Proposal:

Interface Magnetism Investigated with Radioactive Atoms

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Summary

Perturbed Angular Correlation (PAC) measurements in the UHV chamber ASPIC (Apparatus for Surface Physics and Interfaces at CERN) using different PAC probes have resulted (i) in the determination of magnetic hyperfine fields of Se on Fe, Co, Ni and prompted a theoretical study on the magnetic hyperfine fields of 4sp-elements in adatom position on Ni and Fe; (ii) in experiments of ¹¹¹Cd in ultrathin Pd on Ni(001) revealing static magnetic hyperfine fields which indicate an induced magnetic order in Pd; (iii) in the observation of induced fluctuating magnetic interactions in Pd, when thick Pd is in contact with Ni. Future experiments are aimed at the search for the transition from induced static to induced fluctuating magnetic interactions in Pd layers of different thickness and in dependence of the temperature. In addition, further measurements of magnetic hyperfine fields of impurities in adatom position are planned in order to test the predictions of the model calculations which come up with a substantially different behaviour as compared to bulk values.
1. Introduction

The Experiment IS 318 "Surface and Interface studies with radioactive ions" was started to obtain informations on structural and magnetic properties on an atomic scale. The investigations were intended to gain contributions for a better understanding of the physics at interfaces of thin-layer systems.

In brief, the application of the nuclear method PAC (perturbed angular correlation) using suitable radioactive probes provides the following advantages for such studies:

1.) Hyperfine interactions are of short range, therefore monolayer-resolved measurements have been proven to be feasible.
2.) The penetration of radiation is of long range, therefore measurements in any depth of a layer system are possible as shown in the Ni/Pd system.
3.) Measurements with nuclear methods are of high sensitivity, only $10^{-4}$ - $10^{-3}$ of a monolayer (ML) of radioactive probes are necessary for a measurement, therefore the macroscopic properties of the ML, especially of the layer system, are not influenced by the probes.

In our UHV chamber ASPIC at the UHV beam line of ISOLDE we have performed two series of PAC experiments at surfaces 1.) and interfaces 2.):

1.) Investigations of surface site occupation on silicon for $^{77}$Br/$^{75}$Se; investigations of (magnetic) hyperfine interactions on Ni and Pd surfaces with the same probe.

2.) Magnetic hyperfine fields in pure Pd - when Pd is in contact with Ni - revealed induced static and fluctuating magnetic interactions in Pd.

On the basis of these results we present a new proposal concentrating on induced magnetic interactions in nonmagnetic metals, when these metals are in contact with a ferromagnetic metal of the 3d-elements (Chapter III). On the basis of recent calculations for magnetic hyperfine fields of 4sp-elements in adatom position on Fe and Ni [1], we intend to extent our measurements on surface magnetism to further impurities (Chapter II). This might open new possibilities to measure the range of induced magnetic interactions, as will be discussed below.
II. Magnetic Hyperfine Fields at Adatoms on Ferromagnetic Surfaces

In a first experiment we had investigated the surfaces of Ni and Pd single crystals separately [2] which resulted in the first measurement of a magnetic hyperfine field for adatoms.

It was frequently observed that adsorbates, especially chalcogens like O or S - even in amounts of less than one ML - drastically reduce the magnetic moments of the topmost layers of Ni [3]. Our intention was to investigate the local magnetic response at an isolated chalcogen impurity. As a suitable PAC probe atom $^{77}$Se was chosen. Among the 4sp elements, Se has the largest $B_{hf}$ value in Ni bulk (Fig. 1). The evaluation of the PAC spectra revealed a drastic reduction of the magnetic hyperfine fields of the chalcogen Se on Ni with values as small as 1 and 3 T in contrast to the bulk value of 15 T, see Fig. 1. These findings stimulated calculations showing that the magnetic hyperfine fields of the 4sp elements in adatom position exhibit a considerably different behavior than in the bulk [1] and our measurement is in excellent agreement. Furthermore, our preliminary measurements of $^{77}$Se on Fe(001) (which have to be repeated) with the low value of $|B_{hf}| \approx 4$ T (70 T in Fe bulk) also agrees remarkably well with the model calculations.

Request: 2 shifts of $^{77}$Br.

\[ B_{hf} \text{ [T]} \]

\( \bullet \) Exp. Ni bulk
\( \triangle \) Exp. Ni(001) surface
\( \circ \) Exp. Ni(001) surface
\( \times \) Exp. Ni(111) surface

**Fig 1.** Magnetic hyperfine fields for 4sp elements in and on Ni. Ni bulk: experiment [4,5,6] solid line; theory [1]; dotted line; Ni(001) surface: theory [1]; dashed-dotted line. Our experimental results for Se on Ni(001) is inserted as \( \circ \) and for Se on Ni(111) as \( \times \).

The magnetic hyperfine fields of Se in Fe, Co, Ni approximately scale with the magnetic moments of their ferromagnetic hosts. It is of interest, to compare this scaling for Se in the adatom position (Fig.2).
Fig. 2.: Comparison of magnetic hyperfine fields for Se in Fe, Co, Ni [4,5] and on Ni [2]. Fe, Co (not yet published) with the magnetic moments of the hosts. The recent measurements for Co are shown in Fig. 3. The arrows point at the respective axis.

The comparison of the magnetic moments of Fe, Co, Ni with the magnetic hyperfine fields for Se adatoms and Se bulk atoms does not reveal a clear picture at present. In Fig. 2, the presently known data are shown. While the value for Se in Ni [5] is measured with high statistics, the respective values for Co and Fe [4] have rather large error bars and would have to be remeasured for a meaningful comparison. Fig. 2 seems to suggest that Se on Co behaves Ni-like, but in Co it behaves Fe-like. In case of more precise experimental values, this first complete set of data could be the basis for a discussion of the change of the electronic structure for this particular chalcogen impurity, when going from the bulk to the surface (not yet included are the magnetic moments of the hosts at the surface). Because of the reduced symmetry at the (001) surface of Ni (and Fe) as compared with the bulk, the hybridization of the s and p impurity states with the 3d orbitals of the ferromagnetic substrate results in the formation of s-p hybrids which are directed towards the substrate. The s electrons are responsible for the magnetic hyperfine field at the impurity nucleus by the Fermi contact interaction, which is given as \( H_{hf} = \frac{8\pi}{3} m(r = 0) \) [\( m(r = 0) \) is the magnetization density at the nucleus].

In bulk materials there is, of course, not such a preferential orientation, see also Ref. 1.

An interesting aspect can be deduced from Fig. 1, regarding the behaviour of Kr in and on Ni. The bulk value \( B_{hf} = -0.7(1) \) T was measured some time ago by our group at VICKSI in Berlin [6]. On the Ni surface an „enhancement“ a large positive value is predicted. We intend to measure the \( B_{hf}(Kr) \) on Ni for three reasons:

(i) \( B_{hf}(Kr) \) on Ni would be a very sensitive test for the theory of Ref. 1.
(ii) The behaviour of noble gases on metallic surfaces is a widely studied field. Information on magnetic interactions would contribute to the knowledge of the electronic structure.
(iii) In case of successful measurements with \(^{89}\)Rb/\(^{89}\)Kr on Ni, e.g., we obtain a new tool for range measurements of induced magnetic interactions in nonmagnetic metals. For example, Ni will be covered by Pd, monolayer by monolayer, and \(^{89}\)Rb/\(^{89}\)Kr will be used to measure the combined hyperfine interaction on top of the topmost Pd layer until only electric hyperfine interactions occur which are recognizable through a measurement on a pure Pd crystal. Since Kr is bound only by van der Waals interaction this would be a most tender measurement. We have successfully performed such a measurement using Se on a Pd/Fe multilayer system, however, Se is strongly bound and has a strongly reduced \( B_{hf} \) value even for Fe. With a possibly strongly enhanced \( B_{hf} \) value for Kr, these measurements would be much more meaningful and deliver a substantial contribution to our main field of research: induced magnetic interactions in nonmagnetic metals (chapter III).

Request: 10 shifts of \(^{79}\)Rb.
Fig. 3: Temperature dependent PAC measurements of $^{75}$Se on a Co(0001) surface. On the left, the time-differential measurements; on the right, the Fourier transforms and the numerical results for the magnetic hyperfine fields $B_{hf}$ and the electric field gradients $V_{zz}$. The PAC spectrum (300 K) at the bottom was taken with the 0/90° geometry of the counters, proving the presence of magnetic interactions. Bottom: Temperature dependence of the $B_{hf}$ and $V_{zz}$ values.
III. Magnetic Multilayers: The 3d-electron / 4d-electron Systems

Ferromagnetic metals like the 3d-electron elements Fe, Co, Ni induce in
nonspontaneously magnetizing metals magnetic properties (e.g. in other 3d-electron elements
or in 4d- and 5d-electron elements) when they are in contact in a (nonalloyed) multilayer
system. In general, also ferromagnetic elements, Fe, Co, Ni, Gd, and antiferromagnetic
elements, Cr, Eu, Tb, influence each other mutually in respective multilayer systems. The study
of the induced magnetic properties and the coupling through nonmagnetic layers (giant
gainteresistance) as well as of the magnetic influence of spontaneously magnetizing metals has
been the subject of increasing research in recent years, also because of the prospect of potential
applications. – Out of the enormous amount of different magnetic multilayers we have
concentrated on the Pd/Ni system, which consists of isoelectric elements.

A) Superparamagnetic Pd

The strongly exchange enhanced metal Pd was already subject of numerous
investigations on magnetic thin-layer systems [7, 8]. In a series of local investigations (using
the PAC probes $^{100}$Pd/$^{100}$Rh at the HMI and $^{111m}$Cd/$^{111}$Cd at ISOLDE) [9] we have shown that
magnetic moments are induced in a single crystal of Pd up to at least 7 atomic layers away
from the Ni/Pd interface.

From the PAC spectra we obtained the damping constant $\lambda_{d}(^{100}$Rh) $>$ 1x10$^8$ s$^{-1}$ for the
magnetic 4d-probe $^{100}$Rh. Applying the transformation $\lambda_{d} \rightarrow \tau_s$ for magnetically polarized Pd as
it was discussed in Ref. [3, appendix], we obtain a unusually small - paramagnetic spin
fluctuation rate, inserted in Table 1 and compared with other fluctuation rates.

Table 1: Selected Paramagnetic Spin Fluctuation Rates

<table>
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<tr>
<th>System</th>
<th>$\tau_s^{-1}$ [s$^{-1}$]</th>
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<tr>
<td>$^{100}$Rh in Ni/Pd multilayer</td>
<td>&lt; 1x10$^{10}$</td>
</tr>
<tr>
<td>$^{100}$Rh in Pd (pure)</td>
<td>4x10$^{13}$</td>
</tr>
<tr>
<td>$^{100}$Rh in Pd (2% Fe, T=120 K)</td>
<td>2x10$^{12}$</td>
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Pd occupies the properties of superparamagnetism, when thick Pd is in contact with
ferromagnetic Ni. At ISOLDE we had applied the $^{111m}$Cd PAC probe, but an induced magnetic
order was not observed in this system. However, at lower temperatures a transition to a
magnetic order is expected.

We have prepared a low temperature device for ASPIC in order to find the blocking
temperature for the superparamagnetic fluctuations or even the phase transition from
ferromagnetic-like Pd to superparamagnetic Pd. This experiment will be performed with $^{111m}$Cd
probes, because $^{111}$Cd is sufficiently sensitive for detection of a magnetic order, as will be
discussed in the next paragraph.
B) Pd with a Magnetic Order

When Pd is evaporated in amounts of a few atomic layers onto a single crystal of Ni, static magnetic hyperfine fields were measured, indicating the formation of induced static magnetic moments. The magnetic hyperfine fields are oriented inplane as the moments are in the Ni surface.

The induced magnetic hyperfine fields in the 1st and 2nd ML of Pd at the isolated $^{111}$Cd impurity are $|B_{hf}| = 3.9(3) \, \text{T}$ and $|B_{hf}| = 1.2(4) \, \text{T}$, respectively. In Fig. 4 we compare the gradient of the $B_{hf}$ values with the gradient of induced magnetic moments calculated for Pd [10] and find a remarkable agreement with the prediction.

![Graph](image)

**Fig. 4:** Magnetic moments (triangles) for Ni (left hand side) and for Pd in contact with Fe [10] (right hand side) in comparison with $|B_{hf}|$ values (circles) for $^{111}$Cd (center ordinate). In order to compare the gradients for the moments and the $B_{hf}$ values within Ni and Pd the values for Ni are normalized at Ni (bulk), the values for Pd at the 1st ML of Pd.

For ultrathin Pd layers we found static magnetic hyperfine fields already at room temperature. A comparison of the findings for thick Pd in contact with Ni and for ultrathin Pd on Ni suggests the expectation, that there is a transition from induced stable moments to fluctuating moments in dependence on the thickness of Pd on Ni. This shall be investigated using the $^{111}$Cd PAC probe positioned into Pd layers grown on Ni(111) and Ni(001).

By LEED investigations we found that Pd grows on Ni(111) with (111) orientation having the bulk lattice parameters from the first Pd layer on. Therefore, using a Ni(111) crystal only one parameter is varied: the number of atomic layers.

The growth of Pd on Ni(001) reveals a more complicated picture (Fig. 5). The first atomic layer of Pd grows in unit cells of Ni(001)-c(16x2)Pd and it shows a corrugation, which could not be observed in the 2nd layer. From the 4th layer Pd occupies bulk parameters. In this system two parameters are varied.

*Request: 8 shifts of $^{111m}$Cd*
**Fig. 5:** A model of the unit cell for Ni(001)-c(16x2)Pd. The (111) orientation of Pd is indicated by the shadowed pseudo hexagon. Note the accumulation of bridge sites in the middle of the cell. The side view shows the corrugation of the Pd atoms. One of the bridge sites is occupied by the bigger (radioactive) $^{111}$Cd.

In Fig. 5, bottom, the corrugation of the Pd coverage on Ni(001) is shown. This configuration suggests that an impurity of bigger size, sufficiently mobile during the evaporation process, might select special sites within the unit cell. We found Cd to be such an impurity [4.5 in the appendix]. We assume that the bridge site in the unit cell is occupied by Cd which seems to be plausible for reasons of volume and symmetry.

Experiments using scanning tunnelling microscopy for a proof of these assumptions are in preparation at the Humboldt-Universität zu Berlin for the following reasons: If Cd occupies selected sites in such a structure, also other atoms with similar properties might do so. For instance, rare earth atoms are also of much bigger size compared to Pd. If these atoms occupy selected sites in a highly diluted concentration (1%) on the surface incorporated in the structured Pd layer, one might be able to create in a selforganizing process structured layers of unit cells incorporating rare earth atoms also in a structural order. From bulk experiments it is known that rare earth atoms couple magnetically with the magnetic moments of a ferromagnetic host, in our system this would mean a magnetic coupling to the ferromagnetic substrate Ni. If the rare earth atom is able to occupy different states, e.g., a parallel or an antiparallel coupling, a first step to the idea of a „magnetic memory“ with single atoms is achieved.

In order to perform the first basic check on these ideas, we intend to ask for two shifts of $^{149}$Gd/$^{149}$Eu. These tests are required to study the feasibility of $^{149}$Eu as a PAC probe for interface experiments at ISOLDE.

If these tests are successful, we shall apply for beamtime with a separate proposal.

*Request: 2 shifts of $^{149}$Gd.*
References:


[9] See Refs. 3,4,5 of the appendix.


Summary of

Beam-Time Requests for PAC measurements at ASPIC

2 shifts of $^{77}$Br for $^{77}$Se on Fe and in Fe,Co
10 shifts of $^{97}$Rb for $^{79}$Kr on Ni, Ni/Pd.
8 shifts of $^{111m}$Cd for measurements of the polarization of Pd
2 shifts of $^{149}$Gd for tests on a Ni/Pd substrate
Appendix: Publications of experiments performed with ASPIC:

[1] Chemisorption of isolated Br atoms on Si(100)2x1 studied by PAC,
    J. Lohmüller, H. H. Bertschat, H. Granzer, H. Haas, G. Schatz, W.-D. Zeitz,
    and the ISOLDE Coll.,

[2] Magnetic Hyperfine Fields at Se Adatoms on Ni Surfaces,
    H. Granzer, H. H. Bertschat, H. Haas and W.-D. Zeitz, J. Lohmüller, G. Schatz,
    and the ISOLDE Coll.,

    H. H. Bertschat, H. Granzer, H. Haas, R. Kowallik, S. Seeger, W.-D. Zeitz,
    and the ISOLDE Coll.,

[4] Static Magnetic Hyperfine Fields in Magnetically Polarized Pd
    H. H. Bertschat, H.-H. Blaschek, H. Granzer, K. Potzger, S. Seeger, W.-D. Zeitz,
    H. Niehus, A. Burchard, D. Forkel-Wirth, and ISOLDE – Collaboration,

[5] Correlation between local magnetic and structural properties at the Ni/Pd interface,
    K. Potzger, H. H. Bertschat, A. Burchard, D. Forkel-Wirth, H. Granzer, H. Niehus,
    S. Seeger, W.-D. Zeitz, and ISOLDE – Collaboration,

[6] Interface Magnetism Using Radioactive Atoms,
    H. H. Bertschat,

(7) "Nuclei as secret agents"
    James Gillies in
    CERN COURIER 38, 17 (1998).

Diploma and PhD Theses (on the basis of the above listed publications):

1.) S. Seeger; PhD
    Untersuchungen zum Magnetismus in Pd-Ni-Schichtsystemen mit der nuklearen Sonde
    $^{100}$Pd/$^{100}$Rh.

2.) J. L. Lohmüller; PhD
    Chemisorption von Brom auf den Siliziumoberflächen Si(100)2x1 und Si(111)7x7,
    Universität Konstanz, 1995,

3.) H. Granzer; PhD
    PAC-Untersuchungen zum Grenzflächenmagnetismus von Ni und Ni/Pd,
    Freie Universität Berlin, 1996,

4.) K. Potzger; Diploma
    Struktur und Magnetismus an einer NiPd-Grenzfläche
    Humboldt-Universität zu Berlin, 1998.