

## Resistivity study on the production and migration of helium in $\text{LuT}_r$ ( $r \leq 0.20$ )

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$^3\text{He}$  was produced in tritium-doped lutetium specimens by the  $\beta^-$  decay of tritium. This transmutation caused a time-linear increase of the residual electrical resistivity at a rate which indicates that the resistivity per produced He defect is about twice that of T. This result was independent of the speed at which the specimens were cooled to 4.2 K. Recovery in a subsequent annealing experiment started around 26 K, which is attributed to He atoms becoming mobile. Further recovery showed differences between quenched and slowly cooled specimens, indicating that the mobility of helium is strongly affected by short-range ordering of the tritium atoms.

### I. INTRODUCTION

Properties of metals can be severely degraded by small amounts of noble gases. For example, helium, which is produced by  $(n,\alpha)$  reactions in structural materials of nuclear reactors, promotes void swelling and causes embrittlement by the formation of gas bubbles. This has prompted extensive studies on the diffusion and precipitation of helium in metals.<sup>1</sup> Due to its strong interaction with lattice defects (vacancies, dislocations, grain boundaries), it is difficult to study the properties of interstitial helium atoms experimentally. For the investigation of interstitially dissolved helium, two prerequisites must be fulfilled: (i) the specimen must be charged with helium on interstitial sites without producing lattice defects and (ii) the loading must be performed at cryogenic temperatures where the interstitial helium is immobile. Two different approaches are conceivable.

The first is implantation of helium at low temperatures with energies below the threshold for defect production.<sup>2</sup> This limits the implantation depth to a surface layer of about 10 nm.

The second is the so-called tritium trick,<sup>3</sup> which involves charging of a metal with tritium and waiting for the desired helium concentration according to the initial tritium concentration and the half-life of 12.361 years. The recoil of the  $^3\text{He}$  atom from the maximum  $\beta^-$  energy of 18.6 keV is 3.4 keV and thus far below the threshold for defect generation in any metal host lattice.<sup>4</sup> The tritium trick works well for metals which react with tritium exothermically. Furthermore, one has to demand a high solubility in the  $\alpha$  phase where the lattice structures of the metal and the metal tritides are equal. But in most metals with high solubility for protium (H), deuterium (D), and tritium (T) at elevated temperatures [e.g., group-VB metals<sup>5,6</sup>, and Pd (Ref. 7)] the solubility drops drastically at lower temperatures, causing precipitations of highly concentrated hydride phases. One way of avoiding the formation of concentrated hydrides is the charging of these metals with statistically dissolved impurities such as oxygen or nitrogen which trap the hydrogen atoms at lower temperatures. This method was used by Birnbaum and co-workers.<sup>8</sup> Another method is

to choose metals which are believed to have temperature-independent solvus concentrations below certain critical temperatures. The only known metals which such phase boundaries are Sc, Y, Ho, Er, Tm, and Lu.<sup>9</sup>

For the resistivity measurements to be presented in this paper we have chosen Lu because the  $\alpha$  phase concentration of  $\sim 20\%$  (Refs. 9 and 10) is preserved down to 4.2 K for H and D. Due to the fully occupied  $4f$  states, magnetic effects are neither expected nor observed in Lu, in contrast to  $\text{ErH}_x$  or  $\text{TmH}_x$ .<sup>11</sup> A peculiarity of these metal hydrides is the observation of an anomaly in the electrical resistivity<sup>12</sup> during cooling and heating which indicates some microscopic changes. In the case of  $\text{LuD}$ , this anomaly could be attributed to a linear ordering of deuterium pairs along the  $c$  axis of the hcp structure with a Lu atom in between.<sup>13</sup> For the present study, we assume that the isotopic dependence of the solvus line and of the structure of the pairs is negligible. This means that the  $^3\text{He}$  atoms are generated in the  $\alpha$  phase of the Lu-T system, but that locally a nonstatistical distribution of tritium atoms may exist which depends on the details of the cooling procedure.

### II. EXPERIMENTAL PROCEDURE

Lutetium was obtained from Ames Laboratories in the form of ribbons, 1 mm wide and 0.2 mm thick, of 99.99-wt. % purity and less than 0.3-at. % impurity of gases and carbon. Room-temperature to liquid-He temperature residual resistivity ratios ranged from 25 to 50. The specimens were loaded with tritium in a special tritium charging facility<sup>14,15</sup> at temperatures between 800 and 1000 K and pressures up to  $1.5 \times 10^4$  Pa. The  $\text{LuT}_{r-1}$  and  $\text{LuT}_{r-2}$ , and the  $\text{LuT}_{r-3}$  and  $\text{LuT}_{r-4}$  samples (see Table I) were prepared in two different charging runs. To minimize reaction of the Lu samples with impurities during loading, the  $\text{LuT}_{r-3}$  and  $\text{LuT}_{r-4}$  samples were wrapped into thin Lu foils. The purity of the  $\text{T}_2$  gas used was 96.2 at. % T, 0.7 at. %, D, and 3.1 at. % H, as determined by mass spectrometric analysis. The tritium concentration (given as the ratio of tritium to metal atoms) was determined from the pressure drop in the

TABLE I. Resistivity  $\rho$  ( $\mu\Omega$  cm) of four LuT<sub>r</sub> specimens at 4.2 and 296 K, resistivity increase by tritium in Lu at 4.2 K [after slowly cooling (SC) and after quenching (Q)], and 296 K, respectively. Tritium concentrations  $r$  given as ratio of T to Lu atoms are determined from resistivity increases ( $r_p^{4.2}$  and  $r_p^{296}$ ) and pressure drops ( $r_p$ ), respectively.

Specimen	$\rho_{Lu}^{4.2}$	$\rho_{Lu}^{296}$	$\rho_{LuT_r}^{4.2} - \rho_{Lu}^{4.2}$		$\rho_{LuT_r}^{296} - \rho_{Lu}^{296}$	$r_p^{4.2}$		$r_p$
			SC	Q		Q	$r_p^{296}$	
LuT <sub>r</sub> -1	2.44	71.1	58.8	60.8	49.7	0.210	0.216	0.175
LuT <sub>r</sub> -2	2.03	71.8	73.9	75.1	64.8	0.259	0.282	
LuT <sub>r</sub> -3	1.36	63.3	20.1	23.1	20.0	0.080	0.087	0.129
LuT <sub>r</sub> -4	1.33	64.2	24.5	27.6	22.4	0.095	0.097	

known volume, assuming that both samples absorbed the same T concentration, and from the increase in resistivity after loading (see Table I). Resistivity changes were converted to concentrations by dividing by 2.9  $\mu\Omega$  cm/at. % at 4.2 K after quenching, and by 2.3  $\mu\Omega$  cm/at. % at 296 K, respectively. The tritium concentrations determined from the weight gain during the charging procedure were found to be larger than the amount of T gas in the reaction chamber, probably due to the reaction of Lu with heavier gas impurities in the tritium.

The concentrations derived from resistivity measurements and pressure drop are only in poor agreement but show no systematic deviations. The main reason for the poor agreement are probably also additionally absorbed impurities which have a larger specific resistivity than T. As all further results were derived from resistivity measurements, the  $r_p$  values of Table I were used in the following.

Two specimens were mounted on a specimen holder by four gilded clamps which served for current leads and voltage grips. The gauge length was about 1.5 cm. The specimen holder was put into the specimen chamber of a helium-bath cryostat.<sup>16</sup> The specimen chamber was filled with helium gas which was condensed by thermal conductivity from the bath. The separation of chamber and bath avoided contamination of the cooling system by the tritium of the specimens. The helium-gas pressure in the specimen chamber was about  $2.5 \times 10^4$  Pa. Three furnaces along the support tube of the specimen chamber allowed annealing of the specimens at temperatures from about 10 to 800 K. Temperatures above room temperature were not used, to avoid tritium releases of the LuT<sub>r</sub> samples. Before opening the support tube for exchange of specimens it was evacuated through a tubing system containing columns of heated (820 K) CuO granulate and cold (300 K) zeolite<sup>17</sup> to retain traces of tritium.

The temperature of the specimen was measured by standard AuFe-NiCr and Cu-CuNi thermocouples which were calibrated by immersion into liquid helium, liquid nitrogen, and ice water. The temperatures read from the two thermocouples, from a miniature Pt 100 resistor in place of the specimens, and from the temperature gauges of the furnaces (Ge and Pt resistors) agreed within 1 K.

The specimens were cooled to 4.2 K by different treatments: (1) Quenching (Q) from room temperature into

the condensed liquid helium in the specimen chamber at a cooling rate of about 6 K/s, (2) slowly cooling (SC) in the annealing furnace at about 0.04 K/s, (3) quenching to 4.2 K and subsequent aging for 14 h at 161 K (process A1), and (4) quenching to 4.2 K and subsequent aging for 1 h at 181 K (process A2).

After that, the LuT<sub>r</sub> specimens were held for periods of 5–28 days at 4.2 K with resistance measurements at a measuring current of 0.1 A. After this period, the temperature was raised during an isochronal ( $\Delta t = 300$  s) annealing program in logarithmic steps of  $\Delta T/T = 0.2$  with intermediate resistance measurements at 4.2 K.

### III. RESULTS

Figure 1 shows derivatives of the electrical resistivities of LuT<sub>r</sub> specimens during cooling (cooling rate, 0.04 K/s;  $\nabla$ , LuT<sub>r</sub>-1;  $\blacktriangledown$ , LuT<sub>r</sub>-2) and heating (heating rate, 0.02 K/s;  $\triangle$ , LuT<sub>r</sub>-1;  $\blacktriangle$ , LuT<sub>r</sub>-2). After slowly cooling (solid lines) with a rate of 0.04 K/s, the heating curves show a smooth step around 185 K, while after quenching (dashed line), a minimum and a turning point are observed at 175 and 180 K, respectively. This resistivity anomaly compares to results in Ref. 12, where the turning point is found at 170 K in LuD<sub>r</sub> and at 164 K in LuH<sub>r</sub> and is attributed<sup>13</sup> to the pairing of hydrogen

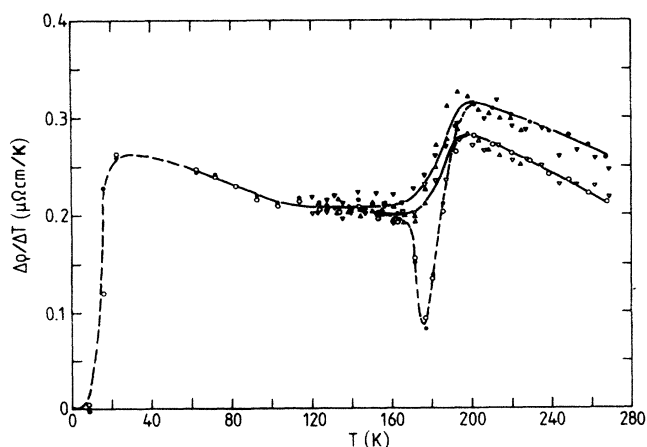


FIG. 1. Derivatives  $\Delta\rho/\Delta T$  of specimens LuT<sub>r</sub>-1 and LuT<sub>r</sub>-2 during cooling at a rate of 0.04 K/s ( $\nabla$ ,  $\blacktriangledown$ ) and subsequent heating at 0.02 K/s ( $\triangle$ ,  $\blacktriangle$ ), respectively. Included are data measured during heating ( $\circ$ ,  $\bullet$ ) after quenching to 4.2 K.

TABLE II. Normalized slope of resistivity increase during aging LuT<sub>r</sub> specimens at 4.2 K and total resistivity change  $\Delta\rho_0$  accumulated during aging time  $t_0$  at 4.2 K. SC, slowly cooling from 296 to 4.2 K with about 0.04 K/s; Q, quenching from 296 to 4.2 K with about 6 K/s; A1, Q plus aging for 14 h at 161 K; and A2, Q plus aging for 1 h at 181 K.

No.	Specimen	Treatment	$\frac{\Delta\rho(t)/\Delta t}{\rho_{LuT_r}^{4.2} - \rho_{Lu}^{4.2}}$ ( $10^{-9}$ per sec)	$\Delta\rho_0$ ( $10^{-9}$ $\Omega$ cm)	$t_0$ ( $10^6$ sec)
1	LuT <sub>r</sub> -1	SC	2.36	186.6	1.41
2	LuT <sub>r</sub> -2	SC	2.13	250.7	1.41
3	LuT <sub>r</sub> -3	Q	1.75	82.24	2.04
4	LuT <sub>r</sub> -4	Q	1.70	99.87	2.04
5	LuT <sub>r</sub> -3	Q	1.48	29.98	0.83
6	LuT <sub>r</sub> -4	Q	1.85	40.47	0.83
7	LuT <sub>r</sub> -3	SC	2.05	37.77	0.82
8	LuT <sub>r</sub> -4	SC	1.79	40.58	0.82
9	LuT <sub>r</sub> -3	A1	1.32	11.10	0.52
10	LuT <sub>r</sub> -4	A1	1.62	24.29	0.52
11	LuT <sub>r</sub> -3	A2	1.94	25.91	0.60
12	LuT <sub>r</sub> -4	A2	1.97	31.20	0.60

atoms. Thus, with heavier hydrogen isotope, the turning point is shifted to higher temperatures. We are unable to explain the difference between the derivatives of the resistivity of the LuT<sub>r</sub>-1 and -2 samples in Fig. 1.

Figure 2 shows the resistivity increase of LuT<sub>r</sub> specimens during holding at 4.2 K after slowly cooling and after quenching. The measured resistivity changes are normalized to the resistivity increase caused by  $T$  immediately after quenching ( $\rho_{LuT_r} - \rho_{Lu}$ , Q) as given in Table I. The averaged normalized slopes are, after slowly cooling,  $(2.24 \pm 0.1) \times 10^{-9} \text{ s}^{-1}$  for the LuT<sub>r</sub>-1 and -2 specimens. The respective values for LuT<sub>r</sub>-3 and -4 are  $(1.70 \pm 0.06) \times 10^{-9} \text{ s}^{-1}$  after quenching and  $(1.92 \pm 0.1) \times 10^{-9} \text{ s}^{-1}$  after slowly cooling. Results of further runs which are not plotted in Fig. 2 are summarized in Table II.

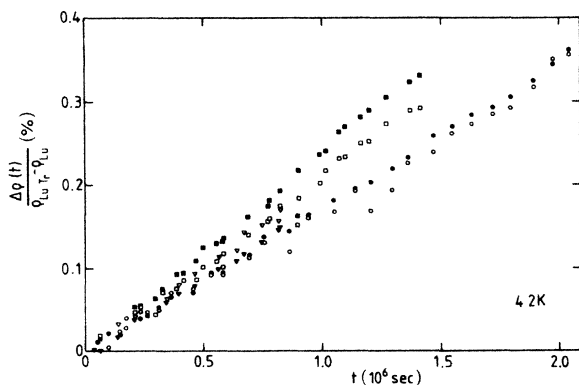


FIG. 2. Resistivity increase during aging at 4.2 K of LuT<sub>r</sub>-1 and LuT<sub>r</sub>-2 after slowly cooling ( $\square, \blacksquare$ ) and of LuT<sub>r</sub>-3 and LuT<sub>r</sub>-4 after slowly cooling ( $\nabla, \blacktriangledown$ ) and after quenching ( $\circ, \bullet$ ), respectively.  $\Delta\rho$  is normalized to the tritium-induced resistivity at 4.2 K immediately after quenching (Table I).

Figure 3 shows the results of isochronal annealing experiments. The dashed (LuT<sub>r</sub>-1) and dash-dotted (LuT<sub>r</sub>-2) lines are results after slowly cooling the specimens to 4.2 K, while the circles give the results after holding them at 4.2 K for 1.41 Msec (see Fig. 2 and Table II). The measurements after cooling only show a sharp dip around 175 K and a nonuniform behavior at higher temperatures. On the other hand, after aging at

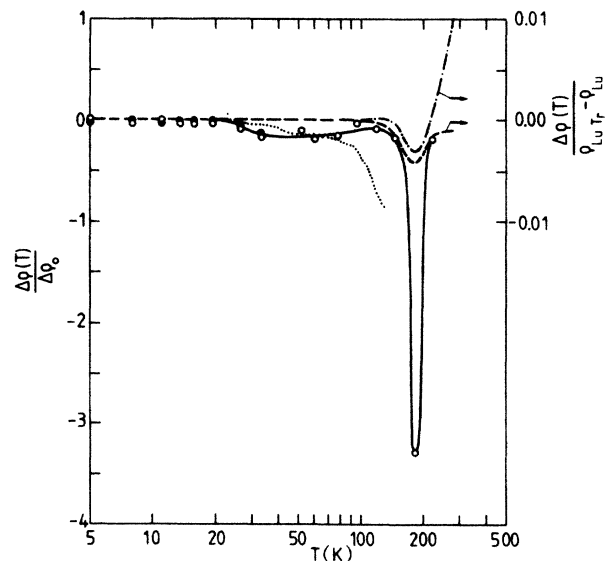


FIG. 3. Relative recovery of the resistivity accumulated during aging of LuT<sub>r</sub> specimens 1 ( $\circ$ ) and 2 ( $\bullet$ ) at 4.2 K after slowly cooling. Included are the results (right-hand-side ordinate) of an annealing experiment immediately after slowly cooling (1, ---; 2, -.-.-). These latter measurements are normalized to the tritium-induced resistivity (Table I). The dotted line gives the recovery after 1.7-MeV electron irradiation (Ref. 18) of pure Lu for comparison.

4.2 K, i.e., when some of the tritium is transformed to  $^3\text{He}$ , the normalized resistivity decreases already around 26 K by about 14% (specimen LuT<sub>r</sub>-2 failed at 50 K). The absolute depths of the resistivity minima at 175 K of LuT<sub>r</sub>-1 are 0.249  $\mu\Omega$  cm before and 0.225  $\mu\Omega$  cm after transformation, respectively. Included in Fig. 3 is a recovery curve of Lu after 1.7-MeV electron irradiation<sup>18</sup> for comparison (see dotted curve).

Figure 4 shows the results of isochronal annealing experiments with specimen LuT<sub>r</sub>-4 before and after aging at 4.2 K. Before aging again only a dip around 175 K is observed, while the resistivity of the  $^3\text{He}$ -containing specimens starts to decrease already around 26 K. This initial step is rather broad, extending up to about 60 K, and its magnitude depends on the cooling procedure. After slowly cooling (SC,  $\nabla$ ) or after quenching and aging for 1 h at 181 K (A2,  $\times$ ) it amounts to about 60–70% of the resistivity accumulated during aging at 4.2 K. After quenching to 4.2 K (Q,  $\circ$  and  $\bullet$ ) or after aging for 14 h at 161 K (A1,  $+$ ) a recovery of 180–200% is observed. The absolute depth of the resistivity minima of specimen LuT<sub>r</sub>-4 around 177 K are, on the average, 0.75  $\mu\Omega$  cm after quenching and 0.21  $\mu\Omega$  cm after slowly cooling, respectively.

As already mentioned, the behavior above the T-ordering temperature ( $\approx 175$  K) is sometimes nonuniform, even in the He-free specimens. This is due to minor differences in cooling rate which produce relatively large changes in residual resistivity. Nevertheless, the measurements in Figs. 3 and 4 indicate that passing through the T-ordering temperature causes no significant changes of the  $^3\text{He}$  contribution to the residual resistivity.

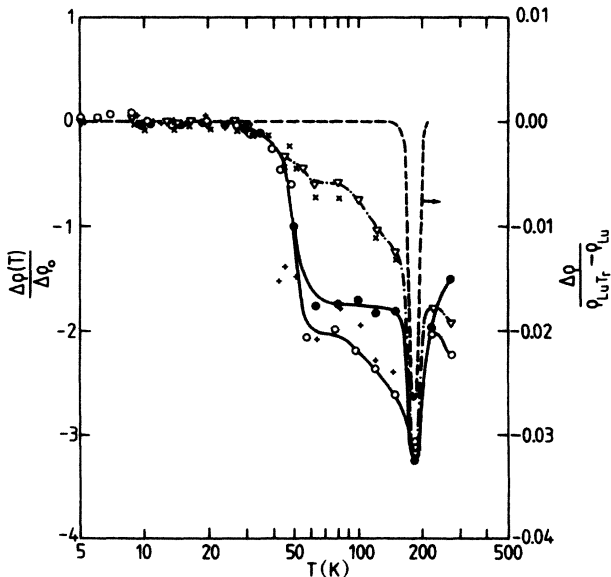


FIG. 4. Relative resistivity recovery of LuT<sub>r</sub>-4 before (---, right-hand-side ordinate) and after aging at 4.2 K. The symbols indicate the different pretreatments (cooling, aging time) as given by numbers in Table II: 4 (●), 6 (○), 8 (∇), 10 (+), and 12 (×).

#### IV. DISCUSSION

The resistance anomaly found in the Lu-H and Lu-D systems<sup>12</sup> is also observed in LuT<sub>r</sub> (Fig. 1). The turning point of the derivative found in our quenched specimens shows a temperature shift from D to T similar to that observed from H to D.<sup>12</sup> This indicates an isotope dependence of the diffusion with a smaller diffusion coefficient for the heavier hydrogen isotopes.

This resistance anomaly has been attributed<sup>13</sup> to linear ordering of hydrogen pairs along the *c* axis with a Lu atom in between, possibly with the formation of larger clusters of such pairs. Thus it is possible to vary the tritium distribution from isolated atoms to pairs or larger clusters by adjusting the cooling rate in the temperature range of the resistance anomaly.

The subsequent increase in residual resistivity  $\Delta\rho$  during aging at 4.2 K (Fig. 2) is attributed to the transmutation of T to  $^3\text{He}$ :

$$\Delta\rho(t) = \Delta c(t)(\rho_{\text{He}} - \rho_{\text{T}}). \quad (1)$$

$\rho_{\text{He}}$  and  $\rho_{\text{T}}$  are the resistivity contributions per unit concentration of He and T, respectively. The produced He concentration,  $\Delta c = -\Delta r$ , is obtained by

$$\frac{\Delta r}{\Delta t} = -\frac{r}{\tau}, \quad (2)$$

where  $\tau = T_{1/2}/\ln 2$  (with  $T_{1/2} = 12.361$  y, the half-life of tritium). The T concentration  $r$  is obtained from (Matthiessen's rule)

$$\rho_{\text{LuT}_r}^{4.2} - \rho_{\text{Lu}}^{4.2} = r\rho_{\text{T}} + \rho_i. \quad (3)$$

The difference on the left-hand side is the resistivity increase after loading with tritium and quenching to 4.2 K. The term  $\rho_i$  takes into account resistivity contributions from contaminations of the tritium gas by other soluble gases. As Table I shows no general difference between the concentrations determined from resistivity and pressure drop, we assume  $\rho_i = 0$  and obtain

$$\frac{\rho_{\text{He}}}{\rho_{\text{T}}} = \frac{T_{1/2}}{\ln 2} \frac{\Delta\rho/\Delta t}{\rho_{\text{LuT}_r}^{4.2} - \rho_{\text{Lu}}^{4.2}} + 1. \quad (4)$$

The  $\rho_{\text{He}}/\rho_{\text{T}}$  values show no significant dependence either on the T concentration or on the cooling procedure, giving an average value of  $2.03 \pm 0.16$ . That means that the resistivity per gas atom is almost exactly doubled during the T- $^3\text{He}$  transmutation.

A possible contribution to the resistivity increase from, e.g., separation of T-T pairs by impacts of electrons produced by the  $\beta^-$  decay can be neglected, as the probability of this process is very low and its contribution to resistivity would be in the percent range.

The above result immediately explains why up to 200% recovery of the transmutation-induced resistivity is observed during annealing (Fig. 4). When an amount  $\Delta c$  of the total transmuted He concentration  $\Delta c_0$  becomes mobile, the resistivity changes as

$$\Delta\rho(T) = \Delta c(T)(\rho'_{\text{He}} - \rho_{\text{He}}), \quad (5)$$

where  $\rho_{\text{He}}$  is the resistivity of the He atom in its original position and  $\rho'_{\text{He}}$  in its position after the annealing stage.  $\rho'_{\text{He}}$  may also contain the resistivity contributions from self-interstitial atoms (SIA's) emitted during the growth of He bubbles. Combining with Eq. (1) gives

$$\frac{\Delta\rho(T)}{\Delta\rho_0} = \frac{\Delta c(T)}{\Delta c_0} \frac{\rho'_{\text{He}} - \rho_{\text{He}}}{\rho_{\text{He}} - \rho_{\text{T}}} \quad (6)$$

Inserting  $\rho_{\text{He}}/\rho_{\text{T}}=2$  and  $\Delta\rho(T)/\Delta\rho_0=-2$  (quenched specimens in Fig. 4), one obtains only one physically meaningful solution, namely, both extreme possible values  $\Delta c(T)/\Delta c_0=1$  and  $\rho'_{\text{He}}/\rho_{\text{T}}=0$ . That is, all He atoms and/or SIA's are involved in the recovery process and their resistivity vanishes in this stage completely. The straightforward explanation is disappearance of helium to unsaturable sinks like surfaces, grain boundaries or large bubbles, and generation of SIA loops. This result excludes formation of smaller He clusters in the quenched specimens, as in this case  $\rho'_{\text{He}}/\rho_{\text{T}}$  would remain finite. The incomplete recovery in the slowly cooled specimens (Fig. 3 and dash-dotted curve in Fig. 4) may indicate that in these specimens some of the helium is, indeed, retained in the matrix. This retention probably is due to trapping of helium at tritium agglomerates (pairs or larger clusters) which are present in these specimens. This assumption is corroborated by the observation that the retention is higher in the more concentrated specimens (Fig. 3) than in the less concentrated ones ( $\nabla$  in Fig. 4). The fact that the He resistivity recovers completely in the quenched specimens shows that He-T complexes are unstable, at least beyond 60 K, otherwise mobile He atoms would be trapped at T with probably only a minor reduction in their resistivity contribution. If, in the slowly cooled specimens, only single and paired T atoms are present, He atoms produced in He-T pairs would readily dissociate. If trapping at T-T pairs would be effective, all He atoms would be trapped again with only a small effect on their resistivity. Thus the recovery in the slowly cooled specimens indicates that only a relatively low concentration of traps is present in these specimens. A comparison of the specimens aged at 161 and 181 K—i.e., at the lower and upper ends of the anomaly region, respectively—gives some indication that it is not T-T pairs but larger complexes which trap helium in the slowly cooled specimens. Figure 5 shows the decrease of resistivity of specimen LuT<sub>r</sub>-4 after quenching during isothermal annealing at 161 and 181 K, respectively. The resistivity change is normalized to a value of  $(\rho_{\text{LuT}_r} - \rho_{\text{Lu}})$  interpolated from the 4.2- and 296-K values given in Table I. While at 181 K the resistivity attains a constant value after about 20 min, the resistivity at 161 K is still decreasing even after 23 h. The curves show that aging for 1 h at 181 K (A2; ●) suffices to attain equilibrium, while 14 h at 161 K (A1; ○) does not. This means that after 14 h at 161 K (A1) very large clusters which form only toward the end of the treatment may not yet have formed despite the fact that a larger fraction of tritium is agglomerated, probably in smaller clusters.

As resistivity recovery after aging takes place in the same temperature regime (Fig. 3) as after electron irradiation<sup>18</sup> it is tempting to assume a process involving self-interstitials: If He is already mobile at 4.2 K and forms bubbles during aging, growth beyond a certain size  $n_0$  (He atoms per bubble) may proceed by emission of SIA's (He self-trapping<sup>19</sup>). The SIA's would contribute to resistivity (Fig. 2), become mobile at 26 K, and then agglomerate, causing resistivity recovery. While SIA's may eventually form and contribute to recovery in both models, the basic question remains as to whether He is mobile and forms bubbles at  $T \geq 26$  K or already at 4.2 K. In the second case, a rather complex behavior during aging should be expected and Eq. (1) would become

$$\Delta\rho = \Delta c (\rho'_{\text{He}} + \nu\rho_{\text{SIA}} - \rho_{\text{T}}) \quad (1')$$

$\rho'_{\text{He}}$  is the average resistivity contribution per He atom in a cluster and is probably decreasing during bubble growth, eventually becoming zero for very large bubbles.  $\nu$  is the number of SIA's emitted per He atom entering a bubble.  $\nu$  is zero for  $n < n_0$  [ $n_0 \approx 5$  according to estimates for fcc Ni (Ref. 19)] and eventually becomes constant [ $\approx 0.5$  (Ref. 19)]. At still higher concentrations, the bubble surface may even become saturated with SIA's and spontaneous formation of loops may occur. As the bubble density and details of the processes involved are not known, no precise predictions are possible at which  $\Delta c$  values the slopes of the resistivity curves in Fig. 2 should be expected to change. For example, bubble nucleation may already have been completed before the onset of measurement, as due to the time needed for loading and mounting the specimens a He concentration of about  $8 \times 10^{-5}$  was present in LuT<sub>r</sub>-2 and -3 at the beginning of the first aging if losses to the surface are neglected. If bubbles have then already nucleated at a density sufficient to suppress further nucleation and of sizes large enough to emit SIA's, this could explain why all curves in Fig. 2 show essentially the same slope, despite He concentrations increasing in LuT<sub>r</sub>-2 and -3

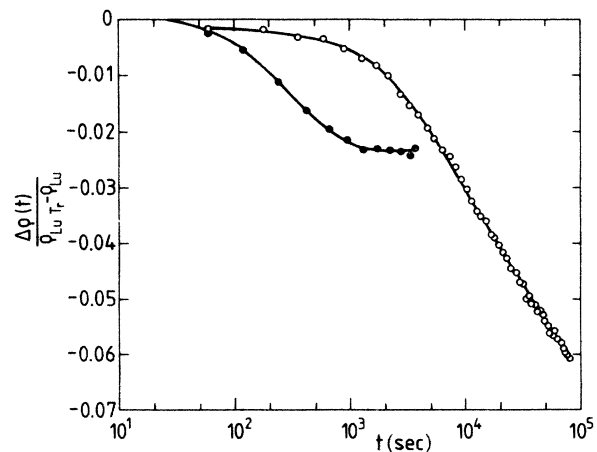


FIG. 5. Relative decrease of electrical resistivity in specimen LuT<sub>r</sub>-4 during aging at 161 K (○) and 181 K (●) after quenching, respectively.

up to about  $1.3 \times 10^{-3}$  during the last experiment. One would therefore have to assume that all measurements in Fig. 2 were taken in a period when bubble nucleation has been completed and agglomeration of SIA's has not yet started. In this case Eq. (1') can be used to determine  $\nu$ : In Ref. 20, 60–90  $\mu\Omega$  cm/at. % was given for the Frenkel-pair resistivity  $\rho_F$  in Lu. If we ascribe, according to results from other metals,<sup>21,22</sup> 70% of  $\rho_F$  to the SIA (i.e.,  $\rho_{\text{SIA}} \approx 50 \mu\Omega$  cm/at. %) and use  $\rho_T = 2.9 \mu\Omega$  cm/at. %, then Eq. (1') yields an upper limit (for  $\rho'_{\text{He}} \approx 0$ ) for  $\nu$  of 0.12, which is considerably below the theoretical estimate<sup>19</sup> of 0.5. Swelling measurements in  $\text{VT}_{0.02}$ ,  $\text{NbT}_{0.59}$ ,<sup>23</sup> and  $\text{TaT}_r$  ( $r = 0.103$  and  $0.42$ )<sup>24</sup> at room temperature conform with  $\nu \approx 0.5$ , while the swelling curves of  $\text{LuT}_{0.13}$  (Ref. 23) at room temperature show a—yet unexplained—change in slope corresponding to  $\nu$  values of 0.5 initially and 0.24 later, respectively. The swelling rates of  $\text{LuT}_{0.1}$  at 4.2 K (Ref. 23) yield  $\nu \approx 0.24$ , if bubble growth at 4.2 K is assumed. This value is lower by a factor of 2 than the corresponding value at room temperature and higher by a factor of 2 than the value derived above. These inconsistencies may indicate that the underlying assumption that He is already mobile at 4.2 K is wrong. Therefore we have to attribute the present resistivity increase during aging (Fig. 2) and the swelling at 4.2 K (Ref. 24) to interstitial He. Thus we obtain for the resistivity per unit concentration of He,  $\rho_{\text{He}} = 5.8 \mu\Omega$  cm/at. % (or  $\rho_{\text{He}}/\rho_{\text{SIA}}$

$\approx 0.12$ ). This value and the relative volume change per interstitial He atom  $(\Delta V/\Omega_0)/(\text{He atom}) = 0.24$  ( $\Omega_0$  is the atomic volume)<sup>23</sup> are not unreasonable when compared to data for interstitial hydrogen atoms in Lu (or in group-Vb metals<sup>25</sup>) which are  $\rho_{\text{H}}/\rho_{\text{SIA}} \approx 0.06$  ( $\sim 0.1$ ) and  $(\Delta V/\Omega_0)/(\text{H atom}) \approx 0.10$  ( $\sim 0.17$ ).

## V. CONCLUSION

The present experiments have shown that resistivity measurements on specimens loaded with tritium yield quantitative information on physical properties and on the mobility of helium in metals. The results clearly indicate that  $^3\text{He}$  is mobile at least at 26 K. Mobility already at 4.2 K cannot be completely ruled out.

The recovery spectra after  $\text{T} \rightarrow ^3\text{He}$  transmutation are sensitive to local arrangement of the T atoms. Therefore He can be used as a probe to study ordering processes of hydrogen in metals in more detail. Similar studies are possible in other metals possessing a temperature-independent solvus line for hydrogenlike Sc, Y, Ho, Er, and Tm.

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<sup>1</sup>Proceedings of the International Symposium on Fundamental Aspects of Helium in Metals, edited by H. Ullmaier [Radiat. Eff. **78**, 1 (1983)].

<sup>2</sup>G. J. Thomas and R. Bastasz, J. Appl. Phys. **52**, 6426 (1981).

<sup>3</sup>G. J. Thomas, W. A. Swansiger, and M. I. Baskes, J. App. Phys. **50**, 6942 (1979).

<sup>4</sup>P. Jung, Radiat. Eff. **35**, 155 (1978).

<sup>5</sup>R. Lässer and K. Bickmann, J. Nucl. Mater. **126**, 234 (1984); R. Lässer and T. Schober, J. Less-Common Met. **130**, 453 (1987).

<sup>6</sup>R. Lässer and K. Bickman, J. Nucl. Mater. **132**, 244 (1985).

<sup>7</sup>R. Lässer, J. Phys. Chem. Solids **46**, 33 (1985).

<sup>8</sup>C. G. Chen, H. K. Birnbaum, and A. B. Johnson, Jr., J. Nucl. Mater. **79**, 128 (1979).

<sup>9</sup>J. P. Burger, J. N. Daou, A. Lucasson, P. Lucasson, and P. Vajda, Z. Phys. Chem. (Neue Folge) **143**, 111 (1985).

<sup>10</sup>J. N. Daou, A. Lucasson, and P. Lucasson, Solid State Commun. **19**, 895 (1976).

<sup>11</sup>P. Vajda and J. N. Daou, J. Less-Common Met. **101**, 269 (1984); J. N. Daou, P. Vajda, A. Lucasson, and P. Lucasson, in *Point Defects and Defect Interactions in Metals*, edited by J. Takumura, M. Doyama, and M. Kiritani (University of Tokyo Press, Tokyo, 1982), p. 325.

<sup>12</sup>P. Vajda, J. N. Daou, J. P. Burger, K. Kai, K. A. Gschneidner, Jr., and I. J. Beaudry, Phys. Rev. B **34**, 5154

(1986); J. N. Daou, P. Vajda, A. Lucasson, P. Lucasson, and J. P. Burger, Philos. Mag. A **53**, 611 (1986).

<sup>13</sup>O. Blaschko, G. Krexner, J. N. Daou, and P. Vajda, Phys. Rev. Lett. **55**, 2876 (1986).

<sup>14</sup>R. Lässer, K.-H. Klatt, P. Mecking, and H. Wenzl, Kernforschungsanlage Jülich, Report No. JÜL-1800, 1982 (ISSN 0366-0885).

<sup>15</sup>R. Lässer and K.-H. Klatt, Phys. Rev. B **28**, 748 (1983).

<sup>16</sup>W. Petry, Ph.D. thesis, Freie Universität Berlin, 1981.

<sup>17</sup>M. Schüller, J. Knieper, and H. Prietz, Kernforschungsanlage Jülich Internal Report ICH 1, 1977 (unpublished).

<sup>18</sup>J. N. Daou, J. E. Bonnet, P. Vajda, A. Lucasson, and P. Lucasson, Phys. Lett. **63A**, 158 (1977).

<sup>19</sup>W. D. Wilson, C. L. Bisson, and M. I. Baskes, Phys. Rev. B **24**, 5616 (1981).

<sup>20</sup>J. N. Daou, J. E. Bonnet, P. Vajda, M. Biget, A. Lucasson, and P. Lucasson, Phys. Status Solidi A **40**, 101 (1977).

<sup>21</sup>R. W. Siegel, J. Nucl. Mater. **69&70**, 117 (1978).

<sup>22</sup>P. Jung, J. Nucl. Mater. **117**, 70 (1983).

<sup>23</sup>T. Schober, R. Lässer, C. Dieker, and H. Trinkaus, J. Less-Common Met. **131**, 293 (1987).

<sup>24</sup>T. Schober, R. Lässer, J. Golczewski, C. Dieker, and H. Trinkaus, Phys. Rev. B **31**, 7109 (1985).

<sup>25</sup>Data reviewed by H. Peisl, in *Hydrogen in Metals I*, edited by G. Alefeld and J. Völkl (Springer-Verlag, Berlin, 1978).