limit as Eq. (1). With these assumptions the slopes dD_T/dM_T are approximately 10 times (for mass) and 5 times (for reduced mass) greater than the value in Eq. (1).

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Anomalously Small Elastic Curie Constant of Hydrogen in Tantalum

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We have measured the change in the absolute value and the change in the temperature dependence of the elastic coefficients of Ta, as caused by loading with hydrogen. From the results it must be concluded that, in contrast to heavy interstitials like O, N, or C, hydrogen has a long-range strain field with cubic symmetry in spite of tetragonal symmetry of the position.

Interstitially dissolved atoms like O, N, or C in bcc metals such as V, Nb, or Ta are examples of elastic dipoles. The orientation of these dipoles in an external strain field causes elastic relaxation or absorption and dispersion and has been called the Snoek effect.[1] In a bcc structure no interstitial site with cubic symmetry exists. Therefore for hydrogen dissolved interstitially in the same metals one expects a Snoek effect as well, provided that H causes an appreciable strain field. That this is the case can be deduced from the lattice expansion (e.g., $\Delta a/a = 5\times10^{-4}$ for 1 at.% H in Ta[2,3]) which is only about a factor of 2–3 smaller than for the heavy interstitials. For $\omega\tau \ll 1$ ($\tau = 1.2\times10^{-11}$ sec for H in Ta at room temperature[4,5]) the change of the elastic coeficients due to the Snoek effect of interstitials on the tetrahedral or octahedral sites in bcc lattices can—for small dipole density ρ and not too low temperatures—be written as[6]

$$\Delta s_{11} = -2\Delta s_{12} = \frac{2}{g}\frac{\rho(A-B)^2(s_{11}-s_{12})^2}{kT}, \qquad \Delta s_{44} = 0. \tag{1}$$

The quantity $A-B$ corresponds to the dipole moment of a magnetic dipole and is a measure for the asymmetry of the long-range strain field. The symmetric part is characterized by the combination $(A+2B)/3$, which is $\frac{1}{3}$ of the trace of the elastic dipole-moment tensor.

For the heavy interstitials in bcc metals the ratio of the asymmetric part to the symmetric part of the strain field, i.e., the ratio $3(A-B)/(A+2B)$, is about 1. Using this number as a guide, we can estimate Δs_{11} for hydrogen and its isotopes, if these interstitials behave similar to the heavy ones. At room temperature one thus would expect $\Delta s_{11}/s_{11} = 8.9\times10^{-2}$ for 1 at.% H in Ta.

We have measured the shear modulus G of Ta single crystals with [111] and [100] orientations, i.e., $G_{[111]}^{-1} = \frac{1}{3}[s_{44}+4(s_{11}-s_{12})]$, $G_{[100]}^{-1} = s_{44}$. In Fig. 1 the sample frequency $f_{[111]}^2$ ($\sim G_{[111]}$), as measured with a torsion pendulum, is plotted versus temperature for two concentrations. The H concentration was determined from the weight increase of the sample as well as from the resistivity change. Both methods gave consistent results. The upper curves in Fig. 1 are the experimental results; the two curves for the loaded samples are corrected for changes in sample dimensions due to lattice expansion. The two lower curves are calculated for the two measured concentrations assuming the above value for Δs_{11}.

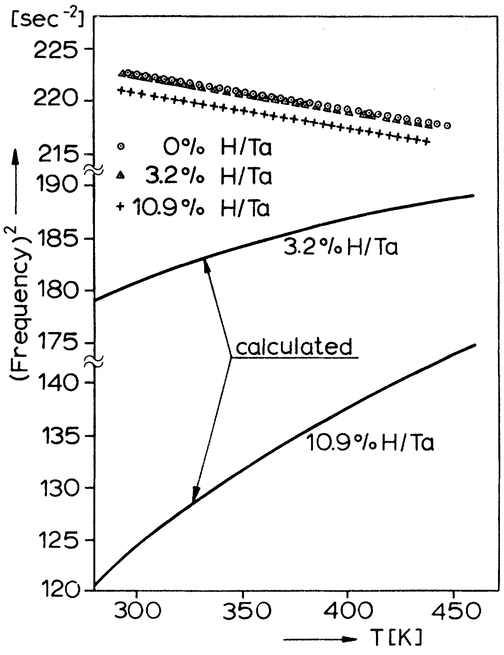

FIG. 1. Shear modulus of Ta loaded with hydrogen. Solid lines, theoretical curves. Note the interrupted scale.

The experimental accuracy was $\Delta f/f = 2\times10^{-6}$. From the change of the modulus at room temperature, we find $\Delta G_{[111]}/G_{[111]} = 5.7\times10^{-4}$ for 1% H in Ta. This value is 2 orders of magnitude smaller than expected (7.3×10^{-2}), yielding $3(A-B)/(A+2B) = 0.09$.

At a given temperature, this small value would still be explainable by assuming that the paraelastic change is accidentally compensated within better than 1% by other effects, e.g., stiffening of the elastic constants by pinning of dislocations or changes in the electronic structure. A more critical test, therefore, is the study of the change in the temperature dependence of the shear modulus. The $1/T$ temperature dependence of the paraelastic part [Eqs. (1)] should show up as an additional temperature dependence which is so strong compared to that of pure Ta $\{G^{-1}dG/dT = 1.425\times10^{-4}\ \mathrm{K}^{-1}$ for [111] orientation$\}$ that for 1% hydrogen the sign of the temperature dependence would be reversed in the temperature range investigated (see theoretical curves in Figs. 1 and 2). In contrast to this prediction the experimental curves show only a very tiny change in slope (1% per 1 at.% H) which has furthermore the wrong sign.

Again it could be argued that the temperature dependence of the Snoek effect is compensated within 2% by another, so far unknown, effect. But that would mean that a process with the same size and temperature dependence as the Snoek effect should exist, but opposite sign. Since this assumption is very unlikely we can again estimate the upper limit for the dipole moment $A-B$ by using the experimental value of the slope

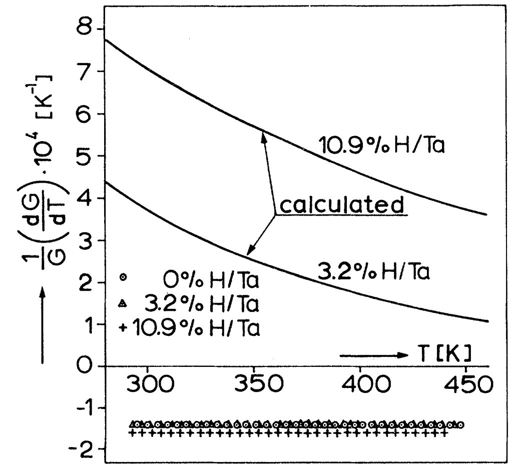

FIG. 2. Temperature coefficient of the shear modulus of Ta with [111] orientation, loaded with hydrogen. Solid lines, theoretical curves.

change, in spite of the wrong sign. Normalized to $(A+2B)/3$ we find $3(A-B)/(A+2B) \leq 0.08$. Since $A-B$ enters squared in Eq. (1), the upper value for the elastic Curie constant is again at least 2 orders of magnitude smaller than expected.

The same measurements have been performed for [100]-oriented samples and we found similar small values both for the absolute change of s_{44} and the change in the temperature dependence of s_{44}. With these results for two independent shear moduli we can exclude a measurable Snoek effect for *all* possible interstitial positions in a cubic structure. It should also be stated that preliminary experiments on Nb have given similar results for $G_{[111]}$ and $G_{[100]}$ for H as well as D.

This surprising absence of a measurable Snoek effect may be discussed under the three following aspects in which properties of H are contrasted to those of heavy interstitials:

(1) For some reason the forces exerted by hydrogen on the host metal may be just such that the strain field stays cubic in spite of a noncubic arrangement of the nearest-neighbor atoms. This can be achieved, for example, if the contribution of long-range forces, as caused by the screening electrons, is more significant than those of short-range forces in determining the dipole moment tensor. It is evidence that as the asymmetric part $A-B$ of the dipole moment tensor becomes smaller, more shells of atoms around the interstitial have to be taken into account. The interstitial site thus loses the special symmetry of the nearest neighbors. Comparing heavy interstitials with H, it is of interest to note that the residual resistivity of H in Ta[4,7,8] or Nb[7,9,10] is a factor of 6–8 smaller than that for heavy interstitials (e.g., 4–5 $\mu\Omega$ cm per 1 at.% for N or O in Nb or Ta[11]).

(2) In Ta as well as in Nb the H isotopes have extremely high jump rates in the temperature range investigated [e.g., $(1-5)\times 10^{11}$ jumps/sec for H in Ta between 300 and 450 K[5]]. The mean time of stay τ is thus comparable with the highest reciprocal frequencies in the lattice.[12] Elastic retardation effects can be expected. For a jumping particle the symmetric part of the long-range strain field will essentially be carried along, whereas for the decay of the original and full development of the new asymmetric part the particle must reside on the new site for a certain time. A very fast jumping particle will have a cubic long-range strain field.

The jumping of a H atom is *not* similar to the reorientation of a dipole with a permanent dipole moment. The dipole moment for the new position exists only after the asymmetric strain has developed. Therefore the interaction with an external strain field is retarded by a time t required for the decay of the original strain field and the buildup of the new strain field. If the mean time of stay is smaller than t, the dipole moment $A-B$ is effectively reduced and thus the number of particles in preferred sites is lowered. No estimate of t is known to us.

In order to make the jump rate as small as possible we have extended the measurements on Ta with 0.2 at.% D to 150 K, at which $\tau = 10^{-8}$ sec, so far without observing a Snoek effect.

(3) As a third point the possibility will be discussed that the position of H isotopes is smeared out. In contrast to point (2) the consequences of this effect would not depend on the jump rate. This third aspect is experimentally supported by the observation of unusually large exponents in the Debye-Waller factor.[13-16] The smearing out would, in a similar way as in point (1), reduce the significance of the local symmetry.

Besides smearing out on one particular site it is also possible that several interstitial sites are occupied simultaneously. Such a defect would have properties similar to those of paraelastic centers in alkali halides, e.g., the off-center Li in KCl.[17] A certain number and combination of interstitial sites are distinguished from those of the rest of the lattice by the fact that the energy for the proton in these sites is lowered due to local strain. So this picture is a compromise between a band model for the proton and a polaron model, in which the particle is caught in just one hole. In Ta and Nb the hydrogen occupies the tetrahedral sites[18,19] which are shown in Fig. 3. Many possible combinations of simultaneously occupied sites can be seen in Fig. 3, e.g., rings, a starlike configuration, or even a sphere. The rings with $\langle 100\rangle$ axes are combinations of two types of tetrahedral sites, whereas the rings with the $\langle 111\rangle$ axes average over all three types of tetrahedral holes. Assuming linear superposition of the strain fields this latter configuration would have a cubic long-range strain field. Such a model of simultaneously occupied tetrahedral sites would not be contradictory to the so-far published structure-analysis results.[18,19]

At present we do not know which of the three aspects discussed is the most important one. Experiments to test consequences of the different models are in progress.

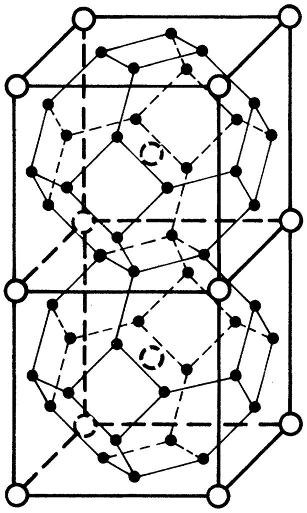

FIG. 3. Tetrahedral interstitial positions in a bcc lattice.

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Approach to Magnetic Saturation of Impurities in Iron: Effects on Nuclear Alignment, Perturbed Angular Correlation, Mössbauer, and γ-Ray Thermometry Measurements*

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Dilute alloys of Mn, Co, Ta, and Ir radioisotopes with iron were cooled to 25 mK; the impurity nuclei were oriented through applied magnetic fields up to $H=15.4$ kOe. At $H\approx 1$ kOe, sufficient to saturate the iron samples, nonalignments of nuclear hyperfine field with $\vec{H}$ of magnitude $\Theta\approx 20°$ were observed with $\sin\Theta \propto 1/H$. Errors in perturbed-angular-correlation measured g factors (10–40%), nuclear alignment parameters (10–30%), and ultralow-temperature Mössbauer and γ-ray thermometry (2 mK) due to insufficient H are discussed.

The large hyperfine fields associated with dilute impurities in ferromagnetic hosts are used in a number of research areas. However, unexplained discrepancies have been noted in nuclear g factors deduced from perturbed-angular-correlation (PAC) measurements,[1] nuclear alignment measurements,[2] in nuclear γ-ray anisotropy thermometry[3] (and its use to measure properties of