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THERMOELECTRIC POWER OF CERIUM UP TO 6 GPa

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The thermoelectric power (TEP) of cerium has been measured up to 6 GPa. The results have been interpreted using the theories developed by Blandin et. al. and Hirst.

Cerium metal undergoes a transformation from f.c.c. (V -phase) to a 'collapsed'f.c.c. (oc -phase)[1] at 0.77 GPa. Associated with the Y-. a transformation are the discontinuities in the various physical properties such as, volume compression[2-7], electrical conductivity[8-10] thermoelectric power[11-13] specific heat 14 magnetic susceptibility etc. 14-15. The variation of these properties with pressure has been explained 16-19 in terms of the promotion under pressure of the 4f-electrons to the conduction band. The valency of cerium in the d -phase,immediately after the)'+ transition Is +3,7[19-20.] Further, o(-cerium undergoes interconfiguration fluctuation (ICF), and the valency of oC-cerium approachQs +4 as the pressure is further increased The thermoelectric power of cerium up to 6,GPa was measured with a view to Investigate further the behaviour cerium under pressure. lfhe results of these measurements are reported in this communication.

The cerium samples used in this study were 99.8;' pure from Research Organic/Inorganic Chem. Corp. USA. The samples in the form of thin strips were cleaned and sealed under vacuum in a quartz tube. Titanium sponge was used as a getter- material to remove the oxygen remaining in the sealed tube. The samples were then annealed at 400 C. The TEP as a function of pressure was measured with a tungsten carbide opposed anvil set-up. The details of technique are published elsewhere[21],

The TEP of cerium as a function of pressure is shown in Fig.l. To start with, TEP is large positive (7f-V/°O. In the y -phase it increases with the increasing pressure and reaches a maximum of 18 /1V/°K. At the -y-9c transition pressure TEP decreases sharply. In the oC -phase TEP decre-

Beyond 2 GPa, TEP exhibits a slight increase with the increase in pressure. At 4.7 cre. TEP increases rapidly; this increase is ass aTEd with the rap ly t.othisoi. eiwidth of the transition is d nearly GPa. . wn the f ,.th e slightly increases with the increase of TEP pressure. The present results are in good agreement with the earlier results up to 2 GPa obtained with a piston-cylinder apparatus 12-133 The break in the TEP versus pressure plot at the y-.a[11] transition observed by Khvostantev et. al. is not observed in the present work.

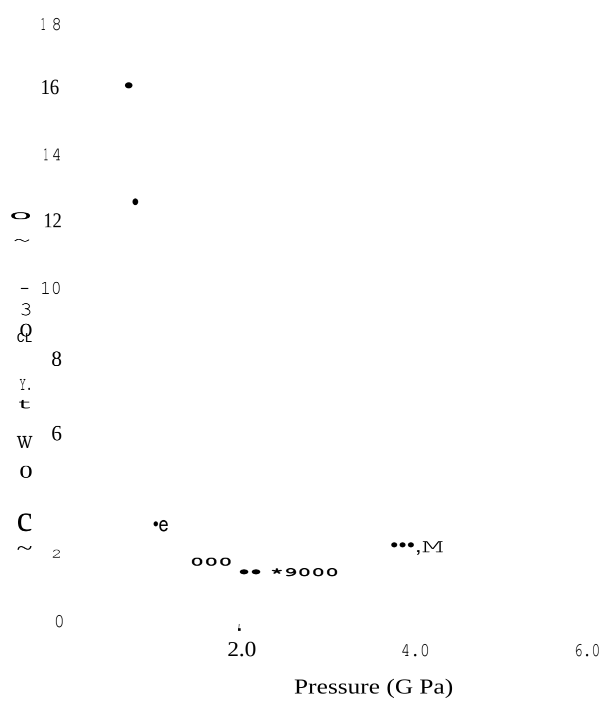

Fig.1. Thermoelectric power of cerium as a function of pressure.

In the earlier study[12] the variation of TEP with pressure was explained on the basis of a Anderson[22,23]. This model proposes six-fold degenerate 4f-level (J=5/2) which is split by the interatomic Coulomb interaction and exchange such that one state lies below and five above the Fermi level. The application of pressure reduces the gap between the lower 4f-level and the Fermi level. The proximity of the 4f-state to the Fermi level in the Y -phase introduces extra density of states[24] at the Fermi level. The contribution to the TEP from this extra density of states is given by12

$$Q = \frac{2\pi^2 k^2 T}{3e} \frac{1}{n(E)} \left[\frac{(E_F - E_f)\Delta}{(E_F - E_f)^2 + \Delta^2} \right] \qquad (1)$$

where k is Boltzman constant, T is temperature in degrees Kelvin, e is absolute electronic charge, n(E) is the density of states, Q is the width of the virtual level, and (F--E f) is the separation between the 4f-level and the Fermi level.

The TEP of cerium at one atmosphere is large positive (7 V/° K) as compared to a small negative for the neighbouring elements lanthanum and praseodymium[25]. The average of the TEP for lanthanum and praseodymium is -2/ 4 V/OK, which should be the value of TEP of cerium if it followed the normal trend. If the difference between the observed value of TEP of cerium and the expected value is attributed to the proximity of 4f-level to the Fermi level, then the contribution from Eq.(1) should 9μV/°K. On substituting 6 '^ 10-2 eV and n(E) = 1.5 states/eV/atom, it E_f - f is required to explain the TEP of cerium.-This value of (E_F-E_f) is close to 0.076 eV - a value derived from the 15.51A absorption peak[16-27] and used quite extensively in the interpretation of the high pressure behaviour of cerium. However, values as high as a few eV for (E_F-E_f) have been estimated theoretically[28]. Recently, a value of 0.04 eV/GPa **was** obtained for the pressure deritive of (E_F-E_f) from the analysis of the compression data of the V -phase[7]. If this value is used together with Eq.(1), the pressure variation of TEP in the y -phase can be explained, The theoretically predicted. variation of TEP with pressure is shown (solid line) in Fig.1.

A large negative contribution to TEP is (EF- Eted from Ei E for negative values of E_f). Thus, if f continuously crosses over EF with pressure, then the TEP will assume large negative values for small negative values of (E_F-E_f). However, experimentally only a small positive value of TEP is observed after the 'V-.a transition. This could be qualitatively explained if it is assumed that (-E)=0 continues to hold even after the pressure exceeds the transition pressure. It is interesting to recall at this stage the suggestion[19] that in the oc -cerium, the 4f virtual bound state gets locked to the Fermi level over a finite pressure range.

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