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Direct proof of the two-phonon character of the dipole excitations in ¹⁴²Nd and ¹⁴⁴Sm around 3.5 MeV

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Resonant inelastic proton scattering experiments were carried out to investigate the γ decay of the 1⁻ state at an excitation energy of around 3.5 MeV in the N=82 nuclei ¹⁴²Nd and ¹⁴⁴Sm. γ -ray branching ratios were determined from γ -proton coincidences in both nuclei. The newly observed γ decay of the dipole excitation at 3225 keV in ¹⁴⁴Sm to the one-phonon states, in particular the $(1^- \rightarrow 3_1^-) E2$ transition to the octupole phonon state, is the first direct proof of the two-phonon character of the 1⁻ state in any N=82 nucleus. [S0556-2813(96)50208-6]

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The firm assignment of a multiphonon structure to the vibrational states of nuclei is one of the fundamental problems in nuclear physics. From the picture of collective low-lying quadrupole and octupole excitations that are considered as "phonons" emerges the idea of multiple-phonon excitations. The coupling of two quadrupole phonons $(2^+ \otimes 2^+)$ leads to a multiplet of three states with spin and parity $J^{\pi}=0^+$, 2^+ , and 4^+ . Such multiplets have been observed in a number of spherical nuclei [1–3] and evidence for $2^+_{\gamma} \otimes 2^+_{\gamma}$ states has been discussed in deformed nuclei [4–8]. In contrast, the knowledge about the $2^+ \otimes 3^-$ [9–17] and the $3^- \otimes 3^-$ [18] multiplets is still rather sparse.

In a simple harmonic picture the members of these multiplets are degenerate in energy and lie at the sum energy of the single-phonon constituents. However, remaining interactions split the multiplet and even the center of gravity of the multiplet states can be shifted. A crucial problem in an experimental investigation of multiphonon multiplets is a reliable identification of the member states. The determination of spin and parity of states in the expected energy region alone is not sufficient because the level density is high and there is often more than one candidate. Therefore the knowledge of the absolute transition strengths and/or γ -decay properties and their comparison with theoretical predictions are important to establish the two-phonon character of a vibrational state.

In the semimagic N=82 isotones which include ¹³⁸Ba, ¹⁴⁰Ce, ¹⁴²Nd, and ¹⁴⁴Sm the $J^{\pi}=1_{ph}^{-}$ member of the $2^+\otimes 3^-$ quadrupole-octupole multiplets has been identified at an excitation energy of around 3.5 MeV [10–12,16,17]. This assignment was based on two observations: First, this state is the only strong electric dipole excitation up to 4.3 MeV and its energy is close to the sum energy of the 2^+ and 3^- state. Second, it shows an enhanced electric-dipole transition strength¹ to the ground state, which is a hint of a large octupole component in the wave function, i.e., a hint of a $2^+\otimes 3^-$ structure.

Still one has to consider that the *E*1 transitions are very weak ($\approx 3 \times 10^{-3}$ W.u.) as compared to the giant dipole resonance (≈ 10 W.u.). Thus, a real proof and a direct evidence of a two-phonon nature of the 1_{ph}^{-} state must come from the observation of an allowed *E*2 or *E*3 decay to the one-phonon states (see Fig. 1). In a simple picture one ex-

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¹Enhanced *E*1 strength means that the transition strength is about 30 times larger than the typical value of $\simeq 10^{-4}$ W.u. (Weisskopf unit).



FIG. 1. Decay pattern expected from the γ decay of a two-phonon 1_{ob}^- state.

pects that the $(1_{ph}^{-} \rightarrow 2_{1}^{+})E3$ transition and the $(1_{nh}^{-} \rightarrow 3_{1}^{-})E2$ transition have transition strengths that are identical to those of the 3_1^- and 2_1^+ states to the ground state, respectively, because the "same phonon" is destroyed [19]. However, the considered E3 transition competes directly with the lower multipolarity E1 transition with the same energy. Therefore it is nearly impossible to measure the B(E3) strength directly. The competing $(1^-_{ph} \rightarrow 2^+_1)E1$ transition is enhanced for the very same reasons as the $(1_{ph}^{-} \rightarrow 0_{g,s}^{+})E1$ ground-state transition. Very recently this electric-dipole transition could be observed for the first time in a N=82 nucleus [15]. Photon scattering (γ, γ') experiments gave previously upper limits for the E1 decay branches to the single-phonon states of the order of 5% or less in ¹³⁸Ba and ¹⁴⁰Ce [16,17].

An observation of the $(1_{ph}^- \rightarrow 3_1^-)E2$ transition is therefore a much more direct proof for the two-phonon structure. This transition was not observed previously in any of the semimagic N=82 nuclei. Only upper limits have been derived from a $(n,n'\gamma)$ study [15]. The situation is quite different for the nuclei with two additional neutrons (N=84). Here the $(1_{ph}^- \rightarrow 3_1^-)E2$ transition strength and furthermore the complete $2^+ \otimes 3^-$ multiplet has been observed in ¹⁴⁴Nd [9,14].

It is the aim of the present paper to show the results of resonant $(p, p' \gamma)$ studies to investigate the γ decay of the $1_{\rm ph}^-$ state in the N=82 nuclei ¹⁴²Nd and ¹⁴⁴Sm in order to establish the two-phonon structure of these states.

The energy of the 1_{ph}^{-} state lies far above the yrast line and an appreciable cross section for its population can only be expected for special reactions such as $(n, n' \gamma)$ and (γ, γ') . These reactions have the disadvantage that they are either not selective (as neutron scattering [13]) or have a very high low-energy background (as photon scattering [16]). Both disadvantages make the observation of very weak γ branches with low transition energies very difficult.

The inelastic proton scattering at proton energies corresponding to the isobaric analogue resonances (IAR) is a fruitful tool to investigate high-lying negative parity states in even-even nuclei (see, e.g., Ref. [20]). It can be expected that the states of the $2^+ \otimes 3^-$ quintuplet can be excited with appreciable strength in this reaction [21-28]. According to the properties of the chosen isobaric analogue resonance only a small spin and parity window will be strongly excited. The selectivity of this reaction is an important advantage and leads to γ spectra containing only a few γ lines. In particular, one is able to detect very weak γ branches.

The protons were scattered from a 1 mg/cm² selfsupporting metallic foil, enriched to 95.7% and 96.3% for ¹⁴²Nd and ¹⁴⁴Sm, respectively. The experiments were carried out at the OSIRIS-Cube spectrometer [29]. It was equipped with six escape-suppressed Ge detectors placed at 45° , 90° , and 135° with respect to the beam axis. The total solid angle covered by these detectors was 4.5% of 4π . In addition two Si(Li) detectors were placed at 90° with respect to the beam axis for particle detection, which covered a relatively large solid angle of 3.2% of 4π [30]. The particle detectors were used at room temperature. A resolution of 45 keV for a 241 Am α source was obtained. The energy resolution of the measured proton spectra was about 90 keV. This loss of resolution comes from the large opening angle of the detectors and from the target thickness. Proton-singles, γ -singles, $\gamma\gamma$ -, and γ -proton coincidences were recorded with the Cologne FERA analyzer system [31] during a 60 hour beamtime for the ¹⁴²Nd target and a 120 hour beamtime for the ¹⁴⁴Sm target. Two million γ -proton coincidences were collected in the ${}^{144}\text{Sm}(p, p' \gamma)$ reaction.

Typical γ -proton coincidence spectra are shown for the 144 Sm $(p, p' \gamma)$ reaction in Fig. 2. In the upper part the proton spectrum is presented. In particular, the strong excitation of states with energies between 3.5 and 5.0 MeV strikes the eye. This is due to the excitation via the IAR and results from the selectivity of this excitation mechanism. The two dotted lines show a cut window in proton energy loss between 3175 and 3275 keV. This is around the excitation energy of the electric dipole 1_{ph}^{-} state in ¹⁴⁴Sm at 3225 keV. The γ rays that were coincident to protons with these energy losses are shown in the lower part of Fig. 2. The two marked γ lines at 1415 keV and 1566 keV correspond (in energy) to the $(1_{ph}^{-} \rightarrow 3_{1}^{-})$ and $(1_{ph}^{-} \rightarrow 2_{1}^{+})$ transitions in ¹⁴⁴Sm, respectively. The other γ lines have been identified as depopulating γ transitions of excited states that are close in energy to the 1⁻_{ph} state in ¹⁴⁴Sm.

To establish these two and other observed γ transitions in the level scheme the γ -proton coincidences were analyzed. Coincident γ spectra were analyzed with gates in proton energy corresponding to definite excitation energy. Within the energy range from 1 to 6 MeV we have chosen 40 keV wide windows with a mean energy every 20 keV in proton energy loss. An example of the coincidence rates in dependence on the excitation energy is shown in Fig. 3. One can see that the coincidence rate of the well known $(2_1^+ \rightarrow 0_{g.s.}^+)E2$ transition in ¹⁴²Nd rises strongly at the excitation energies of 1576 keV



FIG. 2. γ -proton coincidence spectra taken from resonant inelastic proton scattering on ¹⁴⁴Sm. The scattered proton spectrum is shown in the upper part (a). Some strong excitations are labeled with their spin and parity assignment. The label "2⁻&1⁻" indicates a doublet at 4.3 MeV. The resolution obtained in the proton spectrum is about 90 keV. The two dotted lines indicate a 100 keV window around the 3225 keV 1⁻_{ph} state in ¹⁴⁴Sm. The γ spectrum in the bottom part (b) is coincident to the protons in this marked energy range. The two labeled γ lines at 1415 keV and 1566 keV have been established as depopulating $(1^-_{ph} \rightarrow 3^-_1)$ and $(1^-_{ph} \rightarrow 2^+_1)\gamma$ transitions in ¹⁴⁴Sm. The other γ lines depopulate other states in ¹⁴⁴Sm that are close in energy to the dipole 1^-_{ph} state.

and 2084 keV. The maximum at an excitation energy of 1576 keV corresponds to the direct excitation of the 2_1^+ state in ¹⁴²Nd. The second maximum in the coincidence rate indicates the feeding of this 2_1^+ state by the 3_1^- state at 2084



FIG. 3. Coincidence rate of the ground-state transition from the 2_1^+ level in ¹⁴²Nd at 1576 keV plotted versus the excitation energy determined from coincidence γ spectra with a window of 40 keV width in excitation energy. The coincidence rate has maxima at excitation energies of about 1576 keV and 2084 keV. The lowest excitation energy (1576 keV) indicates the direct excitation of the 2_1^+ state in ¹⁴²Nd. The second maximum in coincidence rate indicates the feeding of the 2_1^+ state by the decay of the 3_1^- state at 2084 keV.



FIG. 4. Coincidence rates of the depopulating γ rays of the 1_{ph}^{-} level in ¹⁴⁴Sm at 3225 keV plotted versus the excitation energy in the energy range from 3.0 to 3.5 MeV. These rates were extracted from coincidence γ spectra with a window of 40 keV width in excitation energy. The energies of the observed γ transitions of 1566 keV in the middle part (b), of 1415 keV in the lower part (c), and the strong correspondence to the excitation energy at 3225 keV (a) in all plots establish these γ transitions as the depopulating transitions of the 1_{ph}^{-1} state to the one-phonon states. The dashed curves are plotted to guide the eye.

keV. The shape of this "excitation function" is mainly dominated by the resolution of the proton detectors. For the data with sufficient statistics an additional evaluation of the $\gamma\gamma$ coincidences was done. According to the above procedure we were able to observe very weak γ transitions and to place the corresponding transitions into the level scheme. 161 γ transitions and 61 excited states of ¹⁴⁴Sm could easily be placed.

To establish the two marked transitions in the γ spectrum in Fig. 2 as depopulating γ transitions of the 1_{ph}^{-} state in ¹⁴⁴Sm the coincidence rates of these γ transitions were compared with the coincidence rates of the well-known groundstate γ transition from the 1_{ph}^{-} state. This comparison is

TABLE I. Deduced branching ratios and transition energies of the 2_1^+ , 3_1^- , and 1_{ph}^- states in ¹⁴²Nd. The results obtained in this work are in good agreement with previous data.

	$J^{\pi}_i { ightarrow} J^{\pi}_f$		I_{γ}		$T_{1/2}$ [fs]		
E_x [keV]	Ref. [34]	E_{γ} [keV]	This work	Ref. [15]	Ref. [15]		
1575.8(2)	$2^+_1 \rightarrow 0^+_{gs}$	1575.8(2)	100.0	100.0	159 ± 3		
2084.4(2)	$3_1^- \rightarrow 2_1^+$	508.6(2)	100.0	100.0	630^{+530}_{-200}		
3424.1(4)	$1^{-}_{ph} \rightarrow 0^{+}_{gs}$	3424.2(3)	100.0	100.0	1.8 ± 0.4		
	$\rightarrow 2_1^+$	1847.5(6)	2.7(5)	2.9(8)			
	$\rightarrow 3^{-}_{1}$	(1339.7) ^a	<1.6	< 3.6			

^a γ transition not observed.

TABLE II. Deduced branching ratios, transition energies, and lifetimes of the 2_1^+ , 3_1^- , and 1_{ph}^- states in ¹⁴⁴Sm. The decay from the 1_{ph}^- state to the one-phonon states is observed for the first time.

¹⁴⁴ Sm					
	$J_i^{\pi} \rightarrow J_f^{\pi}$		I_{γ}		
E_x [keV]	Ref. [32]	E_{γ} [keV]	This work	$T_{1/2}$ [fs]	
1660.1(2)	$2_1^+ \rightarrow 0_{gs}^+$	1660.1(2)	100.0	84 ± 3 ^a	
1810.3(2)	$3_1^- \rightarrow 2_1^+$	150.2(2)	100.0	24500 ± 3500^{b}	
	$\rightarrow 0_{gs}^+$	1810.4(3)	7.1(9) ^c		
3225.5(3)	$1^{-}_{ph} \rightarrow 0^{+}_{gs}$	3225.5(2)	100.0	1.94 ± 0.26 ^d	
	$\rightarrow 2_1^+$	1565.8(4)	1.9(3)		
	$\rightarrow 3^{-}_{1}$	1414.9(5)	1.5(3)		

^aReference [35].

^bReference [36].

^cIntensity agrees with the value given in Refs. [11,13].

^dLifetime deduced from $\Gamma_0^2/\Gamma = 220 \pm 30 \text{ meV} [10]$ and the branching ratios determined in this work.

shown in Fig. 4 for the energy range from 3.0 to 3.5 MeV. In the three parts of Fig. 4 the coincidence rates of the 3225 keV, 1566 keV, and 1415 keV γ transitions in ¹⁴⁴Sm are shown. The dashed curves in Fig. 4 are included to guide the eye. We note that the three plots look very similar. Thus we have identified the γ transitions as depopulating γ transitions of the $1_{\rm ph}^{-}$ state in ¹⁴⁴Sm, although the statistics of the low energy γ transitions is low. The branching ratios were deduced from the γ -proton coincidences. Angular correlation effects could be neglected because of the large solid angle of the particle detectors. The complete results of the measurements will be reported elsewhere. In the following the main focus will be on the decay of the $1_{\rm ph}^{-}$ state in ¹⁴²Nd and ¹⁴⁴Sm.

In ¹⁴²Nd we were able to observe the weak *E*1 γ decay of the 1⁻_{ph} state at 3424 keV to the 2⁺₁ state at 1576 keV. The branching ratio is $I(1^-_{ph} \rightarrow 2^+_1)/I(1^-_{ph} \rightarrow 0^+_{g.s.}) = 0.027(5)$. Within the experimental errors this ratio agrees well with the results given in the literature [15]. The branching to the 3⁻₁ state at 2084 keV could not be observed in this work. We deduced an upper limit of $I(1^-_{ph} \rightarrow 3^-_1)/I(1^-_{ph} \rightarrow 0^+_{g.s.}) < 0.016$. In Table I the results are summarized and a comparison with previous data is given.

In ¹⁴⁴Sm neither the γ decay of the 1_{ph}^- state at 3225 keV to the 2_1^+ state at 1660 keV nor its decay to the 3_1^- state at 1810 keV were known. In Table II the deduced γ energies and branching ratios are presented for the 2_1^+ , 3_1^- , and $1_{ph}^$ states. Where possible a comparison is given with branching ratios known from literature. We deduced the ratios $I(1_{ph}^- \rightarrow 2_1^+)/I(1_{ph}^- \rightarrow 0_{g.s.}^+)=0.019(3)$ for the 1565.8(4) γ transition and $I(1_{ph}^- \rightarrow 3_1^-)/I(1_{ph}^- \rightarrow 0_{g.s.}^+)=0.015(3)$ for the 1414.9(5) γ transition from our data.

The lifetime of the 1_{ph}^{-} state was corrected with the new branching ratios. We determined $T_{1/2} = (1.94 \pm 0.26)$ fs from the value $\Gamma_0^2/\Gamma = 220 \pm 30$ meV obtained from the (γ, γ') experiments [10]. Moreover, one obtains the reduced transi-

TABLE III. Reduced transition probabilities of the 2_1^+ , 3_1^- , and 1_{ph}^- states in ¹⁴⁴Sm. The values for the 1_{ph}^- state are calculated from Table II. A comparison is made with previous experiments and results obtained from QPM calculations. The deduced $B(E2;1_{ph}^-\rightarrow 3_1^-)$ value is a proof of the two-phonon structure in the wave function of the 1_{ph}^- state.

		144 Sm				
$J_i^{\pi} \rightarrow J_f^{\pi}$	$B(E\lambda)\downarrow$ [W.u.]					
	Theory	Experiment				
Ref. [32]	Ref. [33]	This work	Others			
$2_{1}^{+} \rightarrow 0_{gs}^{+}$	13.3		11.9(18) ^a			
$3_1^- \rightarrow 0_{gs}^+$	34.4		38(3) ^b			
$3_1^- \rightarrow 2_1^+$	0.33×10^{-2}		$0.28(4) \times 10^{-2}$ b			
$1_{\rm ph}^{-} \rightarrow 0_{\rm gs}^{+}$	1.10×10^{-3}	$3.66(50) \times 10^{-3}$	$3.5(5) \times 10^{-3}$ c			
1 5			$1.3(2) \times 10^{-3}$ d			
$1_{ph}^{-} \rightarrow 2_{1}^{+}$	0.10×10^{-3}	$0.61(13) \times 10^{-3}$				
$1^{-}_{ph} \rightarrow 3^{-}_{1}$	13.7	16.6(40)				

^aReference [32].

^bReference [36].

^cReference [10].

^dReferences [11,13].

tion probabilities from the 1_{ph}^{-} state to lower-lying states. The reduced transition probabilities are $B(E1; 1_{ph}^{-} \rightarrow 0_{g.s.}^{+})$ =3.66(50) mW.u., $B(E1;1_{ph}^{-}\rightarrow 2_{1}^{+})=0.61(13)$ mW.u., and $B(E2;1_{\text{ph}}^{-}\rightarrow 3_{1}^{-})=16.6(40)$ W.u. They are given in Table III. In particular the strong $(1_{ph}^- \rightarrow 3_1^-)E2$ decay to the octupole phonon state at 1810 keV is a compelling proof of the two-phonon character of the 1_{ph}^{-} state at 3225 keV. We stress once more that this γ transition is here established for the first time in a N = 82 nucleus. The deduced B(E2) value for the $(1_{ph}^{-} \rightarrow 3_{1}^{-}) \gamma$ decay is in good agreement with the corresponding $B(E2;2_1^+ \to 0_{g.s.}^+)$ value of 11.9(18) W.u. [32] that is expected for a nearly harmonic two-phonon state [19]. This confirms the dominant two-phonon character of the $1_{\rm ph}^{-}$ state. In Table III a comparison with theoretical predictions [33] is shown for the reduced transition probabilities. These predictions are calculated in the framework of the quasiparticle phonon model (QPM) and agree well with the experimental values.

The new results obtained by the resonant $(p, p' \gamma)$ reaction confirm this method as a fruitful tool to observe extremely weak γ transitions. To obtain a more systematic knowledge about the structure of the 1_{ph}^- state in the N=82 nuclei further investigations will be carried out in the future.

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