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RADIOACTIVE ION BEAMS FOR SOLID STATE RESEARCH

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ABSTRACT

Radioactive isotopes are widely used in many research fields. In some applications they are used as tracers after diffusion or after activation in the material itself through nuclear reactions. For research in solid state physics, the ion implantation technique is the most flexible and convenient method to introduce the radioactive isotopes in the materials to be studied, since it allows the control of the ion dose, the implantation depth and the isotopic purity. The on-line coupling of isotope separators to particle accelerators, as is the case of the ISOLDE facility at CERN, allows the obtention of a wide range of high purity short lived isotopes. Currently, the most stringent limitation for some applications is the low acceleration energy of 60 keV of the ISOLDE beam. In this communication a short review of the current applications of the radioactive beams for research in solid state physics at ISOLDE is done. The development of a post-accelerator facility for MeV radioactive ions is introduced and the advantages of energetic radioactive beams are discussed.

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1. Introduction

The increased application of nuclear techniques to research in solid state physics appeared in the last decades mostly as a result of the conversion and upgrading of well-established nuclear physics oriented laboratories. Relevant examples of this conversion are the electron storage rings which are progressively being used as synchrotron radiation sources. More than 20 of such facilities exist around the world providing sharp and intense synchrotron radiation beams for studies in physics, biology, chemistry and crystallography [1]. The Van de Graaf accelerators, which are nowadays actively used in surface, near surface and thin film analysis, are representative of another type of machines which occupy a relevant position in solid state research. Protons, He^+ and D^+ ion beams accelerated to energies of a few MeV are commonly applied to characterize the crystalline quality, the depth profile and the lattice site location of implanted impurities [2]. Although the first applications of radioactive isotopes to solid state physics started long ago in the late twenties, with the first radio tracer diffusion experiment [3], the first "industrial" mass separator for radioactive ion beams was developed for the Manhattan project only in the late fourties [4]. At the same time, investigators at the Bell Labs proposed the use of ion doping techniques to process silicon based transistors [5].

Neutron activation using nuclear reactors has proved to be an easy way of obtaining radioactive isotopes, with the advantage of being accessible to groups working in different research areas. In this way, radioactive isotopes could be obtained in the materials to be analyzed by neutron irradiation of previously introduced impurities, or by direct implantation of the radioactive species. Both methods were successfully used in wearing studies and they are today routinely used on materials hardness and diffusion studies [6]. These are only a few examples of fundamental scientific and technological ideas that led to the current status and interests of applied research with radioactive ion beams.

More sophisticated techniques that rely on specific properties of excited nuclei were introduced in solid state research in the late sixties. The so called “Hyperfine Interactions” (HI) techniques, which have been developed to investigate the nuclear structure phenomena, are used today to study the interaction of nuclear moments with the local fields in materials. Mössbauer Effect (ME) [7], Perturbed Angular Correlations (PAC) [8], Nuclear Orientation (NO) [9] and the β-Nuclear Magnetic Resonance (β-NMR) [10] are examples of such techniques. Their applications, however, are restricted by the limited number of adequate probe isotopes, particularly, in off-line experiments. Inspite of that, a small number of appropriate nuclei, with halflives of the order of several hours, stimulated new experiments oriented for studies of impurity doping of metals and semiconductors [11, 12]. As a relevant example of such new applications, surface physics should be mentioned as one of the most original development, combining ultra high vacuum conditions and the PAC technique. Radioactive nuclei are deposited onto the top surface or between a two-layer structure grown “in situ”, making possible to probe the fields on these sites and the impurity mobility in surfaces and interfaces of metallic and semiconductor multilayer structures [13].

The relevance of such methods has been well expressed during the last years with the establishment of several proposals and letters of intent for the introduction or reinforcement of solid state physics in the working program of radioactive beam facilities around the world, from which ISOLDE/CERN (Isotope Separator On-Line/ European Organization for Nuclear Research) at Geneva [14, 15], Isospin Laboratory at Oak Ridge [16], ISAC-1/TRIUMF exotic beams facility at Vancouver [17] and the SPIRAL/GANIL (Separator and Post-accelerator of On-line Produced Radioactive Ions) facility at Caen [18] can be mentioned. The success of nuclear solid state physics research depends heavily on the availability of a broad range of appropriate radioactive isotopes and on an adequate sample preparation. The on-line production of radioactive isotopes is the technical approach that best fulfills the first of these

requirements, being its most relevant example the ISOLDE facility at CERN. The combination of a big variety of isotopically clean radioactive ion beams with the possibility to implant the isotopes on-line attracted an increasing number of groups to present projects in materials research at CERN. The second requirement, i.e., the adequate sample preparation, will have a new approach with the recent approval of a project for post-acceleration. The post-acceleration of the radioactive ions until several MeV/u was first proposed at ISOLDE for nuclear and astrophysics studies [19]. Such beams will provide new exotic nuclear configurations, essential to understand the process taking place in the explosive stellar events that led to nucleosynthesis. The nuclear solid state community can also profit from this powerful tool. High energy radioactive ion beams will allow to exploit the advantage of selectively deeper implantations in a unique way that combines the simultaneous modification of materials with a powerful set of nuclear characterization techniques looking into the dopant sites and its interactions with point defects and impurities at the atomic level.

In this paper we include a short survey of the principal features of the on-line isotope production and separation in the ISOLDE facility at CERN. Special attention is given to the recently approved radioactive ion beam post-accelerator project. A review of the principal nuclear solid state techniques is given and its future physics applications with energetic ion beams are introduced.

2. The ISOLDE facility at CERN

2.1. The isotope production

At ISOLDE radioactive nuclei are produced through spallation, fission or fragmentation reactions in thick targets by an external 1 GeV proton beam from CERN's PS-Booster [14]. The target products are vaporized and extracted through a transfer line into a selective ion source. The extracted ions are accelerated to 60 keV and the different masses are separated by analyzing magnets. The ISOLDE group developed different advanced target-ion source combinations that allow the use of radioactive isotopes from about 70 elements [20]. Two different mass separators are available. The first one, the General Purpose Separator (GPS), provides mass separation with a resolving power $M/\Delta M = 2400$. It is designed to provide up to three simultaneous beams, within a mass range of ± 15%, delivered to the beam lines in the experimental hall (fig.1). The second mass separator, the High Resolution Separator (HRS), is being equipped with two bending magnets achieving routinely $M/\Delta M = 10{,}000$ with a maximum resolution of more than 30,000 for special purposes.

2.2. The post-acceleration of radioactive beams

Different projects for energetic radioactive ion beams are currently used around the world [21]. The ISOLDE approach conciliates the large know-how for the production of radioactive beams with a low cost radio frequency quadrupole (RFQ) acceleration Linac [19]. Before the injection into the RFQ, the primary ISOLDE beam is processed in two steps, transforming a continuous (1+) beam in bunches with a high charge to mass ratio. First, the ISOLDE beam will be continuously injected into a Penning trap. After accumulation for about 20 ms, bunches

are transferred to an Electron Beam Ion Source (EBIS). By successive ionization a charge to mass ratio larger than 1/4.5 is obtained in about 10-20 ms. The output ions are then extracted to a voltage of about 1 to 2 kV and the desired charge to mass ratio is selected by dipole magnets and injected in a special low current RFQ accelerator. After the RFQ acceleration to 0.5MeV/u, a linear accelerator based on an Interdigital H structure and three 7-gap resonators provides beams with variable energy between 0.8MeV/u and 2.0 MeV/u. An efficiency of about 10% is expected, essentially due to the EBIS ionization efficiency. For investigations in solid state physics, low energies between 1 and 10 MeV / A can be obtained. For energies below 1 MeV/A, a low charged ion beam (4+) coming out from the EBIS will be injected through an electrostatic high voltage acceleration lens. The samples are mounted in a high voltage platform at a variable potential between 0 and -200kV.

3. Nuclear Solid State physics at ISOLDE

3.1. Present status

The high intensity and wide range of radioactive isotopes have made ISOLDE the ideal facility for solid state physicists using radioactivity in their research. In fig. 2 are shown the elements for which there are radioactive isotopes adequate for several techniques already used in experiments at ISOLDE. Currently, about 35% of all the beam time is dedicated to the solid state physics program, which started during the seventies with some pioneer experiments, in which the short lived ^{111m}Cd and ^{119}Cd isotopes were implanted into non cubic metals [22]. Since then the major effort of the solid state physics program has been the study of impurities in semiconductors [23].

Diffusion studies of radioactive tracers are still today an important tool at ISOLDE, being currently used to study the anomalous diffusion of low concentrations of gold in amorphous silicon [24]. Due to the 60 keV beam energy, the isotopes are implanted to a rather shallow depth. The samples are then submitted to isocronal annealing and the analysis is done by removing layers from the implanted surface, using sputtering techniques, and counting the radiation intensity.

A whole spectrum of more modern techniques which are being used at ISOLDE is essentially divided into two groups: the well-established "nuclear" and the most recent "tracer" techniques. With nuclear techniques like, e.g., Emission Channeling (EC), ME, PAC and β-NMR spectroscopy, the radioactive nuclei are used as local probes of the structural or electronic environment in metals and semiconductors, including surfaces and interfaces.

In EC experiments the lattice site of radioactive nuclei implanted in single crystalline materials is determined by measuring the angular distribution of the emitted particles (α, $\beta^{+/-}$,e^{-}) [25]. In order to decrease the implanted dose and accumulation time, particle detection is done using two dimensional position sensitive detectors, which provide very reliable measurements of the emission channeling/blocking patterns. One of the specific advantages of this technique is a very low minimum implanted dose, of the order of $10^{12}cm^{-2}$, about two orders of magnitude below the doses used by conventional channeling based on Rutherford backscattering. As an example, it is worthwhile to mention the lattice location studies of Li in intrinsic and compound semiconductors of the short lived α-emitter ^{8}Li, after implantation of doses even below $10^{12}cm^{-2}$ [26].

The PAC and ME hyperfine techniques represent the most widespread use of radioactive isotopes for solid state research at ISOLDE. These techniques are sensitive to the local environment of the implanted radioactive probe, providing information on the defect structure,

dynamics and electronic configuration at the atomic level. PAC and ME rely on the interaction of the nuclear quadrupole or magnetic moments of the probe with the electric field gradient (EFG) or magnetic hyperfine field at the probe's location. In particular, the EFG provides information on the local charge density distribution and acts as a fingerprint of a specific defect trapped by the probe nuclei. At ISOLDE several ME experiments have been performed with the first excited state of ^{119}Sn, obtained either by the decay of the long lived ^{119}Sb or the short lived ^{119}In isotopes. Studies of the structure and electronic states of impurity-vacancy complexes in III-V semiconductors and, more recently, studies of Sb doping of Si-based semiconductors such as SiGe alloys are being pursuit at ISOLDE [15, 27]. The sensitivity of the PAC technique, contrary to the Mössbauer effect, is essentially temperature independent and a wider range of probe nuclei can be used [8, 12]. At ISOLDE, the PAC technique is being applied to the study of a technologically relevant problem of impurity-impurity interactions in semiconductors. The trapping of hydrogen at p-type impurities in III-V semiconductors is being studied using the representative ^{111m}Cd probe [28]. The recent introduction and development at ISOLDE of the e^--γ PAC technique enlarged significantly the number of probe nuclei. Isotopes with highly converted cascades, inaccessible to the traditional γ-γ PAC technique, such as ^{197m}Hg and ^{73}As / ^{73}Ge [29] are now available, providing new possibilities for studying defects in compound semiconductors and n-type impurities in intrinsic semiconductors. Further, with the on-line coupling of the e^--γ spectrometer to the ISOLDE beam line, the short lived isotopes ^{127}Ba/^{127}Cs, ^{75}Kr/^{75}Br and ^{83m}Kr/^{83}Kr can, also, be used [30, 31].

Essentially, providing similar information as the ME and PAC techniques, β-NMR works with isotopes of the light alkaline Li, Na and Mg elements. The β-NMR technique has been used at ISOLDE for a long time, essentially, in nuclear physics studies. Recently the

quadrupole moment of ^{11}Li ("halo" nucleus) has been measured with β-NMR measurements after implantation of ^{11}Li in $LiNbO_3$ [32]. A first proposal of β-NMR studies of implanted Li in ZnSe compound semiconductor was recently approved at ISOLDE [33].

The new "tracer" techniques combine the standard electrical or optical measurements, commonly used in semiconductor physics, with the use of radioactive isotopes. It was first demonstrated with the identification of the Deep Level Transient Spectroscopy (DLTS) signals from Au and Pt implanted Si, that when the radioactive decay involves the chemical transmutation of the dopant, the electrical and optical properties of the semiconductors will change in time with the characteristic time constant of the radioactive decay [34]. Currently, Hall effect [35], DLTS, Capacitance-Voltage measurements [36] and Photoluminescence Spectroscopy [37] use radioactive isotopes to overcome the chemical "blindness" of the non-radioactive versions, in studies of the electrical activation of impurities in intrinsic and II-VI compound semiconductors [38].

3.2. Perspectives with the availability of energetic radioactive beams

The availability of energetic radioactive ion beams opens new perspectives at ISOLDE. Due to the present beam energy of 60 keV the probe atoms are localised some tens of nanometers below the surface after implantation. For studies of defects in semiconductors, higher energies would be necessary to localise the radioactive isotopes deeper in the bulk, that is, outside the surface defects and space charge region. By varying the energy of the ions in the 0.1-10 MeV range, implantation profiles can be produced, with a resulting local probe atom concentration of about 10^{15}-10^{16} cm^{-3}, instead of the current 10^{17}-10^{18} cm^{-3}. Phenomena that are highly sensitive to the influence of the Fermi level, such as passivation mechanisms due to the trapping of mobile impurities or defects, can be studied in this way. The radioactive tracer

experiments will also profit with deeper implantations since in this way one should be able to avoid the precipitation of some impurities to the surface [15]. The close vicinity of the surface to the implanted region is, also, a strong limiting factor when comparing data from different techniques, which are sensitive to different depths. The most straightforward case is the DLTS technique, which will have the radioactive isotopes deeper in the sensitive region of the Schottky diode.

Nowadays, the ion implantation technology with energetic ion beams became an essential tool on semiconductor processing for electronic devices. New fields of research are represented by the technological processes where defects are intentionally produced, at selected depths, by ion implantation and used for a controlled stabilization of the properties of semiconductors [5, 15, 39]. The adequate control of the "defect engineering" technologies will need a deeper understanding of the physics involving such defects, and this is an ideal field of future research, which combines energetic radioactive ion beams with the presently well established nuclear techniques.

The future applications of radioactive ion beams are not restricted to semiconductor physics. Radioactive ion beams are currently being applied to study the structure and defects of technologically relevant oxides [40, 41]. The ion beam material modification is being progressively extended to photonics, opening new ways to the doping of a large variety of photo sensitive materials. Waveguides and quantum wells are being produced by the implantation of photo-active impurities at selected depths in different substrate materials like $LiNbO_3$, and Al_2O_3 [42]. The stability of the dopants in such materials represents today a big challenge, both for technological and fundamental reasons, and represents an additional field of research that can be addressed by the combination of the nuclear techniques with energetic radioactive ion beams.

3. Conclusions

The interface between nuclear and solid state physics has always been a fertile ground for discovering new phenomena and attractive applications. During the last 70 years the usefulness of radioactive isotopes applied to solid state physics has been gradually recognized, and a large set of nuclear characterization methods can be used nowadays. The success of nuclear solid state research clearly depends today on two factors: a big variety of intense beams of radioactive isotopes from the appropriate elements, which can only be achieved in on-line mass separators, and on an adequate sample preparation which will be achieved with the post-acceleration of radioactive ion beams. The combination of these factors will allow the use of complementary techniques, which revealed in the past to be the most fruitful way to approach the problems in solid state physics. In the near future, at ISOLDE, the well established nuclear solid state techniques will be combined with energetic radioactive ion beams providing new and unique tools for the characterization of materials.

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References

[1] E.Weihreter, Nucl. Instr. and Meth. B68 (1992) 45

[2] J.A.Davies, G. Amsel and J.W. Mayer, Nucl. Instr. and Meth. B64 (1992) 12

[3] J. Groh and G. v. Hevesey, Ann. d. Phys. 65 (1920) 218

[4] S. Groueff, *Manhattan Project* (Little Brown and Co., Boston, 1967) p. 1

[5] S. Moffatt, Nucl. Instr. and Meth. B96 (1995) 1 *and references therein*

[6] P. Fehsenfeld, A. Kleinrahm and E. Bollmann, KIK-Nachr. Jahrg. 18 (1986) 224; A. Kleinrahm and P. Fehsenfeld, KIK-Nachr. Jahrg. 21 (1989) 22

[7] D.L. Williamson, L. Niesen, G. Weyer, R. Sieleman and G. Langouche, in *Hyperfine Interactions of Defects in Semiconductors*, ed. G. Langouche (Elsevier, Amsterdam, 1992) p. 1

[8] Th. Wichert and E. Recknagel, in *Microscopic Methods in Metals*, ed. U. Gonser, Topics in Current Physics, vol 40, (Springer, Berlin 1986) p. 317

[9] N. J. Stone, in *Low-temperature Nuclear Orientation*, eds N.J. Stone and H. Postma (North-Holland, Amsterdam, 1986) p. 641

[10] J. Christiansen, ed., *Hyperfine Interactions of Radioactive Nuclei* (Springer, Berlin 1983) p. 1

[11] H. Haas, Z. Naturforsch. 41 a (1986) 78

[12] J. C. Soares, in *Semiconductor Device Reliability*, eds. A. Christou and B.A. Unger (Kluwer, Netherlands, 1990) p. 291

[13] G. Krausch, R. Fink, K. Jacobs, U. Kohl, J. Lohmüller, B. Luckscheiter, R. Platzer, B.-U. Runge, U. Wöhrmann and G. Shatz, Hyp. Int. 78 (1993) 261 *and references therein*

[14] E. Kugler, D. Fiander, B. Jonson, H. Haas, A. Przewloka, H.L. Ravn, D.J. Simon, K. Zimmer and the ISOLDE Collaboration, Nucl. Instr. and Meth. B70 (1992) 41; B. Jonson, H.L. Ravn and G. Walter, Nucl. Phys. News 3, 2 (1993) 5

[15] G. Weyer, K.B. Nielsen, M. Fanciulli, M. Lindroos, A. N. Larsen and J.W. Petersen, *Defects Studies in High-energy Ion Implanted Semiconductors*, CERN/ISC 94-24, ISC/ I 12 (1994); N. Achtziger et al., *Energetic Radioactive Ion beams Studies of Hydrogen in Semiconductors*, CERN/ISC 94-27, ISC/ I 13 (1994); H. Haas, J.P. Biersack, D. Fink, H. Grawe, M. Lindroos and K. Nishimura, *Diffusion in Highly Immiscible Systems*, CERN/ISC 94-29, ISC/ I 15 (1994)

[16] D.J. Vieira and M. Wiescher, in *Proc. Workshop on the Production and Use of Intense Radioactive Beams at the ISOSPIN Lab*., ed. J.G. Garret (Oak Ridge, 1992) p. 105; J.A. Sawicki, *idem*, p.149

[17] J. Beveridge and J. D'Auria, ISAC Newsletter 15 (1995)

[18] *The SPIRAL Radioactive Ion Beam Facility*, ed. M.Bex (GANIL R 94 02, 1994) p. 39

[19] P. Van Duppen et al., *Radioactive beam Experiments at ISOLDE*, CERN/ISC 94-25, ISC/P68 (1994)

[20] H.L. Ravn, Nucl. Instr. and Meth. B70 (1992) 107; E. Hagebø, P. Hoff, O.C. Jonsson, E. Kugler, J.P. Omtvedt, H.L. Ravn and K. Steffensen, Nucl. Instr. and Meth. B70 (1992) 165

[21] J.M. D'Auria, Nucl. Instr. and Meth. B99 (1995) 330

[22] H.Haas and M. Menninger, Hyp. Int. 9 (1981) 277

[23] H.Haas, in *Proc. Workshop on the Production and Use of Intense Radioactive Beams at the ISOSPIN Lab*., ed. J.G. Garret (Oak Ridge, 1992) p. 83

[24] M. Horz, P. Scharwaechter, W. Gustin and W. Frank, *Diffusion of Au and Pt in Amorphous Silicon*, CERN/ISC 95-13, ISC/P12 (1995)

[25] H. Hofsass and G. Lindner, Phys. Rep. 201 (1991) 121 *and paper I-15 in this conference*

[26] H. Hofsass, S. Winter, S.G. Jahn, U. Wahl and E. Recknagel, Nucl. Instr. and Meth. B63 (1992) 83

[27] G. Weyer, Nucl. Instr. and Meth. B96 (1995) 315

[28] D. Forkel-Wirth et al., Sol. Stat. Comm. 93 (1995) 425

[29] J.G. Correia, H.Haas, J.G. Marques, A.A. Melo and J.C. Soares, Hyp. Int. 80 (1993) 1321

[30] J.G. Marques, J.G. Correia, A.A. Melo, M.F. da Silva and J.C. Soares, Nucl. Instr. and Meth. B99 (1995) 645

[31] J.G. Correia, H.Haas, J.G. Marques, A.A. Melo, M.F. da Silva and J.C. Soares, *Test of New PAC probes for Hyperfine Interaction Studies at ISOLDE,* CERN/ISC 94-16, ISC/P66 (1994)

[32] E. Arnold et al., Phys. Lett. B 281 (1992) 16

[33] B. Itterman et al., β-*NMR Study on Microscopic Structure and Diffusion Behaviour of Li Implanted ZnSe,* CERN/ISC 95-7, ISC/P67 (1995)

[34] J.W. Petersen and J. Nielsen, Appl. Phys. Lett. 56 (1992) 1122

[35] R.Gwilliam, B.J. Sealy and R. Vianden, Nucl. Instr. and Meth. B63 (1992) 106

[36] J. Bollmann, K. Maass, B. Reinhold, J. Röhrich, M. Wienecke and D. Forkel-Wirth, E-MRS 1995 Spring Meeting (Strasbourg 1995) *to be published*

[37] S.E. Daly, M.O. Henry, K. Freitag and R. Vianden, J. Phys. Cond. Matt. 6 (1994) L643

[38] N. Achtziger et al., *Combined Electrical, Optical and Nuclear Investigations of Impurities and Defects in II-IV Semiconductors,* CERN/ISC 92-35, ISC/P35 (1992)

[39] K.S. Jones et al., Nucl. Instr. and Meth. B96 (1995) 196

[40] B.Hauer, R. Vianden, J.G. Marques, N.P. Barradas, J.G. Correia, A.A. Melo, J.C. Soares, F. Agulló-López and E. Diéguez, Phys. Rev. B51 (1995) 6208

[41] D.Lupascu, M. Neubauer, T. Wenzel, M. Uhrmacher and K.P. Lieb, *paper I-16 in this conference*

[42] Ch. Buchal, Nucl. Instr. and Meth. B96 (1995) 370

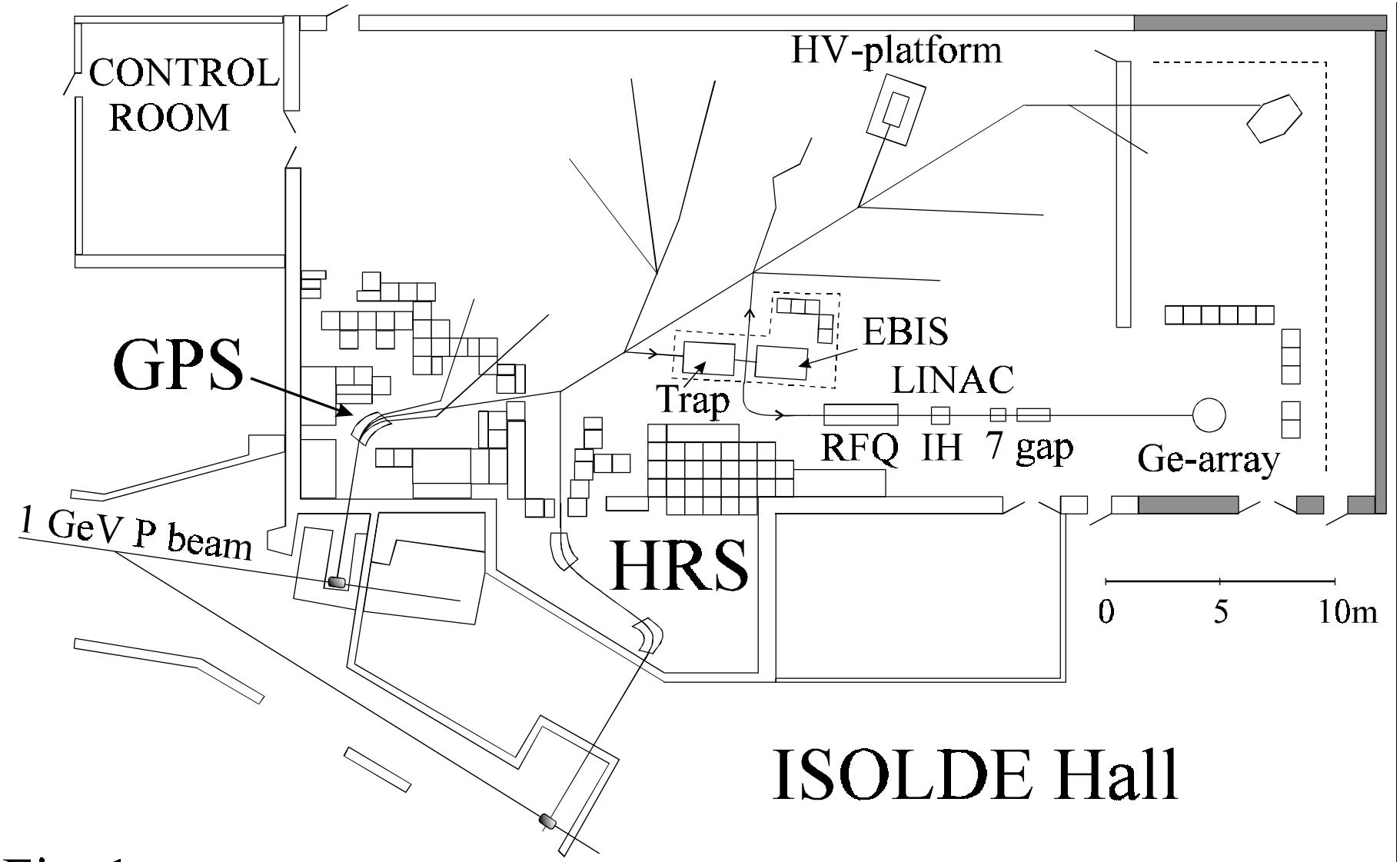
CONTROL
ROOM
HV-platform
GPS
EBIS
LINAC
Trap
RFQ IH 7 gap
Ge-array
1 GeV P beam
HRS
0
5
10m
ISOLDE Hall

Fig. 1

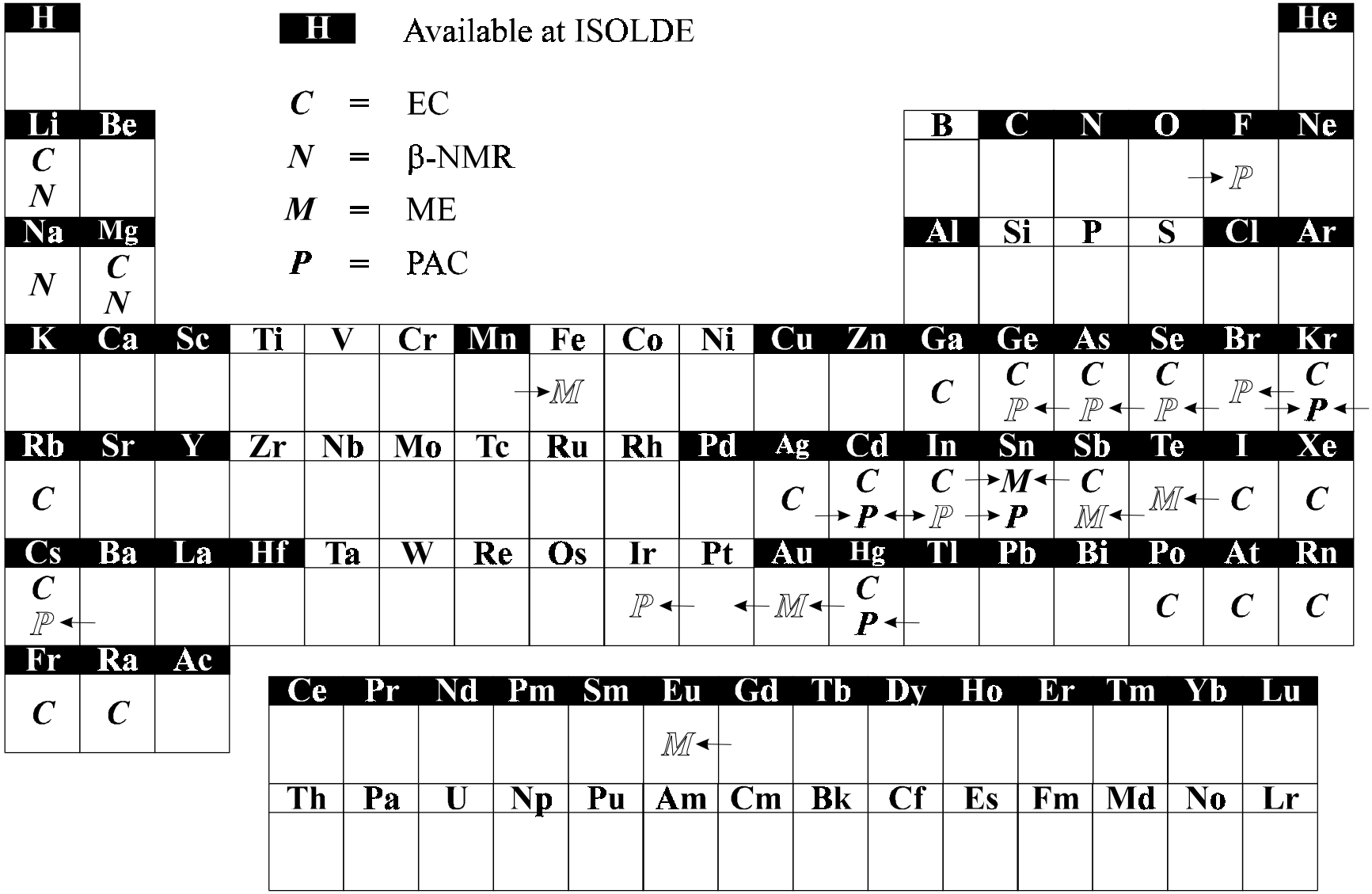
H
Available at ISOLDE
C = EC
N = β-NMR
M = ME
P = PAC
H
He
Li Be
B C N O F Ne
Na Mg
Al Si P S Cl Ar
K Ca Sc Ti V Cr Mn Fe Co Ni Cu Zn Ga Ge As Se Br Kr
Rb Sr Y Zr Nb Mo Tc Ru Rh Pd Ag Cd In Sn Sb Te I Xe
Cs Ba La Hf Ta W Re Os Ir Pt Au Hg Tl Pb Bi Po At Rn
Fr Ra Ac
Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr

Fig. 2