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Abstract: We have precisely investigated the antiferromagnetic transition in the ternary rare-earth metal silicide Er5Ir4Si10 single crystal by performing the high-resolution measurement of the low-temperature specific heat under zero-magnetic field and the AC nagnetization. In the temperature dependence of important physical quantities associated with the antiferromagnetic phase transition, we have observed anomalies associated with the antiferromagnetic long-range ordering at Neel temperature TN. We have confirmed that TN is 3.5 K. In addition, we have first observed two surprising results. Firstly, a shoulder was observed in the vicinity of 2K in addition to the sharp peak at TN corresponding to the antiferromagnetic long-range ordering in the high-resolution measurement of the low-temperature specific heat. Secondly, the anomaly of the AC magnetization at TN depends to the magnetic direction. Though we have clearly observed the anomaly of the AC magnetization assocated with the antiferromagnetic phase transition at TN when the AC magnetic field orientiation is parallel to the c-axis, we have observed no anomaly of the AC magnetization at TN when the AC magnetic field direction is perpendicular to the c-axis. These results clarify that our Er5Ir4Si10 single crystal is a quasi-two-dimensional antiferromagnet and then has no magnetic structure of the Er3+ local moments. However, we have observed a peak of AC magnetization around 2K when the AC magnetic field is perpendicular to the c-axis. This temperature corresponds to that at which a shoulder is observed in the high-resolusion measurement of the low-temperature specific heat.

Furthermore, we have observed no frequency dependency of the AC magnetization which is ordinarily observed in the spin glass state. This result means that there is no disorder in our Er5Ir4Si10 single crystal because the crystal structure of Er5Ir4Si10 has the tetragonal crystal structure in which the octagons of Er3+ ions are stacked. In addition, both the tetragons and the octagons of Er3+ local moments have no magnetic frustration. At last we can conclude that both the shoulder of the low-temperature specific heat in the vicinity of 2K and the peak of the AC magnetization around 2K, which is only observed when the AC magnetic field direction is perpendicular to the c-axis, correspond to the crystallne electric field effet in the plane which is perpendicular to the c-axis of the tetragonal crystal structure.

Dear Dr. K. H. J. Buschow
Editor in Chief
Journal of Alloys and Compounds

I submit my paper entitled "Antiferromagnetic transition in ternary rare-earth metal silicide $Er_5Ir_4Si_{10}$ single crystal "to Journal of Alloys and Compounds.

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Antiferromagnetic transition in ternary rare-earth metal silicide $Er_5Ir_4Si_{10}$ single crystal

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Abstract

   We have precisely investigated the antiferromagnetic phase transition in the ternary rare-earth metal silicide $Er_5Ir_4Si_{10}$ single crystal by performing the high-resolution measurement of the low-temperature specific heat under zero-magnetic field and the AC magnetization. In the temperature dependence of important physical quantities associated with the antiferromagnetic phase transition, we have observed anomalies associated with the antiferromagnetic long-range ordering at Neel temperature T_N. We have confirmed that T_N is 3.5 K. In addition, we have first observed two surprising results. Firstly, a shoulder was observed in the vicinity of 2K in addition to the sharp peak at T_N corresponding to the antiferromagnetic long-range ordering in the high-resolution measurement of the low-temperature specific heat. Secondly, the anomaly of the AC magnetization at T_N depends to the magnetic field direction. *Though we have clearly observed the anomaly of the AC magnetization associated with the antiferromagnetic phase transition at T_N when the AC magnetic field direction is parallel to the c-axis, we have observed no anomaly of the AC magnetization at T_N when the AC magnetic field orientation is perpendicular to the c-axis. These results clarify that our $Er_5Ir_4Si_{10}$ single crystal is a quasi-two-dimentional antiferromagnet and then has no magnetic structure of the Er^{3+} local moments.* However, we have observed a peak of the AC magnetization around 2K when the AC magnetic field is perpendicular to the c-axis. This temperature corresponds to that at which a shoulder is observed in the high-resolution measurement of the low-temperature specific heat.

Furthermore, we have observed no frequency dependence of the AC magnetization which is ordinarily observed in the spin glass state. This result means that there is no disorder in our $Er_5Ir_4Si_{10}$ single crystal because the crystal structure of $Er_5Ir_4Si_{10}$ has the tetragonal crystal structure in which the octagons of Er^{3+} ions are stacked. In addition, both the tetragons and the octagons of Er^{3+} local moments have no magnetic frustration. *At last we can conclude that both the shoulder of the low-temperature specific heat in the vicinity of 2K and the peak of the AC magnetization around 2K, which is only observed when the AC magnetic field direction is perpendicular to the c-axis, correspond to the crystalline electric field effect in the plane which is perpendicular to the c-axis of the tetragonal crystal structure.*

1. Introduction

   The ternary rare-earth metal silicide $R_5Ir_4Si_{10}$ (R = heavy rare-earth metal, Sc and Lu) have been intensively studying. The attractive phenomena in this ternary compound are antiferromagnetic transition [1-7], superconducting transition [1, 8-15], charge-density wave transition [16-29] and the nuclear magnetism [7]. This compound group crystallizes in the tetragonal $Sc_5Co_4Si_{10}$-type structure and the space group is P4/mbm [1, 7, 15]. The projection along the c-axis crystal structure of $Sc_5Co_4Si_{10}$ is shown in Fig. 1. The features of this crystal structure are the absence of the cluster which is composed of transition metals and then the direct bond between the transition metals. These features are in contrast to those of Cheverel phase chalcogenides RMo_6S_8 (R = rare-earth metal) and rhodium boride compounds RRh_4B_4 (R = rare-earth metal). In the $R_5Ir_4Si_{10}$ compound group, the Ir atoms and the Si atoms form planar nets of pentagons and hexagons that are linked in the plane which is perpendicular to the c-axis and then connected along the c-axis via Ir-Si-Ir zigzag chains. On the other hand, the rare-earth metal R^{3+} ions have three sites whose symmetries are different each other. The rare-earth metal R^{3+} ions at two sites of them make the octagonal layer which is perpendicular to the c-axis. We must note that the octagonal layer of the rare-earth metal R^{3+} ions have no magnetic frustration. The rare-earth metal R^{3+} ions at the third site which are present at the center of the octagons which are composed of the Ir atoms and the Si atoms also make another layer which is also perpendicular to the c-axis and a square lattice. We must also note that the square lattice of the R^{3+} ions has no magnetic frustration. Though these both layers are perpendicular to the c-axis, the difference between these two layers is as follows. The one layer contains only the rare-earth metal R^{3+} ions and then separates the pentagon, hexagon and octagon net works which are composed of both Ir atoms and Si atoms. Another includes not only the rare-earth metal R^{3+} but also the Ir atoms and the Si atoms. These results mean that there are two kinds of layers which contain the rare-earth metal R^{3+} ions and   perpendicular to the c-axis. Therefore, we must consider that the character of the $R_5Ir_4Si_{10}$ compounds is quasi-two-dimensional rather than one-dimensional on the heavy rare-earth metal R^{3+} ions

that correspond to the magnetic properties. Furthermore, all Ir-Si and Si-Si distances are clearly short and indicative of the covalent bonding. In many other ternary rare-earth metal silicides such as $ThCr_2Si_2$, $CeNiSi_2$ and $LaRe_2Si_2$, the net work of the Si atoms and transition atoms exists. In this article, we report and discuss the anomalies in the low-temperature specific heat and the AC magnetization measurements associated with the antiferromagnetic transition in our high-quality $Er_5Ir_4Si_{10}$ single crystal.

2. Experiments

The $Er_5Ir_4Si_{10}$ single crystals employed in our study were grown by Czochoralski pulling method with a tetra-arc furnace in the high purity argon atmosphere whose purity is 6N. The purity of starting materials as follows. The purity of Si is 6N and that of Ir is 4N. However, the purity of Tb is 3N. During the single crystal growth a clear faceting has been observed sometimes. We confirmed as-grown crystals to be single crystals by the transmission Laue X-ray photograph method. The single crystal oriented along the c-axis. In addition, in order to improve the quality of the as-grown single crystals we used a solid state electro-transport method (SSE). In the SSE process of the as-grown single crystal, we have kept them at 1273 K for a month.

For the measurement of the temperature dependence of the AC magnetization, we have used the commercial Quantum Design SQUID magnetometer (MPMS) from 1.8 to 20 K.

For the measurement of the temperature dependence of the low-temperature specific heat, a handmade adiabatic method was employed. This non-commercial apparatus enables us the high-resolution measurement of the low-temperature specific heat down to 0.5 K.

3. Results and Discussion

In Figs. 2 and 3, we show the temperature dependence of the AC magnetization from 1.8 to 20 K. We show the result when the AC magnetic field direction is parallel to the c-axis in Fig. 2 and we show the result in Fig. 3 when the AC magnetic field orientation is perpendicular to the c-axis. When the AC magnetic field orientation is parallel to the c-axis, T_N is 3.5 K which is precisely consistent with the result of the low-temperature specific heat measurement as is shown in Fig. 4. However, we have no anomaly of the AC magnetization at T_N when the AC magnetic field direction is perpendicular to the c-axis as is very clear in Fig. 3. This result is a very surprising finding because the result of the temperature dependence measurement of the low-temperature specific heat clearly reveals that the antiferromagnetic long-range ordering occurs at 3.5 K as is clearly shown in Fig. 4. When we take the results mentioned just above into consideration, we must conclude that our $Er_5Ir_4Si_{10}$ single crystal have no magnetic structures of the Er^{3+} local moments. This statement is completely different from those which are described in Ref. [18].

In addition, we have observed the peak of the AC magnetization around 2K when the AC magnetic field direction is perpendicular to the c-axis. We have already reported that a shoulder is observed in the temperature dependence of the low-temperature specific heat in the vicinity of 2 K [7]. The shoulder in the vicinity of 2 K is very precisely consistent with the peak around 2K in the temperature dependence of the AC magnetization when the magnetic field direction is perpendicular to the c-axis. At last we have completely confirmed that the shoulder corresponds to the magnetic property in the plane perpendicular to the c-axis of $Er_5Ir_4Si_{10}$ single crystal. This result reveals that $Er_5Ir_4Si_{10}$ single crystal is a quasi-two-dimensional material and an anisotropic antiferromagnet.

Next, we must discuss on the relation between the shoulder in the temperature dependence of the low-temperature specific heat in the vicinity of 2K and the peak of the AC magnetization around 2K when the AC magnetic field is perpendicular to the c-axis.

The shoulder suggests that the magnetic spatial ordering is a short-range one. The representative magnetic short range ordering is

the spin glass state. The AC magnetization measurement is very powerful for the investigation of the spin glass behavior. We have performed the frequency dependence measurement of the magnetization with the magnetic field whose orientation is perpendicular to the c-axis. The results are shown in Fig. 5. We have observed no spin glass like behavior. Namely, the peak of the AC magnetization does not shift to higher temperature and then the AC magnetization rapidly decreased with increasing the frequency of the AC magnetic field. These results mean that there is no spin glass state in our $Er_5Ir_4Si_{10}$ single crystal. Non-existence of the spin glass state verify that there is no disorder in our $Er_5Ir_4Si_{10}$ single crystal because the crystal structure of $R_5Ir_4Si_{10}$ (R=Tb, Dy, Ho, Er) has no magnetic frustration. Therefore, we can conclude that the shoulder in the vicinity of 2K in the temperature dependence of the low-temperature specific heat and the peak around 2K of the AC magnetization when the AC magnetic field direction is perpendicular to the c-axis originate from the crystalline electric field effect in the plane of $Er_5Ir_4Si_{10}$ single crystal which is perpendicular to the c-axis.

Galli et al. [16-18] had reported that the high-quality single crystal of $Er_5Ir_4Si_{10}$ undergoes the long-range antiferromagnetic transition at 2.8 K from the DC magnetization measurement by using Quantum Design MPMS, the measurement of the resistivity and the specific heat by using Quantum Design PPMS and the neutron diffraction study. These measurements by using Quantum Design MPMS and PPMS only down to 1.8 K cannot clearly detect the shoulder in the low-temperature specific heat [7] and the AC magnetization peak in the vicinity of 2 K which we have first observed and reported in this article. In addition, we show the temperature dependence of the low-temperature specific heat of our as-grown single crystal in Fig. 6. We have clearly observed two successive peaks at 1.8 and 3.5 K. This observation in the temperature dependence of the low-temperature specific heat is very comparable with that of $Yb_5Ir_4Si_{10}$ polycrystalline sample which is reported in Ref. [27]. But we have clearly observed a shoulder in the vicinity of 2 K together with the peak at 3.5 K in our SSE processed single crystal. These results strongly insist that the SSE process is indispensable for the as-grown single crystals grown by

Czochoralski pulling method.

On the other hand, G. J. Li et al. have reported on the superconductivity of $Sc_5Ir_4Si_{10}$ single crystals grown by the floating zone method [13]. Those single crystals showed clearly anisotropic superconducting properties which are very well explained in the standard BCS model. Furthermore, we must note that the observation of the peak effect in the superconductors is the direct evidence of the quasi-two-dimensional nature of the crystal structure [30]. The observation of the peak effect in $Sc_5Ir_4Si_{10}$ single crystal grown by the floating zone method is very consistent with our study of the antiferromagnetic properties in our SSE processed $Er_5Ir_4Si_{10}$ single crystal grown by Czochoralski pulling method with a tetra-arc furnace under high purity argon atmosphere.

4. Conclusions

The AC magnetization measurement of the SSE processed $Er_5Ir_4Si_{10}$ single crystals combined with the experimental results of the low-temperature specific heat have revealed the experimental evidences for the following conclusions.

(1) $Er_5Ir_4Si_{10}$ single crystal is a quasi-two-dimensional material.

(2) $Er_5Ir_4Si_{10}$ single crystal is a quasi-two-dimensional antiferromagnet.

(3) Though $Er_5Ir_4Si_{10}$ single crystal exhibits an antiferromagnetic long-range ordering, there is no magnetic structure of Er^{3+} local moments.

(4) The crystalline electric field effect is only observed in the plane which is perpendicular to the c-axis of the tetragonal crystal structure.

(5) The SSE process is indispensible in order to clarify the intrinsic magnetic properties of the single crystals grown by Czochoralski pulling method.

Finally, we strongly insist that the structural phase transition of $Lu_5Ir_4Si_{10}$ single crystal is much the same as the cubic-tetragonal phase transition in A15 compounds because the nature of the phase transition in both compounds is the first-order one [9, 23, 31].

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Figure Captions

Fig. 1. Projection of $Sc_5Ir_4Si_{10}$ along the c-axis. Filled circles correspond to z = 0, 1 and open circles to z = 1/2 where z is the fractional coordinate along the c-axis of the tetragonal structure.

Fig. 2. Temperature dependence of the AC magnetization in $Er_5Ir_4Si_{10}$ single crystal when the AC magnetic field direction is parallel to the c-axis. Note that there is no anomaly of the AC magnetization around 2 K.
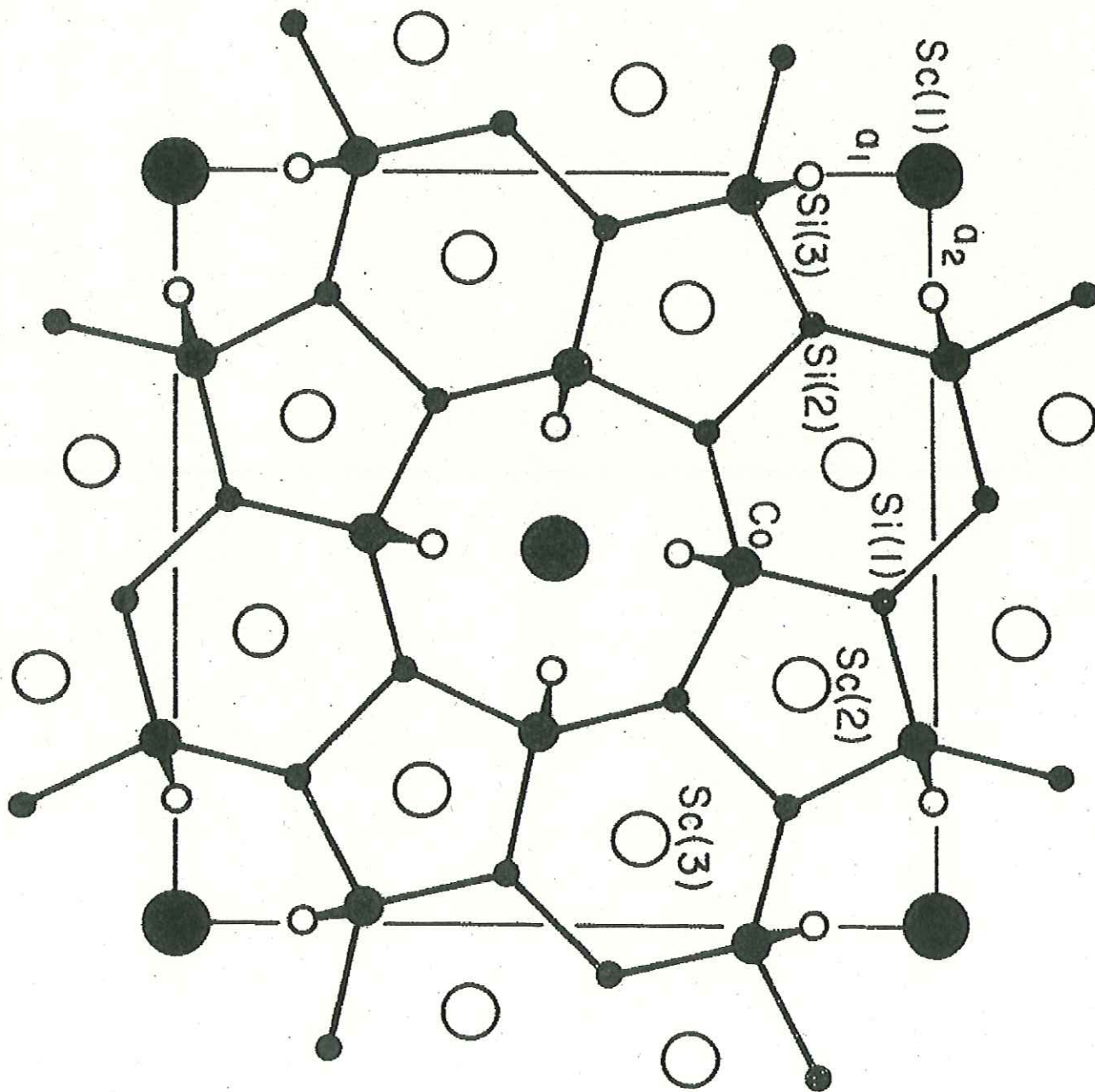
Fig. 3. Temperature dependence of the AC magnetization in $Er_5Ir_4Si_{10}$ single crystal when the AC magnetic field direction is perpendicular to the c-axis. Note that there is no anomaly of the AC magnetization at T_N = 3.5 K.
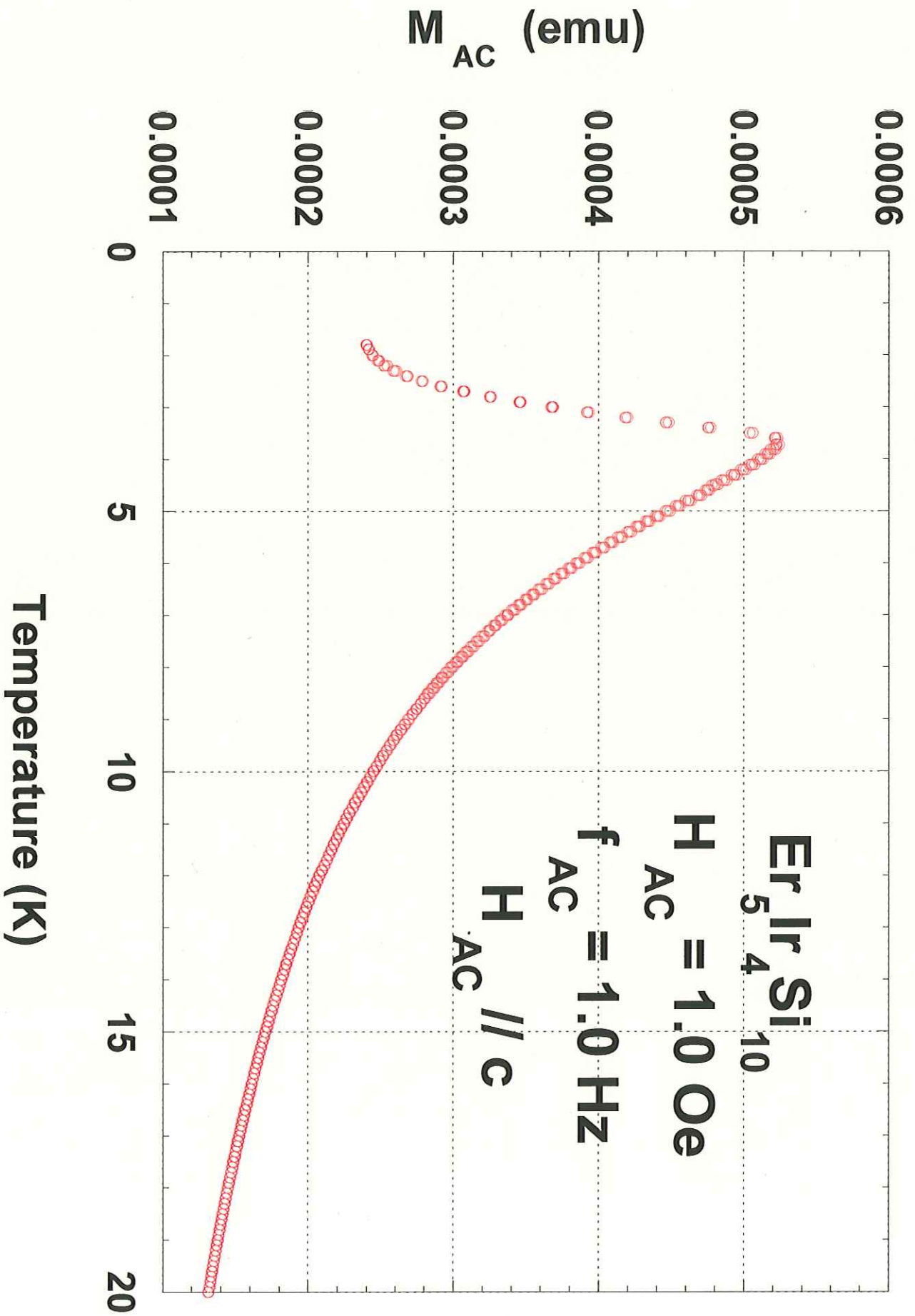
Fig. 4. Temperature dependence of the low-temperature specific heat in the SSE processed $Er_5Ir_4Si_{10}$ single crystal.
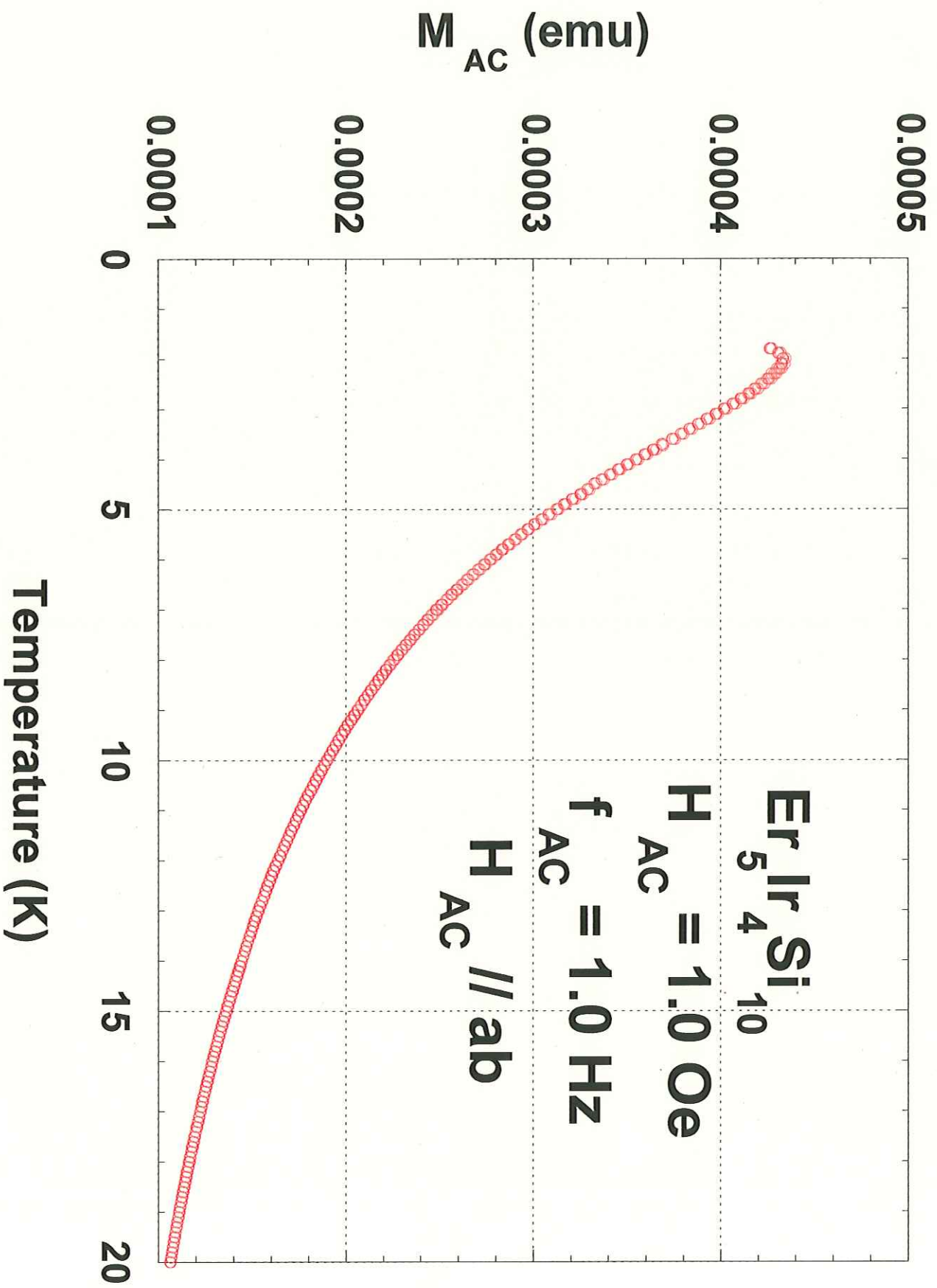
Fig. 5. Frequency dependence of the AC magnetization when the magnetic field direction is perpendicular to the c-axis. The measurements have been performed at 1.0, 10, 100 and 1000 Hz, respectively. No spin glass behavior has observed. We must note that no spin glass behavior clarify non-existence of the disorder in our SSE processed single crystal.
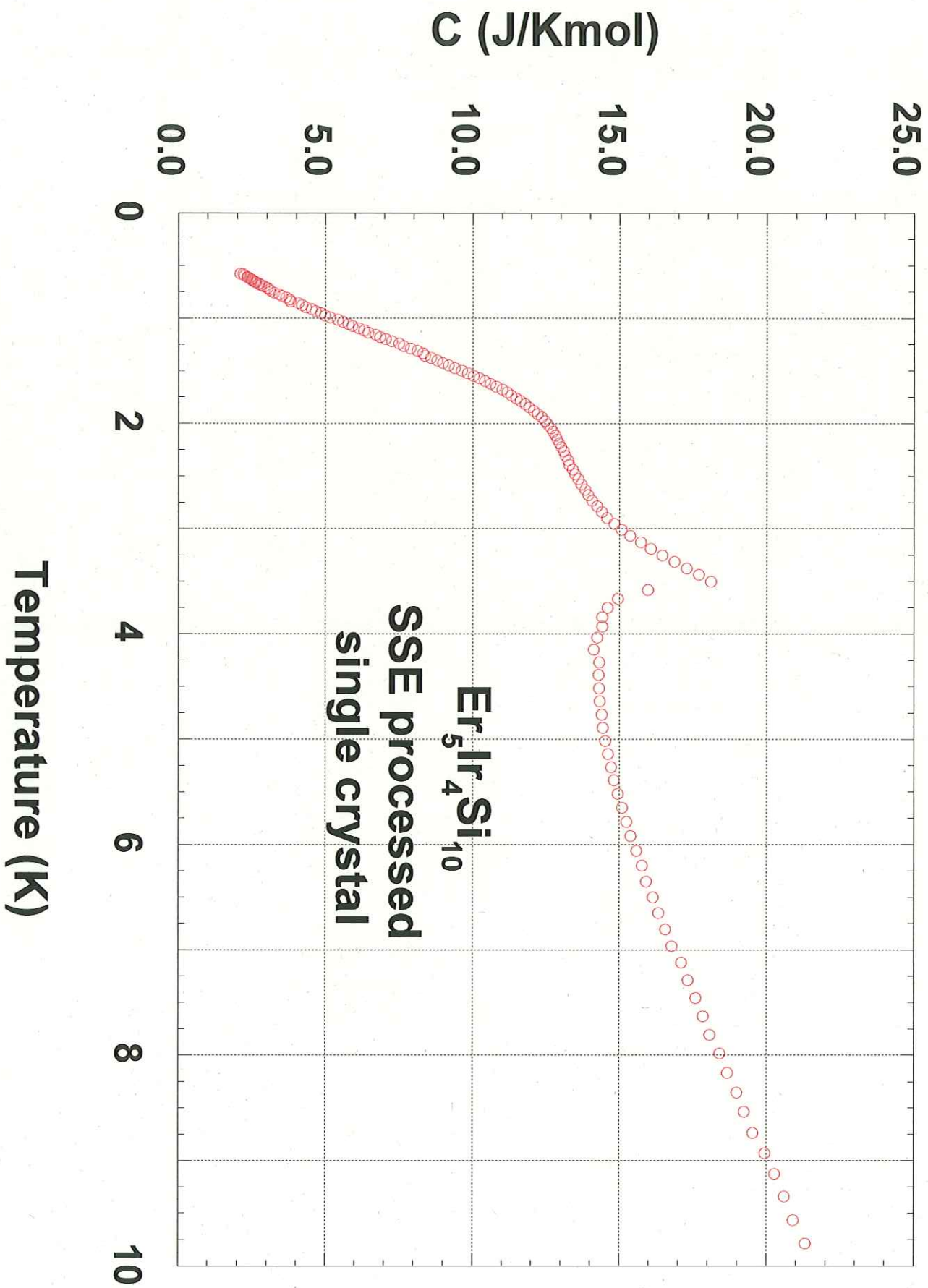
Fig. 6. Temperature dependence of the low-temperature specific heat in an as-grown single crystal of $Er_5Ir_4Si_{10}$ grown by Czochoralsky pulling method. We must note that two successive peaks are observed.

Figure(s)

M $_{AC}$ (emu)

Temperature (K)

Er$_5$Ir$_4$Si$_{10}$
H$_{AC}$ = 1.0 Oe
f$_{AC}$ = 1.0 Hz
H$_{AC}$ // c

C (J/Kmol) vs Temperature (K)

$Er_5Ir_4Si_{10}$
SSE processed
single crystal

M_{AC} (emu)

0.0025

0.002

0.0015

0.001

0.0005

0

f_{AC} = 1.0 Hz

f_{AC} = 10 Hz

f_{AC} = 100 Hz (above), f_{AC} = 1000 Hz(below)

$Er_5Ir_4Si_{10}$

H_{AC} = 1.0 Oe
H_{DC} = 0.0 Oe
H_{AC} // ab

Temperature (K)

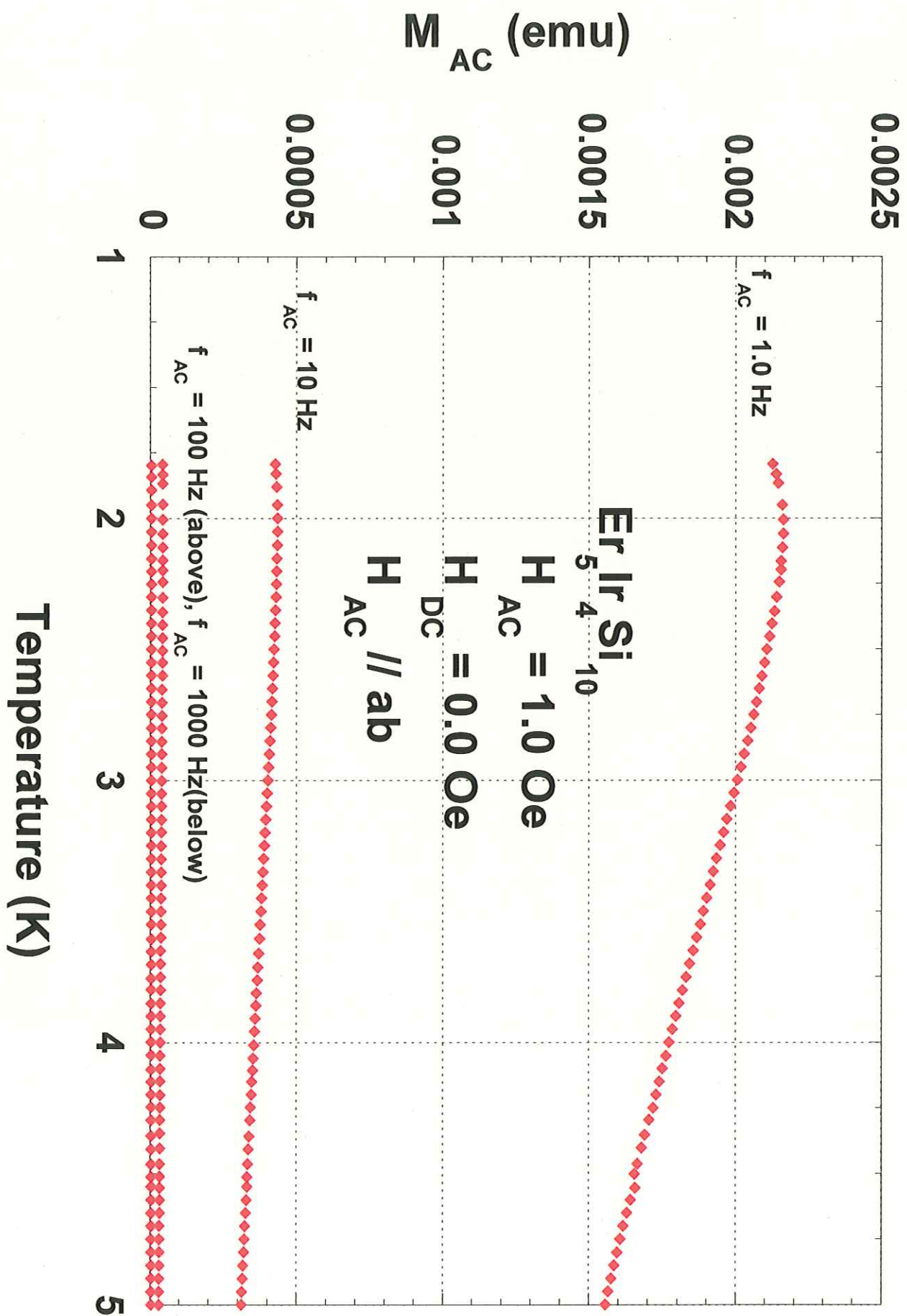0   1   2   3   4   5
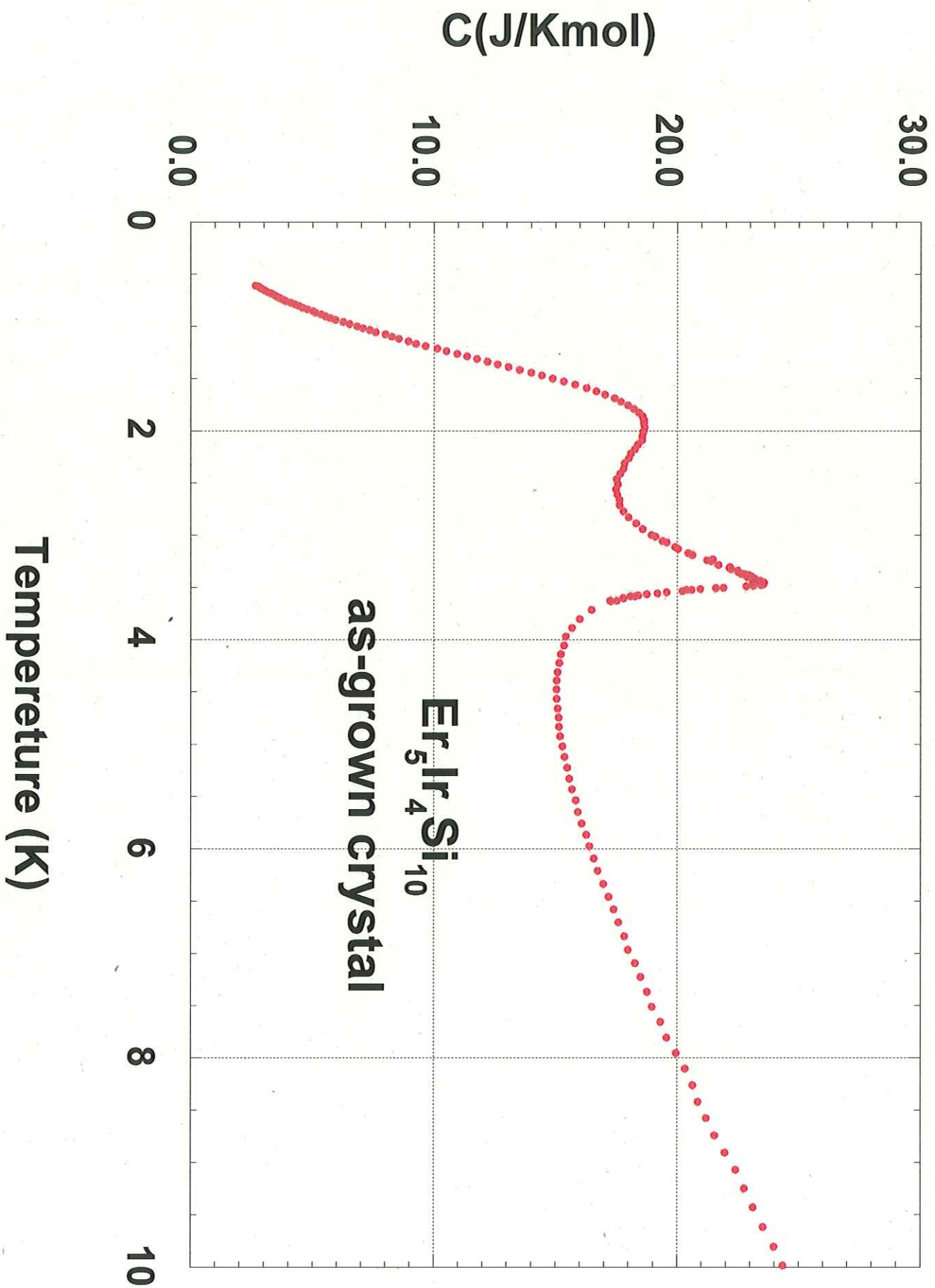
Er$_5$Ir$_4$Si$_{10}$
as-grown crystal

Dear Reviewer

 Thank you very much for your many kind criticisms on my first manuscript. On the benefit of your many comments and criticism, I revised my article.

Thank you very much.

With my best regards

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