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Helium damage in long-aged metal–tritium systems

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Abstract

The evolution of lattice damage in metal–tritium–helium systems is investigated by means of neutron scattering techniques. Polycrystalline samples of tantalum, yttrium and scandium have been loaded with tritium concentrations on the order of ten atomic percent and changes in the positions, intensities and line shapes of Debye–Scherrer lines have been determined over a period of about 15 years. The results are interpreted in terms of a steadily growing number of lattice defects due to helium production. Helium clustering and bubble formation induce self-interstitials and dislocation loops developing into extended dislocation networks at higher helium concentrations. No tritium loss from the samples could be observed even after aging times exceeding one half-life of tritium and giving rise to helium concentrations of several atomic percent. Differences in the results obtained for cubic (tantalum) and hexagonal (yttrium, scandium) host lattices will be briefly discussed. The present work complements results obtained earlier from investigating the same samples after aging times of about one and five years, respectively, and which were presented in Refs. [1,2].

Keywords: Dislocations; Point defects; Neutron diffraction

1. Introduction

Although helium damage in metals is still of considerable technical interest for future fusion reactors (first-wall problem) only few investigations [1–5] applying scattering techniques were done since the late 1980s (for an overview of earlier work see for example [6]). Helium, implanted either using high kinetic energies or by radioactive decay of tritium, induces a complex defect structure in metals due to helium clustering and bubble formation with increasing helium concentration.

We present here a long-term investigation of the evolution of Debye–Scherrer lines due to helium damage in tantalum and in the rare-earth metals scandium and yttrium. Differences between cubic (tantalum) and hexagonal (scandium and yttrium) metal–helium systems will be described as well as differences between these two hexagonal systems.

2. Experimental setup

All measurements were performed on the triple-axis spectrometer VALSE (G43) located at a cold neutron guide position of the reactor Orphée at the Laboratoire Léon Brillouin in Saclay, France. The neutron wavelength used was 0.292 nm and in order to improve resolution in reciprocal space the analyzer was set in second order position. Collimations of 30 min before and after the sample and 60 min before the detector were applied. The setup chosen in this way gives an instrumental resolution which is narrower than the inherent widths of the Debye–Scherrer lines of the unloaded samples. Influences of texture effects which are apparent in the observed scattering intensity distribution of the polycrystalline samples were eliminated by continuous rotation of the specimens during the experiments.

3. Samples

The samples were polycrystalline platelets ($1\times10\times50$ mm^3) loaded with tritium from the gas phase. The initial

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tritium/metal ratios were 0.04 for tantalum, 0.142 for scandium and 0.089 and 0.068 for the two yttrium samples. This implies that at their respective initial tritium concentrations all samples were in the solid–solution phase at room temperature (for further details cf. [1,2]).

All samples except $YT_{0.068}$ were mounted into cylindrical vacuum sealed aluminium containers. The $YT_{0.068}$ sample was mounted into a liquid-nitrogen cryostat and, subsequently, stored and investigated at 77 K till a helium concentration of 2.25% (i.e. a helium/metal ratio of 0.0225) was reached. Subsequently it was mounted into an aluminium container like the other samples and studied at room temperature. For each tritiated sample an unloaded reference sample of identical shape and origin is available. Inspection of the containers and calorimetric determination of the tritium content in 1998 showed that no tritium had been released from the samples.

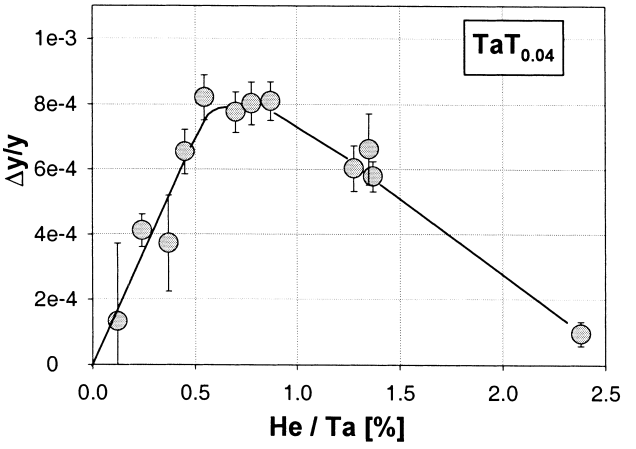

Fig. 1. Helium-induced relative lattice parameter change in $TaT_{0.04}$ as a function of helium concentration, i.e. (helium-to-metal ratio)×100. The interpolating line serves as a guide to the eye, only.

4. Measurements

Debye–Scherrer (DS) lines of all tritiated samples were measured and analyzed in terms of intensity, width and position as a function of helium concentration and were compared to the unloaded reference samples which were investigated each time as a part of the same experiment. To obtain the broadening of the DS lines the line profiles were deconvoluted with the linewidths of the unloaded samples. The measured changes of the lattice parameters were corrected for the lattice contraction due to tritium decay [1,7,8]. For Ta (b.c.c. structure) the (110), (200) and occasionally the (211) lines were studied. For Y and Sc (h.c.p. structure) the (00.2) reflection was scanned along the hexagonal axis, in the basal plane measurements were done regularly at the (11.0) and (20.0) lines. The results presented in the figures were obtained by proper averaging of available data. Error bars represent statistical errors and, where not given, usually do not exceed symbol size.

5. Lattice distortions and Debye–Scherrer lines

Helium is created inside the samples by radioactive tritium decay. The produced helium atoms cluster and at length form helium bubbles. The formation of bubbles is inherently connected with the production of self-interstitials remaining close to the helium bubbles till their growing number gives rise to the punching out of dislocation loops [9]. The continuous formation of such dislocation loops finally leads to the emergence of a connected dislocation network.

Schematically, the above defects influence the DS lines as follows [10]. Since self-interstials and dislocation loops are defects giving rise to distortions of finite size they are leading to a lattice parameter shift of the host lattice and to diffuse scattering but have no influence on the widths of the Bragg peaks or Debye–Scherrer lines of the host lattice. By way of contrast, infinite defects like dislocation networks induce a broadening of the peaks or lines.

6. Results

6.1. Tantalum (Figs. 1 and 2)

The helium-induced lattice expansion increases almost linearly at low He-concentration and saturates near 0.8%He [2]. Up to 2.4%He the lattice parameter decreases to a value similar to that observed at the beginning of the tritium decay.

The line width increases over the entire range of He content examined. First, at low He concentrations till 0.8%, it rises slowly followed by a steeper increase. From 1.3%He on practically no more broadening is found up to about 2.4%He.

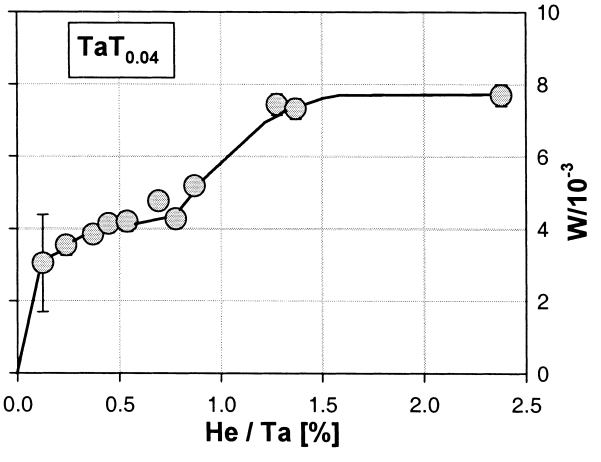

Fig. 2. Relative broadening of Debye–Scherrer lines (W) in $TaT_{0.04}$. The interpolating line serves as a guide to the eye.

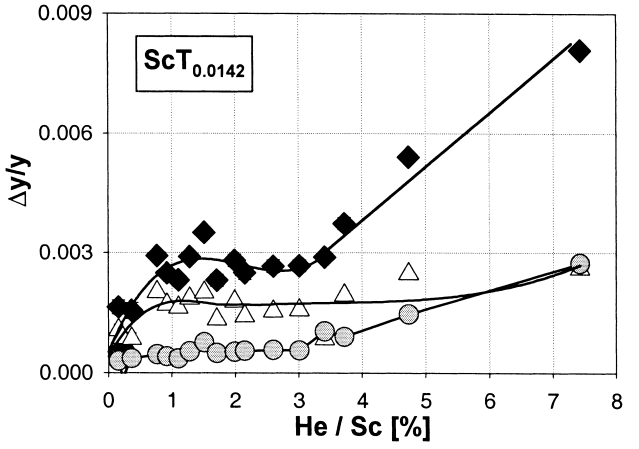

Fig. 3. Helium-induced volume dilatation (♦) and relative lattice parameter change in hexagonal (△) and basal (○) direction as a function of helium concentration in $ScT_{0.0142}$. The expression $\Delta y/y$ labeling the ordinate stands for $\Delta a/a$, $\Delta c/c$ and $\Delta v/v$, respectively. The interpolating lines serve as guides to the eye.

In the hexagonal rare earth systems the evolution of the linewidths as well as the lattice distortion shows different behavior for the reflections lying on hexagonal and basal crystallographic directions, respectively. Therefore, the volume change of the unit cell is displayed for these systems along with the changes observed for the lattice parameters *a* and *c*.

6.2. Scandium (Figs. 3 and 4)

In ScT the lattice volume first increases steeply up to a value of about 0.8%He. Afterwards the evolution of the volume is practically flat till 3.5%He to increase once more drastically steeper. It is interesting to see that the lattice parameter shifts in the hexagonal and basal directions show different behavior which is also borne out by the corresponding volume increase. The lattice parameter shift along basal directions shows a continuous increase till 3.5%He and gets steeper afterwards. The lattice parameter shift along the hexagonal direction can be described in three steps. At low helium concentrations up to 0.8%He the shift increases rather steeply. Then, in the range between 0.8 and 3.5%He, the lattice parameter ceases to grow. At helium concentrations higher than 3.5%He the increase starts again.

The line broadening of the hexagonal direction first rises steeply but appears to level off at higher helium concentrations. Further measurements will be necessary in order to clarify whether the observed slight decrease above 4.5%He is really significant. In the basal plane the observed rate of broadening of the DS lines also decreases continuously with increasing helium content, but generally at a lower level than in the hexagonal direction.

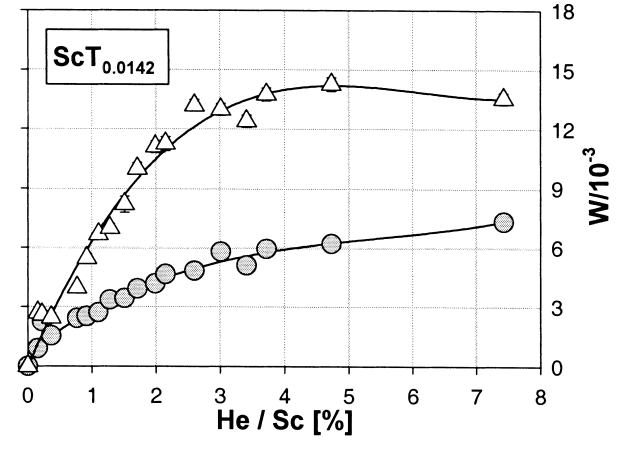

Fig. 4. Relative broadening of Debye–Scherrer lines (W) in $ScT_{0.0142}$. Reflections in the basal plane (○) and the hexagonal (00.2) reflection (△). The interpolating lines are guides to the eye.

6.3. Yttrium

$YT_{0.089}$ (Figs. 5 and 6). The helium-induced lattice expansion increases at lower helium concentrations and saturates somewhere about a helium content of 2.5%. For higher helium concentration the lattice parameters decrease slightly.

The evolution of line broadening exhibits a rather continuous increase, only the broadening along the hexagonal direction shows a steplike behavior around 0.8%He.

$YT_{0.068}$ at liquid nitrogen temperature (Figs. 7 and 8). The lattice expansion is rather steep at small helium concentrations, however, slows down around 0.8%He. After reaching a concentration of about 2.5%He the sample was brought to room temperature and kept there subsequently. Later measurements of the lattice parameters

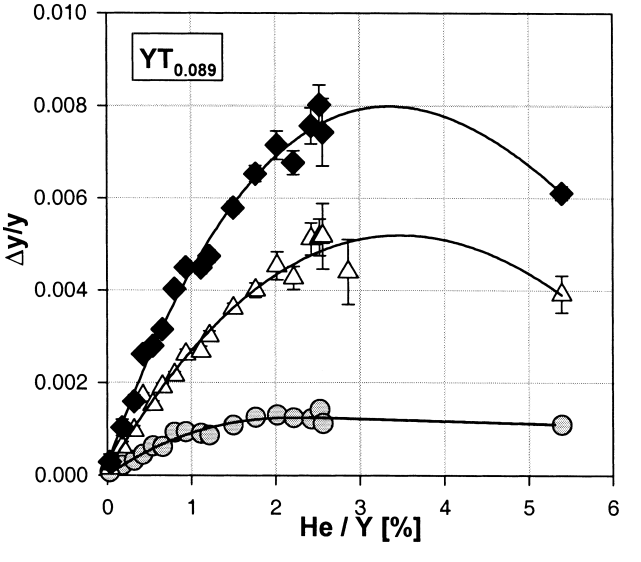

Fig. 5. Helium-induced volume dilatation (♦) and relative lattice parameter change in hexagonal (△) and basal (○) direction as a function of helium concentration in $YT_{0.089}$. The interpolating lines are guides to the eye.

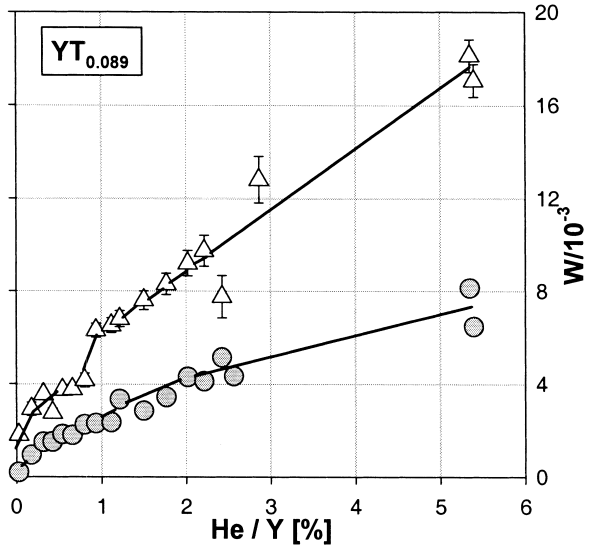

Fig. 6. Relative broadening of Debye–Scherrer lines (W) in $YT_{0.089}$. Reflections in the basal plane (○) and the hexagonal (00.2) reflection (△). The interpolating lines are guides to the eye.

showing a slight contraction of the lattice were corrected for the thermal expansion associated with heating from 77 K to room temperature.

The linewidth along the hexagonal direction obviously did not change or even slightly decrease during all the time the sample was held and measured at liquid-nitrogen temperature. After heating the sample to ambient temperature the linewidth increased in a similar way as found for $YT_{0.089}$. The linewidths in basal directions at lower helium concentrations also did not change significantly but started to increase already at about 0.8%He contrary to the behavior of the hexagonal line.

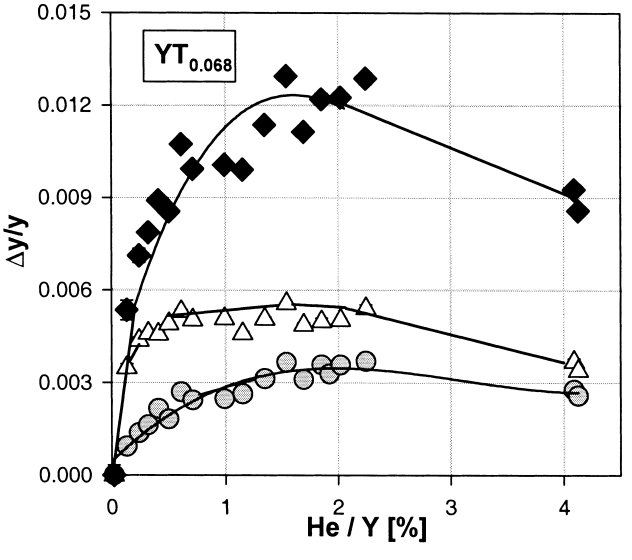

Fig. 7. Helium-induced volume dilatation (♦) and relative lattice parameter change in hexagonal (△) and basal (○) direction as a function of helium concentration in $YT_{0.068}$. The interpolating lines are guides to the eye.

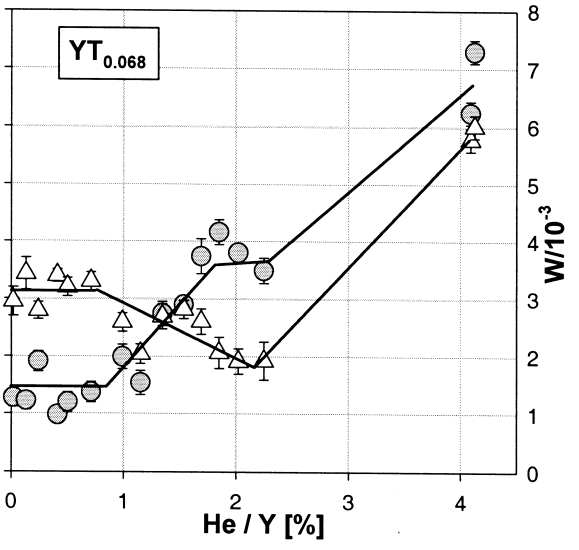

Fig. 8. Relative broadening of Debye–Scherrer lines (W) in $YT_{0.068}$. Reflections in the basal plane (○) and the hexagonal (00.2) reflection (△). The interpolating lines are guides to the eye.

7. Discussion

Generally, all of the examined systems show the following behavior at low helium concentrations. A broadening of the Debye–Scherrer lines and an increase of the lattice parameter(s). In terms of lattice distortion this evolution can be seen schematically as follows [2].

The produced helium atoms cluster and form bubbles inducing self interstitials and later on dislocation loops in the host lattice. Part of the interstitials are incorporated into forming a dislocation network which broadens the lines. The other part of the interstitials remain in the matrix and consequently contribute to the lattice expansion.

The lattice parameter shifts are strongly anisotropic in the examined hexagonal metal–tritium–helium systems exhibiting a much more pronounced shift along the hexagonal direction than in the basal plane. Apart from this, the systems show different behavior with increasing helium concentration.

In $YT_{0.089}$ the formation of interstitials due to helium clustering and bubble formation widens the lattice in both directions in the beginning, but stronger for the hexagonal one. At helium concentrations higher than 1% the lattice expansion occurs mainly in the hexagonal direction while the basal lattice parameter does not change. The increase in the broadening of the widths occurs rather linearly in both directions with increasing helium content but is more pronounced in the hexagonal direction which can be understood as an indication that the building of the dislocation network occurs preferably in the basal planes. The steplike change in the widths of the hexagonal lines at 0.8%He is remarkable.

In scandium the lattice expansion after 0.8%He occurs, contrary to $YT_{0.089}$, mainly because of the increase in the

basal lattice parameter while the hexagonal one rests unchanged. This difference may be explained by the fact that the elastically stiff direction in Sc is the hexagonal one and it is energetically more favourable to expand the lattice in the basal plane while in Y the hexagonal direction is softer.

At about 4.5%He the broadening of the hexagonal lines in scandium ceases while some broadening is still observed in the basal plane. As not all of the newly formed interstitials can be integrated in the network the lattice parameters expand again.

In the $YT_{0.068}$ sample the lattice expansion is much more pronounced than in $YT_{0.089}$ and at low He concentrations no broadening of the lines is observed at all. At liquid nitrogen temperature the He atoms are still mobile and clustering and bubble formation occurs, but the generated interstitials are not mobile enough to lead to a dislocation network and therefore the widths of the lines remain unchanged. As all of the interstitials rest in the host lattice the volume expansion is higher than in the other systems. From a He concentration of approximately 1% on the basal lines broaden while the hexagonal one shows no clear tendency to do so.

In the Ta–He system the lattice parameter expands till 0.8%He and decreases towards higher He contents. The decrease in the lattice parameter may be due to the incorporation of the isolated defects into the dislocation network in agreement with the fact that the widths of the lines broaden. The balance between production of interstitials by He-clustering and bubble formation and integration of defects into the dislocation network seems to be shifting due to less He production with the lowering of the tritium content.

8. Conclusion

In all investigated metal–tritium systems the damage caused by helium production is initially governed by the creation of finite-size defects due to helium clustering and bubble formation which during later stages is increasingly complemented by their successful incorporation into an interconnected dislocation network. The evolution of the damage is strongly anisotropic in the hexagonal systems and influenced by the elastic properties of the host lattices.

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References

[1] O. Blaschko, G. Ernst, P. Fratzl, G. Krexner, P. Weinzierl, Phys. Rev. B 34 (1986) 4985;
O. Blaschko, G. Ernst, P. Fratzl, G. Krexner, P. Weinzierl, J. Nucl. Mater. 141–143 (1986) 540.

[2] O. Blaschko, J. Pleschiutschnig, R. Glas, P. Weinzierl, Phys. Rev. B 44 (1991) 9164.

[3] R. Laesser, K. Bickmann, H. Trinkaus, Phys. Rev. B 40 (1989) 3306.

[4] R. Laesser, K. Bickmann, H. Trinkaus, Phys. Rev. B 34 (1989) 4364.

[5] S. Thiébaut, V. Paul-Boncour, A. Percheron-Guégan, B. Limacher, O. Blaschko, C. Maier, C. Tailland, D. Leroy, Phys. Rev. B 57 (1998) 10379.

[6] R. Laesser, Tritium and Helium-3 in Metals, Springer, Berlin Heidelberg, 1989.

[7] C.K. Saw, B.J. Beaudry, C. Stassis, Phys. Rev. B 27 (1983) 7013.

[8] D. Khatamian, C. Stassis, B.J. Beaudry, Phys. Rev. B 23 (1981) 624.

[9] W.G. Wolfer, Philos. Mag. A 59 (1989) 87.

[10] M.A. Krivoglaz, Theory of X-ray and Thermal Neutron Scattering by Real Crystals, Plenum, New York, 1969.