In-situ (C)DBS at high temperatures at the NEPOMUC positron beam line

Markus Reiner, Philip Pikart and Christoph Hugenschmidt

Technische Universität München, Physik-Department E21 and FRM-II, Lichtenbergstraße 1, D-85748, Germany

E-mail: markus.reiner@frm2.tum.de

Abstract. We report on the current status of the sample heating stage in the CDB-spectrometer at the NEPOMUC positron beam line. The currently installed new sample heating is described in detail and various design aspects are discussed briefly. As an exemplary application, the positron diffusion at high temperatures in Ge was investigated by a depth dependent evaluation of both the Doppler broadening of the annihilation line and free Ps annihilation at the surface. It was confirmed that the temperature dependence of the positron diffusion is extraordinarily strong above 670 K.

1. Introduction

CDBS is outstanding among the various PAS methods due to its elemental selectivity on the vicinity of the positron annihilation site [1]. Performing (C)DBS with a monoenergetic positron beam at high temperatures is an unique experimental technique for the investigation of defect annealing and temperature driven structural changes in thin film systems [2]. However, it requires not only a highly intense positron beam in order to enable sufficiently short measurement times, but also an appropriate sample heating device, which enables the controlled heating of specimens in an UHV condition and biased to high voltages up to -30 kV. For this reason, during the recent years, several heatable sample holders have been applied at the CDB spectrometer [3] of the NEPOMUC positron beam line [4,5]. Within this contribution, we present the currently installed sample heating device as a result of various design studies and recent measurements on the temperature dependence of the positron diffusion in Ge(111).

2. The new sample heating device at the NEPOMUC CDB-spectrometer

The previous sample heaters at the NEPOMUC CDB-spectrometer were integrated in the standard sample holder for room temperature measurements. For this reason, design limitations constrained the maximum reachable temperature to 800 K. In order to increase the temperature further, an extra sample holder for high temperature measurements was constructed and is currently installed at the CDB-spectrometer.

In this device, the sample is heated by the light of a halogen lamp, which is focused onto the bottom of the sample by an elliptic reflector. Compared to the previous designs, the reflecting area has been more than trebled. Thus, the maximum sample temperature, which can be reached by use of the 250 W halogen lamp, will be clearly increased. Besides, in the new set-up, the sample will be movable and hence, spatially resolved measurements will also become possible at high temperatures.

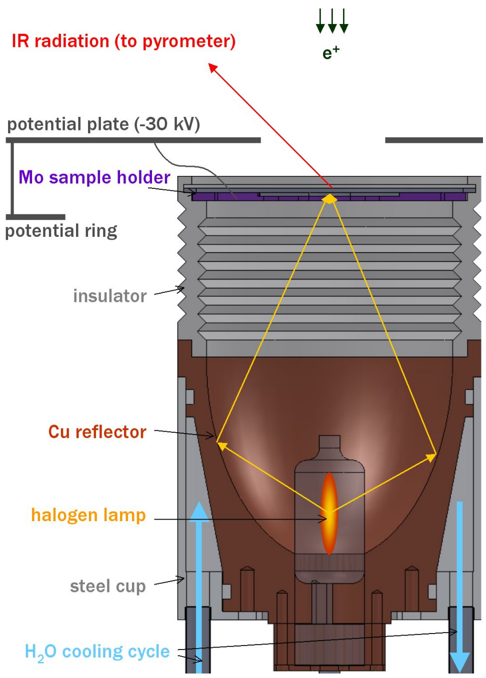

Figure 1. Mirror lamp assembly and heatable sample holder

The detailed sketch in figure 1 shows all relevant parts of the new sample heating device. The sample holder is located above the mirror lamp assembly, which consists of the Au-coated elliptic Cu reflector and the halogen lamp in the lower focal point. In order to run the halogen lamp at full power, a water cycle is used for cooling the mirror lamp assembly. The water will be pumped through the gasketed free volume between reflector and the steel cup, which carries the reflector. In its other focal point, the specimen is placed and hold by an assembly of Mo sheets, which enables fast sample changing. The Mo sheets are fixed on a ceramic insulation, which was designed in order to deal with the electric potential difference of up to 30 kV between reflector and specimen. The potential plate and ring, which enclose the sample holder, lead to a well defined electric potential in the vicinity of the specimen.

It was found, that the two color pyrometer METIS MQ22 from Sensortherm^{TM}, which can be placed outside the sample chamber, is well suited for a contact-free temperature measurement. For this, an IR transparent window is flanged at the sample chamber. Using this method, it is necessary to attach a small temperature control point consisting of colloidal graphite to the sample before heating. In this way, the sample has a well defined emissivity. The results of the pyrometer measurements are in excellent agreement with those obtained by electrical temperature sensors.

In addition, the presented design enables scanning an area of 2 cm x 2 cm on the sample with the NEPOMUC positron beam. The maximum spatial resolution reached by electrostatically focusing the beam onto the specimen will be 0.3 mm. Typical measurement times with an collinear assembly of four pairs of HPGe detectors will be less than 1 min for DBS and 2 h for CDBS. Within these short measurement times, a large variety of samples (with a typical size between 0.5 cm x 0.5 cm and 2 cm x 2 cm) can be investigated at temperatures up to around 1300 K.

3. High temperature behavior of positron diffusion in Ge

3.1. Introduction

As an exemplary measurement, the positron diffusion in single crystalline Ge at temperatures between 673 K and 853 K was investigated. Several studies based on different experimental techniques have been published on this issue. Jorch et al. [6] observed an extraordinarily strong temperature dependence of the positron diffusion length L_+ between 600 and 850 K by the investigation of free Ps annihilation at the surface, as well as Uedono et al. [7] by depth dependent DBS. Soininen et al. [8] found a weaker dependence by depth dependent DBS and investigation of Ps annihilation. These results illustrate that the high temperature behavior of positron diffusion in Ge, which is related to the electron and hole mobility in the semiconductor, is a point of long-term interest and give motivation for the presented study.

3.2. Experimental

A single crystalline Ge(111) sample (1.5 cm x 1.5 cm) was investigated and heated up to a maximum temperature of 853 K. Before the measurements, no surface treatment like polishing or etching has been performed in order to avoid chemical contamination. Instead, the specimen was heated up to the maximum temperature in a first heating cycle in order to remove oxide from the surface. After cooling down to room temperature, the specimen was heated for a second time and the positron diffusion was investigated.

For this, depth dependent DBS (S(E)) and investigation of the free Ps annihilation at the surface (F(E)) were performed with an incident positron energy between 1 keV and 16 keV. The S-parameter was determined by evaluating a central peak region of 511±0.825 keV and a total peak region of 511±11 keV of the broadened annihilation line. The F-parameter was determined by the evaluation of the linearized valley-to-peak ratio (valley region 450 - 500 keV, peak region 500 - 522 keV), which is sensitive to the 3γ-decay of o-Ps with its continuous energy spectrum. The 3γ-decay event can only take place for free Ps, which does not interact with electrons from the bulk. In case of the singlecrystalline Ge, the annihilation of free Ps can be expected to take place only at the surface.

Both the S(E)- and the F(E)-curve were used for a quantitative determination of L_+ by fitting both curves simultaneously. Since the total range of incident energies was accounted for these fits, also epithermal positrons were considered. For performing the fits, the VEPFIT software package [9] was applied. A more detailed description of the method of data evaluation can be found in [2].

3.3. Results and Discussion

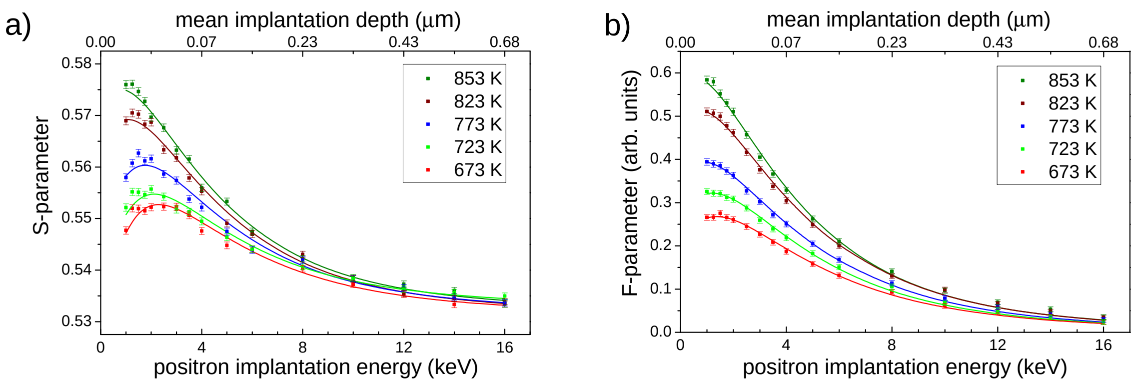

Figure 2. S(E) and F(E) on Ge(111) at high temperatures; solid lines are fit results.

The recorded S(E)- and F(E)-curves (measurement time 12 min per depth profile) at high temperatures are shown in figure 2. At the various temperatures, for high incident positron energies, the S-parameter saturates at a bulk value of 0.532±0.002. For the F-parameter, at all temperatures this bulk value is 0 and hence, no significant 3γ-decay is found for high implantation energies, i. e. in the bulk.

For lower implantation energies, i. e. in the surface region, the F-parameter increases, since positrons, which diffuse to the surface, can desorb as Ps leading to the observed 3γ-annihilation of o-Ps. The fraction of positrons that annihilates as o-Ps increases with temperature and hence, positrons may be thermally desorbed as Ps from surface states. Also the S-parameter at the surface, which takes higher values than in the bulk, increases with the temperature. This observation is directly connected to the increasing fraction of positrons annihilating as o-Ps. The formation of Ps at the surface also leads to the annihilation of p-Ps. Due to its low binding energy, at the annihilation of p-Ps only a small Doppler shift is detected resulting in a high S-parameter. For implantation energies below 3 keV, the S(E)- and F(E)-curves exhibit further features, which can be attributed to the annihilation of epithermal positrons. Here, their behavior is not treated in detail.

The solid lines in figure 2 represent VEPFIT results. The according results for L_+ and its temperature dependence are shown in figure 3. The decrease from 146±5 nm to 106±5 nm between 673 K and 853 K is too large in order to be explained by a $T^{-\epsilon}$ with $\epsilon = 0.25$, which corresponds to acoustical phonon scattering during positron diffusion [10]. This behavior was observed in [8], where, only implantation energies above 6 keV were evaluated in order to exclude any influence of epithermal positrons. This method of data evaluation was also tested on the current data, but led to large uncertainties in the fit results.

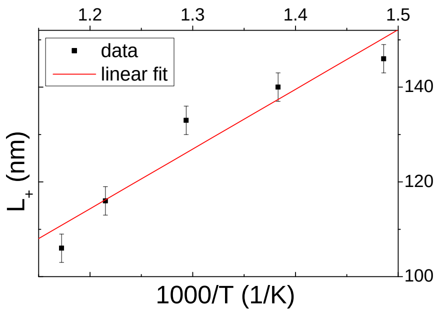

Figure 3. Temperature dependence of L_+ in Ge(111)

A stronger temperature dependence of ϵ=1 can explain the results much better (see fitted line in figure 3). This is also in reasonable agreement with the results published in [6], where positron implantation energies below 5 keV were investigated and ϵ=0.8 was found. In particular, the present study, based on the evaluation of S(E)- and F(E)-scans, confirms the results given in [7], where also epithermal positrons were considered and ϵ=1 was found in the evaluation of S(E)-scans. Possible explanations for the observed strong temperature dependence of L_+ in Ge are given in [6]; there it was tentatively concluded that the positron diffusion is affected by some kind of positron-lattice coupling.

4. Conclusion and outlook

Within this contribution, the development and design of the new sample heater in the CDB-spectrometer at the NEPOMUC positron beam line is presented. Due to the use of the highly intense positron beam, the device will enable novel experiments at temperatures up to 1300 K. Furthermore, the high temperature behavior of the positron diffusion in Ge(111) was investigated exemplary. The depth dependent evaluation of both the Doppler broadening of the annihilation radiation and the free Ps annihilation at the surface revealed an extraordinarily strong temperature dependence of the positron diffusion above temperatures of 670 K.

References

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