RAPID COMMUNICATIONS

PHYSICAL REVIEW C VOLUME 58, NUMBER 5 NOVEMBER 1998

Fragmentation of the two-phonon octupole vibrational states in ^{208}Pb

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(Received 24 June 1998)

An experiment designed to populate two-phonon vibrational states in ^{208}Pb by Coulomb excitation was performed with a ^{136}Xe beam at a bombarding energy of 650 MeV. The γ rays from the decay of the excited states were measured with Gammasphere and scattered particles were detected in the compact heavy-ion counter CHICO. We have not been able to observe any state close to the expected harmonic energy of 5.2 MeV. However, we were able to extract the $B(E3,3_1^- \rightarrow 6_1^+)$ value for the lowest known 6^+ state at 4.424 MeV based on measured γ-ray intensities. About 20% of the expected total $E3$ strength can be found in this state, suggesting a large fragmentation of this second octupole phonon state in ^{208}Pb. Upper limits for the $B(E3)$ strength were determined for higher-lying, but unseen, 6^+ states ranging from 15% of the harmonic value at 5.2 MeV to 100% at 6.0 MeV. [S0556-2813(98)50411-6]

PACS number(s): 21.10.Re, 23.20.Lv, 27.80.+w

A long standing question in nuclear structure physics concerns the existence and the harmonicity of octupole vibrations in ^{208}Pb. The first excited state of ^{208}Pb is one of the ''classic'' examples of a one-phonon vibration of octupole character. The collective nature of this $I^\pi = 3^-$ state is confirmed by the observed large $B(E3)$ value of 34 W.u. [1]. To further establish the phonon description it is particularly important to identify the two-phonon states and to measure their energies and $B(E3)$ strengths. Spherical and doubly-closed shell nuclei like ^{208}Pb were originally expected to show only small anharmonicities [2]. Since the one-phonon state is located at 2.614 MeV, a harmonic two-phonon state is expected at twice that energy, i.e., $\approx$5.2 MeV. However, a deviation from a pure harmonic vibration and a splitting of the $J^\pi = 0^+$, 2^+, 4^+ and 6^+ members of the two-phonon multiplet has been predicted due (i) to the coupling of the octupole vibration to quadrupole phonons [3], (ii) to particle-hole excitations [4], and/or (iii) to the interaction with pairing vibrations [5–8]. While the various theoretical calculations predict different splittings of the two-phonon multiplet, they all agree that the anharmonic effects should be small and of the order of 200 keV or less. Only recently, the first promising experimental evidence for the existence of the 0^+ member of the two-phonon vibration has been reported using a $(n,n'\gamma)$ reaction [9]. The measured γ-ray energy of 2626 keV $(E_{0_2^+}/E_{3_1^-} = 2.005)$ indicates only a slight deviation from a harmonic oscillation. The assignment of this state is based on the measured excitation function, angular distribution, and measured Doppler shift, which is consistent with a lifetime longer than 1 ps. This state had previously been observed in a (p,t) reaction [10] as well as in (p,p') and in (d,p) reactions [11], but no definitive assignment could be made in those cases. The most crucial quantity for the identification of any two-phonon octupole vibrational state, the $B(E3,0_2^+ \rightarrow 3_1^-)$ transition probability, has yet to be measured. The aforementioned $(n,n'\gamma)$ reaction also contains evidence for candidates for the 2^+ and the 4^+ two-phonon octupole excitations [12]. Indeed, the energies of the 2^+ state at 5286 keV $(E_{2^+}/E_{3^-} = 2.022)$ and the 4^+ state at 5216 keV $(E_{4^+}/E_{3^-} = 1.995)$ are very close to the expected harmonic values. While the lifetimes of these states have been measured, they are predominantly determined by large $E1$ decay rates, which are consistent with those expected for the decay of octupole phonon states, but by no means represent an unambiguous identification.

The lifetime is a unique measure of the octupole collectivity only for the decay of the 0^+ state. By contrast, Coulomb excitation is sensitive to all the $B(E3,3^- \rightarrow 0^+,2^+,4^+,6^+)$ values. In Coulomb excitation, the 6^+ state of the multiplet is expected to be the most strongly populated. To date, no heavy-ion induced experiment designed to measure the γ decay following the possible excitation of the second octupole phonon in ^{208}Pb by Coulomb and/or nuclear interaction has been able to observe any reliable candidates. Most of the attempts [13–16] were performed at bombarding energies above the Coulomb barrier, where the nuclear interaction is expected to increase strongly the population probability of the double phonon members [13]. However, due to the uncertainties in calculating the population of the phonon states via deep-inelastic reactions, it is extremely difficult if not impossible to extract sensitive limits for the existence of phonon states [17].

Following upon the results of our previous work [17], this paper reports on an experiment to investigate the two-

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phonon octupole vibrations in ^{208}Pb at a bombarding energy below the Coulomb barrier, where Coulomb excitation dominates. Using the high sensitivity of the 4π-γ-ray spectrometer Gammasphere [18] we planned to search for indications of 6^+ states close to 5.2 MeV. The knowledge of the mechanism of Coulomb excitation enables us to extract $E3$ matrix elements if we observe these states and if not, it allows us to place sensitive limits on their occurrence. It also allows us to determine matrix elements for other states whose population is sufficiently strong that the intensities of their decay can be extracted. The measurement was performed using a thin target (0.9 mg/cm^2) of ^{208}Pb (99.86% enriched) bombarded by a beam of ^{136}Xe delivered by the 88-Inch Cyclotron of LBNL, at an energy of 650 MeV, e.g., 7% below the Coulomb barrier [19]. The γ rays were detected by Gammasphere consisting, at that time, of 93 large-volume Compton-suppressed Ge detectors; in this configuration the photo-peak efficiency is estimated to be 9% at $E_\gamma = 1.0$ MeV and 5% at $E_\gamma = 2.6$ MeV. The choice of a bombarding energy of 650 MeV instead of the "safe" energy of 550 MeV [19] is based on the fact that the excitation cross section of the 6^+ member, for instance, can thereby be increased by an order of magnitude due to the high excitation steps of ≈ 2.6 MeV involved. The newly constructed two-dimensional position-sensitive, parallel-plate avalanche counter CHICO [20] was used to select binary collisions, to correct for the Doppler-shift of the γ rays emitted in-flight, and to determine excitation probabilities as a function of the scattering angle. CHICO covers the angular range $12° \leqslant \vartheta_{\mathrm{lab}} \leqslant 85°$ and $95° \leqslant \vartheta_{\mathrm{lab}} \leqslant 168°$ and 280° in φ_{lab} for a total of 2.7π coverage in solid angle. The forward 20° of the scattering angle were shielded to exclude elastic scattering processes. The angular resolution in ϑ_{lab} is 1° and in φ_{lab} is 9°. Although the beam energy is above the "safe" energy, for forward scattering ($\vartheta_{\mathrm{lab}} \leqslant 85°$) the distance of closest approach is nonetheless large enough to preclude nuclear interactions. For this reaction, forward scattering of the projectile implies the observation of both particles (p) in CHICO and enables the determination of the masses by the relative angles and the time of flight difference of both particles. A mass resolution of about 10% allowed us to uniquely identify the projectilelike and the targetlike reaction products for forward-angle scattering, while particle identification was unambiguous for back scattering.

During the experiment about 8.5×10^7 p-p-γ and 5.8×10^6 p-p-γ-γ events were collected. The hevimet absorbers, normally installed in front of the BGO shields, were removed to allow the measurement of the γ-ray multiplicity and sum energy. The data were sorted into 15 scattering-angle-dependent γ-ray histograms and γ-γ coincidence matrices, Doppler corrected for target-like particles. Each of the scattering-angle regions covered $\Delta\vartheta_{\mathrm{c.m.}} = 10°$. Furthermore, six γ-γ matrices were incremented for the sum of all scattering angles and events restricted to forward scattering, using projectile-projectile, target-target, and projectile-target Doppler corrections. In this type of experiment, involving low γ-ray multiplicity and relatively high γ-ray energies, we had to take into account Compton scattering between Ge crystals by adding the energies of adjacent Ge detectors. This neighbor-add procedure is particularly important when searching for weak transitions by gating on γ-ray energies below the Compton edge of strong γ-ray transitions.[1]

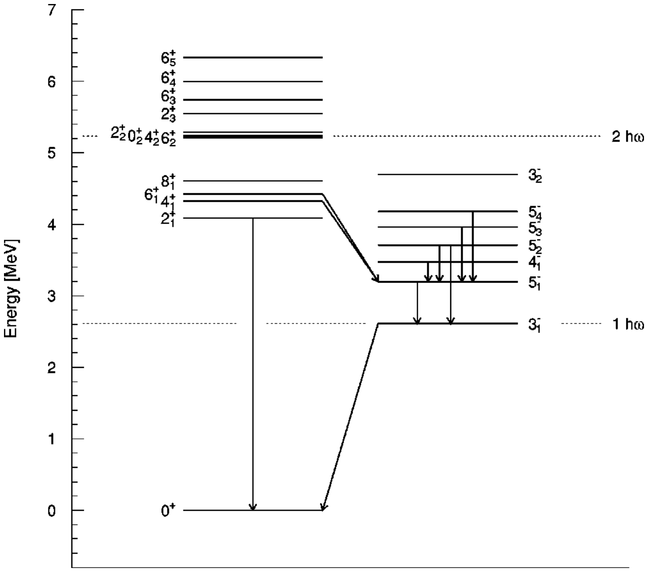

FIG. 1. Partial level scheme of ^{208}Pb used in the Coulomb excitation calculations. The arrows indicate the observed transitions. The dotted lines mark the expected excitation energies for a pure harmonic vibration.

To date, experimental evidence exists for five 6^+ states in ^{208}Pb. They are located at excitation energies of 4.424 MeV [1], 5.213 MeV [1,21], 5.738 MeV [22], 5.993 MeV [1,21], and 6.332 MeV [1]. Figure 1 presents a partial level scheme including all the states which were taken into account in the Coulomb excitation calculations discussed below. Two $E1$ decays of the 4.424 MeV state to the two lowest-lying 5^- levels have been observed whereby the decay to the 3.198 MeV state is one order of magnitude stronger than the decay to 5_2^- level at 3.708 MeV [21]. One $E1$ transition to the lowest 5^- state and one $E2$ transition to the 6_1^+ level at 4.424 MeV has been measured from the 5.213 MeV state. For the 5.738 MeV level only the $E2$ decay to the 4.424 MeV state has been observed, while four different decay pathways are known for the 5.993 MeV 6_4^+ state [21]. We note here that none of the 6^+ assignments above 5 MeV seems well established. The assignment of the 5.213 MeV state is uncertain because a doublet has been suggested at this excitation energy [12]. The 6_3^+ state was measured only once without further explanation supporting the proposed spin assignment [22]. The 6_4^+ assignment at 5.993 MeV was originally only tentative [23] and the measured decay branches in [21] cast some doubts on the 6^+ assignment. Finally, the 6_5^+ state at 6.332 MeV has hardly been mentioned anywhere. Despite these uncertainties in the assignments, the decay branches of the first three 6^+ states to low-lying 5^- and 6^+ states are in agreement with expectations and measurements, e.g., in neighboring nuclei [12]. Whether

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[1] In ^{208}Pb, the cross scattering of the 2.614 MeV γ ray through the individual BGO anti-Compton shields generates background around 2 MeV which overlaps with the predicted energy of one of the strong decays from the 6^+ double-octupole phonon state.

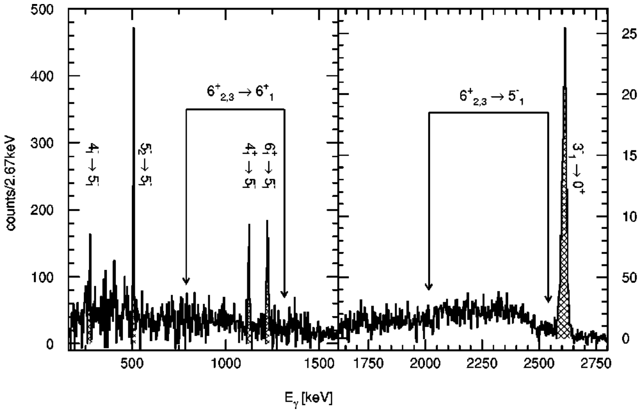

FIG. 2. Different regions of a γ-ray energy spectrum for a targetlike Doppler-shift correction after gating on the $5^- \rightarrow 3^-$ transition at 584 keV. The arrows indicate the expected energies for transitions related to the next two higher-lying 6^+ states (see text).

the $E1$ decays or the $E2$ decays dominate will strongly depend on the hindrance for the $E1$ decays. Based on this discussion we will assume in the estimations below that the decays to the lowest-lying 5^- state and to the lowest-lying 6^+ state are the strongest pathways and we will use the observational limits for these transitions to determine limits for the $B(E3)$ values of the 6^+ member of the two-phonon quadruplet. However, also weaker decay branches like the transition between the 5_2^- level at 3.708 MeV and the 6_1^+ level at 4.424 MeV have also been taken into account according to measured branching ratios.

Figure 2 shows two regions of a spectrum in ^{208}Pb, generated by gating on the $5^- \rightarrow 3^-$ transition at 584 keV, which is supposed to be part of the first dominant decay path for the 6^+ as well as for the 4^+ member of the two-phonon multiplet. The spectrum is summed over all scattering angles. Besides the known 278 keV, 507 keV, 1124 keV, 1225 keV, and 2614 keV transitions, no additional transition could be identified from this coincidence gate. The arrows indicate expected energies from previously suggested 6^+ states at 5.213 MeV and 5.738 MeV. From this spectrum we are able to obtain intensity limits for previously observed transitions and, more generally, for any γ ray corresponding to the decay of a state at any excitation energy assuming similar decay paths as described above. The following estimations of observational limits are based on intensities corresponding to 2 standard deviations above background. These limits can then be used to set limits for the $B(E3,3_1^- \rightarrow 6^+)$ values for 6^+ states at the assumed excitation energies.

Sufficient statistics have been collected for seven transitions to obtain the scattering angle dependence in the Coulomb excitation region of forward scattering. The Coulomb excitation calculations have been performed with the computer code GOSIA developed at the University of Rochester [24], which also calculates γ-ray intensities taking into account matrix elements, conversion coefficients, branching ratios, and lifetimes. Figure 3 shows the experimental angular distribution normalized to the $3_1^- \rightarrow 0^+$ transition as obtained from either γ-ray singles or coincidence measurements. While the $B(E3,0^+ \rightarrow 3_1^-)$, $B(E2,0^+ \rightarrow 2_1^+)$, and $B(E2,3_1^- \rightarrow 5_1^-)$ values are known, the $B(E\lambda)$ values for the other transitions under consideration have been determined from the fits to the experimental data. A full fit of all matrix elements was not possible because the available data are insufficient to overdetermine the many unknown matrix elements. The quoted uncertainties in the matrix elements were determined by varying their magnitude. The solid lines show the results of the GOSIA calculations and, as a reference, the dashed lines indicate the expected yields for a ''safe'' bombarding energy of 550 MeV using the same matrix elements. There is good agreement between the experimental data and the calculations for distant collisions, suggesting, that indeed, Coulomb excitation is still dominating at these angles but with an increase in cross section of about one order of magnitude with respect to the ''safe'' energy. The enhancement for the higher spin states and the suppression for the 2^+ state for closer collisions can be understood in terms of the nuclear interaction, which is expected to set in at larger scattering angles. As shown in Ref. [13], excitations due to the nuclear interaction do not depend on the transferred spin or excitation energy and are also not as sensitive to collective modes as Coulomb excitation. Therefore, we expect a relatively stronger excitation for higher multipolarities. Since the data are normalized to the 3^- decay, the 2^+ decay intensity is reduced while decays of states with higher spin are enhanced.

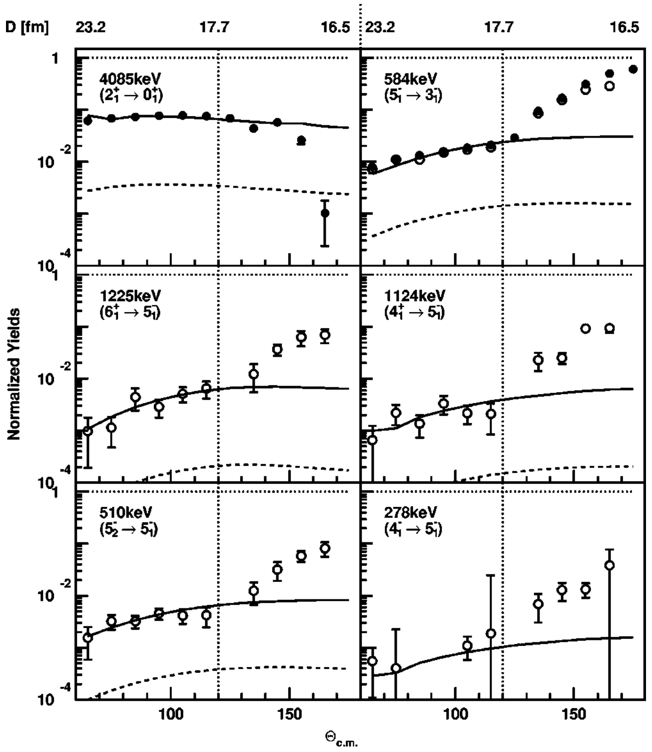

FIG. 3. Measured and calculated γ-ray yields normalized to the $3^- \rightarrow 0^+$ transition at 2614 keV as a function of the projectile scattering angle. The solid circles are measured γ-ray single yields, the open circles are observed γ-γ coincidence yields. The full lines are the results of calculations with GOSIA at a bombarding energy of 650 MeV. The dashed lines are results of calculations using a ''safe'' bombarding energy of 550 MeV. The vertical dotted lines indicate the separation between the Coulomb excitation region and the region of more deep-inelastic processes, including nuclear interactions.

It is not possible to extract unique $E2$ or $E3$ matrix elements for the excitation of the 4_1^+ state at 4.323 MeV. This state can be excited by two $E2$ steps as well as by two $E3$ steps and the two paths interfere. This can be seen by changing the sign of the $E3$ matrix element and recognizing the change in the excitation probability of the 4^+ state. Despite this fact, we can estimate upper limits of $M(E2)=0.24e$ b for the $2^+\rightarrow 4^+$ and $M(E3)=-1.1\ e$ b$^{3/2}$ for the $3^-\rightarrow 4^+$ excitation. The limit of the $E3$ matrix element accounts for about 77% of the expected $E3$ strength in the 4^+ member of the two-phonon multiplet. For the excitation of the 6_1^+ state at 4.424 MeV, we expect three $E2$ steps to be much weaker than two $E3$ steps since no interference is observed when changing the sign. First of all, assuming a direct excitation of the 6_1^+ state at 4.424 MeV we can extract an $E3$ matrix element of $M(E3)=-0.80\pm 0.10\ e$ b$^{3/2}$. However, since we cannot exclude the presence of γ decays from higher lying 6^+ states into the 4.424 MeV state, this value represents an upper limit. We can also determine the matrix element by taking into account the population, and subsequent decay branches of previously suggested 6^+ states. For instance, taking the 6_2^+ state at 5.213 MeV into account, using the quoted branching ratio for the 789 keV and 2015 keV transitions [21] and taking the observational limit for the 2015 keV transition (which is more sensitive than the 789 keV transition) $M(E3)=-0.70\pm 0.10\ e$ b$^{3/2}$ is now obtained for the 6_1^+ state. In turn, a value of $M(E3)=-0.64\pm 0.09\ e$ b$^{3/2}$ was extracted if only the 6_3^+ level at 5.738 MeV is considered. Finally, taking both states at 5.213 MeV and 5.738 MeV into account and assuming the same branching ratios as before, a matrix element of $-0.60\pm 0.09\ e$ b$^{3/2}$ remains for the excitation of the 6_1^+ state. The additional consideration of the higher lying 6^+ states with their corresponding intensity limits would exceed by far the harmonic value of the expected $E3$ value (e.g., the sum of the extracted $E3$ strengths for the latter case including three 6^+ states accounts for about 85% of the total harmonic strength). This result reflects the lacking sensitivity to the higher-lying states in the performed experiment. Further details about the Coulomb excitation calculations will be presented in a forthcoming report. The mentioned values for the $E3$ matrix element of the lowest 6_1^+ state at 4.424 MeV account for 28.2±6.6% [$M(E3)=-0.80\pm 0.10\ e$ b$^{3/2}$], 21.6±5.9% [$M(E3)=-0.70\pm 0.10\ e$ b$^{3/2}$], 18.0±4.8% [$M(E3)=-0.65\pm 0.09\ e$ b$^{3/2}$], or 15.9±4.4% [$M(E3)=-0.60\pm 0.09\ e$ b$^{3/2}$] of the expected strength of the harmonic value which is 1.507 e b$^{3/2}$ [2]. This result is surprising since it implies that about 20% of the expected strength can be found 800 keV below the anticipated harmonic energy, a result which has not been anticipated by theory [4]. The only calculation which predicts a shift of the 6^+ state to lower energies is based on the measured intrinsic quadrupole moment ($Q=0.34\pm 0.10\ e$ b^2) [3] of the one-phonon state. However, even a value of $Q=0.44\ e$ b^2 can only account for a maximum shift of about 200 keV. The 6_1^+ state has been interpreted originally as a mixture of a $(\nu g_{9/2})(\nu i_{13/2})^{-1}$ and a $(\pi h_{9/2})(\pi h_{11/2})^{-1}$ particle-hole excitation [1,21]. The inspection of the systematic given in [21] reveals the interesting fact that this state lies lower than the expected energy. This observation is also true for the 4_1^+ state. It is possible then that this particle-hole state mixes with the two-phonon state and is shifted down in energy while picking up part of the phonon $E3$ strength. It should be stressed that with the information obtained in this experiment a lower limit value of zero for the $B(E3)$ value cannot be ruled out. This is due to the fact that many 6^+ states could exist between 5 MeV and 6 MeV, each only contributing a small fraction of the total $E3$ strength, and feeding the observed 6^+ state with intensities smaller than the observational limits. While in principle possible, this scenario seems unrealistic given the expected density of 6^+ states in this excitation energy region.

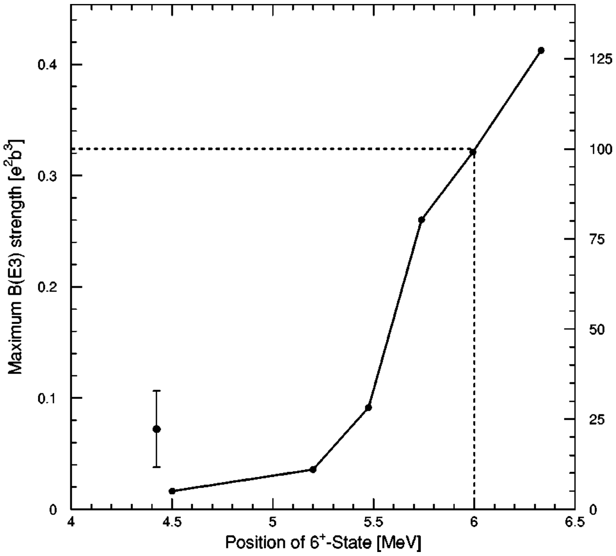

FIG. 4. Limits on the $B(E3)$ values of 6^+ states in ^{208}Pb obtained by combining experimentally observed intensity limits and GOSIA calculations as a function of the excitation energy of the 6^+ state. The strength limits are also given relative to theoretical values expected for a pure harmonic octupole vibration. In addition, the $B(E3)$ value obtained for the only observed 6_1^+ state at 4.424 MeV is shown. The given error follows from the assumption that the maximum 6_1^+ decay intensity is determined only by the direct population of this state, and the minimum by considering the feeding according to the observational limits from the next two higher 6^+ states (see text).

Finally, assuming only one other 6^+ state above 4.424 MeV, the intensity limits imply a maximum $E3$ component. Again, we took two decay pathways to obtain observational limits into account, those corresponding to the $6^+\rightarrow 6_1^+$ and $6^+\rightarrow 5_1^-$ transitions. Weaker decay branches to higher-lying 5^- and 6^+ states have been implicitly taken into account by using the same matrix elements which have been determined for the lower-lying states. We must emphasize at this point that in the unlikely scenario of a strong decay to a different state which we have not considered here could potentially increase the upper limit. Figure 4 summarizes the extracted $E3$-strength limits relative to the theoretical value expected for a pure two-phonon harmonic oscillation as a function of the energy of the assumed 6^+ state. The energy dependence reflects the Coulomb excitation probability of a 6^+ state: just above 5.2 MeV, the adiabatic cutoff for this excitation process sets in and, accordingly, our sensitivity limit decreases. The kink at 5.738 MeV is due to the higher background at 1314 keV (the $6_3^+\rightarrow 6_1^+$ transition) which is close to the very strong $2^+ \rightarrow 0^+$ transition in ^{136}Xe. For an excitation energy of 5.2 MeV the sensitivity limit translates into a maximum $E3$ strength of 15% of the harmonic octupole vibration. This result, when combined with the finding of about 20% of the expected strength at 4.424 MeV, suggests a possible fragmentation of the two-phonon $E3$ strength by mixing with particle-hole states occurring in the range of 5–7 MeV.

In summary, we have performed an experiment to search for two-phonon octupole vibrational states in ^{208}Pb by Coulomb exciting ^{208}Pb with a ^{136}Xe beam at an energy of 650 MeV. Scattered particles were detected by the two-dimensional position-sensitive CHICO array and γ rays were measured with Gammasphere. The experiment did not identify any new state around 5.2 MeV, the energy where the two-phonon members are expected to be located for a harmonic vibration, nor did we observe any previously known 6^+ state of higher excitation energy. However, the lowest lying 6^+ state at 4.424 MeV was populated with a $B(E3)$ value that accounts for about 20% of the harmonic value. Moreover, we have been able to obtain limits for $E3$ matrix elements of states close to the harmonic energy. Based on this analysis we conclude that any 6^+ state at an energy of about 5.2 MeV will have an $E3$ matrix element with less than 15% of the harmonic $E3$ strength. Although the 4_1^+ state at 4.323 MeV has been observed with an intensity comparable to that of the 6_1^+ state at 4.424 MeV, it is not possible to extract the $E2$ nor $E3$ matrix elements for this state, since these excitation pathways interfere with each other. Possibly, the observed distribution of the $E3$ strength in 6^+ states points to a large fragmentation of the octupole vibrational strength of the two-phonon state by mixing with particle-hole states.

We acknowledge fruitful discussions with S. W. Yates. We also wish to thank the accelerator staff for the excellent experiment conditions. One of us (K.V.) is indebted to the Deutsche Forschungsgemeinschaft for financial support. This work was supported by the Department of Energy under Contract no. DE-AC03-76SF00098 (LBNL) and W-31-109-ENG-38 (ANL).

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