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To cite this version:

L.V.B. Diop, Z. Arnold, J. Kamarád, O. Isnard. Pressure dependence of the magnetic properties and phase diagram of HoCo12B6 ferrimagnetic compound. Journal of Magnetism and Magnetic Materials, 2019, 476, pp.106-110. 10.1016/j.jmmm.2018.12.042 . hal-02380614

HAL Id: hal-02380614

https://hal.science/hal-02380614v1

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Pressure dependence of the magnetic properties and phase diagram of $HoCo_{12}B_6$ ferrimagnetic compound

L. V. B. Diop[1†], Z. Arnold[2], J. Kamarád[2] and O Isnard[1]

[1]*Univ. Grenoble Alpes, CNRS, Institut Néel, 38000 Grenoble, France*

[2]*Institute of Physics AS CR v.v.i., Na Slovance 2, 182 21, Prague 8, Czech Republic*

ABSTRACT

The magnetic properties of $HoCo_{12}B_6$ ferrimagnetic compound are investigated versus the magnetic field, temperature and pressure. The magnetic phase diagram exhibits three remarkable temperatures: the ordering temperature T_C, the spin reorientation temperature T_{SR} and the compensation temperature T_{COMP} at 147 K, 76 K and 47 K respectively. In $HoCo_{12}B_6$ intermetallic system the spontaneous magnetization resulting from the antiparallel coupling of the magnetic moments of Ho and Co sublattices is pressure insensitive at lowest temperatures whereas it decreases with pressure at temperature range above T_{SR}. The mean Co magnetic moment is small (< $0.5\mu_B$) in the studied compound and exhibits a weak pressure dependence. In addition, significant pressure effects have been observed on both the spin reorientation transition and the Curie temperatures. A value of dT_C/dp = -3.5±0.4 K GPa^{-1} is dominated by the Co sublattice contribution. The spin reorientation transition temperature is also found to decrease versus pressure dT_{SR}/dp = -4.2±0.4 K GPa^{-1}. The compensation temperature where the Ho and Co sublattices have the same magnitude but opposite direction does not vary significantly with applied pressure in the investigated pressure range (up to 1GPa). The results of pressure dependence of the magnetic properties are compared to earlier results on other R-Co-B borides. $HoCo_{12}B_6$ compound confirmed the remarkably high volume stability of the magnetic properties of the $RCo_{12}B_6$ compounds with the itinerant character of magnetism and low values of both the T_C and magnetization.

[†] Present adress : Institut für Materialwissenschaft, Technische Universität Darmstadt, D-64287 Darmstadt, Germany

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1. Introduction

Intermetallic compounds based on rare-earth (R) and transition metals (T) form an important class of materials with various applications such as permanent magnets, magnetostrictive sensors and magneto-optical devices. In addition substitution or addition of other, namely light elements (H, B), can be used to improve their functional properties. Some important intermetallic compounds present a compensation point at which the R and the T sublattices have the same magnitude of magnetization but they are coupled antiparallel. This makes such magnetic systems interesting for the magneto-transport effects in spintronics [1] and has triggered a renewed interest in such ferrimagnetic materials. R–T alloys with heavy rare-earth elements are also investigated by means of optical switching experiments [2].

Very interesting magnetic properties have been recently reported for the $RT_{12}B_6$ antiferromagnetic and ferrimagnetic compound (R = rare-earth, Y and T = Co, Fe) [3-8, 24]. The $RT_{12}B_6$ compounds crystallize in the rhombohedral $SrNi_{12}B_6$-type structure, space group $R\bar{3}m$ [9,10]. In this structure, rare-earth element occupies 3a crystal site. Transition metal atoms are located on two inequivalent positions 18g and 18h. The boron atoms are localized on another 18h site. Earlier studies showed that the $RCo_{12}B_6$ compounds are stable for the majority of rare-earths. Their low Curie temperatures and low Co magnetic moment in comparison with metallic cobalt (T_C = 150 K and μ_{Co} = 0.42 μ_B/Co atom for $YCo_{12}B_6$) [11] are attributed to hybridization of B(p) and Co(d) electrons bands as a consequence of small interatomic distances between Co and B atoms (2.05 Å). Nuclear magnetic resonance (NMR) studies on $RCo_{12}B_6$ compounds showed significantly different magnetic moments on the two Co crystallographic sites with a lower value of the Co magnetic moment on 18g [12].

The effect of volume changes on intrinsic magnetic properties of several $RCo_{12}B_6$ compounds ($YCo_{12}B_6$, $LaCo_{12}B_6$, $CeCo_{12}B_6$ and $GdCo_{12}B_6$) has been investigated by studies under hydrostatic pressure up to 1 GPa [6,7,8,11]. Such pressure corresponds to the decrease of volume around 1%. Only moderate pressure effects have been observed on both magnetization and Curie temperature for all studied compounds. The Curie temperature decreases with pressure for all studied compounds. Moderate decrease of T_C is comparable for all of them and can be attributed mostly to the volume dependence of both, the Co magnetic moment and the exchange interactions. The saturation magnetization of $YCo_{12}B_6$, $LaCo_{12}B_6$ and $CeCo_{12}B_6$ compounds decreases with pressure in the whole temperature range below T_C. As regards $CeCo_{12}B_6$ [7,8,11], this effect can be ascribed to the increasing delocalization of the 4f-electrons of Ce and consequently to a following shift of Ce toward

the tetravalent state under pressure. The situation is different for $GdCo_{12}B_6$ where, due to the antiparallel coupling of the Gd and Co sublattices and to the negligible pressure effect on localized Gd magnetic moment, the observed increase of magnetization with pressure at temperatures below compensation temperature is attributed predominately to the pressure induced decrease of Co magnetic moments [7,8]. The estimated decrease of μ_{Co} is also quite comparable with pressure induced decrease of M_S in $YCo_{12}B_6$. The decrease of μ_{Co} is also responsible for the observed increase of compensation temperature with pressure. In general, the $RCo_{12}B_6$ compounds studied up to now under pressure exhibit remarkably higher volume stability of the magnetic properties than the RCo_4B compounds, where more than two times higher values of dT_C/dp and $dlnM_S/dp$ were observed [13]. The relative volume stability of magnetic properties of $RCo_{12}B_6$ compounds with the itinerant character of magnetism and low values of both the T_C and magnetization can be attributed to the remarkable role of the hybridization as a consequence of small distances between Co and B atoms. Similarly the volume stability of the highly reduced μ_{Co} was recently observed for tetravalent Th state in $ThCo_4B$ compound [14].

$HoCo_{12}B_6$ compound (space group $R\bar{3}m$, a = 9.451 Å, c = 7.454 Å) exhibits antiparallel coupling of the magnetic moments of Ho and Co sublattices. Studies of its magnetic phase diagram showed that this compound is characterized not only by the magnetic ordering temperature (T_C = 147 K) and compensation temperature (T_{COMP} = 47 K) but also by spin reorientation temperature (T_{SR} = 76 K). This behavior is a consequence of remarkable magnetic anisotropy of Ho sublattice. Recently, using the combination of magnetization measurements, ac susceptibility and neutron diffraction the magnetic phase diagram of $HoCo_{12}B_6$ has been determined [15]. These extended studies showed that $HoCo_{12}B_6$ compound orders its magnetic moments close to the crystallographic *c*-axis at 1.5 K and they undergo a spin reorientation from *c*-axis to basal plane at T_{SR} = 76 K. An incommensurate magnetic structure with a long incommensurate period along the *c*-axis has been found above T_{SR}. This configuration preserved up to the Curie temperature. The Rietveld refinement analysis of neutron diffractions revealed that Ho atoms carry a magnetic moment of 8.9 μ_B at 1.5 K. Co moments are coupled antiparallel to the Ho one. At the temperature of 1.5 K, the Co in 18 h position exhibits a moment of 0.63 μ_B whereas moment of the Co in 18 g site exhibits a value as low as 0.14 μ_B.

The high pressure studies of $RCo_{12}B_6$ compounds have been performed up to now on compounds with nonmagnetic R (Y, La, Ce) or R with negligible magnetocrystalline anisotropy (Gd) [6,7,8,11]. Holmium has magnetic moment comparable with Gd but at the

same time it exhibits remarkable magnetocrystalline anisotropy. Consequence of this property is the existence of spin reorientation transition in $HoCo_{12}B_6$ compound [15,16]. Motivation for our high pressure studies of $HoCo_{12}B_6$ compound was to get information not only about volume stability of Co magnetism and the effect of Ho sublattice on the itinerant character of Co magnetism but also about the effect of volume change on anisotropy of the $HoCo_{12}B_6$ compound and about the role of anisotropy of Ho – sublattice on character of magnetism and exchange interactions. To reach this goal we have studied several magnetic quantities – compensation temperature, spin reorientation temperature, Curie temperature and magnetization under hydrostatic pressure.

2. Experimental methods

A polycrystalline $HoCo_{12}B_6$ ingot sample was synthesized by melting high-purity starting elements (Ho - 99.9%, Co - 99.95%, B - 99.9%) in a high frequency induction furnace under a purified argon gas atmosphere. To promote homogeneity, the ingot was remelted five times, then pieces of the obtained sample were wrapped in a Ta foil sealed in an evacuated quartz tube and annealed at 900 °C for three weeks. The crystallographic (phase) purity was checked by standard x-ray diffractometry. A careful look at the diffraction pattern indicates the presence of residual traces of the binary Co_2B compound as minority impurity phase. The specific heat measurements have been carried out on a commercial PPMS instrument (Quantum Design). The magnetization measurements at high hydrostatic pressure up to 1 GPa were performed in a SQUID magnetometer (Quantum Design Co.) using a home-made miniature piston-cylinder CuBe pressure cell with a mixture of mineral oils as a pressure transmitting medium. The pressure was determined at low temperatures using the known pressure dependence of the critical temperature of the superconducting state of the pure Pb (5N) sample [17]. For comparison, ambient pressure magnetic measurements were performed on the above described set-up as well as using the extraction method.

The evolution of the Curie temperature under different pressures was determined from temperature dependence of the d.c. susceptibility measured in magnetic field of 0.1 T at a heating rate of 1 K per minute. T_C was defined as the inflexion point of low field magnetization vs. temperature curve. The high pressure studies were performed at magnetic fields up to 7 T in the temperature range from 5 K to 200 K. The spontaneous magnetization $M_S(T,P)$ at different temperatures and pressures was determined by an interpolation of the isothermal magnetization curves to zero internal field. The magnetization data were corrected for the presence of the minor ferromagnetic Co_2B impurity in order to get the intrinsic

magnetization values of $HoCo_{12}B_6$. The measurements were realized just above the ordering temperature of $HoCo_{12}B_6$ in order to remain far below the Curie point of Co_2B, which is 429 K. The Co_2B impurity was consequently considered as carrying a saturated magnetic moment simplifying the correction for its ferromagnetic contribution.

3. Results and discussion

The magnetic ordering temperature of $HoCo_{12}B_6$, T_C = 147 K, at ambient pressure is clearly seen and confirmed by the anomaly observed at the specific heat data presented on Figure 1. This ordering temperature decreases significantly upon applying pressure as can be seen on Figure 2 recorded at 0.1 T. A pronounced peak appears just below the Curie temperature that recalls the large Hopkinson effect reported earlier for isostructural compounds like $YCo_{12}B_6$. Interesting transport properties have been reported to occur around such Hopkinson effect [18,19]. Temperature dependence of isofield magnetization curve (μ_0H = 2 T) recorded for $HoCo_{12}B_6$ compound at different pressures is presented on Figure 3. The minimum of the magnetization observed at about 47 K corresponds to the compensation temperature.

The pressure dependence of the ordering temperature T_C, as derived from isofield thermomagnetic measurements at field of 0.1 T, is illustrated in Figure 4. The pressure induced decrease of T_C is linear in the used pressure range and a sensitivity of dT_C/dp = -3.5±0.4 K GPa^{-1} is obtained. This value is very close to that reported for $YCo_{12}B_6$ (dT_C/dp = -3.9±0.2 K GPa^{-1}) but slightly smaller than that reported for $LaCo_{12}B_6$ (dT_C/dp = -4.7±0.2 K GPa^{-1}), a compound having much larger unit cell too. The unit cell volume difference most probably contributes to the observed difference of pressure evolution of the ordering temperature T_C since the magnetization of the $RCo_{12}B_6$ compounds is found to be sensitive to the dlnM/dlnV = 1.8 ±0.1 [20]. The above results prove the dominant role of the Co magnetic sublattice on the magnetic ordering temperature and its pressure dependence. Consequently the decrease of T_C versus pressure can be attributed mostly to the effect of volume change on the Co-Co exchange interactions.

As shown by the minimum value of the magnetization at about 47 K a compensation temperature T_{COMP} is observed at ambient pressure in good agreement with earlier reports [21]. This indicates that at T_{COMP} the two opposite magnetic sublattices (the R and the Co ones) have the same magnitude and cancel out. As can be seen in Figure 2 and in Figure 3 the compensation temperature T_{COMP} does not evolve with pressure and can be considered as

pressure insensitive. This is significantly different pressure behaviour of T_{COMP} in comparison with that of the $GdCo_{12}B_6$ compound where, T_{COMP} increases with increasing pressure due to an increase of magnetization below T_{COMP} and a decrease of one above this temperature. The investigation of the magnetization curves recorded at different pressures reveals that the spontaneous magnetization M_S of $HoCo_{12}B_6$ at 5 K is pressure insensitive within the experimental error. The situation is more complex at higher temperatures (typically above T_{SR}) where the pressure slope of M_S is becoming negative, reaching the value of dln M_S/dp = -0.041 GPa^{-1} at 130 K. The very negligible pressure sensitivity of the low temperature magnetization indicates that the difference between the Ho and Co sublattice magnetization does not change upon applying pressure. Since the Ho magnetic moment is of localised type being carried by the 4f electronic shells, we can expect low pressure influence on these orbitals the Ho moment being preserved at low temperature. Consequently, the zero Kelvin magnetization of the Co sublattice magnetization is deduced to be also very little sensitive to the applied pressure in the investigated pressure range (below 1 GPa). This shows a remarkably large stability of the low Co magnetic moment in the $HoCo_{12}B_6$. The significant magnetization decrease observed upon applying pressure between T_{SR} and T_C results from the dominant Co sublattice magnetization, but keeping in mind that in this temperature range an incommensurate magnetic structure has been derived from neutron diffraction [15], one can expect this magnetic structure to evolve versus pressure too.

The magnetization isotherms of $HoCo_{12}B_6$ compound recorded at different pressures and different temperatures from 5 K up to 70 K are presented in Figure 5. As can be seen from this Figure 5, the isothermal magnetization of $HoCo_{12}B_6$ compound is featured by an unusually large susceptibility at high magnetic field. This behaviour is opposite to the easy magnetization saturation observed previously at 5 K for other ferrimagnetic like $GdCo_{12}B_6$ compound [8]. We interpret this as resulting from a stronger Gd-Co exchange interaction and an overall smaller anisotropy as expected from the spherical 4f electronic shell of Gd orbital moment [21]. According to the estimation of the R-Co exchange interaction in $RCo_{12}B_6$ compound a reduction of about 20 to 25% is derived from R = Gd to R = Ho.

The magnetization isotherms of $HoCo_{12}B_6$ compound recorded up to 7 T at temperatures 90 K and 130 K under different pressures are given in Figure 6. According to earlier reported neutron diffraction data at low temperature 1.5 K, both the Co 18g and the Co 18h sites are contributing to the magnetization. The Co 18g exhibits a much smaller magnetic moment of 0.14 μ_B against 0.63 μ_B for the Co 18h. On the contrary, at about 78 K and above, the Co18g position moment has been shown to vanish and these sites no longer carry a

significant magnetic moment [15]. This very different magnetic behaviour of the two inequivalent Co positions arises from their different atomic local environment. Indeed the 18g position has only 7 Co neighbours but 4 B atoms and twice more R neighbours than the 18h position (9 Co, 3 B and only one R neighbours). It is worth pointing out that in the temperature range where (namely around 70 K and above) Co 18h atoms are carrying all the magnetization that remains on the Co magnetic sublattice. We can consequently assign the pressure dependence of spontaneous magnetization to the dominant behaviour of Co at 18h position. The change of parameter dM_S/dp with increasing temperature is illustrated on Figure 7.

Similarly to what has been observed for the ordering temperature, the spin reorientation transition is also found to decrease upon applying pressure. Figure 2 plotting temperature dependence of low field magnetization (μ_0H = 0.1 T) at different pressures indicates a linear reduction of T_{SR} under pressure with a moderate slopes dT_{SR}/dp = -4.2±0.4 K GPa^{-1}. This indicates that the balance between the Co and Ho anisotropy evolves with pressure. The higher decrease of the T_{SR} comparison to T_C indicates a complex effect of volume change on the anisotropy behaviour of both sublattices. The main influence of pressure is to reduce the axial anisotropy temperature domain where the Ho sublattice prevails. At present, the mechanism leading to the observed reduction of T_{SR} is not completely clear. It is the Co magnetic anisotropy reinforced upon applying pressure or it is the reduced Ho sublattice anisotropy. However the above discussed pressure induced reduction of the Co exchange interactions should play a role.

It is also remarkable that the moderate sensitivity of the T_{SR} observed here for $HoCo_{12}B_6$ compound is in opposition to the extreme sensitivity of the ordering direction to Fe content in $GdCo_{12-x}Fe_xB_6$ compounds as reported recently by Diop *et al* [22] and Lee-Hone *et al* [23]. This can be taken as an indicator of the large influence of the electronic effect of the Fe for Co substitution in the $GdCo_{12-x}Fe_xB_6$ series of compounds.

The pressure effect on T_C and M_S can be attributed mostly to the volume dependence of both the Co magnetic moment and the exchange interactions. Indeed a comparison with non magnetic rare-earth containing $RCo_{12}B_6$ compounds shows that the pressure dependence of the magnetization is of the same order of magnitude for $YCo_{12}B_6$ and $LaCo_{12}B_6$ than for $HoCo_{12}B_6$ [6,7,8,11].

The results obtained on $HoCo_{12}B_6$ compound confirmed the remarkably high volume stability of the magnetic properties of the $RCo_{12}B_6$ compounds with the itinerant character of magnetism and low values of both the T_C and magnetization. It can be attributed to the

remarkable role of the hybridization of B(p) and Co(d) electrons bands as a consequence of small interatomic distances between Co and B atoms and the consequently low Co magnetic moment magnitude.

4. CONCLUSION

The magnetic phase diagram of $HoCo_{12}B_6$, presented in Figure 8, exhibits three remarkable temperatures: the ordering temperature at 147 K corresponding to the para to ferromagnetic transition upon cooling, the spin reorientation temperature and the compensation temperature where the two sublattice magnetization cancel out. The spin reorientation observed on $HoCo_{12}B_6$ compound is confirming earlier neutron powder diffraction results as well as previously reported ^{57}Fe Mössbauer data on Fe doped sample. The spin reorientation from *c*-axis to basal-plane occurring at T_{SR} = 76 K is found to be pressure sensitive to an external applied pressure. The Curie temperature is also pressure sensitive. On the contrary the compensation temperature is found to be mostly insensitive to pressure and kept a value of about 47 K.

The observed pressure behavior of the magnetic properties of $HoCo_{12}B_6$ confirmed itinerant character of Co magnetism and the remarkable role of the hybridization as consequence of small distances between Co and B atoms. The T_C reduction versus pressure can be attributed to both the combine effect of decreases of exchange interactions and Co magnetic moment.

Acknowledgements

The authors gratefully acknowledge the CNRS-ASCR French-Czech collaboration program.

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Figure captions:

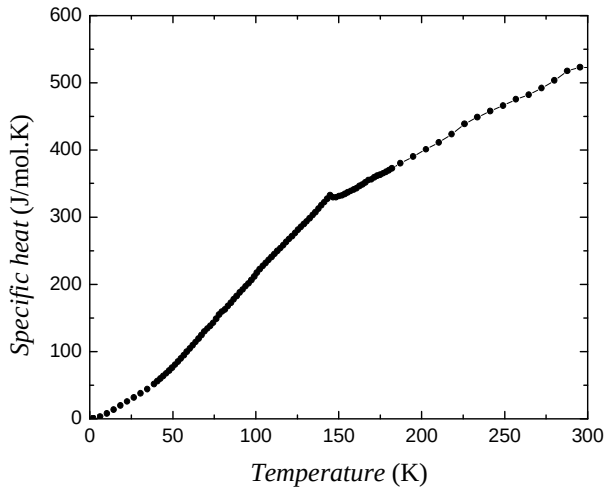

Figure 1: Temperature dependence of specific heat of $HoCo_{12}B_6$ with an anomaly at $T_{C.}$

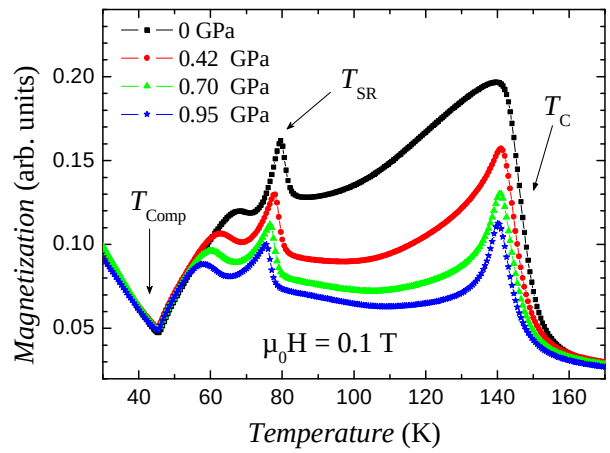

Figure 2: Temperature dependence of low field magnetization (μ_0H = 0.1 T) of the polycrystalline $HoCo_{12}B_6$ compound at different pressures.

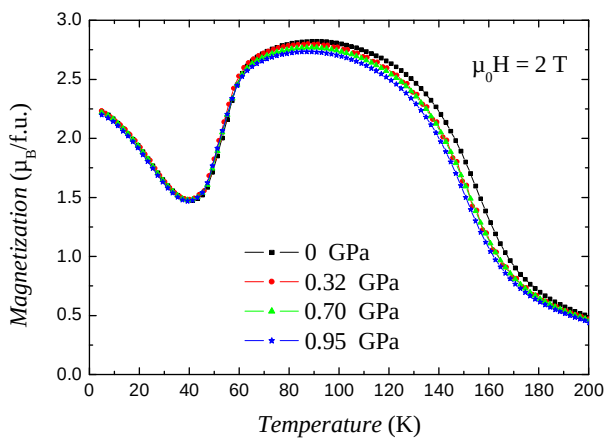

Figure 3: Temperature dependence of isofield magnetization curve (μ_0H = 2 T) of $HoCo_{12}B_6$ compound recorded at different pressures. The minimum in the magnetization observed at about 47 K corresponds to the compensation temperature.

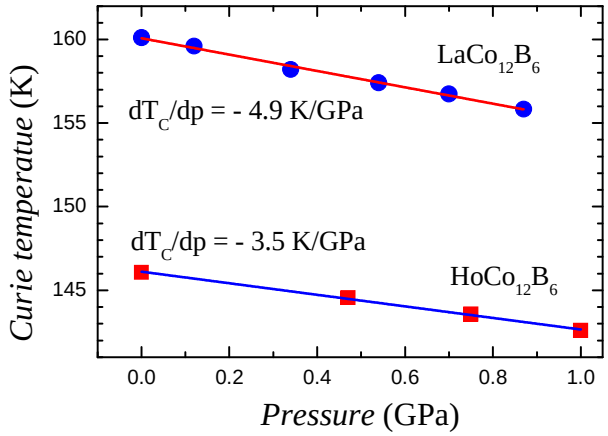

Figure 4: Pressure dependence of magnetic ordering temperature of $HoCo_{12}B_6$ compound with a comparison to that of $LaCo_{12}B_6$

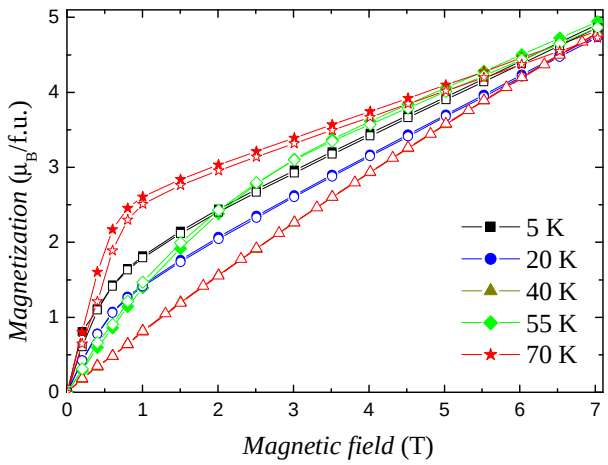

Figure 5: Magnetization isotherms of $HoCo_{12}B_6$ compound measured at the indicated temperatures (below T_{SR}) under different pressures.

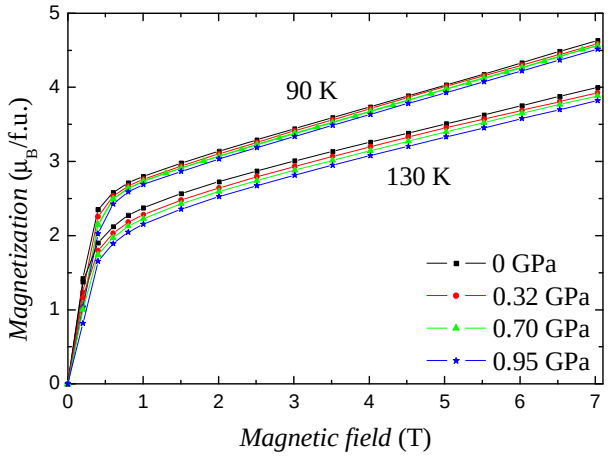

Figure 6: Magnetization isotherms of $HoCo_{12}B_6$ compound measured at the indicated temperatures (between T_{SR} and T_C) at different pressures.

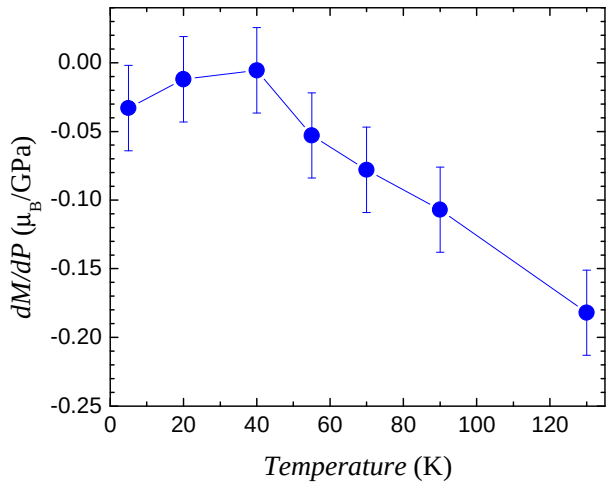

Figure 7: Pressure dependence of magnetization of $HoCo_{12}B_6$ compound recorded up to 7 T at different temperatures.

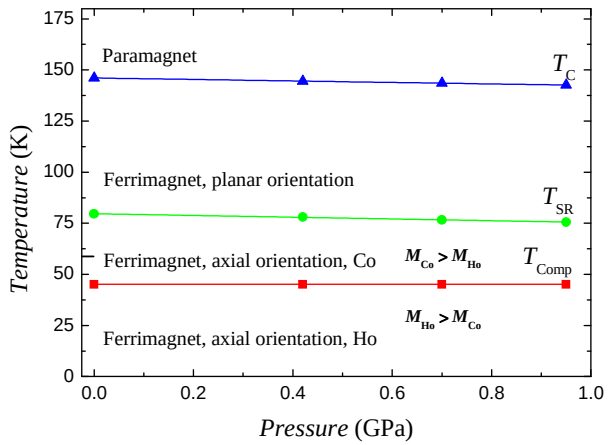

Figure 8: Magnetic phase diagram of $HoCo_{12}B_6$ compound and its temperature and pressure dependence.