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RECENT ADVANCES IN LASER SPECTROSCOPY AT ISOLDE*

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The production of rare isotopes at facilities such as ISOLDE is often compromised by the presence of intense isobaric contaminant beams. This has necessitated extensive research and development into cleaner production methods, such as the laser ion source, as well optimization of target materials and design. In a few special cases, it has been possible to use these techniques to even produce isomeric beams for spectroscopy and post acceleration. Such selectivity is not possible for all elements and there are still many cases prevented by isobaric contamination. During the last three years a new laser spectroscopy experiment CRIS has been established at ISOLDE that aims to produce a universal method of isotope purification for secondary experiments, such as nuclear spectroscopy. This new technique offers an efficient method for selecting either the ground state or long lived isomeric state > 1 ms.

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1. Introduction

The frontiers of various physics research areas are often defined by the limits of resolution or sensitivity. This is perhaps most striking in the discipline of modern optics. Since Newton first dispersed solar radiation into a spectrum of colours, large improvements in resolution have revealed new physics and paradigm shifts in our understanding. The dark lines in the solar spectrum discovered by Fraunhofer took almost a century and the development of quantum mechanics to be fully understood. In 1924, Pauli first suggested the existence of nuclear spin and the associated atomic hyperfine structure (hfs) that would result [1], which was soon after experimentally

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verified [2–4]. The hyperfine states resulting from the coupling of the nuclear spin to the electronic total angular momentum are non-degenerate if the nucleus has a non-zero magnetic dipole moment or electric quadrupole moment. By probing this hfs with optical techniques it is possible to measure the nuclear moments and spin without introducing nuclear model dependence. When these optical measurements are extended across a chain of isotopes, the change in electronic binding energy associated with changes in the mass and nuclear charge distribution (isotope shift) can be studied. With even higher resolution techniques the distribution of nuclear magnetism [5, 6] and magnetic octupole moments can be determined [7].

State-of-the-art on-line facilities, such as ISOLDE, are now pushing rare isotope production to the very limits of nuclear existence. Through innovative new techniques, such as collinear resonant ionization spectroscopy (CRIS) it will soon be possible to further test nuclear theories at the extremes of isospin with optical measurements.

There exists a large variety of laser spectroscopy techniques for rare isotope research [8]. These can be broadly categorized into three subgroups: in-source, collinear beams and trapped. The majority of radioactive isotopes measured with laser spectroscopy have been achieved with either in-source or collinear beams techniques. There has been a continuous programme of research and development in both techniques at ISOLDE [9–13]. Since the last Zakopane conference in 2010, collinear experiments at ISOLDE have extended the beryllium charge radii to include ^{12}Be [14] and studied the charge radii of magnesium isotope chain [15]. The copper isotopes from $^{58-78}$Cu [16–19] and gallium isotopes from $^{63-82}$Ga [20–24] have been studied. The neutron deficient polonium isotopes have been studied down to ^{191}Po [25, 26], permitting alpha and beta-spectroscopy to be performed on ^{195}Po [27] and ^{199}Po [26].

2. Collinear resonant ionization spectroscopy

In-source laser spectroscopy utilizes resonant ionization spectroscopy (RIS), which step-wise excites atoms through one or more resonant transitions to a state from which the atom can be ionized by the application of a DC electric field infra-red radiation (for Rydberg states) or with an intense laser pulse [28]. The process of non-resonant excitation into the continuum has typical cross-sections that are more than seven orders of magnitude smaller than resonant atomic transitions. Excitation to an autoionizing state (if one exists for the particular element) increases the cross-section and is, therefore, relatively easier to saturate. Of the existing techniques, in-source laser spectroscopy is currently the most sensitive and has demonstrated the ability to measure isotopes with production yields down to 1 atoms/s for ^{182}Pb [29] and 0.01 atoms/s for ^{191}Po [30]. The technique is limited in resolution by the Doppler broadening of the transition linewidth (3–4 GHz), which is associated with the thermal motion of the atoms in the hot ionizer-cavity ($\sim 2000°$C). Collinear laser spectroscopy overlaps the laser with an ion beam that has been accelerated from the ion source to 30–60 keV [31]. Since the energy spread of the beam is conserved under acceleration, the product of velocity and velocity spread must also be conserved resulting in a compression of the Doppler broadening by three orders of magnitude at typical acceleration energies. Exciting an in-flight ensemble permits many variations on the basic concept such as β-NMR, coincidence and bunched beam techniques [8]. These techniques are typically limited by relatively low detection efficiency of resonantly scattered light and/or large background counting rates (in the case of β-NMR) which have, in general, prevented measurements on cases with yields below 100 ions/s [32].

To overcome these limitations in collinear laser spectroscopy, a novel method that performs RIS in a collinear geometry (CRIS) was developed off-line by Kudriavtsev and Letokhov in 1982 [33]. CRIS has potentially the same experimental efficiency as in-source laser spectroscopy and due to the suppression of Doppler broadening, a resolution down to a few MHz. The technique is schematically presented in Fig. 1. The radioactive ion beam is first mass separated and then cooled and bunched in a gas filled linear Paul trap (ISCOOL) [34]. By introducing a bunched structure to the beam, it is possible to avoid duty cycle losses associated with using pulsed lasers to ionize the ensemble. The ISCOOL cooler/buncher at ISOLDE can trap ions for 5 ms to 1 s, with typical bunch temporal widths of between 1–6 μs and up to 10^7 ions per bunch [35]. The bunched structure also reduces any background associated with the dark counting rate of the ion detector by the ratio of the trapping time to the temporal bunch width, identical to the bunched beam method used in fluorescence detection [36]. The bunched

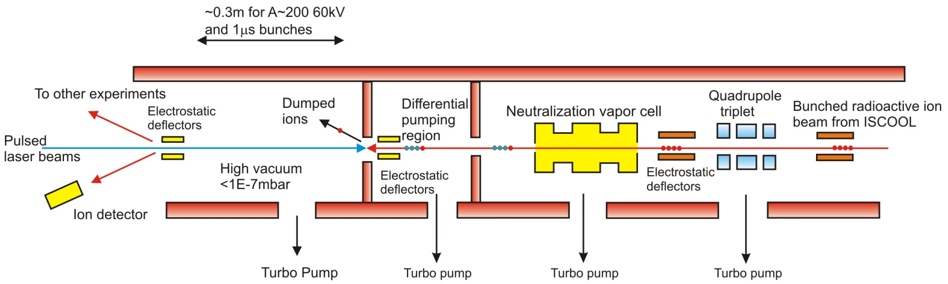

Fig. 1. (Colour on-line) Schematic drawing of the CRIS method. The interaction region has a length of 1.2 m. The neutralization vapour cell is filled with potassium and heated to 150°C. A detailed description can be found in reference [37].

beam is then neutralized in an alkali vapour cell before it is transported to the laser-atom interaction region. A scanning voltage is applied to the cell to accelerate (or decelerate) the ion bunch before neutralization, which Doppler tunes the atoms onto resonance with the laser. The pressure within the interaction region must be kept below 10^{-7} mbar in order to limit the non-resonant collisional ionization rate. This is achieved by placing a differential pumping region after the alkali vapour cell. The non-neutralized fraction of the beam is deflected within this differential pumping section. The resonantly produced ions are deflected at the end of the interaction region and transported to a charge particle detector such as a microchannel plate (MCP). A detailed description of the set up is given in Ref. [37].

The ultra high sensitivity of the CRIS method is due to three key aspects: high ionization efficiency of RIS schemes (up to 27% [9]), high detection efficiency of the resonant ions (close to 100%) and a very low rate of non-resonant ionization. The non-resonant ionization rate at 10^{-9} mbar is estimated to be lower than 10^{-6} per meter of the interaction. For rare isotopes produced without isobaric contamination, this equates to background free detection. If isobaric contamination is present at 10^7–10^8 particles per second it will be suppressed to just a few ions/s in the detection region. Previous attempts at CRIS have never combined all of three parts simultaneously. The ytterbium isotope chain was studied using the CRIS method 20 years ago and demonstrated the ability to detect resonant ions with yields down to 10^7 s^{-1} while suppressing the isobaric contamination by a factor of more than 10^8 [38]. However, this first on-line experiment suffered from a low experimental efficiency due to duty cycle losses and low metastable state population in the neutralization process. An off-line test of the CRIS technique on ^{27}Al at the IGISOL facility in 2001 utilized the then recently commissioned gas-filled linear Paul trap [39] to avoid duty cycle losses [40]. This test demonstrated an experimental efficiency of 1:30, an improvement compared to resonant fluorescence detection of ^{27}Al by more than three orders of magnitude. The overall sensitivity was limited by the poor vacuum in the collinear beam line during this test ($\sim 10^{-5}$ mbar), which resulted in a non-resonant ionization rate of almost 1:40. The CRIS beam line [37] at ISOLDE was designed and constructed to over come these previous difficulties and fully realize the sensitivity of the method first proposed 30 years ago [33].

3. Status of the CRIS project

The experiment was proposed to and accepted by the ISOLDE and Neutron Time-of-Flight Experiments Committee (INTC) in Feburary 2008. This proposal aims to study the neutron deficient isotopes $^{201-206}$Fr and shortlived isotopes ^{218,219}Fr [41]. The major parts of the beam line were designed and constructed at the University of Manchester and installed into the ISOLDE hall in April 2009. During 2009 the vacuum system (provided by KU Leuven) and the differential pumping effect were tested, which demonstrated a factor of up to 1000 isolation between the alkali vapour cell and the interaction region. The ion optics, MCP detector and alkali vapour cell were installed and tested during 2010. A test beam time in 2010 demonstrated a neutralization efficiency of $\sim 50\%$ using radioactive isotopes of rubidium with a vapour of potassium heated to 150°C. The first on-line commissioning run was carried out in November 2011, which successfully detected resonant ionization of ^{207}Fr and demonstrated a suppression of non-resonant ionization by a factor of less than 10^{-4} m^{-1} at 10^{-8} mbar. During the commissioning experiment a pulsed 1.4 GeV proton beam impinged onto a UC$_x$ target, which produced the neutron deficient francium isotopes through spallation reactions. The atoms were ionized within a rhenium surface ionizer cavity and subsequently accelerated to 30 keV. A frequency-doubled injection-seeded Ti:sapphire laser system [42] was used to generate laser light at 422.7 nm to excite the $7s(^2S_{1/2}) - 8p(^2P_{3/2})$ transition in the francium atom. The fundamental wavelength from a Nd:YAG laser (1064 nm) was used to ionize the excited state of francium. The second harmonic of the Nd:YAG laser was used to pump the injection-seeded Ti:sapphire laser system. A sample spectrum of ^{207}Fr is shown in Fig. 2. This commissioning

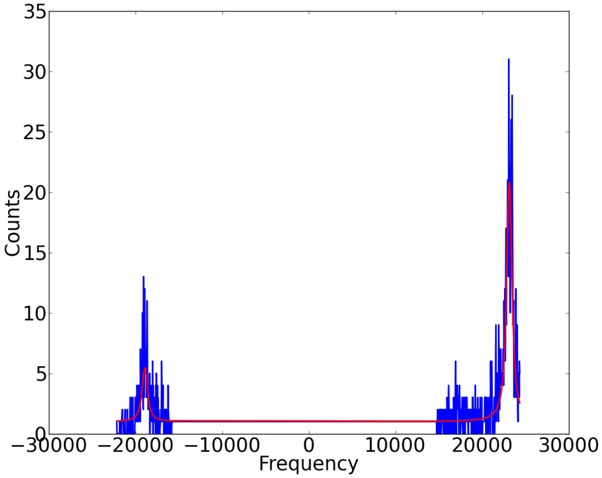

Fig. 2. (Colour on-line) First collinear resonant ionization spectrum of ^{207}Fr, shown with a fit to the data. The zero frequency position was arbitrary defined. From this spectrum the A-factor of the $^2S_{1/2}$ state was determined to be 8390(100)[200] MHz and compares closely to literature value of $A(S_{1/2}) = 8484(1)$ MHz [43].

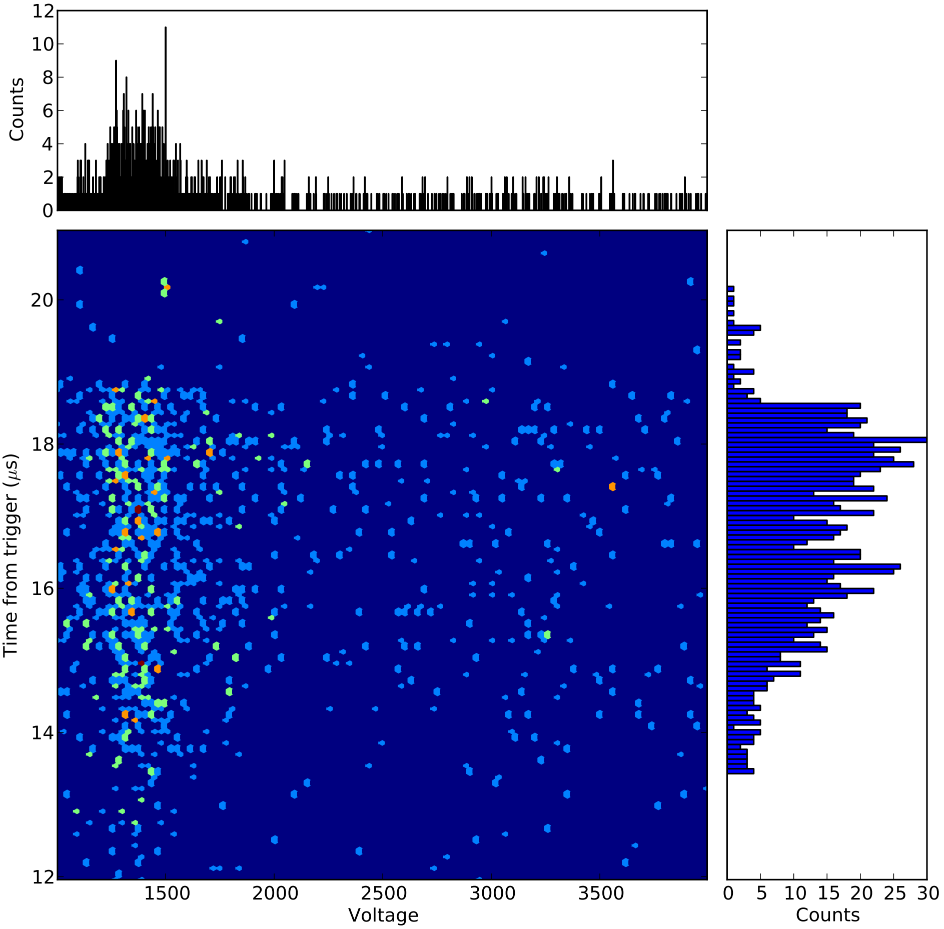

Fig. 3. (Colour on-line) Time projection plot of the voltage scan of ^{207}Fr shown. In the 2011 commissioning experiment the CRIS data acquisition system recorded data event-by-event, allowing the voltage scan and time-of-flight to be studied simultaneously.

run detected ^{207}Fr with a total experimental efficiency of 10^{-6}. It was estimated that a factor of 10^3–10^4 could be accounted for by a low fluence of photons in the non-resonant ionization step, noise in the MCP and poor transmission associated with the misalignment of the alkali vapour cell, which introduces a steering effect when a tuning voltage is applied. The signal detected by the MCP was digitized by a LeCroy WavePro 725Zi oscilloscope and time stamped with respect to the master trigger signal from the Nd:YAG laser. The time projection spectrum of the high energy peak in Fig. 2 is shown in Fig. 3, where the projections of the voltage scan and time-of-flight are presented. The interaction region in the CRIS beam line is 1.2 m in length, which requires that the temporal bunch length is kept below 7 μs for $A = 200$ at 30 keV, so that an entire bunch can be positioned within the interaction region when the pulsed laser fires. It can be seen in Fig. 3 that there is a cut off in the resonant signal beyond 18.9 μs, which corresponds to the part of the atomic bunch being located before the electrostatic deflector in the differential pumping section (see Fig. 1) when lasers are present.

During 2012 the source of the noise in the MCP signal was located and eliminated. The laser system for the resonant step was changed to a narrow band width Ti:sapphire laser (600 MHz) pumped with a 10 kHz Nd:YAG laser [10]. The 422.7 nm laser light was fiber coupled from the RILIS cabin and synchronized to the 30 Hz Nd:YAG laser used to non-resonantly ionize the francium beam. A preliminary test beam time has demonstrated a total experimental efficiency of better than 1:400 for ^{202}Fr and a suppression of non-resonant ionization by $\sim 3 \times 10^5$.

4. Isomer selective decay spectroscopy

The multi-step resonant ionization process can efficiently select one isotope (or long-lived nuclear state) from another due to the fact that the transitions of each isotope are modified by changes in the hfs and isotope shift. This selectivity can be defined by the ratio of the separation of resonances to the linewidth of the resonances. A selectivity of greater than 10^4 per step in a RIS scheme is possible. The total selectivity of a multi-step ionization scheme is the product of the individual selectivity of each step. The CRIS method introduces an additional kinematic shift between isotopes, which further enhances the selectivity. A selectivity of greater than 10^{14} is possible, making CRIS a comparable technique to trace analysis methods such as accelerator mass spectrometry [44]. This permits the CRIS method to be used to purify a contaminated beam or select nuclear isomeric state for decay spectroscopy experiments [45]. In-source laser spectroscopy has been used previously to produce isomeric beams of ^{68,70}Cu for Coulomb excitation, mass spectrometry and decay studies [46]. During this previous work the Doppler broadening within the ion source limited the achievable selectivity between nuclear states and there was almost 50% isobaric contamination of gallium.

The CRIS technique can be used to purify the beam and suppress isobaric contamination as well as the ground state by more than a factor of 10^6. A particular interesting area of investigation is the decay spectroscopy of low energy isomeric states that cannot be easily separated by other techniques such as trap assisted decay spectroscopy [47, 48]. The measurement of additional hyperfine components with laser spectroscopy is a unique observable for discovering new isomeric states. An example of one such case is the isomer in ^{80}Ga which was discovered with laser spectroscopy, while previously missed by Penning trap mass spectrometry and β-decay studies [21, 49, 50]. A new decay spectroscopy station has been constructed to demonstrate the technique of isomer selection with CRIS (see Fig. 4) [51]. The first physics case for this device will study the high-spin isomeric states in ^{202,204,206}Fr [52]. The CRIS method will be used to produce pure isomeric beams, which will be transported to a decay spectroscopy station [51].

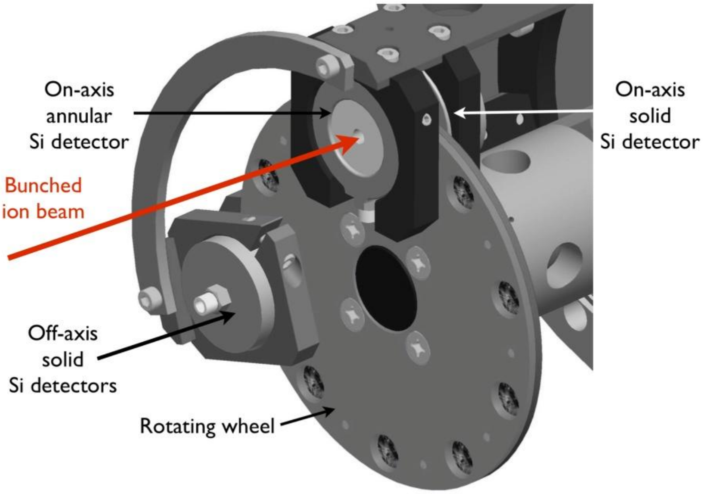

Fig. 4. (Colour on-line) The new decay spectroscopy set up on the CRIS beam line. The radioactive beam is transported through an annular Si PIPS detector to a carbon foil mounted on rotatable wheel. A standard PIPS detector is located behind the carbon foil position. There are ten carbon foil positions on the wheel. A pair of Si PIPS detectors are positioned off axis to study longer-lived states.

5. Summary

The CRIS experiment at ISOLDE has been constructed and commissioned. The first run measured the hfs of ^{207}Fr with a total experimental efficiency of 1:10^6. A second test beam time has improved this to 1:400 with a non-resonant ionization rate per interaction meter of 3×10^{-6} m^{-1}. A new decay spectroscopy station has been commissioned to demonstrate the technique of isomer selection with CRIS.

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REFERENCES

[1] W. Pauli, *Naturwiss* **12**, 741 (1924).

[2] H. Nagaoka, T. Mishima, *Proc. Imp. Acad.* **2**, 249 (1926).

[3] E. Back, S. Goudsmit, *Z. Phys.* **43**, 321 (1927).

[4] E. Back, S. Goudsmit, *Z. Phys.* **47**, 174 (1928).

[5] A. Bohr, V.F. Weisskopf, *Phys. Rev.* **77**, 94 (1950).

[6] H.H. Stroke, R.J. Blin-Stoyle, V. Jaccarino, *Phys. Rev.* **123**, 1326 (1961).

[7] V. Jaccarino, J.G. King, R.A. Satten, H.H. Stroke, *Phys. Rev.* **94**, 1798 (1954).

[8] B. Cheal, K.T. Flanagan, *J. Phys. G* **37**, 113101 (2010).

[9] B.A. Marsh *et al.*, *Hyperfine Interact.* **196**, 129 (2010).

[10] S. Rothe *et al.*, *J. Phys. Conf. Ser.* **312**, 052020 (2011).

[11] V.N. Fedosseev *et al.*, *Rev. Sci. Instrum.* **83**, 02A903 (2012).

[12] K.T. Flanagan, *AIP Conf. Proc.* **1377**, 38 (2011).

[13] W. Nörtershäuser, *Hyperfine Interact.* **198**, 73 (2010).

[14] A. Krieger *et al.*, *Phys. Rev. Lett.* **108**, 142501 (2012).

[15] D.T. Yordanov *et al.*, *Phys. Rev. Lett.* **108**, 042504 (2012).

[16] K.T. Flanagan *et al.*, *Phys. Rev.* **C82**, 041302 (2010).

[17] P. Vingerhoets *et al.*, *Phys. Rev.* **C82**, 064311 (2010).

[18] P. Vingerhoets *et al.*, *Phys. Lett.* **B703**, 34 (2011).

[19] U. Köster *et al.*, *Phys. Rev.* **C84**, 034320 (2011).

[20] B. Cheal *et al.*, *Phys. Rev. Lett.* **104**, 252502 (2010).

[21] B. Cheal *et al.*, *Phys. Rev.* **C82**, 051302 (2010).

[22] E. Mané *et al.*, *Phys. Rev.* **C84**, 024303 (2011).

[23] T.J. Procter *et al.*, *Phys. Rev.* **C86**, 034329 (2012).

[24] B. Cheal *et al.*, *J. Phys. Conf. Ser.* **381**, 012071 (2012).

[25] T.E. Cocolios *et al.*, *Phys. Rev. Lett.* **106**, 052503 (2011).

[26] T.E. Cocolios *et al.*, *J. Phys. Conf. Ser.* **381**, 012072 (2012).

[27] T.E. Cocolios *et al.*, *J. Phys. G* **37**, 125103 (2010).

[28] V.N. Fedosseev, Yu. Kudryavtsev, V.I. Mishin, *Phys. Scr.* **85**, 058104 (2012).

[29] H. De Witte *et al.*, *Phys. Rev. Lett.* **98**, 112502 (2007).

[30] M. D. Seliverstov *et al.*, submitted to *Phys. Lett.* **B**, 2012.

[31] W.H. Wing, G.A. Ruff, W.E. Lamb, J.J. Spezeski, *Phys. Rev. Lett.* **36**, 1488 (1976).

[32] K.T. Flanagan *et al.*, *J. Phys. G* **39**, 125101 (2012).

[33] Yu.A. Kudriavtsev, V.S. Letokhov, *Appl. Phys.* **B29**, 219 (1982).

[34] H. Frånberg *et al.*, *Nucl. Instrum. Methods Phys. Res. B* **266**, 4502 (2008).

[35] E. Mané *et al.*, *Eur. Phys. J.* **A42**, 503 (2009).

[36] P. Campbell *et al.*, *Eur. Phys. J.* **A15**, 45 (2002).

[37] T.J. Procter *et al.*, *J. Phys. Conf. Ser.* **381**, 012070 (2012).

[38] C. Schulz *et al.*, *J. Phys. B* **24**, 4831 (1991).

[39] A. Nieminen *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **469**, 244 (2001).

[40] K.T. Flanagan, Ph.D. Thesis, University of Manchester, UK, 2004.

[41] J. Billowes *et al.*, "Collinear Resonant Ionization Laser Spectroscopy of Rare Francium Isotopes", CERN-INTC-2008-010.

[42] M. Hori, A. Dax, A. Soter, *Hyperfine Interact.* **212**, 179 (2012).

[43] A. Coc *et al.*, *Phys. Lett.* **B163**, 66 (1985).

[44] L.K. Fifield, *Rep. Prog. Phys.* **62**, 1223 (1999).

[45] V.S. Letokhov, *Opt. Commun.* **7**, 59 (1973).

[46] I. Stefanescu *et al.*, *Phys. Rev. Lett.* **98**, 122701 (2007).

[47] S. Rinta-Antila *et al.*, *Eur. Phys. J. A* **31**, 1 (2007).

[48] M. Kowalska *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **689**, 102 (2012).

[49] J. Hakala *et al.*, *Phys. Rev. Lett.* **101**, 052502 (2008).

[50] P. Hoff, B. Fogelberg, *Nucl. Phys.* **A368**, 210 (1981).

[51] K.M. Lynch *et al.*, *J. Phys. Conf. Ser.* **381**, 012128 (2012).

[52] M. Huyse *et al.*, *Phys. Rev.* **C46**, 1209 (1992).