

Effect of pressure on magnetic properties of $\text{ThFe}_{11}\text{C}_x$

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The $\text{ThFe}_{11}\text{C}_x$ carbides crystallize in the tetragonal BaCd_{11} type structure (I4/amd space group) [1]. Carbon atoms occupy pseudo-octahedral sites with 4 iron atoms and two thorium atoms at the corners. Iron atoms form three sublattices. Direct bonds exist between the carbon and iron atoms.

It turns out that the carbon concentration has a large effect on all magnetic properties of the $\text{ThFe}_{11}\text{C}_x$ compounds. The Curie temperature and the Fe–Fe exchange interactions are found to increase significantly upon insertion of carbon into the crystal lattice. On the contrary, the saturation magnetization and the paramagnetic effective magnetic moment are found to decrease when the carbon concentration increases [2]. The hybridization between the C and Fe states in $\text{ThFe}_{11}\text{C}_x$ seems to be very similar to that in binary Fe_3C compound, leading to similar changes of the magnetic properties. In particular the 3d magnetism is significantly more delocalized when compared to $\alpha\text{-Fe}$ [1].

Also the magnetocrystalline anisotropy is very sensitive to the carbon concentration. The spin reorientation of the first-order type from planar to uniaxial anisotropy has been demonstrated to take place below room temperature. It is supposed that it results from a competition between the Fe sublattices anisotropy terms [1].

To get a better insight into the peculiar magnetic behavior of the $\text{ThFe}_{11}\text{C}_x$ compounds, namely the volume dependence of the magnetization and spin reorientation transition temperature T_{SR} we performed high pressure magnetization studies in the pressure range up to 10 kbar. The magnetization measurements were performed in SQUID magnetometer using a

CuBe pressure cell in temperature range 5–300 K and at magnetic fields up to 5 T. The studies were performed on samples $\text{ThFe}_{11}\text{C}_{1.5}$ and $\text{ThFe}_{11}\text{C}_{1.8}$, the same as in the work [2].

The ambient pressure magnetization and T_{SR} values are in good agreement with the results of [2]. The magnetization of both samples decreases with pressure in the whole temperature range. The magnetic isotherms obtained for 5 K and 300 K for different pressures on $\text{ThFe}_{11}\text{C}_{1.5}$ are presented on Fig. 1.

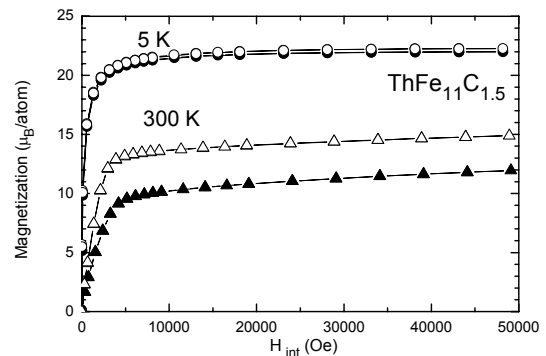


Fig. 1. Magnetic isotherms measured on $\text{ThFe}_{11}\text{C}_{1.5}$ at 0 kbar (empty symbols) and 7 kbar (full symbols) for 5 K and 300 K

The decrease of magnetization with pressure at 5 K is quite low on both samples, the value obtained on $\text{ThFe}_{11}\text{C}_{1.5}$ ($d\ln M_s/dp = -1.3 \times 10^{-3} \text{ kbar}^{-1}$) is higher than that obtained on $\text{ThFe}_{11}\text{C}_{1.8}$ ($d\ln M_s/dp = -0.3 \times 10^{-3} \text{ kbar}^{-1}$). Significantly larger decrease of magnetization was observed on both samples at room temperature. More than six times higher value of $d\ln M_s/dp$ was observed on $\text{ThFe}_{11}\text{C}_{1.5}$ ($d\ln M_s/dp = -25 \times 10^{-3} \text{ kbar}^{-1}$) than on $\text{ThFe}_{11}\text{C}_{1.8}$ ($d\ln M_s/dp = -3.8 \times 10^{-3} \text{ kbar}^{-1}$).

The different pressure effects on magnetization at room temperature indicate also different pressure behavior of T_{C} on

these two samples. Using the Kouvel relation we can estimate the pressure effect on T_C , $dT_C/dp \sim -6$ K/kbar for $\text{ThFe}_{11}\text{C}_{1.5}$ and $dT_C/dp \sim -3.5$ K/kbar for $\text{ThFe}_{11}\text{C}_{1.8}$.

The relation between increase of volume of the lattice cell (2.5 %) and increase of T_C (98 K) [1,2] caused by the increase of carbon content from $\text{C}_{1.5}$ to $\text{C}_{1.8}$ – is quite well comparable with the estimated values of pressure effects on T_C (decrease of volume of 1 % will cause the decrease of T_C by 35 – 60 K).

The observed pressure effects on both magnetization and T_C agree with conclusions derived in [1] that 3d magnetism of $\text{ThFe}_{11}\text{C}_x$ is significantly more delocalized when compared to α -Fe. On the other hand we have to mention that the studied compounds cannot be considered as a typical itinerant ferromagnet where the relation $d \ln M_S/dp \sim d \ln T_C/dp$ has to be valid. Higher values of pressure effect on T_C than that on M_S are similar to the behavior of $\text{Fe}_{72}\text{Pt}_{28}$ invar alloys.

The pressure effect on spin reorientation transition temperature measured at low magnetic field of 100 Oe was found to be surprisingly large, as it illustrated on Fig. 2 for $\text{ThFe}_{11}\text{C}_{1.8}$.

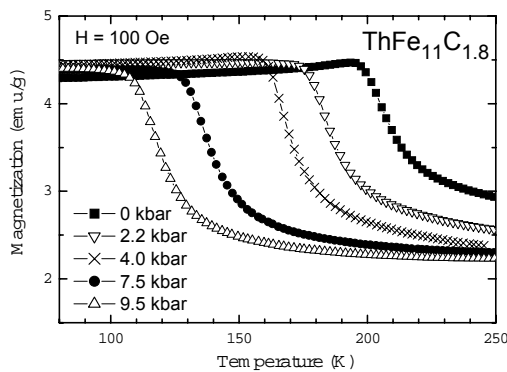


Fig. 2. Temperature dependence of low field magnetization measured at 100 Oe in the vicinity of spin reorientation temperature for different pressures in $\text{ThFe}_{11}\text{C}_{1.8}$.

Quite comparable values of dT_{SR}/dp were observed for the two compounds - $dT_{SR}/dp = -8.0$ K/kbar and -9.5 K/kbar for $\text{ThFe}_{11}\text{C}_{1.5}$ and $\text{ThFe}_{11}\text{C}_{1.8}$ respectively. The complex behavior of magnetic anisotropy as a

function of volume are visualized on Fig. 3, where the changes of T_{SR} connected with the evolution of volume obtained by changes of carbon concentration and by high pressures are presented.

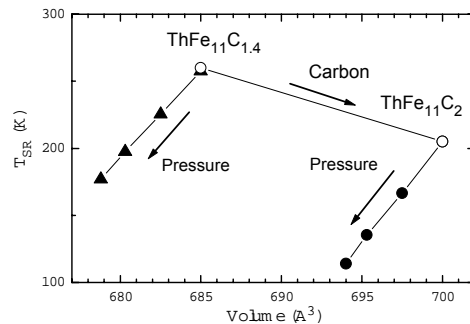


Fig. 3. The changes of T_{SR} connected with the evolution of volume obtained by changes of carbon concentration and by high pressure

To explain such behavior is not simple. Assuming that there is no anisotropy contribution from the Th ions we conclude that the carbon insertion plays a significant role in the different terms of anisotropy on the iron sites. According to Mössbauer spectroscopic measurements [3], the iron hyperfine fields range from 397 kG for the 4b sites to 385 kG for the 8d sites and 311 kG for the 32i sites. This indicates a large difference from one crystal site to the others and has been explained by differences in the local environment of the iron atoms. One can speculate that the changes of local environment connected with insertion of carbon atoms and creation of the Fe – C bonds on certain crystallographic positions can affect the crystal field parameters and can be substantially different than the effect of high pressures, where the crystal lattice is only deformed.

Acknowledgements

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