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PROPOSAL
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(n,p) emission channeling measurements on ion-implanted beryllium

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Abstract

We propose to perform emission-channeling measurements using thermal neutron induced proton emission from ion-implanted ^{7}Be. The physics questions addressed concern the beryllium doping of III-V and II-VI semiconductors and the host dependence of the electron capture half-life of ^{7}Be.

Technique

Emission-channeling (EC) is a powerful experimental technique to pin down the exact site of atoms in a crystal lattice, see e.g. ref. [1,2]. Typically radioactive isotopes are implanted that subsequently emit low-energy betas, conversion electrons or alphas.
Unfortunately several elements have no radioisotope with suitable half-life and charged particle energies. However, if the element in question has an isotope with high cross-section for (n,p) or (n,α) reactions, the charged particles can be produced by exposing the ion-implanted sample to an intense neutron flux.

This technique has been explored by Biersack et al. already over 30 years ago [3,4]. However, only for lithium (^{6}Li) and boron (^{10}B) doped samples the count rates were sufficient. EC with beryllium had been proposed also [5], making use of the huge cross-section for thermal neutron capture of ^{7}Be: ca. 40000 barn for ^{7}Be(n,p). Unfortunately no intense radioactive ion beams of ^{7}Be were available at that time to achieve the required sample doping. Meanwhile intense ^{7}Be beams can be produced at ISOLDE, allowing the doping of samples with several 10^{15} atoms of ^{7}Be within few hours [6].

We want to use this opportunity to study via ^{7}Be(n,p) EC several fundamental questions that could not be addressed so far.

Physics aim

The first two physics motivations come from applied physics:

1. Today Mg is used as the acceptor impurity in GaN (III-V) semiconductors with important commercial applications, although its acceptor activation energy of 210 meV is not ideal. Theoretical predictions claim that Be might work better or at least as good, with predicted electrical acceptor activation energies ranging from 90 to 250 meV. However, in experiments Be does not work well as acceptor, but it is not exactly known why. One theory put forward in that respect is that Be is not incorporated on Ga sites in GaN but interstitially, where it is not electrically active [7]. We therefore propose to study this intriguing case of the lattice location of Be in GaN and AlN semiconductors.

2. Be has also applications in the field of II-VI semiconductors where it is used, for instance, in ternary $Zn_{1-x}Be_xO$ compounds [8,9]. While pure ZnO is a technologically promising hexagonal II-VI semiconductor with a band gap of 3.3 eV, BeO is a hexagonal compound with a bandgap of 10.6 eV. Hence, by means of replacing Zn with Be and forming a ternary alloy, the stochiometry x allows to adjust the band gap to higher values than in pure ZnO, which is an essential feature for the realization of quantum-well based devices based on ZnO. Here we propose to study the open question to what level Be is incorporated on Zn sites.

3. Another motivation comes from nuclear physics: The decay rate of low-Z elements undergoing electron capture may be susceptible to changes of the surrounding environment [10]. If ^{7}Be is hosted in a matrix that causes the loss of part of its 2s electrons, its half-life may be affected noticeably. The astrophysical interest (the ^{7}Be decay rate is an important parameter in the Standard Solar Model [11]) and the availability of pure ^{7}Be beams for implantation triggered recently great experimental and theoretical activity in this field [12-17]. A promising attempt to explain the observed half-life changes quantitatively relates them to the calculated number of lost 2s electrons for ^{7}Be implanted into different media. The first-principles electronic structure calculations use a density functional technique [14]. However, a big uncertainty (up to a factor two!) arises due to the unknown distribution of the implanted ^{7}Be over different possible lattice sites. To solve this problem we propose to measure the lattice site distribution of ^{7}Be implanted into different samples commonly used for half-life measurements: Al, Al_2O_3 and graphite. The samples will contain after these measurements still enough ^{7}Be activity to measure the half-life precisely by following the decay over up to a year. Since the decay measurements will be

performed with exactly the same samples used for emission channeling, the uncertainties for a theoretical modeling are thus minimized.

If successfully commissioned with the proposed first experiments, this unique method can in future be applied to study the lattice location of Be in other hosts.

Alternative methods

For heavier impurity atoms also the channeling of Rutherford backscattered ions from an external beam could be used to study the lattice location, but in case of dopants with lower Z than the matrix this method is not usable.

Beryllium has six particle-stable isotopes:

- ^{7}Be is purely decaying by electron capture, and is thus not directly useful for EC.
- ^{10}Be emits betas with Q_{β}-=556 keV that could well be used for electron EC. However, with a half-life of $1.5 \cdot 10^{6}$ years the specific activity is very low. If ^{10}Be was implanted at the same dose as that we propose for ^{7}Be, the beta activity would be only few Bq: too low to measure an EC pattern within reasonable time.
- ^{11}Be has 13.8 s half-life, i.e. would need to be used on-line. However, the electron energies are far too high (Q_{β}-=11.5 MeV) to show appreciable channeling effects. ^{11}Be has also a 2.9(4)% branch of beta-delayed alpha emission with energies of 770 keV (dominantly) and 466 keV [18]. Such an experiment might be feasible at ISOLDE, but the available ^{11}Be yield is one to two orders of magnitude lower compared to the ^{8}Li yield, the latter decaying via ^{8}Be breakup with 200% branching ratio to alphas. Thus, compared to the previous ^{8}Li EC experiments [2] about thousand times longer beam times would be required. This is clearly not realistic.
- ^{12}Be and ^{14}Be have very high Q_{β} values and very low yields. Thus they are not useful for EC experiments.
- In principle nuclear reactions on stable ^{9}Be could be used to generate charged ejectiles for channeling measurements. Photo-dissociation, i.e. ^{9}Be(γ,n)^{8}Be followed by breakup to two alpha particles has cross-sections in the sub-millibarn range. Thus, at equal Be concentration, a gamma flux of at least 10^{15} cm^{-2} would be required to achieve similar count rates as with the proposed ^{7}Be(n,p) experiment. However, such a huge gamma flux would create an unacceptable background for the charged particle detectors. Alternative reactions like (p,d), (d,p), (p,α), (d,α) etc. do not show a pronounced resonance and have cross-sections of the order of 0.1 b only. Thus elastic scattering of the projectile beam and charged particle reactions on the host matrix atoms would create enormous background for the particle detectors and the intense primary beam would create considerable damage to the samples.

We can conclude that emission channeling with ^{7}Be(n,p) is the most promising way to study the lattice location of beryllium impurities.

Realization of the experiment:

We propose to implant the samples with a ^{7}Be beam at ISOLDE and then use them as soon as possible (since ^{7}Be is decaying with 53 days half-life) for (n,p) EC measurements at ILL. We aim for implantation of the samples up to a ^{7}Be concentration of 0.5% in the peak of the deposition. Similar dopant concentrations are used for EC measurements with beta or conversion-electron-emitters implanted at ISOLDE. Since these are often far heavier isotopes, the damage in the implantation region is higher than in our case.

We have already tested at ILL a setup including a vacuum chamber with external neutron beam collimation and internal beamstop for scattered neutrons, a remote controlled goniometer holding the sample, a neutron camera for sample positioning and the Medipix-2 silicon pixel detector for high resolution position-sensitive charged particle detection. An algorithm to suppress background

from fast neutrons, betas and gammas [19] was successfully tested. The detector has also been successfully tested by direct exposition to a thermal neutron beam of 10^7 n/cm^2/s [20].

The angular resolution with the given setup is limited by the size of the implanted beam spot and not by the position resolution of the detector. With the detector at 15 cm from the sample, a good compromise between count rate and angular resolution is found for beam spots of 1 to 2 mm diameter. Thus, for a sample implanted on 2 mm diameter 10^9 protons will be emitted into 4π per day beam time with a neutron beam intensity of $3 \cdot 10^9$ n/cm^2/s. This corresponds about to the minimum number of events in electron EC experiments at ISOLDE. Another ILL beam line even provides a capture flux of up to $2 \cdot 10^{10}$ n/cm^2/s. Thus even higher statistics could be obtained with the given ^{7}Be concentration.

There are only few isotopes that may emit charged particles with significant cross-sections when exposed to a thermal neutron beam. Among the host atoms of the proposed matrices only the ^{14}N(n,p) reaction with 1.93 barn cross-section could be of concern for the nitride samples, but the resulting 0.58 MeV protons can be easily discriminated from the 1.44 MeV protons stemming from ^{7}Be(n,p).

Beam request:

^{7}Be beam intensities up to 300 nA have been measured before at ISOLDE [6]. However, to prevent a significant growth in beam emittance that would reduce the fraction of the beam that can be focused through a 1 to 2 mm diameter diaphragm, we plan to operate the ion source at a very conservative current of only 10 nA. Thus one sample with 10^{14} atoms (in 2 mm diameter) could be collected within less than an hour. Additional implantations with ^{9}Be under exactly the same conditions are required for proper sample characterization.

We request in total 6 shifts to implant about two dozen samples (for different annealing cycles) with ^{7}Be and ^{9}Be respectively, plus 2 shifts to tune the beam and, if needed, optimize the Be/Li ratio by microgating with the beam chopper on the RILIS pulses. When produced off-line from PSI graphite, the ^{7}Li contained in the graphite needs to be outgased before the run to achieve an acceptable ^{7}Be/^{7}Li ratio > 1.

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