CERN-EP/80-127

July 15, 1980

APPLICATION OF ON-LINE ABMR TECHNIQUES TO NUCLIDES FAR FROM STABILITY

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ABSTRACT

The possibilities and limitations of the atomic-beam magnetic resonance (ABMR) method in hyperfine structure measurements on nuclides far from stability are discussed with respect to nuclear and atomic properties of the nuclides to be studied, half-lives, transmission of the apparatus, oven and detector efficiencies, etc. The ABMR apparatus, connected on-line with the ISOLDE isotope separator is described, and the elements and production yields available at ISOLDE are compared with the requirements of the ABMR method.

Geneva, July 1980

Contribution to the 10th International Conference on Electromagnetic
Isotope Separators and Techniques related to their Applications.

Zinal, Switzerland, September 1-6, 1980

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1. INTRODUCTION

The atomic-beam magnetic resonance (ABMR) method has been used during the last decades to study systematically the hyperfine structure of free atoms. The main interest in measurements on unstable nuclides is associated with the nuclear part of the hyperfine interactions, i.e. in the determination of nuclear spins and moments. The majority of experimental data of this kind are due to ABMR experiments; e.g. 80% of the about 500 directly determined nuclear spins of ground and isomeric states in unstable nuclides¹⁾.

Off-line techniques may be used to study nuclides with half-lives down to about 10 min. These experiments include isotope production in a cyclotron or reactor, transfer to the ABMR apparatus and counting of the exposed collectors in external detectors. The investigation of more short-lived nuclides requires some kind of on-line procedure, and, for nuclides far from stability an intermediate isotope separation step is indispensable.

The first on-line ABMR experiments were performed in 1964 at the Princeton 18 MeV proton cyclotron²⁾. In the low-energy proton reactions, it is evident that only nuclides close to the beta-stability line could be reached for study. Due to the pure reaction products no isotope separation was required.

The second ABMR apparatus to be put on-line³⁾ was the former Uppsala machine^{4,5)}, which in 1974 was moved to the ISOLDE facility at CERN. The wide range of elements available in high yields has made possible an important extension of ABMR measurements to nuclides far from stability. So far, the experiments have given 57 new spin values and 35 magnetic moments of the elements ^{35}Br , ^{37}Rb , ^{49}In , ^{53}I , ^{55}Cs , ^{63}Eu , ^{69}Tm , ^{79}Au , ^{81}Tl and ^{89}Fr (see e.g. ref. ⁶⁾). Nuclides produced in yields down to 10^6 atoms/s and with half-lives down to 0.38 s have been reached for study.

2. NUCLEAR QUANTITIES ACCESSIBLE IN ABMR EXPERIMENTS

The results from ABMR experiments give information on the hyperfine structure (hfs) of free atoms, i.e. on the energy splittings due to the interaction between the nuclear multipole moments and the corresponding field quantities from the electrons. Measurements performed in weak external magnetic fields reveal the Zeeman-splitting of the hyperfine sub-levels, from which the nuclear spin I is unambiguously determined, once the electronic angular momentum J and splitting factor g_J are known. The deviations from the Zeeman-frequencies, observed in successively stronger fields, give the zero-field hyperfine separations with increasing accuracy. High precision values of the separations may subsequently be obtained by recording the so-called direct transitions between the hyperfine levels

in very weak fields.

The zero-field hyperfine levels, characterized by the quantum number F , ($|I - J| \leq F \leq I + J$), may be written as linear combinations of the hyperfine coupling constants and known functions including the quantum numbers I , J and F .

$$W_0^F = a f_1^F + b f_2^F + c f_3^F + d f_4^F + \dots \quad (1)$$

Here

$$\begin{aligned} a &\propto \mu_I \langle B \rangle / IJ & I, J &\geq 1/2 \\ b &\propto Q_S \langle \delta^2 V / \delta z^2 \rangle & I, J &\geq 1 \\ c &\propto \Omega \langle \delta^2 B / \delta z^2 \rangle & I, J &\geq 3/2 \\ d &\propto Q_{4S} \langle \delta^4 V / \delta z^4 \rangle & I, J &\geq 2 \\ &: \end{aligned} \quad (2)$$

represent the contributions from the dipole, quadrupole, octupole and hexadecapole interactions, respectively, with the given restrictions on I and J . The quantities B and V are the magnetic field and the electric potential, respectively, due to the electronic shell.

Experimental data on the hyperfine separations thus give values for the coupling constants, from which the corresponding nuclear moments, magnetic dipole μ_I , electric quadrupole Q_S , magnetic octupole Ω and electric hexadecapole Q_{4S} , may be evaluated.

The evaluation of the magnetic dipole moments from the measured a -factors is generally made through a direct comparison with a stable isotope in which the values of I , μ_I and a are known. Neglecting the hyperfine anomaly (cf. below), the following relation holds within a given element;

$$\left(\frac{a}{g_I} \right)_1 = \left(\frac{a}{g_I} \right)_2 \quad (3)$$

Because of the limited number of directly determined quadrupole moments, the evaluation of Q_S from measured b -factors has to rely to a large extent on theoretical calculations of the electronic part of the quadrupole interaction. The accuracy of these calculations range between 1% and 30% depending on the complexity of the electronic wave functions.

The energy contributions from the next two higher order hfs interactions are small and generally negligible (a factor $10^4 - 10^6$ smaller than the ones already discussed). In high precision ABMR experiments, however, the c and d -factors have been determined in a few stable isotopes¹⁾ (cf. also ref. ⁷⁾). Evidently, the

evaluation of the corresponding nuclear moments, Ω and Q_{ns} , involves large uncertainties due to the complicated calculations of the electronic parts. Their impact on nuclear theory, therefore, has been limited to date.

In addition to nuclear spins and moments discussed above, ABMR experiments may also yield a direct measure of nuclear g-factors through their interaction with the external magnetic field. Combined with experimental a-factors, one gets in this case information on the differential hyperfine anomaly Δ , defined by :

$$\left(\frac{a}{g_I}\right)_1 = \left(\frac{a}{g_I}\right)_2 (1 + {}^1\Delta^2) \quad (4)$$

The anomaly, probing the difference in distribution of nuclear magnetism in the two isotopes, is generally small, but may reach a 10% level in ${}^2S_{\frac{1}{2}}$ electronic states in heavy elements.

3. THE ON-LINE ABMR APPARATUS AT ISOLDE

Systematic ABMR investigations of the nuclear quantities, described in the preceeding section, require on-line operation at a powerful ISOL (isotope separator on-line) facility. In this section, we will describe the ABMR apparatus³⁾ connected on-line with the ISOLDE isotope separator⁸⁾ at CERN.

The design of the ABMR apparatus is shown in Fig. 1. It consists of an oven section (8), a main section with three magnets (9-11) and a collector-detector section (12-14). The A-magnet (9) acts as a polarizer and the B-magnet (11) as an analyser of the beam, the resonance transitions being induced in the intermediate homogeneous C-field (10). The lower part of the figure shows the narrow beam space including beam trajectories for atoms at resonance. The obstacles are arranged in such a way that only those atoms having undergone a transition from a focusing to a defocusing state may reach the collector and give rise to a resonance signal.

3.1 The oven section

The 60 keV ion beam from the ISOLDE separator is deflected 90° by a pair of electrostatic deflector plates (3) into the oven section of the ABMR apparatus. A stringent requirement in ABMR work is to have a small, well defined source position. This is accomplished by the focusing and deflecting elements of the ISOLDE beam line, and in particular by the electrostatic quadrupole triplet (7) incorporated in the ABMR machine. The strong focusing force of this lens has made it possible to obtain a beam spot smaller than 0.5 mm^2 at the source position.

The closed-oven systems used in previous off-line works have had to be exchanged to new arrangements fitting the on-line running procedure. We have chosen to use simply a foil mounted between two electrodes on an oven tube,

which may be introduced into the ABMR apparatus through an air lock. The foil is mounted with a 45° angle to both the incoming ion beam and to the optical axis of the apparatus. In the on-line operation, the foil is heated and the collected activity is evaporated continuously from the foil. The evaporation follows the cosine distribution law; i.e. 70% of the forward direction activity is evaporated at 45° , along the optical axis of the ABMR machine.

The oven foil system has been used successfully in on-line experiments, and also in cases with an intermittent running procedure, i.e. collection on a cold foil and a subsequent heating off. The latter technique is particularly suited for the study of daughter products.

The choice of oven foil material has to be made with great care in order to obtain an efficient conversion of the incoming ion beam to an atomic beam, useful in the ABMR experiment. Materials making chemical reactions or alloys with the activity under investigation have to be avoided. Furthermore, in some cases, a proper surface work function of the oven foil is of decisive importance to get a high ion-to-atom ratio. The alkali elements, having a low ionization potential, require a low work function surface to be evaporated in the form of atoms. The halogens, on the other hand, require a high work function surface to avoid negative ion formation. In our experiments, we have chosen a tantalum foil covered by a layer of yttrium for the alkalies⁹⁾ and a rhenium foil for the halogens¹⁰⁾.

3.2 The main section

The atoms entering the main section first find themselves in the inhomogeneous field of the sixpole A-magnet. A force will be felt due to the interaction between the effective magnetic moment and the field gradient, focusing or defocusing depending on the sign of the moment. Only those atoms being focused may pass the A-magnet and enter the C-region. They will be focused once more in the fourpole B-magnet, hit the central obstacle and be lost, unless a change to a defocusing state occurs in the C-region. The change is induced by a rf-field superimposed on the homogeneous magnetic C-field. Once this resonance condition is fulfilled the atomic-beam may transverse the full length of the ABMR apparatus and reach the annular collector.

The sixpole-fourpole focusing magnet configuration of the present apparatus is optimized^{5,11)} with respect to transmission for elements which are difficult to deflect; i.e. elements which have small g_J -values (small effective magnetic moments) and high evaporation temperatures T (high velocities through the apparatus). Compared to the transmission of conventional twopole-twopole machine, the focusing principle alone gives a gain of a factor 10 for elements with large g_J/T -values. In the optimized version, this gain is obtained also for elements with small values of g_J/T . The present apparatus thus gives a high transmission for a large range of elements; a necessary condition in experimental work on different elements produced in minute amounts.

The choice of the sixpole-fourpole configuration is based on the following considerations : i) A sixpole A-magnet is well suited to accept a beam of large solid angle since the restoring force is progressively stronger with increasing radius. ii) A fourpole B-magnet, with the constant field gradient, defocuses the atoms close to the beam axis more, and those at large radii less than a sixpole B-magnet. The latter property results in a small collector area, which is of importance in on-line work. The collector area is about a quarter of that in a sixpole-sixpole configuration with similar transmission properties. The detector area may thus be made correspondingly smaller, reducing the background counting rate. From this point of view, a sixpole-twopole machine would be even more advantageous. However, the lack of axial symmetry is expected to cause severe adjustment problems¹¹⁾.

3.3 The collector-detector section

The detection of the atomic-beam transmitted through the ABMR apparatus is sensitively made by monitoring the nuclear radiation. Depending on the decay characteristics of the nuclides under investigation, different kind of detectors are used; thin plastic scintillators for β^- and β^+ , surface barrier detectors for α , Ge(Li) and NaI detectors for γ and X-rays.

The collectors, normally thin aluminium foils, are exposed to the atomic-beam for a certain period, after which they are moved to the detector positions, either automatically within an internal collector-disc system or manually to external set-ups. Nuclides with half-lives down to about 20 s are generally monitored on the primary decay. More short-lived nuclides may be studied by exposing the collectors long enough to saturate a longer-lived daughter, and to measure the activity of the latter in well shielded external counters. In cases where no daughter activity is available, the detectors may be placed directly at the collector position of the apparatus, monitoring continuously the collected activity. In spite of a higher background counting rate, this detection technique has turned out to be very rapid and efficient.

4. ATOMIC PROPERTIES OF IMPORTANCE IN ABMR WORK

The atomic properties of an element are often decisive for a successful investigation by the ABMR method. Generally, atomic-beam work is simplified by elements having :

a) small electronic J-values (different from zero), resulting in a simple hyperfine structure,

b) only electronic ground state thermally populated; i.e. no interfering resonances from excited states,

c) large electronic g-values and low evaporation temperatures, resulting in a high transmission (cf. section 3.2).

Inspecting now the periodic table of the elements in Fig. 2, one finds the alkali elements (group IA) to have all the properties a)-c) above. They are consequently well suited for atomic-beam work. The elements of group IB (Cu, Ag and Au) have much the same properties. A drawback is of course the higher evaporation temperatures needed which result in a lower transmission.

Elements with closed electronic shells or subshells have 1S_0 ground states, i.e. angular momentum and hence magnetic moment equal to zero. These elements (the groups IIA, IIB and VIIIA, and the elements Pd and Yb) cannot be studied by conventional atomic-beam techniques, since the effective magnetic moments are too small to cause a sufficient deflection in the focusing magnets. The excited states are too high in energy for thermal population. Different oven systems have, however, been developed to produce beams of atoms in metastable states (cf. e.g. ref. 7)).

The p-electron elements (the groups IIIA-VIIA) are generally rather suitable for atomic-beam work. There are some exceptions, e.g. the element Pb with a 3P_0 electronic ground state.

The elements of the groups IVB-VIIIB are very difficult because of the complicated electronic structures and the high evaporation temperatures needed. The 4d and 5d elements require special evaporation techniques to be studied (cf. e.g. ref. 7)).

The atomic properties of the elements of group IIIB (Sc, Y, the lanthanides and the actinides) show a large diversity. The simplest elements to study are the rare-earths Eu, Dy, Ho, Er and Tm with moderate evaporation temperatures and a rather simple electronic structure.

In summary, the elements which are relatively simple to study with the atomic-beam method are : the alkali metals (group IA), the noble metals (group IB), the p-electron elements (the groups IIIA-VIIA) and some rare-earths (Eu, Dy, Ho, Er and Tm).

In the periodic table of Fig. 2, we have also indicated the elements which have appeared as primary products in scheduled production runs at ISOLDE and those tested on-line. It may be noted, however, that further elements are available as daughter products and that several elements, particularly in the groups IIIA-VIIA, are within reach with present target ion source techniques.

The beam production at the separator and at the ABMR apparatus has much in common, and there is consequently a large overlap between the elements produced at ISOLDE and those which are possible to study by the ABMR method. The large

range of 1S_0 elements are, of course, inaccessible to ABMR. Due to their low vapour pressure, the d-electron elements (the groups IVB-VIIIB) evidently pose large problems to both the separator and ABMR group.

So far, the ABMR experiments at ISOLDE have been concentrated to the primary products Rb, Cs, Fr; In, Tl; Br, I and Eu and to the daughter products Au and Tm (cf. Fig. 2). In a near future we foresee an extension of the measurements to further p-electron and rare-earth elements.

5. OVERALL EFFICIENCY OF THE ABMR METHOD

It is evident that experiments including population inversion and external perturbation of atomic systems will suffer from a low efficiency. In the ABMR measurements, the main reducing factor, 10^{-4} , is due to the low transmission of the polarizing and analysing magnets. The rf-transitions involve only a fraction of the hyperfine sublevels, typically 10^{-1} , with a transition probability of 5×10^{-1} . There are also significant losses in the oven and detector systems, say 10^{-1} and 2×10^{-1} , respectively. Including further reductions, due to e.g. the transmission from the separator, of 5×10^{-1} , we end up with a typical overall efficiency of 5×10^{-8} . This efficiency is defined as the count rate at the ABMR detector divided by the saturation count rate in the collector chamber of the separator (atoms per second produced). The efficiency, of course, shows large variations depending on the properties of the nuclide under investigation. Assuming an overall efficiency of 5×10^{-8} and a lower limit of 10 cpm for the detection of a resonance signal, it is possible to make a measurement with a production rate in the separator of 3×10^5 atoms/s.

As examples of production yields at ISOLDE, we show in Fig. 3 those of the alkali elements rubidium, cesium and francium. In spallation reactions with a 1 μ A, 600 MeV proton beam, rubidium is obtained from a niobium target, cesium from a lanthanum target and francium from a uranium target. In cesium, the peak yield is about 7 nA. The gap in the francium yield curve is due to the region of extremely short-lived isotopes just above the neutron shell closure at $N = 126$.

The lower curves for rubidium and cesium, displaced towards the neutron-rich side, are the yields obtained in proton-induced fission in a target of graphite cloth impregnated with 25 g uranium. The combination of spallation and fission targets thus gives a wide range of isotopes in high yields.

Since the production yields decrease strongly when going away from the beta-stability line, it is the intensities rather than the half-lives which limit the number of nuclides to be studied by the ABMR method. The most short-lived nuclide studied to date is the 0.38 s rubidium isotope ^{95}Rb .

The horizontal line at 10^6 - 10^7 atoms/s shows the lower yield limit required in our measurements. The nuclides in which the ABMR group at ISOLDE has made ground state hfs measurements are indicated by black dots at the bottom of the figure. The open rings denote nuclides first investigated by other groups, and the half-filled rings nuclides in which previous spin determinations have been followed by our moment measurements. Two symbols at the same mass number represent ground and isomeric state measurements.

Experimental results on the hfs of excited states and on isotope shifts have recently been obtained in these alkalies at Mainz^{12,13,14)} and at ISOLDE^{15,16)} by laser spectroscopic methods. Thus information on spectroscopic quadrupole moments and on changes in nuclear radii are available in addition to spins and magnetic moments. The collinear beam-laser apparatus of the Mainz group has now been installed at ISOLDE and given the first results on the 1S_0 elements barium and ytterbium¹⁷⁾.

6. CONCLUSION

As stated in the introduction, the main objective of hfs experiments on unstable nuclides lies in the field of nuclear physics. The results from systematic spin and moment measurements give important information on nuclear level assignments and on changes in nuclear deformation. Of particular value are investigations of long isotopic chains, mapping the changes in nuclear structure as a function of neutron number. To extend previous hfs measurements and to reach short-lived nuclides far from stability, on-line operation at a powerful ISOL-facility is required.

In this paper, we have discussed the possibilities and limitations of the ABMR method in hfs investigations of short-lived nuclides far from stability. As an illustration of the results already obtained, we have indicated in the nuclear chart of Fig. 4 the extent of our on-line hfs measurements at ISOLDE. Also indicated in the nuclear chart are the nuclides previously studied by the Gothenburg-Uppsala atomic-beam group using off-line ABMR techniques. Obviously, the results of the combined efforts, e.g. 216 nuclear spins representing more than 40% of the number directly determined spins in unstable nuclides, have had a great impact on the present knowledge of nuclear structure.

ACKNOWLEDGEMENTS

The author would like to thank Prof. Ingvar Lindgren, founder and leader of the Swedish atomic-beam group, for his constant support of the on-line ABMR project at ISOLDE. Present and former members of the atomic-beam and ISOLDE groups are sincerely thanked for their important contributions to the project, in particular Drs. Sten Ingelman and Gudmund Wannberg in constructing and setting up the equipment at ISOLDE.

This work has been supported financially by the Swedish Natural Science Research Council.

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FIGURE CAPTIONS

Fig. 1 : Top view of the ABMR apparatus connected on-line with the ISOLDE isotope separator. (1) Beam line from the separator, (2) off-line collector position, (3) vacuum chamber with electrostatic deflector plates, (4) beam line continuing to the collinear beam-laser experiment, (5) beam scanner, (6) automatic valve, (7) electrostatic quadrupole triplet, (8) oven section, (9) polarizing sixpole magnet, (10) dipole magnet and rf-loop, (11) analysing fourpole magnet, (12) collector section, (13) collector disc feeder and (14) detector section. The lower part of the figure shows the beam space of the ABMR apparatus, including a beam bundle of atoms in resonance.

Fig. 2 : The elements which are inaccessible to "conventional" ABMR studies, due either to $J = 0$ electronic ground states (the groups IIA, IIB, VIIIA and the elements Pd, Pb and Yb) or to too high evaporation temperatures to form an atomic-beam (the 4d and 5d elements of the groups IVB-VIIIB), are given in the periodic table by shaded squares. The elements within squares have been obtained as primary products in scheduled production runs at ISOLDE, and those within circles have been tested on-line (H. Ravn, private communication). On-line ABMR experiments at ISOLDE have been performed in elements indicated by a black triangle.

Fig. 3 : Production yields of rubidium, cesium and francium from different reactions and target systems at the ISOLDE isotope separator (courtesy H. Ravn). Nuclides produced in yields higher than 10^6 - 10^7 atoms/s have been studied (black dots) by the ABMR-group.

Fig. 4 : Conventional nuclear chart including the nuclides experimentally observed. The naturally occurring nuclides are denoted by black squares and those studied by the Gothenburg-Uppsala-CERN atomic-beam group by circles. At the ISOLDE Facility, CERN, on-line ABMR experiments have been performed in nuclides of the elements Br, Rb, In, I, Cs, Eu, Tm, Au, Tl and Fr (cf. e.g. ref.⁶). Shell closures and isodeformation curves ($\epsilon \approx 0.2$) are also indicated to give a rough idea of the nuclear shape and structure to be expected in different regions.

Fig. 1

PERIODIC TABLE OF THE ELEMENTS

[illegible]

Fig. 2

Fig. 3

Fig. 4

18