskopf units, respectively, suggesting that they are indeed collective octupole excitations. The unusual properties of these two nuclei have led to a number of searches (e.g., Refs. 5-7) for the expected $3^- \times 3^-$ quartet of states with spins and parities of 0^+, 2^+, 4^+, and 6^+ at about twice the energy of the 3^- phonon. No clear-cut identification of the members of the two-phonon quartet has emerged in either nucleus.

In ^{208}Pb, the large level density in the vicinity of the predicted energy of the two-phonon states contributes to the difficulties in identifying these levels. The octupole phonon in ^{146}Gd lies about 1 MeV lower in energy than the 3^- octupole state in ^{208}Pb. The smaller level density expected in the region of the predicted $(3^- \times 3^-)_I$ states has led to ^{146}Gd as the site of searches[7,8] for two-phonon octupole excitations.

Only in recent years has convincing evidence for two-phonon octupole states been reported. The most compelling data are the observed[9,10] stretch-coupled states of this type in ^{147}Gd and ^{148}Gd. The identifications of these states by their characteristic cascades of two $E3$ transitions was possible because, serendipitously, they occur as yrast states in these nuclei and lower multipolarity decays do not occur readily. But, since these states involve the coupling of one or two neutrons to the two-phonon octupole excitation ($J^\pi = \frac{19}{2}^-; \nu f_{7/2} \times 3^- \times 3^-$ in ^{147}Gd and $J^\pi = 12^+; \nu^2 \times 3^- \times 3^-$ in ^{148}Gd), their descriptions are not as straightforward as would be the expected case in a doubly closed-shell nucleus. Because of these ambiguities, the identification of two-phonon octupole multiplets in even-even nuclei remains an important goal.

Since strong octupole excitations should persist in nuclei near closed shells, we have attempted to locate two-phonon octupole states in ^{144}Sm, which is only two protons removed from ^{146}Gd, has as its second excited state a 3^- level, and is stable. Recent measurements[11] indicate that the octupole excitation in this nucleus, with a $B(E3;3^- \rightarrow 0^+) = 38 \pm 3$ Weisskopf unit (W.u.), is very similar to that of ^{146}Gd.

Inelastic neutron scattering (INS) experiments were performed with the University of Kentucky 7.0 MV Van de Graaff accelerator to search for two-phonon octupole states in ^{144}Sm. The scattering sample was 20.9 g of Sm_2O_3, enriched to 85.57% in ^{144}Sm and contained in a thin polyethylene cylindrical vial of 2.5 cm diameter and 2.7 cm height. An n-type HPGe detector of 35% efficiency and 1.8 keV at 1.33 MeV energy resolution was used to observe the reaction γ rays. The sample-to-detector distance was about 1 m and neutron-induced background was suppressed by time-of-flight discrimination.

The INS technique offers several advantages over other nuclear techniques in examining the low-lying, low-spin ($J \leq 7$) states.[12] The $(n,n'\gamma)$ reaction is not restricted by spin and parity selection rules and has good sensitivity for levels not strongly populated in other reactions, a necessity for the detection of possible two-phonon levels.

Gamma rays were placed on the basis of their excitation functions, and the analysis of γ-ray angular distribution and cross section data permitted the assignment of spins and parities to most of the levels in ^{144}Sm. In addition, since enhanced $E1$ transitions emerge as one of the characteristic signatures of octupole phonon transi-

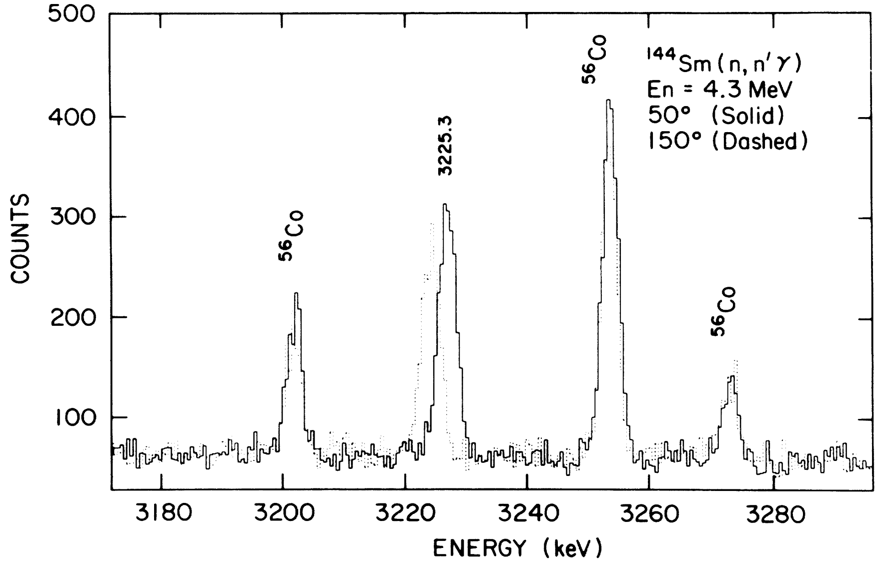

FIG. 1. Portion of the γ-ray spectrum measured following the $^{144}Sm(n,n'\gamma)$ reaction induced by 4.3 MeV neutrons. The shift in energy of the 3225.3 keV γ ray between the two angles of measurement is evident. The additional peaks are γ rays from a ^{56}Co calibration source which was placed near the detector during the measurements.

tions,[13,14] level lifetimes were measured following the INS reaction with the Doppler-shift attenuation method (DSAM). An example of the Doppler shift for a ^{144}Sm γ ray is shown in Fig. 1. The analysis methods of the DSAM data were the same as that described by Belgya *et al.*,[15] except that the Winterbon description[16] of the Doppler-shift attenuation process was used. The results of previous studies[17] were considered in making the spin-parity assignments.

In Fig. 2, a partial level scheme for ^{144}Sm containing the proposed two-phonon octupole states and quadrupole-octupole levels is displayed. The transition rates presented in Fig. 2 are for transitions of pure $E1$ and $E2$ multipolarity. Table I provides a listing of the pertinent levels and the associated γ-ray information. To demonstrate the degree of confidence one can hold in the measured transition probabilities, Table II gives a comparison of some level lifetimes we have determined using DSAM with those previously deduced in ^{144}Sm from nuclear resonance fluorescence measurements.[18]

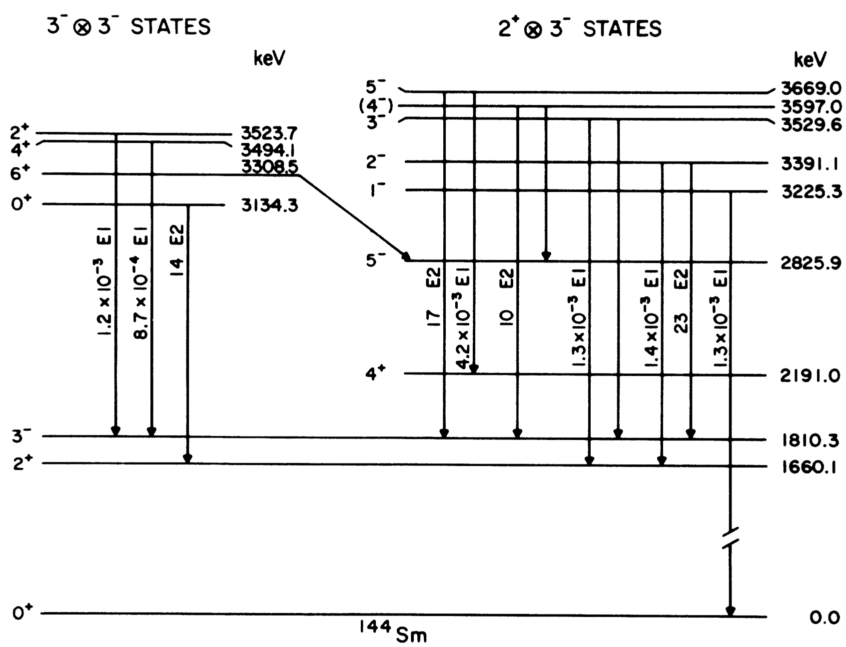

FIG. 2. Partial ^{144}Sm level scheme containing proposed octupole-octupole and quadrupole-octupole states. Measured transition rates in Weisskopf units are given on the transition arrows. Table I contains additional information about the deexciting γ rays.

TABLE I. Properties of levels attributed to octupole-octupole (positive parity) and quadrupole-octupole (negative parity) excitations in ^{144}Sm.

E_x (keV)	$J_i^\pi \rightarrow J_f^\pi$	E_γ (keV)	a_2	a_4	Branching	τ (fs) [a]	$B(\sigma L)$ [b]
3134.3	$0^+ \rightarrow 2^+$	1474.2	0.08(13)	0.06(18)	100	190^{+340}_{-80}	14^{+11}_{-9} $E2$
3308.5	$6^+ \rightarrow 5^-$	482.6	−0.65(32)		100		
3494.1	$4^+ \rightarrow 3^-$	1683.8	−0.47(6)	0.24(8)	100	86^{+15}_{-12}	$8.7^{+1.4}_{-1.3} \times 10^{-4}$
3523.7	$2^+ \rightarrow 3^-$	1713.4	−0.23(4)	−0.01(5)	100	61 ± 4	$(1.2 \pm 0.1) \times 10^{-3}$
3225.3	$1^- \rightarrow 0^+$	3225.3	−0.23(7)	−0.02(9)	100	$8.5^{+1.2}_{-1.1}$	$(1.3 \pm 0.2) \times 10^{-3}$
3391.1	$2^- \rightarrow 3^-$	1580.9	−0.48(12)	−0.18(19)	39(1)	31^{+5}_{-4}	23^{+4}_{-3} $E2$
	$\rightarrow 2^+$	1730.9	0.06(6)	−0.17(8)	61(1)		$(1.4 \pm 0.2) \times 10^{-3}$
3529.6	$3^- \rightarrow 3^-$	1719.3	0.41(13)	−0.03(17)	28(2)	30^{+5}_{-4}	c
	$\rightarrow 2^+$	1869.5	−0.28(8)	0.05(10)	72(2)		$1.3^{+0.3}_{-0.2} \times 10^{-3}$
3597.0	$(4^-) \rightarrow 5^-$	770.5	0.04(19)	−0.21(22)	25(2)	76^{+57}_{-27}	c
	$\rightarrow 3^-$	1786.7	0.13(12)	−0.01(16)	75(2)		10^{+6}_{-4} $E2$
3669.0	$5^- \rightarrow 4^+$	1478.0	−0.14(11)		64(8)	17^{+10}_{-7}	$4.2^{+4.4}_{-2.1} \times 10^{-3}$
	$\rightarrow 3^-$	1858.6	0.28(39)		36(8)		17^{+19}_{-9} $E2$

[a]Statistical uncertainties only are indicated.
[b]$E2$ transitions are indicated. All others for which values are reported are of $E1$ multipolarity.
[c]Mixed multipolarity transition.

Perhaps the most characteristic signature of two-phonon octupole structures we can hope to observe is the occurrence of fast $E1$ transitions from 2^+ and 4^+ members of the two-phonon octupole quartet to the 3^- octupole phonon.[14] Additionally, these states are not expected to exhibit extensive γ-ray branching to other levels. Only two levels in ^{144}Sm, the 4^+ level at 3494.1 keV and the 2^+ state at 3523.7 keV, are observed to decay solely by $E1$ transitions to the octupole state. Support for the assertion that these are collective two-phonon octupole levels comes from the observation of $E1$ transitions which are considerably faster [$B(E1)$'s$\simeq 10^{-3}$ W.u.] than the majority of $E1$ transitions in this mass region.[19] Moreover, these transitions are the fastest $E1$ transitions observed from any of the positive-parity states in ^{144}Sm. The fact that these levels have not been observed previously,[17] particularly in particle transfer reactions, is also consistent with their being of a more complex, two-phonon origin. While the γ-ray transitions from these levels are considerably faster than "normal" $E1$ transitions, there are a number of other fast $E1$ transitions from excited states of ^{144}Sm to the 3^- phonon.

Candidates for the 0^+ and 6^+ members of a two-phonon octupole multiple are difficult to identify, particularly since clearly distinguishing features, such as direct $E3$ transitions to the 3^- phonon, are not anticipated.[14] Low-lying 0^+ states at 2477.6, 2822.6, and 3134.3 keV have been reported[17] previously and are observed in our INS measurements. Each decays by γ-ray emission to the first excited state. The lowest of these has been suggested[20] as a $\pi(h_{11/2})^2_0 j_0^{-2}$ excitation of the ^{146}Gd core. The 2822.6 keV level is the excited state most strongly populated in the (p,t) two-neutron transfer reaction[21] and can be identified as the neutron pairing vibration. The 3134.3 keV state is also strongly populated, although with considerably smaller cross section than the 2822.6 keV state, in the two-neutron transfer reaction and is the best candidate for the $(3^- \times 3^-)_{0^+}$ state. However, from the decay of this state, one could certainly argue that it might be of two-phonon quadrupole origin. The large two-neutron transfer strength to this state can be taken as an indication of substantial interaction between the pairing vibrational and two-phonon modes. There are no other candidates in this energy region for the 0^+ member of the multiplet, with the possible exception of the 3823.6 keV level which decays to the first 2^+ level by a γ ray with an isotropic angular distribution, but a definite spin could not be assigned to this level.

The only definite 6^+ level in this region is the state at 3308.5 keV which deexcites to the 5^- level at 2925.9 keV by an $E1$ transition. The energy of this γ ray is too low to permit a meaningful lifetime determination by the DSAM.

Coupling between the quadrupole and octupole vibrational modes should produce a quintet of negative-parity states with angular momenta ranging from 1 to 5. The predictions of Vogel and Kocbach[22] indicate that the $2^+ \times 3^-$ states should lie in the same energy region as the two-phonon octupole states in ^{144}Sm. An additional search was therefore undertaken for the expected quintet of quadrupole-octupole states having spins and parities of 1^-, 2^-, 3^-, 4^-, and 5^-.

The 1^- state at 3225.3 keV clearly does not have a simple shell model interpretation, and several authors[11,23,24]

TABLE II. Comparison of additional ^{144}Sm level lifetimes with those previously measured by nuclear resonance fluorescence (see footnote).

E_{level} (keV)	τ (fs) DSAM	τ (fs) (γ,γ') [a]
2423.3	37^{+5}_{-4}	43 ± 6
2799.7	82^{+8}_{-7}	140 ± 27
3225.3	$8.5^{+1.2}_{-1.1}$	3.0 ± 0.3
3890.1	$2.0^{+1.9}_{-1.0}$	3.1 ± 0.5

[a]Reference 18.

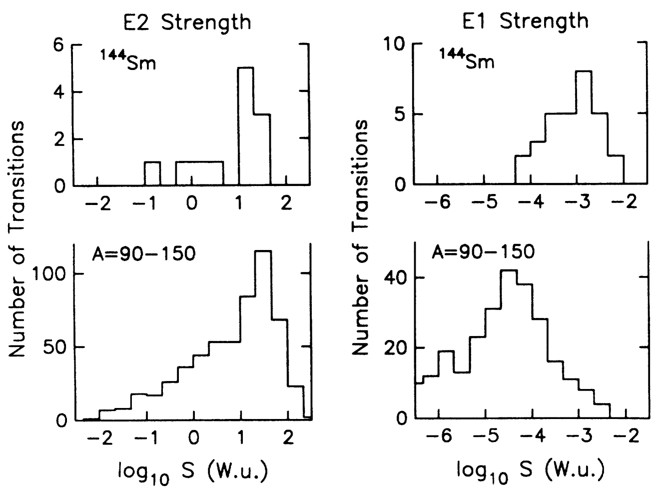

FIG. 3. Comparison of $E1$ and $E2$ γ-ray strengths in the $A=90$–150 region (Ref. 19) with measured values in ^{144}Sm. The logarithmic scale of the abscissa indicates the transition strengths in Weisskopf units.

have suggested this level as the 1^- member of the quadrupole-octupole multiplet. In particular, the arguments of Barfield and co-workers,[11] which are based primarily on the previously observed[24] fast $E1$ ground-state transition from this level and the $B(E1)$ they measured for the $3_1^- \rightarrow 2_1^+$ transition, for the interpretation of this state as a $(2^+ \times 3^-)_{1^-}$ excitation are quite compelling. The $B(E1;1^- \rightarrow 0^+)$ we deduce from our measured lifetime of the 3225.3 keV level is not as large (see Table II) as that reported by Metzger,[18,24] but it clearly indicates a fast $E1$ transition. Although a branch to the 2_1^+ quadrupole phonon had not been observed,[18,24] Barfield *et al.*[11] suggest that branching from this 1^- level to the first excited state should occur with a $B(E1;1^- \rightarrow 2^+)/B(E1;1^- \rightarrow 0^+)$ ratio of unity. Neither this $1^- \rightarrow 2^+$ branch nor a γ-ray decay to the 3^- one-phonon level are observed in the present measurements. With Barfield's assumption of a $B(E1)$ ratio of unity, though, the branch to the 1660.1 keV 2^+ level should have been observed. We have determined an upper limit of this ratio to be 0.3.

While the 2^- and 3^- members of the quintet will not likely decay directly to the ground state, transitions to the 2_1^+ and 3_1^- levels are expected. Despite the fact that it is more than 1 MeV lower than the predicted[22] energy, the best 2^- candidate is at an energy of 3391.1 keV. This level is the only identified 2^- state which decays to both of the one-phonon states, and the $E1$ transition to the 2^+ quadrupole phonon is fast (see Fig. 2). On similar grounds, the best 3^- candidate is the state at 3529.6 keV. We should point out, however, that there are several other 2^- and 3^- levels which decay solely to the first excited state by fast $E1$ transitions.

The most likely 4^- multiplet member, based on the observed fast $E2$ transition to the 3^- octupole level, is the 3597.0 keV state, although the spin assignment is tentative.

There are several well-established 5^- levels in ^{144}Sm; however, only one is observed to decay to both the 3_1^- and 4_1^+ levels. A decay to the first 4^+ level might be anticipated since this state is of similar structure as the 2_1^+ state.[25] Even in view of the large uncertainties, the fast transitions from the 3669.0-keV level support the interpretation of this level as a quadrupole-octupole two-phonon state.[23]

We should also note that we observe no good candidates for the 2^+ or 4^+ members of a two-phonon quadrupole triplet in ^{144}Sm. Since the quadrupole phonon in this nucleus is not very collective,[26] this result may not be too surprising.

In Fig. 3 we compare the $E1$ and $E2$ γ-ray strengths determined in our experiments with those compiled[19] for nuclei in the $A=90$–150 region. While we have measured the lifetimes of only a few $E2$ transitions, their strengths appear to mirror the strength histogram in this mass region. On the other hand, the measured $E1$ strength distribution clearly indicates that the $E1$ transitions in ^{144}Sm are fast. The observed $E1$ transition rates are generally comparable to the values obtained in octupole-deformed actinides,[27,28] and are substantially faster than the average $E1$ rate of 5×10^{-5} W.u. extracted for the $A=90$–150 mass region. We should also note here that many of the fastest transitions in the compilation—i.e., those comprising the "tail" of the strength histogram shown in the lower portion of Fig. 3—are from states of known octupole character in the $A=140$–150 region. This fact lends further credence to the assertion that the fast $E1$ transitions observed in ^{144}Sm can be attributed to the presence of strong octupole correlations. Similar behavior has also been observed[29] in ^{142}Nd, another $N=82$ isotone.

While fast $E1$ transitions may be interpreted as direct evidence for the existence of two-phonon octupole vibrational states in ^{144}Sm, the abundance of fast $E1$ transitions observed suggests substantial fragmentation of the two-phonon octupole strength and considerable mixing with other states. The observation of related positive and negative parity states in the 3–4 MeV excitation region could also be taken as evidence of clustering;[30] however, the fragmentation suggested above would make the identification of possible candidates for these states tenuous. Further experimental tests, like inelastic charged-particle scattering with special emphasis on two-step processes or multiple $E3$ Coulomb excitation, may help clarify the viability of the present proposals which are based mainly on the observation of electromagnetic decay properties.

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