Elsevier Editorial System(tm) for Journal of Alloys and Compounds
Manuscript Draft

Manuscript Number:

Title: The role of crystal chemistry in YbCu5−xAux

Article Type: Full Length Article

Keywords: A. intermetallics. C. magnetoresistance. C.kondo effect.

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Abstract: The role of crystal chemistry in YbCu5-xAux crystallizing in the FCC AuBe5-type structure is highlighted through the study of the $0.7 \geq x \geq 0.4$ region. Magnetic frustration, due the magnetic Yb-ions forming a network of edge-sharing tetrahedra, seems to play an important role in the evolution of the ground state of the system. Significant differences are found among the YbCu5-xAux compounds compared to the cubic isotypic systems YbCu5-xAgx and YbCu5

Prime Novelty Statement

To

The Editor in chief of

Journal of Alloys and Compounds

In the submitted manuscript, we are for the time reporting results on the study of a new set of samples prepared at x= 0.4, x=0.5, x=0.6 and x=0.7 of the $YbCu_{5-x}Au_x$ solid solution. New hints and a new perspective are evident from this study. For instance we have clearly shown that the properties of the sample at x=0.4 deviates from those of the other samples because it is on the inferior border of the solid solution. Even most importantly, for the first time the role of magnetic frustration in the evolution of the ground state of this cubic fcc system is evidenced, at difference from the other analogous system $YbCu_{5-x}Ag_x$, where physics is dominated by Kondo effect and crystal field splitting. In fact, for $YbCu_{5-x}Au_x$ we evaluated the Kondo temperature from scaling analysis of magnetoresistence data, and for the first time its value was found to be very low through the entire system, at difference from previous thinking (of T_K = 40 K for low x-values), proving that Kondo interactions is not very important for the system, and magnetic frustration competes with RKKY interactions for defining the ground state. In view of these facts, we think that the publication of this work in Journal of Alloys and Compounds will be valuable.

With many thanks

Yours Sincerely

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The role of crystal chemistry in $YbCu_{5-x}Au_x$

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Abstract

The role of crystal chemistry in $YbCu_{5-x}Au_x$ crystallizing in the FCC $AuBe_5$-type structure is highlighted through the study of the $0.7 \geq x \geq 0.4$ region. Magnetic frustration, due the magnetic Yb-ions forming a network of edge-sharing tetrahedra, seems to play an important role in the evolution of the ground state of the system. Significant differences are found among the $YbCu_{5-x}Au_x$ compounds compared to the cubic isotypic systems $YbCu_{5-x}Ag_x$ and $YbCu_5$

Key words: A. intermetallics. C. magnetoresistance. C.kondo effect.

1 Introduction

Heavy fermions are known to show rather rich phase diagrams with phase transitions which depend on the relative magnitude of some competing energy scales. A qualitative scenario for explaining the ground state properties of heavy fermions was proposed by Doniach [1]. In this scenario two important energy scales are in competition, the Kondo interaction and the inter-site Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between local moments via the conduction electrons. If Kondo interaction is small compared to RKKY interaction, a magnetic ground state is favored, vice-versa a magnetic order is suppressed by strong Kondo interaction due to the screening of magnetic moments by conduction electrons. Chemical composition, as well as pressure and magnetic field, can play an important role in the competition between these two interactions and, consequently, in the ground state evolution of these compounds.

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In some cases, magnetic ordering can be inhibited also by the strong frustration of magnetic interactions. In fact, there is a growing evidence that frustration is an important additional tuning parameter in the Kondo lattice model of heavy fermion materials. Thus, more general phase diagrams extending the qualitative scenario proposed by Doniach to include frustration effects have been proposed [2-4].

Until a few years ago frustration in metals was not expected to play an important role due to the fact that for intermetallic compounds exchange interactions cannot be reduced to nearest neighbors (nn), and the effect of including additional couplings besides nn was shown, in case of localized spin systems, to remove the degeneracy of the ground state [5]. Nevertheless, in recent times various metallic systems showing frustrated behaviour have been studied and novel states of matter induced by frustration were suggested for some of these systems [5].

In this work we will present the case of the $YbCu_{5-x}Au_x$ system as an example where various aspects related to crystal chemistry (such as competition in compound formation and geometric frustration) have to be considered in the study of ground state properties of materials. The interest in the cubic $YbCu_{5-x}T$ (T = Au, Ag) systems crystallizing in the $AuBe_5$-type was triggered by the previous study of the heavy fermions $YbCu_4Au$ and $YbCu_4Ag$ [6]. These compounds were initially supposed to be stoichiometric and subsequently it was clear that they are points of crystallographic order of $YbCu_{5-x}T$ solid solutions. A comparison between these two systems is very useful since Cu/Au and Cu/Ag substitution are both isoelectronic. This is done in section 2.1, where some peculiar features of two ordered compounds are compared with $YbCu_5$, which is the common point of the solid solutions with x=0. In section 2.2 literature data on $YbCu_{5-x}Au_x$ are reported and finally in section 4 we present new results on the investigation of $YbCu_{5-x}Au_x$ for x=0.4,0.5,0.6,0.7.

2 Literature data

2.1 $YbCu_5$, $YbCu_4Au$, $YbCu_4Ag$: a brief comparison

In a recent update of the Yb-Cu phase diagram at ambient pressure [7] the compound $YbCu_5$ is not reported since it forms as cubic $AuBe_5$ crystal structure only at high pressure [8] or by melt spinning [9]. Instead, at compositions very close to $YbCu_5$ (1.5 at. % Yb shifted towards Yb-side), the $YbCu_{4.5}$ compound exists in a stable range of temperature (it melts congruently at the relative maximum of 935°C). The crystal structure of the two compounds are closely related since $YbCu_5$ crystallizes in the cubic $AuBe_5$-type and $YbCu_{4.5}$ forms in a complex monoclinic superstructure of several thousands of atoms per unit cell derived from the cubic $AuBe_5$-type via the introduction of planar defects parallel to {hhh}[10].

Physical properties of the cubic $YbCu_5$ prepared under high pressure were investigated and show heavy fermion behavior without magnetic order and with an electronic Sommerfeld coefficient of 550 mJ/mol K^2 [11].

$YbCu_4Au$ and $YbCu_4Ag$ crystallize in the cubic $MgCu_4Sn$-type (an ordered ternary derivative of the $AuBe_5$-type). Significant differences exist between the two compounds from the viewpoint of physical properties, mainly due to the difference in Kondo interactions. In fact, the crystalline field splitting in $YbCu_4Ag$ (on the order of 80 K) appears to be small compared to the Kondo temperature (T_0=150 K), whereas the opposite is valid for $YbCu_4Au$ which shows a $T_0 \approx 2K$ [12]. Consequently, $YbCu_4Au$ orders magnetically below 1 K, whereas no order was found for $YbCu_4Ag$ down to lowest temperatures [6]. The magnetic structure of $YbCu_4Au$ has been studied by elastic neutron scattering and it was found an incommensurate antiferromagnetic structure with a propagation vector k=(0.553,0.415,0.303). On the basis of a spiral structure as the best model, the ordered magnetic Yb moment are considerably reduced with respect to those expected for the ground state doublet [13]. Finally, a study made on the $YbCu_4$ (Ag,Au) solid solution reveals that the differences in physical properties between $YbCu_4Au$ and $YbCu_4Ag$ cannot be attributed only to chemical pressure, but may arise from a different hybridization between the two border compounds [14].

2.2 $YbCu_{5-x}Au_x$ vs. $YbCu_{5-x}Ag_x$ systems

After the studies on the above-mentioned heavy fermions, it was realized that $YbCu_4Au$, like the analogous $YbCu_4Ag$, is actually a point of crystallographic order of a $YbCu_{5-x}Au_x$ solid solution. Starting from the ordered lattice of $YbCu_4Au$, where Yb, Cu and Au atoms are located on the 4*a*, 16*e* and 4*c* crystallographic sites of the $F\bar{4}3m$ space group, in $YbCu_{5-x}Au_x$, by decreasing values of *x*, Cu substitute Au in the 4*c* site up to the lowest value of *x* [15].

The differences in $YbCu_4Ag$ and $YbCu_4Au$ ordered compounds reflect also in the $YbCu_{5-x}Ag_x$ and $YbCu_{5-x}Au_x$ solid solutions. At first, let us consider the compound formation of the cubic phase. In the first system this phase is stable up to the very low limit of solubility x=0.15 [16], whereas in $YbCu_{5-x}Au_x$ the cubic phase become already unstable for values below x=0.4 [15]. This last feature is shown in Fig. 1 which reports a detailed study in the Yb-Cu-Au system of the homogeneity field for the cubic $AuBe_5$-type. For the $YbCu_{5-x}Au_x$ system a homogeneity range of $0.4 \leq x \leq 1.8$ was found, whereas for lower values of *x* down to x=0 its monoclinic superstructure is formed [7]. From Fig. 1 one can see that for compositions around 7 at. % Au, corresponding to $x \approx 0.4$, there is a small overlapping region where the two phases are stable at stoichiometries of Yb shifted of 1.5 at. %.

Concerning the physical properties, in case of $YbCu_{5-x}Ag_x$ the characteristic temperature T_0 of the Kondo effect decreases continuously from 150 K to 70 K in the $1 \geq x \geq 0.15$ range with a corresponding enhancement of the Sommerfeld value from 210 mJ/mol K^2 in $YbCu_4Ag$ to 460 mJ/mol K^2 for the lower limit, this last value being close to the value of 550 mJ/mol K^2 of $YbCu_5$. In case of $YbCu_{5-x}Au_x$, the Kondo temperature slightly increases by x decreasing but its values keep always low. On the other hand, the γ values, already considerable for $YbCu_4Au$ (γ = 1 J /mol K^2), reach typical values of very high heavy fermions in the lower limit of x [17].

Most of attention in the $YbCu_{5-x}Au_x$ system was focused in searching for a quantum phase transition (QPT) separating a magnetically ordered ground state at x=1 from a non-magnetic heavy fermion (shown virtually at x=0). In fact, starting from $YbCu_4Au$, with substitution of Au by Cu in the 4c-site (i.e. for decreasing x-values), a decreasing of T_N was inferred from resistivity and specific heat measurements [18, 19]. Since Cu is smaller than Au, this evolution cannot be due to chemical pressure as it occurs in case of the system with Ag. The similarities between the XRD patterns of $YbCu_{4.5}$ and $YbCu_5$ (see Fig. 2) lead to a misintepretation of some data. In particular, the existence in this system of a lower limit for the cubic phase already at x=0.4 make questionable the finding of a possible QPT around (x=0.2−0.4) evaluated from resistivity measurements [18]. In fact, in contrast with this claiming, a recent NQR and NMR measurement done on the sample at x=0.6 found no evidence of a magnetic long range order down to 20 mK [20].

In order to extract more information on the evolution of the ground state of this intriguing system we were motivated to prepare a new set of samples in the region close to the inferior limit of existence of the cubic phase. In the following of the paper we report some of our results obtained on the investigation of the $YbCu_{5-x}Au_x$ system in this region.

3 Experimental details

The polycrystalline samples of $YbCu_{5-x}Au_x$ (x = 0.4,0.5,0.6,0.7) have been prepared by weighting the stoichiometric amount of elements with the following nominal purity: Yb (99.9 pct mass), Cu (99.999 pct mass) and Au (99.99 pct mass). The elements were enclosed in small tantalum crucibles, sealed by arc welding under pure argon, in order to avoid the loss of Yb with a high vapor pressure. The samples were then melted in an induction furnace, under a stream of pure argon. To ensure homogeneity during the melting, the crucibles were continuously shaking. The samples were then annealed in a resistance furnace at 700°C for two weeks and finally quenched in cold water. The alloys were characterized by optical and electronic microscopy and by quantitative electron probe microanalysis (EPMA). The crystalline structure was examined by X-ray diffraction (XRD).

Electrical resistivity was measured with PPMS device using 4-wire AC technique in 0.4−300 K temperature range. Magnetic susceptibility (at excitation field 100 Oe) and magnetization were measured by MPMS commercial device (Quantum Design) in the temperature range 2−300 K and in an applied magnetic field up to 5 T.

4 Results and Discussion

Fig.2 reports XRD powder patterns for the four samples, together with data for x=0.2 taken from our previous work [7]. The patterns of the samples prepared in this work were indexed on the basis of the cubic $AuBe_5$-type. In particular, the satellite peaks of the monoclinic superstructure of the cubic $AuBe_5$-type, present for the alloy at x=0.2, are absent in all the XRD patterns. Fig. 3 shows the variation of the lattice parameter a of the cubic $AuBe_5$ phase of $YbCu_{5-x}Au_x$ as a function of Au content compared with data taken from the literature. The values for the samples prepared in this work are in line with those reported in the literature [15,18]. As it was already evidenced in a previous paper [7], the change of slope at the stoichiometric point of $YbCu_4Au$ is due to the fact that YbCu4Au is at the crossover between two kinds of Cu/Au substituted sublattices, 16c for x>1 and 4c for $x\leq1$.

The inverse magnetic susceptibility $1/\chi(T)$ for the samples of the $YbCu_{5-x}Au_x$ solid solution is shown in Fig.4 as a function of temperature between 2 K and 300 K. All the compounds exhibit Curie-Weiss (CW) behavior above 50 K and the susceptibility data can be accounted for with a CW law and a Pauli-like contribution χ_0:

$$\chi = \chi_0 + \frac{C}{(T - \Theta_p)} \qquad (1)$$

For all the samples the value of the effective moments are close to the free ion value 4.54 μ_B. Furthermore, the paramagnetic Curie-Weiss temperature Θ_p is negative and its absolute value increases by decreasing x. This is shown in Table 1 where Θ_p is compared with the temperature T_M (T_N for x=1), which is the temperature of the anomaly of the specific heat measurements reported elsewhere [17]. One can notice that $|\Theta_p|$ is much larger than T_M. For a non frustrated magnet it is expected to be $|\Theta_p| \approx T_M$, whereas frustrated magnets are characterized by $|\Theta_p| \gg T_M$. An empirical measure of frustration was defined as $f = -\Theta_p/T_M$ where T_M represents any cooperative-ordering transition temperature [21]. For T<20 K the inverse magnetic

susceptibility starts to deviate from CW behavior due to crystal field splitting thus, in an evaluation of the empirical parameter *f* for this system, it is more appropriate to use the Θ_p value extracted at low temperature which involves only the doublet ground state. Proceeding in this way we obtained an increase of *f* from 1.9 to 8.4 in the $0.7 \geq x \geq 0.4$ compositional range [17].

These results indicate that the $YbCu_{5-x}Au_x$ solid solution has all the characteristics of a system where frustration plays an important role, also supported by specific heat measurements, which reveals a change of the ground state from a long range order, for x=1 and x=0.8, to one with a high density of excitations [17]. In fact the crystal structure of the system is a face-centered cubic (FCC) lattice with a $AuBe_5$ structure type. As seen in Fig. 5 the FCC lattice can be viewed as a network of edge-sharing tetrahedra with the Yb magnetic ions located at the vertices. This is indeed a three-dimensional analogue of a triangular lattice. Analogous cubic FCC compounds RCu_4In (R = Gd, Dy, Ho and Er), isotypic with $YbCu_{5-x}Au_x$, show negative Θ_p significantly larger than their respective Néel temperatures, and exhibit strong signs of geometrical frustration [22].

In Fig.6 the temperature variation of the electrical resistivity in the $YbCu_{5-x}Au_x$ system are depicted, compared with previous literature data at very low temperatures (see inset of that figure) and complemented with literature data for x=0.8 and 1 [18]. From the figure we can notice that, whereas the ordered composition $YbCu_4Au$ exhibits typical Kondo lattice features, incoherent Kondo scattering develops for all the other compositions. Furthermore, for T<1 K the curves have a tendency to flatten as the temperature decreases (see the inset of Fig. 6). The effect of geometrical frustration seems to be stronger for low values of *x*, where the electrical conductivity at low temperatures is lower. This may be due to the change of RKKY-interactions which compete with the frustration effect. The sample prepared at x=0.4 behaves differently either from literature data than from our measurements. In fact, the resistivity at T around 0.1 K of the two samples at x=0.4 and 0.5 are very close (see inset of Fig.6), and for the sample prepared in this work the trend of ϱ (T) deviates from the trend of the other samples already for T around 10 K. The reason of this deviation is that in this region a mixture of the two phases is probably formed (as indicated in Fig. 1). We believe that, due to similarity of XRD patterns between the cubic and its monoclinic superstructure, even a 10−20% of a monoclinic phase may be present together with the cubic phase without showing the extra peaks of the superstructure.

The magnetoresistance (MR) was measured at various constant temperatures as a function of magnetic field H (up to 9 T) for $YbCu_{4-x}Au_x$ (x=0.5, 0.6, 0.7). The relative MR [(ϱ (H)- ϱ (0))/ ϱ (0) * 100] of all the samples measured is negative for all fields and temperatures studied, and its absolute values increases by decreasing of temperature and increasing of magnetic field. This trend is consistent with a single impurity Kondo behavior. As an example, the relative MR for the sample at x =0.7 at different temperatures ranging from 0.4 K to 20 K is presented in the inset of Fig. 7. The absolute value of the relative MR reaches about 60% at 1 K in a field of 9 T, suggesting a low Kondo temperature (T_K). In order to take profit of these

measurements, we have evaluated T_K performing a scaling analysis derived from the Bethe-ansatz studies [19]. In particular, the relative MR at different temperatures was scaled to a single curve by plotting it as a function of H/(T + T*), where T* is an energy scale related to T_K. The values of T* were adjusted in order to get the best overlap. This scaling procedure is shown in Fig. 7 for the sample $YbCu_{4.3}Au_{0.7}$. The relative MR(H) isotherms measured above T ≥ 0.8 K collapse onto a single curve with T* = 1.0 K. A change in T* by ±0.3 K makes this overlap considerably worse. The fact that the MR isotherm at T = 0.4 K does not overlap well is probably due to the vicinity of the magnetic transitionat T ≈ 0.35 K. The same scaling analysis was performed on samples with x = 0.6 and x = 0.5. We obtain good overlaps of relative MR isotherms above T≥ 0.8 K with slightly increasing values of T* up to 1.8 K by decreasing of *x* down to *x* = 0.5. These remarkably low values for a Kondo energy scale extracted from these MR data are consistent with the values evaluated from specific heat [17] by applying the Desgranges and Schotte [24] criterion used for Kondo impurities and valid for a doublet ground-state. Comparatively, a much higher Kondo scale (T_K ≈ 40 K) was obtained from MR measurements on $YbCu_{4.8}Au_{0.2}$ [19] due to the fact that this alloy is within the mentioned superstructure domain and therefore it may behave differently.

5. Conclusions

A new set of samples of $YbCu_{5-x}Au_x$ for *x*=0.4, 0.5, 0.6, 0.7 was synthesized and characterized in order to extract more information on the evolution of the ground state of this system. We have shown that a detailed crystallographic investigation of the cubic $AuBe_5$-type $YbCu_{4-x}Au_x$ solid solution provides valuable information on the homogeneity range close to the inferior limit and, therefore, on the interpretation of the physical properties of the system close to this limit. In particular, it was shown that the sample prepared at *x* = 0.4 exhibits deviations of physical properties compared to the samples with compositions x>0.4 likely due to the presence of a minor part of the $YbCu_{4.5}$-based monoclinic phase.

Significant differences were found among these compounds compared to the cubic isotypic $YbCu_{5-x}Ag_x$ and $YbCu_5$. In fact, whereas in $YbCu_{5-x}Ag_x$ the ground state is that of a Fermi liquid Kondo lattice, and vary continuously from *x*=1 down to the lowest limit of *x* = 0.15 (tending to merge with the physical properties of $YbCu_5$), in the case of $YbCu_{5-x}Au_x$ magnetic frustration seems to play a significant role in the evolution of the ground state. The Kondo scale, evaluated from a scaling analysis on MR data, increases slightly by *x* decreasing but it remains always ≤ 2 K, in contrast to what was previously thought. Therefore in this system the Kondo effect is much less important compared to the case of Ag and, starting from $YbCu_4Au$, the effect of frustration seems to increase by decreasing of *x*, i.e. by increasing of atomic disorder, causing an evolution of the

ground state in the system towards a system without long range magnetic order. Furthermore, the properties in the low region of *x* are clearly different from the properties of the cubic $YbCu_5$ synthesized at high pressure.

Acknowledgements

This work was supported by the project VEGA 2/0070/12 and 2/0106/13, CFNT MVEP - the Center of Excellence of the Slovak Academy of Sciences and the EU ERDF-ITMS26220120047 and ITMS26220120005. The liquid nitrogen for the experiment was sponsored by the U.S. Steel Kosice, s.r.o .

M G and J S acknowledge the VI Executive Program of Scientific and Technological Cooperation between Italy and Argentina for the years 2014-2016. J S acknowledges FONCyT support through Project PICT 1060-2010 and UNCuyo through Project 06/C457.

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Captions

Figure 1: Range of compositions of $YbCu_{5-x}Au_x$ and $YbCu_{4.5}$-based solid solutions

in a schematic isothermal section at 700°C of the Yb-Au-Cu system (from [7]).

Figure 2: (Color online) X-ray diffraction (Cu K_α) powder patterns of $YbCu_{5-x}Au_x$ of (a) x=0.2 [7] (b) x=0.4 (c) x=0.5 (d) x=0.6 (e) x=0.7

Figure 3: Lattice parameters a versus x for $YbCu_{5-x}Au_x$.

Figure 4: (Color online) Magnetic susceptibility vs. T of $YbCu_{5-x}Au_x$ for x=0.4−0.7. Inset: Temperature dependence of inverse susceptibility.

Figure 5: (Color online) (a) Crystal structure of $YbCu_4Au$ seen as edge-sharing tetrahedra with Yb ions located at the vertices (unit cell indicated by black lines).

(b) Schematic magnetic moments interacting antiferromagnetically in a tetrahedral environment

Figure 6: (Color online) Temperature dependence of the electrical resistivity of $YbCu_{5-x}Au_x$ (x=0.4,0.5,0.6,0.7) complemented with literature data for x=0.8 and x=1 [18]. Inset: Low temperature resistivity data for (x=0.4,0.6,0.8,1) taken from [18].

Figure 7: (Color online) Relative magnetorsistance vs H/(T+T*) for $YbCu_{4.3}Au_{0.7}$: T* = 1 K. The inset shows the relative magnetoresistance vs H at different temperatures.

Table 1: Magnetic data of $YbCu_{5-x}Au_x$.obtained by CW fitting above 50 K (a) These data were taken from [17]; (b) data from [18]. T_M indicates the temperature of the specific heat maxima for $x \leq 0.7$.

x	Θ_p	$T_N(T_M)$[(a)]
0.4	-17 K	0.15 K
0.5	-13 K	0.16 K
0.6	-11 K	0.30 K
0.7	-10 K	0.35 K
1.0[(b)]	-5.5 K	0.7 K

***Highlights (for review)**

- A new set of samples close to the inferior limit of the cubic phase was prepared
- Physical properties of $YbCu_{4.6}Au_{0.4}$ already deviates from the other samples
- Magnetic frustration seems to play a big role in the system
- Kondo temperature is always less than 2 K at difference from previous data

Figure 1

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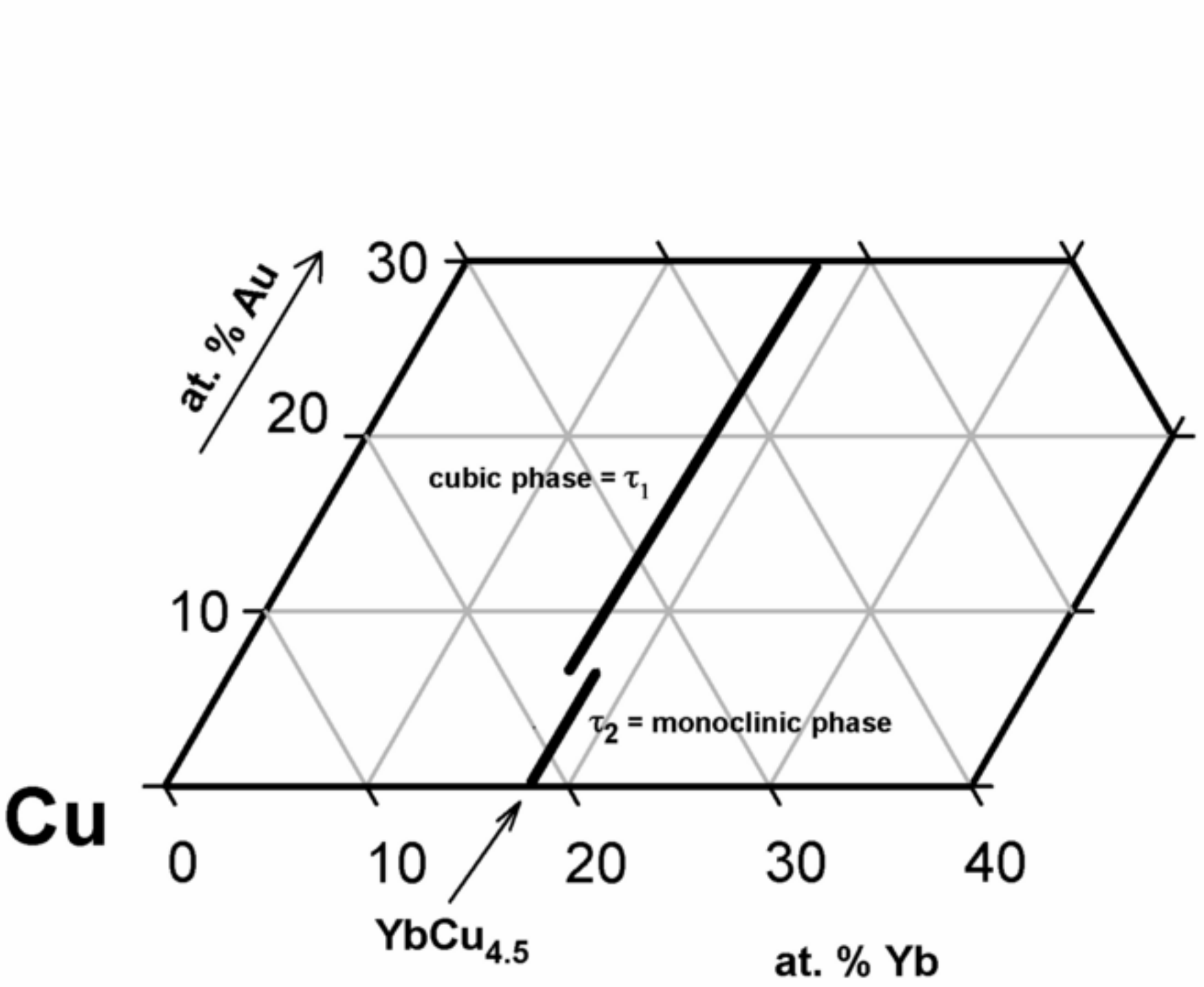

Figure 2

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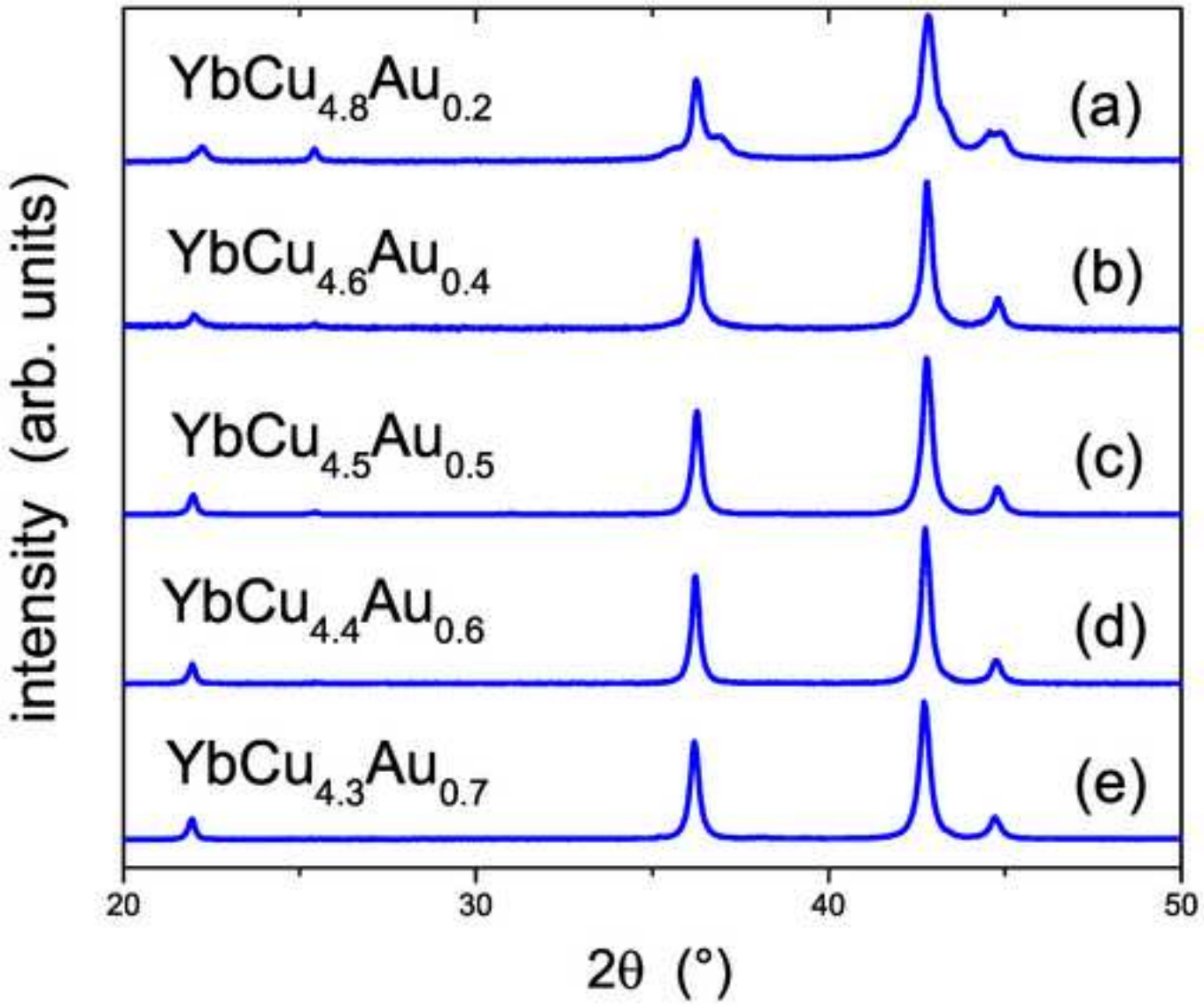

Figure 3

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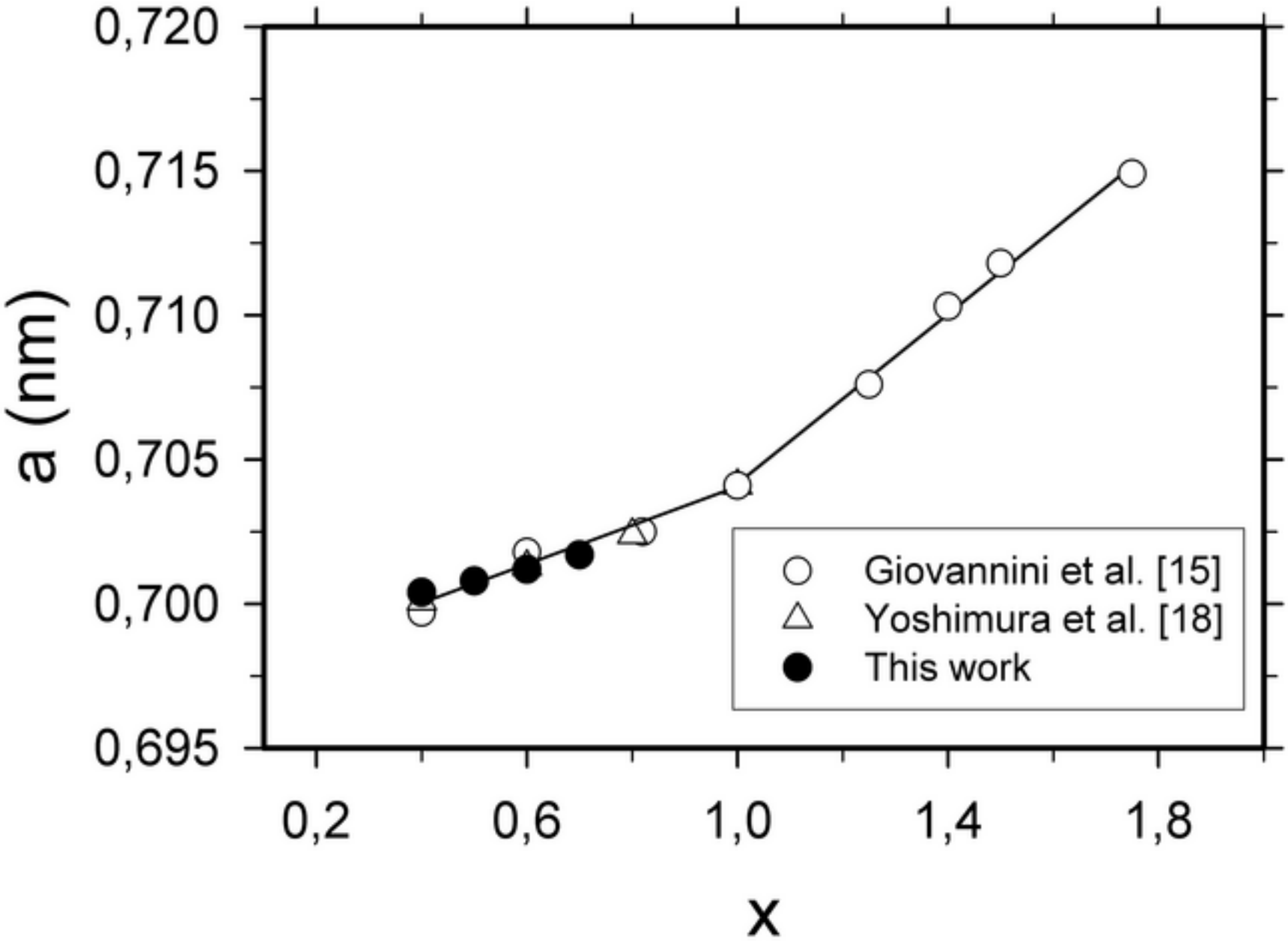

Figure 4

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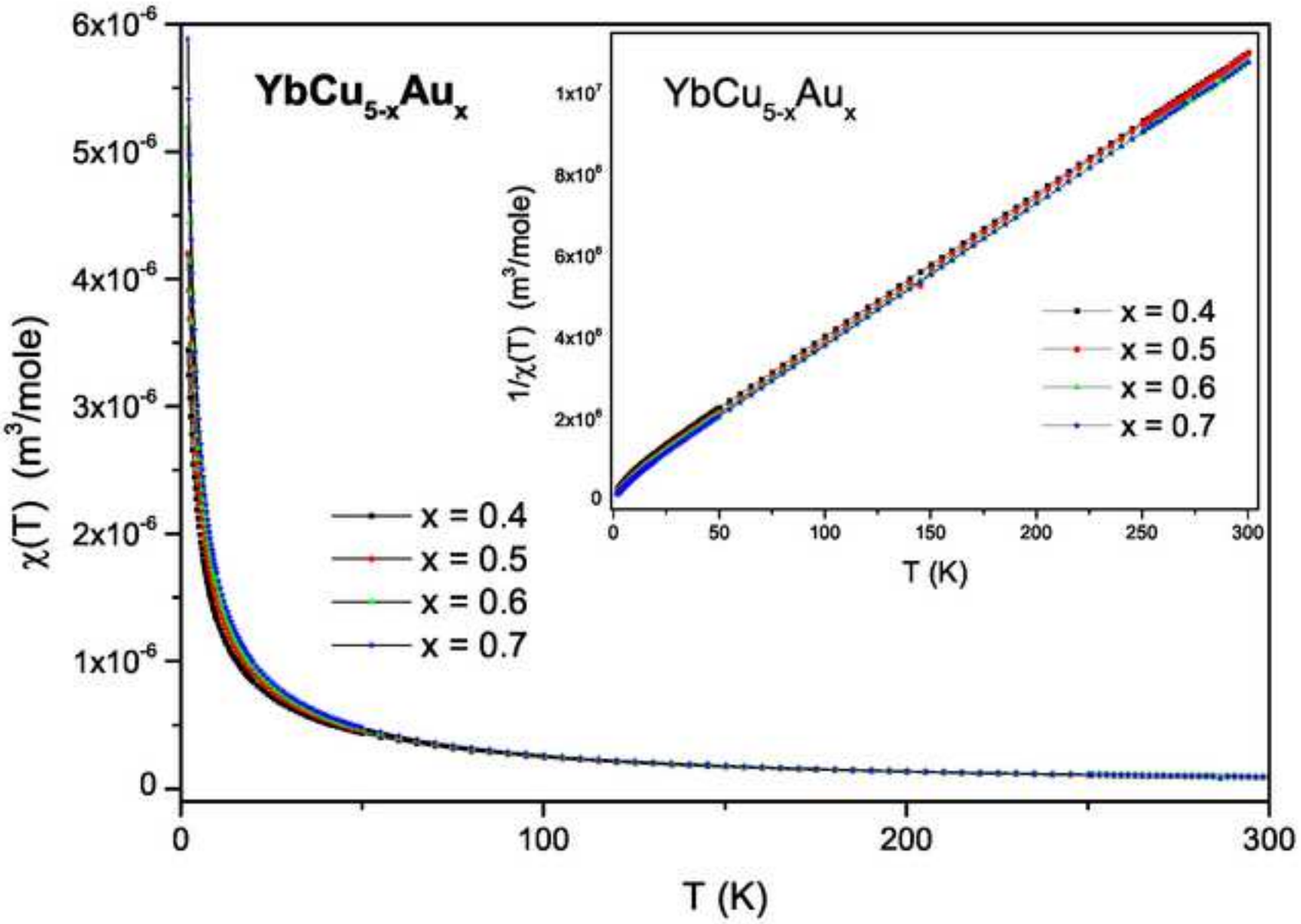

Figure 5A

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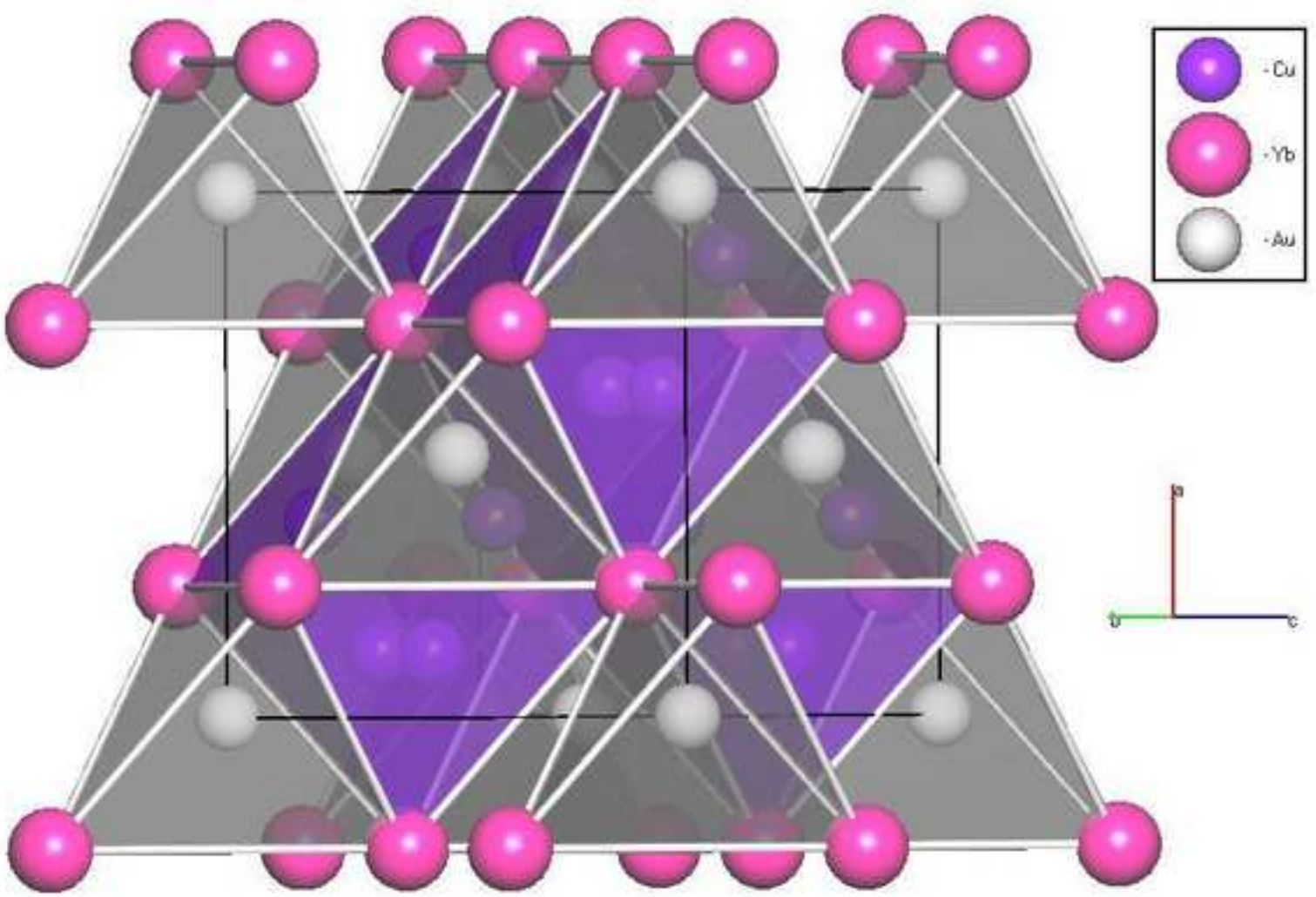

Figure 5B

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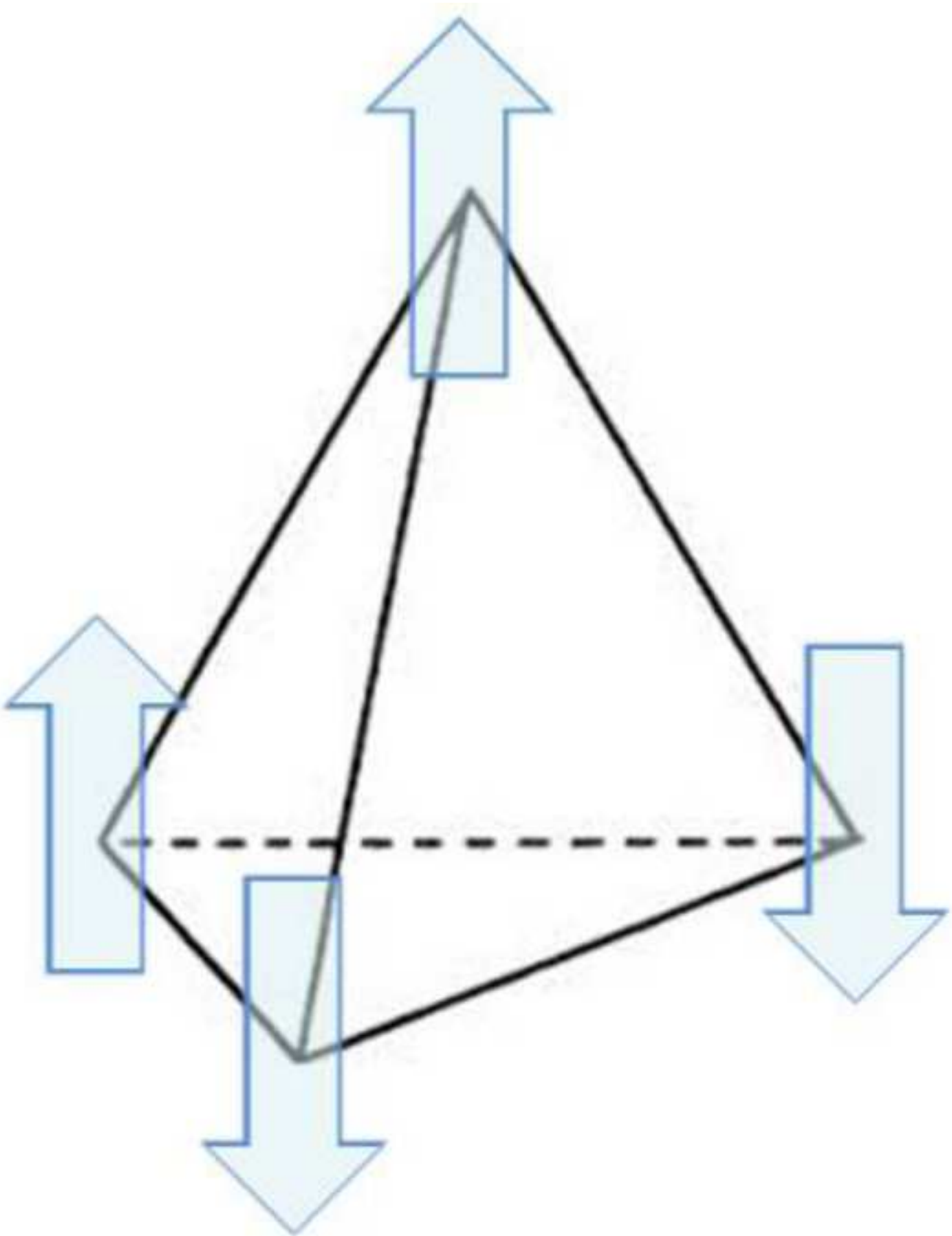

Figure 6

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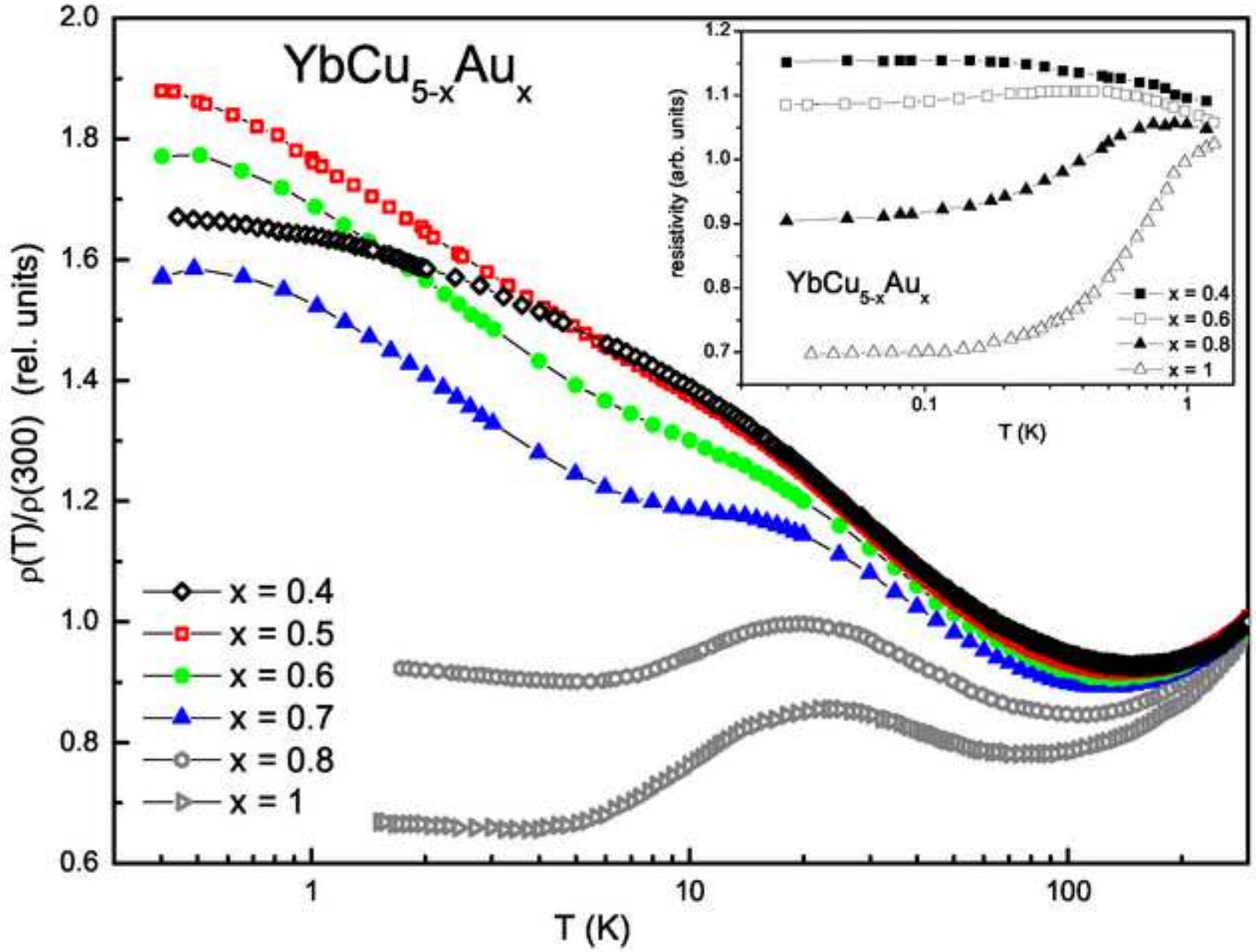

Figure 7

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