High-Precision Spectroscopy of ²⁰O Benchmarking Ab Initio Calculations in Light Nuclei

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states were firmly established. From the γ -ray branching and E2/M1 mixing ratios for transitions

deexciting the 2_2^+ and 3_1^+ states, the B(E2) and B(M1) were determined. Various chiral effective field theory Hamiltonians, describing the nuclear properties beyond ground states, along with a standard USDB interaction, were compared with the experimentally obtained data. Such a comparison for a large set of γ -ray transition probabilities with the valence space in medium similarity renormalization group *ab initio* calculations was performed for the first time in a nucleus far from stability. It was shown that the *ab initio* approaches using chiral effective field theory forces are challenged by detailed high-precision spectroscopic properties of nuclei. The reduced transition probabilities were found to be a very constraining test of the performance of the *ab initio* models.

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Introduction.-Nuclear structure studies aim at understanding the properties of atomic nuclei based on nucleons interacting in the nuclear medium by combined strong, electromagnetic, and weak interactions. Chiral effective field theory (EFT) provides a framework for nuclear forces based on quantum chromodynamics which, together with ab initio many-body approaches, allows one to perform first-principle nuclear structure calculations including twoand three-nucleon forces in various regions of the Segré chart [1–4]. Previous studies of neutron-rich isotopes have proven to be especially suitable to establish advanced theoretical calculations based on chiral EFT forces. In particular, the neutron drip line for oxygen presents a strong anomaly with ²⁴O being the last bound isotope, whereas theoretical predictions positioned the drip line at doubly magic ²⁸O [5–9]. This puzzle was solved by the introduction of chiral EFT three-body forces [10]. These contributions have been studied extensively in subsequent works, especially in comparison with mass [11-16], charge radius [17–23], and electromagnetic moment [24–26] measurements of neutron-rich systems. The present challenge is to obtain unambiguous experimental measurement to compare to different *ab initio* calculations to improve their accuracy and predictive power. Electromagnetic transition probabilities play a major role in testing the quality of the chiral EFT interaction with *ab initio* approaches [4,27], since they are connected to the nuclear wave functions. The comparison between high-precision measurements in excited states and state-of-the-art ab initio calculations provides a sensitive probe of the nuclear structure details comparable to nuclear masses or charge radii. The isotopic chain of oxygen was identified as an ideal laboratory to benchmark state-of-the-art *ab initio* theory [1]. In neutronrich oxygen, the introduction of three-body forces induces a repulsion between the neutron $1s_{1/2}$ and $0d_{3/2}$ orbitals defining the drip line [10]. Detailed spectroscopy of these orbitals at the drip line remains an experimental challenge, but relevant information can be obtained in slightly less exotic nuclei, such as ²⁰O [28]. In this Letter, we present the spectroscopic study of non-yrast states in ²⁰O using stateof-the-art instrumentation.

Experimental details.—The ²⁰O nucleus was populated in the ${}^{19}O(d, p){}^{20}O^*$ direct reaction in inverse kinematics,

using a pure radioactive beam postaccelerated to 8 AMeV, with an average intensity of 4×10^5 pps, delivered by the SPIRAL1 accelerator complex in GANIL and impinged on a deuterated polyethylene target (CD_2) . Two types of targets were employed in the experiment: a 0.3-mg/cm²thick self-supporting CD₂ target for spectroscopy measurements and a 0.3-mg/cm²-thick target deposited on a 24.4-mg/cm²-thick Au backing (hereinafter mentioned as CD_2 and $CD_2 + Au$, respectively). Using the CD_2 target, detailed spectroscopy was performed and the $CD_2 + Au$ was used for measuring the lifetime of the populated excited states using the Doppler shift attenuation method (DSAM) [29]. The measurements were performed in triple coincidence: The beamlike recoils were detected in the VAMOS++ magnetic spectrometer [30] to reject the background coming from fusion-evaporation and fusionfission events, protons were measured at backward angles by the MUGAST array [31], and, at backward angles, the AGATA array [32] was employed for the detection of the γ rays emitted by the excited nucleus.

The coupling of these three instruments provides a large solid angle for detection of recoiling nuclei, a highprecision kinematic reconstruction, and a unique sensitivity for γ rays emitted in flight thanks to γ -ray tracking algorithms [33], resulting in unprecedented Doppler correction capabilities. This unique combination of direct reaction and state-of-the-art spectrometers allows one to perform a combined charged particle and γ -ray spectroscopy, along with the measurement of subpicosecond lifetimes. In particular, the MUGAST array allows for the selection on an event-by-event basis of the excited states directly populated in the final ²⁰O* nucleus and measurement of its velocity at the reaction moment for each of the populated states to perform a feeding-free, fully controlled, and high-accuracy lifetime measurement. More details on the experimental apparatus and data analysis procedure can be found in Ref. [31].

Spectroscopic study.—The energy and angle of the protons detected in MUGAST allowed for the reconstruction of the ²⁰O excitation energy spectrum using NPTool [31,34] (see also Supplemental Material [35]). The ground state, the 2_1^+ at 1.67 MeV, the 4_1^+ at 3.55 MeV, the 2_2^+ at 4.07 MeV, and 3_1^+ states at 5.23 MeV were observed. Moreover, two



FIG. 1. Two-dimensional correlation between the ²⁰O excitation energy on the *y* axis and the corresponding γ -ray decays on the *x* axis. The transitions from which the lifetimes are extracted are highlighted in red. The weakest transitions depopulating the $2^+_{2,3,4}$ states are marked with black boxes.

additional states, already identified in [41] and both tentatively assigned as $J^{\pi} = (2^+_{3,4})$, were observed. Excited states above the neutron separation threshold in ²⁰O were populated [42]. The correlation between the excitation energy of ²⁰O and the emitted γ rays is shown in Fig. 1. The transitions used in the line-shape analysis for the lifetime extraction are highlighted in red in the figure.

The prompt Doppler-corrected tracked γ -ray spectrum measured in AGATA in triple coincidence using the CD₂ target is shown in Fig. 2. The decays of the 2_1^+ , 4_1^+ , 2_2^+ , and 3_1^+ excited states are clearly visible. The reconstruction of the level scheme obtained via particle- γ spectroscopy combining MUGAST and AGATA is reported in detail in [42–44]. The complete spectroscopic information is summarized in Supplemental Material [35]. The branching ratios of the $2_2^+ \rightarrow 2_1^+$ and $2_2^+ \rightarrow 0_1^+$ transitions were measured to be 0.88(1) and 0.12(1), respectively. For the $3_1^+ \rightarrow 2_2^+$ and $3_1^+ \rightarrow 2_1^+$ transitions, the measured branching ratios were 0.28(1) and 0.72(1), respectively.

Lifetime measurements.—Previous experiments provided the lifetime measurements of the 2_1^+ [$\tau = 10.5(4)$ ps [45]]



FIG. 2. Prompt γ -ray spectrum following the ¹⁹O(d, p)²⁰O reaction measured in AGATA in coincidence with MUGAST and VAMOS++.

and 2_2^+ ($\tau = 150_{-30}^{+80}$ fs [28]) states. In the present work, lifetimes were extracted by fitting the line shape of the transitions in the γ -ray spectra of the CD₂ + Au dataset with realistic Monte Carlo simulations. The simulations have been performed using the AGATA GEANT4 simulation code [46], that includes the geometry of the array and the reaction event with the emission of the beam- and targetlike particles as well as γ rays. Line-shape analysis based on Monte Carlo simulations for AGATA was already performed in the femtosecond range [47–49], proving the capabilities of the apparatus in this range of lifetimes.

The simulation has been optimized by adjusting the parameters corresponding to the response function of the detectors at the time of the experiment to reduce the sources of systematic errors. The velocity distributions at reaction point were measured and used as an input of the simulation of the decay for each of the investigated states. The reproduction of the energy loss in the target and degrader was tested for both CD_2 and $CD_2 + Au$ datasets on the $2_1^+ \rightarrow 0_1^+$ transition. The energy and full width at half maximum of the transition were in agreement within the detector resolution. The comparison is provided in Supplemental Material [35] (see also Refs. [36–38] therein).

The lifetime of the 2_2^+ state was extracted by fitting the line shape of the simulated $2_2^+ \rightarrow 2_1^+$ transition to the experimental one, obtained by requiring the coincidence with the 2_2^+ state in the excitation energy and, thus, removing the influence of feeders. The simulations were performed by varying two parameters: the energy of the transition at rest and the lifetime of the 2_2^+ state. The minimum was attested at 70(10) fs [70(14) fs for 90% confidence]. In Fig. 3, the red hatched area represents the simulation output within the 1σ limit. The two-dimensional χ^2 surface is shown in Supplemental Material [35] (see also Refs. [39,40] therein). To extract the lifetime of the 3_1^+ state, the $3_1^+ \rightarrow 2_1^+$ transition at 3552.6 keV was chosen. Similarly to the procedure for the 2_2^+ state, the simulated spectrum was fitted to the experimental one after gating on



FIG. 3. Comparison between experimental data and simulation for the lifetime of the 2^+_2 state in ²⁰O.

the excitation energy of the 3_1^+ state. Using the least- χ^2 procedure, a lifetime of $\tau = 54(9)$ fs was obtained [54(12) fs for 90% confidence].

Discussion.-The present measurement confirms the short lifetime of the 2^+_2 state and the conclusion drawn in [28] in spite of tension between the results of the two experiments. The origin of the discrepancy is not completely understood. The computation of a longer lifetime can arise from systematical effects in the initial velocity distribution model or unobserved side-feeding contribution. Such systematic errors are canceled in our improved experimental approach. The use of the (d, p) reaction combined to a thin target layer where ²⁰O is produced, followed by an optimized gold thick foil to develop the DSAM profile, allows one to determine, on an event-byevent basis, the entry point in the recoil from the measured excitation energy and the initial velocity distribution for each state used to extract the lifetimes. The reduced transition probabilities have been extracted from the measured transition energies, branching ratios, and lifetimes and are reported in Table I. A measured value of the mixing ratio $\delta(E2/M1)$ for the $2^+_2 \rightarrow 2^+_1$ transition reported earlier is -0.18(8) [50]. The mixing ratios of the $3^+_1 \rightarrow 2^+_{1,2}$ transitions are experimentally unknown, preventing us to extract model-independent transition probabilities.

The present experimental results have been compared to *ab initio* calculations using the valence-space in-medium similarity renormalization group (VS-IMSRG) [52]. The calculations were performed at the VS-IMSRG(2) level, building an effective shell-model Hamiltonian for ²⁰O in the $0d_{5/2}$, $1s_{1/2}$, $0d_{3/2}$ configuration space for protons and neutrons. The *E*2 and *M*1 transition operators were evolved consistently with the VS-IMSRG keeping up to two-body operators, but meson-exchange currents, explored for *M*1

TABLE I. Comparison between experimental, shell-model (USDB), and *ab initio* [N³LO_{*lnl*}, 1.8/2.0(EM), N²LO_{*GO*}] transition probabilities. The *B*(*E*2)'s are given in e^2 fm⁴ and the *B*(*M*1)'s in μ_N^2 . The experimental transition probabilities of the $3_1^+ \rightarrow 2_1^+$ ($3_1^+ \rightarrow 2_2^+$) have been calculated using a theoretical mixing ratio of $\delta = 0.13$ ($\delta = 0.019$), obtained from USDB. The binding energy with respect to ¹⁶O is presented in MeV.

	Exp.	USDB	N ³ LO _{lnl}	1.8/2.0(EM)	$N^2 LO_{GO}$
$B(E2; 2^+_1 \to 0^+_1)$	5.9(2)	3.25	0.79	0.89	0.80
$B(E2; 2^+_2 \to 0^+_1)$	1.3(2)	0.77	0.21	0.20	0.26
$B(E2; 2_2^+ \to 2_1^+)$	4(2)	0.0005	0.089	0.070	0.18
$B(M1;2^+_2\rightarrow 2^+_1)$	0.05(2)	0.019	0.014	0.017	0.12
$B(E2;3^+_1 \rightarrow 2^+_1)$	0.32(7)	0.57	0.16	0.17	0.17
$B(M1;3^+_1 \rightarrow 2^+_1)$	0.016(4)	0.029	0.023	0.028	0.0089
$B(E2; 3_1^+ \to 2_2^+)$	0.7(2)	1.24	0.14	0.15	0.11
$B(M1;3^+_1\to2^+_2)$	0.19(4)	0.32	0.53	0.55	0.56
Binding energy	-23.74 [51]	-23.63	-19.67	-20.51	-22.71

transitions in very light nuclei [53–55], were not included. As a starting point, three well-established nuclear Hamiltonians based on chiral EFT with three-nucleon forces were used: (i) 1.8/2.0(EM) [56,57], which reproduces very well ground-state energies up to heavy nuclei [3,58] and was used for ²⁰O in Ref. [28]; (ii) the more recent N³LO_{*lnl*} [59] and (iii) N²LO_{GO} [60], which includes explicit Δ -isobar degrees of freedom. In addition, standard shell-model calculations using the USDB interaction [61] in the same configuration space were performed. Unlike in the VS-IMSRG calculations, the bare *M*1 and *E*2 operators and, therefore, neutron effective charges $e_n = 0.4$ [62] were used along with the USDB interaction. For the VS-IMSRG and configuration-interaction calculations, the codes IMSRG++ [63] and KSHELL [64] were used, respectively.

Figure 4 compares the experimental low-lying excitation spectra of ²⁰O with the results of the theoretical calculations. Additionally, the experimental and calculated level scheme of ¹⁹O are also shown in Supplemental Material [35]. The excitation energies obtained in the ab initio approaches and the shell model are, in general, in good agreement with experiment, within hundreds of keV. The 1.8/2.0(EM) and USDB results are in the best agreement with the data. We emphasize the good agreement for the $1/2^+$ excited state corresponding to the $(d_{5/2})^2(s_{1/2})^1$ configuration in ¹⁹O. Consistently, the evolution of its excitation energy, for different chiral EFT Hamiltonians, is correlated to the $2^+_1 \rightarrow$ 2^+_2 energy difference in ²⁰O (Supplemental Material [35]). It should be noted that there was no nuclear structure information on excitation energies, etc., in oxygen or similar systems used for the derivation of the chiral EFT Hamiltonians in *ab initio* approaches, while the shell model USDB interaction resulted from the fit to the selected nuclear structure data. The calculated wave functions of the states observe two main structures: the 0_1^+ , 2_1^+ , and 4_1^+ yrast states are mainly due to the neutron $(0d_{5/2})^4$ configuration, while the 2^+_2 and 3^+_1 states are dominated by the $(0d_{5/2})^3(1s_{1/2})^1$ configuration. The color code on the level shows the amplitude of the main configurations ($\geq 10\%$). The ab initio and shell-model calculations are in good agreement, but the shell model suggests more fragmented wave functions.

The mixing ratio of transitions from the 3_1^+ state are experimentally unknown. For the following discussion, the theoretical values obtained using the USDB [61] interaction were used to obtain the corresponding B(E2) and B(M1). The experimental and theoretical reduced transition probabilities are presented in Table I and reported in Fig. 4. The B(E2) between the 2_2^+ state and the 0_1^+ and 2_1^+ states, and from the 3_1^+ state, were found experimentally small, consistently with their single-particle character, interpreted as $(0d_{5/2})^4 \rightarrow (0d_{5/2})^3(1s_{1/2})^1$ single-particle transition.

The B(E2) values are systematically underestimated in the *ab initio* calculations, as already observed in Ref. [62]



FIG. 4. Experimental ²⁰O excited states compared to theoretical USDB shell-model calculations and VS-IMSRG results obtained with three different Hamiltonians. For theoretical states, the leading configurations are reported with a color code for each level: $(d_{5/2})^4$ (black), $(d_{5/2})^3(s_{1/2})^1$ (green), and $(d_{5/2})^2(s_{1/2})^2$ (violet). Other configurations are shown in gray. The colored bar length is proportional to each contribution in the wave function. The measured and calculated B(E2)s (blue) and B(M1)s (red) are also reported proportionally to the arrow thickness.

for ²¹O and discussed first in Ref. [65] and recently in much detail in Refs. [66,67]. The likely reason is the restriction to the VS-IMSRG(2) level, which leaves many-particlemany-hole correlations out of the evolved VS-IMSRG operator. With a neutron effective charge $e_n = 0.4$, USDB B(E2) results present a good agreement with experimental values, in particular, those involving the 3^+_1 state. An even better agreement for some transitions is observed in USDB calculations using an effective charge $e_n = 0.5$. However, high-precision spectroscopy data reveal that simple effective charges in the shell model seem not to reproduce all transitions simultaneously (see also the discussion in Ref. [62]) and a more sophisticated treatment would be desirable. In addition, the $B(E2; 2_2^+ \rightarrow$ 2_1^+) is underestimated by orders of magnitude in all the models, which suggests some deficiency of the wave functions. It could be related to the limited configuration space or to the insufficient configuration mixing. The latter is consistent with the results of the measurements of the spectroscopic factor (S) by Hoffman et al. [41]. The cross sections for 0^+_1 and 4^+_1 states were computed with L=2transfer and large S factor obtained are compatible with the occupancy of the d orbitals only. Similarly, the cross section for 2^+_2 state is dominated by L = 0 transfer consistent with the single-particle excitation into the $1s_{1/2}$ neutron orbital. In contrast, the cross section for the 2_1^+ state was obtained with large contributions of L = 0(S = 0.19) and L = 2 transfers (S = 0.43) corresponding to a more fragmented wave function of the 2^+_1 state.

The theoretical B(M1) reduced transition probabilities for the $2_2^+ \rightarrow 2_1^+$ transition are about a factor of 3 smaller than those obtained in the experiment, with a small difference between USDB and 1.8/2.0(EM) and lower values for the other two VS-IMSRG calculations. The B(M1) for the 3^+_1 state are in reasonably good agreement with the experiment, especially for USDB, where there is agreement within $(1-2)\sigma$. The three chiral EFT Hamiltonians well reproduce the $3_1^+ \rightarrow 2_1^+ B(M1)$ value, but they overestimate the $3_1^+ \rightarrow 2_2^+$ reduced transition probability by about a factor of 2. Overall, for the M1 transitions there is a better agreement for the phenomenological USDB interaction. The *ab initio* results are of similar quality, with a slight preference for 1.8/2.0(EM)over the other two chiral EFT Hamiltonians. The agreement may improve when including meson exchange currents. Hence, the measurements of the B(M1) reduced transition probabilities appear to be very pertinent for testing *ab initio* calculations based on chiral EFT Hamiltonians. It should be noted that the short lifetime measured in this work (lower than 100 fs) for the 2^+_2 state is incompatible with having at the same time a low B(E2) and a low B(M1), in the range of the theoretical predictions, for the $2^+_2 \rightarrow 2^+_1$ transition.

Conclusion.—The lifetimes of the 2^+_2 and 3^+_1 excited states in ²⁰O were measured by means of the DSAM technique via the direct (d, p) reaction in inverse kinematics using a radioactive postaccelerated beam of ¹⁹O. A feeding-free lifetime for the 2^+_2 and the 3^+_1 states was extracted. For the first time in the key isotopic chain of oxygen, all spectroscopic observable obtained for yrast and non-yrast excited states in the neutron-rich ²⁰O were compared simultaneously to the results of ab initio calculations using chiral EFT forces and provide the results in reasonable agreement with the experimental data. The reduced transition probabilities, B(M1) and B(E2), in particular, provide a very constraining test of the performance of the *ab initio* models. Many improvements in ab initio calculations are still to be envisaged like including meson exchange currents or many-particle-many-hole correlations by releasing the restriction to the VS-IMSRG(2) level, so that the predictive power can reach and exceed that of the conventional phenomenological shell-model approaches. The comparison between data and predictions will be strengthened by systematic estimation of the theoretical uncertainties. This observation serves as motivation for future endeavors aimed at quantifying uncertainties in VS-IMSRG calculations. This work paves the way for lifetime measurements in exotic nuclei using next-generation radioactive beam facilities under construction worldwide to be compared with state of the art *ab initio* calculations.

The supporting data for this article are from the e775s experiment and are registered [68] following the GANIL data policy.

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