Realization of simultaneous channeling and nuclear-hyperfine-interaction experiments

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With the use of conversion electrons emitted during the decay of ^{111}In atoms diffused into a copper single crystal, it is shown that the electrons are steered by lattice rows and that the impurities are localized at substitutional sites. The damage produced by implantation of the radioactive atoms was investigated in a simultaneously performed channeling and perturbed-$\gamma\gamma$ angular-correlation experiment using ^{111}In as probe atom. The formation of defect clusters at the probes' site can be observed by both techniques.

By channeling and nuclear-hyperfine-interaction experiments information about the lattice site of impurity atoms (probes) is obtained on a microscopic scale. The channeling effect is sensitive to the *geometrical position* of probe atoms, but different sites occupied at the same time are difficult to resolve; this problem predominantly exists in the case of probe atom defect interactions in a damaged lattice. On the other hand, the measurement of the hyperfine interaction at the probe atoms one allows to determine the *number of* different lattice *sites* and their *relative population,* but due to the lack of an appropriate theory the geometrical information is not directly accessible. So it is obvious that both techniques are complementary, especially if they are simultaneously applied at the same sample. We use the radioactive atom ^{111}In, which is suited to determine the hyperfine interactions by the perturbed-$\gamma\gamma$-angular-correlation technique (PAC) and also to perform a channeling experiment with its conversion electrons emitted in competition to the γ rays. A few channeling experiments for lattice location of radioactive atoms were performed with α or β particles,[1,2] but the employment of conversion electrons, appearing in nearly all radioactive decays, should open a more general way for such combined experiments.

The probe atom ^{111}In decays by electron capture to the excited daughter nucleus ^{111}Cd which reaches its ground state via a $\gamma\gamma$ cascade composed of a 171- and a 245-keV transition. The observation of this $\gamma\gamma$ cascade is the salient point of the PAC experiment.[3] However, with a probability of 8.7 and 5.4% the two transitions take place by the emission of conversion electrons of the *K* shell with an energy of 145 keV (*K* 171) and 219 keV (*K* 245), respectively, which are easily resolved by a silicon surface barrier detector. In Fig. 1 we show the results of a channeling experiment where the intensities of the *K* 171 and *K* 245 electrons are recorded around low index crystal directions: A single crystal of copper cut parallel to a {100} plane and electropolished, was diffused in a closed, H_2-filled quartz ampoule (60 min at 680 K) with 500 μCi ^{111}In, corresponding to an average concentration of 1.5 ppm, so that the In atoms should be on substitutional sites in the copper lattice. For both electron energies maxima of intensities of the emitted particles are clearly visible in [100], [110], and [111] lattice directions as determined by H^+ backscattering. The angular scans correspond to a random-axial-random scan; the tilting plane and a low-index plane enclose a small angle of about 10 or 14° in each case. The flux peaking observed for the three lattice directions reflects the substitutional position of the emitter and is in agreement with the results obtained by Uggerhøj *et al.*,[2] who showed that the channeling behavior of electrons and positrons, produced by the β^- and β^+ decay of ^{64}Cu implanted in a copper crystal, is just opposite: In case of a substitutional emitter a flux peak for electrons is observed due to the steering by the atomic strings in contrast to a blocking dip for positrons. The width of the flux

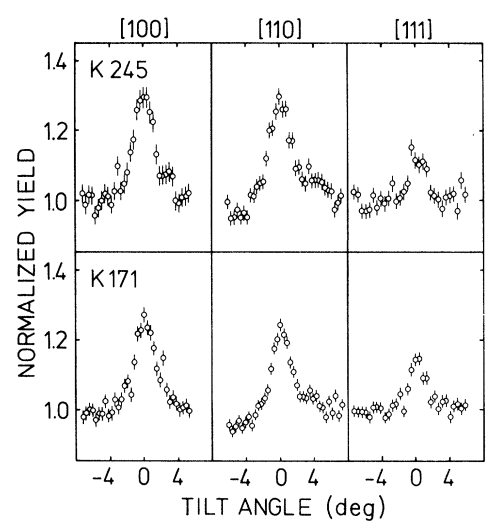

FIG. 1. Normalized emission yields around low-index directions of 145- and 219-keV *K*-conversion electrons from the decay of ^{111}In diffused into a Cu crystal.

peaks of the conversion electrons is in agreement with the critical angle ψ_1 from the Lindhard theory[4] taking into account the limited angular resolution of about 1° in our measurement. The clear results of this experiment establish conversion electrons to be suited for channeling experiments.

To investigate the influence of radiation damage on the observed channeling effect, we doped copper crystals with radioactive atoms by implantation producing lattice defects at the same time. Provided a sensitivity to lattice defects exists in the case of conversion electrons emitted from probe atoms inside the crystals, the PAC can be used as a second completely independent method to gain information about the damage situation of the same system, for this method can detect lattice defects via their induced electric field gradient (efg) at the probes's site. Therefore, we have combined both techniques to study the behavior of the lattice defects during an isochronal annealing sequence. Copper crystals were implanted with ^{111}In at 300 K to a dose of about $2\times10^{14}/\mathrm{cm}^2$ corresponding to an average local impurity concentration of about 100 ppm. The ^{111}In ions were implanted at three different energies resulting in different mean depths of the emitters. As a result of dechanneling the heights χ_M of the flux peaks are different and χ_M values of 2.2, 1.5, and 1.3 were measured for K 245 electrons in a [100] direction after implantation with 30 keV (mean depth =70 Å), 120 keV (200 Å), and 350 keV (520 Å), respectively. From these values a dechanneling length of about 500 Å is deduced in agreement with an estimate given by Andersen *et al.*[5]

Results of the PAC and channeling (K 245 around [110]) experiments performed during isochronal annealing of the copper crystal are collected in Figs. 2 and 3. The results obtained directly after the implanation with 120 keV and after annealing at 500 and 590 K are shown in Fig. 2 for PAC (left side) and channeling (right side). For the channeling experiment, a change of the three flux peaks is clearly visible. Because the only difference consists in the annealing treatment, which influences the structure and/or concentration of the produced lattice defects, the sensitivity to defects is established. The complete results of the annealing program are collected in Fig. 3, where the maximum yield of the channeling effect is displayed for a 120- and a 350-keV implantation. Concerning the PAC experiment the spectra in Fig. 2 belonging to $T_A=500$ and 590 K exhibit a strong modulation in contrast to the first spectrum

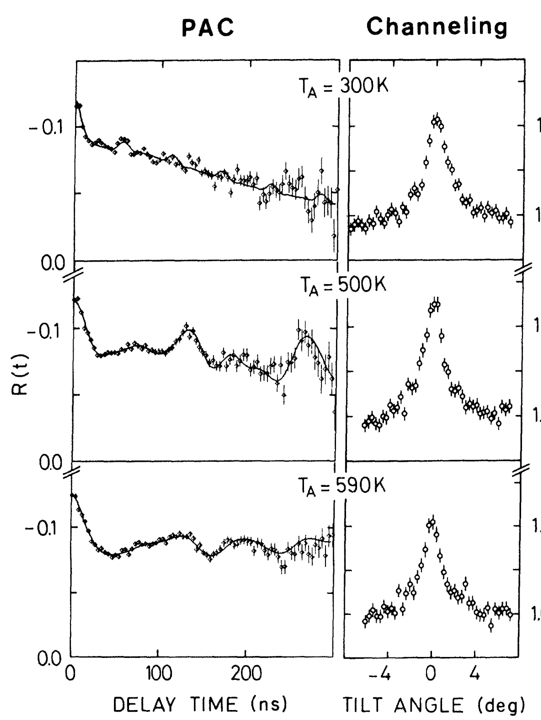

FIG. 2. PAC time spectra $R(t)$ and flux enhancement of K 245 conversion electrons around a [110] axis simultaneously measured for different annealing temperatures T_A at a Cu crystal implanted with 120-keV ^{111}In ions.

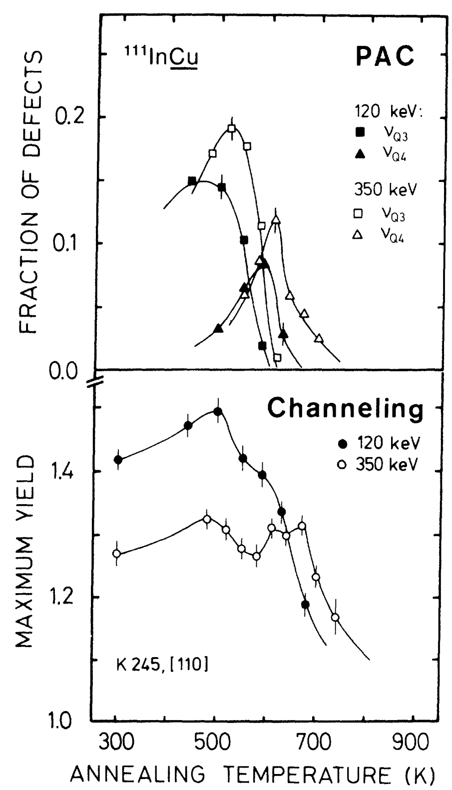

FIG. 3. Fractions of In atoms in defect configurations ν_{Q3} (squares) and ν_{Q4} (triangles) compared with [110] channeling peak heights for K 245 conversion electrons as a function of annealing temperature for two implantation energies.

($T_A = 300$K). The time spectra $R(t)$ are obtained by a combination of the different coincidence spectra recorded by the four γ detectors.[6] The $R(t)$ spectra show the existence of three different ^{111}In sites: The time-dependent modulation reflects two defect associated In sites distinguishable by two different electric quadrupole interaction frequencies $\nu_Q = eQV_{zz}/h$, which are caused by the defect specific field gradient V_{zz} (eQ is the quadrupole moment of the excited ^{111}Cd state enclosed by the observed γ cascade). The third In site is readable from the time-independent offset in the $R(t)$ spectrum and corresponds to probe atoms without a defect in their vicinity. The two defects formed at the In site are known from earlier defect investigations in copper (they are labeled $\nu_{Q3} = 50$ MHz and $\nu_{Q4} = 54$ MHz, respectively) and are identified as defect clusters.[6] The two fractions of ^{111}In atoms decorated with these clusters are given in Fig. 3.

The isochronal annealing program was started at 300 K, just above the recovery stage III in Cu, where preferentially small defect clusters are present in the lattice, randomly distributed. During annealing a dissolution of the smaller and a growth of larger clusters occurs leading to a decrease of the randomly distributed damage. This decrease is accompanied by an increase of the channeling effect. However, since the growth of the clusters partially happens at the site of the ^{111}In emitters, as indicated by the increase of the cluster configurations, the channeling effect should be influenced by a *second process* which tends to decrease χ_M due to a perturbation of the lattice in the next neighborhood of the emitter. Caused by this second process the increasing χ_M tips over around 500 K so that at higher temperatures χ_M decreases, though the average defect concentration in the lattice is reduced. Around 600 K and PAC shows that the In cluster configurations dissolve and at the same time χ_M increases again in case of the 350-keV implantation. The collapse of the flux peak at still higher temperatures (680 K) can be explained by a strong increase of the dechanneling probability, because it is accompanied by an observed loss of radioactive emitters via the surface so that it has to be assumed that the emitters also diffuse to a greater depth. In the 120-keV-implantation case, the loss of activity and the disappearance of χ_M above 600 K is observed at lower annealing temperatures, because (a) the local defect concentration is higher, so that the probe atoms are more mobile due to irradiation enhanced diffusion and (b) the dechanneling probability shows a stronger depth dependence. As a consequence the second increase of χ_M remains hidden. The annealing experiment demonstrates that two processes have to be taken into account in order to explain the behavior of the channeling effect: (i) The reduction of lattice damage randomly distributed with respect to the radioactive emitters and (ii) the formation of defects at the probes' site which is controlled by the PAC experiment.

As discussed by Andersen *et al.*[7] the theoretical description of the lattice steering for light particles, especially low energetic electrons, is more complicated than for positive ions, because the application of the classical continuum model seems to be not justified. However, our results obtained for a diffused Cu crystal show a clear steering effect for conversion electrons having energies around 200 keV. The flux peaks observed in different lattice directions agree with the channeling behavior as derived from a classical point of view for substitutional emitters. Owing to our limited angular resolution it might be that the observed flux peaks represent only an envelope of a more complex structure caused by diffraction effects. Even if a more quantitative description of the steering effect of conversion electrons should demand calculations based upon diffraction theory, like in the case of electron microscopy, the use of conversion electrons remains still attractive: The large number of nuclear hyperfine experiments, which always relate their information to the lattice site of the used radioactive atom, can be combined with the site information of channeling experiments. On the other hand, the analysis of the position of impurities by the channeling effect can be improved by nuclear hyperfine experiments determining the number and population of different sites occupied by the impurities. This point is of particular interest in the case of defect studies by channeling via the determination of the displacement of an impurity caused by a trapped defect.[8] As compared to the backscattering channeling technique the use of radioactive emitters has the advantage that problems caused by lattice damage due to the analyzing ions and by kinematical restrictions are absent and low impurity concentrations of 100 ppm or less are sufficient. It is obvious that the investigation of a trapped point defect—interstitial or vacancy—at ^{111}In atoms in different matrices[9] is a fruitful application of such combined experiments: With help of the PAC the formation of a distinct defect-impurity configuration can be facilitated so that their geometrical structure can be determined by the channeling effect in a unique way.

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[1]B. Domeij and K. Bjorkqvist, Phys. Lett. 14, 127 (1965).
[2]E. Uggerhøj and J. U. Andersen, Can. J. Phys. 46, 543 (1968).
[3]H. H. Rinneberg, At. Energy Rev. 17, 2 (1979).
[4]J. Lindhard, *Atomic Collision Phenomena in Solids* (North-Holland, Amsterdam, 1970), p. 1.
[5]S. K. Andersen, F. Bell, F. Frandsen, and E. Uggerhøj, Phys. Rev. B 8, 4913 (1973).
[6]O. Echt, E. Recknagel, A. Weidinger, and Th. Wichert, Z. Phys. B 32, 59 (1978).
[7]J. U. Andersen, S. K. Andersen, and W. M. Augustyniak, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 39, No. 10 (1977).
[8]M. L. Swanson, L. M. Howe, and A. E. Quenneville, J. Nucl. Mater. 69&70, 372 (1978).
[9]E. Recknagel and Th. Wichert, Nucl. Instrum. Methods 182/183, 439 (1981)