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ANELASTIC EFFECTS IN LUTETIUM CONTAINING HYDROGEN (OR DEUTERIUM) IN SOLID SOLUTION

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Abstract.—The internal friction of $\alpha$-LuH(D)$_x$, with $0 \leq x \leq 0.2$, has been measured in the range $4.2 \leq T \leq 450$ K both in the annealed state and after deformation. Three main peaks were observed and designated according to their characteristics: (1)"H"-peak — at 215-225 K, with an $x$-dependent amplitude and an isotope-dependent temperature; (2)"d-H"-peak — at 250-260 K, appears after deformation in the H(D)-containing samples only; (3)"d"-peak — at 340-350 K, exists in all deformed samples. A tentative interpretation implying H-pairs at low temperature is proposed.

Introduction.—The systems rare earth-hydrogen have provoked the interest of research workers for quite some time now (1), since these metals absorb hydrogen readily and are forming hydrides upto LnH$_3$ under normal conditions. Among the lanthanides, the three heaviest trivalent ones — Lu, Tm and Er — exhibit an additional particularity. These h.c.p. metals retain hydrogen in solid solution ($\alpha$-phase) at room temperature and below in important amounts: upto 20, 11 and 3 at.% respectively (2). Moreover, the above solubility limits seem to be independent of temperature down to liquid helium, designating these metals as excellent candidates for the study of hydrogen behaviour at low temperatures. A first investigation (3) of the electrical resistivity of $\alpha$-Lu(H) had revealed an anomaly around 170 - 180 K. To determine its origin and to look for eventual analogies in the neighbours of lutetium, a series of detailed studies has been started on the systems $\alpha$-RE(H), with RE = Lu, Tm or Er. The influence of a quench and/or irradiation with electrons of subthreshold energies as concerns the metal atom displacement upon the systems $\alpha$-Tm(H,D) (4) and $\alpha$-Er(H,D) (5) as well as a pronounced isotope effect have been interpreted in the framework of a model, which assumes a short-range order (possibly pairs) of the hydrogen atoms at temperatures below the anomaly. The present study of internal friction in specimens of $\alpha$-Lu(H,D) is intended as a complementary investigation, which might allow to throw additional light upon the configuration of hydrogen in these systems.

Experimental.—The specimens were prepared from 250 $\mu$m thick lutetium foils with a stated metallic purity of 99.9 at.%. The main contamination stems from neighbouring lanthanides, Mg, Si and Pb. Gaseous impurities have not been determined, but do certainly include several tenths of at.% O$_2$ and N$_2$. The initial hydrogen contents has
been eliminated by outgassing at 1000°C in a vacuum of \(<10^{-6}\) Torr. The charging procedure consisted in heating at 650°C in a H (or D) atmosphere of a corresponding pressure. The final specimens had the dimensions of 60 x 1.5 x 0.25 mm\(^3\) and included \(\text{Lu, LuH}_{0.05}, \text{LuH}_{0.1}, \text{LuH}_{0.2}, \text{and LuD}_{0.05}, \text{LuD}_{0.1}, \text{LuD}_{0.2}\). Internal friction was observed by means of a torsional pendulum working in the temperature range 4 - 450K, with frequencies between 0.18 and 1.5 Hz\(^{6}\). The torsional deformation was made in situ at room temperature.

Results and Discussion.

1) Role of hydrogen - In Fig. 1 we show the temperature dependence of the internal friction and of the frequency square for annealed (at 470 K) specimens of \(\text{Lu, LuH}_{0.1}\) and \(\text{LuD}_{0.1}\) selected for comparison. In the entire measured temperature range between liquid helium and 450 K, the only significant structure is the appearance of a peak in the hydrogenated specimens, at 215 K in \(\text{LuH}_{0.1}\) and at 225 K in \(\text{LuD}_{0.1}\). (At the peak temperature, we have \(\frac{QAG}{G}\approx 5\) and a 40\% larger width than Gaussian.)

If cooled right down from room temperature, the samples exhibited the same peak, which we shall call "H"-peak, but superposed by other structures due to cold work to be discussed in subsection 2.

The peak temperature is isotope- but not concentration-dependent, while the peak amplitude is, as demonstrated in Fig. 2. Here, we have plotted the amplitude of the "H"-peak as a function of the H (or D)-contents in the specimens, and we note that its evolution can be reasonably well represented by a linear growth.

![Figure 1](image1.png)

**Fig. 1.** - Internal friction and square of the frequency as a function of temperature for the annealed specimens of \(\text{Lu, LuH}_{0.1}\) and \(\text{LuD}_{0.1}\).

![Figure 2](image2.png)

**Fig. 2.** - Amplitude of the "H"-peak as a function of \(\text{H(D)}\) concentration. To the right is plotted the "H"-peak, with its background subtracted, for the three pairs of specimens to show the isotope effect on its temperature.
Finally, in the case of the LuH$_{0.2}$ specimen, we have varied the applied frequency by a factor of about 8 and determined the activation energy of the mechanism giving rise to the "H"-peak as $E_a = 0.54 \pm 0.05$ eV, with a preexponential factor of $3 \times 10^{-13}$ s (Fig. 3). An extrapolation towards zero frequency would situate this process in a "static" regime near 200 K.

The fact that the "H"-peak exists only in the hydrogenated specimens, the linear concentration dependence of its amplitude as well as the isotope effect on the peak temperature, qualify it as due to the relaxation of hydrogen in the lutetium lattice. The role of other impurities can be excluded, since the linear peak growth (upto 20%) precludes an interaction with foreign atoms, unless one assumes non-saturable trapping. The latter is not likely, in view of the fact that oxygen and nitrogen - the only impurities which could play a role due to their abundance - are most probably bonded in oxide and nitride compounds and thus only available to a limited extent.

From channeling investigations (7) it was determined that deuterium (and thus probably hydrogen) atoms are occupying the tetrahedral site through the whole investigated temperature range between 25 and 400 K, with no specific feature around 200 K. Now it is known from symmetry considerations that, in a hexagonal lattice, an isolated defect in a tetrahedral site should not give rise to an internal friction peak like that observed in this study. On the other hand, the temperature of the "H"-peak when extrapolated towards zero frequency, $\sim$200 K, corresponds closely to the end of the anomaly region in resistivity studies (3-5), thus probably reflecting the same mechanism. There (in particular in ref. 4 and 5) it had been suggested that H-atoms might interact at short distance to form pairs when the sample was at low temperature. When approaching the anomaly region upon heating the pairs would dissociate to yield isolated H-atoms of the true $\alpha$-phase solid solution. The latter process is rather gradual, finishing close to 250 K, and indicates that some H-H interaction seems to remain even after the pair had been separated by one site. The deviation from the trigonal symmetry of the tetrahedral site is probably big enough to give the observed effects in internal friction (quasi-Snoek process in a strain field) and in resistivity (anomaly), but not sufficient to be detected in channeling experiments where the site location is determined to $\leq 0.1 \AA$.

2) Role of cold working. - Torsional deformation applied to the specimens gave internal friction spectra as shown in Fig. 4 for the metal and for the LuH$_{0.1}$ and LuD$_{0.1}$ specimens. We note the appearance in the pure Lu-specimen of two minor peaks centered near 160 K and 220 K (called "$d_1$" and "$d_2$") and a big structure named "$d$"-peak,
with a maximum at 340-350 K. The solid solutions exhibit the same "d"-peak, but instead of the "d₁₂"-peaks - they possess a peak at 250-260 K labeled "d-H"-peak, which is superposed upon the "H"-peak discussed in subsection 1. This "d-H"-peak also shows an isotope effect, the LuD₀.₁ sample having the maximum 5-10 K higher. (This observation resembles closely to the peak found in deformed hydrogenated iron.)

Fig. 5 presents the strain dependence of the various peaks for selected specimens: to the left, we show the "d" and "d₁₂" peaks for the metal and the two hydrogenated specimens of Fig. 4; to the right - the "d-H" peak of the three deuterated samples. All peaks exhibit a saturation behaviour at higher strains. In general, the LuDₓ specimens have a somewhat larger "d"-peak than the LuHₓ samples; this is not unreasonable in view of the slightly more expanded lattice of the latter - a deformation will have more effect in a denser structure. The smaller effect in the metal sample, on the other hand, is due to its "softer" lattice, with no interstitial hydrogen to strain it. All the described structures disappear or are, at least, strongly diminished after an anneal at 470 K.

These characteristics suggest that the peaks could be representative of various types of Hasiguti processes: interactions of different types of dislocations with point defects either existing (H-atoms) or generated during the deformation in the crystals. The vanishing of the "d₁₂"-peaks in the hydrogenated samples would be due to a complete blocking of some of these processes by hydrogen. The "d-H"-peak can be attributed to some relaxation of H-atoms on dislocations. Further studies of the influence of cold work on these materials are indicated.
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References.


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