## $^{138}$ Ba $(d,\alpha)$ Study of States in $^{136}$ Cs: Implications for New Physics Searches with Xenon Detectors

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B. M. Rebeiro, 1.2 S. Triambak, 1.* P. E. Garrett, 3.1 G. C. Ballo, 4 B. A. Brown, 5 J. Menéndez, 6 B. Romeo, 7
P. Adsley, 8 B. G. Lenardo, 9 R. Lindsay, 1 V. Bildstein, 3 C. Burbadge, 3.† R. Coleman, 3 A. Diaz Varela, 3 R. Dubey, 1.10
T. Faestermann, 11 R. Hertenberger, 12 M. Kamil, 1 K. G. Leach, 13 C. Natzke, 13 J. C. Nzobadila Ondze, 1
A. Radich, 3 E. Rand, 3 and H.-F. Wirth, 12

1 Department of Physics and Astronomy, University of the Western Cape, P/B X17, Bellville 7535, South Africa

2 Department of Physics, McGill University, Montréal, Québec H3A 278, Canada

3 Department of Physics, University of Guelph, Guelph, Ontario N1G 2W1, Canada

4 TRIUMF, 4004 Wesbrook Mall, Vancouver, British Columbia V6T 2A3, Canada

5 Department of Physics and Astronomy and National Superconducting Cyclotron Laboratory, Michigan State University,
East Lansing, Michigan 48824-1321, USA

6 Department of Quantum Physics and Astronomy and Institute of Cosmos Sciences, University of Barcelona, 08028 Barcelona, Spain

8 Cyclotron Institute and Department of Physics and Astronomy, Texas A&M University, College Station, Texas 77843, USA

10 Institute of Physics, University of Szczecin, 70-451 Szczecin, Poland

11 Physik Department, Technische Universität München, D-85748 Garching, Germany

12 Fakultät für Physik, Ludwig-Maximilians-Universität München, D-85748 Garching, Germany

13 Department of Physics, Colorado School of Mines, Golden, Colorado 80401, USA
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We used the  $^{138}$ Ba(d,  $\alpha$ ) reaction to carry out an in-depth study of states in  $^{136}$ Cs, up to around 2.5 MeV. In this Letter, we place emphasis on hitherto unobserved states below the first  $1^+$  level, which are important in the context of solar neutrino and fermionic dark matter (FDM) detection in large-scale xenon-based experiments. We identify for the first time candidate metastable states in  $^{136}$ Cs, which would allow a real-time detection of solar neutrino and FDM events in xenon detectors, with high background suppression. Our results are also compared with shell-model calculations performed with three Hamiltonians that were

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Our results are also compared with shell-model calculations performed with three Hamiltonians that were previously used to evaluate the nuclear matrix element (NME) for <sup>136</sup>Xe neutrinoless double beta decay. We find that one of these Hamiltonians, which also systematically underestimates the NME compared with the others, dramatically fails to describe the observed low-energy <sup>136</sup>Cs spectrum, while the other two show reasonably good agreement.

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It has been pointed out [1] that double beta decaying atomic nuclei provide the necessary framework to perform real-time spectroscopic studies of solar neutrinos, with high background suppression. In such cases, the parent nucleus has an even number of protons (Z) and neutrons (N), with A = Z + N, and total angular momentumparity  $J^{\pi} = 0^+$ . Consequently, the attractive nuclear pairing interaction renders it more bound than its isobaric (A, Z+1) neighbor, which has odd Z and N. This scenario precludes single  $\beta$  transitions of the type  $(A, Z) \rightarrow (A, Z + 1)$  and presents a "stable" target for the solar  $\nu_e$  flux,  $\phi_e$ . It also results in low thresholds for charged-current (CC)  $\nu_e$  capture to  $J^{\pi} = 1^+$  states in the (A, Z + 1) system. As this intermediate nucleus is oddodd, its low-lying structure is mainly defined by twoquasiparticle configurations for the unpaired proton and neutron. Such configurations may lead to the existence of metastable states, with long half-lives that permit a nearly

background-free identification of CC solar  $\nu_e$  captures, via a delayed coincidence analysis [1].

In this regard, xenon-based detectors [2–8] present a unique opportunity for solar neutrino detection, both at the tonne scale and beyond. As examples, the nEXO [2], KamLAND-Zen [3], and NEXT [4] experiments rely on isotopically enriched xenon to search for lepton-numberviolating (LNV) neutrinoless double beta decays  $(0\nu2\beta)$  of <sup>136</sup>Xe. The low  $\nu_e$  reaction threshold for <sup>136</sup>Xe presents a compelling case to use such xenon detectors for solar neutrino astronomy at energies ≤1 MeV. A previous study [9] showed that the dominant CC  $\nu_e$  captures on  $^{136}$ Xe will be through the two lowest-energy 1<sup>+</sup> states in <sup>136</sup>Cs, at 591 and 845 keV respectively [10], with the former 1<sub>1</sub><sup>+</sup> state being the most significant ( $Q_{\nu} = 681.3 \text{ keV}$ ). As a result, detectors loaded with 136Xe will be sensitive to  $\phi_e(\text{CNO}, {}^7\text{Be}, {}^8\text{B}, pep)$ . Of particular interest are  ${}^7\text{Be}$ electron-capture neutrinos and those emitted from the solar CNO cycle, whose detection will offer insight into the innermost core of the Sun [11–13]. Additionally, such experiments can also identify similar CC-type excitations to 1<sup>+</sup> states in <sup>136</sup>Cs, caused via MeV-scale fermionic dark matter (FDM) absorption [14,15] on <sup>136</sup>Xe.

Based on the above, a search for FDM absorption on <sup>136</sup>Xe was recently performed [16]. However, the analysis was severely challenged by the meager experimental information [17] available for the low-lying level scheme of <sup>136</sup>Cs. Only three states have thus far been experimentally verified below the  $1_1^+$  level, with assigned  $J^{\pi}$  values of 4<sup>+</sup>, 8<sup>-</sup>, and 9<sup>-</sup>, respectively [17–19]. Independently, shellmodel calculations [9] were performed to predict  $\gamma$ -ray deexcitation paths from the  $1_1^+$  level in  $^{136}$ Cs. The results showed promise for solar  $\nu_e$  detection in both current and next-generation xenon experiments, mainly because of feeding to the predicted first excited state in <sup>136</sup>Cs  $(E_x = 23 \text{ keV}, J^{\pi} = 3^+)$ , which connects to the ground state via a slow ( $\tau = 851 \text{ ns}$  [20])  $3_1^+ \rightarrow 5_1^+$  electricquadrupole (E2) transition. As this level has not been experimentally validated to date, a more comprehensive elucidation of the low-lying structure of <sup>136</sup>Cs is essential to make further progress in this regard.

There is additional widespread interest to accurately determine the nuclear matrix elements (NMEs) for various  $0\nu2\beta$  candidates, including <sup>136</sup>Xe [21–24]. The calculated NME for this particular case ranges from  $M^{0\nu} = 1.11-4.77$ [25], for light Majorana neutrino exchange. This theoretical limitation translates into an inevitable uncertainty band [3] on the LNV parameter responsible for the decay, which is hoped to be extracted from future experiments. Within the nuclear shell model, the NME is in the range  $M^{0\nu} = 1.63-2.45$ , depending on the Hamiltonian used for the calculation. This spread is primarily because one of the Hamiltonians (QX) yields a systematically lower value for  $M^{0\nu}$ , by about 40%, as shown in Table I. This systematic discrepancy appears to persist [26] even when recently acknowledged short-range NMEs [27] are taken into consideration. Therefore, an accurate understanding of the low-energy level scheme in <sup>136</sup>Cs also presents a robust testing ground for theory calculations of the  $^{136}$ Xe  $0\nu2\beta$ NME. This is because comparisons with experiment are much more sensitive to details of the nuclear Hamiltonian in odd-odd nuclei. Such details can be masked in even-even systems such as <sup>136</sup>Xe and <sup>136</sup>Ba, because of the dominant pairing interaction and other collective effects.

With these motivations in place, this Letter reports a detailed high-resolution investigation of low-lying states in  $^{136}\mathrm{Cs}$ . We used the  $^{138}\mathrm{Ba}(d,\alpha)^{136}\mathrm{Cs}$  two-nucleon transfer reaction, which is well suited for such a study.

The experiment was performed at the Maier-Leibnitz Laboratorium in Garching, Germany. A 600 nA, 22 MeV deuteron beam was incident on a 99.8% enriched  $40~\mu g/cm^2$ -thick  $^{138}BaO$  target, evaporated on a carbon

TABLE I. Shell-model-evaluated NMEs for  $^{136}$ Xe  $0\nu2\beta$ .

| Hamiltonian             | $M^{0 u}$       |
|-------------------------|-----------------|
| GCN5082 [28]            | 2.28, 2.45 [29] |
| $V_{\text{low-}k}$ [30] | 2.39 [30]       |
| JJ55t (SN100t) [31]     | 2.06, 2.21 [31] |
| QX (SVD, MC) [32]       | 1.63, 1.76 [33] |

foil. The reaction ejectiles were momentum analyzed with a high-resolution Q3D magnetic spectrograph [34]. The  $\alpha$ particles were selected by comparing the partial energy losses of the reaction products in two gas proportional counters and the total energy deposited in a plastic scintillator detector at the focal plane. For energy calibration, we used the  ${}^{94}\text{Mo}(d,\alpha){}^{92}\text{Nb}$  and  ${}^{92}\text{Zr}(d,\alpha){}^{90}\text{Y}$  reactions on enriched 94MoO3 and 92Zr targets that had thicknesses of 100 µg/cm<sup>2</sup> and 50 µg/cm<sup>2</sup>, respectively. The calibrations explicitly took into account differences in reaction kinematics and energy losses within the target foils, as described in Refs. [35,36]. A sample calibrated  $^{138}$ Ba $(d, \alpha)$  spectrum is shown in Fig. 1. The measured full widths at half maxima (FWHM) of the  $\alpha$  peaks were ~10 keV, vastly superior than the 40 keV resolution reported in a previous  ${}^{136}Xe({}^{3}He, t)$  study [10,37] that mainly investigated 1<sup>+</sup> states in <sup>136</sup>Cs.

The  $^{138}$ Ba $(d,\alpha)$  spectra were collected at different angles in the range  $\theta_{\rm lab}=5^{\circ}$ –45°, at 5° intervals. Additionally,  $^{138}$ Ba(d,d) elastic scattering data were acquired in the range  $\theta_{\rm lab}=15^{\circ}$ –115°, also at 5° intervals. We used these datasets to determine the target thickness and obtain differential scattering cross sections, as described in Refs. [25,38]. The measured angular distributions were then compared to distorted wave Born approximation (DWBA) predictions, provided by the DWUCK5 computer code.

The selectivity of the  $(d, \alpha)$  reaction is such that the transferred np pair is in a relative l=0 state, with spin S=1 and isospin T=0 [39]. If both nucleons are picked up from the same single-particle  $(j^2)$  configuration, the

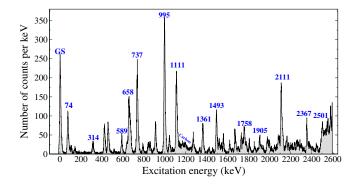


FIG. 1. Sample  $^{138}$ Ba $(d,\alpha)$  spectrum obtained at  $\theta_{\rm lab}=10^{\circ}$ . A few prominent peaks are labeled.

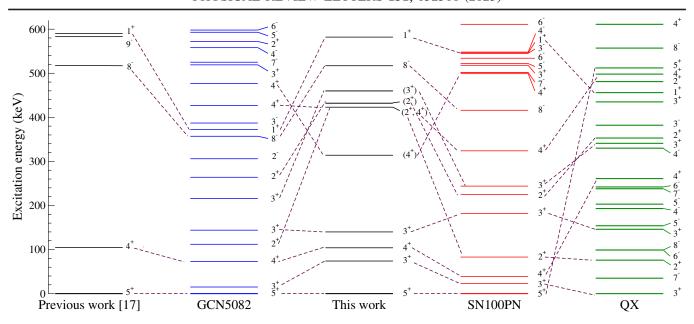


FIG. 2. Comparison between theory and experiment for the low-lying energy spectrum of <sup>136</sup>Cs. The shell-model results were obtained with the GCN5082, SN100PN, and QX effective interactions.

total angular momentum J of the final state is necessarily odd. However, if the neutron and proton are picked up from different configurations, with  $L = l_n + l_p$ , then J = L and  $J = L \pm 1$  states, with parity  $(-1)^{l_n + l_p}$  are produced [40].

For the DWBA analysis, we chose appropriate optical model parameters (OMPs) for the incoming  $d + {}^{138}\text{Ba}$ channel [41] by comparing our measured elastic scattering angular distribution with DWBA results from using different global OMPs. For the outgoing  $\alpha + {}^{136}\text{Cs}$  channel we chose the OMPs of Ref. [42], which were optimized for the  $^{136}$ Ba $(\alpha, \alpha)$  reaction at 20 MeV [43]. The  $^{138}$ Ba $(d, \alpha)$ calculations were performed assuming the "cluster" deuteron-transfer approximation [44,45], with form factors for a deuteron in a Woods-Saxon potential well, at the correct separation energy for each state in <sup>136</sup>Cs. We also took into consideration finite-range corrections [46,47] and nonlocality effects, using the prescription from Ref. [48]. Next, our measured cross section angular distributions were overlaid with normalized best-fit DWBA results. The latter were obtained assuming various L-transfer values for given J, and allowed incoherent summations of two different values L and L'. Identified states were then compared with shell-model predictions and previous measurements.

For the shell-model calculations we used a configuration space comprising the  $0g_{7/2}$ ,  $1d_{5/2}$ ,  $1d_{3/2}$ ,  $2s_{1/2}$ , and  $0h_{11/2}$  orbitals for neutrons and protons, and three different Hamiltonians: SN100PN [49], GCN5082 [28], and QX [32]. The SN100PN interaction is very similar to the JJ55t Hamiltonian [25], and was used by the authors of Ref. [9] to evaluate the level scheme of  $^{136}$ Cs. Independently, the GCN5082 and QX Hamiltonians were used to calculate the  $^{136}$ Xe  $0\nu2\beta$  NME [29,33].

Figure 2 compares calculated energy levels of <sup>136</sup>Cs to those identified from this experiment, up to the  $1_1^+$  level. Our results are summarized in Fig. 3 and Table II. We also used two-nucleon transfer amplitudes (TNAs) [50] obtained with the GCN5082 and SN100PN Hamiltonians for critical comparative cross-checks. This was feasible because most of the low-lying states had TNA dominated by simple two-nucleon configurations. For example, both calculations showed that the dominant orbitals involved in the transfer to the  $J^{\pi} = 5^{+}$  ground state [51,52] are  $g_{7/2}$  and  $d_{3/2}$  for proton ( $\pi$ ) and neutron ( $\nu$ ) pick up, respectively. This state can be produced by both L = 4 and L = 6 transfer. The relative L contributions can be evaluated via the *jj* to LS transformation that involves the normalized 9*j* coefficient [39],

$$\sqrt{3(2j_n+1)(2j_p+1)(2L+1)} \begin{cases} l_n & 1/2 & j_n \\ l_p & 1/2 & j_p \\ L & 1 & J \end{cases}.$$
 (1)

This yields a predominantly L=6 transition for the ground state, which is consistent with our observations. The same two-nucleon configuration dominates transfer to the  $3_1^+$  and  $4_1^+$  states. For the former, the intensity of the L=2 transition is nearly 17 times weaker than the L=4 transfer. This agrees with the measured angular distribution of the first excited state, observed at 74 keV. Next, we compared the measured cross section for this level relative to the ground state (after accounting for the difference in their predicted DWBA yields), with the relative scaling of their calculated transfer intensities. The reasonable agreement

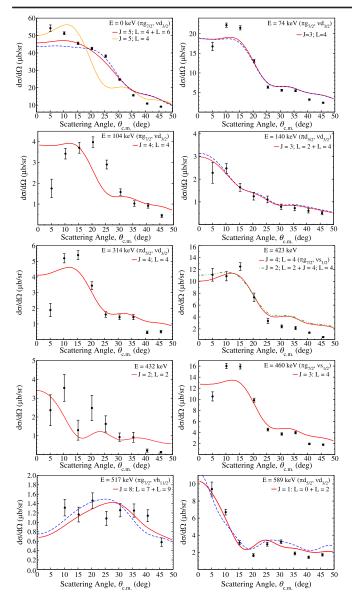


FIG. 3. Measured  $^{138}$ Ba( $d, \alpha$ ) angular distributions compared with best-fit DWUCK5 DWBA predictions (solid red curves). The blue dashed curves are from using fixed relative L contributions from Eq. (1). The dominant orbitals involved in the pair transfer are specified in each plot.

between these two values validated the  $3_1^+$  assignment for this state. In comparison, we identify the 140-keV state as  $3_2^+$ , whose dominant TNA corresponds to the  $(\pi d_{5/2})(\nu d_{3/2})$  orbitals. Both L=2 and L=4 transfer contribute for this state, which agrees well with the measured distribution. Spin-parity assignments for the remaining states identified in Table II were made through similar analysis of the shapes of the angular distributions, relative cross sections, and L-transfer intensities predicted by theory.

We do not observe explicit signatures of the low-lying 2<sup>+</sup> states, which are predicted to be weakly populated. We also do not observe the known 9<sup>-</sup> state at 583.9 (5) keV. This can be explained by the DWBA calculations,

TABLE II. Observed <sup>136</sup>Cs levels up to the 1<sup>+</sup><sub>1</sub> state.

| Refs. [10,17] |           | This Letter  |   |    |                    |  |
|---------------|-----------|--------------|---|----|--------------------|--|
| $E_x$ (keV)   | $J^{\pi}$ | $E_x$ (keV)  | L | L' | Assigned $J^{\pi}$ |  |
| 0.0           | 5+        | 0.0          | 4 | 6  | 5+                 |  |
|               |           | 74(2)        | 4 |    | $3^+$              |  |
| 104.8(3)      | $4^{+}$   | $104(2)^{a}$ | 4 |    | $4^{+}$            |  |
|               |           | 140(3)       | 2 | 4  | $3^+$              |  |
|               |           | 314(2)       | 4 |    | $(4^{+})$          |  |
|               |           | $423(3)^{b}$ | 4 |    | (4+)               |  |
| 431(2)        | $(3^{+})$ | 432(3)       | 2 |    | $(2^{+})$          |  |
|               | ` /       | 460(3)       | 4 |    | (3+)               |  |
| 517.9(1)      | 8-        | 517(3)       | 7 | 9  | 8-                 |  |
| 583.9(5)      | 9-        | •••          |   |    |                    |  |
| 591(2)        | $1^+$     | 589(3)       | 0 | 2  | 1+                 |  |

<sup>a</sup>Although the measured angular distribution for this state is dissimilar to other L=4 cases, our spin-parity assignment is consistent with a previous  $\gamma$ -ray measurement [18].

<sup>b</sup>Possible unresolved  $(4^+)$ ,  $(2^+)$  doublet. See text for details.

which show that L = 9 transfer for this state is significantly weaker than the dominant L = 7 transfer to the 8<sup>-</sup> state. A tentative 3<sup>+</sup> state was reported at 431 keV [10], but excluded from Ref. [17]'s compilation. We investigated this state's possible existence by refitting the 423-keV peak with fixed line shape parameters, based on previous knowledge of the detector response [53]. This analysis indicated a possible level at  $E_x = 432(3)$  keV, whose angular distribution is shown in Fig. 3. Although it is statistics limited, the measured distribution appears to be consistent with L=2 transfer. The intensity of this possible transition is comparable to those predicted for the  $2_1^+$  and  $2_2^+$  levels. We also observe that the  $\theta_{\rm lab} = 5^{\circ}$  cross section for the 423-keV state is enhanced compared with the other L=4 transitions. This can be attributed to an additional L=2component which is  $\sim 20\%$  of the L=4 contribution, as shown in Fig. 3. Thus, one cannot rule out an unresolved state at  $\sim$ 423 keV, with an L=2 contribution that corresponds to one of the  $2^+$  levels.

Figure 2 shows that the SN100PN and GCN5082 results are overall very similar and could be matched to our identified levels from this experiment. A recent independent calculation performed with the proton-neutron quasiparticle random-phase approximation [54] also shows reasonable overall agreement with our measured spectrum. However, there is a stark disagreement with the QX results where the  $5_1^+$  ground state shows up at a significantly higher energy. The QX interaction also shows several lowlying negative parity states in <sup>136</sup>Cs that are not predicted by the other Hamiltonians or verified by experiment. These observations underscore the importance of testing model predictions in intermediate odd-odd nuclei for  $0\nu2\beta$  candidates. Under such requirement, the QX interaction may be considered less reliable and likewise disfavor  $^{136}$ Xe  $0\nu2\beta$ NME values determined with this Hamiltonian.

In the context of solar  $\nu_e$ /FDM detection in xenon-based detectors, this Letter presents the first unequivocal identification of the predicted long-lived excited 3<sup>+</sup><sub>1</sub> state in <sup>136</sup>Cs, with a firm spin-parity assignment. The measured excitation energy,  $E_x = 74$  keV, is more than 3 times higher than the shell-model prediction in Ref. [9]. Nevertheless, in the absence of competing branches [55], the 3<sup>+</sup><sub>1</sub> state at 74 keV is expected to still have a long enough lifetime for a feasible delayed coincidence tagging of solar  $\nu_e$ /FDM interactions on  $^{136}$ Xe. As this level can deexcite to the  $5_1^+$  ground state via both internal conversion (IC) and  $\gamma$ -ray emission, its total transition rate is proportional to  $E_{\gamma}^{5}(1+\alpha)$ , where  $\alpha$  is the IC coefficient [56]. Based on our measured energy and simple scaling arguments, the shell-model predicted lifetime of the state is ~280 ns, 3 times shorter than the value obtained with  $E_x = 23 \text{ keV } [9,20]$ .

In conclusion, we used  $^{138}\text{Ba}(d,\alpha)$  angular distribution measurements, together with shell-model calculations to report the location of possible metastable states with  $J \ge 1$  in the odd-odd <sup>136</sup>Cs nucleus. The new states observed in this Letter offer an opportunity for high background rejection, and open new possibilities for the detection of solar  $\nu_e$  events and/or FDM interactions in large xenon detectors. We unambiguously identify the first excited state in <sup>136</sup>Cs, which has spin-parity 3<sup>+</sup> and would decay to the 5<sup>+</sup> ground state via a slow E2 transition. Our findings are supported by a recent independent study [57] that measured the lifetime of the  $3_1^+$  state to be  $\tau = 157(4)$  ns. We also compare our experimental results with shell-model predictions made with three Hamiltonians that were previously used to evaluate the  $^{136}$ Xe  $0\nu2\beta$  NME. It is shown that one of the Hamiltonians (QX), which also systematically underestimates the NME compared with the others, fails to accurately describe the <sup>136</sup>Cs spectrum. As this inadequacy may have been obscured when predictions were compared with experimental data on even-even nuclei, one might disfavor  $0\nu2\beta$  results obtained with this Hamiltonian.

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- \*striambak@uwc.ac.za †Deceased.
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