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Hyperfine interaction studies with monolayer depth resolution using ultra-low energy radioactive ion beams

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Abstract

A variety of nuclear techniques rely on the incorporation of radioactive atoms to investigate the microscopic structural, electronic and magnetic properties of a material. In the past, ion implantation has been utilized to introduce these radioactive probes, resulting in a depth distribution of typically several hundreds of Å, and damaging the sample. Both implantation-related deficiencies are incompatible with the ever shrinking sizes relevant in nanostructures. This problem can be circumvented by using ultra-low energy ion beams – of the order of 5 eV, i.e. below the displacement energy of the substrate atoms. Consequently, the radioactive probes are "deposited" on top of the sample, without generating damage to the substrate. Since the implantation chamber is in vacuo connected with the molecular beam epitaxy deposition chamber, the probe layer can be introduced at any stage during the sample growth (from surface to interface)—with monolayer depth resolution. As an example, we discuss the ultra-low energy ion deposition of ^{111}In in Cr, followed by analysis with perturbed angular correlation spectroscopy. The aim of the study is to explore the magnetic ordering of Cr thin films. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

For several decades, nuclear methods have provided a powerful tool in the study of the structural, electronic and magnetic properties of materials [1,2]. Nuclear orientation either by low temperature or perturbed angular correlation (PAC) techniques as well as Mössbauer spectroscopy, provide microscopic information on the vicinity of a radioactive probe nucleus. Due to their high sensitivity (in many cases an amount of 10^{12}–10^{14} atoms, a small fraction of a monolayer, is sufficient), these techniques have shown to yield very valuable information in thin film studies, where the amount of material is often too small to apply conventional techniques [3]. Because thermal diffusion cannot result in control of the depth profile, these radioactive tracers are mainly introduced by ion implantation, typically using energies of the order of several tens to several hundreds of keV. Two major drawbacks of this technique, however, are the depth distribution of the

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implanted species (i.e. the roughly Gaussian implantation profile) and the damage caused by the deposited energy. For instance, implantation of In in Cr, with an energy of 50 keV, results in a distribution located 135 Å below the sample surface, with a FWHM of 130 Å; and the formation of approximately 570 vacancies per incoming ion. Driven by nanotechnology, both for the study of surfaces and interfaces, there has been an increasing need for monolayer (ML) sensitivity and consequently, for other ways of introducing the radioactive probes. Some groups use indirect evaporation techniques, e.g. thermally evaporating the probes from a Cu [4] or Mo foil [5] target, in which the radioactive probes are initially introduced by diffusion or implantation respectively. Alternatively, one can combine the mass selectivity and controllability of ion implantation with the extremely low energies of deposition, i.e. *low energy ion deposition* (LEID) or *soft landing* [6–8]. LEID has been used in the past for the study of surface phenomena (e.g. diffusion, incorporation, nucleation,...) [9,10]. In the present work, we present the use of low-energy radioactive ion beams for the study of thin film properties with ML depth resolution and we will show that LEID can be put forward as an ideal tool for depth dependent studies of structural, electrical and magnetic properties in state of the art materials. In order to illustrate the general concept, we will describe the deposition trace amounts of radioactive In probes on Cr for the study of the magnetic properties of Cr thin films.

2. LEID: the technique and the setup

A schematic overview of the LEID setup is shown in Fig. 1. An isotopically pure ion beam with an energy of typically 50 keV is delivered by the Leuven isotope separator. A proper mass resolution is obtained by bending the ion trajectory over an angle of 55° in the 1.5 m radius analysis magnet, thereby separating the 2.5 mm FWHM ^{111}In beam over 13.5 mm from its neighbouring masses. Subsequently, the ion beam can be swept, yielding a typical beam spot of 4×4 mm^2 on the target, and finally entering into an electrostatic deceleration lens. This deceleration stage consists of five metal electrode rings with increasing diameter and electrostatic potential along the path of the ions. The specific configuration was chosen based on simulations of the ion path depending on the number, shape and voltage of the electrodes using the program SIMION [11]. The high voltage supply to the sample holder is derived directly from the source section of the isotope separator, to which a variable negative voltage $V_2 = 0$–200 V is superimposed. An additional permanent offset $V_1 = 50$ V is used, shifting the zero point for deposition to $V_2 = 50$ V. This approach enables us to check that no ions can reach the sample for

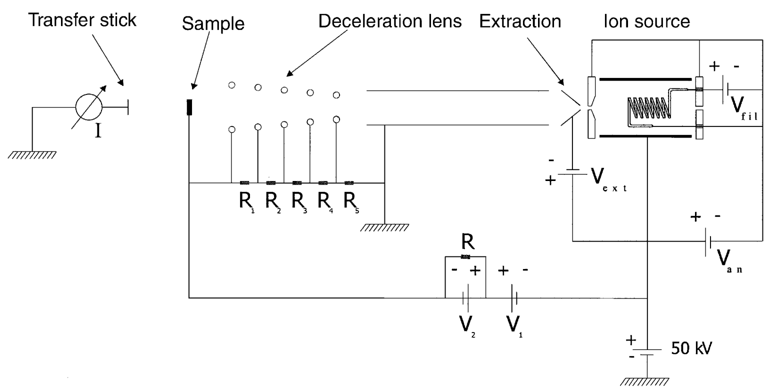

Fig. 1. Schematic diagram of the LEID set-up. The deposition energy is selected by adjusting the output voltage of the power supply V_2.

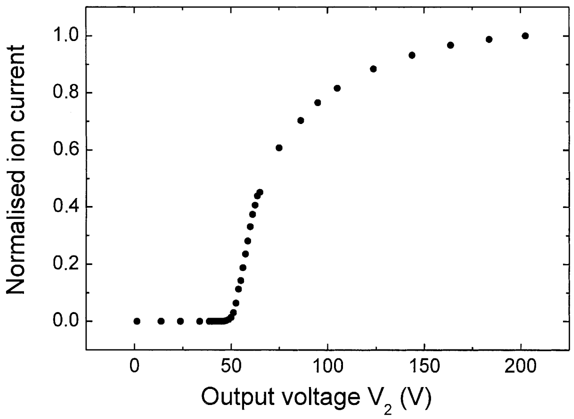

Fig. 2. Ion current through the grid as a function of output voltage V_2. The zero-point energy is set at approximately 50 V using power supply V_1.

'negative' deposition energies. A firm calibration of the deposition energy is obtained by measuring the current transmitted through a grid, put at the sample position (Fig. 2). This current is measured on a Faraday cup mounted on the transfer stick behind the grid, and is normalised to the current measured when no retarding field is applied. Below approximately 50 V (i.e. $E_{dep} = 0$ eV), no ions pass, whereas at a higher voltage, the ion current gradually increases. This *gradual* increase is related to the mesh size of the grid, and is not caused by a spread of the ion energy [7].

So far, LEID has been used to study the behaviour of individual atoms gently deposited onto the surface of a crystal. Surface diffusion mechanisms, nucleation of deposited ions, incorporation of ions in terraces, the formation of quasi-one-dimensional wires, etc. have been investigated by combining conventional and hyperfine interaction techniques [3,9,10]. To incorporate LEID in thin film and multilayer studies, a variety of sample preparation and characterization facilities should be available in the soft landing chamber, or via an in vacuo coupling. The LEID setup of the Ion and Molecular Beam Laboratory at KU Leuven is in vacuo connected with two thermal deposition units, a scanning tunneling microscope (STM), Auger electron spectroscopy (AES), low energy electron diffraction, Mössbauer spectroscopy and Rutherford backscattering (RBS) spectrometry facilities. Details on the experimental setup can be found in [7].

3. Low energy ion deposition of In on Cr

Below the Néel temperature ($T_N = 311$ K), bulk Cr exhibits a so called spin density wave (SDW), i.e. an antiferromagnet of which the magnitude of the magnetic moments vary in a sinusoidal way, with a period of approximately 21 unit cells ($\sim$61 Å). This incommensurate SDW is characterized by a spin vector $\vec{\mathbf{S}}$ and a wave vector $\vec{\mathbf{Q}}$, and is labelled longitudinal or transversal for $\vec{\mathbf{S}} \| \vec{\mathbf{Q}}$ and $\vec{\mathbf{S}} \perp \vec{\mathbf{Q}}$ respectively. In bulk Cr, the polarisation of the SDW changes from longitudinal below to transversal above $T_{SF} = 123$ K, the spin flip temperature. Commensurate antiferromagnetic ordering (i.e. no magnitude variation of the magnetic moments) has been reported for Cr alloys or in strained Cr lattices. The magnetic phase diagram for Cr thin films is found to be more complicated [12]. PAC experiments using conventional implantation of the ^{111}In probes have shown that the magnetic ordering in low dimensional chromium depends on the growth conditions. For instance, a shift of T_N and T_{SF} have been reported, as well as the collapse of the SDW for film thicknesses below the SDW period [13].

In the past, PAC experiments on Cr thin films have been performed using high energy ion implantation. It has been shown that the technique is very sensitive to the orientation of the spin. However, until now, PAC is not *directly* sensitive to the direction of the $\vec{\mathbf{Q}}$-vector. In order to derive the polarisation of the SDW consistently from a PAC experiment one should be able to probe the variation of the magnetic hyperfine field from layer to layer. Here we present a first attempt to reach that goal.

3.1. Deposition of stable ^{115}In ions

Before depositing extremely low concentrations of radioactive ^{111}In probes, extensive tests were performed with 5 eV stable ^{115}In ion beams onto a variety of substrates, using fluences of the order of 10^{15} at./cm^2 or more. First, the beam profile as well as the uniformity of the implanted region was investigated by RBS mapping. After deconvoluting the He beam profile used for RBS (approximately 1×1 mm^2), a homogeneous In profile was observed in a region of about 4 mm wide, i.e. equal to the sweeping area of the ion beam. Channeling measurements indicated that the implantation process did not induce any noticeable damage to the substrate, as expected for these extremely low energies. The absence of implantation induced craters on the surface, and the presence of small aggregates deposited onto the substrate, were confirmed by in vacuo STM. No traces of any contamination could be observed within the sensitivity of AES.

3.2. Deposition of radioactive ^{111}In ions

In order to study the polarisation of the SDW in Cr thin films, it is essential to confine the radioactive probes to 1 ML, as illustrated in Fig. 3(c) and (d). To achieve optimal crystallinity, a 100 Å thick Cr film was grown onto an epitaxial ⟨MgO⟩/Fe (75 Å)/Ag (300 Å) substrate/seed-layer structure. The roughness of the Cr layer could potentially impose a limit to the depth sensitivity of the study. Therefore, the sample surface was examined with in vacuo STM. From a scanned area of 400×400 nm^2, an *rms* roughness of approximately 0.4–0.5 nm was deduced, i.e. negligible compared to the SDW wavelength. This Cr film was covered with 2×10^{13} ^{111}In/cm^2 at an energy of 5 eV, using a beam current of approximately 150 pA. Subsequently, a second 100 Å thick Cr film was deposited, after which the sample was covered with a Ag capping layer. Hence, a 200 Å thick Cr layer is obtained, halfway decorated with ^{111}In probes as schematically illustrated in Fig. 3(c) and (d). The lateral uniformity of the deposited probes was confirmed by measuring the activity as a function of position across the sample.

3.3. Perturbed angular correlation experiment

The PAC measurement was performed at room temperature, in a four-detector set-up and with the sample normal in the plane of the detectors and oriented at 45° relative to the start detectors. The PAC spectrum generally reflects a more or less periodic oscillation with a frequency directly related to the magnetic hyperfine field at the probe nucleus. Furthermore, the spectrum can contain two harmonics, their relative presence being a direct measure for the orientation of the hyperfine field ($\vec{S}$-vector). Several possible models were considered when analysing the PAC time spectrum (Fig. 4). The shape of the spectrum excludes the presence of a commensurate SDW phase [3], which would result in a periodic oscillation of the amplitude with little damping. Alternatively, an incommensurate SDW should be considered for

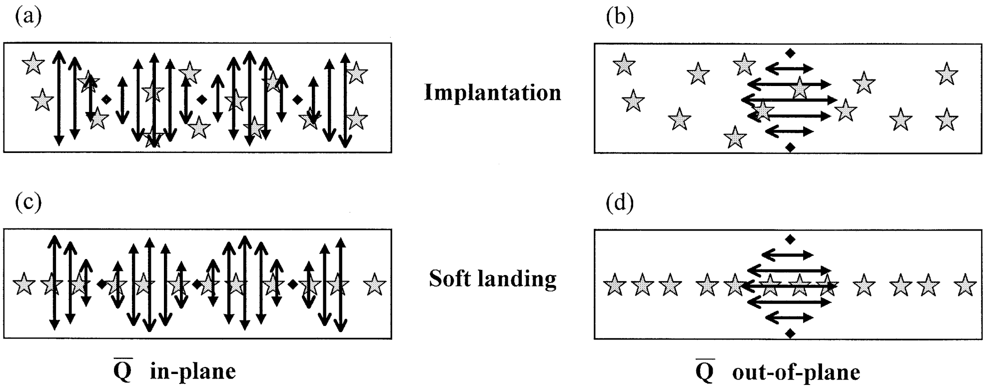

Fig. 3. Schematic configuration of a transversal SDW with its wave vector in the plane (a,c) or out of the plane (b,d) of the Cr layer. The confinement of the radioactive probes in case of soft landing (c,d) results in monolayer depth sensitivity, which cannot be obtained when using conventional ion implantation (a,b).

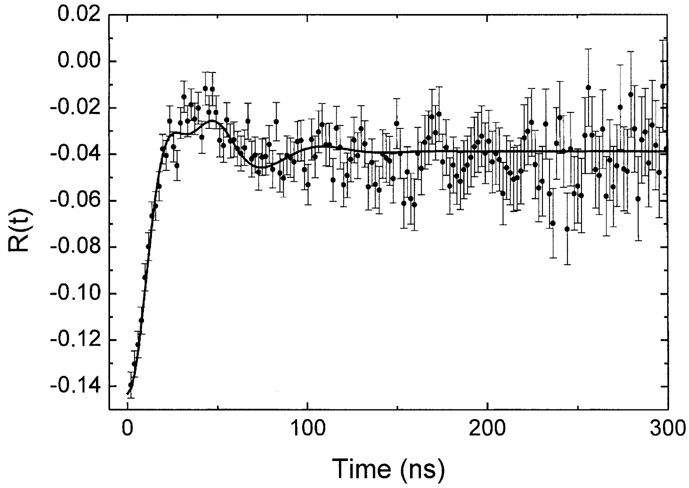

Fig. 4. PAC time spectrum of a 200 Å thick Cr layer, which was halfway decorated with ^{111}In probes, using ion beam deposition at 5 eV. The solid line is a fit assuming a longitudinal SDW.

which the spins ($\vec{\mathbf{S}}$-vector) are in-plane with the Cr-film, a feature that is readily derived from this PAC experiment. Distinction between either a transversal SDW ($\vec{\mathbf{Q}}$ out-of-plane) or a longitudinal SDW ($\vec{\mathbf{Q}}$ in-plane) is made by a fit procedure with the magnitude of the frequency (hyperfine field) as the free parameter. If the SDW were propagating along the surface normal ($\vec{\mathbf{Q}}$ out-of-plane, Fig. 3(d)), obviously, there would be no information on the position of the probe layer with respect to the (anti)nodes of the SDW. To account for this, a possible phase shift is entered in the analysis. Moreover, the depth distribution of the In probes, as inferred from STM (see above), was taken into account. For the model consisting of a SDW propagating along the surface normal, the experimental spectrum could be reproduced for a phase shift of approximately $\pi/4$, but assuming a hyperfine field substantially different from the values known in literature [3]. Much more consistent results were obtained using the model assuming an in-plane $\vec{\mathbf{Q}}$ vector, i.e. a longitudinal SDW, yielding a hyperfine field in agreement with the literature.

Although neither of the two alternatives could be ruled out completely as yet, measurements after In depositions at various depths in the Cr layers will allow full characterization of the SDW polarization.

4. Conclusion

LEID allows to deposit, with ultra low energies – down to a few eV, small amounts of radioactive tracers in a controlled and non-destructive manner. Based on the present and existing PAC studies of the Cr magnetism, we illustrated that LEID gives way to the so far experimentally almost inaccessible observation monolayer resolved hyperfine fields. Consequently, for other nanostructures, the technique of soft landing of radioactive probes can become an essential tool for the study of the microscopic properties of magnetism or the magnetic moment profile, as well as the phase formation at interfaces and near surfaces. These data are excellent testing grounds for ab initio model calculations which recently became very reliable for electron density distributions. It is expected that this LEID facility will form the basis for hyperfine interaction studies of structural, electrical and magnetic properties of thin films, nanostructures and their interfaces, where ML depth sensitivity is crucial.

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References

[1] G. Schatz, A. Weidinger, Nuclear Condensed Matter Physics, Wiley, West Sussex, 1996.
[2] G. Langouche (Ed.), Hyperfine Interaction of Defects in Semiconductors, Elsevier, Amsterdam, 1992.
[3] J. Meersschaut, J. Dekoster, S. Demuynck, S. Cottenier, B. Swinnen, M. Rots, Phys. Rev. B 57 (1998) R5575.
[4] T. Klas, R. Fink, G. Krausch, R. Platzer, J. Voigt, R. Wesche, G. Schatz, Surf. Sci. 216 (1989) 270.
[5] ISOLDE Collaboration, E. Kugler, D. Fiander, B. Jonson, H. Haas, A. Przewloka, H.L. Ravn, D.J. Simon, K. Zimmer, Nucl. Instr. and Meth. B 70 (1992) 41.
[6] J. Dekoster, B. Degroote, H. Pattyn, G. Langouche, A. Vantomme, S. Degroote, Appl. Phys. Lett. 75 (1999) 938.
[7] B. Degroote, Ph.D. Thesis, Katholieke Universiteit Leuven, 2001, available at www.fys.kuleuven.ac.be/iks/nvsf/nvsf.htm.
[8] C.R. Laurens, L. Venema, G.J. Kemerink, L. Niesen, Nucl. Instr. and Meth. B 129 (1997) 429.
[9] C.R. Laurens, M.F. Rosu, F. Pleiter, L. Niesen, Phys. Rev. Lett. 78 (1997) 4075.
[10] B. Degroote, A. Vantomme, H. Pattyn, K. Vanormelingen, submitted to Phys. Rev. B.
[11] D.A. Dahl, D.E. Delmore, A.D. Appelhans, Rev. Sci. Instr. 61 (1990) 607.
[12] P. Bödeker, A. Schreyer, H. Zabel, Phys. Rev. B 59 (1999) 9408.
[13] J. Meersschaut, J. Dekoster, R. Schad, P. Beliën, M. Rots, Phys. Rev. Lett. 75 (1995) 1638.