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CERN/ISC 95-4
ISC/P29 Add. 1
19.01.1995



SEP
CERN-ISC
95-4

SURFACE AND INTERFACE STUDIES WITH RADIOACTIVE IONS

Berlin¹-CERN²-Konstanz³-Århus⁴ Collaboration

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Berlin
1995

SUMMARY

Monolayer-resolved measurements at interfaces by means of magnetic and electric hyperfine interactions were proposed in our ISOLDE experiment IS 318. First results on silicon surfaces and on magnetic multilayer systems are obtained and briefly reported. In order to complete these experiments, several runs at ISOLDE are requested and the experimental procedures are described.

INTRODUCTION

The experiment IS 318 "Surface and interface studies with radioactive ions" was started to obtain information on structural and magnetic properties at microscopic scales for several model systems in order to develop a better understanding of the properties of two classes of materials:

- 1) semiconductor-metal contacts,
- 2) metallic multilayer systems.

During the first year (1994), exploratory experiments were performed ad 1) on semiconductor *surfaces*, ad 2) on *ferromagnetic* multilayer systems.

In our UHV chamber ASPIC, carrier-free radioisotopes provided from the BOOSTER-ISOLDE mass separator can be deposited *with extreme cleanliness* on surfaces. This is essential for all investigations on surfaces and interfaces, especially for semiconductors. After the samples are loaded with radioactive probe atoms, no additional contamination can generally be detected. In laboratory experiments (University Konstanz or HMI Berlin) contaminations by accompanying elements could often not be avoided to a comparable degree and were thus the cause for severe problems.

We describe the specific motivation for the particular experiments in progress, then we give a brief status report on the results achieved so far, and finally we propose further experiments with beam time requests.

1 Semiconductor Surfaces

Our final aim in this project is to study semiconductor-metal contacts with radioactive ions, since this method is most suitable to provide information *exactly* from the interface. Before a semiconductor surface covered with a metal can be successfully investigated, knowledge about the position of the probe atom on a clean surface is a necessary prerequisite. Therefore, we have started to study the adsorption sites of various probe atoms on semiconductor surfaces as a function of temperature.

1.1 Status of Experiments with ^{77}Br on $\text{Si}(100)2\times 1$ surfaces

The halogen probe ^{77}Br (half-life 57 h) was obtained either from a ^{77}Kr beam or a ^{77}Rb beam. Isolated ^{77}Br atoms were adsorbed on $\text{Si}(100)2\times 1$ surfaces at a temperature of 90 K. With ^{77}Br decaying to the PAC nucleus ^{77}Se , a single electric quadrupole interaction frequency of $\nu_Q=526$ (7) MHz was measured (see Fig. 1), indicating the occupancy of a well defined position at the surface. With increasing temperature, a depopulation of this position in favor of another one with $\nu_Q=482$ (7) MHz was observed around 320 K (see Fig. 2a). These measurements yield detailed information about the silicon-bromine system, which is also of interest for technical applications [1]. With the help of theoretical models [2], the different diffusive displacements could be explained. At lower temperatures, the bromine atoms occupy sites at the dimer rows, characteristic for this reconstructed surface. Above RT diffusion along these rows to missing dimer sites occurs ending up at step sites (tentative assignment). Thermal desorption occurs at 650 K (see Fig. 2b).

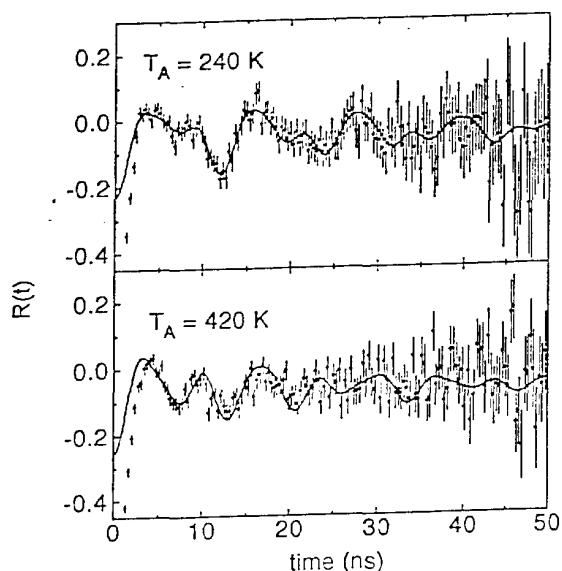


Fig. 1: PAC time spectra for ^{77}Br on $\text{Si}(100)2\times 1$ surface for two different annealing temperatures.

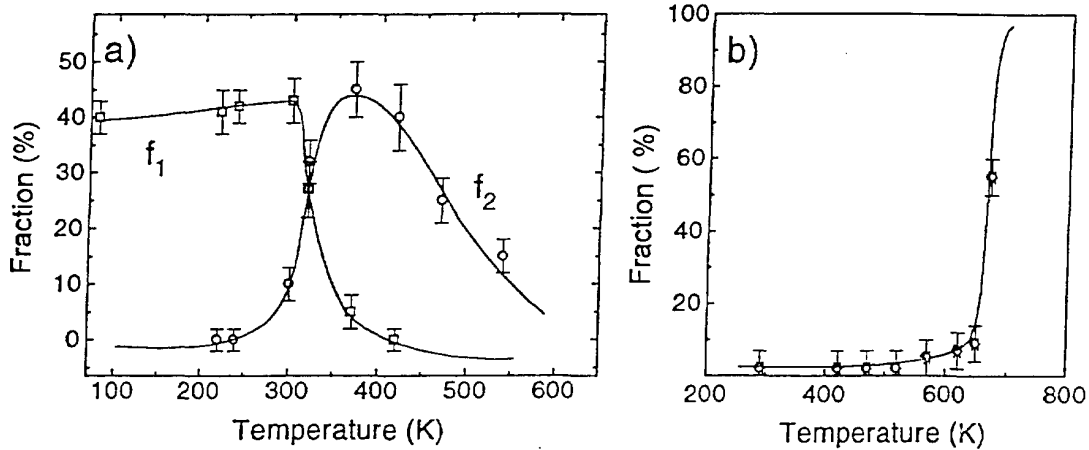


Fig. 2: a) Fractions f_1 and f_2 of probe atoms exposed to two different field gradients as a function of annealing temperature. The solid lines are guides to the eye.
b) Fraction of desorbed ^{77}Br probe atoms as a function of annealing temperature.

1.2 Future experiments on silicon surfaces

- In order to verify the above mentioned findings, we plan experiments on vicinally cut crystals where stepped surfaces are obtained. Thus preferential surface positions are offered to the probe atoms.
- At a Si(111) surface with the well known 7×7 reconstruction signals characteristic for Br atoms at two distinct sites have already been observed in our preliminary experiments. Here we plan a complete temperature dependent measurement.
- The system In on Si-surface has been subject of successful experimental PAC studies using the isotope $^{111}\text{In}(\text{Cd})$ [3, 4], but diffusion experiments on isolated probe atoms on the Si(100) 2×1 surface were problematic until now. This is probably due to carbon and chlorine contaminations caused during the deposition. At ISOLDE, investigations on very clean surfaces with the probe nucleus $^{111\text{m}}\text{Cd}$ on the Si(100) 2×1 surface offer a promising alternative to investigate isolated adatoms.

2) Ferromagnetic multilayer systems

Out of the huge amount of magnetic multilayer systems of interest, we have concentrated on Ni-Pd-systems as mentioned in our original proposal. In simple cases, they consist of Ni or Pd single crystals covered with a few monolayers of Pd or Ni, respectively. It is theoretically predicted [5] and experimentally confirmed [6], that Pd polarizes magnetically in contact with a ferromagnetic metal. Ni-Pd is a most interesting model system, where *monolayer-resolved* measurements are necessary for the quantitative determination of the magnetic profile in the region close to the interface. It is of great importance to study the magnetic properties (in our case the magnetic hyperfine field at the probe nuclei) in Pd in dependence of the distance from the Ni crystal as well as to study the magnetic properties within the Ni crystal close to the interface in dependence with the number of Pd monolayers grown on it.

In a laboratory experiment at HMI Berlin we had started to measure the Pd polarization with the PAC probe $^{100}\text{Pd}/^{100}\text{Rh}$ and found a magnetic interaction of a surprisingly long range [7]. However, no detailed study of the depth dependence was possible since no *resolved* spin precession of the 4d element probe ^{100}Rh could be observed. One possible explanation is the assumption of very strong magnetic hyperfine fields at the ^{100}Rh nuclei in polarized Pd. The magnetic hyperfine field of ^{100}Rh in Ni has been measured to be 20.2T at RT [8] leading to precession frequencies just at the limit of time resolution of conventional PAC detector systems. If the magnetic field exceeds considerably this value in polarized Pd due to the larger lattice constant, the frequencies are not resolved. It therefore seems advisable to repeat these experiments with nonmagnetic probe atoms such as Cd or Se, and ^{111}Cd and ^{77}Se are easily available at ISOLDE.

2.1 Status of Ni-Pd experiments

- a) Magnetic hyperfine fields at impurities incorporated in the Ni surface have been measured earlier in laboratory experiments using ^{111}In [9]. In order to take advantage of these results, we have used $^{111\text{m}}\text{Cd}$ from ISOLDE, without any contaminations as mentioned above. Our magnetic measurements in the Ni surface and electric quadrupole interaction measurements in the Pd surface essentially confirmed the earlier data [9, 10].

Considerable progress has been achieved in the present experiment series when the Ni surfaces were covered with one or several monolayers of Pd. In contrast to our earlier measurements with $^{100}\text{Pd}/^{100}\text{Rh}$ [7], we did observe spin precession frequencies, usually showing several fractions. This is expected, because close to the surface of ferromagnetic crystals combined interaction

$$\omega_i = \omega_i(\omega_Q, \omega_L)$$

will be observed, consisting of

$$\begin{aligned} \omega_Q &= e Q V_{ZZ} / 4I(2I-1)\hbar && \text{electric quadrupole interaction and} \\ \omega_L &= g_N \mu_N B_{hf} / \hbar && \text{magnetic dipole interaction.} \end{aligned}$$

Fig. 3 shows an example, where a Ni(111) surface (after Cd probe atoms had been deposited on it) was covered with three monolayers of Pd, at a temperature of 90 K.

The frequencies obtained cannot be identified with any of the known surface positions nor with the known magnetic interaction frequencies. Although the PAC pattern suggests a new result for magnetic interactions, a definite conclusion cannot be drawn from these exploratory experiments .

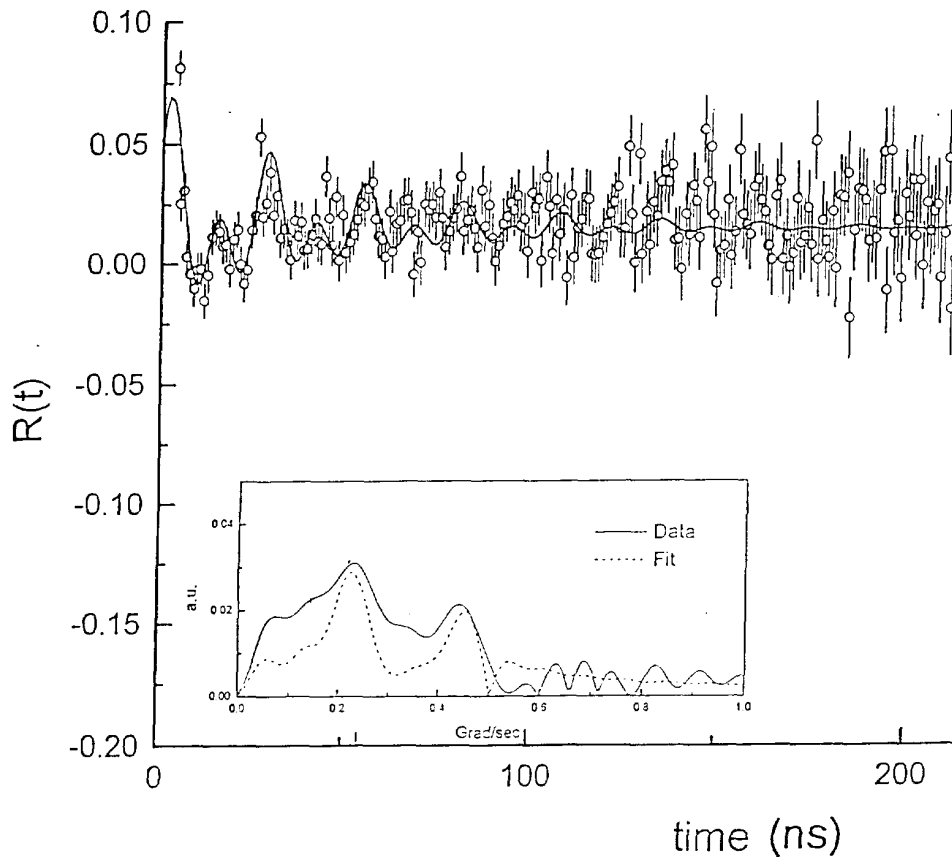


Fig. 3: PAC spectrum of $^{111m}\text{Cd} / ^{111}\text{Cd}$ on Ni(111) covered with 3 monolayers of Pd at 90 K.

- b) When Rb or Kr beams were produced at ISOLDE, we have used $^{77}\text{Br}/^{77}\text{Se}$ probe nuclei as a new possibility for surface and interface studies. ^{77}Br was positioned on Ni(111) surfaces with the aim to measure for the first time the magnetic hyperfine field for adatoms on top of a ferromagnetic surface. Magnetic hyperfine fields are known for most of the elements of the periodic table as impurities *in* the ferromagnetic elements Fe, Co, Ni. These values are quite well reproduced within theoretical models [e.g. 11]. In no case any hyperfine field has yet been measured for impurities *on top* of a surface. There the most drastic change in coordination number should lead to a drastic change in the hyperfine field value. In our experiment for $^{77}\text{Br}/^{77}\text{Se}$ on Ni(111) at different temperatures a single interaction frequency was observed (see Fig. 4). However, from this experiment alone it cannot be concluded to which extent the interaction is of magnetic or of electric origin.

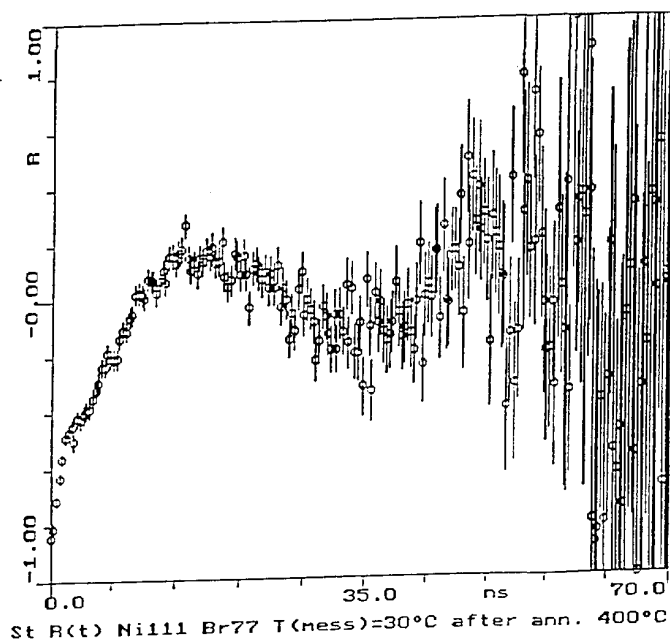


Fig. 4: PAC spectrum of $^{77}\text{Br}/^{77}\text{Se}$ on Ni(111) surface at 300 K.

2.2 Future experiments on Ni-Pd

- a) A systematic investigation of the depth dependence of the *magnetic* interaction of Cd in palladium grown on a nickel single crystal is planned. In order to disentangle magnetic and electric interactions, the nickel crystal will also be replaced by a respective copper one. For a cross-check of these experiments, ^{111}Cd will be embedded into a Pd single crystal and Ni layers will be grown on top of it in order to polarize Pd magnetically.
- b) The $^{77}\text{Br}/^{77}\text{Se}$ -on-nickel experiment shall be continued to higher temperatures. Above the Curie temperature, magnetic contributions to the hyperfine interactions are excluded.

3. Beam-time request

1 x 3 shifts of ^{111}Cd	1. half of 1995	Ni-Pd multilayers
1 x 3 shifts of ^{77}Br	1. half of 1995	Si(100)
1 x 3 shifts of ^{77}Br	2. half of 1995	Ni surface
1 x 3 shifts of ^{111}Cd	2. half of 1995	Ni-Pd multilayers
1 x 3 shifts of ^{111}Cd	1. half of 1996	Si(111)
1 x 3 shifts of ^{77}Br	1. half of 1996	Ni-Pd multilayers

18 shifts

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