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# Evidence for radiative coupling of the pygmy dipole resonance to excited states

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The photoabsorption cross section and ground-state branching ratio of $^{142}$Nd were measured using quasimonoenergetic $\gamma$-ray beams at several beam energies. Two peaks corresponding to the pygmy dipole resonance (PDR) were identified. The branching ratios were compared to statistical-model calculations. We found that the Brink hypothesis is violated, and that the branching ratios are only reproduced by introducing a possible new decay mode of the observed PDR.

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The pygmy dipole resonance (PDR) is an $E1$ excitation in atomic nuclei with a large neutron excess and has been identified to be an oscillation of the neutron skin [1]. The PDR is a mixed isovector and isoscalar mode [2], and is excited in $(\gamma, \gamma')$ experiments at the excitation energy of $E_x = 5{\sim}8$ MeV in the $N = 82$ isotones [3]. It is also excited in $(\alpha, \alpha'\gamma)$ experiments, being observed at $E_x = 5{\sim}6$ MeV in $^{140}$Ce [4] and $^{138}$Ba [5]. It is still not known how the PDR radiatively couples to excited states. An investigation of its radiative coupling can lead to a better understanding of its structure as well as the potential impact it may have on reaction rates that are important to nuclear astrophysics [6] and advanced nuclear fuel cycles [7].

The radiative coupling of nuclear states within the statistical model of the nucleus is typically described using the Brink hypothesis (BH) [8,9]. According to it, the giant dipole resonance (GDR; a collective oscillation where the neutrons and the protons in the nucleus are out of phase of each other) is built on excited states, and the properties of the GDR are independent of the nature of the state it is built on Ref. [10]. Such a GDR built on a state with energy $E$ will have its mean energy shifted up by $E$, but will otherwise retain the same properties.

The BH has been shown to be a reasonable concept for the GDR at low excitation energies in a variety of experiments, including those measuring average intensities of primary transitions from (n,$\gamma$) [11], (p,$\gamma$) [12], and $(\gamma, \gamma')$ reactions [13]. Reactions with light ions which double excite the GDR also substantiate the BH [14,15]. Reactions with heavy ions at relativistic energies demonstrate that the GDR is built on excited states, but that the width increases as the total temperature of the nucleus increases [16,17]. This means a violation of the strict form of the hypothesis, but this is supposed to be describable as a smooth and slowly varying function of excitation energy. The violation of the BH in the region of the low-energy tail of the GDR has not been demonstrated yet, to our knowledge. If it is, it could affect essentially all calculations using the statistical model.

The BH was shown to be at least approximately valid also for the $M1$ scissors mode (i.e., a collective excitation observed at about 3 MeV in deformed nuclei) using data from the measurement of two-step $\gamma$ cascades following thermal-neutron capture [18]. In practice, the BH is usually assumed to be valid not only for the GDR, but for photon emission and photon absorption in general [10].

In this Rapid Communication, we studied the radiative coupling of the PDR to excited states and the applicability of the BH in a $(\gamma,\gamma')$ experiment on $^{142}$Nd using a quasimonoenergetic beam. This was done by measuring the photoabsorption cross section and ground-state branching ratio at several beam energies, and testing their mutual consistency within the nuclear statistical model [13,19]. The $^{142}$Nd$(\gamma,n)$ cross section was additionally measured to constrain the low-energy tail of the GDR above the neutron separation energy, since the extrapolation of the GDR can affect statistical-model calculations. We show that the BH is violated, and the only way we found to explain the observed branching ratios is by introducing a resonance built only on excited states at $E_\gamma = 4.9{\sim}6.3$ MeV, which may be a newly observed decay mode of the PDR.

The $^{142}$Nd$(\gamma, \gamma')$ reaction (nuclear resonance fluorescence) was studied at 18 energies at $E_\gamma = 4.2{\sim}9.7$ MeV using the quasimonoenergetic 100% linearly polarized $\gamma$-ray beam at the

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High Intensity $\gamma$-ray Source (HI$\gamma$S) at Duke University [20], with an average beam width of 3%. The target was 30 g of $Nd_2O_3$ powder enriched to 96% in $^{142}$Nd. It was encapsulated in an acrylic cylinder with an inner diameter of 2.54 cm and an inner length of 2.65 cm. The wall thickness was 0.3 cm. The experimental setup and procedure were similar to that reported in Ref. [21]. In the present experiment, the high-purity Germanium (HPGe) detector array consisted of four clover detectors [22] arranged perpendicular to the beam, with two in the horizontal plane and two in the vertical plane. The efficiency was measured using a calibrated $^{56}$Co source up to 3.5 MeV, and simulated up to 10 MeV using the Monte Carlo *N*-particle transport code [23].

The $E1$ $(\gamma,\gamma')$ ground-state (elastic) cross section $\sigma_{\gamma 0}$ and transition cross sections from several low-lying excited states to the ground state were measured. From these, the $E1$ photoabsorption cross section $\sigma_{\gamma T}$ and the average ground-state branching ratio $\langle b_0 \rangle$ were extracted. The $M1$ elastic cross section was also measured, and the results will be reported elsewhere along with a comparison of our data with previously observed discrete states [3].

The intensity of transitions directly to the ground state used in determining the $E1$ $\sigma_{\gamma 0}$ was obtained by fitting the vertical-detector spectrum in the energy region of the incident beam. The fit function accounted for the observed discrete states as well as the contribution from the many other states excited by the beam, but not individually resolved. The detector response including the Compton edge and the first and second escape peaks was included. The $E1$ $\sigma_{\gamma 0}$ was determined with an average uncertainty of 7% and was corrected for coherent atomic scattering [24]. The transition multipolarity was uniquely determined because of the polarized nature of the beam (see Fig. 1) [25].

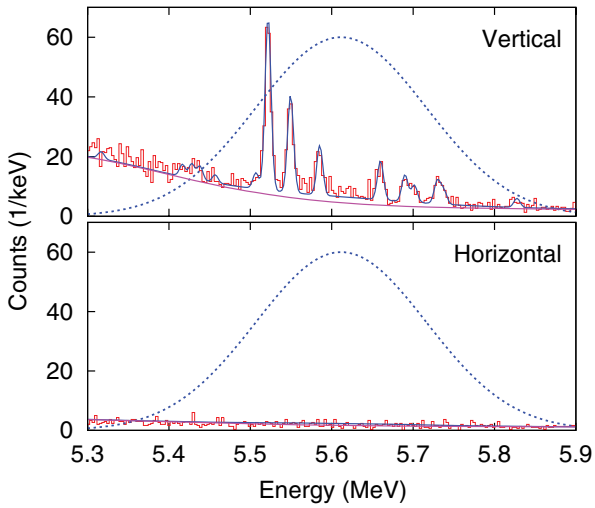


FIG. 1. (Color online) Experimental $^{142}$Nd$(\gamma, \gamma')$ spectra at $E_{\text{beam}} = 5.6$ MeV. The fit to the peaks and the detector response (primarily the Compton edge) is overlayed. The dashed line is the beam energy profile. The vertical (horizontal) spectrum corresponds to $E1$ ($M1$) transitions with a small (4%) overlap of the two angular distributions. Ground-state $E1$ transitions for several discrete states are seen in the vertical-detector energy spectrum. At this energy, no discrete transitions are seen in the horizontal detectors.

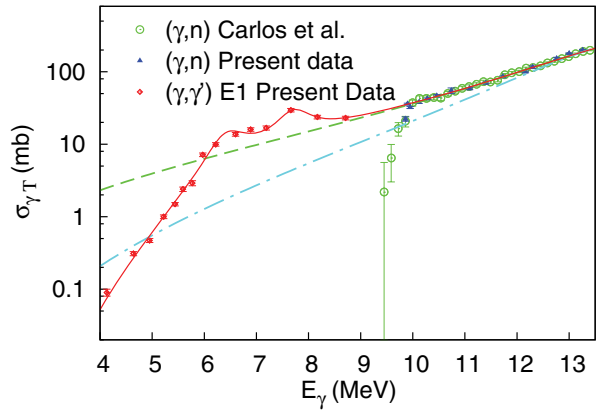


FIG. 2. (Color online) The $^{142}$Nd $(\gamma, n)$ cross section and the $E1$ photoabsorption cross section $\sigma_{\gamma T}$. The data from Carlos *et al.* [28] was scaled by 0.86 [29]. The dashed (dash-dotted) line is a fit to the combined $(\gamma, n)$ data using a Lorentzian (temperature-dependent width) function for the GDR [30]. The solid line is a fit to the experimental data.

The $E1$ $\sigma_{\gamma T}$ was determined from the sum of the $E1$ $\sigma_{\gamma 0}$ and the transition cross sections from up to seven low-lying excited states (i.e., first six $2^+$ states and the first $1^-$ state, all at $E_x < 3.5$ MeV) to the ground state. The low-lying states were fed via cascade from the states initially excited by the beam. Individual ground-state transitions at $E_x > 3.5$ MeV were too weak to be directly observed. The possible contribution from such sidefeeding to the photoabsorption cross section was accounted for in the simulations described below. The calculated contribution from sidefeeding averaged about 10% in the region of the PDR and reached a maximum value of about 25% at $E_\gamma = 9.7$ MeV. The cross sections were corrected for the attenuation of the beam flux through the target, which varied between 11% and 20%. The cross sections at $E_\gamma \geqslant 9$ MeV were not obtained because of experimental difficulty in measuring the beam flux. $\langle b_0 \rangle$ was obtained by dividing $\sigma_{\gamma 0}$ by $\sigma_{\gamma T}$, and is independent of the absolute flux normalization and correction for attenuation of the flux through the target. $\langle b_0 \rangle$ is shown in Fig. 3.

The $^{142}$Nd $(\gamma, n)$ cross section was measured at 15 energies at $E_\gamma = 9.86 \sim 13.3$ MeV (see Fig. 2). The quasimonoenergetic $\gamma$-ray source at the National Institute of Advanced Industrial Science and Technology in Tsukuba, Japan [26] was used. The experimental setup, method, and analysis techniques employed here have been previously reported in Ref. [27]. The measured cross sections agree with results reported in Ref. [28] when they are multiplied by a normalization factor of 0.86, as recommended by Ref. [29]. The combined $(\gamma, n)$ data is well described by using a Lorentzian function for the GDR.

The $(\gamma,\gamma')$ photoabsorption cross section $\sigma_{\gamma T}$ deviates strongly from the Lorentzian function of the GDR, as shown in Fig. 2. At $E_\gamma \geqslant 6$ MeV, the measured cross section exhibits resonancelike structures interpreted as two components of the PDR at $E_x = 6.5$ and 7.8 MeV. The component at $E_x = 6.5$ MeV was observed previously as a cluster of discrete states [3], while the component at $E_x = 7.8$ MeV is observed for the first time. A suppression, or drop off, of $\sigma_{\gamma T}$ is seen at $E_x < 6$ MeV and goes below the value expected for a

Lorentzian function of the GDR and other known functions for the GDR [30]. A similar suppression in strength in the $(\gamma, \gamma')$ cross section was seen in $^{138}$Ba [31] at $E_\gamma < 5.5$ MeV and in $^{139}$La [32] at $E_\gamma < 6.0$ MeV, indicating that this is a common feature of the $N = 82$ isotones. In addition, both Refs. [31] and [32] see a similar enhancement in the cross section near 7.8 MeV, which may be due to the possible presence of a second component of the PDR.

To study the coupling of the PDR to excited states and the validity of the BH, we simulated $\langle b_0 \rangle$ using the statistical model. Simulations were based on the DICEBOX algorithm [33], which was modified to allow simulations of the $(\gamma,\gamma')$ reaction. The only difference of the algorithm with respect to Ref. [33] is in the definition of the initial states for $\gamma$ decay. The population of initial states was accounted for by knowing the energy profile of the flux and the partial radiation widths of all states to the ground state. The beam profile was taken from experiment, while the radiation widths were calculated as described in the following paragraph. The simulated $\langle b_0 \rangle$ was reconstructed using only contributions from transitions, which were also observed experimentally to reproduce the experimental values of $\langle b_0 \rangle$.

DICEBOX uses the $E1$, $M1$, and $E2$ radiative strength functions (RSF) (i.e., average radiative width as a function of energy for a given transition multipolarity) to calculate the radiative transition widths between all states in a simulated level scheme. The used $E1$ strength function is taken from the experimental $E1$ $\sigma_{\gamma T}$ [13,19]. $\sigma_{\gamma T}$ was fit with a phenomenological function, which reproduced the shape at all energies including the PDRs and the exponential drop at low energy. The exponential decrease was extrapolated for $E_x \rightarrow 0$ MeV. The $M1$ and $E2$ strength functions used the single-particle models [34] and they only significantly affect the branching ratios at $E_x < 5$ MeV.

The simulated level scheme was constructed using the back-shifted Fermi gas (BSFG) model for the level density [35]. The level-density model was constrained by the set of known discrete states at $E_x < 3.5$ MeV, which is believed to be complete. It was then adjusted within this constraint to minimize $\chi^2$. A parity dependence to the level density was assumed at $E_x < 4.8$ MeV, assuming entirely positive parity states. We found the parity dependence to be necessary to satisfy the two constraints on the level density mentioned above for each case.

An advantage of DICEBOX in comparison with all other codes is that it takes into account the fluctuation properties of individual partial radiation widths [36]. Individual widths are expected to fluctuate around their expectation value according to a $\chi^2_\nu$ distribution with $\nu = 1$ degrees of freedom. Inclusion of the fluctuation of radiative widths to the simulations allowed us to estimate the uncertainty that arises from not knowing precisely the radiation widths for each state.

Various models of how the GDR and PDR are built on excited states were tested with the DICEBOX code in an attempt to reproduce the experimental data. Using the BH, it is not possible to reproduce the experimental $\langle b_0 \rangle$ at all measured energies [see Fig. 3(a)], with the largest disagreement at $E_\gamma = 6\sim8$ MeV, which is the same region corresponding to the PDR. Neither modifying the level density further nor

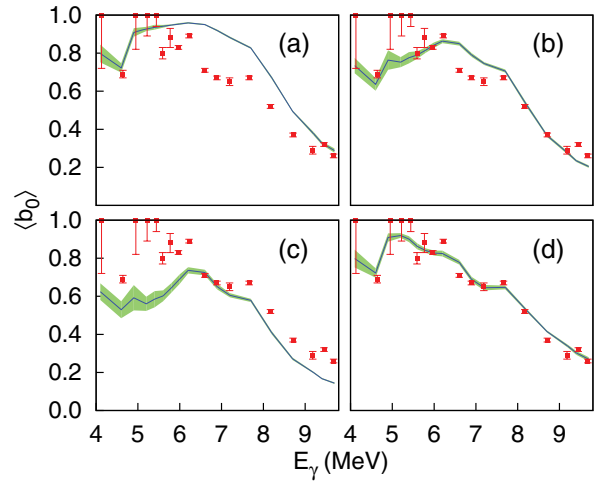


FIG. 3. (Color online) The experimental and calculated $\langle b_0 \rangle$ for four models: (a) the experimental $\sigma_{\gamma T}$ built on excited states (the BH model), (b) the $T$-dependent width model for the extrapolation of the tail of the GDR for $E_x \rightarrow 0$ MeV, but using the experimental cross section where it is larger than the extrapolation function, which occurred at about $E_\gamma > 5.3$ MeV, (c) similar to model (b), but using the GLO model for the GDR, and (d) similar to model (a), but including a resonance built only on excited states between about 5 and 6 MeV, and able to reproduce $\langle b_0 \rangle$ at all energies. The simulation is shown as a solid line and its uncertainty is given by the shaded region.

$\nu$ of the $\chi^2_\nu$ distribution improved agreement. According to our knowledge, this is the first time that the BH has been demonstrated to be violated for the low-energy tail of the GDR.

Changing the way the GDR is built on excited states failed to produce agreement with the experimental $\langle b_0 \rangle$. Radiative strength functions were tested, which used different functions for the extrapolation of the tail of the GDR for $E_x \rightarrow 0$ MeV, but used the experimental cross section where it is larger than the extrapolated tail of the GDR, which occurred at about $E_\gamma > 5.3$ MeV. Two functions were tested for the extrapolation [30]: the temperature-dependent width ($T$-dependent) function and the generalized Lorentzian (GLO) function [see Figs. 3(b) and 3(c), respectively]. The $T$-dependent function was chosen because it gives the least amount of strength at low energies out of all known functions describing the GDR. The GLO function was chosen as it best describes radiative widths measured using a $(n, \gamma)$ reaction in the neighboring nucleus, $^{143}$Nd [30]. It has a nonzero extrapolation of the RSF for $E_x \rightarrow 0$ MeV.

The $T$-dependent function cannot reproduce $\langle b_0 \rangle$ at all $E_\gamma$, significantly disagreeing at $E_\gamma = 6\sim8$ MeV [see Fig. 3(b)]. The disagreement is even larger for the GLO function as the additional strength at low energies further lowers $\langle b_0 \rangle$ [see Fig. 3(c)]. These two functions for the radiative strength function exclude all known proposed functions for the low-energy tail of the GDR [34], as the behavior of other functions at $E_x < 5$ MeV is either similar to the GLO function or includes more strength at low energies, increasing the disagreement.

Simulations, which included the PDR coupled only to the ground state and used the available extrapolations of the GDR for coupling the PDR region to excited levels, did not

significantly affect the branching ratios in the energy region of the PDR.

In an attempt to reproduce $\langle b_0 \rangle$ at all energies, a resonance modeled as a Lorentzian function was added to the strength function at low energies. The resonance parameters and the energy range of the excited states that it is built on were varied freely in a $\chi^2$ fit to the experimental $\langle b_0 \rangle$. The level density used for Fig. 3(a) was used for the fit. To optimize fitting time, $\langle b_0 \rangle$ was calculated using a simplified model, which was sufficient for calculating branching ratios [13,19]. Assuming either an $E1$ or $M1$ multipolarity produced the same results. The fit results were then reproduced and confirmed using DICEBOX.

The resulting resonance successfully reproduces $\langle b_0 \rangle$ at all energies [see Fig. 3(d)]. It has a mean energy of $E_r = 1.0 \pm 0.4$ MeV, a width of $\Gamma = 2.6 \pm 0.4$ MeV, a maximum cross section of $\sigma_0 = 0.6 \pm 0.1$ mb, and is built only on excited states from $E_x = 4.9 \pm 0.1$ to $6.3 \pm 0.1$ MeV. Many different random initial conditions were tested for $E_r$, $\Gamma$, $\sigma_0$, and $E_x$, confirming the above results. This resonance is at a similar location to those of the "soft-pole" resonances (Er $< 2\sim3$ MeV) observed in nuclei with $A < 100$ [37–40] in the $\gamma$-ray cascade following excitation by a light ion beam, and could similarly impact calculated reaction rates for nuclear astrophysics [41]. This is the first evidence of such a soft resonance in a spherical nucleus with $A > 100$, as far as we know. The present result suggests it may be a decay mode of the PDR.

The resonance may be a transition between two possible modes of the PDR, as suggested by the presence of two PDR peaks, and the energy region of states that the resonance is built on. This would require a transition multipolarity of $M1$ or $E2$ such that states with $J^\pi = 1^-$ would be exclusively transitioned to. This was tested and reproduced the experimental $\langle b_0 \rangle$ for either $M1$ or $E2$. It requires $\sigma_0 \approx 10$ mb, but all other parameters stayed the same. Subsequent decay to the ground state would be preferred, which could be investigated by a $\gamma$-$\gamma$ coincidence measurement.

The low-energy $\gamma$ rays from this resonance were not directly observable in the present experiment because of the large $\gamma$-ray background at the transition energy due to beam interaction with electrons in the target. The direct transition may be observable in charged-particle experiments which directly excite the PDR and measure the resulting $\gamma$-ray cascade (see, e.g., Ref. [38]).

The BH is violated by the proposed resonance as it is built only on states in a specific energy range [10]. In other words, the RSF depend on $E_x$. If universal, this could affect all calculations using the nuclear statistical model and improve their accuracy as our understanding of the violation of the BH grows.

The origin of the proposed resonance may be understood by considering the nature of the PDR. In heavy nuclei due to Coulomb and symmetry energies, excitations generated by isoscalar and isovector vibrations are mixed, including for the PDR [42,43]. The PDR includes partly a toroidal surface excitation [44,45] where the neutron skin may play a part in enhancing isoscalar-isovector mixing [46]. Theoretical calculations show that because the proton number is far from a magic number, the proton system in the ground state of $^{142}$Nd is deformed, resulting in the PDR splitting into $K = 0$ and $K = 1$ parts (oscillations along and perpendicular to the proton symmetry access, respectively), with $K = 0$ at lower energy [47,48]. This indicates that the inclusion of deformation will be necessary to fully explain the properties of the PDR.

In summary, the $^{142}$Nd photoabsorption cross section $\sigma_{\gamma T}$ was measured using the $(\gamma,\gamma')$ and the $(\gamma,$n) reactions at $E_\gamma = 4.2\sim13.3$ MeV. Two distinct peaks of the PDR were observed, as well as a suppression in $\sigma_{\gamma T}$ at $E_x < 6$ MeV, going lower than any known function used to describe the tail of the GDR. The ground-state branching ratio $\langle b_0 \rangle$ was measured in the $(\gamma,\gamma')$ experiment. Simulations using the statistical model testing the mutual consistency of $\sigma_{\gamma T}$ and $\langle b_0 \rangle$ indicate that the BH is violated for the radiative decay of $1^-$ states in the energy region corresponding to the PDR. This is the first observation of the violation of the BH in the low-energy tail of the GDR, as far as we know. If the violation of the BH is universal, then it could affect all calculations using the statistical model, including those for nuclear astrophysics and nuclear reactors. The only model we found to reproduce the experimental $\langle b_0 \rangle$ introduces a resonance built solely on excited states between about 5 and 6 MeV, which may be a newly observed decay mode of the PDR.

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