Notas de Física - Volume XI - Nº 18

ON THE SUSCEPTIBILITY OF LIQUID GADOLINIUM *

R. M. Xavier, X. A. da Silva, W. Baltensperger **
Centro Brasileiro de Pesquisas Físicas
Rio de Janeiro, Brasil

(Received 5 February, 1965)

In the rare earth metals the ionic magnetic moments are coupled via the conduction electrons. This has often been studied for the solid state. [1, 2, 3, 4] In this work the effects of a liquid structure and of a short mean free path are evaluated. Although precise empirical data are lacking, the results clearly show that at high temperatures the short mean free path drastically decreases the paramagnetic Curie temperature, which however is larger in the liquid compared to the solid state.

The coupling between two ion spins has the form

$$I_{ij} = \left[9\pi Z^2 \Gamma^2/4\right](g-1)^2 F_\lambda(2k_m R_{ij}) J_i J_j \qquad (1)$$

where Z is the valency, $-\Gamma/2$ the exchange integral between a 4f and a conduction electron, g the Landé factor, $(g-1)J_i$ the

* Accepted for publication in Physics Letters.

** Present address: Eidgenössische Technische Hochschule - Zürich.

component of the spin directed along the angular momentum of the ion i, $^{1}k_m$ the wave number corresponding to the Fermi energy μ. For an infinite mean free path λ, $F_\infty(x) = x^{-4}$ (x cos x - sin x) is the Ruderman - Kittel function. When λ is finite, F_λ is the integral expression given by Kaplan (ref. 5, equation 19 multiplied by $(4\pi K_m^4 R^2)^{-1}$).

In solids the total interaction energy is a sum of (1) over all lattice positions. In a liquid this energy is obtained by integrating (1) over the radial distribution function. Thus in the presence of a magnetic induction B along the z-direction, the energy per ion in the paramagnetic region becomes

$$E = -g\,\mu_b\,B\langle J_z\rangle + (9\pi\,Z^2\Gamma^2/8\mu)\,(g-1)^2\langle J_z\rangle^2\,S \qquad (2)$$

where μ_b is the Bohr magneton, $\langle\ldots\rangle$ the statistical average. The structure factor is

$$S = \sum_{j\neq 0} F(2\,k_m\,R_{oj}) \qquad (3)$$

for solids, or

$$S = \int_0^\infty 4\pi r^2 \rho(r)\,F(2\,k_m\,r)\,dr \qquad (4)$$

for liquids, $\rho(r)$ being the atomic density at a distance r from an ion. The paramagnetic Curie temperature in the Weiss approximation becomes

$$T_c = -3\pi\Gamma^2\,Z^2\,S(g-1)^2J(J+1)/(4k_B\mu) \qquad (5)$$

(k_B is the Boltzmann constant).

$\rho(r)$ can be determined by x-ray or neutron diffraction. For liquid rare earths however, these data are not available. We shall construct a distribution fucntion with the assumption that two elements with the same crystal structure at high temperature have similar distribution functions in the liquid state. Thus, given $4\pi r^2 \rho_A(r)$ the radial distribution function for a liquid A, and a scale factor $\alpha = (\rho_{oA}/\rho_{oB})^{-}$ where ρ_o is the atomic density, the proposed distribution in the liquid B is $4\pi(\alpha r)^2 \alpha^{-3}\rho_A(\alpha r)$. As a test the radial distributions proposed for Cs based on the data of Li, Na K and Rb [6] agree with the empirical distribution better than 4 and 6% regarding the positions of the first and second peak, respectively, while the heights show a disagreement of 18% in the worst case. Since Gd has a b.c.c. structure at high temperatures, [7] we use the data of Cs.

The mean free path varies slowly at high temperatures. A value of $\lambda = 5$ Å has been quoted for the solid phase [4]. Kaplan's expression $F_\lambda(2 k_m R)$ has been evaluated with an electronic computer.

The structure factor S is an invariant of the scale transformation. For the b.c.c. structure and $\lambda = \infty$ the sum is known [3]. In the case $\lambda = 5$Å the sum has been evaluated to the 8^{th} nearest neighbour. For the liquid state the integrations were carried out graphically up to 8.65 Å, which is beyond the

second peak of the radial distribution. In the case $\lambda = \infty$, an integration up to infinity using a constant density was added, which gave a contribution of 10%. This could not be done for finite λ, where, however, this correction should be much smaller. The Curie temperature depends on $\Gamma^2/\mu = 0.014$ ev. This parameter was obtained from the value $\Gamma = 0.314$ ev, [2] and from the density of the high temperature solid [7].

Table 1 shows that a short mean free path strongly diminishes the interaction both in the solid and liquid state. On the other hand the liquid structure increases the interaction. It should be mentioned that several of the data assumed for the liquid state are taken from the solid, and that e.g. a shorter mean free path in the liquid state may somewhat reduce the discontinuity of T_c on melting.

* * *

Acknowledgements

The authors are indebted to Dr. E. M. Ferreira for help with the programming and for performing the computation at the Centro de Calculo, U. N. V., in Caracas. They gratefully acknowledge a grant by the Ford Foundation in support of post graduate lectures at this institute (W.B.).

λ	∞		5 A	
	S	T_c	S	T_c
Unit	10^{-4}	^oK	10^{-4}	^oK
Solid^{bcc}	-25	130	-7.8	41
Liquid	-73	390	-30	160

Table 1: Structure factors and paramagnetic Curie temperatures of Gadolinium at high temperatures calculated for infinite and finite mean free path.

References

1. P. G. de Gennes, C. R. Acad. Sci., Paris, 247, 1836 (1958).
2. J. Chevalier et W. Baltensperger, Helv. Phys. Acta 34, 859 (1961).
3. Y. A. Rocher, Thèse, Advances in Phys. (Phil. Mag. Supl.), 11, 233 (1962).
4. P. G. de Gennes, Jour. Phys. et le Radium 23, 510 (1962).
5. J. I. Kaplan, Phys. Rev. 129, 644 (1963).
6. N. S. Gingrich and L. Heaton, J. Chem. Phys. 34, 877 (1961).
7. E. M. Savitskii, V. F. Terekhova and O. P. Naumkin, Sov. Phys., Uspekhi, 6, 123 (1963).

* * *