Two-Phonon Octupole Excitation in ^{208}Pb

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Experimental evidence for identification of the 5241-keV 0^+ state in ^{208}Pb as the lowest-spin member of the $3^- \otimes 3^-$ quartet is reported. This state, observed previously in neutron transfer reactions and inelastic proton scattering, has been populated with the inelastic neutron scattering reaction. Its decay to the 3^- octupole phonon in ^{208}Pb is confirmed by γ-γ coincidence measurements, and the spin-0 assignment is substantiated by the population cross section. The cascade of two $E3$ transitions thus established is an expected signature of a two-phonon octupole excitation.

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The low-lying 3^- excitations observed in even-even nuclei near closed shells have frequently been interpreted as collective surface vibrations of the octupole type, and the $E3$ strengths of the $3^- \rightarrow 0^+$ transitions in these nuclei support this view [1]. Moreover, a two-phonon $3^- \otimes 3^-$ quartet of states $0^+, 2^+, 4^+$, and 6^+) is expected at about twice the energy of the one-phonon octupole excitation. The identification and characterization of these states should provide valuable information about the effects of the Pauli principle and phonon-phonon coupling between elementary modes of excitation.

In recent years, a number of searches for two-phonon octupole excitations in spherical even-even nuclei such as ^{146}Gd [2,3] and ^{208}Pb [4–10] have been reported, but no clear-cut identification of the members of the two-phonon quartet has emerged. Individual, but more complex, states of the two-phonon type have been established in ^{147}Gd [11] and in the $N = 84$ isotones ^{144}Nd [12], ^{146}Sm [12], and ^{148}Gd [13,14]. However, because these states involve the coupling of particles to the two-phonon octupole excitation, e.g., $\nu^2 \otimes (3^- \otimes 3^-)_{6^+}$ in ^{148}Gd, their descriptions are not straightforward. The identification of these states by the characteristic cascade of two enhanced $E3$ transitions has been possible because they occur as yrast states and lower multipolarity decays do not occur readily.

The 3^- first excited state of ^{208}Pb with an $E3$ transition strength of 34 W.u. [1] has long been interpreted as a collective vibrational state. In this nucleus, 0^+ excited states are known [5,15,16] at 4866 and 5237 keV, approximately twice the energy of the octupole phonon, whereas it is anticipated that three such states should be present, i.e., the neutron and proton pairing vibrations and the lowest-spin member of the two-phonon octupole quartet [17]. Julin and co-workers [5] searched for the missing 0^+ state by observing $E0$ internal conversion transitions to the ground state following the ^{207}Pb(d,p) and ^{208}Pb(p,p') reactions, but found only the two aforementioned states and no additional decay branches from these states. The lower of these two states now seems firmly established [16–18] as the neutron pairing vibrational state in ^{208}Pb, but, despite arguments [5,18] the higher state is predominantly of two-phonon octupole character, the situation remains unclear.

The most convincing evidence for the 0^+ member of the two-phonon octupole quartet would be the measurement of a large $B(E3; 0^+_{2\text{ phonon}} \rightarrow 3^-_{1\text{ phonon}})$ value. However, the predicted lifetime of the two-phonon 0^+ state (from tens to hundreds of picoseconds, depending on the assumptions used), combined with its nonobservation in Coulomb excitation or other reactions where the recoil distance lifetime method could be used [6,7,9,10], indicates that the measurement of this lifetime will be difficult. In the absence of this $B(E3)$ determination, the best evidence for the two-phonon state would be the observation of a 0^+ state which preferentially decays by an $E3$ transition to the one-phonon state over other, lower multipolarity transitions.

Inelastic neutron scattering with γ-ray detection should be an excellent probe in search for two-phonon octupole excitations in ^{208}Pb. Statistical model calculations indicate that all states with spins ≤ 6 below 6.0 MeV in excitation should be populated with sufficient cross sections to permit their observations. The nonselective nature of this reaction, coupled with the excellent resolution obtained through γ-ray detection, thus provides a good opportunity to observe two-phonon octupole excitations. With the $(n, n'\gamma)$ reaction, the identification of a spin-0 state may be accomplished if the excitation functions of transitions from the level agree in both shape and magnitude with that expected from the statistical model calculations and the angular distributions of the transitions are isotropic. Furthermore, the expected two-phonon 0^+ state should lie at approximately twice the energy of the one-phonon excitation, exhibit no Doppler shift since the lifetime is expected to be $\gg 1$ ps, and have a branching ratio favoring

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the $E3$ transition to the one-phonon state. Therefore, a detailed study of ^{208}Pb in the 4.5- to 6.0-MeV energy region was undertaken with the $(n, n'\gamma)$ reaction, the ultimate goal being to identify all states with spins ≤ 6 that decay by γ-ray emission, with special emphasis on two-phonon octupole candidates.

Gamma-ray excitation functions, angular distributions, Doppler-shift attenuation lifetimes, and γ-γ coincidences were measured with the $(n, n'\gamma)$ reaction and accelerator-produced monoenergetic neutrons. The experimental apparatus and methods used for these studies at the University of Kentucky have been reported on many occasions, and the latest developments have been described recently [19]. The γ rays from the $(n, n'\gamma)$ reaction (Fig. 1) were detected using a BGO-Compton-suppressed, *n*-type HPGe detector having a relative efficiency of 57% and an energy resolution of 2.0 keV at 1.33 MeV. Time-of-flight suppression was used to reduce extraneous background events. For the excitation function and angular distribution measurements, a large (77.6 g), highly enriched (99.08%) isotopic sample of metallic ^{208}Pb was used.

In addition to these singles experiments, γ-γ coincidence measurements have been performed using "beams" of fast neutrons produced with a forced-reflection neutron collimator. A 2-kg sheet of natural lead served as the target for this $(n, n'\gamma\gamma)$ coincidence measurement. Coincidences were recorded with three HPGe detectors of 32%, 52%, and 57% relative efficiencies in a close geometrical arrangement located as near as possible to the sample which was irradiated with the collimated neutrons; the sample and detector array were slightly over one meter from the source of neutrons. Figure 2 illustrates the gate on the 2614-keV $3^- \rightarrow 0^+$ transition in ^{208}Pb from the ^{nat}Pb$(n, n'\gamma\gamma)$ reaction at $E_n = 6.5$ MeV. This gate, in combination with others on transitions from low-lying states, and threshold determinations (Fig. 3) permit the unambiguous placement of γ rays that populate the first excited state of ^{208}Pb. With the γ-ray singles measurements, these data provide an excellent opportunity for detecting even weakly populated levels; the decays of all known $J \leq 6$ states below 5.8 MeV in ^{208}Pb were observed and a number of new levels in ^{208}Pb, including two below 5.0 MeV, have been established [20].

The coincidence of a 2626.5 ± 0.4-keV γ ray with only the 2614-keV transition (Fig. 2) and its excitation threshold (Fig. 3) confirm that this γ ray is from a state at 5241 keV in ^{208}Pb. (It should be noted that a γ ray of this energy has also been observed in the $(n, n'\gamma)$ reaction with reactor fast neutrons and placed from a state at 5241 keV [21], but no further characterization was reported.) Two known, well-established states in this energy region are expected to be populated by inelastic neutron scattering: the aforementioned 0^+ state and a 3^- state at 5245 keV. The 2631-keV γ ray from the latter state is clearly evident in Figs. 1 and 2; no evidence is found for additional states in this region. The measured angular distribution (Fig. 4) of the 2626-keV γ ray is isotropic, as expected for a spin-0 state. Moreover, no Doppler shift is observed for this transition (Fig. 4), indicating that the lifetime of the state is >1 ps. For comparison, also shown in Fig. 4 are the angular distribution and the Doppler shift of the 2631-keV ($3^- \rightarrow 3^-$) γ-ray transition.

The magnitude of the cross section for production of the 2626-keV γ ray (Fig. 3) also demonstrates that the spin of the 5241-keV state is zero. If the spin of this state were 1 or 2, ground-state transitions would be anticipated, as are observed [20,22] for all other known states of these spins in ^{208}Pb. Based on the cross section (Fig. 3), angular distribution (Fig. 4), and Doppler shift (Fig. 4) data, other

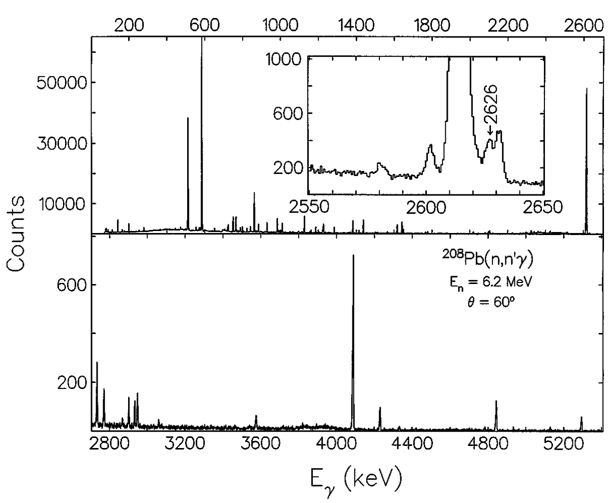

FIG. 1. Gamma-ray spectrum from the ^{208}Pb$(n, n'\gamma)$ reaction measured at 60° relative to the incident 6.2-MeV neutrons. The 2626-keV γ ray is indicated in the inset.

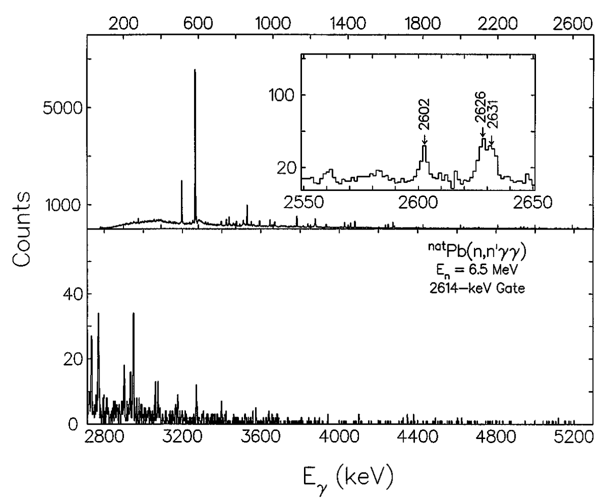

FIG. 2. Gamma-ray coincidence spectrum obtained by gating on the 2614-keV ($3^- \rightarrow 0^+$) γ ray from the ^{nat}Pb$(n, n'\gamma\gamma)$ reaction at an incident neutron energy of 6.5 MeV. The 2626-keV γ ray and other nearby coincident γ rays are indicated in the inset.

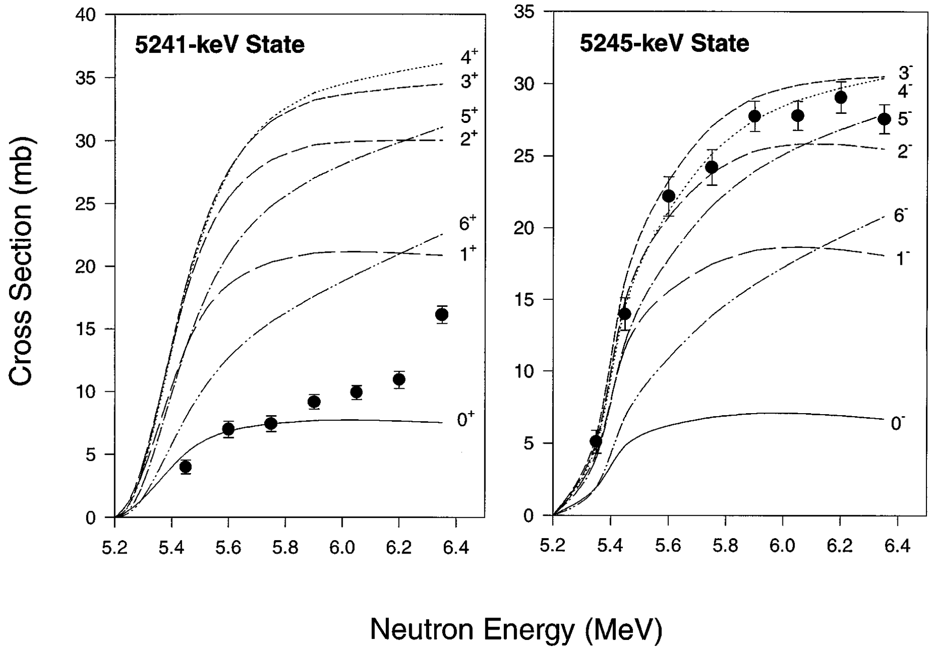

FIG. 3. Excitation functions of the 2626-keV γ ray from the 5241-keV state and the sum of all γ rays from the 5245-keV 3^- state. Comparisons are made with calculated cross sections for various spin possibilities. The increasing cross sections for the 5241-keV state at the highest measured energies are likely the effect of feeding from higher-lying states.

(higher) spin possibilities are extremely unlikely. In short, all of the experimental data are consistent with the conclusion that the 5241-keV state has zero spin, and the 0^- possibility, with an $M3$ transition to the 3^- first excited state, seems remote.

The energy of the known 0^+ state, 5237 ± 2 keV from internal conversion electron measurements [5] of the ground-state transition, is slightly different from the energy of 5241 keV established in the present work; however, a reevaluation of the energy calibration of these internal conversion data [23] indicates that the energy is in good agreement with that obtained in this work. It must be concluded that only one 0^+ state is present and that the 2626-keV γ ray represents the $0^+ \rightarrow 3^-$ transition. When this transition is considered with the 2614-keV $3^- \rightarrow 0^+$ transition, *a cascade of two E3 transitions, a characteristic signature of a two-phonon octupole excitation, is established.* This $E3$-$E3$ cascade is the first observed in a nucleus outside the $N = 82$ region and the first leading directly to the ground state of an even-even nucleus. This observation offers strong support for suggestions [5,16,18], based primarily on energy and population arguments, that the second excited 0^+ state in ^{208}Pb is the 0^+ member of the two-phonon octupole quartet. A search has been conducted for other possible transitions from this state, i.e., an $E1$ transition to the 4842-keV 1^- state or an $E2$ transition to the 4085-keV 2^+ state, but none were found. The absence of other decay branches could be taken as evidence of the collective nature of the $0^+ \rightarrow 3^-$ transition.

Julin *et al.* [5] estimated theoretically that K conversion of the $E0$ transition from the two-phonon octupole 0^+ state to the ground state should represent about 20% of the deexcitation of such a state. Unfortunately, no relationship between the population of the 5241-keV state in their reactions and the $(n, n'\gamma)$ reaction is possible, so a decay branching ratio cannot be established.

It would also be desirable to measure the multipolarity of the 2626-keV transition by detecting internal conversion electrons or internal e^+e^- pairs, but this determination is not possible using the massive scattering sample required for inelastic neutron scattering. Measurements with other reactions and thin targets are being pursued; however, it is not clear that the cross section [5] for population of the 5241-keV state is sufficient and that other technical difficulties can be overcome. The limit of $\tau > 1$ ps for the lifetime of the 5241-keV state established in this work is not inconsistent with the 2626-keV transition being a collective $E3$, but a measurement of the collectivity of the 5241-keV state would, of course, prove very meaningful.

Different theoretical approaches [18,24,25] lead to somewhat differing conclusions about the location of the other members of the two-phonon quartet. The identification of these higher-spin members would provide a measure of the anharmonicity of this vibration and is of fundamental importance, but their characterization is expected to be difficult, since decays by $E1$ transitions will likely predominate. It should also be noted that no γ rays deexciting the 4866-keV 0^+ neutron paring vibrational state were observed in the present work. The identification of the proton pairing vibration in ^{208}Pb is also of great interest. Additional experimental studies to

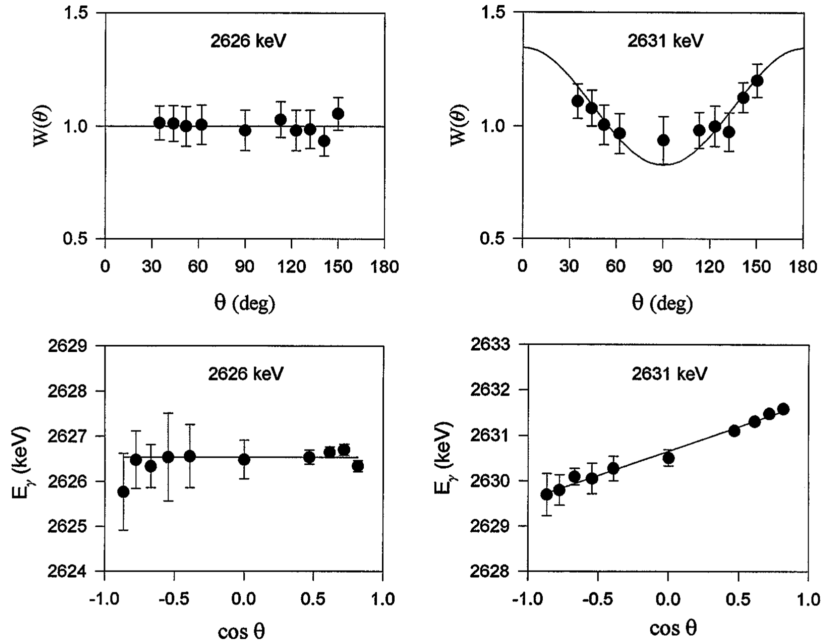

FIG. 4. Gamma-ray angular distributions with Legendre polynamial fits (top) and Doppler shifts (bottom) of the 2626- ($0^+ \rightarrow 3^-$) and 2631- ($3^- \rightarrow 3^-$) keV γ rays. The larger energy uncertainties at backward angles are attributed to difficulties in deconvolution of the close-lying doublets produced by the Doppler shift of the 2631-KeV γ ray to lower energies.

resolve these questions are in progress.

In summary, all of the data for the 2626-keV γ ray are consistent with its assignment as a deexcitins $E3$ transition from the previously known 0^+ state in ^{208}Pb. The resulting cascade of $E3$ transitions (2626- and 2614-keV γ rays) thus represents the most compelling evidence yet presented that the 5241-keV state has a large two-phonon octupole component and evinces the role of the double octupole phonon in ^{208}Pb.

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[1] R. H. Spear, At. Data Nucl. Data Tables **42**, 55 (1989).
[2] S. W. Yates *et al.,* Z. Phys. A **234**, 417 (1986).
[3] S. W. Yates *et al.,* Phys. Rev. C **36**, 2143 (1987).
[4] M. A. J. Mariscotti *et al.,* Nucl. Phys. **A407**, 98 (1983).
[5] R. Julin *et al.,* Phys. Rev. C **36**, 1129 (1987).
[6] H. J. Wollersheim *et al.,* Z. Phys. A **341**, 137 (1992).
[7] M. Schramm *et al.,* Z. Phys. A **344**, 121 (1992).
[8] B. D. Valnion *et al.,* Z. Phys. A **350**, 11 (1994).
[9] C. Fahlander *et al.,* Phys. Scr. **T56**, 243 (1995).
[10] E. F. Moore *et al.,* Nucl. Instrum. Methods Phys. Res., Sect. B **99**, 308 (1995).
[11] P. Kleinheinz *et al.,* Phys. Rev. Lett. **48**, 1457 (1982).
[12] L. Bargioni *et al.,* Phys. Rev. C **51**, R1057 (1995).
[13] S. Lunardi *et al.,* Phys. Rev. Lett. **53**, 1531 (1984).
[14] M. Piiparinen *et al.,* Phys. Rev. Lett. **70**, 150 (1993).
[15] J. H. Bjerregaard, O. Hansen, O. Nathan, and S. Hinds, Nucl. Phys. **89**, 337 (1966).
[16] G. J. Igo, P. D. Barnes, and E. R. Flynn, Phys. Rev. Lett. **24**, 470 (1970); Ann. Phys. (N.Y.) **66**, 60 (1971).
[17] J. Blomqvist, Phys. Lett. **33B**, 541 (1970).
[18] P. Curutchet *et al.,* Phys. Lett. B **208**, 331 (1988).
[19] T. Belgya *et al., Proceedings of the 8th International Symposium on Capture Gamma-ray Spectroscopy and Related Topics, Fribourg, 1993,* edited by J. Kern (World Scientific, Singapore, 1994) p. 878.
[20] Minfang Yeh *et al.* (to be published).
[21] L. I. Govor, A. M. Demidov, and V. A. Kurkin, Izv. Akad. Nauk SSSR Ser. Fiz. **54(3)**, 526 (1990).
[22] M. Schramm, Ph.D. thesis, Hahn-Meitner Institute, Berlin, 1993.
[23] R. Julin (private communication).
[24] J. Dehuang and F. Deji, J. Phys. G **16**, 1685 (1990).
[25] K. Takada and Y. R. Shimizu, Nucl. Phys. **A523**, 354 (1991).