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High field magnetization of TiBe_2 and related compounds

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Résumé. — La mesure de l'aimantation en hauts champs (19 T) du composé intermétallique de phase Laves TiBe_2 a été faite à 1,8, 4,2 et 9,6 K et montre une relation non linéaire entre le champ et l'aimantation. L'aimantation commence par augmenter linéairement, puis plus vite que le champ magnétique jusqu'à environ 7 T où une courbure vers l'axe des champs prend naissance. Par comparaison les composés $\text{Ti}(\text{Be}_{0,95}\text{Cu}_{0,05})_2$ et $\text{Ti}(\text{Be}_{0,9}\text{Cu}_{0,1})_2$ sont partiellement ferromagnétiques (ce qui confirme les données de Matthias) mais le composé $\text{Ti}(\text{Be}_{0,99}\text{Cu}_{0,01})_2$ est paramagnétique et sa susceptibilité est inférieure à celle de TiBe_2 . Nous concluons de ces mesures que TiBe_2 est un paramagnétique dont la susceptibilité est fortement augmentée par échange et nous comparons ses propriétés en haut champ à celles de Pd et YCo_2 .

Abstract. — Magnetization measurements at 1.8, 4.2 and 9.6 K at fields up to 19 tesla on the intermetallic Laves phase compound TiBe_2 reveal a non linear behaviour of the susceptibility : the magnetization increases at first faster than the magnetic field up to 7 T and then curves towards the field axis. By comparison the compounds $\text{Ti}(\text{Be}_{0,95}\text{Cu}_{0,05})_2$ and $\text{Ti}(\text{Be}_{0,9}\text{Cu}_{0,1})_2$ display a ferromagnetic character whereas the compound $\text{Ti}(\text{Be}_{0,99}\text{Cu}_{0,01})_2$ appears less magnetic than TiBe_2 . We infer from these facts that TiBe_2 is a very strongly exchange enhanced paramagnet and compare it with Pd and YCo_2 .

Since its discovery [1] TiBe_2 has posed an intriguing puzzle concerning its magnetic properties. It was called an *itinerant antiferromagnet* by Matthias [2] on the evidence of a flat susceptibility below about 20 K and a Weiss temperature also about 20 K. However neither neutron scattering [3] nor specific heat [4] could detect a phase change in the system at low temperatures. On the contrary very recent electron spin resonance experiments [5] shed considerable doubt on the initial interpretation of Matthias.

In an effort to better understand the magnetic properties of TiBe_2 we have performed high field magnetization measurements at low temperatures and compared our results, with those compounds doped with 1 %, 5 % and 10 % Cu, the latter two having ferromagnetic properties [6].

The samples have been prepared by induction melting of the constituents without further annealing [7]. The residual resistivity ratio of our TiBe_2 sample between room temperature and 4.2 K was 19 (compared to ~ 13 in Ref. [2]). It thus appear that

the comparatively slow cooling associated with the induction melting in a crucible (compared to the more rapid one inherent to the arc melting process on a cold substrate) is enough to ensure a reasonable homogeneity for the stoichiometric compound. A special Debye-Scherrer X-ray examination was performed on the 5 % Cu compound and showed only a single phase to be present. Atomic absorption analysis could not detect Cu in TiBe_2 and measured 2.01 % at. Cu in $\text{Ti}(\text{Be}_{0,99}\text{Cu}_{0,01})_2$. The samples weighed about 0.1 g and were of irregular, highly polycrystalline shape. Two independent measuring systems could be used on the same samples in three different magnets : two water-cooled Bitter type solenoids capable of 15 T and 20 T [8] in which the magnetization is measured by extraction of the sample between two counter wound pick up coils whose voltage is integrated, giving an overall sensitivity of 3×10^{-3} emu. The magnetic field is strictly proportional to the current in the coil. The samples are immersed directly in He at 4.2 K. The second apparatus is a superconducting coil capable of 1.3 T at 1.8 K. The magnetization is obtained from the change of the pick up signal of a vibrating coil assembly through which the sample is slowly swept [9]. The

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absolute calibration of the vibrating coil and the field current relationship are established with a palladium reference sample.

Figure 1 shows the ensemble of our results for the magnetization (in emu/mole) at 4.2 K *versus* field for TiBe_2 and the compounds with 1 %, 5 % and 10 % Cu substituted for Be. Concerning the *low* field part of figure 1 ($H < 3$ T) we first note the excellent agreement of the initial susceptibility of TiBe_2 found here : $(8.6 \pm 0.05) \cdot 10^{-3}$ emu/mole at 4.2 K compared with that inferred from figure 1 of reference [2] i.e. $(8.5 \pm 0.1) \cdot 10^{-3}$ emu/mole although no detail is available in reference [2] concerning the field used for the susceptibility determination. This susceptibility is indicated by the straight line of figure 1. No correction is made for demagnetization effect as a rough estimate shows that for TiBe_2 the demagnetization factor is of the order of 10^{-3} .

Fig. 1. — High field magnetization curve at 4.2 K for TiBe_2 and compounds with 1 %, 5 % and 10 % Cu substituted for Be. The straight line represents the initial susceptibility found by Matthias *et al.*, reference [2]. Note that although the compounds with 5 % and 10 % Cu are partly ferromagnetic the compounds with 1 % Cu is paramagnetic and has lower susceptibility than TiBe_2 . For sake of comparison the magnetization of palladium is indicated on the same scale which is in emu/mole (left) and Bohr magneton/formula unit (right).

This very good reproducibility is quite encouraging considering that the samples are of different origins. The striking fact of course is the non linear increase of the magnetization *versus* field relation for TiBe_2 above roughly 3 T. In order to display this effect

more precisely and to study its temperature dependence we have plotted in figure 2 the apparent susceptibility M/H as a function of magnetic field for TiBe_2 at 1.8, 4.2 and 9.6 K (for sake of clarity we have only plotted the 4.2 K magnetization results in figure 1). As is evident from figure 2 this increase is temperature dependent, being most pronounced at 1.8 K where the initial susceptibility is

$$(8.6 \pm 0.05) \cdot 10^{-3} \text{ emu/mole}$$

and increases up to $(9.3 \pm 0.05) \cdot 10^{-3}$ emu/mole at 6.6 T. This maximum apparent susceptibility is already less at 4.2 K $(9.0 \pm 0.05) \cdot 10^{-3}$ emu/mole at 6 T, and barely detectable at 9.6 K near 4 T. This maximum (field induced) susceptibility should be compared with the maximum of susceptibility observed near 10 K [2].

Fig. 2. — Apparent susceptibility M/H of TiBe_2 as a function of field for temperatures 1.8, 4.2 and 9.6 K.

The second remarkable feature of figure 1 is the apparent tendency towards *saturation* of the magnetization exhibited by all compounds measured for fields greater than 10 T. This effect manifests itself by a strong curvature of the initial magnetization towards the field axis until a common slope of $(3.5 \pm 0.5) \cdot 10^{-3}$ emu/mole is reached depending on which compound is considered. This curvature is quite unexpected as the highest magnetization reached is only $0.2 \mu_B$ /formula unit and is very far from saturation if the maximum moment is derived from the Curie constant of reference [2] i.e. $1.64 \mu_B$ /formula unit. The results concerning the Cu 5 % and Cu 10 % TiBe_2 compounds are somewhat less disturbing yielding a very steep magnetization increase at low field as noted previously [6]. However two facts are worth noticing : first the magnetization curve for the $\text{Ti}(\text{Be}_{0.9}\text{Cu}_{0.1})_2$ compound falls roughly a factor 2 below that of reference [6] for the same compound. We have at the moment no explanation for this apparent discrepancy. We note however that the high field slope of the magnetization (above 2 T) of the compound is parallel to the susceptibility curve

of TiBe₂ : from the difference of the two curves one can infer an order of magnitude for the ferromagnetic moment of 300 emu/mole i.e. 0.05 Bohr magneton per formula unit or 0.5 Bohr magneton per Cu atom. We note that the same analysis applied to reference [6] yields about 1 Bohr magneton per Cu atom. Concerning the less doped compound Ti(Be_{0.95}Cu_{0.05})₂ the same analysis performed below 4 T allows as well to determine a ferromagnetic moment of 0.5 Bohr magneton per Cu atom. The second fact we want to emphasize is the apparent merging of the TiBe₂ magnetization curve with that of the Ti(Be_{0.95}Cu_{0.05})₂ system at fields near and above 10 T which appears as a result of the upward curvature of TiBe₂ magnetization curve. Finally the apparent lowering of the initial susceptibility of Ti(Be_{0.99}Cu_{0.01})₂ which is 5.6×10^{-3} emu/mole compared to 8.6×10^{-3} emu/mole for TiBe₂ confronts us with a real puzzle.

We have at present no convincing explanation for this ensemble of results. We consider that the hypothesis of TiBe₂ being an itinerant antiferromagnet has been already very much shaken and the non linear behaviour of the magnetization curve observed here is of little help for this interpretation as none of the orders magnitudes for a combination of spin flop field and saturation effect agree with anything reasonable. On the other hand the interpretation of TiBe₂ as a very strongly exchange enhanced material [5] although in much better position for explaining the specific heat, the neutron diffraction, the occurrence of ferromagnetism and the E.S.R. data, will require considerable effort to cope with the non linear M^3 and M^5 terms in the free energy derivatives. Such an attempt has been already tried by Bloch *et al.* [10] in order to account for the H^3 terms present in the magnetization curves of Laves phases compounds YCo₂ and LuCo₂ (further studied by [11]), Pd [12] and was anticipated for *antiferromagnetic* TiBe₂ [13]. In this description the relation between M and H is derived from a Landau expansion as :

$$\frac{dF}{dM} = H = \frac{1}{\chi} M + BM^3$$

where both χ and B may be temperature dependent.

Assuming a small deviation from linearity this relation is inverted to yield

$$M = \chi H - B\chi^4 H^3.$$

Table I. — Comparison of cubic magnetization terms.

Metal	Pd ^(a)	YCo ₂ ^(b)	TiBe ₂ ^(c)
B (emu/mole) (4.2 K)	-1.6×10^{-4}	-5×10^{-4}	-0.34×10^{-4}
χ (emu/mole) (4.2 K)	0.72×10^{-3}	1.7×10^{-3}	8.6×10^{-3}
$B\chi M_0^2$ ^(d)	- 3.6	- 26	- 9.1

^(a) From reference [12].

^(b) From reference [10].

^(c) This work.

^(d) We have assumed 1 Bohr magneton/formula unit.

We present in table I a compilation of the parameter B for Pd, YCo₂ and TiBe₂ from the available data. We note that in our case this term is only well apparent in a restricted field interval as it is overwhelmed by higher order terms quite soon, contrary to Pd and to the Co compounds. We remark however that the order of magnitude of B varies inversely with the susceptibility of the host thus indicating that such a cubic curvature may be a more general property of strongly paramagnetic systems as soon as a given fraction of the saturated magnetization is reached by the appropriate field. This is clearly seen by rewriting the M - H relation in a dimensionless manner :

$$\frac{M}{M_0} = \frac{\chi H}{M_0} - (B\chi M_0^2) \left(\frac{M}{M_0} \right)^3.$$

There is *a priori* no relation between B and χ as they belong to different terms of the series expansion of the magnetic free energy so that other cases are needed to test this fact.

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