# Magnetic susceptibility of liquid heavy rare earth metals

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**Résumé.** — Nous avons mesuré pour la première fois la susceptibilité magnétique de Gd, Tb, Dy et Ho dans l'état solide à hautes températures et dans l'état liquide jusqu'à 1 700 °C. Nous n'observons qu'un faible changement aux transitions de phase et aux températures de fusion. Nous discutons les moments magnétiques dus aux électrons localisés 4f et la contribution magnétique à la résistivité électrique des terres rares lourdes liquides.

**Abstract.** — The magnetic susceptibility $\chi$ of Gd, Tb, Dy and Ho has been measured for the first time in the high temperature solid and liquid state up to 1 700 °C. There are only very small changes at the high temperature phase transitions and at the melting points. The magnetic moments due to the localized 4f electrons and the magnetic contribution to the electrical resistivity $\rho$ of liquid heavy rare earths (RE) are discussed.

The aim of this paper is to investigate the unknown magnetic properties of the heavy RE metals in the high temperature solid and liquid state [1] in order to see the variation of $\chi$ and the magnetic moments $\mu_{eff}$ across the RE series. Apart from its own right, such magnetic data have interesting consequences for the discussion of the electrical resistivity of the heavy RE at high temperatures and in the liquid state.

The electrical resistivity $\rho$ [2] increases rather monotonically across the trivalent rare earth series from La to Lu in the liquid state. This is contrary to the behavior at room temperature where $\rho$ has a maximum for Gd [3]. This maximum has been explained by the magnetic contribution to $\rho$ in terms of large spin-disorder scattering. This is due to electron scattering from the disordered 4f-spins. Calculations of this contribution based on s-f exchange interaction [4] show this term to be temperature independent and proportional to the de Gennes factor $(g_J - 1)^2 J(J + 1)$, which is largest for Gd. $J$ is the quantum number of the total angular momentum given by $J = S + L$, where $S$ is the spin quantum number and $L$ is the quantum number of the orbital angular momentum. On the other hand, $\chi$ and $\mu_{eff}$ are proportional to $g_J[J(J + 1)]^{1/2}$, which is largest for Dy. Hence, in order to understand the magnetic contribution to $\rho$ of the liquid heavy RE one has to know the magnetic properties above the melting point. Therefore, we have measured $\chi$ of Gd, Tb, Dy and Ho in the high temperature solid and liquid state.

The measurements have been performed by a novel high-temperature, high-vacuum pendulum balance working in a Faraday configuration of the magnetic field. The liquid samples have been contained in Mo crucibles. The absolute accuracy is estimated to 2 %, whereas the relative accuracy is given by 1 %. The measurements have been performed by increasing and decreasing the temperature $T$. More experimental details will be reported elsewhere [5].

Figure 1 shows the inverse of $\chi$ for Gd in the solid and liquid state. $\chi^{-1}$ follows a Curie-Weiss law. From the slopes we have determined a magnetic moment of $\mu_{eff} = 8.2\ \mu_B$ in the high temperature solid and of $\mu_{eff} = 8.6\ \mu_B$ in the liquid state. At the high temperature phase transition $T_P$ and at the melting point $T_M$ only a very small change of $\chi$ occurs. From this experimental result we draw the conclusion, that the magnetic properties of Gd do not change drastically at the melting point. Therefore we should have a very similar 4f-configuration and quantum number $J$ at room temperature and in the liquid state. Figure 2 shows $\chi^{-1}$ for Tb in the solid and liquid state. The inverse of $\chi$ follows a Curie-Weiss law. From the slopes we have determined a magnetic moment of $\mu_{eff} = 9.5\ \mu_B$ in the solid and of $\mu_{eff} = 9.0\ \mu_B$

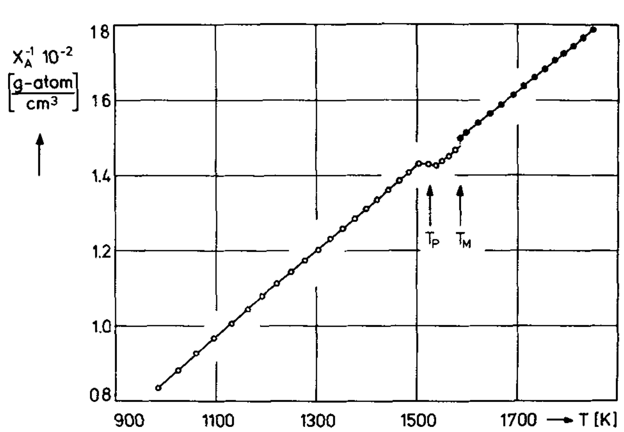


Fig. 1. — Inverse magnetic susceptibility of Gd.

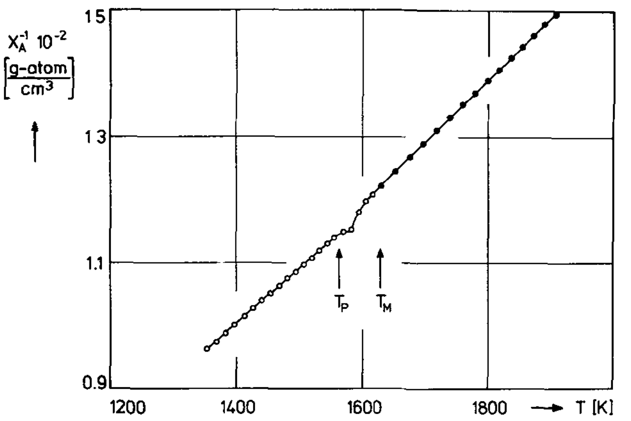


Fig 2. — Inverse magnetic susceptibility of Tb.

in the liquid state. Again the changes at $T_P$ and $T_M$ are very small. The observed data of Dy and Ho confirm the conclusions drawn above. A plot of $\chi$ and $\mu_{eff}$ in the liquid state as a function of $Z$, the number in the periodic table, gives very similar trends known for the RE in the solid state.

An overview of the experimental data is given in table I. In general the magnetic moments of the liquid state are very similar to the one known from the solid state. The magnetic moments in the solid state can be accounted for by the typical values for $S$, $L$ and $J$. For comparison the magnetic moments determined by magnetization in the magnetically ordered state and by neutron diffraction are given. They differ in the conduction electron polarization, which is not seen in neutron diffraction investigations, but is believed to account for the excessive moment values obtained for the heavy rare earth metals.

Table I

*Magnetic properties of heavy rare earths*

| | $\mu_{cal}$ | $\mu_{eff}$ $T \gtrsim T_c$ | $\mu_{eff}$ $T \lesssim T_M$ | $\mu_{eff}$ $T > T_M$ | $\mu_{mag}$ $a$ | $\mu_n$ $b$ | $g_J J$ |
|---|---|---|---|---|---|---|---|
| Gd | 7.94 | 7.98 | 8.2 | 8.6 | 7.63 $c$ | 7.0 | 7.0 |
| Tb | 9.72 | 9.77 | 9.5 | 9.0 | 9 34 | 9.0 | 9.0 |
| Dy | 10.64 | 10.64 | 10.7 | 10.5 | 10.65 | 9.5 | 10.0 |
| Ho | 10.60 | 11.2 | 10.9 | 10.6 | 10.34 | 9.9 | 10.0 |

all $\mu$ in $\mu_B$

$\mu_{cal} = g_J[J(J+1)]^{1/2}$

For general references see ref. [1], p. 132, 133.

$a$ magnetic moment from magnetization measurements

$b$ magnetic moment from neutron scattering

$c$ see ref. [6].

We do not see how significant the enhanced magnetic moment of liquid Gd is. Usually, such enhanced moments may be understood in terms of the Van Vleck theory by considering the small energy separation between the ground state and the first excited state. Thermal population of this higher level causes an admixture of the higher $J$ value and results in an enhanced $\mu_{eff}$.

Having confirmed, that the $J$ values do not change from the solid to the liquid state, the only conclusion seems to be, that the magnetic contribution to $\rho$ of the heavy RE is small in the liquid state. This has been confirmed by calculations presented in reference [7].

Financial support of the Swiss National Science Foundation is gratefully acknowledged.

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