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Thermal expansion and spin density fluctuations in $\text{TiBe}_{2-x}\text{Cu}_x$ itinerant ferromagnets

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Résumé. — Nous présentons des mesures de dilatation, de magnétostriction et d'aimantation sur des alliages ferromagnétiques itinérants $\text{TiBe}_{2-x}\text{Cu}_x$. Le modèle de Stoner ne permet pas d'interpréter nos résultats, mais ceux-ci sont en bon accord avec les prédictions du modèle de fluctuations de densité de spin de Moriya et Usami.

Abstract. — We have made thermal expansion, magnetostriction and magnetization measurements on the itinerant ferromagnetic alloys $\text{TiBe}_{2-x}\text{Cu}_x$. The results cannot be interpreted on the Stoner model, but are in good agreement with the predictions of the spin density fluctuation model of Moriya and Usami.

The Stoner model [1] has long been one of the basic conceptual tools in metal magnetism, particularly in the field of itinerant ferromagnets. However recent theoretical and experimental work [2-5] has indicated that while the Stoner model gives a good representation of the $T = 0$ state, behaviour at finite T should be understood in terms of spin density fluctuations (SDF). One physical property for which the two approaches give explicit and different predictions is the thermal expansion [6, 7]. Previous experimental results have been inconclusive : thermal expansion work on the band ferromagnet MnSi [8] has suggested that agreement with SDF is better than with the Stoner model while results on certain other systems have been interpreted

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in terms of the Stoner model [9]. By choosing a particularly favourable system we have been able to make a stringent quantitative check of the models. We have made magnetization, magnetostriction and thermal expansion measurements on the itinerant ferromagnetic alloys $\text{TiBe}_{2-x}\text{Cu}_x$ ($x > 0.15$); we observe behaviour which is quite different from the Stoner predictions while being in good agreement with a simple version of the SDF model. The results demonstrate that the SDFs cause the fluctuating mean square local moments to increase strongly with temperature for $T > T_c$.

TiBe_2 is a strongly enhanced paramagnet and $\text{TiBe}_{2-x}\text{Cu}_x$ alloys show properties typical of itinerant ferromagnets above a critical composition $x \simeq 0.15$ [10-14]. Samples with $x = 0, 0.16, 0.20$ and 0.24 were prepared using the same technique as described by Smith [13] and were each in the form of a small cylinder cut from the centre of a melted and annealed button. At each concentration, thermal expansion, magnetostriction [15] and magnetization measurements were all done on one and the same sample. The magnetization results were in good agreement with previous work [10-14]. The alloys are somewhat magnetically inhomogeneous, as can be seen from low field curvature in the Arrott plots for $T > T_c$. In addition, the coefficient γ (the slope $(M/m)/m^2$ in the Arrott plots) is field dependent, as was remarked by Acker *et al.* [12].

Magnetostriction was strongly positive throughout for all samples, figure 1. We find :

$$\frac{l(H, T) - l(0, T)}{l(0, T)} = \lambda(m^2(H, T) - m^2(0, T))$$

with a value of λ which varies little with temperature. The value of λ was similar for the three ferromagnetic samples.

The thermal expansion results are shown in figure 2. Pure TiBe_2 shows a positive thermal expansion which is closely proportional to T^2 up to about 18 K, and slightly faster than T^2 beyond. The ferromagnetic alloys show negative thermal expansion at low temperatures becoming strongly positive for $T > T_c$.

Before analysing the results, we will outline what we can expect to see on the Stoner and SDF models. On the Stoner model [6] the magnetic length term is $Cm^2(H, T)$ where C is a constant

Fig. 1. — Magnetostriction of $\text{TiBe}_{1.8}\text{Cu}_{0.2}$. The length change $[l(T, H) - l(T, 0)]/l(T, 0)$ is plotted against the magnetization squared $m^2(T, H)$ in $(\mu_B \text{ per Ti atom})^2$, for $T = 1.5 \text{ K}$ and $T = 21.6 \text{ K}$.

Fig. 2. — Thermal expansion of TiBe_2 , $\text{TiBe}_{1.84}\text{Cu}_{0.16}$, $\text{TiBe}_{1.8}\text{Cu}_{0.2}$ and $\text{TiBe}_{1.76}\text{Cu}_{0.24}$. The origins for the different curves are arbitrary, but the scales are all the same.

for the material and m is the overall magnetic moment. Magnetostriction results are of this form, figure 1, and can be used to calibrate C (which we can identify with the experimental λ). In addition, at $H = 0$ the spontaneous magnetic length change should be $Cm_s^2(T)$ where $m_s(T)$ is the spontaneous magnetization as estimated from Arrott plots. Using the λ value from the magnetostriction we can predict the spontaneous length change with no free parameter.

From among the SDF models, we will outline the phenomenological itinerant ferromagnetic limit model of Moriya and Usami [7], because of its simplicity. In analogy with the Stoner relation $\delta l/l = Cm^2$ we now have $\delta l/l = C \langle m_i^2 \rangle$ where the m_i are the fluctuating instantaneous local moments on the different sites. Moriya and Usami write an energy functional :

$$F = \sum_q \frac{1}{2 \chi_q^{\text{HF}}} m_q^2 + \frac{\gamma}{4N} \sum_i m_i^4,$$

where $m_q = \frac{1}{N} \int d\mathbf{r} e^{i\mathbf{q} \cdot \mathbf{r}} \mathbf{m}(\mathbf{r})$.

At $T = 0$ there is a uniform magnetization state where all m_i are equal with $m_i^2(0) = -\frac{1}{\chi_0^{\text{HF}} \gamma}$. Above T_c , they assume that χ_q^{HF} is approximately constant for the q values of interest, and they make the mean field type of approximation [16] :

$$\langle m_i^4 \rangle \simeq \frac{5}{3} \langle m_i^2 \rangle \langle m_i^2 \rangle.$$

This model predicts for the spontaneous magnetic length change between $T = 0$ and $T = T_c$

$$\frac{l_0 - l_{T_c}}{l_0} = \frac{2}{5} C m_s^2(0),$$

and above T_c

$$\frac{l_T - l_{T_c}}{l_0} = \frac{3}{5} C \frac{1}{\gamma \chi(T)}.$$

Here γ can be determined from Arrott plots and C is obtained as above from magnetostriction so we again have a prediction with no free parameters for the magnetic thermal expansion.

We now turn back to the experimental data. A major difficulty other workers have run into in attempting to isolate the magnetic thermal expansion is the correction for background phonon and electronic terms which rapidly become important with increasing temperature. Here we are in a more satisfactory position as we have data for the pure TiBe_2 which we will consider to give us the paramagnetic « background ». To be more explicit, we can consider the different types of term. First, the phonon term : this is weak at these temperatures as we are in a high Debye θ system, and any way this term should be similar in TiBe_2 and in the alloys. Secondly the electronic term : this is certainly strong — the $\delta l/l = \alpha T^2$ behaviour suggests it is dominant in the pure TiBe_2 . However the electronic specific heats of TiBe_2 and $\text{TiBe}_{1.8}\text{Cu}_{0.2}$ are almost identical [14] so this term should be much the same in the alloys as in the pure compound (the electronic term in the thermal expansion is directly related to the volume dependence of the electronic specific heat). Finally, anticipating our results on the alloys we can expect an SDF term in the pure TiBe_2 , but for the moment we will assume this term is zero. We will take :

$$\frac{\delta l^*}{l}(T) = \left[\frac{\partial L}{L}(T) \right]_{\text{alloy}} - \left[\frac{\partial L}{L}(T) \right]_{\text{TiBe}_2},$$

as giving us the magnetic thermal expansion of the alloys.

Results for the sample $x = 0.20$ are given in figure 3, where $\frac{\delta l^*}{l}(T)$ is compared to the Stoner and SDF predictions. We can see that the Stoner prediction fails badly — it gives much too high an estimate of $[l^*(0) - l^*(T_c)]/l(0)$, and does not predict the strong positive thermal expansion above T_c . The SDF prediction is much closer to experiment. The predicted $[l^*(0) - l^*(T_c)]/l(0)$ while still too high is much closer to the observed value, and the model predicts the positive thermal expansion above T_c .

Two experimental considerations could improve agreement with the model. First, as the T_c of the sample is somewhat smeared out, we should smear out also the sharp minimum at T_c in the model curve. Secondly, for the model curve above T_c we have used a constant low field value of γ ; if we had chosen to use the high field γ (γ seems to reach an almost temperature independent value above about 40 kG) the calculated model curve above T_c would lie slightly above the experimental curve. Finally, to be consistent we should allow for a SDF term in the thermal expansion of the pure TiBe_2 . As the χ of TiBe_2 in this temperature range varies little [14] this contribution should be small, but if it exists the « background » phonon plus electronic term we subtract from the alloy data would be slightly lower.

On the theoretical side, the model [7] is certainly over simplified, particularly around T_c . If we take the model literally we would expect Curie-Weiss behaviour for the susceptibility right down to T_c , which is not the case. However the model has the great virtue of introducing no free fitting parameter; we have not made specific comparisons with other versions of the SDF approach [3, 5] which do not have this advantage.

Fig. 3. — The magnetic thermal expansion for $\text{TiBe}_{1.8}\text{Cu}_{0.2}$, $\delta l^*(T)/l$ as defined in the text. Curves calculated using the Stoner model and the Moriya-Usami model are given for comparison. Between $T = 0$ and $T = T_c$ the form of the Moriya-Usami model curve is only schematic.

Neutron diffraction results on itinerant ferromagnets at $T > T_c$ give complementary information on the spatial correlations between these fluctuating moments [17, 18]. Measurements on TiBe_2 alloys would be difficult but interesting.

In conclusion, the SDF model of Moriya and Usami [7] seems to give an excellent physical picture for the behaviour of this typical itinerant ferromagnetic system. At $T = 0$, the magnetization is uniform with all $\mathbf{m}_i$ equal to $\mathbf{m}_s(0)$. By T_c , $\langle \mathbf{m}_i \rangle$ is zero but there are local fluctuating $\mathbf{m}_i$, random in direction and varying considerably in modulus, with $\langle \mathbf{m}_i^2 \rangle_{T_c} \simeq 0.6 m_s^2(0)$. As the temperature is further increased and the susceptibility drops, the moduli of the fluctuating local moments increase regularly, so $\langle \mathbf{m}_i^2 \rangle$ rapidly overtakes $m_s^2(0)$. The thermal expansion clearly reflects this behaviour.

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- [15] Throughout we will refer to relative length changes, which are just 1/3 of relative volume changes.

We have checked that the magnetostriction is isotropic.

- [16] The factor 5/3 is the ratio $\left[\int_0^\infty x^6 e^{-x^2} dx / \int_0^\infty x^2 e^{-x^2} dx \right] \left[\int_0^\infty x^4 e^{-x^2} dx / \int_0^\infty x^2 e^{-x^2} dx \right]^{-2}$.
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