View metadata, citation and similar papers at core.ac.uk

brought to you by CORE

provided by Publications of the IAS Fellows

arXiv:cond-mat/0005399v1 [cond-mat.str-el] 24 May 2000

First order transition from ferro- to antiferromagnetism in $CeFe_2$ based pseudobinary alloys

Meghmalhar Manekar, S. B. Roy and P. Chaddah

Low Temperature Physics Laboratory,

Centre for Advanced Technology,

Indore 452013, India

(August 2, 2011)

Abstract

We present results of ac susceptibility measurements highlighting the presence of thermal hysteresis and phase coexistence across the ferro-to antiferromagnetic transition in various $CeFe_2$ based pseudobinary systems. These results indicate that the ferro-to antiferromagnetic transition in these systems is first order in nature.

Typeset using REVTEX

The C15-Laves phase compound $CeFe_2$ retains its identity amongst the other members of RFe_2 family (where R=Y,Zr and heavy rare earth elements). First, magnetic moment of $CeFe_2$ per formula unit ($\approx 2.4\mu_B$) is distinctly smaller than that found in other RFe_2 compounds [1]. Second,its Curie temperature $T_C(\approx 235$ K) is relatively small in comparison to the T_C of other RFe_2 compounds [1].However, short range magnetic order is detected in its paramagnetic state even in the temperature regime upto four times T_C [2].All these aspects drew the attention of the experimentalists during last thirty years, and amongst other things the role of Ce in the magnetic properties of $CeFe_2$ has been a subject of both theoretical [3] and experimental investigations [4,5];this in turn led to the discovery of newer interesting properties [6]. The most recent neutron measurements on single crystal sample of pure $CeFe_2$ have now revealed the presence of low temperature antiferromagnetic fluctuation in this otherwise ferromagnetic compound [6]. From the study of doped-$CeFe_2$ it is already known for quite sometime that the ferromagnetism of $CeFe_2$ is quite fragile in nature and a low temperature antiferromagnetic state can be established easily with small amount of doping with elements like Al,Co,Ru,Ir,Re,Os [7–13]. It should be noted, however, that the destabilization of ferromagnetism in $CeFe_2$ is not a simple disorder induced one, since the doping with other elements like Ni,Mn,Rh,Pd leads to simple dilution of ferromagnetism [8,10].

Most of the early experimental activities in $CeFe_2$ were focussed to establish the exact nature of the low temperature magnetic phase, whether it is a re-entrant spin-glass [14,15] or an antiferromagnetic state [9,10,16,17] and except in few cases [17,18] not much emphasis was given on the exact nature of this phase transtion. With the antiferromagnetic nature of the low temperature state being more or less established [16,17], in the present work we shall specifically address the question –what is the nature of this ferro- to antiferromagnetic transition? While there exists no complete theory (to our knowledge) to explain the interesting magnetic properties of $CeFe_2$, a phenomenological model dealing with itinerant electron systems [19] has often been invoked to explain the para-to ferro- to antiferromagnetic transition in the doped-psuedobinary alloys of $CeFe_2$.This phenomenological model of

Moriya and Usami predictd that the ferro- to antiferromagnetic transition would be a first order transition, while para- to ferromagnetic transition would be a second order transition [19]. With our high resolution ac-susceptibility measurement across this ferro- to antiferromagnetic transition in two doped samples of $CeFe_2$, we shall report characteristics which are typically associated with a first order transition.On the other hand the higher temperature para- to ferromagnetic transition can be characterized as a standard second order phase transtion. We believe that such a clear cut characterization of the various phase transitions in $CeFe_2$ based pseudobinaries is necessary, either for an appropraite extension of Moriya-Usami model [19] or for the development of newer theory for the proper understanding of the magnetic properties of $CeFe_2$.

Two samples–$Ce(Fe,5\%Ir)_2$ and $Ce(Fe,7\%Ru)_2$–used in the present study were prepared by argon arc melting from metals of at least 99.99% purity.Details of sample preparation, heat treatment and characterisation can be found in Ref. 10.The same samples have earlier been used in some other studies [13,20,21].

The AC susceptibility setup consists of a coil system having a primary solenoid and two oppositely wound secondaries each consisting of 1500 turns. The coil is dipped in liquid nitrogen to ensure that the temperature of the coil remains constant during the entire experiment to avoid drifts in the value of the applied field. The sample is mounted in a double walled quartz insert and its temperature is raised by heating the exchange gas by a heater wound on a seperate teflon mounting.A temperature controller (Lake Shore–DRC-91CA) is used for controlling the temperature.A copper-constantan thermocouple is used in differential mode to monitor the small temperature lag between the sample and the sensor.The sinusoidal output of a lock in amplifier (Stanford Research–SR830) is given to a voltage to current convertor which drives the current through the coil to generate the neccessary ac magnetic field. The signal from the pickup coil which is proportional to the susceptibility is measured by the same lock in amplifier. The field and frequency values were 4 Oe rms and 333Hz respectively.

Fig.(1) shows the AC susceptibility (χ) for both $Ce(Fe,5\%\ Ir)_2$ and $Ce(Fe,7\%\ Ru)_2$ as a function of temperature (T). The para- to ferromagnetic transition is characterized by a sharp increase in susceptibility (χ) with the decrease in T at T_{Curie} ≈185K in the 5% Ir doped sample and T_{Curie} ≈165K in the 7% Ru doped sample. Below T_{Curie} the susceptibility more or less flattens out for both the samples, before decreasing sharply at around 135K in Ce(Fe,5% Ir)$_2$ and at around 125K in Ce(Fe,7% Ru)$_2$. This low temperature decrease in χ has earlier been taken as a signature of ferro- to antiferromagnetic transition [10,13,20], and the estimated transition temperatures (T_N) from our present study agree well with the existing literature [10,13,20].

Our aim now is to find out the exact nature of these two magnetic transitions oberved in CeFe$_2$-based pseudobinaries. Experimentally, the indication of a first order transition usually comes via a hysteretic behaviour of various properties, not necessarily thermodynamic ones. As an example, the first indication of a first order melting transition from elastic solid to vortex liquid in a vortex matter came via distinct hysteresis observed in transport property measurements [22,23]. The confirmatory tests of the first order nature of a transition ofcourse involve the detection of discontinuous change in thermodynamic observables and the estimation of latent heat, and this has subsequently been achieved for vortex lattice melting in vortex matter [24,25]. There also exists a less rigorous class of experimental tests which invloves the study of phase inhomogeneity and phase coexistence across a first order transtion. This kind of experiment has also come out to be pretty informative for the melting transtion [26] as well as ordered solid to disordered solid transition [27,28] in vortex matter. In our present study we shall use hysteresis and phase coexistence to investigate the nature of the magnetic transitions in CeFe$_2$ based systems; our observable will be ac susceptibility (χ).

In order to observe a hysteresis in the transition, if any, we have chosen to sweep the temperature at a slow rate (0.006K/sec typical and slower when needed) instead of stabilizing at each temperature. This was done to ensure that the temperature is varied unidirectionally during both the heating and cooling cycles. The signal was measured at a temperature interval of 0.2K. The time constant of the low pass filter of the lock in amplifier was chosen such that the temperature changes negligibly(compared to our temperature step) within a time interval of 10 times the time constant. The temperature difference between the sensor and the sample, as monitored by the differential thermocouple, was always less than 1% of the sensor temperature and is used to obtain the correct value of the sample temperature.

First, we show the effect of temperature cycling on the para- to ferromagnetic transition in fig(2).In case of Ce(Fe,5% Ir)$_2$ the transition is reversible within an error of 0.15K to 0.2K.In case of Ce(Fe,7% Ru)$_2$ the reversibility is even better. The lack of hysteresis in para- to ferromagnetic transition within an error bar smaller than our temperature step, is indicative of a second order phase transition.

We then focus our attention on the ferro- to antiferromagnetic transition which has been shown to be associated with a structural distortion from cubic to rhombohedral [16,17], hinting towards a first order transition.The same protocol of sweeping the temperature and measuring the signal at closely spaced temperature values is followed during this measurement also.

Fig(3) shows the result of our measurements on both 5% Ir and 7% Ru doped CeFe$_2$ samples. Both the samples show a distinct thermal hysteresis in the ac-susceptibility across the ferro- to antiferromagnetic transition.The width of the hysteresis is about 2K which is well beyond the error in our measurements.

To study the phase coexistence we use the technique of minor hysteresis loop (MHL) [29]. We first define the "envelope curve" as the curve enclosing the thermally hysteretic susceptibility beteween the lower and higher temperature reversible region (see Fig.3). We can draw a MHL during the heating cycle i.e. start heating and increase T from the lower temperature reversible (antiferromagnetic) region and then reverse the direction of temperature before reaching the higher temperature reversible (ferromagnetic) region. We can also draw a MHL in the cooling cycle i.e. start cooling from the reversible ferromagnetic region and reverse the direction of temperature before reaching the lower temperature reversible antiferromagnetic region. If the heating is reversed at sufficiently 'low' temperatures the minor loop does not coincide with the cooling part of the 'envelope curve'. Here in the lower part of the hysteretic regime the high temperature ferromagnetic phase phase is not formed in a sufficient quantity; so when the temperature is decreased the curve does not fall on the cooling part of the envelope curve which represents the curve along which the high temperature phase is supercooled. The MHL's initiated from temperatures well inside the hysteretic regime coincide with the cooling part of envelope curve indicating that the high temperature phase has formed in a sufficient quantity. In Fig. 4 and 5 we present some representative MHLs both for the Ce(Fe,5%Ir)$_2$ and Ce(Fe,7%Ru)$_2$ alloys. We have drawn similar MHLs from the cooling branch of the enevelope curve, which are not shown here for the sake of clarity and conciseness. We have reproduced this behaviour of MHLs over many experimental cycles. The presence of these MHLs clearly suggest the existence of phase coexistence across the ferro- to antiferromagnetic transition. Had there been no phase coexistence we would have followed the cooling part of the envelope curve reversibly on increasing T.Very similar minor hysteresis loop technique has been used to study the phase coexistence associated with a first order metal-insulator transitions in NdNiO$_3$ [30].

It should be noted here that the pinning of solitons (domain walls) by lattice defects can also give rise to a thermal hysteresis [31] in magnetic measurements. However, the observed thermal hysteresis in our present study is confined to a relatively narrow temperature window and this argues against such a possibility.

In conclusion we have shown that the ferro- to antiferromagnetic transition in the compounds Ce(Fe,5%Ir)$_2$ and Ce(Fe,7%Ru)$_2$ is accompanied by distinct thermal hysteresis as well as signatures of phase-coexistence. We argue that these observations are indicative of the first order nature of the concerned phase transition. The higher tempeature para- to ferromagnetic transition appears to be a typical second order phase transition. These results would support the applicability of Moriya-Usami's model [19] in explaining the double magnetic transitions in various CeFe$_2$ based pseudobinary systems. A calorimetric study is now required to confirm the conjecture that this ferro- to antiferromagnetic transition is first order in nature. However, it should be noted that in the case of small latent heats it might be difficult to distinguish a first order transition through calorimetric studies [32];

in such cases the observed hysteresis and phase coexistence would remain a useful tool for identification of a first order transition.

REFERENCES

[1] Buschow K H J, in Ferromagnetic Materials, edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1980) Vol.1.

[2] Deportes J, Givord D and Ziebeck KRA, 1981, J. Appl. Phys. **52** 2074.

[3] Erikkson O, Nordstrom L, Brooks MSS and Johansson B, 1988, Phys. Rev. Lett. **60** 2523.

[4] Kennedy SJ, Brown PJ and Coles BR, 1993, J. Phys.:Condens. Matter, **5** 5159.

[5] Cooper MJ et al, 1996, Phys. Rev. **B54**, 4068.

[6] Paolasini L et al, 1998, Phys. Rev.**B58** 12117.

[7] Franceschini DF and Da Cunha SF , 1985, J. Magn. Magn. Matr. **52** 280.

[8] Rastogi AK and Murani AP, in Theoretical and Experimental Aspects of Valence Fluctuations and Heavy Fermions edited by L. C. Gupta and S. K. Malik (Plenum, New York) 1687.

[9] Roy SB and Coles BR, 1989, J. Phys. :Condens Matter,**1**, 419.

[10] Roy SB and Coles BR, 1989, Phys. Rev. **B39**, 9360.

[11] Rastogi AK, Hilscher G, Gratz E and Pillmayr N, 1988, J. Phys. Colloq. **C8** 277.

[12] Roy SB, Kennedy SJ, Coles BR, 1988, J. Phys. Colloq. **C8** 271.

[13] Rajarajan AK, Roy SB and Chaddah P, 1997, Phys. Rev. **B56** 7808.

[14] Roy SB and Coles BR, 1987, J. Phys. **F17** L215.

[15] Pillay RG, Grover AK, Balasubramanian V, Rastogi AK and Tandon PN, 1988, J. Phys. **F18** L63.

[16] Kennedy SJ, Murani AP, Cockcroft JK, Roy SB and Coles BR, 1989, J. Phys. :Condens

Matter, **1** 629.

[17] Kennedy SJ and Coles BR, 1990, J. Phys. :Condens Matter **2** 1213.

[18] Ali N and Zhang X, 1992, J. Phys. :Condens Matter **4** L351.

[19] Moriya T and Usami K, 1977, Solid St. Commun. **23** 935.

[20] Wang D, Kunkel HP and Williams Gwyn, 1995, Phys. Rev. **B51**, 2872.

[21] Kunkel HP, Zhou XZ, Stampe PA, Cowen JA and Williams G, 1996, Phys. Rev. **B53** 15099.

[22] Safar H et al, 1992, Phys. Rev. Lett. **69** 824.

[23] Kwok WK et al, 1994, Phys. Rev. Lett. **72** 1092.

[24] Zeldov E et al, 1995, Nature, **375** 373.

[25] Schilling A et al, 1996, Nature, **382** 791.

[26] Soibel A et al, Proc. of LT-22 Conf. Helsinki 1999, Physica **B** (in press).

[27] Roy SB and Chaddah P, 1997, J. Phys. :Condens Matter, **9** L625.

[28] Roy SB, Chaddah P and Chaudhary S, 1998, J. Phys. :Condens Matter, **10** 4885.

[29] Roy SB and Chaddah P, 1997, Physica **C279** 70.

[30] Granados X, Fontcuberta J, Obradors X and Torrence JB, 1992, Phys. Rev. **B46** 15683.

[31] Levanyuk AP and Sigov AS in Defects and Structural Phase transitions (Gordon and Breach Science Publishers,1988).

[32] White RM and Geballe TH, in Long Range Order in Solids (Academic Press, 1979).

FIGURES

FIG. 1. AC susceptibility (χ) versus temperature (T) plots for (a)Ce(Fe,5%Ir)$_2$ (b)Ce(Fe,7%Ru)$_2$.

FIG. 2. χ vs T plot highlighting the thermal reversibility of the para- to ferromagnetic transition in (a)Ce(Fe,5%Ir)$_2$ (b)Ce(Fe,7%Ru)$_2$.

FIG. 3. χ vs T plot highlighting the thermal irreversibility of the ferro- to antiferromagnetic transition in (a)Ce(Fe,5%Ir)$_2$ (b)Ce(Fe,7%Ru)$_2$.

FIG. 4. Minor hysteresis loops (MHL) in χ vs T plot highlighting phase coexistence in Ce(Fe,7%Ru)$_2$: (a)representative MHL initiated from the lower part of the hysteretic regime, (b)representative MHLs initiated from well inside the hysteretic regime. See text for details.

FIG. 5. Minor hysteresis loops (MHL) in χ vs T plot highlighting phase coexistence in Ce(Fe,5%Ir)$_2$.

Fig1(a)

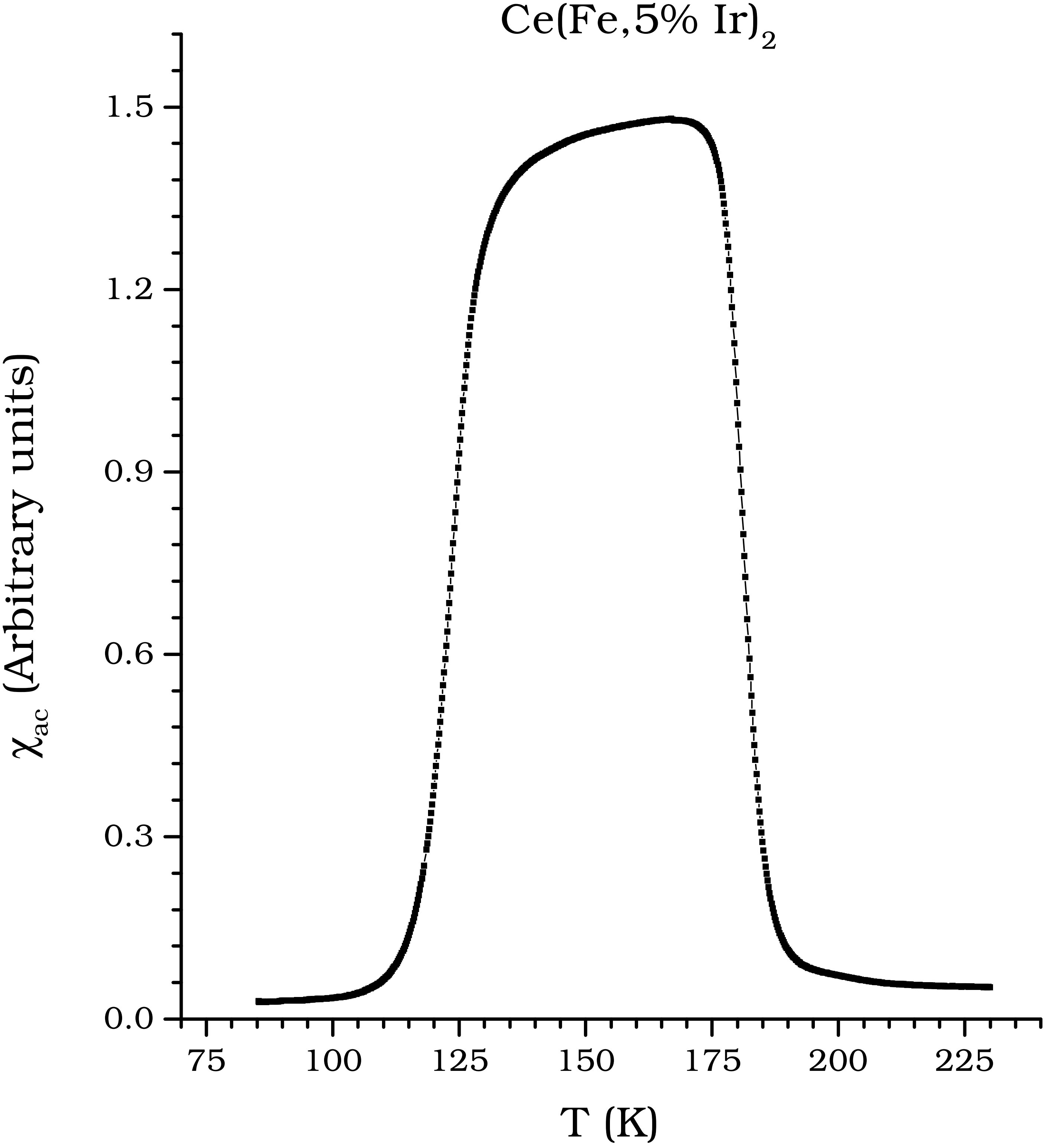

Ce(Fe,5% Ir)$_2$
χ_{ac} (Arbitrary units)
0.0
0.3
0.6
0.9
1.2
1.5
75
100
125
150
175
200
225
T (K)

fig1(b)

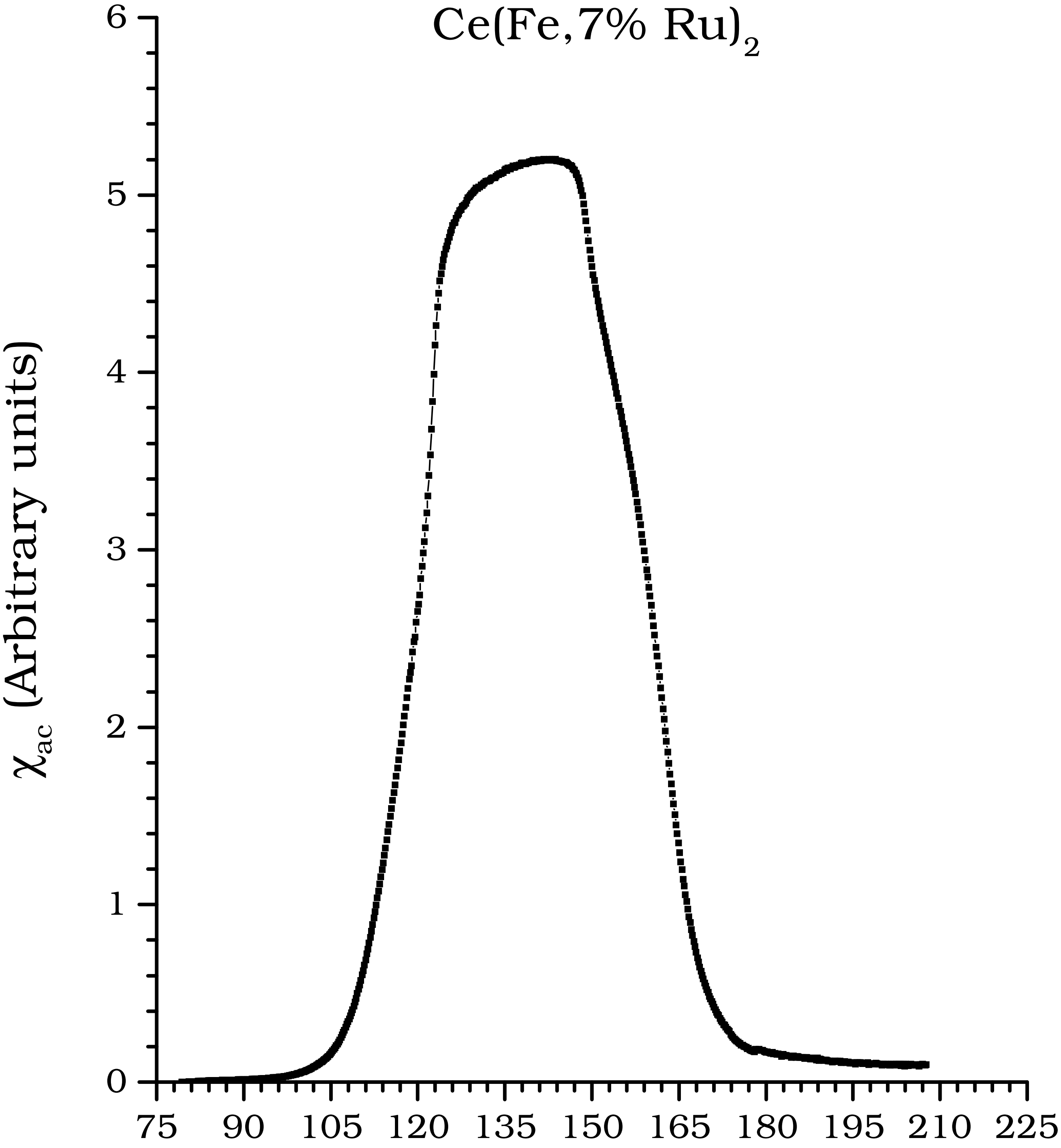

Ce(Fe,7% Ru)$_2$
χ_{ac} (Arbitrary units)
T (K)
0
1
2
3
4
5
6
75
90
105
120
135
150
165
180
195
210
225

fig2(a)

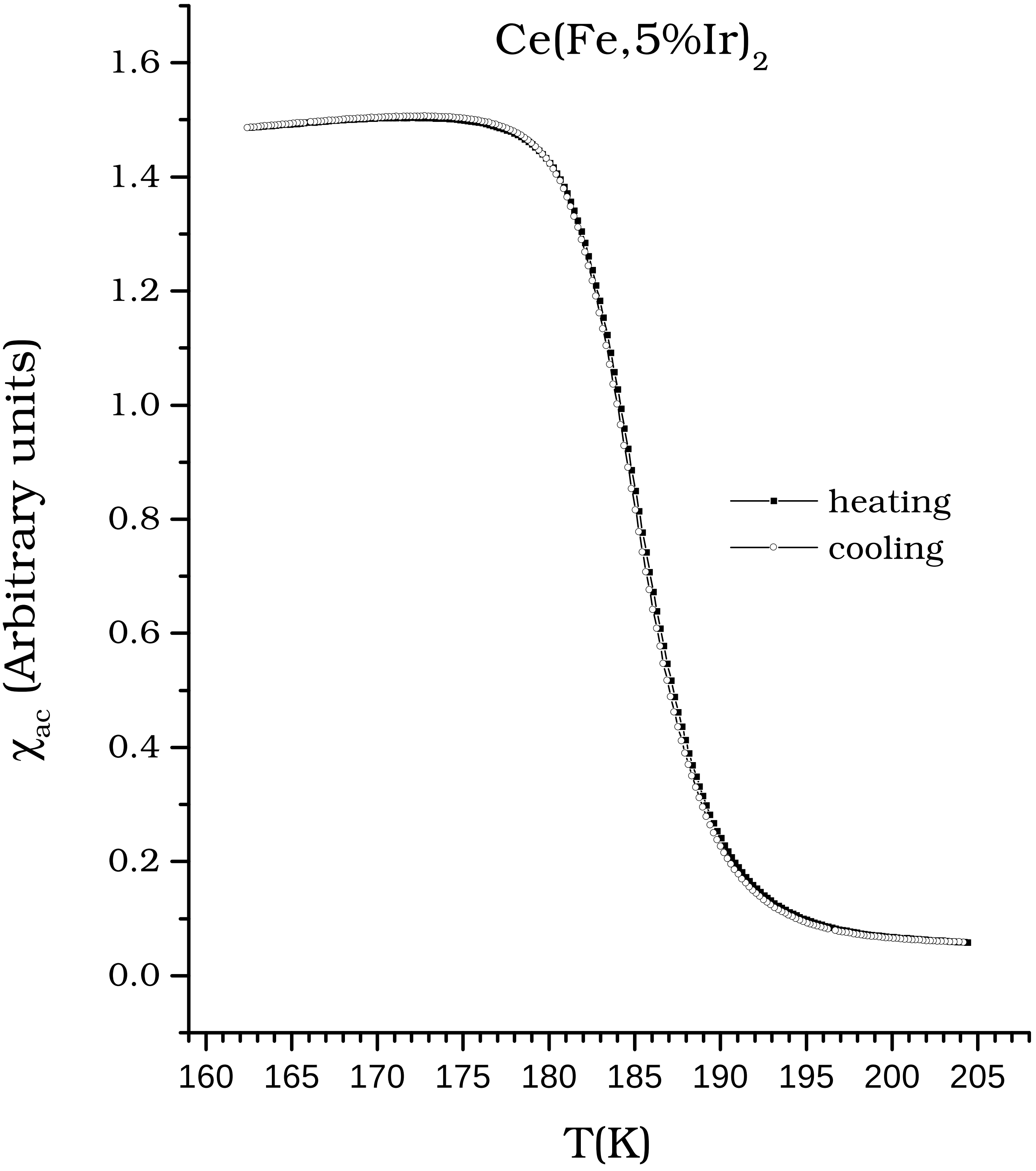

Ce(Fe,5%Ir)$_2$
χ_{ac} (Arbitrary units)
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
160
165
170
175
180
185
190
195
200
205
T(K)
heating
cooling

fig2(b)

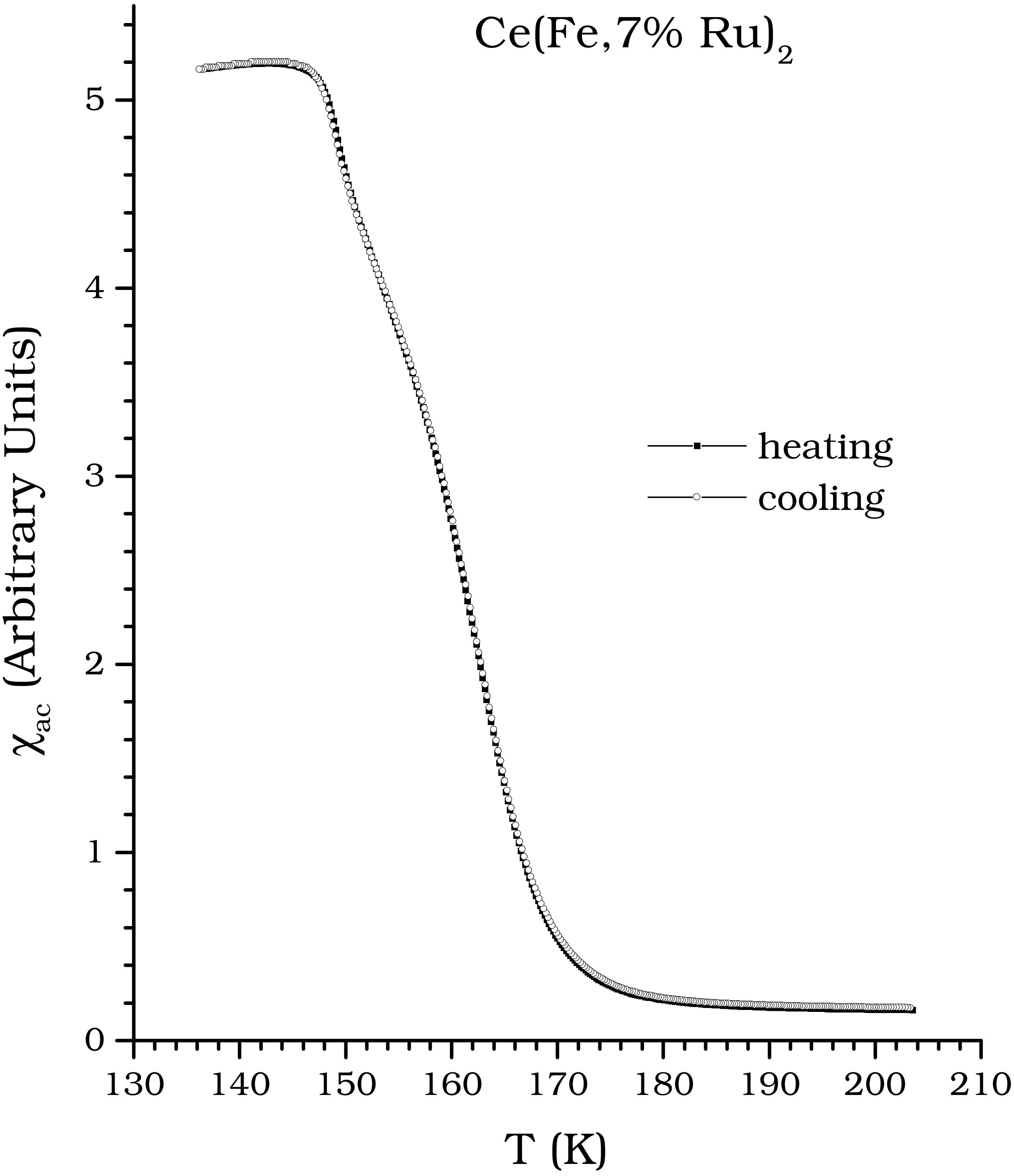
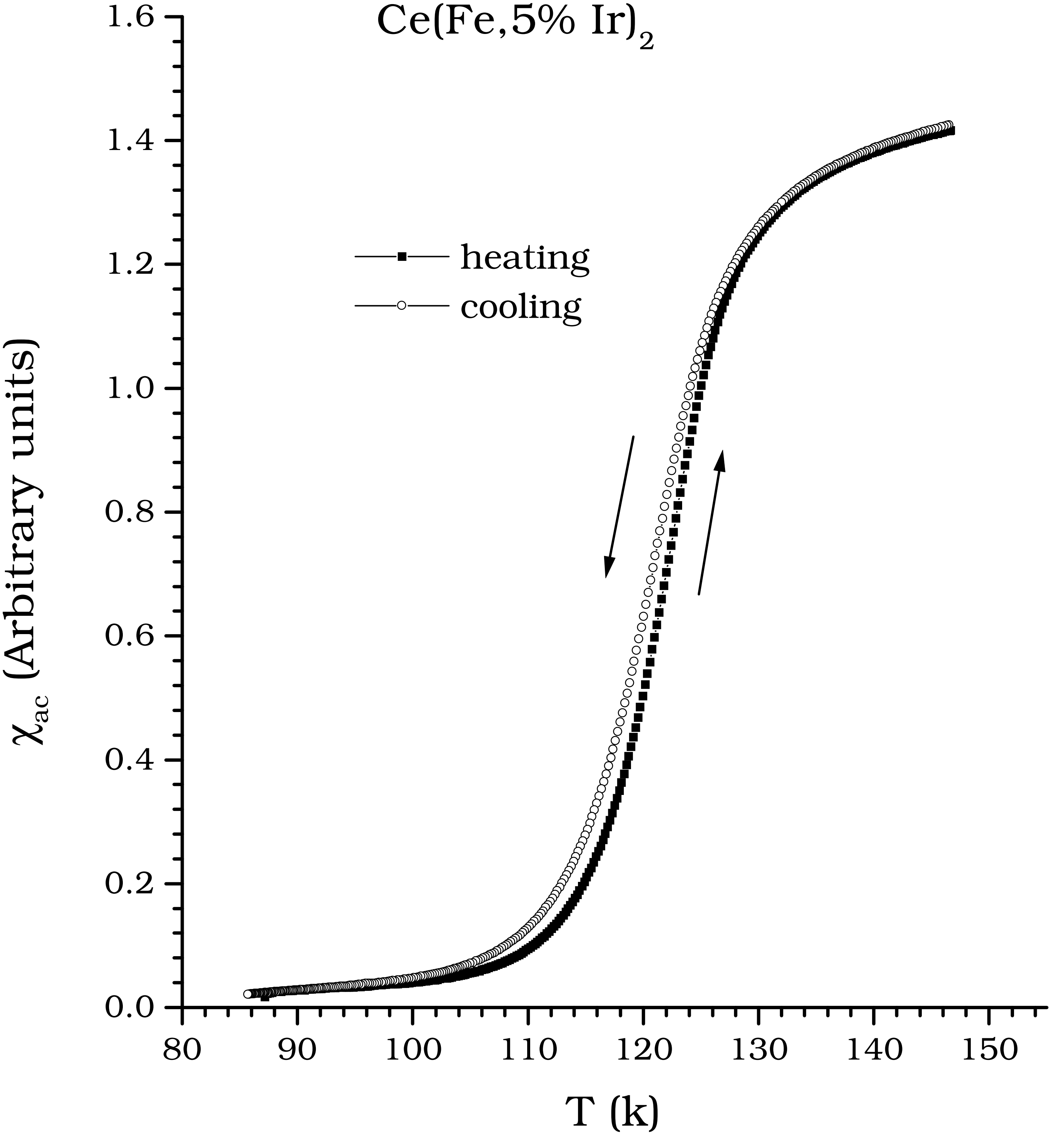

Ce(Fe,7% Ru)$_2$
heating
cooling
χ_{ac} (Arbitrary Units)
T (K)
0
1
2
3
4
5
130
140
150
160
170
180
190
200
210

fig3(a)



Ce(Fe,5% Ir)$_2$
heating
cooling
χ_{ac} (Arbitrary units)
T (k)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
80
90
100
110
120
130
140
150

fig3(b)

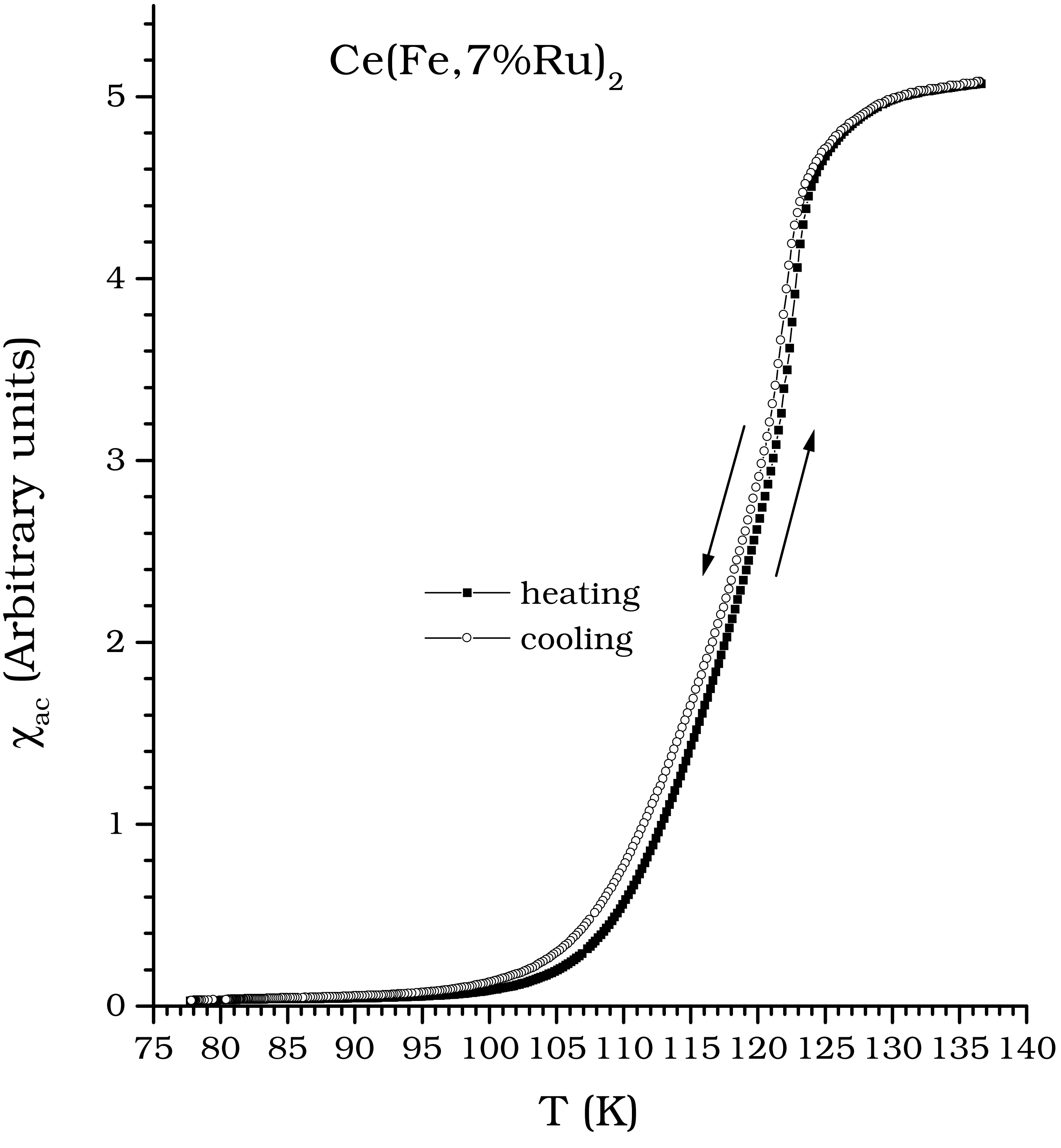

Ce(Fe,7%Ru)$_2$
heating
cooling
χ_{ac} (Arbitrary units)
0
1
2
3
4
5
T (K)
75
80
85
90
95
100
105
110
115
120
125
130
135
140

fig4(a)

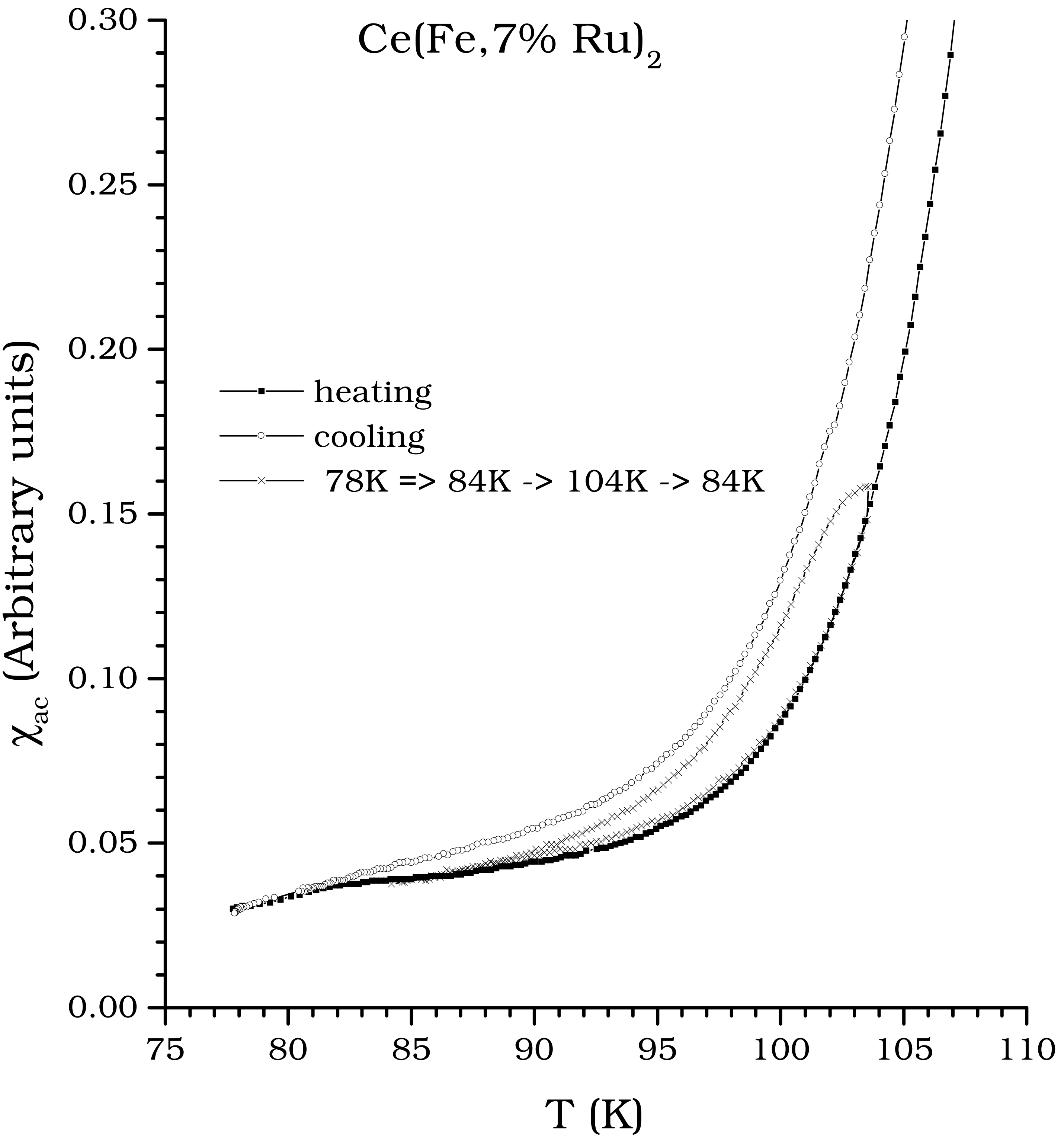

Ce(Fe,7% Ru)$_2$
heating
cooling
78K => 84K -> 104K -> 84K
χ_{ac} (Arbitrary units)
0.00
0.05
0.10
0.15
0.20
0.25
0.30
75
80
85
90
95
100
105
110
T (K)

fig4(b)

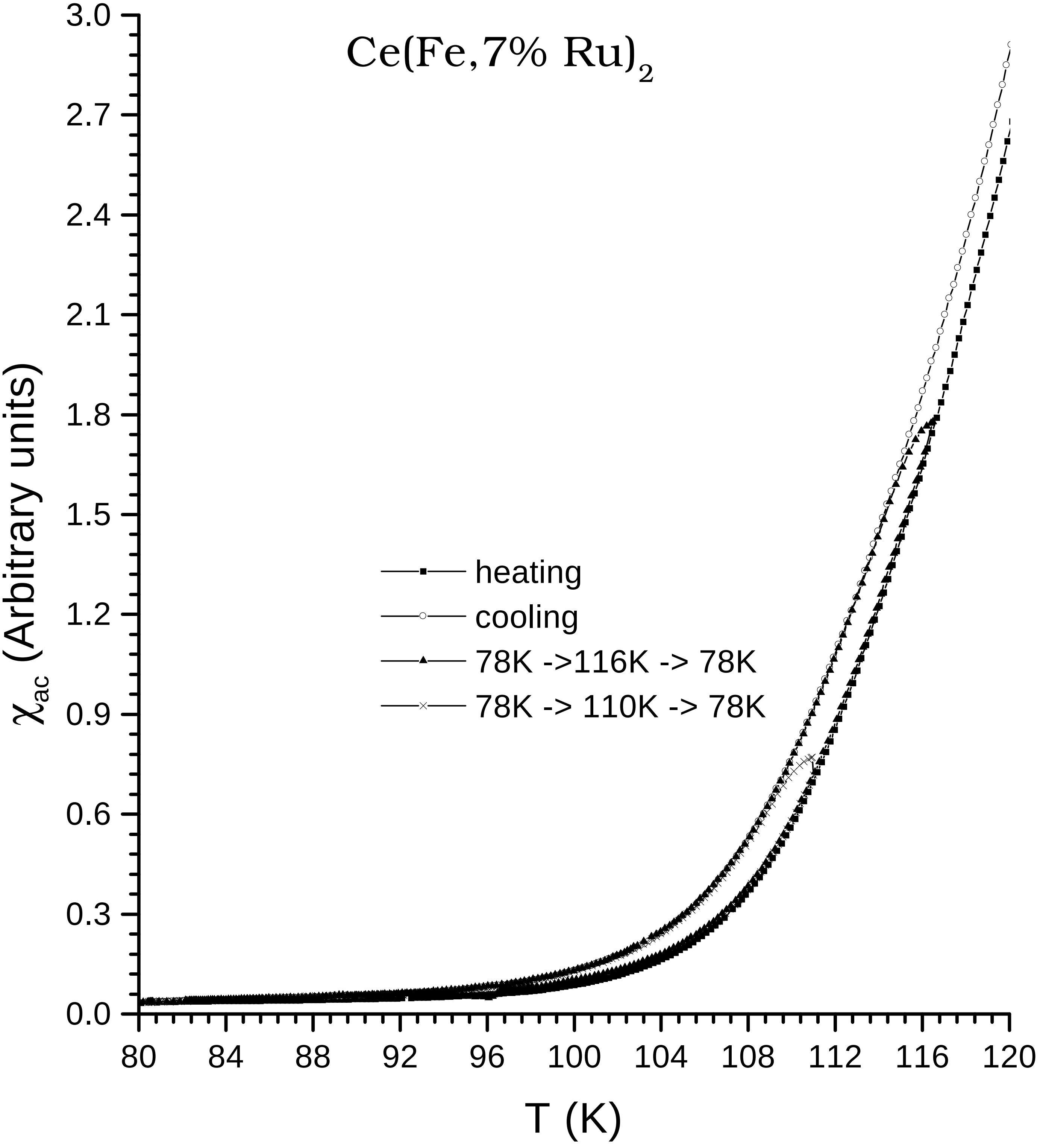
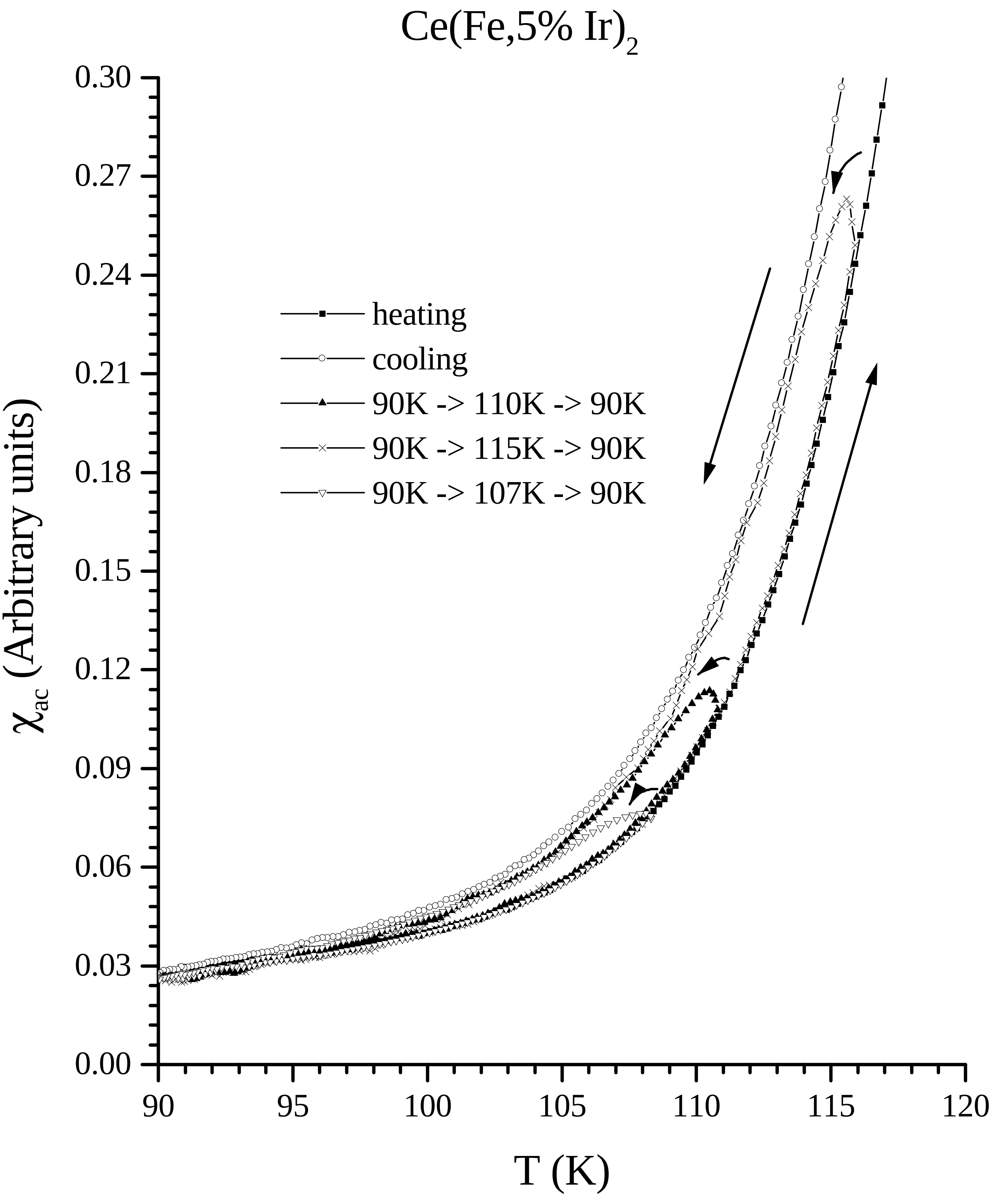

Ce(Fe,7% Ru)$_2$
heating
cooling
78K ->116K -> 78K
78K -> 110K -> 78K
χ_{ac} (Arbitrary units)
T (K)
0.0
0.3
0.6
0.9
1.2
1.5
1.8
2.1
2.4
2.7
3.0
80
84
88
92
96
100
104
108
112
116
120

fig. 5



Ce(Fe,5% Ir)$_2$
heating
cooling
90K -> 110K -> 90K
90K -> 115K -> 90K
90K -> 107K -> 90K
χ_{ac} (Arbitrary units)
T (K)
0.00
0.03
0.06
0.09
0.12
0.15
0.18
0.21
0.24
0.27
0.30
90
95
100
105
110
115
120