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Itinerant metamagnetism in TiBe_2

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Résumé. — On discute les mesures de Monod, Felner, Chouteau et Shaltiel [1] de l'aimantation du composé défini TiBe_2 sous champs intenses en termes d'une transition métamagnétique sous champs de 5 T environ.

Abstract. — The high field magnetization data of Monod, Felner, Chouteau and Shaltiel [1] on TiBe_2 are discussed in terms of a metamagnetic transition in fields about 5 T.

Monod *et al.* [1] have measured the high field magnetization of the cubic Laves phase compound TiBe_2 and observed a positive deviation from the low field linear dependence of this magnetization at fields above 3 T. At fields above about 7 T the corresponding curves begin to deviate towards the field axis. It is proposed here that this set of phenomena as well as other related results which will be summarized below are a manifestation of itinerant metamagnetism in this compound. TiBe_2 was at first [2] thought to be an itinerant antiferromagnet. It now appears, however, that it is in fact a strongly exchange enhanced paramagnet similar to Pd and YCo_2 . The antiferromagnetism was felt to arise since the magnetic susceptibility has a maximum at about 10 K. The absence of this spin ordering is deduced from neutron [3], specific heat [4] and ESR [5] data. Metamagnetism, i.e. a first order magnetic process, was predicted [6] to occur in TiBe_2 prior to the measurements [1], but at that time the metamagnetic process was suggested to occur from an antiferromagnetic to a ferromagnetic state in a high field, as first proposed in [7]. However, as first proposed in [8], such an effect may equally well occur from a paramagnetic to a ferromagnetic state. Both phenomena have more recently been observed, as summarized in [6] and [9], and TiBe_2 thus joins an interesting list of itinerant metamagnets. The prediction in [6] was made since TiBe_2 was shown in [10] to transform into an itinerant ferromagnet when substituting more than about 6% Be by Cu; this substitution was felt [6] to lead to an internal magnetic field due to local environment effects on the magnetic moments which reside on the Ti atoms [11]. This internal field may lead to metamagnetism as first observed with Gd doped

YCo_2 [12], [6], and more recently with Gd doped Y_4Co_3 [13] and other materials [9]. The process should thus, it was argued, also occur in high external fields. The local environment effects thus envisaged in Cu doped TiBe_2 are not at all clear and will have to be studied further, both experimentally and theoretically, since this set of phenomena is interesting enough to deserve further investigation.

It is now proposed to enlarge slightly on the above discussion, in particular the detailed findings of [1] : Itinerant metamagnetism was shown to be most likely [8] if the low field susceptibility has a temperature maximum. At that time Pd was thus regarded as a good candidate but it now appears that the critical magnetic field is too high. Nevertheless, the occurrence of such maxima continues to point to this phenomenon in both YCo_2 [12] and TiBe_2 [2]. The critical fields may be estimated from the data tabulated in [1]. Using a Landau free energy expansion

$$F = \frac{1}{2} AM^2 + \frac{1}{4} BM^4 - HM, \quad B < 0, \quad (1)$$

two critical fields may be defined [6] : H_c is the field where $\partial^2 F / \partial M^2 = 0$ and H_c^* where

$$F[M(H_c^*)] = F[0] = 0,$$

giving

$$\left. \begin{aligned} H_c &= (2/3\sqrt{3}) (A^3/|B|)^{1/2}, \\ H_c^* &= (1/\sqrt{2}) H_c. \end{aligned} \right\} \quad (2)$$

The tabulation in [1] leads to the following values of H_c^* :

$$\text{Pd} : 110 \text{ T}; \quad \text{YCo}_2 : 17 \text{ T}; \quad \text{TiBe}_2 : 5.8 \text{ T}. \quad (3)$$

The « low » value for YCo_2 is in disagreement with earlier estimates [12], [14] but the value for TiBe_2 lies in the centre of the anomalous increase of M observed in [1]. Of course, this increase is not sharp but spread over about 3 T and this effect will be briefly discussed below, again in terms of local environment effects. Nevertheless, this « low » value of H_c^* for TiBe_2 makes this substance a more ideal itinerant metamagnet than Pd or YCo_2 . All three substances have susceptibility maxima in line with theory [8]. Such maxima may occur if the density of states curve has a local minimum or dip at the Fermi energy. Precisely this result was obtained in a band calculation for TiBe_2 [15]. However, susceptibility maxima may also arise for other reasons, a case in point being the cubic material TiH_2 ! Here such a maximum occurs at 300 K [16] and this was for many years regarded as an indication of antiferromagnetism (I was myself guilty of this heresy [17]). It is now clear, as shown by Ducastelle *et al.* [16] and Gupta [18], that an electronically driven tetragonal distortion at this temperature gives rise to the maximum and this possibility needs to be kept in mind also for TiBe_2 and perhaps elsewhere.

Other features found in [1] and stressed there as particularly interesting include the low value of the magnetization at high fields compared to the value derived from the Curie-Weiss constant. This is, however, completely in line with the itinerant electron model : Above the critical field the material is in effect a « weak itinerant ferromagnet » and here the ratio of the Curie-Weiss to the saturation moment is always large [19]. In high fields the observed ratio tends *very roughly* to $1.64/0.2 = 8.2$, which would correspond [19] to an itinerant ferromagnet with Curie point about 30 K. The Curie points of Cu doped TiBe_2 increase with increasing copper content, reaching [10] 33 K for $\text{Ti}(\text{Be}_{0.75}\text{Cu}_{0.25})_2$. Hence this value of 30 K, just crudely estimated, could be taken to be the limiting Curie temperature for the weak itinerant ferromagnet TiBe_2 whose order is produced by very large internal or external fields. Monod *et al.* [1] also investigated several Cu doped TiBe_2 specimens. For the sample $\text{Ti}(\text{Be}_{0.9}\text{Cu}_{0.1})_2$ the magnetization curve lies roughly a factor 2 below that reported in [10]. If this difference is real it might be a manifestation of the sensitivity of the transition to local environment effects, briefly discussed below. In the same way it may be possible to explain the fact that the magnetization curve of $\text{Ti}(\text{Be}_{0.99}\text{Cu}_{0.01})_2$ has an initial susceptibility below that of pure TiBe_2 , namely [1] 5.6×10^{-3} emu/mole compared to 8.6×10^{-3} . The authors of [1] also point out that the magnetization curves for TiBe_2 and $\text{Ti}(\text{Be}_{0.95}\text{Cu}_{0.05})_2$ tend to merge at fields above about 10 T. Since this is already roughly $2H_c^*$ it seems reasonable to expect this tendency of mutual approach of the magnetization

curves in *total* (external + internal) fields of this order.

Reference has been made several times to the importance of local environment effects in this problem. It is envisaged that TiBe_2 carries no moments in zero and low fields $< H_c^*$. Adding copper atoms produces magnetically active Ti atoms [11], the magnetic distribution being rather well concentrated on these sites. However, it is clearly stated in [11] that further experiments are required to assess whether an average Ti site is the result of a uniform or a widely non-uniform environment of copper atoms. The effective internal fields would, in the second case, also vary sensitively, giving rise to the spread of the metamagnetic transition and to the differences of the magnetic results for different specimens of Cu doped TiBe_2 alluded to earlier. The magnetic heterogeneities can, however, not be too severe since Arrott plots (M^2 vs. H/M) have been found to be straight for $\text{Ti}(\text{Be}_{0.9}\text{Cu}_{0.1})_2$ [10]. On the other hand, such heterogeneities seem to show up in the high pressure data of Chu *et al.* [20].

Local environment effects can be assessed theoretically using modern approaches to the alloy problem [21]. What is at issue the effect of replacing Be by Cu atoms and the resulting effective fields on the magnetically active sites. The detailed mechanism is unknown and would require further theoretical considerations. Experimentally, it would be necessary to assess these fields and their distribution by several methods such as neutron scattering [11], Mössbauer, NMR, etc. The magnetic fields involved have been shown to be of the order of 5 T, i.e. relatively low, and hence the sensitivity of the magnetic properties to these environmental effects must be considerable. The effects of heat treatment on these properties should thus also be investigated. I am grateful to the authors of [1], [5], [11] and [15] for their kindness in sending me preprints.

Note added in proof. — High field measurements on pure and Cu doped TiBe_2 have also been reported by Acker *et al.* [22]. The results are similar to those of reference [1] and were interpreted in terms of exchange enhanced paramagnetism in all fields, in contrast to the present proposals. However, the Arrott plots for TiBe_2 continue to curve in the highest fields applied (21.3 T), making it impossible to distinguish the two proposals. Furthermore, the χ versus H maximum, here regarded as manifesting spread first order transitions, were proposed in [22] to follow from Fermi liquid theory, giving a term in χ going as $H^2 \ln H$. The status of this term is, however, still completely uncertain, as is that of the $T^2 \ln T$ term used in [22] to explain the susceptibility versus temperature maximum.

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