Magnetic susceptibility as a tool for studying the phenomenon of mixed valence in SmB6

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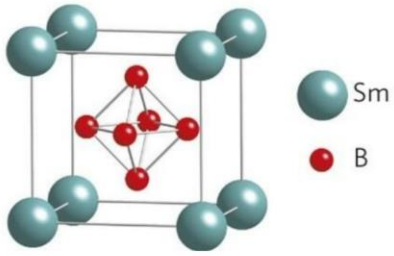
In the present work, the static magnetic susceptibility of classical intermediate-valence compound SmB6 is measured at a purely hydrostatic gaseous pressure. Based on the analysis of the obtained data within the framework of the existing model of magnetism, the initial baric derivative of the intermediate valence of Sm is determined for SmB6, which is consistent with the results of other methods.

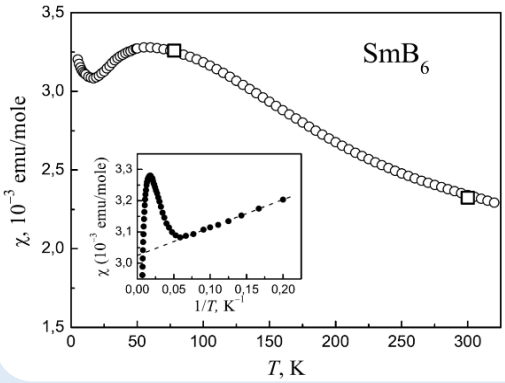
Crystal structure of SmB6 (CsCl-type)



Sm^{2+} ($4f^6$) and Sm^{3+} ($4f^5 5d^1$)

z = 2.6 (T = 300K)

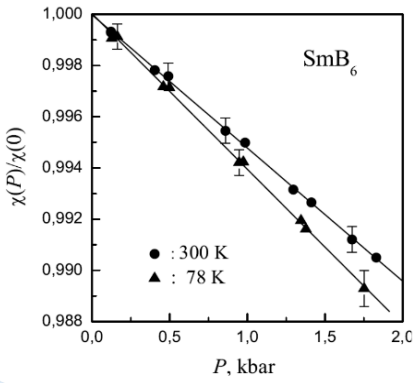
Temperature dependencies of magnetic susceptibility



The symbol (□) marks data of pendulum magnetometer at P=0.

In insert: χ vs 1/T dependence

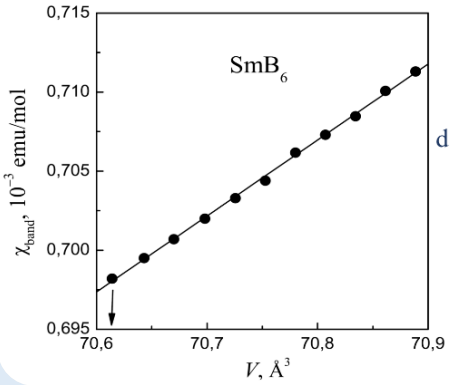
Pressure dependence of magnetic susceptibility



T (K)	χ (10^{-3} emu/mole)
78	3,26
300	2.325

T (K)	$d\ln\chi/dP$ ($Mbar^{-1}$)
78	-6.1 ± 0.5
300	-5.2 ± 0.3

Calculated band magnetic susceptibility vs unit cell volume



$\chi_{band} \simeq 0{,}7 \cdot 10^{-3}$ emu/mol ;

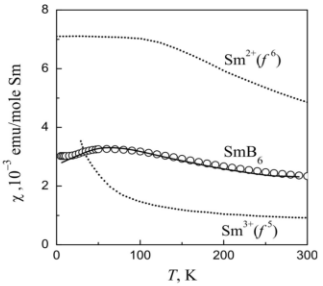
$d\ln\chi_{band}/d\ln V \simeq 4.7$ (T = 0 K),

↓

$d\ln\chi_{band}/dP \simeq -3$ $Mbar^{-1}$

(at B≃1.5 Mbar)

Temperature dependence of magnetic susceptibility of SmB6



Temperature dependence of χ of Sm2+ and Sm3+ ionic states together with experimental susceptibility of SmB6 (after subtraction of the impurity low temperature contribution) and model description (solid line).

$$\chi(T) \simeq \nu\,\chi_{Sm2+}(T) + (1-\nu)\,\chi_{Sm3+}(T),$$

$4f^6$ configuration	$4f^5 5d^1$ configuration
$^7F_0$: $L = S = 3$ and $J = 0$	$^7H_2$ ($4f^5$||): $L = 5$, $S = 3$ and $J = 2$ => $g_J = 0$
$\chi_{VV}(0) \simeq 7.1 \cdot 10^{-3}$ emu/mole	$^5H_3$ ($4f^5$#): $L = 5$, $S = 2$ and $J = 3$.

$\nu = 3 - z$, ν =0.4

Pressure effect on valence

- Considering the ionic contribution of Sm to magnetism as a dominant:

$$\frac{d\nu}{dP} = \frac{\chi(T)(1-\nu)}{\chi_{Sm^{2+}}(T) - \chi(T)} \frac{d\ln\chi(T)}{dP.} \qquad \frac{d\nu}{dP} = (-2.9 \pm 0.15)\ Mbar^{-1}$$

$$\frac{dz}{dP} = -\frac{d\nu}{dP} = (+2.9 \pm 0.15)\ Mbar^{-1}.$$

- Refinement estimate Including the contribution of the band states (χ_{band}):

$$\frac{d\nu}{dP} = \frac{\chi(1-\nu)}{\chi_{Sm^{2+}} - \chi}\left(\frac{d\ln\chi}{dP} - A\right). \quad A \approx \frac{0.5}{\chi}\frac{d\chi_{band}}{dP} \simeq -0.7\ Mbar^{-1}$$

$$\frac{dz}{dP} = -\frac{d\nu}{dP} = (+2.5 \pm 0.15)\ Mbar^{-1}$$

***Summary*:**

The observed pressure effect value demonstrates a noticeable decrease in susceptibility under pressure, which weakly depends on temperature. From model analysis of the experimental data, combined with the supplemented LSDA + U calculations of the electronic structure and van Vleck paramagnetism of the band states of SmB6, we have estimated the value of the initial pressure derivative for the intermediate valence of Sm. It was shown that the valence of Sm and its pressure dependence are closely related to the detailed characteristics of the conduction band and 4 f states.