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Proposal to the ISOLDE Committee

Radioactive Ions for Surface Experiments

Berlin[1]-CERN[2]-Konstanz[3]-Århus[4] Collaboration

H.-H. Bertschat[1], D. Forkel[2], H. Haas[2], R. Kowallik[1], B. Luckscheiter[3], A. Mayer[3], B. Moore[a], G. Schatz[3], S. Seeger[2], G. Weyer[4], and W.-D. Zeitz[1]

Spokesman: H. Haas

Contactman: H. Haas

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[a] McGill University, Montreal, Canada

Summary

The results of experiment IS31 at ISOLDE and a series of complementary experiments at Konstanz have demonstrated the great potential of perturbed angular correlation (PAC) measurements to study isolated impurities on surfaces. Up to now only metallic systems could be investigated in detail using the probe isotopes ^{111m}Cd and ^{111}In.

It is now proposed to use the unique possibilities offered by the combination of the Booster ISOLDE Facility equipped with a UHV beamline and the new UHV chamber for surface physics (ASPIC) recently constructed by the collaboration and successfully tested at ISOLDE-3 to extend these measurements to other probe isotopes, semiconductor surfaces, and magnetic systems. The planned PAC experiments include:

1) Investigation of the surface sites and diffusion properties of ^{111m}Cd on Si and Ge surfaces via the electric field gradient with special emphasis on segregation phenomena.

2) Study of the magnetic nature of non-metallic impurities on ferromagnets with ^{79}Rb and ^{77}Kr sources.

3) Measurement of the magnetic structure of Pd-Ni interfaces using isolated ^{100}Pd nuclei.

Information about the local dynamical behaviour of surface atoms can be obtained via Mössbauer spectroscopy (MS). It is therefore intended to study some of the systems characterized by PAC also with emission MS of ^{119}In-^{119}Sn and ^{119}Sb-^{119}Sn, a technique developed in experiment IS20.

A parasitic test of a novel concept for direct deposition of ion beams on surfaces is being planned.

I. MOTIVATION

Surfaces and interfaces are a field of great interest in several areas of physics. For the study of pure surfaces or for monolayer coverages a wide variety of experimental methods has been developed. Very sparse data, however, exist for isolated impurities or defects on surfaces. It therefore would be quite important to get microscopic information on electrical and lattice structure for such systems.

In recent years the corresponding problem for impurities and defects in bulk metals has been very successfully attacked with the use of nuclei as probes. Particularly experiments with perturbed angular correlation (PAC) and Mössbauer spectroscopy (MS) have in this case given detailed insight. Both techniques require as little as 10^{11} atoms in favourable cases, much less than a monolayer (~ $10^{15}/cm^2$). The application of these methods to study surfaces therefore can give unique microscopic information, particularly when combined with an on-line isotope separator like ISOLDE. For PAC the technical problems of such experiments could be overcome in laboratory experiments at Konstanz and in the pilot experiment IS31 performed at ISOLDE-2, where in particular a quantitative study of surface diffusion for Cd and In on stepped Pd(111) surfaces could be completed. It has thus been established that PAC measurements at least with ^{111}Cd are well suited to characterize isolated impurity sites on metal surfaces. The number of systems investigated up to now is very small, however, calling for further systematic measurements. In this context it is also of great interest to use other probe isotopes like. ^{79}Rb and ^{77}Kr, only available at ISOLDE. Clearly the next stage should be the investigation of semiconductor surfaces and magnetic materials as well as possibly noble gas covered surfaces, where the primary interest are phase transitions in 2-dimensional systems. In view of their catalytic activity, the alkali-covered Pd and Pt surfaces are also prime candidates for further experiments.

The nuclear techniques PAC and MS should be particularly well suited for the investigation of interfaces, where the standard surface techniques are not applicable at all. Both the lattice structure and the magnetism at interfaces are a topic of great present interest. Well defined interfaces may be produced by controlled coverage of previously characterized surfaces.

The direct deposition of radioactive ions at surfaces would furthermore open up many new possibilities for surface and interface studies. Further progress can be expected in the development of new surface techniques like recoil angular distribution or surface channeling.

II. STATUS

In the first experiments performed at ISOLDE the system ^{111}Cd on Mo(110) was studied in detail /1/. We have demonstrated that the

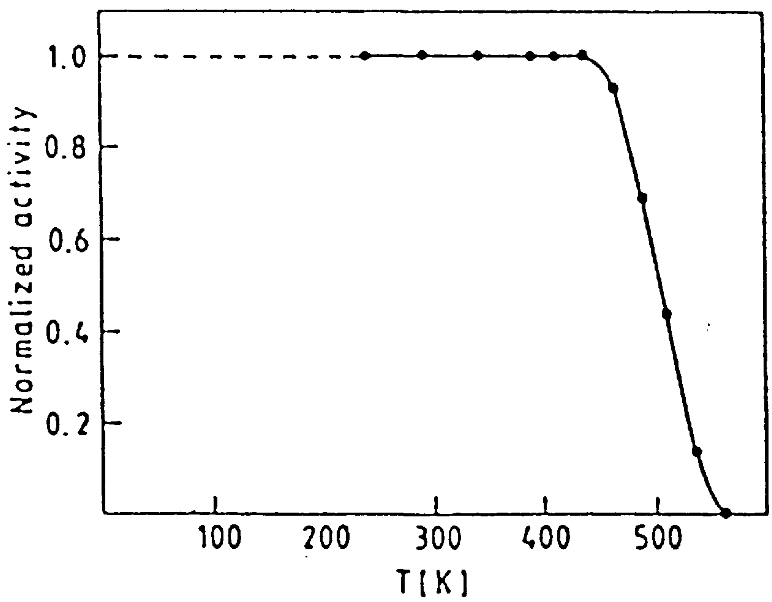

Fig.1: Desorption isochrone for ^{111m}Cd from Mo(110)

deposition of isolated radioactive ^{111m}Cd (48 min) atoms and their observation via well defined PAC spectra is possible. Fig.1 shows the simple desorption isochrone observed for this system. Such data have also been recorded earlier with stable isotopes, but due to the large coverage necessary, these curves are much more complex and very difficult to interpret. Also the case of ^{79}Rb (20 min) has been successfully tested.

In a further series of experiments at ISOLDE-2 we have been able to characterize the sites taken up by isolated In and Cd atoms on the model surface Pd(111) from the first landing position on the terrasses up to their incorporation into the top surface layer. The electric field gradient (efg) as observed with PAC was used as signature /2/. In Fig.2 the identified sites are shown schematically. From the temperature dependence of their occupation the adatom diffusion for this system could be studied in detail /3/.

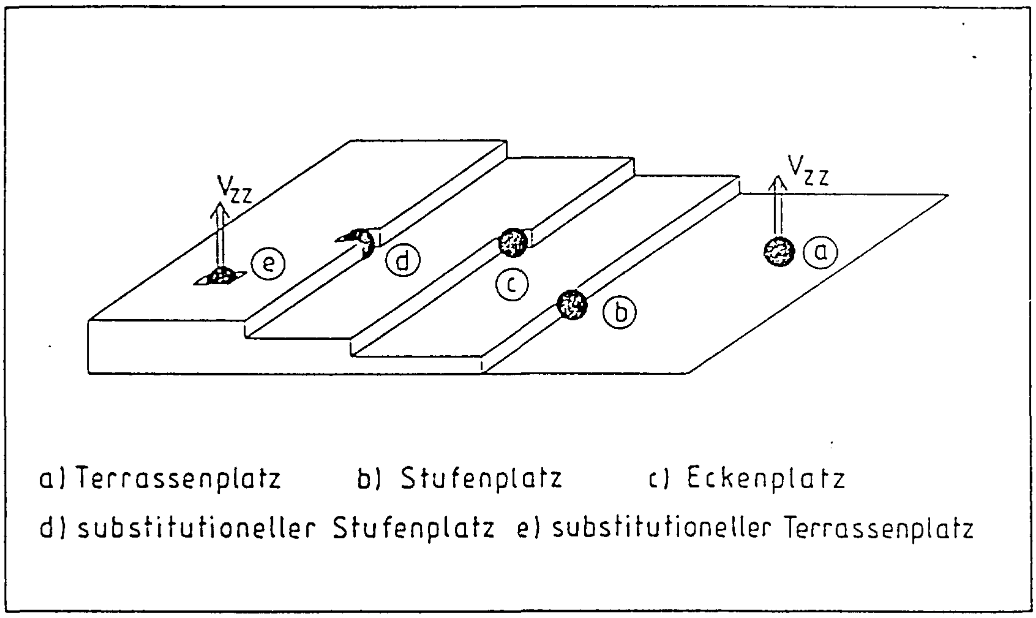

Fig.2: Identified surface sites occupied by In and Cd adatoms on Pd(111)

This complete picture of surface diffusion for the model system clearly has demonstrated the great potential of the nuclear techniques at ISOLDE to investigate isolated impurities at surfaces and interfaces on a microscopic scale.

In a series of off-line experiments at Konstanz the isotope ^{111}In (2.7 d) has been deposited in a similar way. Monolayer coverages on In /4/ as well as isolated impurities on several metallic surfaces /5/ could be studied by PAC. Also first results for a magnetic system, In on Ni /6/, could be obtained.

III. APPARATUS

The central problem in the use of radioactive atoms for the study of surfaces is the sample preparation and the deposition of the probe nuclei.

For the project IS31 we had installed at ISOLDE-2, beam line 2A, a somewhat improvised UHV apparatus. This equipment is still available and could be used in some of the parasitic tests foreseen in the present proposal.

Our new Apparatus for Surface Physics at ISOLDE CERN ("ASPIC"), specially designed for operation at ISOLDE, is a versatile system for surface and interface measurements (see Fig.3). It is fully based on the technique of two-stage deposition as already developed in experiment IS31: The ISOLDE isotopes are evaporated in a small chamber out of the rotatable implantation oven onto a carefully cleaned and degased transfer foil mounted on a special transportation manipulator that takes the activity into the main chamber. The samples, mechanically and chemically polished, can be transferred from a 12 sample stack onto the low temperature measuring manipulator with an additional small handling manipulator. Surface cleaning is accomplished with the

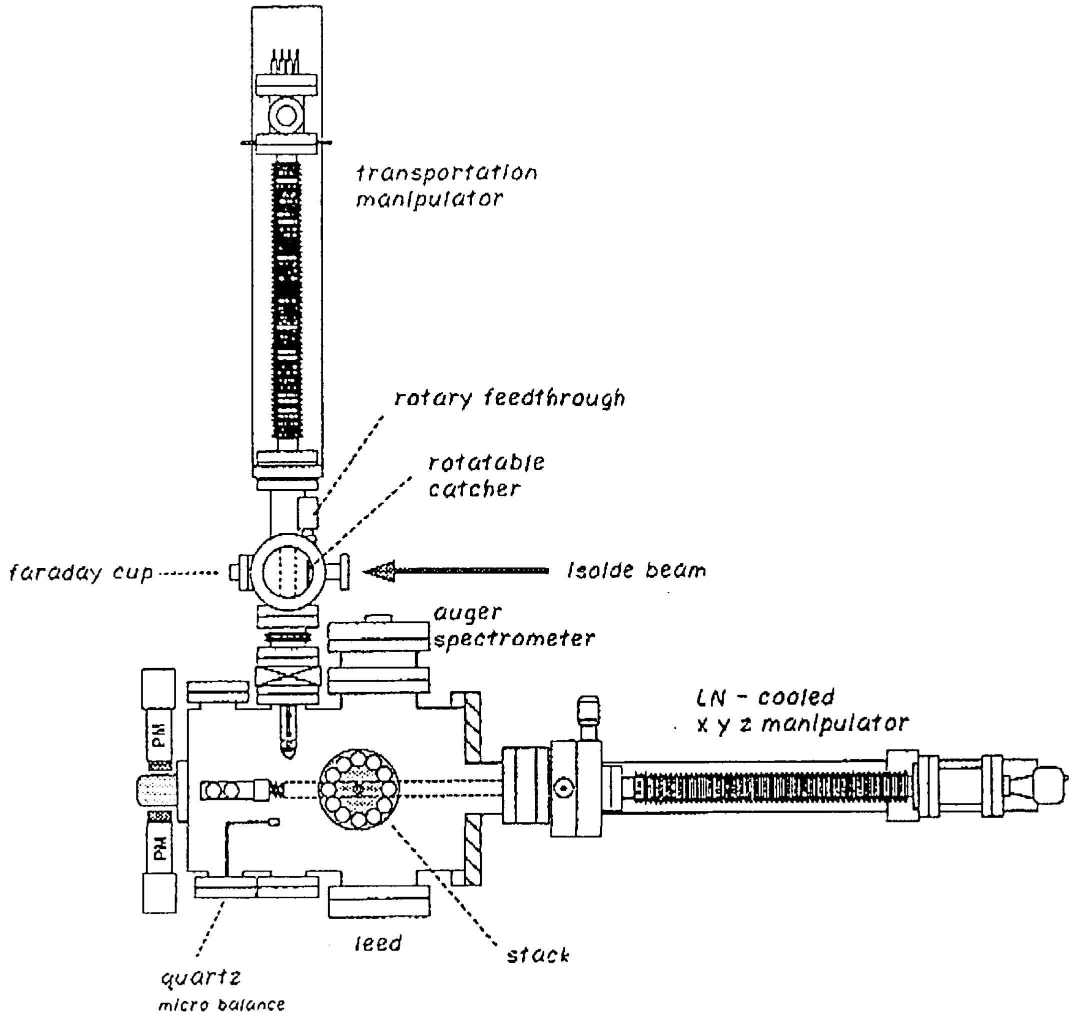

Fig.3: Schematic layout of the new surface physics chamber ASPIC

use of a sputter gun, electron beam heating, resistive heating, and a gas inlet system. Evaporation devices provide the option to deposit additional non-active materials to produce covered surfaces or thin films, the growth of which is controlled by a quartz microbalance. The sample purity may be checked with a CMA Auger spectrometer. The system also contains a LEED system for surface structure characterization. To eliminate vibrations that might impair the MS experiments proposed, the chamber can be fully operated with nonmoving pumps. This feature will be also of great importance for the planned installation of a tunneling microscope, now being developed in Konstanz. Sample manipulations and data taking are controlled by a UNIX computer with an XWINDOWS program.

When installed at the new ISOLDE facility this instrument will allow a very systematic use of radioactive isotopes for surface and interface studies. In December 1990 ASPIC was successfully tested at ISOLDE-3, where a first result for ^{111m}Cd on a Cu(100) surface could be obtained. At present we are using the slightly modified system in Berlin with commercially obtained radioisotopes. The installation there will allow us to perform PAC measurements of monolayers on magnetic surfaces, the planned next step in our research programme. These measurements will also permit us to test all the possibilities offered by the new setup. After the interruption of ISOLDE operation, however, the full exploitation of its potential for on-line work can be resumed at the Booster ISOLDE facility. The RISE Collaboration is prepared to assist in the installation of the UHV beamline there. In particular the use of short lived PAC isotopes and other nuclear techniques like Moessbauer spectroscopy, recoil desorption, or surface channeling shall benefit from the versatility of ASPIC.

IV. PAC EXPERIMENTS

The PAC technique for surface studies has now reached a stage where the systems to be investigated may be selected due to their theoretical or technological relevance and not only their experimental feasibility. Out of the wide spectrum of possible applications three examples have been chosen for the first measurements at the Booster ISOLDE Facility.

1)Semiconductor surfaces

The contact between semiconductors and metals is of utmost technological importance in modern electronics. PAC is a unique tool to investigate the geometrical and electronic structure of such interfaces on the atomic scale. As a first step in this direction we intend to study isolated atoms and clusters of simple metals like In and Cd on the elemental semiconductors Si and Ge. Very interesting diffusion and clustering phenomena have been observed by tunneling microscopy /7/ at higher coverage. Exactly the same systems can be studied with ^{111}Cd in the incipient phase of association. Obviously one has first to characterize the various surface sites via the efg in the same way as done earlier for metals. An interesting aspect is the problem of surface wetting and the growth of islands on clean semiconductor surfaces. Here especially the access to short-lived nuclear probes at ISOLDE is of great advantage.

In future experiments we aim at investigations of semiconductor materials which are of importance for photovoltaic applications. Local electronic structures (e.g. charge states) of matrix and impurity atoms are of special interest and can be investigated by the nuclear methods.

2)Nonmagnetic adatoms at Ni surfaces

While the magnetic hyperfine fields at metallic impurities in and on metals appear to reflect simply the magnetization of the nearest neighbour atoms, there is evidence that more complicated behaviour will be observed for non-metallic adsorbates.

With the probe atoms ^{79}Kr and ^{77}Br we have recently at ISOLDE measured the hyperfine fields in nickel /8/. Especially for Kr the extremely small field may be understood as a consequence of a cancellation of conduction electron and nearest neighbour contribution.

As both of these will be quite different on the surface, the accidental cancellation should be removed, leading to a strongly enhanced hyperfine field at surface sites relative to the bulk value.

Having accomplished these basic experiments, we plan to study the behaviour of a noble gas on metallic surfaces as function of temperature. As the deposited radioactive ^{79}Rb (decaying to ^{79}Kr) will be much stronger bound, these investigations of noble gases will not be restricted to temperatures below 50K, the desorption temperature, but may possibly be extended up to room temperature. Furthermore the behaviour of noble gases close to surfaces can be studied, because the parent Rb atoms may be covered with matrix atoms, which is not possible for the noble gases themselves.

3)^{100}Pd in Ni-Pd interfaces

Interface magnetism is a very promising topic for studies with nuclear techniques. For the non-magnetic probe atom Cd /9/ and the strongly magnetic Fe probe /10/ first data already exist.

To complete our ongoing investigation of the Ni-Pd interface, of particular interest from the theoretical viewpoint, we intend to study at ISOLDE the magnetic hyperfine field for isolated atoms on nickel surfaces using the PAC technique with thermally deposited ^{100}Pd.

Such experiments can be further extended, since impurity probe atoms may be brought into any arbitrary layer close to an interface by proper deposition techniques. In this way a monolayer resolved investigation of the magnetic behaviour of interfaces will be possible using the PAC technique in connection with ASPIC at ISOLDE.

V. MÖSSBAUER EXPERIMENTS

Mössbauer spectroscopy is a very powerful tool for obtaining microscopic information at the nucleus not only through hyperfine interaction (e.g. isomer shift, magnetic splitting, quadrupole coupling) but also through the Debye-Waller factor (DWF) that is particularly sensitive to the bonding environment. Such data for surfaces would be extremely valuable.

Only MS measurements on clean monolayer surfaces have been reported up to now /11/. No data for isolated impurities exist, demonstrating the difficulty of such experiments with conventional absorber methods. The strong sources obtainable at ISOLDE should make many measurements feasible, however.

The ^{119}Sn state, very successfully applied for questions in solids /12/, is the optimal candidate. As it is produced in the decay chain from ^{119}Cd, the same sample preparation as for ^{111}Cd PAC may be employed, resulting in completely complementary information.

A strong anisotropy of the DWF parallel and perpendicular to the surface is expected in most cases. Of particular interest is the (110) surface of bcc metals, where even stronger anisotropy in the plane could occur due to atomic jump processes at low temperature.

Very favorable conditions for ^{119}Sn MS are also found for ^{119}Sb sources that may be produced at ISOLDE in the decay chain of ^{119}Xe. In this case it would be necessary to try the method for deposition in a preliminary test.

VI. PARASITIC TEST EXPERIMENTS

Radioactive atoms at surfaces offer the possibility for a number of new experiments. The surfaces characterized via the PAC method may be used to test such methods in a controlled way. Out of several possibilities we propose to test ion deposition

The possible uses of nuclei for surface studies could be greatly enhanced if a direct deposition of the radioactive ion beam from ISOLDE on surfaces could be accomplished. With the high beam quality from ISOLDE this should in principle be possible. The blow-up of the beam spot size accompanying deceleration can be largely avoided if the retardation is performed in a very strong magnetic field that can coil up the ion path. Beam-optical calculations have confirmed this idea. The suggested deceleration system is shown in Fig.4. With the combination of a 10T magnet available at HMI Berlin and the surface chamber ASPIC

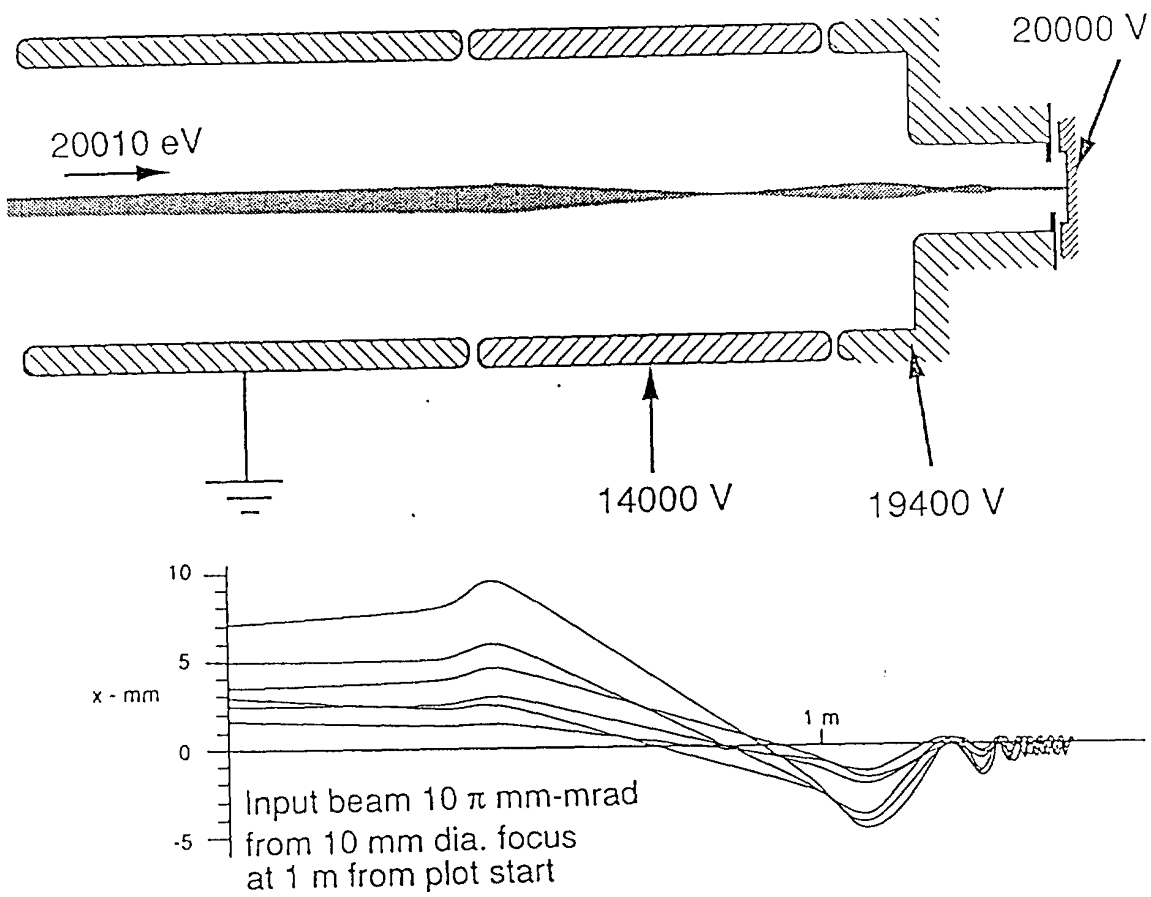

Fig.4: Ion optical system for "soft landing" experiment. Top: Electrode configuration; Bottom: Ion trajectories into 10T magnet

we are in a perfect situation to test this concept at the Booster ISOLDE Facility. To demonstrate that "soft landing" has occurred we intend to deposit ^{111m}Cd ions on Pd(111), where we know the efg signature of all surface sites from our previous experiments.

VII. BEAM TIME REQUEST

To perform the experimental programme as sketched above, we would need access to the ISOLDE separator for 4 to 6 periods of 2-3 days per year. All experiments could be performed with standard target/ion source units and in most cases the beamtime could be shared with other users, as the actual collection times will generally only be a few minutes every 1 to 3 hours.

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