Calculated thermopower for Ti_3SiC_2.

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Abstract

Measurements have shown that polycrystalline Ti_3SiC_2 has an almost zero thermopower over a wide temperature range. A thermopower calculation indicates that the basal component is positive, while the c axis component is negative and about twice as large.

Introduction

Ternary carbides and nitrides with hexagonal structure and the general formula $M_{N+1}AX_N$, where N=1-3, M is an early transition metal and A is an A-group element (mostly IIIA and IVA) form a class of compounds which have attracted attention for their interesting mechanical properties [1]. They are all good thermal and electrical conductors.

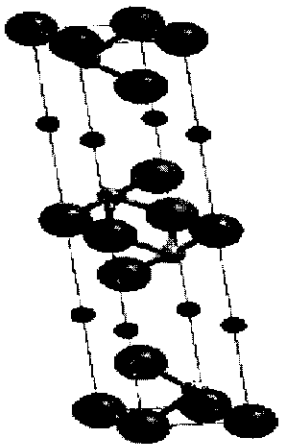

Fig.1: Ti_3SiC_2 structure

Among them, Ti_3SiC_2 (Fig 1.) has a metallic conductivity incresing linearly with temperature in the 300-850 K range and which is about half that of pure titanium [1]. Most noteworthy, polycrystalline Ti_3SiC_2 has a thermopower that is essentially zero over at least the same temperature range [2].

We present here a thermopower calculation starting from the band structure of the material, which may explain this peculiar behaviour.

Thermopower calculation

The thermopower S_u has been calculated using Boltzmann transport theory, with the relaxation time approximation valid at high enough temperature for phonon scattering

$$S_u = \frac{L_1^{-1} L_{12}}{T}$$

where u stands for the basal or the c axis direction and the Onsager coefficients are given by

$$L_{11} = \int dE \left\{ \frac{\partial f_0}{\partial \mu} \sigma_u(E) \right\}$$

$$L_{12} = L_{21} = -\frac{1}{e} \int dE \left\{ \frac{\partial f_0}{\partial \mu} (E-\mu) \sigma_u(E) \right\}$$

here $\sigma_u(E)$ is the transport distribution

$$\sigma_u(E) = \frac{e^2 \tau}{(2\pi)^3} \int_{Ek=E} \frac{dS}{|\nabla E_k|} v_u v_u$$

(v_u beeing the electron velocity along u).

The relaxation time has been choosen to be energy dependant $\tau(\varepsilon) \sim \frac{1}{g(E)}$. This Mott approximation would be exact for a constant scattering matrix element. So the only anisotropy considered in the calculation comes from the band structure and not from the scattering process.

The calculation of the transport fonction then proceeds as explained in [3]. The band structure calculation used to obtain the velocities has been performed with the LAPW Wien2k program [4].

Anisotropy and thermopower

The results of the calculation is shown in Fig.2. The basal component of S is negative and about twice as large as the c-axis component, which is positive. Moreover both components have only a weak variation with temperature, so that they nearly compensate for a polycrystal on a large temperature range. For comparison we also show the experimental results of Yoo et al [2].

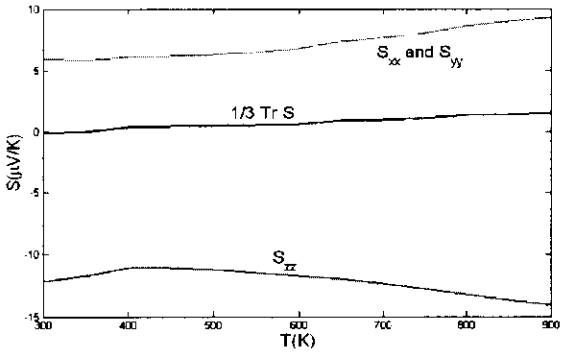

Fig2: Calculated Ti_3SiC_2 thermopower

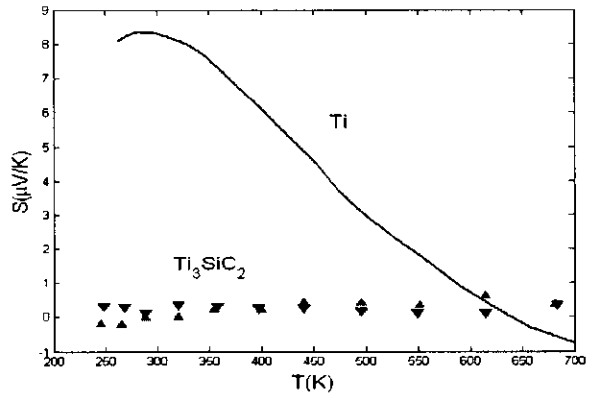

Fig3 : Experimental Ti_3SiC_2 polycrystal thermopower from Ref. [2]

Conclusions

In spite of its shortcomings (oversimplified treatment of the scattering matrix element) the present calculation suggests that the surprising near zero thermopower of polycrystalline Ti_3SiC_2 is due to a compensation between the c-axis and basal components. This anisotropy comes from the band-structure and can even be related to the shape of the Fermi surface parts which dominate the transport distribution fonction [5].

References

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