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FORMATION OF SMALL VACANCY CLUSTERS IN TUNGSTEN AROUND SILVER AND
INDIUM IMPURITIES STUDIED BY PAC AND THDS

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Vacancy clustering at Ag and In impurities in W was investigated combining
Thermal Helium Desorption Spectrometry (THDS) and Perturbed Angular Correlation
(PAC) measurements. The results of these experiments are compared with each
other and with those obtained from Monte Carlo calculations.

1. Results of PAC measurements

Almost perfectly (100) oriented polycrystalline foils of W were
implanted at room temperature with 25 keV and 110 keV [111]Ag and [111]In
ions, at a dose of $2 \times 10^{13}/cm^2$ or less. The samples were annealed for
15 min in vacuum at step-wise increasing temperatures, and studied by
PAC after each step. From these measurements information was obtained
about the symmetry of each lattice defect trapped at the impurity atom,
and the number of defects as a function of annealing temperature. For
details of the method, see ref. [1] and references quoted therein.
Apart from substitutional impurities, i.e. atoms with no defects
within 1 lattice unit (LU), 3 different impurity-defect configurations
were distinguished. The corresponding fractions are shown in fig. 1.
For [111]In$\underline{W}$ the results agree with those obtained by Pütz et al. [2].
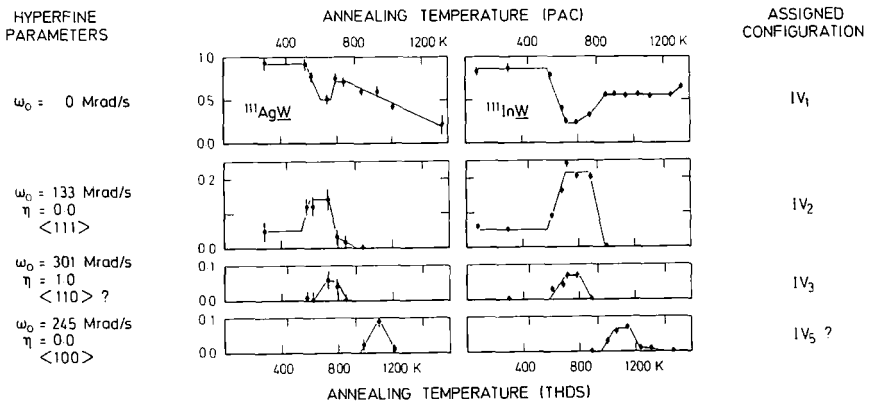

Fig. 1. Annealing behaviour of defects in W, observed in PAC measurements on samples
implanted with [111]Ag and ^{111}In, respectively. Corresponding hyperfine parameters are
indicated on the left-hand side. Assigned impurity-vacancy clusters are given on the
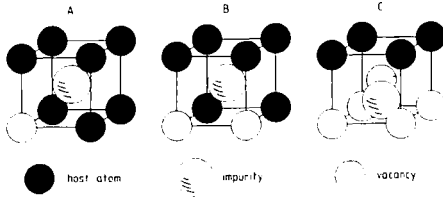right hand side. Annealing time was 15 min.

Fig. 2. Proposed impurity-vacancy configurations: (A) IV_2, (B) IV_3 and (C) IV_5.

To facilitate the comparison between PAC and THDS results, the tempe-
rature scale of fig. 1 is renormalized according to the annealing con-
ditions in the THDS experiments. The latter (see sec. 2) clearly show
vacancy mobility at about 800 K (stage III). Therefore, the formation
and break-up of defects observed by PAC in this temperature region
have to be associated with trapping and detrapping of vacancies. Ac-
cording to THDS vacancy-type traps in W persist up to 1400 K after
implantation with 20-25 keV Ag or In ions, trap concentrations being
twice as large as in self-implanted samples. Therefore, the defect
observed by PAC at about 1100 K is likely to be a relatively small
vacancy cluster, too.
    In fig. 2 we give some vacancy configurations that are consistent
with the observed quadrupole frequencies (ω_0), asymmetry parameters
(η), and orientations (<...>) of the defect symmetry axis. In the
case of IV_5 relaxation of the impurity atom over 0.3 LU along the
<001> direction has to be assumed.

2. Results of THDS measurements

    A (100) single crystal of W was implanted with 5 keV and 25 keV
Ag and In ions, at doses ranging from $2 \times 10^{12}/cm^2$ to $2 \times 10^{13}/cm^2$. The
sample was annealed by increasing the temperature to a value T_a at
a rate of 40 K/s, and subsequently injected with 250 eV He ions. De-
fects bind He atoms with characteristic energies which were determined
by monitoring the He release during the linear heating. From the num-
ber of He atoms desorbing in a certain peak information was obtained
about the trap concentration. A survey of the THDS method is given in
ref. [3]. The equipment used for the experiments in this study is
described in ref. [4].
    Some typical helium desorption spectra for AgW are shown in fig.
3. Different peaks are clearly visible. The peaks A-C correspond to
He atoms releasing from substitutional Ag atoms (interstitial-type
traps). He atoms are bound to vacancy-type traps with much higher
binding energies (>2.5 eV in W). Release of these He atoms gives rise
to peaks G-H. In fig. 4 the relative concentrations of the two classes
of traps are shown as a function of the annealing temperature.
    For the In implanted samples no interstitial-type traps were ob-
served, which implies that substitutional In atoms do not bind He atoms
at 300 K. We notice that for the samples implanted with 5 keV (20 keV)
Ag ions an appreciable number of interstitial-type traps is present
only after annealing at 850 K (1350 K). This seems to contradict the
PAC results which indicate a nearly 100% substitutional fraction of
Ag and In atoms immediately after implantation.

3. Model calculations

    A simulation of the binary collision cascade following implan-
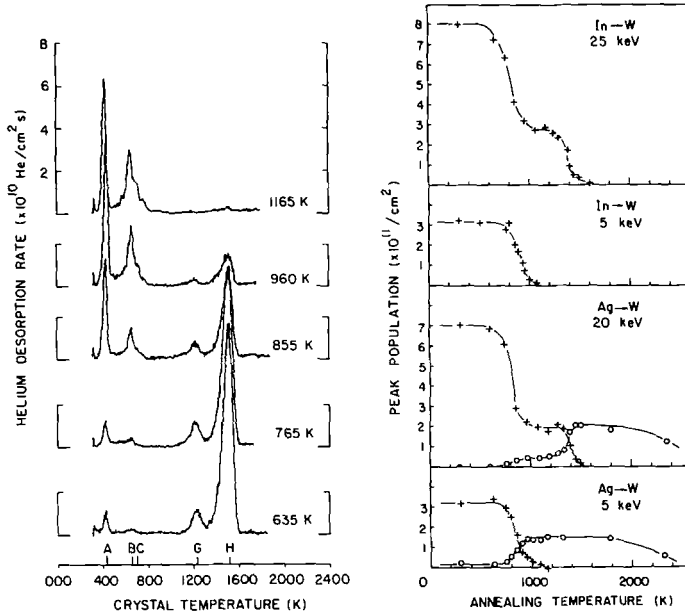tation was performed using the programme MARLOWE [5]. Radial vacancy

Fig. 3. Helium desorption spectra of (100) W implanted with 2×10^{12}/cm^2 of 5 keV Ag ions, annealed at the indicated temperatures, and subsequently injected with 3×10^{12}/cm^2 of 250 eV He ions. Heating rate was 40 K/s.

Fig. 4. Relative concentrations of vacancy-type (O) and interstitial type (+) traps in (100) W implanted with 2×10^{12}/cm^2 Ag or In ions.

distributions around the implanted atom were calculated for 25 keV ions impinging on (100) W at 10° and 0° with respect to the [001] d rection. The number of implantation events was limited to 20 in eac case. The fraction of the formed Frenkel pairs (FP) that will spon- taneously recombine depend strongly on the recombination distance. In the case of 1-3 keV noble gas implantation of Mo, Hou et al. [6] found that for an FP recombination distance of 3.7 LU and a displac ment energy of 33 eV calculated vacancy concentrations were in agr ment with experimentally observed values. We used parameter values 40 eV and 3.7 LU, respectively, in our calculations for W, the results of which are shown in fig. 5.
    A Monte Carlo approach was applied to estimate the fraction of near vacancies migrating towards the implant during early stage III Only vacancies within a sphere with radius of 4 LU were considered. A jump probability of 1/8 was assumed for each <111> direction. For each non-equivalent lattice point the fate of 2000 vacancies was fc lowed. The vacancy was considered to be trapped once it had jumped a first or second neighbour position with respect the the impurity. The vacancy was considered to be lost for short range capture once it had migrated more than 4 LU from the trap. We found that 56% of the vacancies starting from less than 2 LU were trapped after about jumps, and that 8% of the vacancies starting from 2 to 4 LU were trapped after about 13 jumps. The calculated fractions of In atoms
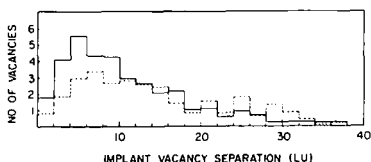
Fig. 5. Radial vacancy distributions
for 25 keV In ions implanted into (100)
W. Dashed line: implantation in channel
direction. Full line: implantation 10°
off the [001] channel.

associated with 0, 1 and 2 extra vacancies in early stage III are 40%,
30% and 15%, compairing fairly well with the experimental PAC values
30%, 20% and 7% (In) and 50%, 15% and 6% (Ag), respectively.

4. Discussion

The most striking difference in the results of the two applied
methods is that PAC shows a large substitutional fraction in
as-implanted samples, where virtually no substitutional atoms are
seen by THDS. Apparently no He atoms are trapped at substitutional
Ag atoms because these are effectively shielded by nearby vacancies.
In the case of substitutional In atoms, the binding energy is too
small to cause trapping at room temperature.

Using the observed annealing temperature T_a = 550(10) K at which
vacancy trapping is observed by PAC, the time Δt = 15 min during
which the sample was annealed, the calculated number of vacancy
jumps n = 5–13 required for short-range capture, and the jump fre-
quency ν_0 = 4.4×10^{13}/s derived from the data given in ref. [7], we
calculate the monovacancy migration enthalpy H_{1V}^m = $kT_a \ln (\nu_0 \Delta t/n)$
= 1.71(5) eV.

The defect in W observed by PAC at higher temperatures may be
the same as the one suggested by Weidinger et al. [8] for In-implan-
ted Mo. In the latter case the impurity atom was assumed to be sur-
rounded by a nearly tetrahedral cluster of 4 vacancies. However,
symmetry and magnitude of the electric field gradient tensor are
also consistent with the configuration InV_5.

Acknowledgements

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