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Radioactive ions for solid-state investigations at magnetic surfaces and interfaces

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Abstract. Hyperfine interactions observed at isomeric states of radioactive probe nuclei are used as a tool for solid-state investigations. This method is sensitive to atomic-scale properties. In recent years surface and interface investigations using radioactive probes delivered many results which can hardly be achieved by any other method. Several groups, *e.g.*, from Konstanz, Leuven, Groningen, Aarhus, Uppsala, Tel Aviv, Pennsylvania, contributed to this field. Our group studies magnetic properties at surfaces and interfaces performing perturbed angular correlation (PAC) measurements in the UHV chamber ASPIC (Apparatus for Surface Physics and Interfaces at CERN). We take advantage of the enhanced variety of PAC probes delivered by the on-line mass separator ISOLDE. First, we report on measurements of magnetic hyperfine fields (B_{hf}) at Se adatoms on a ferromagnetic substrate using ^{77}Se as a PAC probe. The investigation of *induced* magnetic interactions in nonmagnetic materials is a further subject of our studies. Here the nonmagnetic $4d$ element Pd is investigated, when it is in contact with ferromagnetic nickel. An outlook will be given on studies to be done in the future. The experiments were performed at the HMI, Berlin, and at CERN, Geneva.

PACS. 75.70.-i Magnetic properties of thin films, surfaces, and interfaces – 75.70.Rf Surface magnetism – 76.80.+y Mössbauer effect; other γ-ray spectroscopy

1 Introduction

The use of radioactive ions for investigations of condensed matter is world-wide spread during several decades and is still in an increasing mode. More recent are investigations of surfaces and interfaces of solids, in particular of metallic multilayers. Around the world, there are about a dozen of research groups applying hyperfine-interaction measurements for the study of *local* structural and electronic properties at surfaces and in ultrathin multilayer systems. Pioneering experiments were performed in the beginning of the eighties of the last century with Mößbauer spectroscopy on Fe using isotopically enriched ^{57}Fe layers [1]. These experiments focused the interest especially on *local* magnetic properties at surfaces, near surfaces and at interfaces. Magnetism at surfaces and interfaces became a rapidly growing field of interest and many different methods were applied, in particular the use of circular polarised synchrotron radiation opened new insights. Nevertheless, local magnetic properties still may be studied best with local methods like hyperfine-interaction measurements (*e.g.*, Mößbauer spectroscopy). Noniron systems were accessible applying radioactive ions in a further type of hyperfine-interaction measurements: perturbed angular correlation spectroscopy (PAC). First experiments on Ni [2] and Co [3] with PAC were performed by the Konstanz group.

Of special interest are induced magnetic interactions in magnetic multilayer systems, where magnetic interactions are induced into nonmagnetic metals, when these are in contact with a ferromagnet. One of the first experiments addressing induced magnetic interactions was performed on the Ni/Pd system [4] applying a (nonlocal) magnetometry measurement. It was found that the total magnetic moment of a thin Ni layer increases when Ni is covered by several atomic layers of Pd. The interpretation attributed the increase of the total magnetic moment to additionally induced magnetic interactions in Pd. By incorporating radioactive probes into the Pd layers and applying PAC, our group could prove that, indeed, Pd is magnetically polarised for a few atomic monolayers [5].

The investigation of induced magnetic interactions has become a large field especially with respect to technical applications. Local investigations are necessary in order to understand the underlying physical mechanisms in particular at the interfaces in a multilayer system. Also here, the measurements of hyperfine interactions at suitable radioactive ions are in a favourable situation, since hyperfine interactions are of short range, consequently monolayer-resolved measurements are possible; the gamma radiation of the probe ions is of long range, consequently, measurements can be performed in any depth of the sample; nuclear methods are of high sensitivity, only a small amount

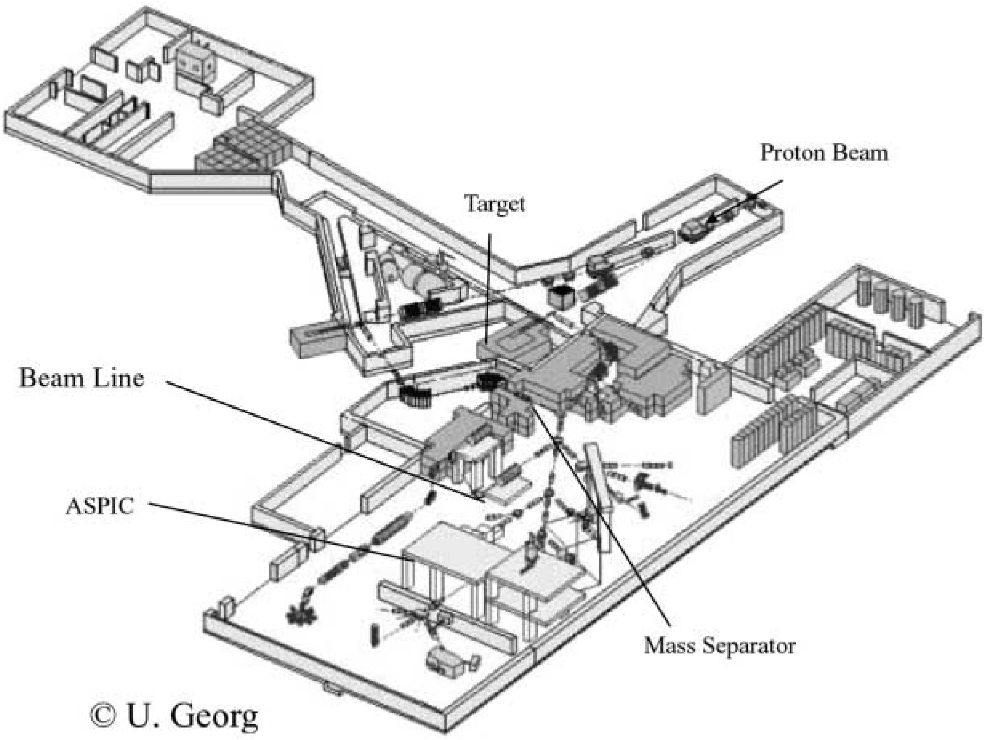


Fig. 1. Experimental hall of the on-line mass separator ISOLDE at CERN, Geneva. In the centre of the hall the ultra-high vacuum (UHV) chamber ASPIC (Apparatus for Surface Physics and Interfaces at CERN) is connected to the mass separator by a UHV beam line. (By courtesy of U. Georg, ISOLDE).

of radioactive probe ions (10^{-4}–10^{-5} of an atomic layer) are necessary for a PAC measurement, consequently, the overall properties of a magnetic layer will not be disturbed.

In this review article we shall present studies of surface magnetism investigated with radioactive atoms; induced magnetic interactions in interface systems studied with monolayer resolution will follow and finally we speculate on possible applications far beyond the presently modern nanoscopic technologies. We start with a very brief introduction of the PAC method lining out the most important features in order to demonstrate how the position of the probe atoms is determined and the magnetic properties are measured simultaneously.

2 Perturbed angular correlation spectroscopy (PAC) for combined interactions

Radioactive ions are obtained from the on-line mass separator ISOLDE at CERN, fig. 1. The mass separator delivers a variety of suitable nuclei in or close to the valley of stability of the nuclear chart in a sufficient amount for solid-state investigations. Three advantages in comparison to other supplies of radioactive sources should be mentioned: i) Because of the *on-line* mode short-lived isotopes are available. ii) The *separator* provides the desired isotopes without contamination. iii) Our experimental set-up, the ultra-high vacuum (UHV) chamber ASPIC (Apparatus for Surface Physics and Interfaces at CERN; base pressure 2×10^{-9} Pa) is *connected* to the mass separator by a UHV beam line, therefore all experiments can be performed without breaking the vacuum. Details of the experimental procedures, in particular of how the radioactive precursor is produced and separated in ISOLDE and positioned onto metallic surfaces of single crystals are described elsewhere [6]. We emphasise that the sample preparation is done with the radioactive precursors, whereas the hyperfine-interaction measurement applying PAC spectroscopy is performed with the decay products when the sample is moved into the PAC position of ASPIC within the 4-detector array. The main features of the PAC method may be followed up in the graph of fig. 2. The ratio function $R(t)$ delivers the PAC time spectra as it is shown schematically in fig. 2, bottom. Measured $R(t)$ functions are given in fig. 5 in the following. From these functions the EFG and the B_{hf} are extracted [7]; the EFG serves as a finger print of the probe location, the B_{hf} may serve as a measure for the s-electron polarisation in magnetic systems.

3 Surface magnetism

The magnetic hyperfine fields of most of the elements of the periodic table implanted as isolated impurities in the ferromagnetic hosts Fe, Co, Ni were measured by different methods during the last 4 decades. In fig. 3, the experimentally obtained bulk B_{hf} values for the $4sp$ elements are plotted [8,9]. Theoretical calculations reproduce these

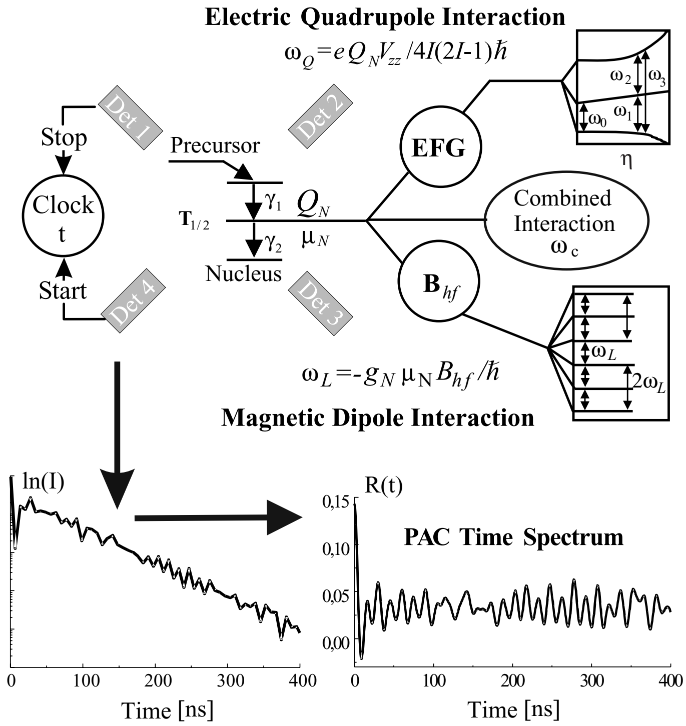


Fig. 2. Perturbed angular correlation (PAC) method as a diagram. Top centre: Suitable PAC probes as decay products of the precursor are excited nuclei having a gamma-gamma cascade with an isomeric state with a half-life in the range of ns or μs. During the lifetime of this intermediate state its nuclear moments are interacting with the immediate environment. Top right: The quadrupole moment Q_N is interacting with the electric-field gradient (EFG) (which is always present at surfaces and interfaces) leading to the electric quadrupole interaction frequency ω_Q. V_{zz} is the largest component of the EFG tensor. The nuclear moment μ_N is interacting with the magnetic hyperfine field, B_hf, present in magnetic systems, resulting in the Larmor frequency ω_L. In most cases both interactions occur simultaneously as combined interaction which can be treated only numerically [7]. Top left: In the 4-detector array the decay function of the isomeric state is measured modulated by the interaction frequencies as shown bottom left. Bottom right: The life time is eliminated in the ratio function $R(t) = 2[(C(180°)-C(90°))/(C(180°)+2C(90°))]$. The expression C stands for the coincidence count rates of the gamma-ray detectors which are positioned at 90° and 180° to each other.

measurements quite well [10]. Close to the half-filled $4sp$ shell where the strongest s-electron polarisation is possible, Se occupies the largest B_hf value in bulk Ni [11]. Placing Se (respectively its precursor Br) as adatom on Ni surfaces, considerably reduced B_hf values were measured, as indicated in fig. 3 [12]. Calculations on the B_hf values for adatoms on Ni(001) reveal a completely different behaviour [13] for sp-elements, in contrast to transition elements where the magnetic hyperfine fields show a scaling behaviour to bulk values [14]. In fig. 3 the calculated adatom values are plotted as a double bump curve in relation to the element number and the calculated value for Se is in obvious agreement with the measurement. In

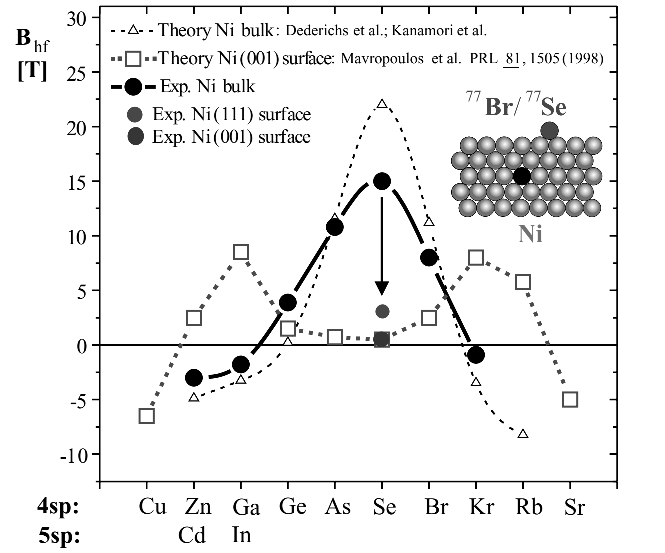


Fig. 3. Magnetic hyperfine fields of the $4sp$-elements as impurities in Ni bulk, black circles [8,9,11]. Open triangles represent the respective calculations [10]. Open squares represent the calculations for the elements in the adatom position [13]. Below the arrow the experimental values for Se as adatom are plotted [12].

order to test the calculation more sensitively, measurements at Zn, Ga or Kr would be necessary where the bulk values are small and negative, whereas the predicted adatom values are large and positive. Measurements with Kr could be very difficult, because the recoil energy of the decay product Kr as a result of the β-decay could cause desorption [15]. Well-suited probe nuclei for PAC spectroscopy exist with ^{111}Cd and ^{117}In nuclei which represent the isoelectric $5sp$-elements with respect to Zn and Ga. Although no calculations exist for the $5sp$-elements

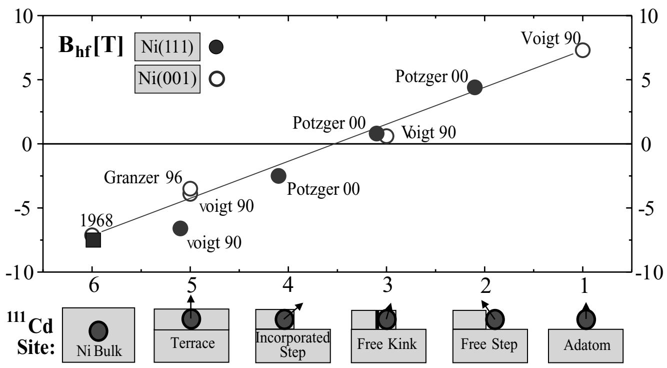


Fig. 4. Increasing magnetic hyperfine field at ^{111}Cd *versus* decreasing contact with ferromagnetic Ni. The bulk value [8] and the adatom value on Ni(001) (Voigt 90, [16]) are connected by a straight line. The values for the positions at terrace, incorporated step, free kink, and free step follow this line. The probe positions are identified by the size and the angle of the simultaneously measured EFGs, as indicated with arrows in the icons. See also fig. 5, where examples of the EFGs are explicitly given.

we anticipate a similar behaviour. Then we expect strong positive B_{hf} values for Cd in the adatom position on Ni. When going from the negative bulk value ($B_{hf} = -7$ T) to the adatom value, several in-between steps are possible. Figure 4 shows the first and recently —with our preliminary data— completed series of such measurements. The temperature-dependent positions of the probe atoms were identified by their EFGs. The sign is only measured for the bulk value, for the adatom values the signs were calculated [17].

These experiments on the study of surface magnetism are part of knowledge-oriented investigations in basic research. Application-oriented research on magnetic systems has to be focused on interface systems, where two different metals are in contact. Whereas for many investigation methods stacks of magnetic multilayers are necessary, investigations with radioactive ions can be performed with exactly one interface, because of the high sensitivity of nuclear methods and because of the short range of hyperfine interactions. With respect to fig. 4, position 5, we now discuss terrace magnetism and the magnetic properties of induced magnetic interactions, when a different ultrathin metal, in this case Pd, is in contact with a Ni(001) terrace at room temperature.

4 Interface magnetism

The combined interaction of ^{111}Cd incorporated in the topmost layer of Ni with (001) orientation is shown in fig. 5, top. A kind of beat pattern is observed caused by the electric quadrupole and the magnetic dipole interaction frequencies. These interactions are of no surprise because the probes are in a nonsymmetric position and Ni is ferromagnetic. The magnetic hyperfine field is reduced to about 50% of its bulk value. Evaporating one atomic monolayer of Pd on top of this Ni surface by MBE

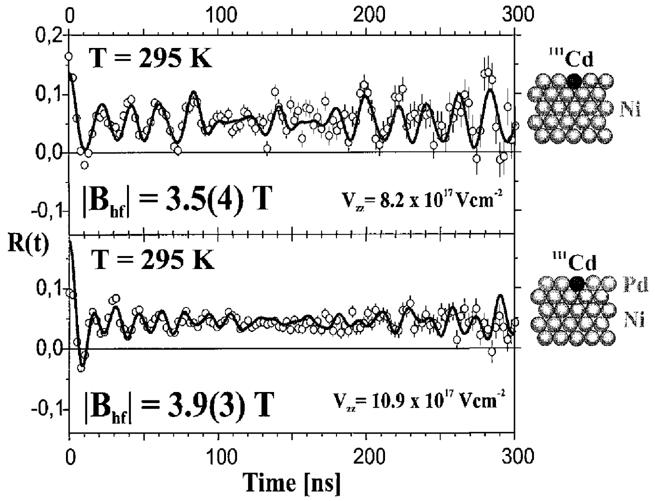
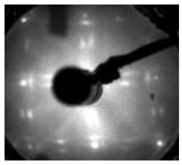


Fig. 5. PAC time spectra of ^{111}Cd incorporated in the topmost layer of Ni(001), top, and in the monolayer of Pd, grown on Ni(001), bottom.



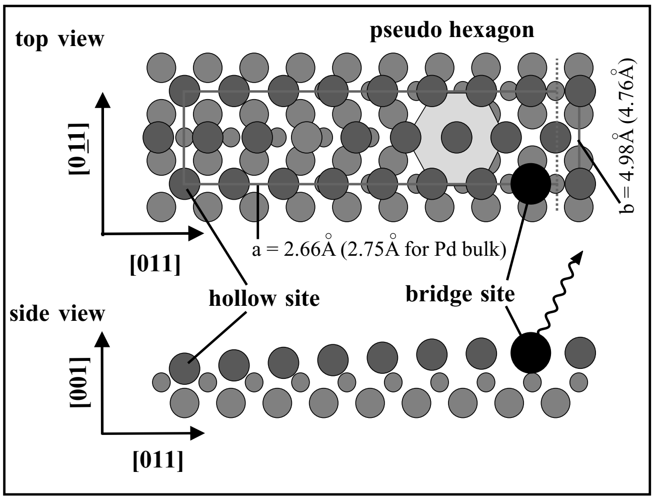


Fig. 6. Model of a Ni(001)c(16 × 2)Pd unit cell, only one half is shown. Light gray circles represent Ni, dark gray represent Pd and black the Cd impurity.

techniques (molecular beam epitaxy) and incorporating the radioactive probes into Pd, the PAC spectrum (fig. 5, bottom), was obtained which resembles the top spectrum surprisingly well. The magnetic hyperfine field is the same indicating that Pd is magnetically polarised. It is proven that the probes are incorporated in the Pd layer because their EFG is different from the one of ^{111}Cd in the topmost layer of Ni and the EFG is reproducing the value which was measured for ^{111}Cd in the topmost layer of a single crystal of Pd(111) [18]. However, the lattice parameters of Ni and Pd differ by approximately 10%, therefore a study of the structure of Pd grown on Ni(001) is necessary.

Figure 6 represents the result of a LEED study (low-energy electron diffraction). Pd grows on Ni(001) in unit cells as Ni(001)c(16 × 2)Pd. The immediate structural environment with respect to the Ni substrate varies from Pd atom to Pd atom. Therefore the $3d$-$4d$ electron hybridisation between Ni and Pd varies, consequently the induced magnetic moments and the transferred magnetic hyperfine fields vary from atom to atom which would result in a broad B_{hf} distribution. Nevertheless, for ^{111}Cd we measure a discrete B_{hf} value. The interpretation of this surprising result is that the Cd atoms occupy selected sites within the unit cell of Pd [19]. Regarding fig. 6, bottom, a corrugation of the Pd atoms is to be seen, starting from hollow sites and going to unique sites in the middle of the cell. With respect to the bigger size of the Cd atoms the occupation of bridge sites during the evaporation process is likely. Performing a PAC experiment with a self-element

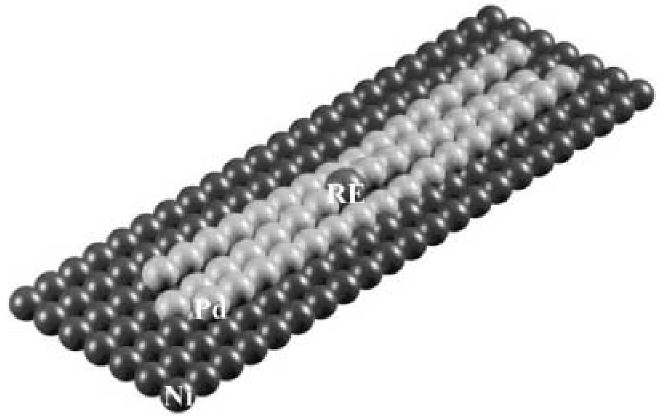


Fig. 7. 3-dimentional view of a complete Ni(001)c(16 × 2)Pd unit cell incorporating one rare-earth atom on an expected unique site.

probe of Pd, ^{100}Pd, which unselectively occupies random sites within the unit cell, a broad distribution of magnetic hyperfine fields was observed [19].

5 Possible applications

During the last decades magnetic memories have been developed with smaller and smaller magnetic units. Nowadays, these units have a size of about 10^4 nm^2. There are many attempts in many laboratories around the world to achieve further miniaturisation. One goal is the terabit memory, *i.e.*, 10^{12} bit per cm^2. It might be worth speculating whether the finding within the experiments with isolated radioactive Cd ions in the Pd-Ni interface might point at a possible solution. On the atomically structured Ni surface a structure of Pd unit cells is obtained which is due to the fact that Ni and Pd have rather different lattice parameters. Speculating that rare-earth atoms evaporated onto the Ni/Pd system occupy selected sites within the unit cells (see artist's view in fig. 7) one would obtain an ordered structure of isolated magnetic atoms in a self-organising procedure. These magnetic atoms couple with the ferromagnetic substrate. A first task is the investigation whether such structures exist. Secondly, the magnetic behaviour of isolated rare-earth atoms in such structures has to be studied. The on-line mass separator ISOLDE delivers various radioactive rare-earth beams suitable for these investigations [20].

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