

energy to the walls. However, if, as is suggested by neutral-particle measurements,<sup>6</sup> a small proportion of the ions were heated to very high energies, the loss of these could more easily account for the lack of energy containment. The magnetic surface disruption could allow the particles to move to the walls along the magnetic field lines or, if the particle energy were preferentially transverse, as might arise from ion-cyclotron heating, the large "banana" orbits may intersect the vacuum walls. In the latter case improved energy containment might be achieved with larger toroidal currents.

The present results confirm the importance of MHD considerations in the triggering of the disruptive instability but the extremely high induced electric field and associated anomalous resistivity suggest that microinstabilities may have considerable importance for the development of the instability. The electric field is the means of dissipating into the plasma the poloidal magnetic field energy released at the disruption. This energy does not remain in the plasma but appears to be lost very rapidly.

It is a pleasure to acknowledge many fruitful discussions with M. G. Bell [who first pointed out the importance of the velocity of expansion in Eq.

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## Electrotransport of Interstitial H and D in V, Nb, and Ta as Experimental Evidence for the Direct Field Force

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Steady-state electrotransport measurements were performed for H and D interstitials in V, Nb, and Ta. The measurements were made in the temperature range between 49°C and 245°C and with hydrogen concentrations up to an atom ratio of 0.054. The results yield strong experimental evidence for the existence of a direct field force.

The forces in electrotransport are the subject of new theoretical interest; however, the occurrence of a direct field force remains controversial, even in the case of the simple jellium model of Das and Peierls<sup>3</sup> for which an exact result for the electrotransport force has been found.<sup>3-6</sup> In this paper, we present an experimental study of the electrotransport of H and D interstitials in V, Nb, and Ta. A new experimental technique was applied by use of resistance measurements under steady-state conditions. The experiments were performed in a broad temperature range and on a set of samples with different electronic

structures. The results yield straightforward experimental evidence for the direct field force, and also show how the electrotransport behavior is modified by the hydrogen-induced variations of the electronic structure.

The concentration  $c$  of the hydrogen interstitials in an electrotransport experiment under steady-state conditions is a function of the local value of the electric potential  $\varphi$ . The effective charge number  $Z^*$  is defined<sup>7</sup> by

$$eZ^* = -c(\partial\mu/\partial c)\partial \ln c / \partial\varphi, \quad (1)$$

where  $e$  denotes the (positive) elementary charge

and  $\mu$  the chemical potential of the hydrogen. In our experiments,  $Z^*$  was determined from this equation by measuring  $\partial \ln c / \partial \varphi$ . The advantage of steady-state experiments, in which transport quantities such as the diffusion coefficient do not enter the data-evaluation procedure, is demonstrated in Eq. (1).

The samples consisted of 3.5-cm-long, 0.3-cm-wide, and 130- $\mu\text{m}$ -thick foils doped with H or D. Current leads were attached to the sample ends, and five potential contacts were spot welded along the sample at distances of about 0.7 cm from each other. Thus, a sample was divided into four inner sections each confined by two potential contacts, and two smaller outer sections leading to the current leads. The potential contacts were used in order to determine the potential  $\varphi$  and the hydrogen concentration  $c$  along the sample. The hydrogen concentration in each of the four inner sample sections was determined from the resistivity increase caused by the hydrogen (the resistivity increase and the concentration are proportional for the low concentrations of this study).

Prior to the measurements, the foils [Materials Research Corp., nominal purity 99.9% (V), 99.99% (Nb), and 99.996% (Ta)] were annealed under ultrahigh-vacuum conditions (better than  $10^{-9}$  mbar) at temperatures of about 1100°C (V), 2050°C (Nb), and 2200°C (Ta) in order to remove cold-work dislocations, and in the case of Nb and Ta also for purification. The hydrogen doping was performed electrolytically (0.1%  $\text{H}_2\text{SO}_4$  in  $\text{H}_2\text{O}$  or  $\text{D}_2\text{SO}_4$  in  $\text{D}_2\text{O}$ ). The concentrations were at first estimated from the hydrogen-induced resistivity increase.<sup>8,9</sup> After the measurements were completed, the exact concentrations were determined from the amount of dissolved hydrogen with a vacuum extraction device. The concentrations (atom ratios from 0.0039 to 0.054) were low enough to ensure that all samples were in the  $\alpha$  phase within the temperature range of the experiments.<sup>10</sup> The experiments were performed in a commercial oil-filled thermostat. The resistance of hydrogen-free dummy samples was also measured in order to reduce the influence of temperature drifts. During the experiments, the hydrogen content of the samples remained unchanged due to an impermeable surface oxide layer. (Hydrogen was particularly not entering the samples during the experiments since vacuum extraction of dummy samples yielded no observable hydrogen contamination.)

Figure 1 shows results obtained from a  $\text{VD}_{0.012}$

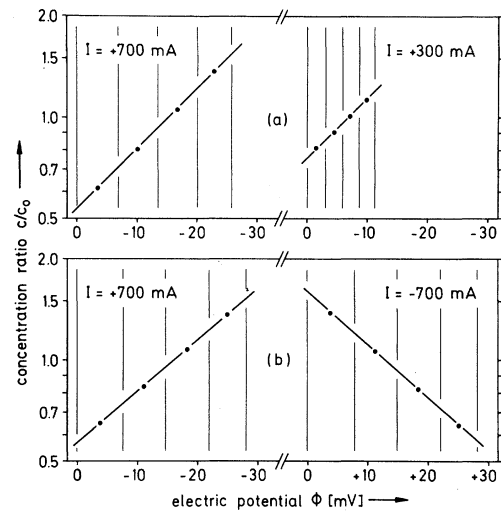


FIG. 1. Experimental results for a  $\text{VD}_{0.012}$  sample at (a) 196°C and (b) 245°C. The plots show the logarithm of the hydrogen concentration  $c$  versus the electric potential  $\varphi$ .  $c_0$  is the mean concentration of the sample and  $I$  is the electrotransport current. The experimental errors are smaller than the size of the data points.

sample. In each plot, the logarithm of the D concentration is plotted versus the electric potential  $\varphi$ . The vertical lines refer to the potential values at the potential contacts. The data points represent the mean value of the hydrogen concentration between two neighboring potential contacts.<sup>11</sup> The effective charge number was, according to Eq. (1), obtained from the slope  $\partial \ln c / \partial \varphi$ , with values for  $c \partial \mu / \partial c$  given by Veleckis and Edwards.<sup>12</sup> The deviations of  $c \partial \mu / \partial c$  from the thermal energy,  $k_B T$ , were smaller than about 20%, with one exception ( $\text{VD}_{0.054}$  sample) where they reached a maximum of about 40%. The slope  $\partial \ln c / \partial \varphi$  was confirmed to be independent of the size [Fig. 1(a)] and of the direction [Fig. 1(b)] of the applied voltage. It was seen in the experiments that the concentration differences within a sample remained small enough to ensure that the data-evaluation procedure was not impaired by concentration-dependent values of  $c \partial \mu / \partial c$  and  $Z^*$ .<sup>13</sup>

In Fig. 2, the effective charge numbers of all samples are plotted versus temperature. The results show, for each metal, a distinct dependence on temperature, hydrogen concentration, and isotope. The subsequent discussion will essentially be devoted to the temperature dependence. For all samples, independent of metal, hydrogen isotope, or concentration, the effective

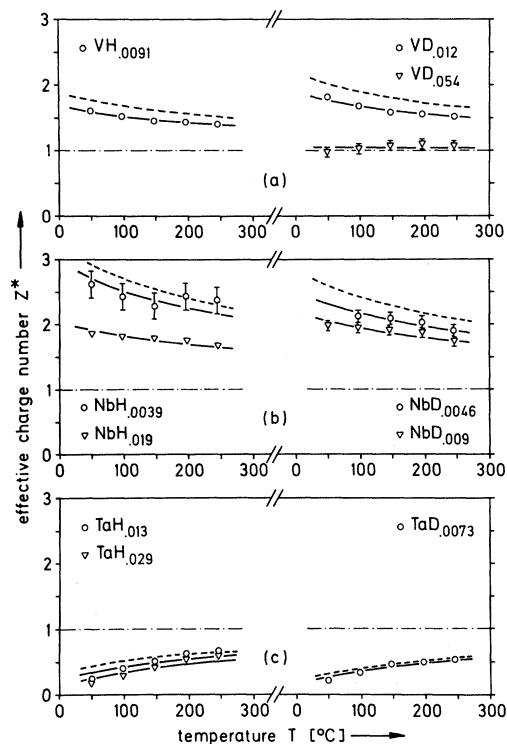


FIG. 2. The effective charge number  $Z^*$  versus temperature for different H and D concentrations in (a) V, (b) Nb, and (c) Ta. The solid and dashed lines are fits using Eq. (2) where the dashed lines correspond to  $Z^*$  values extrapolated to a H or D concentration of zero. The dash-dot-dashed lines are for  $Z^*=1$ . Experimental errors are only indicated when exceeding the size of the data points.

charge numbers at high temperatures approach a value of about +1, corresponding to the electric charge of a proton or deuteron. Such a behavior is predicted in the theories of Fiks<sup>14</sup> and of Huntington and Grone,<sup>15</sup> where the total force on the dissolved ion is split into a direct-force term (contributing +1 to  $Z^*$  in the present case), and an electron-drag term. The temperature dependence of the electron-drag term, especially for metals with a complex electronic structure, cannot be expected to be exactly inversely proportional to the resistivity  $\rho$ . In spite of this, we show in Fig. 2 least-squares fits for  $Z^*$  (solid lines) according to

$$Z^* = 1 + K/\rho, \quad (2)$$

with  $K$  as a fitted parameter. It is seen that these fits provide a fair description of the experimental results.

Previous measurements of Hérold, Marêché, and Rat<sup>16</sup> support our observation of  $Z^*$  values

approaching +1 at high temperatures. From their measurements, together with data for the diffusion coefficient,<sup>10</sup> the effective charge numbers of H and D in V, Nb, and Ta can be estimated to lie, at 600° C, in the range between 0.8 and 1.4. We note, however, that such an estimate involving diffusion-coefficient values embodies relatively large uncertainties.

The present results seem incompatible with significant and temperature-independent screening effects for the direct field force as proposed by Bosvieux and Friedel.<sup>17</sup> This is particularly striking for the Ta results which would require a drag force to increase in magnitude with increasing temperature if no direct force was assumed. It is precisely the fact that for ten samples of three different metals +1 appears to be the approximate value of  $Z^*$  at high temperatures that makes a theory without a direct-force term highly improbable (as, for example, assuming that electron and hole drag forces have very different temperature dependences). The direct force represents, in the present case, a substantial part of the total force since the drag forces due to electrons and holes partially compensate each other, and are small because of the relatively small scattering cross sections of the hydrogen interstitials. (The resistivity increase due to the hydrogen is, for instance, almost an order of magnitude smaller than that due to other interstitials.) Hydrogen interstitials are, therefore, especially suitable for studies such as the present one.

Of the previous experiments on metal-hydrogen systems, those of Oriani and Gonzalez<sup>18</sup> should also allow conclusions to be made about the existence of the direct force because of their large temperature range and the experimental accuracy they claim. The results seem to rule out  $Z^*$  values approaching +1 at high temperatures. However, we wish to point out that the  $Z^*$  values measured in these experiments are systematically too small because the electrotransport current was shunted by electrodes at the end sections of the sample thereby making the current only partially effective for hydrogen transport. A more careful estimate shows that the  $Z^*$  values of Ref. 18 might be too small by a factor of about 2,<sup>19</sup> thus providing no adequate criterium for the direct force.

We conclude with a comment on the concentration dependence of our results. In order to perceive this dependence more easily, and in order to obtain comparable data for the two hydrogen

isotopes, we show in Fig. 2  $Z^*$  values extrapolated to vanishing H or D concentrations (dashed lines).<sup>20</sup> For all metals,  $Z^*$  decreases with increasing concentration. The decrease reflects the hydrogen-induced variations of the electronic structure on the electron drag, and is caused by modifications in number and mobility of both electrons and holes. To study these effects more closely, we are presently extending the experiments to higher hydrogen concentrations.

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## Phonon Spectra of Chevrel-Phase Lead and Tin Molybdenum Sulfides: A Molecular-Crystal Model and Its Implications for Superconductivity\*

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Heat-capacity (2–400 K) and inelastic-neutron-scattering studies of the phonon spectra are reported for superconducting, high-critical-field, Chevrel-phase, lead and tin molybdenum sulfides ( $T_c \sim 11$ –15 K). Nine phonon modes per unit cell are identified whose frequencies are in the immediate region Bergmann and Rainer indicate is necessary to optimize  $T_c$ . Acoustic-phonon softening on cooling is observed. A molecular-crystal model is proposed for the lattice dynamics of these materials.

Chevrel-phase ternary molybdenum sulfides were discovered<sup>1</sup> in 1971 and soon thereafter were found to possess superconducting transition

temperatures  $T_c$  as high as  $\sim 15$  K (for  $\text{PbMo}_6\text{S}_8$ ).<sup>2</sup> They also have the highest upper critical fields  $H_{c2}(T=0)$  known: in the 500–700 kG range for