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# Field-dependent ac susceptibility of $Ce_2Fe_{17}$

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**Abstract**

Over part of the $H$–$T$ plane, the field- and temperature-dependent ac susceptibility, $\chi(H, T)$, of $Ce_2Fe_{17}$ is found to exhibit maxima, the amplitude of which increase with increasing field. While the appearance of such a maximum seems intuitively correct along a so-called 'S-shaped' magnetization ($M$) versus field ($H$) curve characterizing a metamagnetic/first-order phase transition, it has not been previously observed in the ac susceptibility of other systems exhibiting such transitions. In an attempt to identify the origin of this effect, detailed magnetization data, $M(H, T)$, are used to reconstruct a phase diagram for this system—specifically by using the derivatives $\partial M/\partial T$ and $\partial M/\partial H$—as detailed recently by Janssen and co-workers. This behaviour is then shown to occur on crossing the boundary from a *ferromagnetic* into a *spin flop* (SFI) phase in $Ce_2Fe_{17}$. As a corollary, this previously atypical behaviour of $\chi(H, T)$ may be a defining characteristic of such transitions; specifically, it may reflect the possibly unique combination of ferromagnetic and antiferromagnetic interactions that are present in this system.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

The field- and temperature-dependent ac susceptibility, $\chi(H, T)$, of magnetic materials has been widely used to provide fundamental information about phase transitions, paramagnetic to ferromagnetic transitions in particular [1]. Here we report a detailed investigation of the magnetic properties of the $Ce_2Fe_{17}$ alloy system which reveals a complex behaviour of the ac susceptibility as a function of field and temperature, a perhaps not unexpected result in view of the complicated (magnetic) structures that this system displays. Of particular note however, is that over part of the $H, T$ plane, $\chi(H, T)$ is found to exhibit not only a maximum, but one with a magnitude which increases with increasing field. While the appearance of a maximum per se may appear intuitively appropriate near a metamagnetic/first-order transition around which magnetization ($M$) versus field ($H$) curves display an 'S-shape' characteristic of such transitions (namely, above the metamagnetic field the magnetization is initially an increasing function of field, so the differential susceptibility (the local slope of the $M$–$H$ curve [2]) passes through a maximum), it has not been observed previously in ac susceptibility measurements, and thus no field dependence for it has been established. Specifically, detailed measurements of the ac susceptibility, $\chi(H, T)$, in systems such as $Gd_2In$ [3] and $Ce(Fe_{0.93}Ru_{0.07})_2$ [4], which both display metamagnetic transitions, failed to reveal the presence of such a maximum; consequently the influence of increasing applied fields on it remain unexplored. This situation probably arises because of the fundamental difference between measurements of the differential and ac susceptibilities, the latter measuring the averaged slope of an ac minor loop, the average being taken over the amplitude of the ac driving field [2, 5]. That these procedures produce markedly different results is illustrated by their behaviour near a conventional second-order/continuous paramagnetic to ferromagnetic transition. Above the ordering temperature of such a transition, the magnetization–field curves exhibit a slope that decreases monotonically with decreasing field; in particular, along the critical isotherm at $T_c$ conventional scaling theory describes the field dependence of the magnetization in the power-law form [6]

$$M(H, T_c) \sim H^{1/\delta} \qquad (1)$$

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which, for the critical exponent $\delta > 1$, yields a monotonically decreasing differential susceptibility/slope with increasing field. In contrast, the temperature-dependent ac susceptibility measured in fixed applied fields exhibit maxima above $T_c$, a behaviour which has not been replicated in the differential susceptibility found from measurements of the (dc) magnetization in fixed field versus temperature over the same regime. Such ac susceptibility maxima have been shown [1, 7] to decrease in amplitude while their location in temperature increases above the ordering temperature, $T_c$, as the applied field increases; theoretically, the emergence of this peak structure can be understood [8] on the basis of the fluctuation-dissipation theorem, with the locus of such peaks defining a crossover line in the $H$–$T$ plane below which the response is field dominated, while above this line the response is temperature dominated [8].

The response reported here, at least for a section of the $Ce_2Fe_{17}$ phase diagram, is in marked contrast, and this may be a determining factor for certain ferromagnetic–spin flop transitions, as discussed below.

Rare earth–transition metal alloys of the type $R_2Fe_{17}$, with the most abundant rare earth, Ce, and the cheapest transition metal, Fe, i.e. $Ce_2Fe_{17}$, have been investigated extensively, a consequence of both their unusual magnetic properties and potential applications. The magnetic properties of $Ce_2Fe_{17}$ are anomalous, as investigations using a wide range of techniques confirm. Early studies indicated that on cooling, $Ce_2Fe_{17}$ has a paramagnetic to antiferromagnetic/helical transition at $T_N \approx$ 220 K, and subsequently an unusual antiferromagnetic/helical to ferromagnetic/fan structure transition around $\Theta_T \approx$ 90 K [9]. Subsequent Mössbauer studies over an extended temperature range, and including the effects of doping with hydrogen [10], confirmed this result. The most recent study [11], encompassing the orientational response in single crystals, concluded that this system displays not one, but two transitions: a paramagnetic to antiferromagnetic (AF) phase at a Néel temperature, $T_N$, $\approx$ 208 K, and a second transition at $\Theta_T \approx$ 124 K. Doping with 0.5% Ta raises the Néel temperature slightly to 214 K, with a subsequent transition to a ferromagnetic phase occurring near $T_\Theta \sim$ 75 K. This latter, comprehensive study by Janssen *et al* [11] included phase diagrams in the $H$–$T$ plane for both undoped and Ta-doped $Ce_2Fe_{17}$ single-crystal samples, and elucidating the conclusion of the emergence of differing ordered low temperature states from earlier studies [9, 12, 13]. The phase diagrams deduced from this recent study displayed several complicated features, including a field-induced spin flop (SF) transition to a low temperature ferromagnetic state. Aspects of these phase diagrams appear well described by Moriya–Usami-like phenomenological theories [14] which, appropriately, include competing antiferromagnetic and ferromagnetic interactions prevalent in this system.

For the present sample, that region of the $H$–$T$ plane over which the ac susceptibility maxima exhibit an increase in amplitude with increasing applied field is shown to map onto the boundary of the field-modulated SFI intermediate phase, identified using the techniques advocated by Janssen *et al* [11]. As the latter authors suggest, this boundary is quite

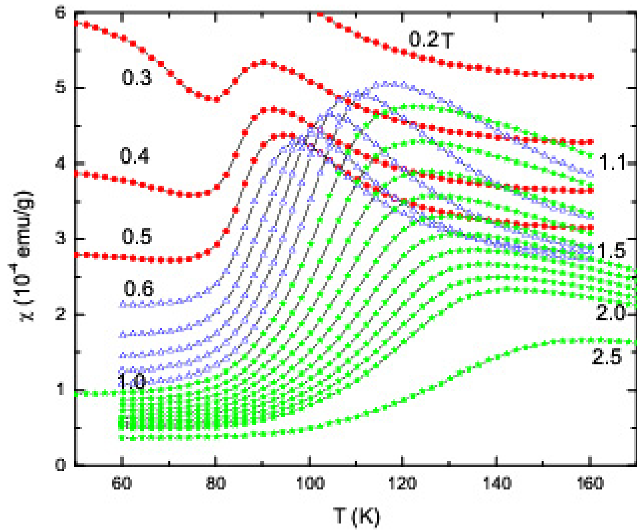


**Figure 1.** The temperature dependence of the ac susceptibility, $\chi(H, T)$, of $Ce_2Fe_{17}$ in the vicinity of the lower transition(s), near 100 K, measured in various static applied fields (marked) between 0.2 and 2.5 T. The blue open triangles designate the regime of response where the susceptibility peak amplitudes increase with increasing applied field, 0.6 T $< H <$ 1.0 T.

different from the conventional paramagnetic to ferromagnetic boundary. The current data thus not only are consistent with the presence of this recently proposed SFI intermediate phase, but, and more importantly, also may provide a criterion for identifying the boundary to such a phase in future studies.

## 2. Experimental details

Polycrystalline samples of $Ce_2Fe_{17}$ were prepared from Ce and Fe with a purity better than 99.99% by arc melting in an Ar atmosphere; the samples were inverted and remelted several times to ensure homogeneity, and this was followed by annealing, as detailed previously [15]. X-ray powder diffraction data were collected at room temperature using Cu K$\alpha$ radiation. These data confirmed that the sample was single phased with $Th_2Zn_{17}$ structure with lattice constant $a =$ 8.495 Å and $c =$ 12.412 Å.

A Quantum Design Model 6000 PPMS was used to measure the ac susceptibility (at 2.4 kHz with an ac driving field amplitude of 10 $\mu$T) in zero field and various non-zero static applied fields as a function of temperature; also detailed measurements of both the isothermal magnetization as a function of field at numerous selected temperatures and the variation of the magnetization with temperature in different constant fields were made. All fields were applied along the largest sample dimension to minimize demagnetization effects.

## 3. Results and discussion

The temperature dependences of the ac susceptibilities measured in various applied fields in the vicinity of the lower transition near 100 K are presented in figure 1. Figure 1 reproduces ac susceptibility data over the field

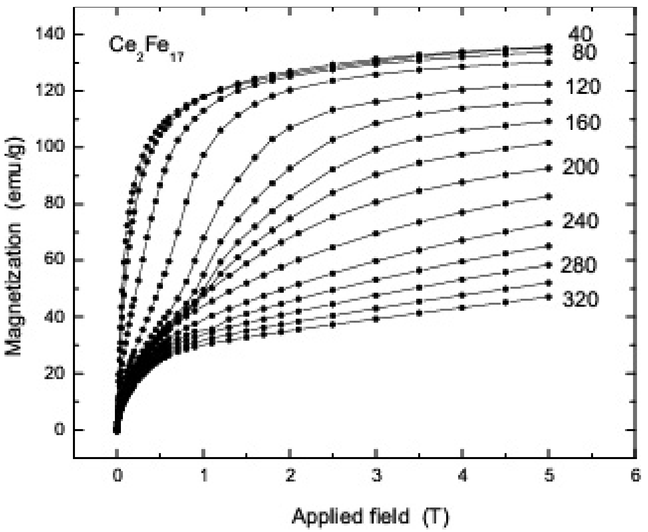


**Figure 2.** The temperature dependence of the isothermal magnetization of $Ce_2Fe_{17}$ in the temperature range 40–320 K in fields from 0 to 5 T.

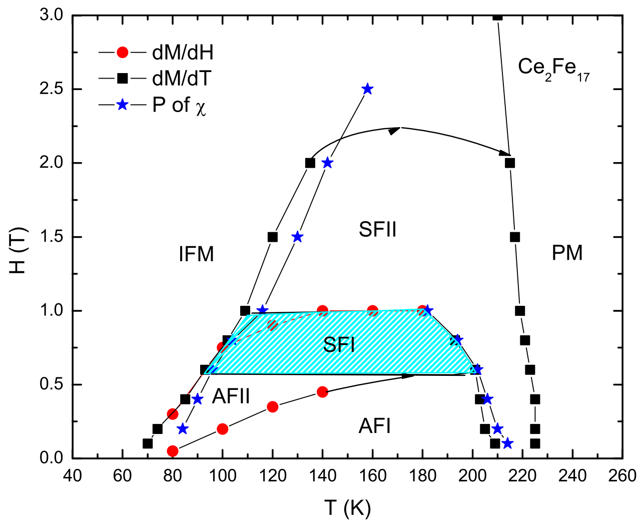


**Figure 3.** The phase diagram—in the $H$–$T$ plane—for the present $Ce_2Fe_{17}$ sample, constructed using magnetization and susceptibility data. The phase boundaries determined from $\partial M/\partial H$ are represented by filled red circles, those from $\partial M/\partial T$ by filled black squares (both following the protocol established in [11]); the boundaries corresponding to the ac susceptibility maxima are represented by filled blue stars using data over the complete range of field and temperature (shown in figure 4). The area of the $H$–$T$ plane designated SFI (shown in the hatched area) corresponds to the small window marked in figure 4.

range from 0.2 to 2.5 T, showing the emergence of maxima near a metamagnetic transition (as figure 2 confirms), in contrast to the behaviour reported for polycrystalline $Gd_2In$ [3] and $Ce(Fe_{0.93}Ru_{0.07})_2$ [4] (and confirmed by a detailed re-examination of the ac susceptibilities in both systems), both of which exhibit field-modulated, metamagnetic antiferromagnetic to ferromagnetic transitions. The variation in the peak height and temperature with field evident here is complex; while the peak temperature increases monotonically with increasing applied field, the peak amplitude first decreases, then increases, before finally decreasing again with increasing field. As mentioned in section 1, the ac susceptibility measured in fixed field as a function of temperature immediately above $T_c$ at a paramagnetic to ferromagnetic transition in numerous systems [1, 7] displays a maximum, the temperature of which increases while the amplitude decreases with increasing field. This behaviour is predicted on the basis of standard scaling theory, and enables estimates of the associated critical exponents to be made [1, 7, 8]. A similar response has also been reported in superparamagnetic systems [16], and can be similarly explained. In contrast, the previously unreported increase in peak amplitude with field occurring in $Ce_2Fe_{17}$ is shown in greater detail in figure 1 by the blue open triangles, covering the field range from 0.6 to 1.0 T.

In an attempt to identify the source of this effect, a phase diagram for the present sample has been constructed using the criteria adopted by Janssen *et al* [11]. Specifically, numerical estimates for $\partial M(T)/\partial T$ were made as a function of temperature using the temperature dependence of the magnetization measured in various applied fields, while the isothermal magnetization measured as a function of field provided estimates of $\partial M(H)/\partial H$ at various fields. Combining these two data sets enables a phase diagram to be constructed in an analogous manner to that of Janssen *et al* [11]; given the previous detailed discussion of this procedure, just a summary of the resulting phase diagram is reproduced in figure 3. This phase diagram is in good overall agreement with those in [11]. Actually figure 3 is slightly less complicated than that reported for an undoped single crystal with applied field $H$ perpendicular to the $c$ axis (on which the present phase nomenclature is based), though it displays some features reminiscent of those for the Ta-doped $Ce_2Fe_{17}$ single crystal of that reference, including several 'domed' $H$–$T$ boundaries. This probably arises due to the polycrystalline nature of the present specimen, which might also contain trace amounts of W and Cu (from the arc melting process). Indeed, low temperature magnetization data ($T < 50$ K) for the present sample are also similar to those for the Ta-doped specimen of [11] (and hence are not reproduced here), and these were interpreted as being consistent with a fan-like structure with a ferromagnetic component at low temperature and low field.

For completeness, figure 4 presents a comprehensive summary of the temperature dependences of ac susceptibilities over the temperature interval from 10 to 300 K and in fields between 0.1 to 5 T. The mapping of the characteristic features evident in this figure with the phase diagram constructed from the corresponding magnetization data using the prescription advocated by Janssen *et al* [11] is striking. At higher temperatures—near 200 K—the ac susceptibility exhibits a peak which, while decreasing in amplitude, shows a slight decrease in temperature between 0.1 and 0.6 T; this peak can be seen to correspond closely with the upper phase boundary of the antiferromagnet AFI phase established using magnetization data, specifically $\partial M(T)/\partial T$. This association appears physically reasonable; increasing the uniform applied field—the antithesis of the conjugate field for antiferromagnetic

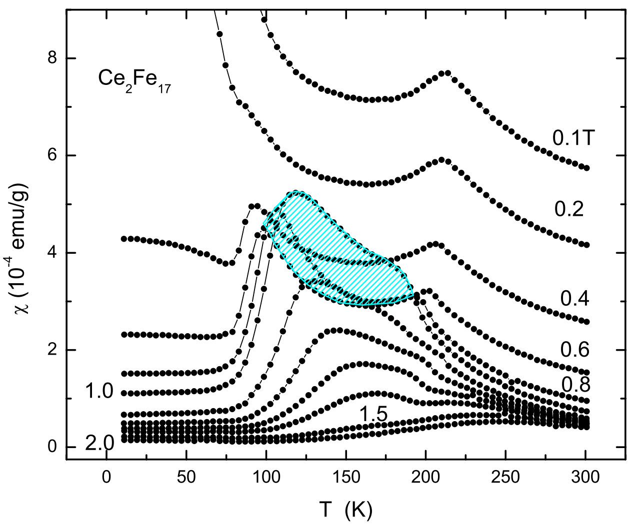


**Figure 4.** The field and temperature dependence of the ac susceptibility, $\chi(H, T)$, over the entire range examined; $10\text{ K} \leqslant T \leqslant 300\text{ K}$, $0.1\text{ K} \leqslant H \leqslant 2.5\text{ T}$ for the present $Ce_2Fe_{17}$ specimen. The regime (see the text and figure 1) corresponds to the SFI phase, $90\text{ K} \leqslant T \leqslant 190\text{ K}$, $0.6\text{ T} \leqslant H \leqslant 1.0\text{ T}$, and is shown as the hatched area.

order—should suppress such order, but given the magnitude of the ordering temperature compared to that of available fields, the suppression of the transition temperature should be slight. Beyond 0.6 T this peak evolves into a shoulder, a feature discussed in more detail below.

The field and temperature dependence of the ac susceptibility near 80 K is more complicated; this reflects the fact that several complex phase boundaries (determined, following [9], from both $\partial M(T)/\partial T$ and $\partial M(H)/\partial H$) are in close proximity, if not coincident, here. In this region the ac susceptibility peak amplitudes first decrease ($H = 0.2$–$0.6$ T), then increase ($H = 0.6$–$1$ T) before decreasing again (from 1.1 to 5 T) with increasing applied field, as mentioned earlier. In contrast, the susceptibility peak temperatures increase monotonically with field, tracking closely along the phase boundaries between the IFM–AFII, SFI and SFII regimes determined from $\partial M(T)/\partial T$ and $\partial M(H)/\partial H$ estimates. Furthermore, the ac susceptibility peak temperature–field profile over the 'atypical' regime (that region of the $H$–$T$ plane where the peak amplitudes increase with field) maps directly onto the field-induced ferromagnetic IFM–SFI boundary line. It should be noted that the zero-field ac susceptibility falls monotonically with increasing temperature between 50 and 150 K. Here the SFI and SFII phases are not present; they emerge only in the presence of non-zero applied fields. On the basis of the argument that the uniform applied field is the conjugate field for a collinear magnetization, one might initially anticipate that the ac susceptibility (peak) would display a monotonic variation with field along the entire low temperature boundary shown in figure 3. It does not. The ac susceptibility peak falls with increasing field on exiting the IFM phase along both the IFM–AFII and the IFM–SFII transition lines, which is reminiscent of the case for conventional ferromagnetic to paramagnetic transitions. That the corresponding peak amplitude increases with increasing field along the IFM–SFI boundary surely reflects the more subtle influence of this field on the complex spin rearrangements evolving along this boundary, which, in turn, manifest the underlying competing spin–spin interactions in this system.

Finally it should be noted that in this same low temperature region of nearly overlapping phase boundaries near 80 K, the low field ($0.2\text{ T} < H < 0.6\text{ T}$) decrease in ac susceptibility peak amplitude, accompanied by a slight increase in the peak temperature, tracks the boundary to the AFII phase in this region. This mimics the field dependence of the response along the AFI boundary near 200 K, mentioned earlier, with increasing uniform applied fields again suppressing the regime of accessibility of the $H$–$T$ plane to this AFII phase. (The high temperature boundary of the SFI phase near 200 K tracks the shoulder structure, mentioned above.)

It would clearly be advantageous if the underlying spin configurations in the various phases mentioned above, and shown in the associated phase diagram, could be established. In this way it might be possible to identify the spin reorientational processes responsible for the unusual field dependence in the ac susceptibility reported above, and subsequently investigate the field dependence of the physical mechanisms/interactions driving these reorientation processes. In this context, the present study suffers from the same limitations as the study reported by Janssen *et al* [11] in utilizing macroscopic probes; such probes do not yield information from which definitive conclusions regarding microscopic spin configurations can be drawn. The latter would require detailed single-crystal neutron scattering studies, for example. Under the present circumstances therefore, it is only possible to suggest possible scenarios on the basis of comparisons with theories that predict phase diagrams with some similarities to those observed experimentally—Moriya–Usami-like approaches [14], for example. This latter theory utilized a single underlying ferromagnetic (FM) and a second competing antiferromagnetic (AFM) coupling in an itinerant electron system, and the resulting phase diagram exhibited a high temperature paramagnetic regime, separated from a ferromagnetic ground state by an intervening antiferromagnetic phase; these predictions exhibit a striking resemblance to the IFM, AFI and PM phases appearing as ground states in various temperature intervals in zero applied field for the present sample (figure 3). Differences emerge, however, between model predictions and observations for finite field. The observed phase diagram(s) display considerably more complexity than the single enveloping dome-like feature delineating the antiferromagnetic regime in model calculations. The results reported above indicated the emergence of a field-induced antiferromagnetic AFII phase along with two intermediate field-induced spin flop phases (SFI and SFII); the atypical response which is the subject of this report occurs only along the low temperature boundary of the first of these spin flop phases. Such differences probably reflect theoretical limitations imposed by the use of two competing/interacting modes representing uniform ferromagnetism and staggered antiferromagnetism (wavevector $q = 0$ and $q \neq 0$, respectively), with the ensuing free energy expansion being terminated at fourth order. As suggested by Janssen *et al* [11], the inclusion of higher order terms and/or the introduction of more than two types of competing interaction might bring us closer to replicating the observed phase diagrams. Furthermore, for the emergence of SF modes which are canted, some form of magnetic anisotropy would seem a basic ingredient; indeed, while such states can display non-zero magnetization, the possibility that the latter could be characterized by a non-zero wavevector would also need to be considered. To reiterate, such possibilities remain to be verified by microscopic probes.

## 4. Summary and conclusions

In summary, over a portion of the $(H, T)$ plane, the ac susceptibility of a polycrystalline $Ce_2Fe_{17}$ sample is shown to exhibit a peak, the amplitude of which *increases* with increasing static biasing field over a small applied field range (0.6–1 T) between 90 and 190 K; such a behaviour was not previously reported to our knowledge. In an attempt to establish the origin for this effect, a phase diagram has been constructed from detailed magnetization data following the procedures outlined previously [11]. The latter indicates that fields in the range 0.6–1 T induce an SFI phase intermediate (in temperature) between a low temperature IFM region and a high temperature disordered paramagnetic phase, and a comparison of the magnetization and ac susceptibility data indicates that the region over which this field-induced increase in ac susceptibility peak amplitude occurs tracks the IFM–SFI boundary. This peak amplitude–field relationship is the inverse of that predicted and observed at conventional ferromagnetic to paramagnetic phase boundaries, and in (super)paramagnetic systems. Despite the polycrystalline nature of the present specimen, not only is its saturation moment close to that reported for single-crystal samples [13], but also its phase diagram exhibits the predominant features evident in those recently reported [11] for both undoped and Ta-doped single crystals.

This behaviour in $\chi(H, T)$ may be a defining characteristic of such transitions, specifically reflecting the unusual—possibly unique—combination of ferromagnetic and antiferromagnetic interactions in the $Ce_2Fe_{17}$ system that leads to the complicated influence of applied fields on the changing spin configurations along the IFM–SFI boundary. Certainly it is not a universal characteristic of systems with competing ferromagnetic and antiferromagnetic interactions, even when these lead to metamagnetic transitions [3, 4, 17]. The present results thus suggest further study to determine whether such a response is a universal feature along spin flop boundaries—or their analogue—occurring in other systems.

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