First-order metamagnetic transition in $CeFe_2$-based pseudobinary alloys

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First-order metamagnetic transition in $CeFe_2$-based pseudobinary alloys

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Abstract. We present results of a direct-current magnetization study showing that the low-temperature antiferromagnetic state in various $CeFe_2$-based pseudobinary alloys can be transformed into a ferromagnetic state through a magnetic field-induced phase transition. We highlight the presence of hysteresis and phase coexistence across this metamagnetic transition and argue that the observed phase transition is first order in nature.

1. Introduction

The interesting magnetic properties of the C15–Laves phase compound $CeFe_2$ [1–6] and its pseudobinaries [7–20] have been attracting attention almost continuously for the last twenty years. Most of the studies are mainly focused on the proper understanding of the magnetic ground states of the parent as well as the pseudobinary compounds, and less emphasis is given to the exact nature of the phase transitions. In a recent study [21] we have addressed this latter question in the context of double magnetic transitions in $CeFe_2$-based pseudobinary systems. With temperature-dependent ac susceptibility measurements we have shown that while the paramagnetic-to-ferromagnetic transition (as a function of decreasing temperature) is a second-order phase transition, the lower-temperature ferromagnetic-to-antiferromagnetic transition carries the signature of a first-order phase transition. The presence of thermal hysteresis and phase coexistence across this ferromagnetic-to-antiferromagnetic transition was highlighted [21]. With the existing knowledge [13, 15, 17–20] that the lower-temperature antiferromagnetic state can be caused to revert back to the ferromagnetic state with the application of an external magnetic field, the question naturally arises as to what the nature of this field-induced antiferromagnetic-to-ferromagnetic transition is. In this paper we address this question in detail through our dc magnetization measurements. We shall argue that this metamagnetic transition is first order in nature.

2. Experimental procedure

We have used in the present study the same two samples—Ce(Fe, 5% Ir)$_2$ and Ce(Fe, 7% Ru)$_2$ compounds—as were used in our earlier measurements [21]. The dc magnetization is measured using a commercial SQUID magnetometer (Quantum Design-MPMS5). We have used a scan length of 2 cm, with each scan containing 32 data points. The 2 cm scan length is used to ensure minimum sample movement in the inhomogeneous magnetic field of the superconducting

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magnet. This magnetic field inhomogeneity in a 2 cm scan is about 1 Oe in an applied field of 20 kOe [22].

3. Results and discussion

In figure 1 we present magnetization (M) versus temperature (T) plots for the Ce(Fe, 5% Ir)$_2$ and Ce(Fe, 7% Ru)$_2$ samples obtained in an applied field of 100 Oe. The double magnetic transitions are clearly visible and the transition temperatures agree well with those obtained earlier in ac susceptibility measurements [21]. The data shown in figure 1 are obtained while warming up unidirectionally from low temperatures after zero-field cooling. We have also obtained data while cooling and a distinct thermal hysteresis of width 5 K is observed across the ferromagnetic-to-antiferromagnetic transition. This is to be contrasted with the relatively small thermal hysteresis of 2 K obtained earlier in the ac susceptibility measurements [21]. We note that a smaller hysteresis is expected in an ac measurement because the ac field assists a metastable-to-stable-state transformation. Our SQUID magnetometer cannot, however, monitor possible temperature lags between the sample and the sensor—as we were able to do in our ac measurements [21]. For this reason we shall not emphasize thermal hysteresis in this report.

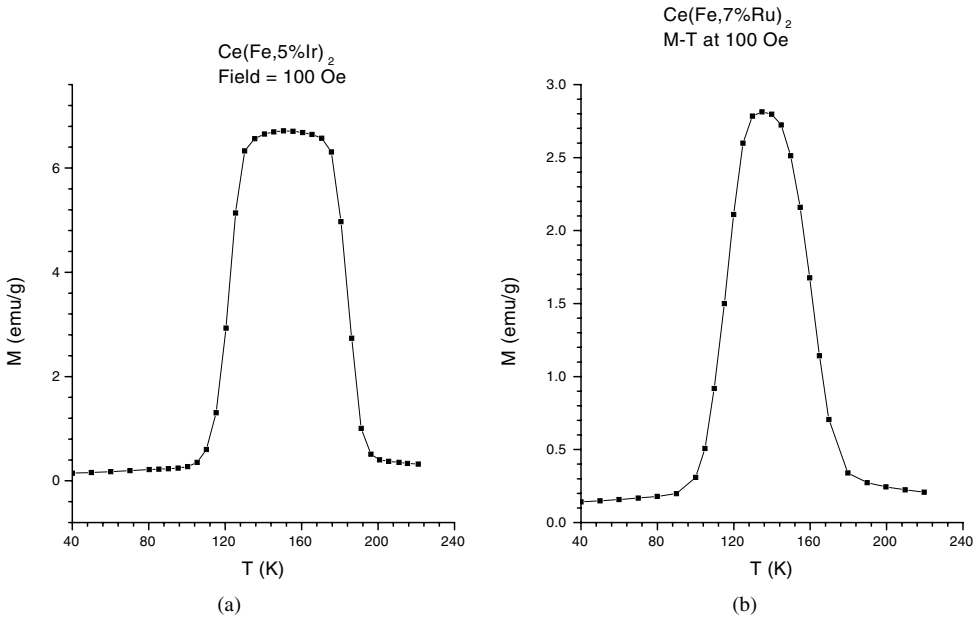


Figure 1. Magnetization versus temperature plots for (a) Ce(Fe, 5% Ir)$_2$ and (b) Ce(Fe, 7% Ru)$_2$.

In figure 2 we present isothermal magnetization (M) versus field (H) plots for Ce(Fe, 5% Ir)$_2$ at various temperatures obtained after zero-field cooling the sample from a temperature above the Curie temperature ($T_C \approx 185$ K). Above 130 K the M–H plot shows the typical behaviour of a ferromagnet, reaching technical saturation by 10 kOe at 150 K. Below 80 K, although there is a small non-linearity in the low-field ($H < 5$ kOe) regime, the characters of the M–H plots are drastically different. This we attribute to the antiferromagnetic nature of the low-temperature phase. The M–H behaviour in the T-range 130 K $> T >$ 80 K, however, is quite interesting (see figure 2). While in the low-field regime there is a distinct

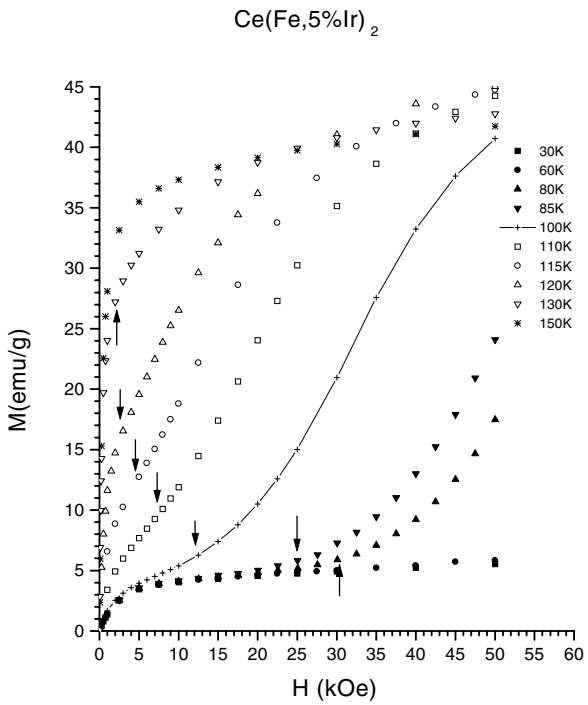


Figure 2. Magnetization versus field plots for Ce(Fe, 5% Ir)$_2$ at various temperatures. The arrows mark the onset field H_M of the metamagnetic transition. The line for the M–H curve at $T = 100$ K serves as a guide to the eye.

deviation from the characteristics of the higher-temperature ferromagnetic state, a sharp rise in M occurs in the high-H regime indicative of a field-induced ferromagnetic transition or a metamagnetic transition. We mark the onset field of this metamagnetic transition as H_M. H_M is estimated as the field at which the M–H curve changes curvature from convex to concave, i.e. where $\mathrm{d}M/\mathrm{d}H$ shows a minimum. With the decrease in T the value of H_M increases and goes beyond the range of existing field strength (55 kOe) in our SQUID magnetometer by 60 K.

We have obtained qualitatively similar results from the isothermal field dependence of the magnetization at various temperatures for the Ce(Fe, 7% Ru)$_2$ sample measured after zero-field cooling the sample from a temperature above the Curie temperature ($T_C \approx 165$ K). These M–H plots are shown in figure 3.

We shall now focus on this metamagnetic transition, and look for signatures typically associated with a first-order phase transition, namely hysteresis and phase coexistence. The control variable inducing the transition is the magnetic field. We shall work in the temperature ranges 80 K $\leqslant T \leqslant$ 130 K for Ce(Fe, 5% Ir)$_2$ and 90 K $\leqslant T \leqslant$ 130 K for Ce(Fe, 7% Ru)$_2$, so that the metamagnetic transition remains clearly visible within the upper limit of our magnetic field range. We present in figures 4, 5 M–H curves obtained in the ascending- and descending-field cycles for both Ce(Fe, 5% Ir)$_2$ and Ce(Fe, 7% Ru)$_2$, showing distinct hysteresis associated with the metamagnetic transition. A sharp rise in magnetization accompanied by hysteresis is traditionally attributed to the first-order magnetic process [23, 24]. However, it can still be argued that the observed hysteresis may be an intrinsic property of the field-induced ferromagnetic state and originate from the domain wall pinning and/or freezing of domain

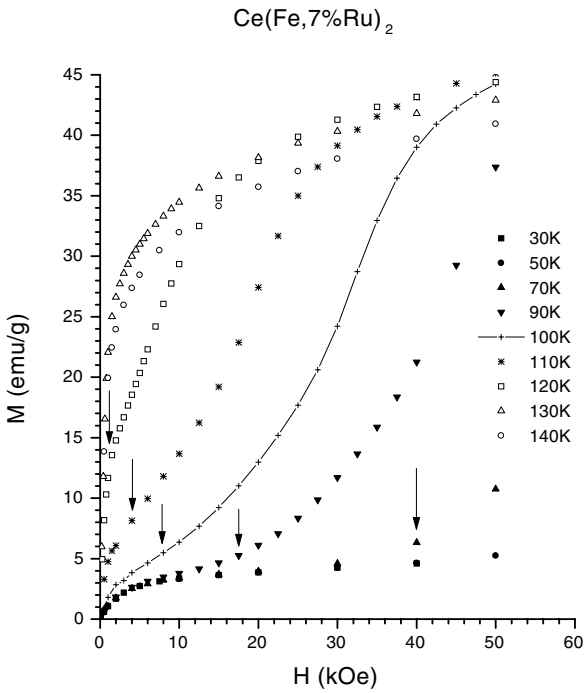


Figure 3. Magnetization versus field plots for Ce(Fe, 7% $Ru)_2$ at various temperatures. The onset field H_M of the metamagnetic transition is marked by arrows. The line for the M–H curve at $T = 100$ K serves as a guide to the eye.

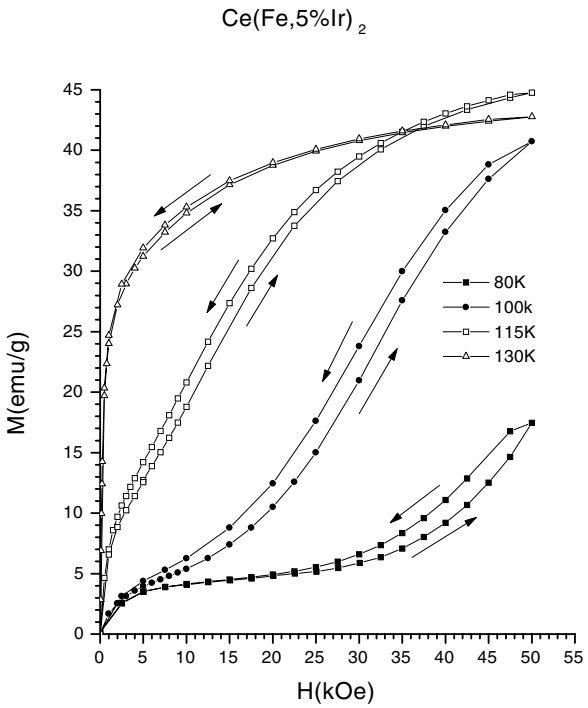


Figure 4. M–H curves for Ce(Fe, 5% $Ir)_2$ showing hysteresis associated with the metamagnetic transition. The arrows show the direction of field change.

rotation [25]. To exclude these possibilities we have measured carefully the M–H curves for both of the samples in the temperature regime where the ground state is ferromagnetic at all

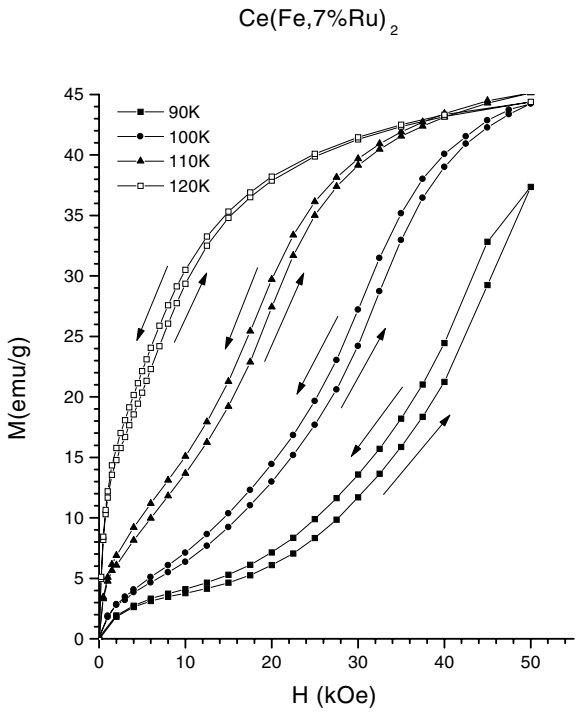


Figure 5. M–H curves for Ce(Fe, 7% Ru)$_2$ showing hysteresis associated with the metamagnetic transition. The arrows show the direction of field change.

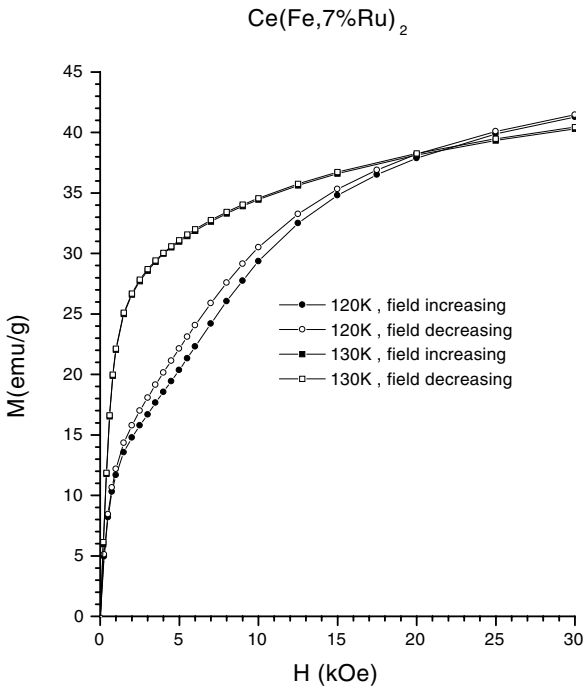
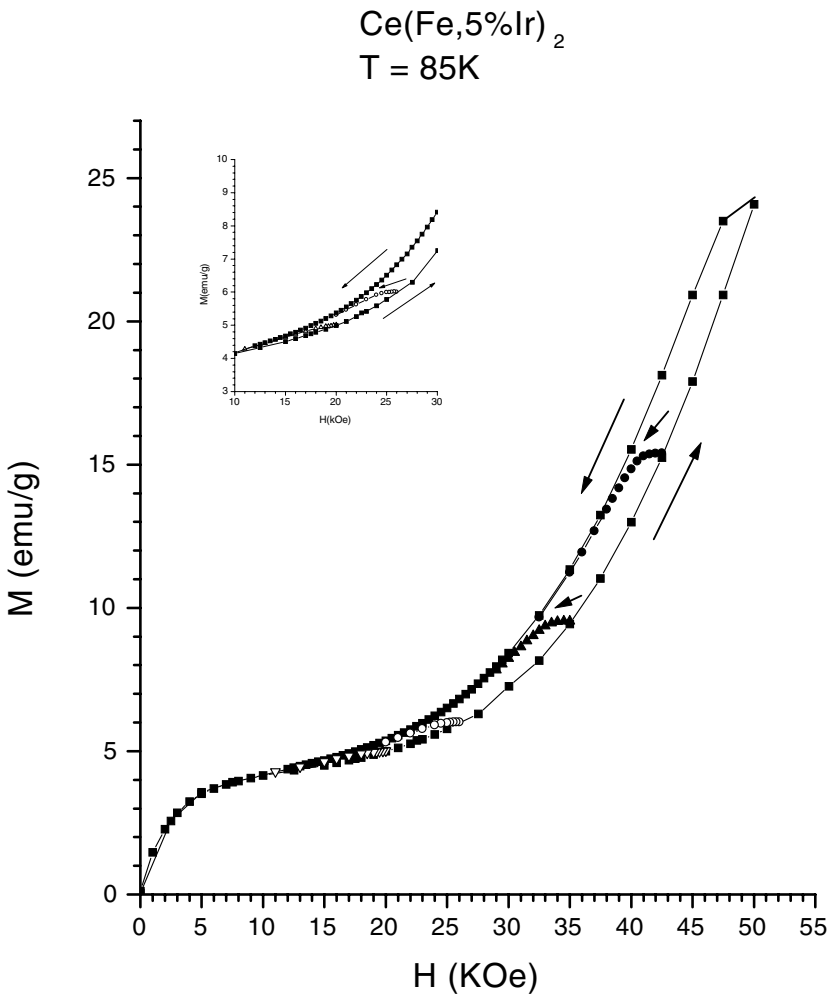


Figure 6. M–H curves for Ce(Fe, 7% Ru)$_2$ at temperatures of 120 K and 130 K. See the text for details.

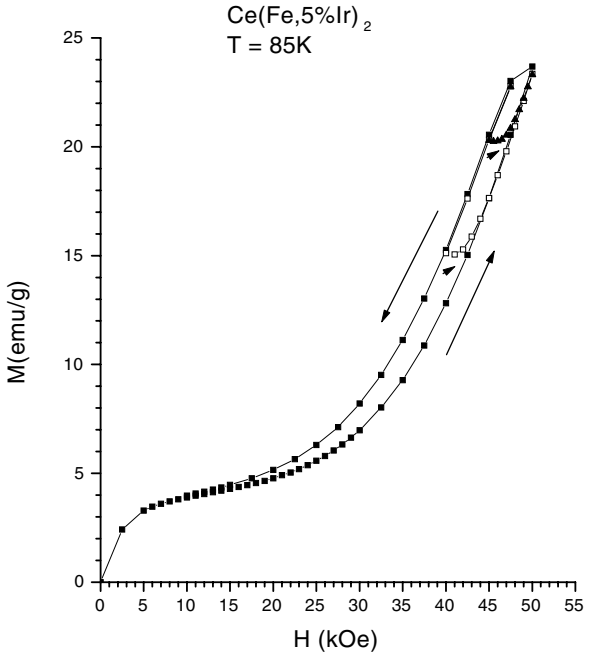
H-values. M–H curves in the ferromagnetic regime show negligibly small hysteresis with the coercivity field (H_C) of the order of 5 Oe. To show the contrast of the hysteresis intrinsic to the ferromagnetic regime of these samples with the hysteresis associated with the metamagnetic



(a)

Figure 7. Minor hysteresis loops generated during: (a) the ascending-field cycle for Ce(Fe, 5% Ir)$_2$ at $H = 20$ kOe (open triangles), $H = 26$ kOe (open circles), $H = 35$ kOe (solid triangles) and $H = 42.5$ kOe (solid circles); (b) the descending-field cycle for Ce(Fe, 5% Ir)$_2$ at $H = 40$ kOe (open squares) and $H = 45$ kOe (solid triangles). The envelope curve is represented by solid squares. The measurements were done at $T = 85$ K. The inset shows an expanded view.

transition, in figure 6 we present M–H curves for Ce(Fe, 7% Ru)$_2$, measured at $T = 130$ K and 120 K. (The ferromagnetic-to-antiferromagnetic transition temperature for this sample is approximately 127 K (see figure 1(b)). At $T = 120$ K, the onset of the metamagnetic transition takes place at a relatively small value of $H_M \approx 1.5$ kOe. The hysteresis associated with the metamagnetic transition shows up as a distinct bubble in the field range 4 kOe $< H <$ 30 kOe, with relative reversibility in the field regime above (see figure 5) and below. This is to be contrasted with the M–H curve at $T = 130$ K which is quite reversible (on the same scale) for all H-values of the measurement (see figure 6). We have also checked the field history dependence of the hysteresis associated with the metamagnetic transition by cycling the field isothermally (after initial zero-field cooling) more than once between zero and the maximum



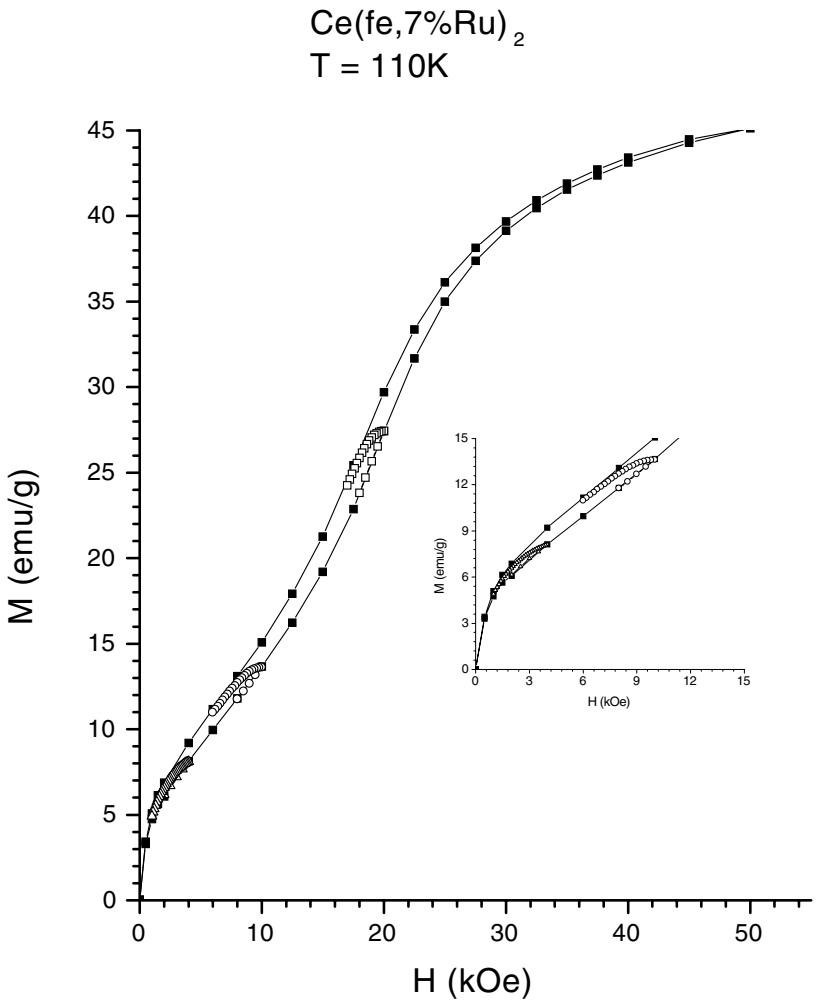
(b)

Figure 7. (Continued)

applied field (50 kOe). Unlike in the case of ferromagnetic hysteresis loops, no virgin curve is observed here, and the hysteresis loop obtained in the first field cycle is reproduced in all subsequent cycles.

While the hysteresis associated with the ferromagnetic state is relatively small and does not change much in the temperature range 130 K $< T <$ 160 K (measured but not shown here for the sake of conciseness), the hysteresis associated with the metamagnetic transition is observed below 130 K and grows relatively rapidly with the decrease in T. It should be noted here that at $T = 80$ K for Ce(Fe, 5% Ir)$_2$ and at $T = 90$ K for Ce(Fe, 7% Ru)$_2$ the formation of the higher-field ferromagnetic state is probably not completed by 50 kOe, and accordingly the magnetization, and the associated hysteresis, has not reached saturation (see figures 4 and 5).

Having established the hysteretic nature of the metamagnetic transition, we shall now look for phase coexistence in the transition region. To study phase coexistence we use the technique of minor hysteresis loops (MHLs) [21,26]. We shall first define the hysteretic M–H curve obtained by isothermal field cycling between 0 and $H_{max} = 50$ kOe as the 'envelope curve'. The field-increasing curve (0 to 50 kOe) corresponds to the antiferromagnetic phase transforming to the ferromagnetic phase, with the antiferromagnetic phase persisting as a metastable phase over some field region. Similarly the field-decreasing (50 kOe to 0) curve corresponds to the ferromagnetic phase transforming to the antiferromagnetic phase with the ferromagnetic phase persisting as a metastable phase over some field region. We can now generate an MHL during the ascending-field cycle, i.e. start increasing H from the lower-field reversible (antiferromagnetic) regime and then reverse the direction of change of H before reaching the higher-field saturation magnetization regime. We can also produce an MHL in the descending-H cycle, i.e. start decreasing H from the saturation magnetization regime and reverse the direction of change of H before reaching the low-H antiferromagnetic

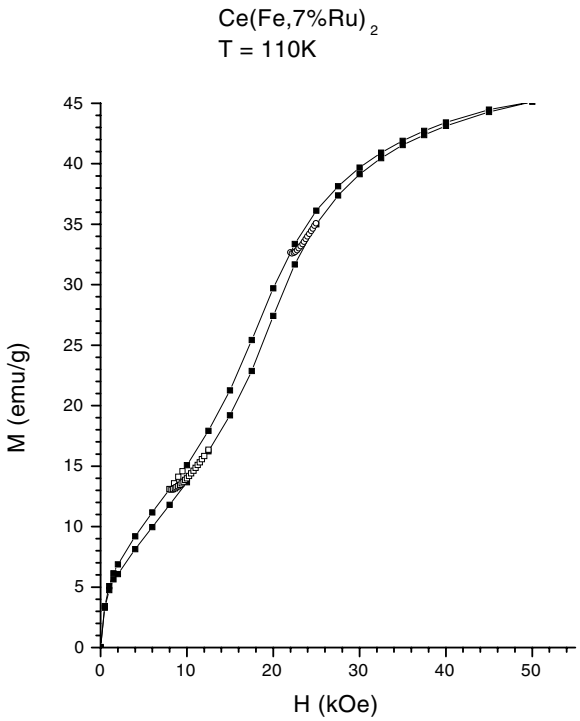


(a)

Figure 8. Minor hysteresis loops generated during: (a) the ascending-field cycle for Ce(Fe, 7% Ru)$_2$ at $H = 4$ kOe (open triangles), $H = 10$ kOe (open circles) and $H = 20$ kOe (open squares); (b) the descending-field cycle for Ce(Fe, 7% Ru)$_2$ at $H = 8$ kOe (open squares) and $H = 22$ kOe (open circles). The envelope curve is represented by solid squares. The measurements were done at $T = 110$ K. The inset shows an expanded view.

regime. We show in figures 7 and 8 examples of these MHLs for Ce(Fe, 5% Ir)$_2$ and Ce(Fe, 7% Ru)$_2$ samples.

At field values close to H_M in the ascending-field cycle, the high-field ferromagnetic phase is not expected to be formed in sufficient quantity, and complete transformation occurs only at much higher fields. When we initiate an MHL from a field close to H_M, this partially formed ferromagnetic phase 'supercools' and persists as a metastable phase. We see only a small amount of hysteresis as H is lowered from field values close to H_M (see the insets of figures 7(a) and 8(a)). At fields well above H_M a much larger fraction (close to 100%) of the sample has transformed to the ferromagnetic phase. When we now lower H and initiate an MHL, the entire sample 'supercools' in the ferromagnetic phase, and the hysteresis observed



(b)

Figure 8. (Continued)

should be much larger. This is brought out in figures 7 and 8 where MHLs initiated from field values well inside the hysteretic regime coincide with the upper envelope curve, indicating that the high-field ferromagnetic phase has formed in sufficient quantities. To show further evidence of supercooling of the high-field phase, in figure 9 we show MHLs at $H = 2$ kOe drawn from the lower envelope curve and at $H = 1.6$ kOe drawn from the upper envelope curve of the Ce(Fe, 7% Ru)$_2$ sample at $T = 110$ K. These field values are chosen such that the lower and upper envelope curves have the same magnetization value. Note that $H = 2$ kOe is lower than the estimated $H_M \approx 4$ kOe and accordingly the MHL drawn from the lower envelope curve shows almost no irreversibility. The high-field ferromagnetic phase is thus not yet formed. On the other hand the MHL drawn from the upper envelope curve at a lower-field value of 1.6 kOe shows distinct irreversibility. This clearly shows that the high-field phase persists (as a metastable 'supercooled' phase) in this field regime in the descending-field cycle. There are similar results for the Ce(Fe, 5% Ir)$_2$ sample, but they are not shown here for the sake of conciseness.

It is to be noted here that the low-field magnetic response of the low-temperature (supposedly) antiferromagnetic state is for both of the samples quite non-linear in nature (see figures 2 and 3). This behaviour definitely indicates the presence of some ferromagnetic correlation in this low-temperature phase. While it was pointed out earlier that such a behaviour probably arose due to an impurity ferromagnetic phase [20], an intrinsic origin of such a behaviour cannot be ruled out entirely [19]. Careful microscopic measurements (such as neutron diffraction and/or Mössbauer measurements) are now necessary to resolve this problem for the $CeFe_2$-based pseudobinaries.

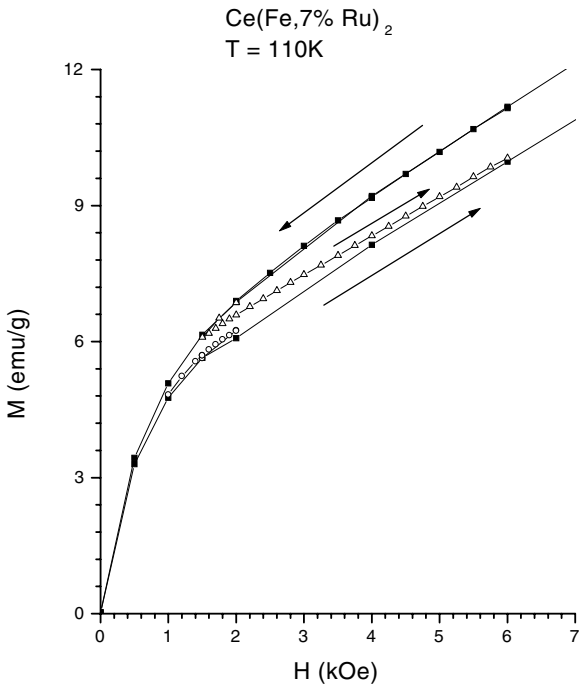


Figure 9. Comparison of minor hysteresis loops generated during the ascending-field cycle ($H = 2$ kOe—open circles) and descending-field cycle ($H = 1.6$ kOe—open triangles) at approximately the same values of the magnetization for Ce(Fe, 7% Ru)$_2$ at $T = 110$ K. The envelope curve is represented by solid squares. See the text for details.

4. Conclusions

Summarizing our results, we can say that the field-induced antiferromagnetic-to-ferromagnetic transition in Ce(Fe, 5% Ir)$_2$ and Ce(Fe, 7% Ru)$_2$ samples is accompanied by field hysteresis as well as phase coexistence. These are the typical characteristics of a first-order transition. Hence we argue that the observed metamagnetic transition in these $CeFe_2$-based pseudobinaries is a first-order transition. The present study complements our earlier study of temperature variation in the same systems, and establishes the existence of a first-order ferromagnetic-to-antiferromagnetic phase transition in the $CeFe_2$-based systems in a more general H–T plane.

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