Pygmy and core polarization dipole modes in $^{206}$Pb: Connecting nuclear structure to stellar nucleosynthesis

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Abstract

A high-resolution study of the electromagnetic response of $^{206}$Pb below the neutron separation energy is performed using a ($\gamma$, $\gamma'$) experiment at the HILiS facility. Nuclear resonance fluorescence with 100% linearly polarized photon beams is used to measure spins, parities, branching ratios, and decay widths of excited states in $^{206}$Pb from 4.9 to 8.1 MeV. The extracted $\Sigma B(E1) \uparrow$ and $\Sigma B(M1) \uparrow$ values for the total electric and magnetic dipole strength below the neutron separation energy are 0.9 ± 0.2 e² fm² and 8.3 ± 2.0 $\mu_n^2$, respectively. These measurements are found to be in very good agreement with the predictions from an energy-density functional (EDF) plus quasiparticle phonon model (QPM). A detailed theoretical analysis allows to separate the pygmy dipole resonance from both the tail of the giant dipole resonance and multi-phonon excitations. Combined with earlier photonuclear experiments above the neutron separation energy, one extracts a value for the electric dipole polarizability of $^{206}$Pb of $\sigma_0 = 122 ± 10$ mb/MeV. When compared to predictions from both the EDF-QPM and accurately calibrated relativistic EDFs, one deduces a range for the neutron-skin thickness of $R_{skn} = 0.12–0.19$ fm and a corresponding range for the slope of the symmetry energy of $L = 48–60$ MeV. This newly obtained information is also used to estimate the Maxwellian-averaged radiative cross section $^{206}$Pb($\gamma$, $\gamma'$)$^{206}$Pb at 30 keV to be $\sigma = 130 ± 25$ mb. The astrophysical impact of this measurement—both on the s-process in stellar nucleosynthesis and on the equation of state of neutron-rich matter—is discussed.

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The commissioning of both powerful telescopes and sophisticated terrestrial facilities has resulted in a strong synergy between nuclear physics and astrophysics. Where do the chemical elements come from? and what is the nature of matter at extreme densities? are a few of the fundamental questions animating nuclear astrophysics today. However, progress in answering these questions relies on our understanding of nuclear structure far away from the valley of stability where the neutron–proton asymmetry is large and our knowledge is poor. The dynamics of such exotic systems is imprinted in the nuclear symmetry energy:

$$ S(\rho) = \frac{1}{2} \left( \frac{\partial^2 E(\rho, \delta)}{\partial \delta^2} \right)_{\delta=0} \approx E(\rho, \delta = 1) - E(\rho, \delta = 0). $$

Here $E(\rho, \delta)$ is the energy per nucleon as a function of the total baryon density $\rho$ and the neutron–proton asymmetry $\delta = \rho - n$. 

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\( \frac{(N - Z)}{A} \). Note that to a very good approximation the symmetry energy represents the energy cost of turning symmetric nuclear matter (\( \delta = 0 \)) into pure neutron matter (\( \delta = 1 \)). In particular, the density dependence of the symmetry energy around nuclear saturation density (\( \rho_0 \)) is encoded in a few bulk parameters [1]:

\[
S(\rho) = J + Lx + \frac{1}{2}K_{sym}x^2 + \ldots \quad \text{with} \quad x = \frac{\rho - \rho_0}{3\rho_0}.
\]  

An observable that is particularly sensitive to the density dependence of the symmetry energy is the neutron–skin thickness of a heavy nucleus, defined as the difference between the neutron and proton rms radii \( R_{skin} = R_n - R_p \). In particular, the neutron skin of \(^{208}\text{Pb}\) is strongly correlated to the slope of the symmetry energy \( L \). The Lead Radius Experiment (PREX) has provided the first model independent evidence in favor of a neutron-rich skin in \(^{208}\text{Pb}\) [4,5]. Unfortunately, unforeseen technical issues compromised the statistical accuracy of the measurement. And whereas a follow-up experiment (PREX-II) is envisioned to reach the original sensitivity, a complementary observable has been identified that is also strongly correlated to the slope of the symmetry energy: the electric dipole polarizability \( \alpha_D \) [6,7].

The Pygmy Dipole Resonance (PDR)—the emergence of low-energy dipole strength with neutron excess—has motivated a great deal of experimental and theoretical effort [8]. Observed as a concentration of electromagnetic strength overlapping the low-energy tail of the Giant Dipole Resonance (GDR), the PDR has been identified in a broad range of neutron-rich systems, ranging from light [9,10], to transitional [11,12], up to heavy nuclei [7,13–15] far away from stability. In this regard the PDR—and perhaps higher-multipolarity modes [16–18]—may provide useful constraints on such fundamental properties as the neutron–skin thickness of medium to heavy nuclei [7,19,20], the nuclear symmetry energy [10,21], and the properties of neutron stars [22].

The heavy \(^{206}\text{Pb}\) nucleus containing 42 excess neutrons is expected to have a robust neutron skin, and as a consequence, should exhibit an appreciable amount of low-energy dipole strength. “A systematic study of the nuclear dipole strength in the lead isotopes, including \(^{206}\text{Pb}\), is presented in Ref. [14] for excitation energies up to 6.5 MeV.” At excitation energies below the neutron separation energy (\( S_n = 8.087 \) MeV) the PDR coexists with a variety of modes, such as the tail of the GDR, magnetic dipole transitions, and multi-phonon excitations [23–26]. So far, isolating the low-energy photoabsorption spectra from these other contributions has proved elusive [27–29]. As a result and despite significant experimental and theoretical effort, see e.g. [29], a direct model-independent determination of the neutron–skin thickness from the PDR is not yet possible [8]. However, rather than concentrating on the fraction of the PDR that contributes to the cross section, a more robust observable is the “inverse-squared” energy weighted sum \( \langle \sigma_2 \rangle \). That is [30],

\[
\alpha_D = \frac{1}{2\pi^2 \alpha} \int_{0}^{\infty} \sigma_0(E) \frac{dE}{E^2} = \frac{\sigma_{2-}}{2\pi^2 \alpha} = 6.942 \sigma_{2-},
\]  

where \( \sigma_{2-} \) is directly proportional to the electric dipole polarizability \( \sigma_0 \) (both given in mb/MeV) and \( \alpha \) is the fine-structure constant.

As far as the impact of the present measurement on stellar nucleosynthesis is concerned, the PDR in \(^{206}\text{Pb}\) might affect the \(^{205}\text{Pb}\) radiative neutron capture cross section, a reaction of relevance to the destruction of \(^{205}\text{Pb}\) during the s-process [26,31]. The now extinct radionuclide \(^{205}\text{Pb}\) with a half-life of 15.3 Myr may be of significant cosmochemical interest due to its pure s-process nature [32]. Further, it could provide key information on the formation of the solar system, particularly on the time span between the last s-process nucleosynthetic events that have modified the composition of the solar nebula and the formation of the solid bodies in the solar system. The presence of \(^{205}\text{Pb}\) in the early solar system was demonstrated recently and it was suggested that the \(^{205}\text{Pb}\) \(^{205}\text{Tl}\) pair is well suited for chronological studies, complementing information provided by other extinct short-lived radionuclides [33,34].

The abundance of \(^{205}\text{Pb}\) in the early solar system inferred from carbonaceous chondrites data can also be used to assess whether asymptotic giant branch (AGB) stars and massive Wolf–Rayet stars are the most likely sites of the s-process [35,36]. However, this investigation remains sensitive to the amount of freshly produced s-process material, including the survival of \(^{205}\text{Pb}\) in AGB stars [37] through both neutron capture and weak interaction processes. Due to its astrophysical importance, the Maxwellian-averaged capture section cross section in \(^{205}\text{Pb}\) needs to be known accurately, and if not measurable directly, at least it should be experimentally constrained.

In the present study we were able to decompose the multipolarity and the decay pattern of the PDR and the GDR in \(^{206}\text{Pb}\) below the neutron separation energy. We studied the structure of \(^{206}\text{Pb}\) using monoenergetic and 100% linearly polarized photon beams from the High-Intensity Gamma-ray Source (HI\(\gamma\)S) facility [38]. Nuclear resonance fluorescence (NRF) measurements were performed in the energy range from 4.9 to 8.1 MeV. The sample consisted of 4 g of metal powder enriched to 99.3% in \(^{206}\text{Pb}\), and contained in a thin-walled and ultra-pure quartz ampule with a diameter of 0.9 cm. Possible contributions of resonance states from the \(\text{SiO}_2\) ampule were searched for and found to be negligible, in agreement with literature data [39]. At each energy, an average photon flux of \(10^5\) s\(^{-1}\) bombardied the \(^{206}\text{Pb}\) target for approximately four hours. Six HPGe detectors (four with 60% and two with 25% relative efficiency to a standard 7.62 cm \(\times\) 7.62 cm NaI detector) were used to measure the \(\gamma\) rays emitted from the NRF process. Two detectors were arranged in the horizontal plane at 90° to the incident beam, two were at 90° in the vertical plane, while the remaining two detectors were placed at 135° in the horizontal plane. The polarization plane of the beam was the horizontal plane. The energy distribution of the photon beams was measured using a 123% efficiency HPGe detector placed a short distance behind the target position. During beam profile measurements, the beam was attenuated by a series of copper absorbers.

![Fig. 1.](image)
mounted upstream to avoid pileup and long dead times. A portion of the spectra of NRF γ rays from a photon beam with centroid energy of 5.750 MeV is displayed in Fig. 1 showing unambiguously a clear distinction between E1, M1, and E2 multipolarities [27,40]. More detailed information about the NRF technique used at the HfJ/S facility may be found in Refs. [12,28,41–43].

The distribution of electric and magnetic dipole states in 206Pb from 4.9 to 8.1 MeV is shown in Fig. 2. A total of one hundred 1− states were observed at low energies, with most of them located at $E_γ > 6.5$ MeV where the level density is very high. Our results for the electric dipole strength are consistent with the presented values in Refs. [14,44]. However, there are many dipole states missing in the last two references due to the low sensitivity associated with bremsstrahlung beams. In turn, twenty-six M1 states were identified with the strength localized largely in two regions around 6 and 7.5 MeV, assumed to be associated with a spin-flip resonance [27,28,45]. However, it is clear that in this energy region the dipole response is predominantly electric in nature. The vast majority of these transitions decay directly to the ground state.

Needless to say, a proper description of the rich and complex experimental spectrum depicted in Fig. 2 requires a highly sophisticated theoretical treatment. Thus, displayed in Fig. 2 is a detailed comparison of the experimentally observed E1 and M1 strength distributions against theoretical predictions from three-phonon EDF+QPM calculations [23,24]. The faithful reproduction of the experimental data is fully consistent with earlier EDF+QPM calculations that successfully reproduced the experimental photoneutron cross sections—albeit in the GDR region—in 206Pb, 207Pb, and 208Pb [46].

We emphasize that in contrast to the QPM calculations of Ref. [14], our EDF+QPM calculations are performed with single-particle energies obtained in a self-consistent manner from our EDF approach based on fully self-consistent Hartree–Fock–Bogoliubov (HFB) calculations. In this sense our QPM calculations are considerably more elaborate than those of Ref. [14]. They account for nuclear ground-state properties like binding energies, root-mean-square radii and the difference between them [24,26]. This is found to be very important for the reliable description of low-energy dipole and higher-multipole pygmy resonances, which are strongly connected to the neutron skin thickness. More details on the comparison between our and other theoretical methods are given in Refs. [27,47].

Following our previous EDF+QPM calculations [27,28,40], the M1 transitions in 206Pb are calculated with a quenched effective spin-magnetic factor $g^\text{eff} = 0.75 g^{\text{bare}}$ where the bare spin-magnetic moment $g^{\text{bare}}$ has been adopted. Similar to our findings in the $N = 50$, $\text{R}_2$ isotones [27,40] and in $\text{S}_2\text{Cr}$ [28], the M1 strength below the neutron separation energy is dominated by spin-flip excitations. A small admixture of about 7% from orbital contributions is obtained.

The calculated EDF+QRPA 1− excitations in the $E_γ ≤ 6.8$ MeV region in 206Pb are dominated by almost pure neutron, weakly bound two-quasiparticle configurations which may be associated with neutron skin vibrations. The predicted structure of the states strongly resembles our previous findings on PDR systems obtained in a large variety of atomic nuclei [8,23,24,27,28,40,43]. Similar conclusions can be also drawn by means of detailed studies of transition densities, indicating a skin mode with dominantly neutron oscillations at the nuclear surface as is characteristic of the PDR. With the increase of the excitation energy ($E_γ ≥ 6.8$ MeV) the isovector contribution to the E1 strength also increases as indicated by the out-of-phase relations of the proton and neutron contributions to the QRPA states [24,43]. Moreover, the EDF+QRPA calculations of 206Pb demonstrate that in the vicinity of the neutron threshold and above the theoretical dipole strength function fits a Lorentzian shape, as is generally assumed for the GDR [45]. Thus, the combined analysis of the structure, transition densities, and energy-weighted sum rules of the 1− states provides a clean separation of the PDR and GDR excitation within the QRPA framework. The QPM model basis is built of QRPA phonons with spin and parity $J^p = 1^+, 2^+, 3^−, 4^+, 5^−$. The model Hamiltonian is diagonalized on an orthonormal set of wave functions constructed as a superposition of one-, two- and three-phonon components [23,24]. The theoretical method allows for sufficiently large configuration spaces such that a unified description of one-phonon – PDR and GDR and multi-phonon states is feasible. Finally, from the individual transitions one may also compute experimental and theoretical cumulative (or integrated) strengths below the neutron separation energy in order to visually assess the spectral contribution to the overall strength; see Fig. 3. As it is seen from the figure, the EDF+QPM calculations strongly suggest that the PDR dominates the distribution of E1 strength up to about 7 MeV, at which point the tail of the GDR starts making an important contribution. Overall, the PDR and the GDR account for about 77% and 12% of the E1 strength below the neutron separation energy, respectively. Below 8.1 MeV there is significant impact from multi-phonon states to the total E1 strength and to a lesser extent to the M1 strength: ≈31% and ≈3%, respectively. For these theoretical estimates interference terms have been neglected but they are, of course, taken into account in the full calculations. A detailed description of the elaborate multi-quasiparticle and multi-phonon approach has proven successful in various scenarios as well as ad-

### Table 1

Summary of the E1 and M1 strengths in 206Pb.

| Parameter | Present data | EDF+QPM |
|-----------|--------------|---------|
| Energy interval (MeV) | 4.9–8.1 | 4.9–8.1 |
| Number of E1 states: | | |
| Within the exp. sensitivitya | 100$^a$ | 94 |
| Total | 340 | |
| Number of M1 states: | | |
| Within the exp. sensitivityb | 26$^b$ | 28 |
| Total | 170 | |
| $\Sigma B(M1) \uparrow (\mu_N^2)$ | 8.3 ± 2.0 | 8.9 |

$^a$ The sensitivity limit for a single E1 transition is $\sim 5 \times 10^{-4}$ e² fm$^2$.

$^b$ The sensitivity limit for a single M1 transition is $\sim 5 \times 10^{-2}$ $\mu_N^2$. |
ditional improvements to the model for heavy nuclei displaying a high level density near threshold are beyond the scope of this letter. For a detailed discussion of the technical aspects of the model see Refs. [16–18,23,24,27,40,43,48]. The comparison of the theoretical results for low-energy electric and magnetic spectral distributions and cumulative strengths in $^{208}$Pb presented here with calculations given in Ref. [14] clearly reveals that the EDF+QPM approach provides a better description of the experimental data.

Having fully characterized the distribution of low-energy dipole strength in $^{208}$Pb, we now return to the main motivation behind this letter: what is the impact of this nuclear experiment on astrophysics? We start by assessing the impact of this measurement on the density dependence of the symmetry energy. Although the $E^{-2}$ weighting in Eq. (3) makes the low-energy component of the dipole response of paramount importance to the evaluation of $\alpha_D$ (or $\sigma_{-2}$), knowledge of the dipole response above the neutron separation energy is also required. Thus, to complement the present low-energy experiment we rely on the measurements of the photoabsorption cross section for $E_\gamma > S_{\text{In}}$ reported in Refs. [46,49]. Using the combined experimental information, we list in Table 2 a few moments of the experimental photoabsorption cross section. Approximately 90% of the quoted uncertainties originate from the systematic uncertainties associated with the photon flux and detector efficiency measurements. As a reference, values for the classical TRK sum rule are also provided. Beside these moments, additional predictions are displayed in Table 3 for the neutron-skin thickness of $^{206}$Pb (and $^{208}$Pb) together with a few bulk parameters of the symmetry energy; see Eq. (2). These predictions were performed using a small set of accurately calibrated relativistic EDFs [51,52], with ground state properties computed in a mean-field approach and the dipole response in a self-consistent RPA and the EDF+QPM (GiEDF) [23,24] approaches; for a recent review on the imprint of the symmetry energy on the dipole response see Ref. [53]. Table 3 illustrates powerful correlations between nuclear observables. In the case of the isovector dipole mode, the out of phase oscillation of protons against neutrons, the symmetry energy acts as the restoring force. In particular, theoretical models with a soft symmetry energy, namely models that predict a slow increase of the symmetry energy with density (i.e., small $L$) predict large values for the symmetry energy at the sub-saturation densities of relevance to the excitation of this mode. In turn, this induces a quenching and hardening of the dipole strength relative to their stiffer counterparts (i.e., models with large $L$). Because the distribution of dipole strength is sensitive to the density dependence of the symmetry energy, the energy weighted sum (or total photoabsorption cross section $\sigma_D$) is not, as it is “protected” by the classical TRK sum rule. Indeed, Table 3 indicates a mild model dependence (of a few percent) in $\sigma_D$. Instead, the $E^{-2}$ weighting enhances the low-energy response and reveals a large sensitivity of $\sigma_{-2}$ (of $\sim 25\%$) to the density dependence of the symmetry energy. These findings suggest the following insightful correlation: the stiffer the symmetry energy, the larger the neutron-skin thickness of heavy nuclei, and the larger the electric dipole polarizability [63–55]. Based on the limited set of relativistic and nonrelativistic EDFs displayed in Table 3, the measured experimental value of $\sigma_{-2}$ in $^{206}$Pb suggests a fairly soft symmetry energy, with values of its slope at saturation density in the range $48 \lesssim L \lesssim 60$ MeV. Correspondingly, the extracted range of values of the neutron skin in $^{208}$Pb is $0.12 \lesssim \Delta r_{\text{skin}}^{208} \lesssim 0.19$ fm, which translates into $0.13 \lesssim R_{\text{skin}}^{208} \lesssim 0.21$ fm for $^{208}$Pb. From the EDF+QPM calculations of both, $E1$ and $M1$ strengths up to $25$ MeV in $^{208}$Pb (the corresponding $\sigma_D$, $\sigma_{-1}$ and $\sigma_{-2}$ values are given in Table 3 with the notation GiEDF), we obtained for the total dipole polarizability $\alpha_D^{\text{GiEDF}}(^{208}\text{Pb}) = 127$ mb/MeV = 18.3 fm$^2$/eV. The comparison of the $\sigma_D$ value with the one obtained in $^{208}$Pb [7] shows a decrease of $3\%$ in $^{208}$Pb. This is found correlated with the decrease of the neutron skin thickness of $^{208}$Pb, which is $4\%$ less than that of $^{206}$Pb (see in Table 3).

We now proceed to assess the impact of the present measurement on the neutron capture cross section, and possibly on stellar nucleosynthesis. The intensity and the energy distribution of the nuclear dipole response, including both the low-energy PDR plus the contributions from core polarization below the neutron separation energy, are fundamental ingredients for the determination of the neutron-capture rates [26,31,56]. To estimate the radiative neutron cross section of $^{205}$Pb, statistical model calculations using the TALYS-1.8 code [57] have been carried out, with the results displayed in Fig. 4. The “Recommended” curve corresponds to the radiative cross section obtained with the presently measured $E1$ and $M1$ $\gamma$-ray strength, complemented by EDF+QPM predictions outside of the experimental energy range. The uncertainty band in Fig. 4 stems from the experimental uncertainties associated with the $\gamma$-ray strength, but is also due to the use of different models to predict the nuclear level density [58,59]. The recommended cross section is obtained with the combinatorial model of Ref. [60]. Note that level densities are constrained on the cumulative number of low-lying levels, including the $J = 1$ states measured in the present experiment. As shown in Fig. 4, the QPM model supplemented with QRPA calculations generate a cross section that is in excellent agreement with the calculations based on the experimental strength. Whereas the $M1$ contribution is found to be rather insignificant (less than 5\%), the combined PDR plus core polarization contribution is crucial for a proper description of the cross section. Indeed, excluding the PDR contribution, QRPA predictions by themselves yield negligible $E1$ strength below 6 MeV, leading to a cross section about 5 times lower relative to the one involving the combined contribution, as shown in Fig. 4. Note that the contribution of higher multipoles has also been included in the calculation of the radiative neutron capture cross section but remains negligible. Based on these results, the experimentally constrained Maxwellian-averaged cross section at 30 keV is estimated to be $130 \pm 25$ mb—a value that is consistent with the prediction of $125 \pm 22$ mb estimated solely by theoretical means. This latter value has been recommended in Ref. [61] and has been traditionally used in s-process calculations. With this updated cross section, the dynamics involved in the survival and destruction of $^{205}$Pb in AGB stars is put on much more solid ground.
Table 2
Summary of a few moments of the photoabsorption cross section of $^{206}\text{Pb}$ and $^{208}\text{Pb}$.

| Nucleus | $E_{\text{cm}}$ (MeV) | G0NZ(A) (mb MeV) | $\sigma_0$ (mb MeV) | $\sigma_{-1}$ (mb) | $\sigma_{-2}$ (mb MeV) | Ref. |
|---------|------------------------|------------------|-------------------|------------------|----------------------|-----|
| $^{206}\text{Pb}$ | 26 | 2962 | 3544±294 | 241±17 | 18±1 | Present+ [46,49] |
| $^{208}\text{Pb}$ | 25 | 2980 | 3981±331 | 287±18 | 20±1 | [50] [

Table 3
Moments of the photoabsorption cross section of $^{206}\text{Pb}$ as in Table 2, but now as predicted by a series of accurately calibrated relativistic EDFs [51,52] and the non-relativistic EDF (GiEDF) underlying the QPM approach. Also shown is the neutron-skin thickness of $^{208}\text{Pb}$ (and $^{206}\text{Pb}$ not displayed in square brackets) as well as values of the symmetry energy ($J$) its slope ($L$) and its curvature ($K_{\text{sym}}$) at saturation density [see Eq. (2)]. The large negative GiEDF value for $K_{\text{sym}}$ is typical for non-relativistic approaches, see e.g. [29].

| Model | $\sigma_0$ (mb MeV) | $\sigma_{-1}$ (mb) | $\sigma_{-2}$ (mb MeV) | $R_{\text{skin}}$ (fm) | $J$ (MeV) | $L$ (MeV) | $K_{\text{sym}}$ (MeV) |
|-------|---------------------|-------------------|-----------------------|------------------------|----------|----------|------------------|
| RMF012 | 3653 | 237 | 17 | 0.12 [0.13] | 29.8 | 48.3 | 98.7 |
| FSU Garnet | 3689 | 243 | 18 | 0.15 [0.16] | 30.9 | 51.0 | 59.5 |
| FSU Gold | 3638 | 251 | 19 | 0.19 [0.21] | 32.6 | 60.5 | 51.3 |
| RMF028 | 3711 | 265 | 21 | 0.26 [0.29] | 37.5 | 112.6 | 26.2 |
| RMF032 | 3812 | 262 | 21 | 0.30 [0.32] | 41.3 | 125.6 | 28.6 |
| GiEDF | 3060 | 230 | 18 | 0.15 [0.16] | 33.4 | 53.9 | $-188.4$ |

Fig. 4. (Color online.) Radiative capture cross section $^{206}\text{Pb}(n,\gamma)^{207}\text{Pb}$ using as input the experimental $E1$ and $M1$ dipole strength (red curve) or the three-phonon EDF-QPM plus EDF-QRPA predictions (blue curve). The dotted line is obtained with the EDF-QRPA strength excluding the PDR contribution.

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