First order metamagnetic transition in $CeFe_2$ based pseudobinary alloys

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Abstract

We present results of dc magnetisation study showing that the low temperature antiferromagnetic state in various $CeFe_2$-based pseudobinary alloys can be transformed into 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 of first order in nature.

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I. INTRODUCTION

The interesting magnetic properties of the C15-Laves phase compound $CeFe_2$ [1–6] and its pseudobinaries [7–20] have been drawing almost continuous attention during last twenty years. Most of these studies are mainly focussed on the proper understanding of the magnetic ground states of the parent as well as the pseudobinary compounds, and less emphasis is given on 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 the 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 information [13,15,17–20] that the lower temperature antiferromagnetic state can be reverted back to the ferromagnetic state with the application of an external magnetic field, the question naturally arises - what is the nature of this field induced antiferromagnetic to ferromagnetic transition ? In this paper we address this question in details through our dc magnetisation measurements. We shall argue that this metamagnetic transition is of first order in nature.

II. EXPERIMENTAL

We have used in the present study the same two samples – $Ce(Fe,5\%Ir)_2$ and $Ce(Fe,7\%Ru)_2$– used in our earlier measurements [21]. Dc magnetisation is measured using a commercial SQUID magnetometer (Quantum Design-MPMS5). We have used a scan length of 2cm, with each scan containing 32 data points. The 2cm scan length is used to ensure minimum sample movement in the inhomogeneous magnetic field of the superconducting magnet. This magnetic field inhomogeneity in a 2 cm scan is about 1 Oe in an applied field of 20 kOe [22].

III. RESULTS AND DISCUSSION

In Fig. 1 we present magnetisation (M) versus tempearture (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 are well in accord with those obtained earlier in ac-suceptibility measurements [21]. The data shown in Fig. 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 smaller thermal hysteresis of 2 K obtained earlier in the ac-suceptibility 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.

In Fig. 2 we present isothermal magnetisation (M) versus field (H) plots for Ce(Fe,5%Ir)$_2$ at various temperatures obtained after zero field cooling the sample from tempearture above the Curie tempeature (T$_C$ ≈185K). Above 130 K the M-H plot shows the typical behaviour of a ferromagnet, reaching technical saturation by 10 kOe at 150K. Below 80K, although there is a small non-linearity in the low field (H < 5 kOe) regime the character 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 regime 130K $> T >$ 80K, however, is quite interesting (see Fig. 2) . While in the low field regime there is a distinct deviation from 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 dM/dH 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 60K.

We have obtained qualitatively similar results from the isothermal field dependence of magnetisation at various temperatures for the $Ce(Fe,7\%Ru)_2$ sample measured after zero field cooling the sample from temperature above Curie temperature (T_C ≈165K). These M-H plots are shown in Fig. 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 magnetic field. We shall work in the temperature regime, 80K$\leq T \leq$130K for $Ce(Fe,5\%Ir)_2$ and 90K$\leq T \leq$130K 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 Fig.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 magnetisation accompanied by hysteresis is traditionally attributed to the first order magnetic process [24]. However, it can still be argued that the observed hysteresis may be the intrinsic property of the field induced ferromagnetic state and originates from the domain wall pinning and/or freezing of domain rotation [25]. To negate these possibilities we have measured carefully the M-H curves for both the samples in the temperature regime where the ground state is ferromagnetic at all 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 transition, in Fig. 6 we present M-H curves for $Ce(Fe,7\%Ru)_2$, measured at T=130 K and 120K. (The ferro- to antiferromagnetic transition temperature for this sample takes place approximately at 127 K (see Fig. 1 (b)). At T = 120K, the onset of the metamagnetic transtion takes place at a relatively small value of H_M ≈1.5kOe. The hysteresis associated with the metamagnetic transition shows up as a distinct bubble in the field regime 4kOe$< H <$30kOe, with relative reversibility in the field regime above (see fig.5) and below. This is to be contrasted with the M-H curve at T=130K which is quite reversible (in the same scale) for all H values of measurement (see Fig.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 applied field (50 kOe). Unlike in the case of ferromagnetic hysteresis loops, no virgin curve is observed here, and the obtained hysteresis loop in the first field cycle is reproduced in all the subsequent cycles.

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

After establishing the hysteretic nature of the metamagnetic transition, we shall now look for the phase coexistence in the transition region. To study the phase co-existence 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 magnetisation regime. We can also produce an MHL in the descending H cycle i.e. start decreasing H from the saturation magnetisation regime and reverse the direction of change of H before reaching the low H antiferromagnetic regime. We show in Fig. 7 and 8 examples of these MHLs in $Ce(Fe,5\%Ir)_2$ and $Ce(Fe,7\%Ru)_2$ samples.

At field values close to H_M on the ascending field cycle, the high field ferromagnetic phase is not expected to be formed in sufficient quantitiy, 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 small amount of hysteresis as H is lowered from field values close to H_M (see inset of fig. 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 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 fig. 9 we show MHLs at H = 2kOe drawn from the lower envelope curve and at H = 1.6kOe drawn from the upper envelope curve of the $Ce(Fe,7\%Ru)_2$ sample at T = 110K. These field values are chosen such that the lower and upper envelope curves have the same magnetisation value. Note that H= 2 kOe is lower than the estimated H_M ≈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. Similar results exist for the $Ce(Fe,5\%Ir)_2$ sample also, but 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 for both the samples is quite non-linear in nature (see Fig. 2 and 3). This behaviour definitely points out 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 (like neutron diffraction and/or Mossbauer measurements) are now necessary to resolve this problem of the $CeFe_2$-based pseudobinaries.

IV. CONCLUSION

Summarising our results we 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 compliments 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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FIGURES

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

FIG. 2. Magnetisation 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=100K serves as a guide to the eye.

FIG. 3. Magnetisation versus field plots for Ce(Fe,5%Ir)$_2$ at various temperatures. The onset field H_M of metamagnetic transition is marked by arrows.The line for the M-H curve at T = 100K serves as a guide to the eye.

FIG. 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.

FIG. 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.

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

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

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

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

Fig. 1(a)

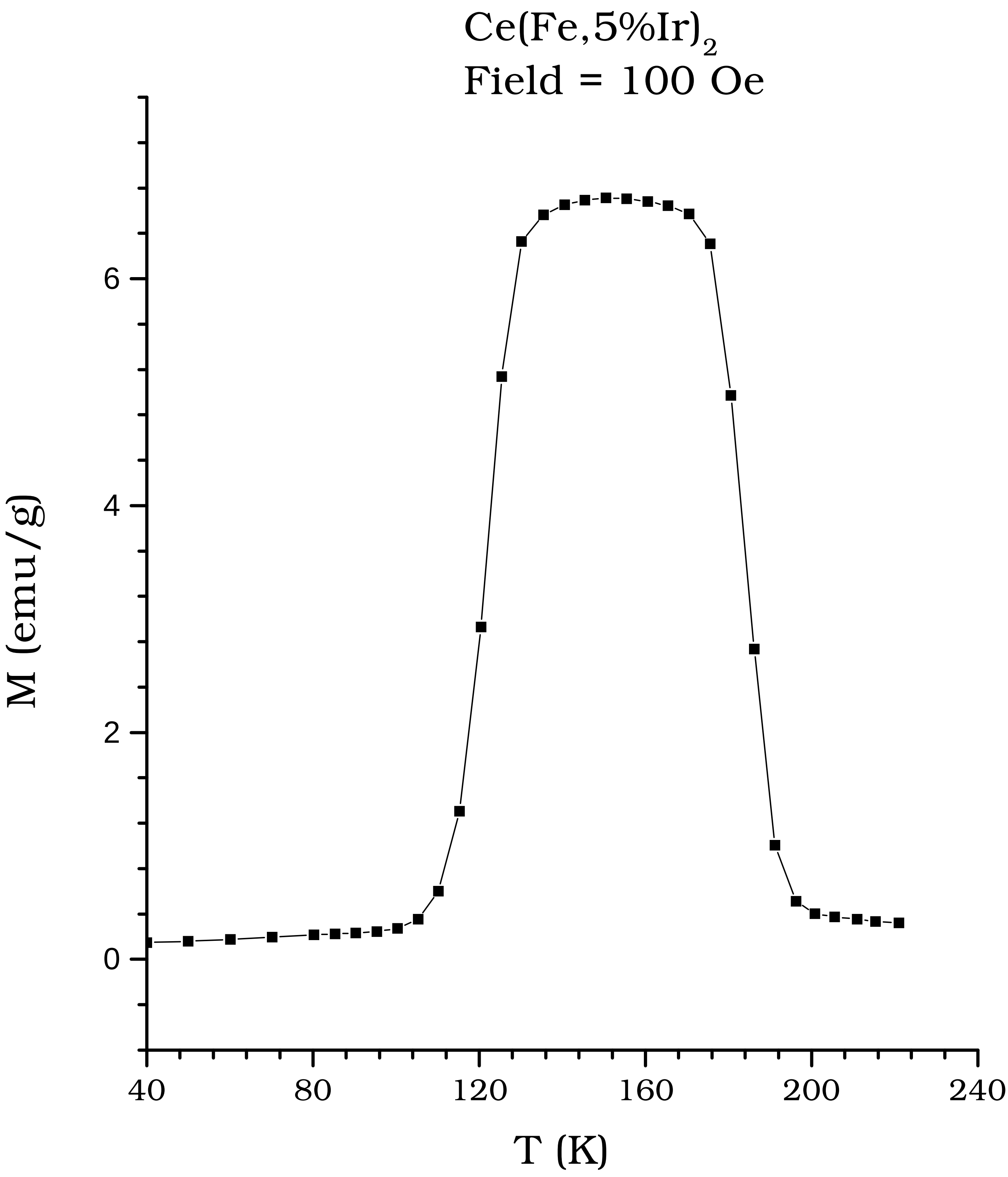


Fig. 1(b)

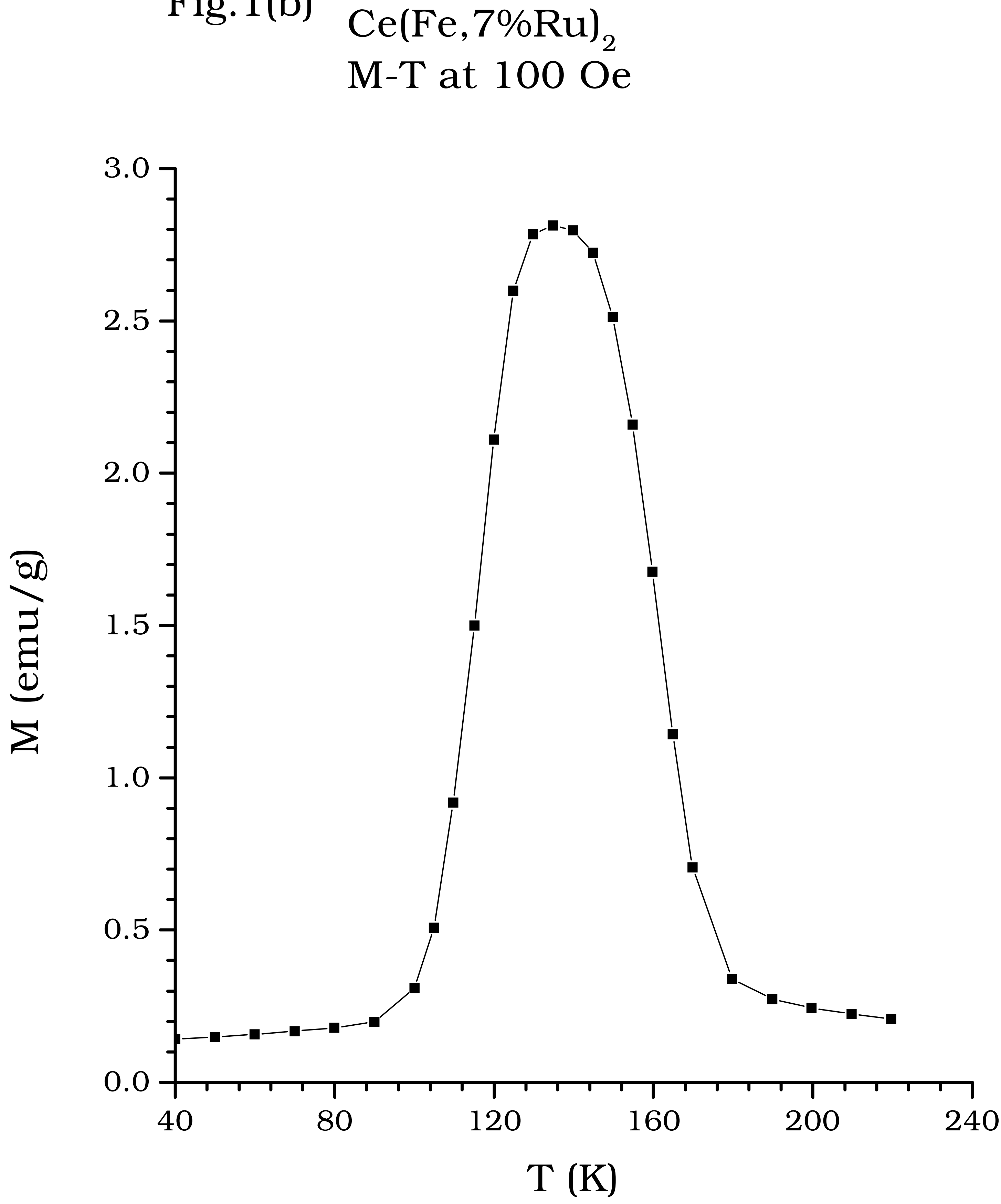
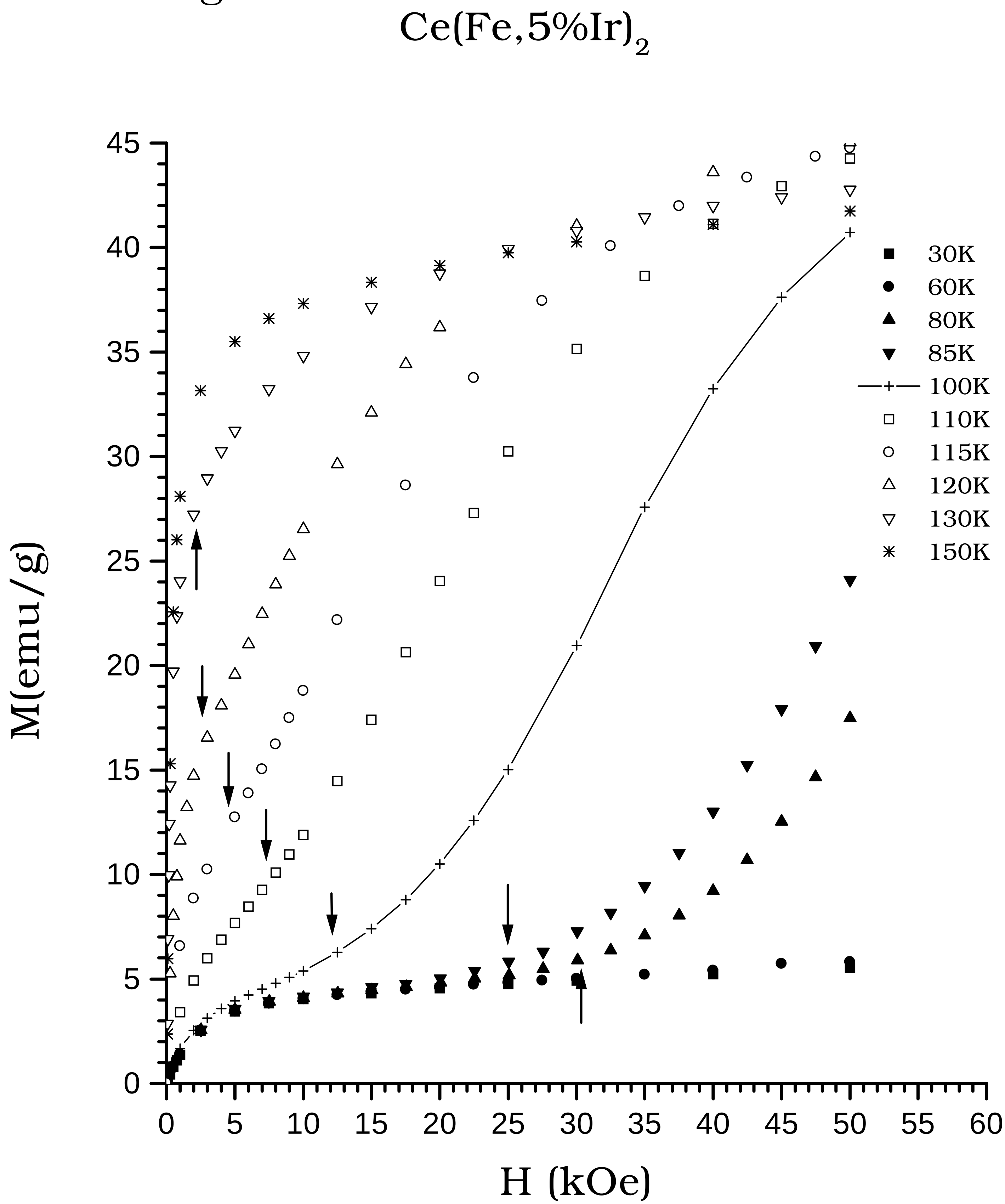
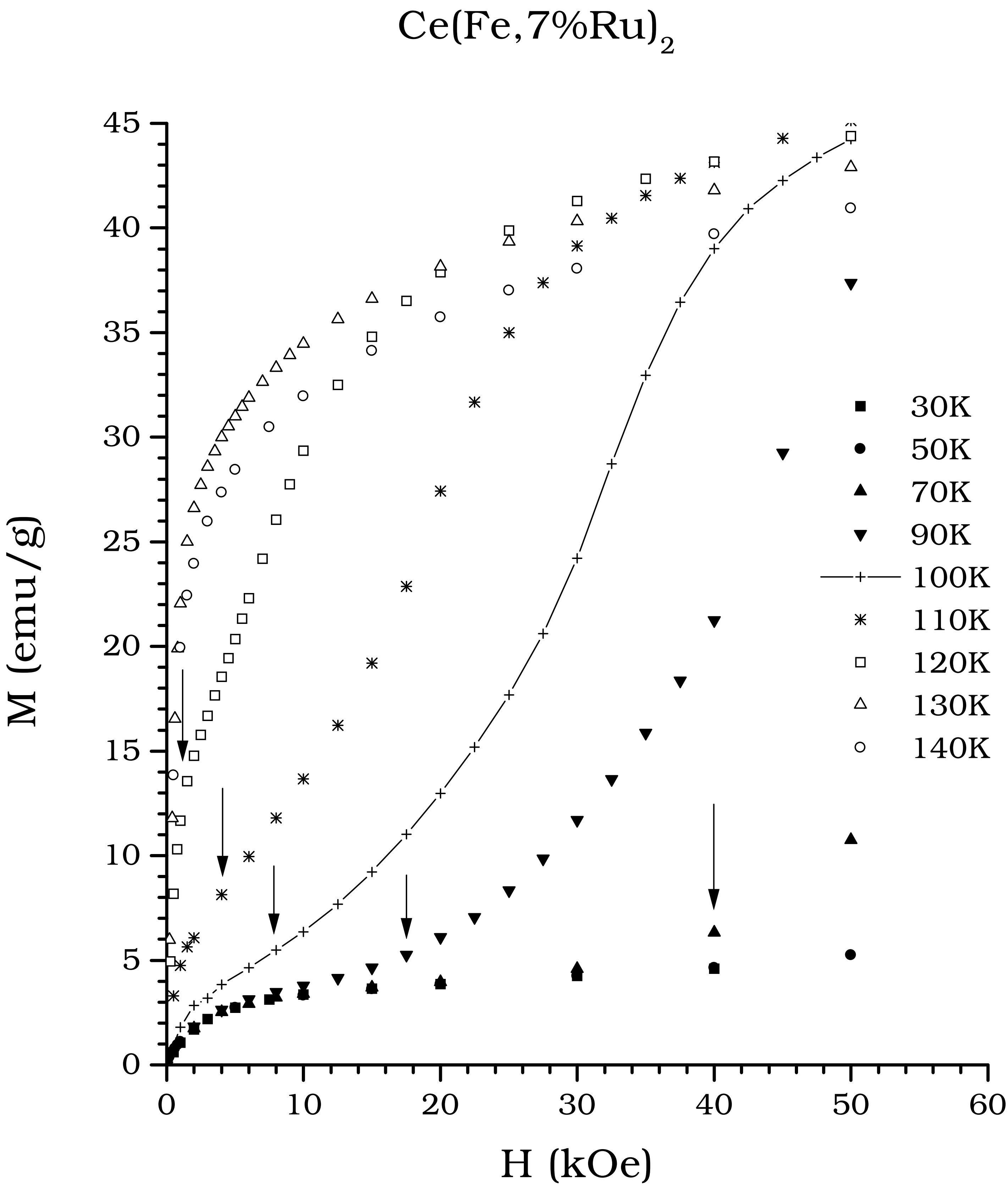

Ce(Fe,7%Ru)2
M-T at 100 Oe
M (emu/g)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
40
80
120
160
200
240
T (K)

Fig.2



Ce(Fe,5%Ir)₂
M(emu/g)
H (kOe)
0
5
10
15
20
25
30
35
40
45
50
55
60
30K
60K
80K
85K
100K
110K
115K
120K
130K
150K

Fig.3



Ce(Fe,7%Ru)$_2$
M (emu/g)
H (kOe)
0
5
10
15
20
25
30
35
40
45
0
10
20
30
40
50
60
30K
50K
70K
90K
100K
110K
120K
130K
140K

Fig.4

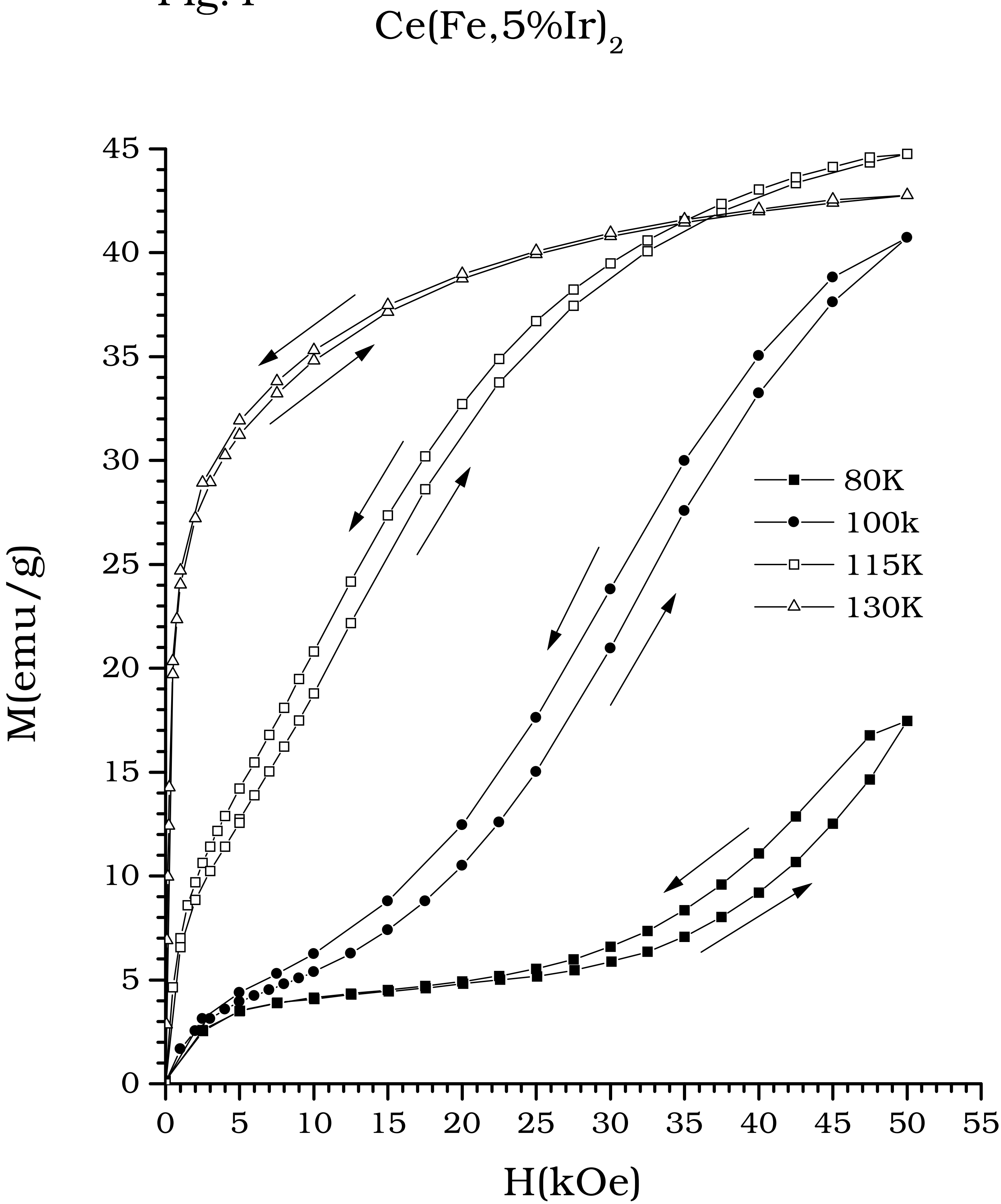
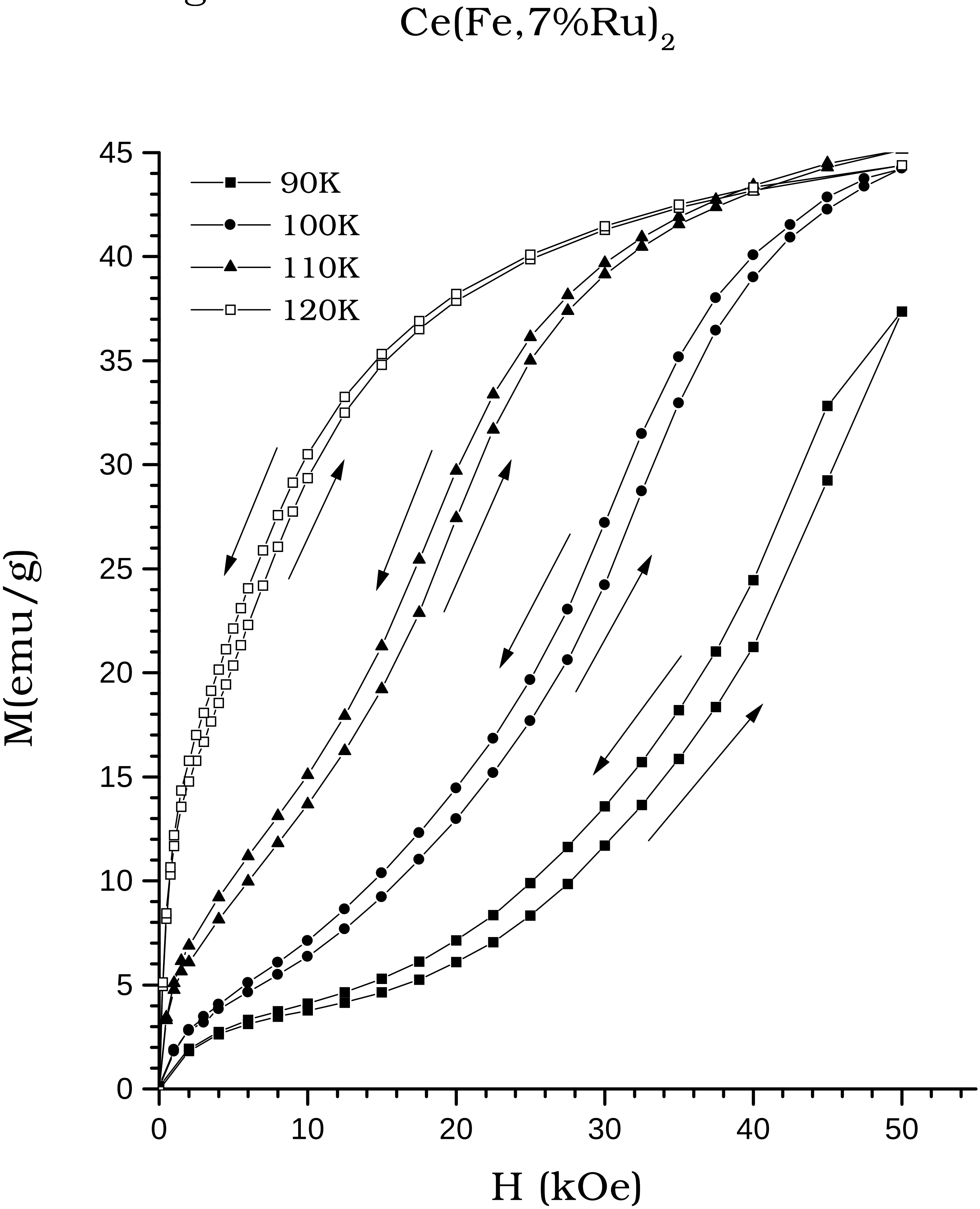

Ce(Fe,5%Ir)2
M(emu/g)
H(kOe)
80K
100k
115K
130K
0
5
10
15
20
25
30
35
40
45
50
55

Fig.5



Ce(Fe,7%Ru)$_2$
90K
100K
110K
120K
M(emu/g)
H (kOe)
0
5
10
15
20
25
30
35
40
45
0
10
20
30
40
50

Fig.6

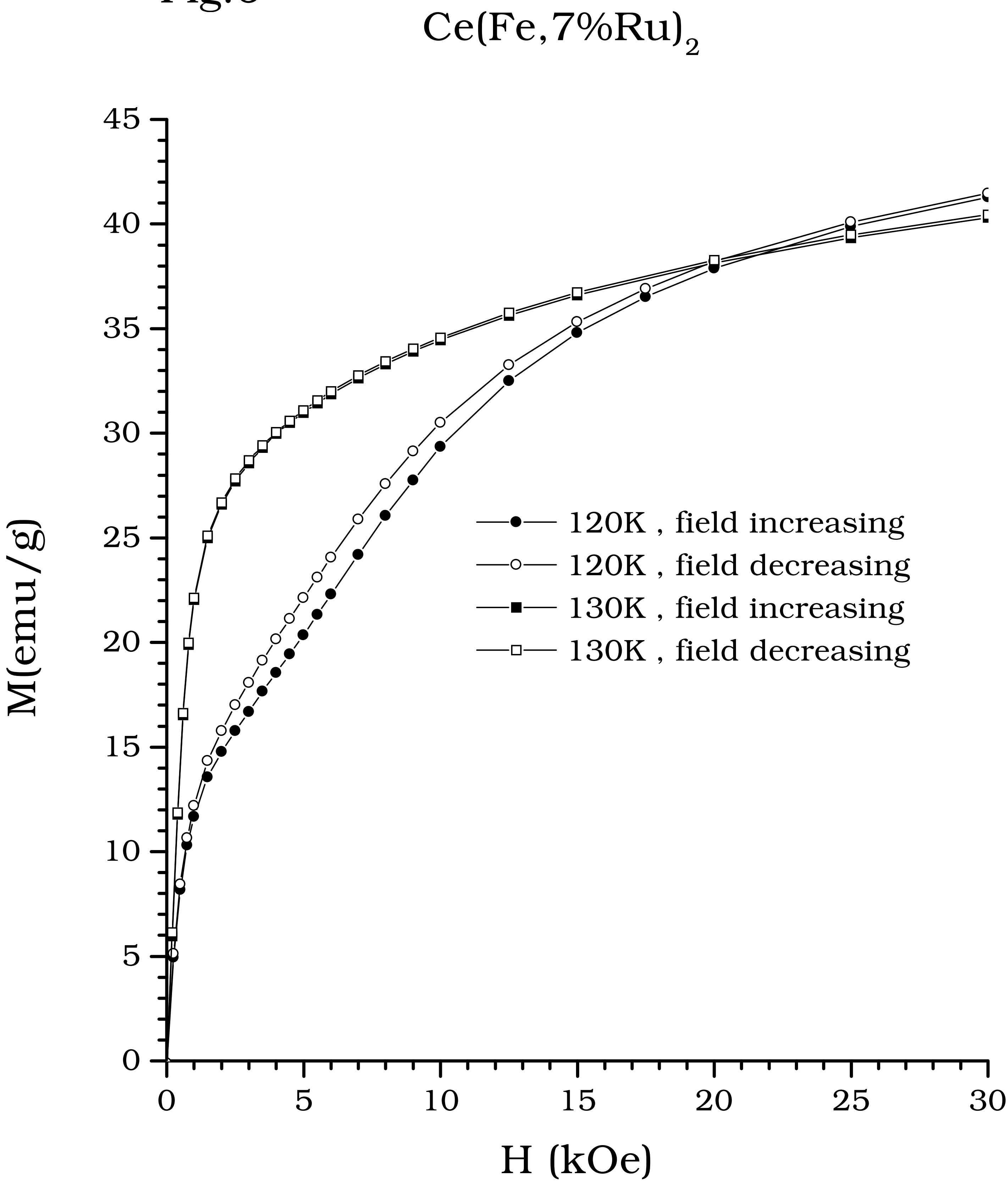
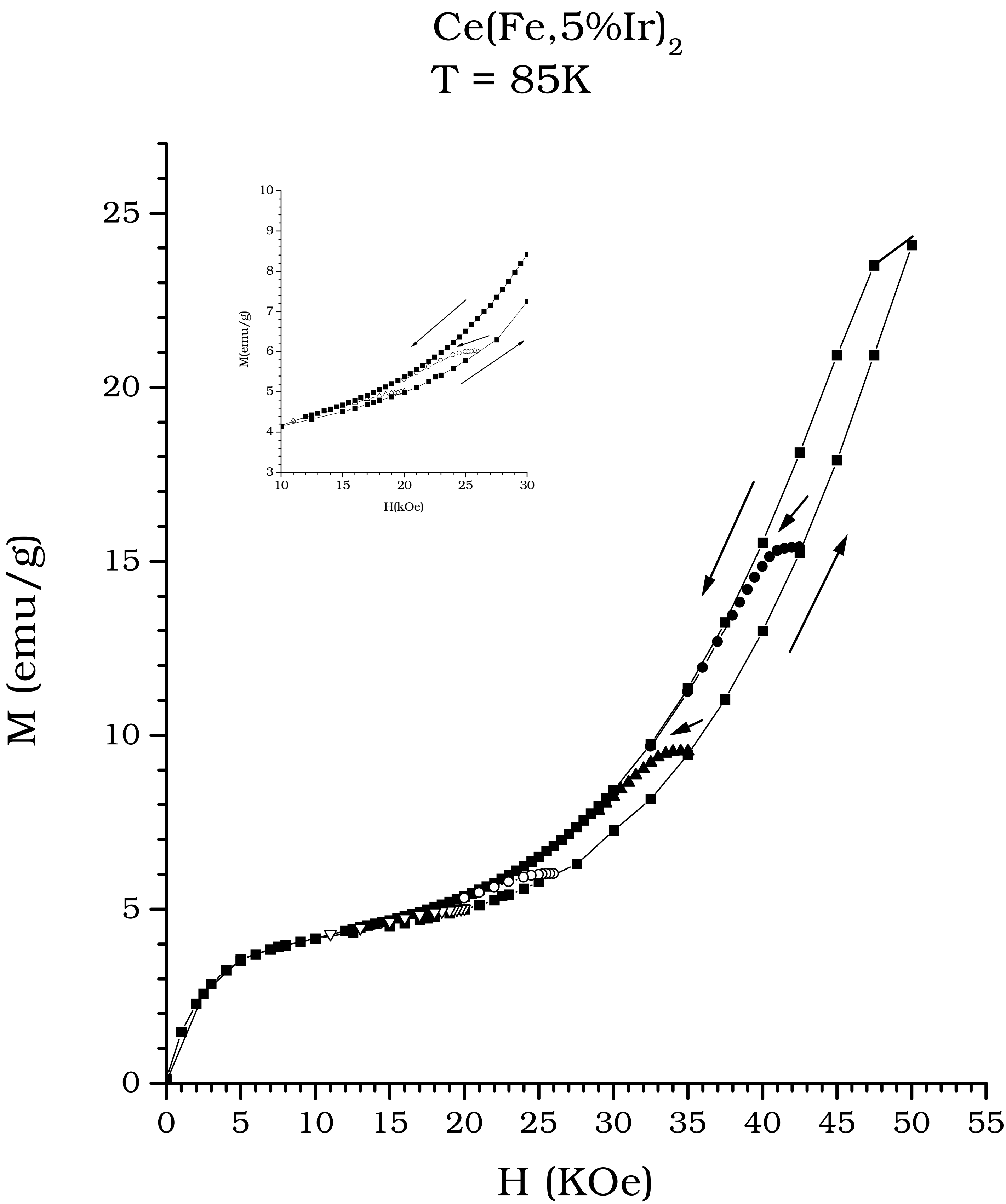
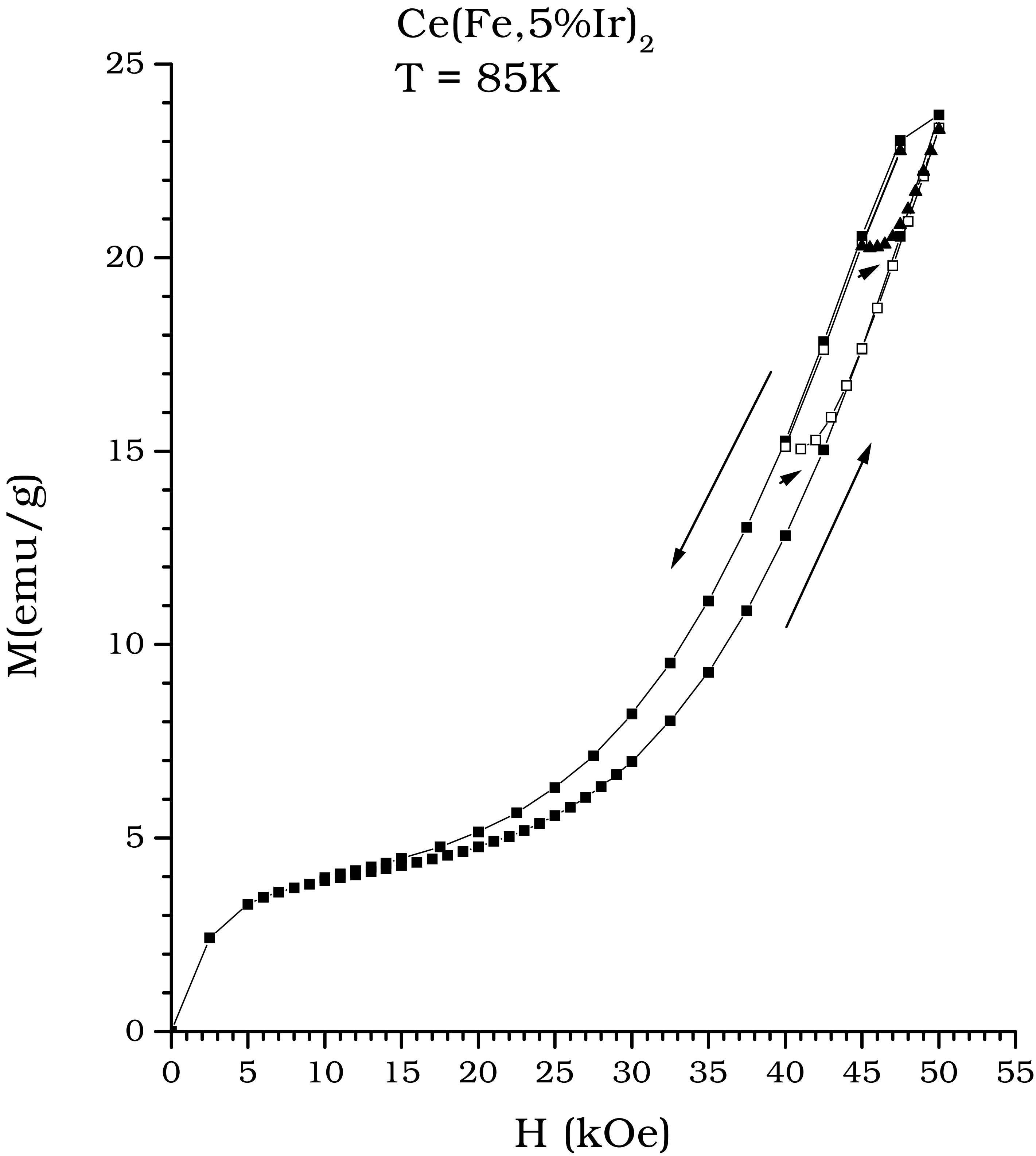

Ce(Fe,7%Ru)2
M(emu/g)
H (kOe)
0
5
10
15
20
25
30
35
40
45
120K , field increasing
120K , field decreasing
130K , field increasing
130K , field decreasing

Fig. 7(a)



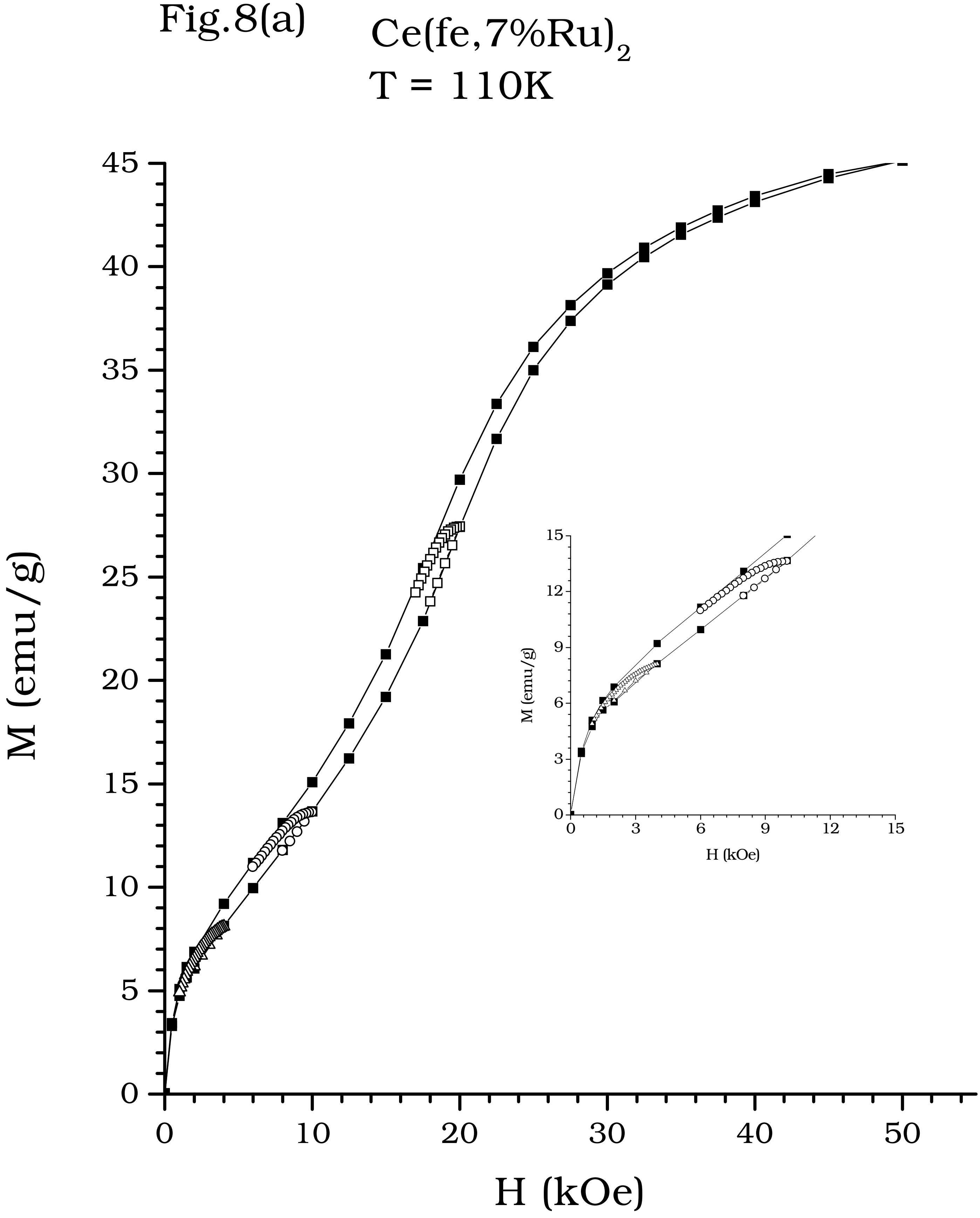
Ce(Fe,5%Ir)$_2$
T = 85K
M (emu/g)
H (KOe)
0
5
10
15
20
25
30
35
40
45
50
55
M(emu/g)
H(kOe)
3
4
5
6
7
8
9
10

Fig.7(b)



Ce(Fe,5%Ir)$_2$
T = 85K
M(emu/g)
H (kOe)
0
5
10
15
20
25
0
5
10
15
20
25
30
35
40
45
50
55

Fig.8(a)



Ce(fe,7%Ru)$_2$
T = 110K
M (emu/g)
H (kOe)
0
5
10
15
20
25
30
35
40
45
0
10
20
30
40
50
M (emu/g)
H (kOe)
0
3
6
9
12
15
0
3
6
9
12
15

Fig.8(b) $Ce(Fe,7\%Ru)_2$
T = 110K

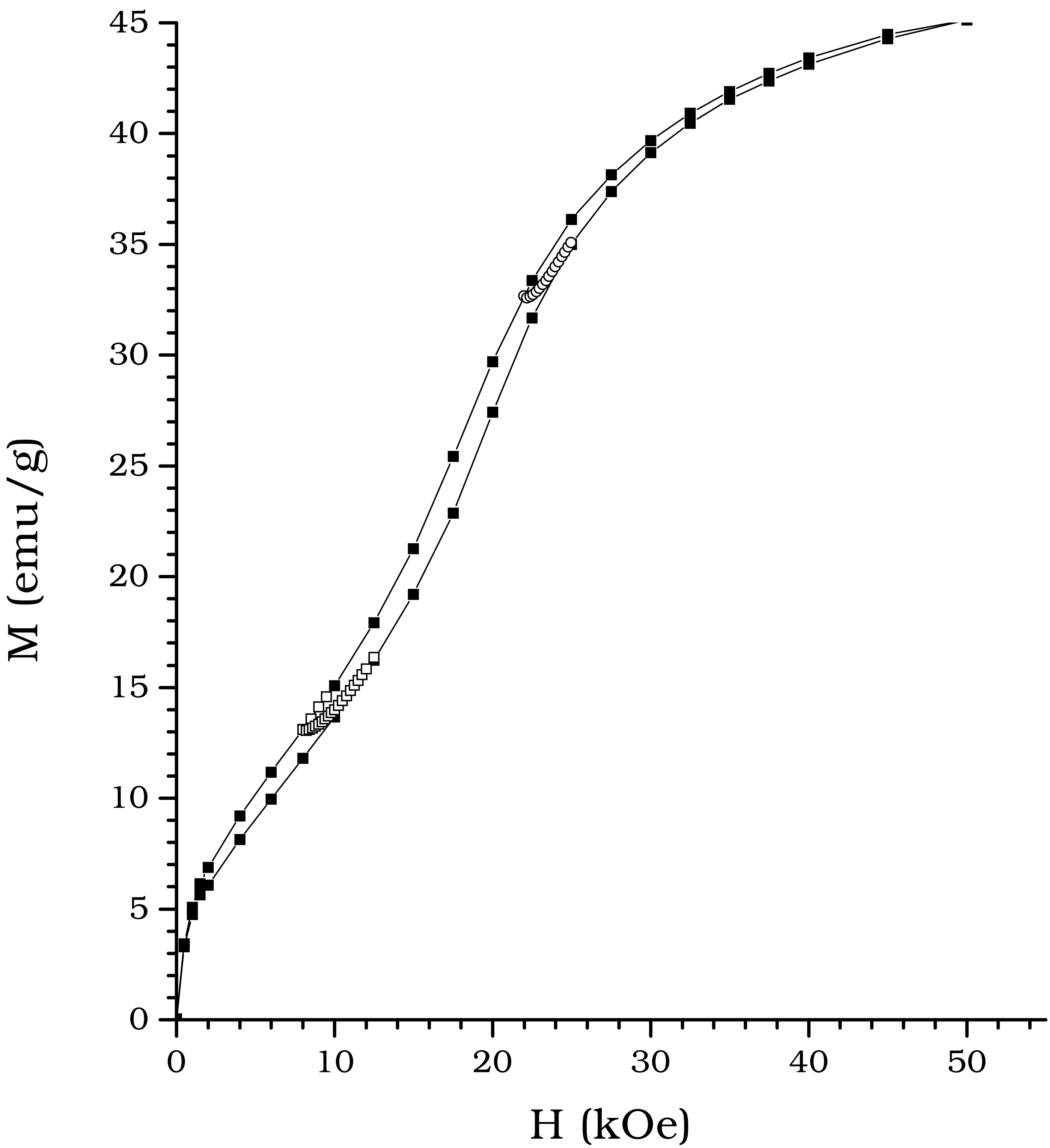


Fig.9