

# Influence of the electronic structure on the plastic properties of the single crystal $\text{YbxY}_{1-x}\text{InCu}_4$ and $\text{YbAgyIn}_{1-y}\text{Cu}_4$ systems

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Source / Izvornik: *Croatica Chemica Acta*, 2005, 78, 627 - 632

Journal article, Published version

Rad u časopisu, Objavljena verzija rada (izdavačev PDF)

Permanent link / Trajna poveznica: <https://urn.nsk.hr/urn:nbn:hr:217:631045>

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Download date / Datum preuzimanja: **2025-01-08**



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## Influence of the electronic structure on the plastic properties of the single crystal $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ and $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$ systems\*

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RECEIVED NOVEMBER 5, 2004; REVISED JUNE 6, 2005; ACCEPTED JUNE 7, 2005

### Keywords

- Influence of electronic structure
- Vickers microindentation hardness testing
- Single crystal of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  systems with  $0 \leq x, y \leq 1$ 
  - Lattice parameter
  - Electrical resistivity

We report the results of Vickers microindentation hardness measurements on two series of the alloy systems:  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$ , where the parameters  $x$  and  $y$  vary from 0 to 1. In both systems the microhardness decreases with increasing the concentration parameters  $x$ , or  $y$ . Almost the same behavior exhibits the lattice parameter of these systems, which governs evolution of the systems from semi-metallic towards more metallic character. In accordance with that, we ascribe the observed concentration dependence of the Vickers microhardness data to the change of the electronic structure of these systems.

## INTRODUCTION

It was shown recently that the results of measurements of the Vickers microhardness on numerous polycrystalline alloy systems containing U (5f) or Ce (4f) elements reflect intrinsic properties of the investigated materials,<sup>1</sup> although, it is well known that the grain boundaries: homo-phase or hetero-phase interfaces separating grains, which exist in polycrystalline samples, might exert profound effects on the mechanical and the other properties of the system investigated.<sup>2</sup> In spite of using the polycrystalline samples in the recent work the results of these investigations show: first, that they do not depend significantly on the surface of samples and on the crystallographic orientation and we have observed clear concentration dependence of the microhardness; second, one can compare hardness of various alloy systems among

themselves; third, the results of the measurements can be explained by existing theories. In our searching to reveal possible influence of the open f-shell, which is responsible for many interesting electrical and magnetic properties, on the hardness of materials containing elements with open f-shell, we have investigated now microhardness of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  alloy systems. The spatial extent of f-orbital is reduced relative to d-orbital, and this fact enhances solubility, *i.e.*, mixing with non-magnetic ion in a wide concentration range. Therefore, we have expected that in such systems one could observe the influence of electronic structure on mechanical properties much clearly than in the systems containing elements with the open d-shell. It is well established that the effect of the interface appears in a wide variety of forms and stem from a wide range of sources.<sup>2</sup> There-

\* The paper is dedicated to the late Prof. Dr. Vladimir Šips.

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fore, to avoid as much as possible these effects we used single crystals of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  alloy systems.

Among many available methods for the investigation of mechanical properties micro-indentation hardness (or microhardness) measurement seems to be very convenient and useful technique, which gives a qualitative indication of the strength of material, especially its resistance to a plastic deformation, or resistance to dislocations movement.<sup>3–14</sup>

Both alloy systems  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  crystallize in the same crystallographic system: the cubic C15b,  $\text{MgCu}_4\text{Sn}$  type. The main parameter, which governs the change of the electronic structure in these systems is the lattice parameter, which in both systems generally decreases with the concentration parameters  $x$ , or  $y$ .<sup>15</sup> In the present work, we show the results of the measurements on both systems and we show that in both systems one can observe direct influence of electronic structure on plastic mechanical properties, *i.e.*, on microhardness.

Transport, magnetic and other interesting properties of these systems have been examined and the results were published in numerous papers. Besides the other interesting points, they are valence fluctuating systems at lower  $x$  and  $y$ <sup>15–19</sup> (and references therein). Some of details of concentration dependence of the microhardness, we explain by mixing  $\text{Yb}^{3+}$  and  $\text{Yb}^{2+}$  ions in some alloys.

## EXPERIMENTAL

The single crystal samples of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  (with  $x = 0, 0.1, 0.3, 0.5, 0.7, 0.875, 0.9, 0.95, 1$ ) and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  (with  $y = 0, 0.15, 0.27, 0.5, 0.6, 0.75, 0.8, 1$ ) systems were prepared using a flux method.<sup>16</sup> For the microhardness measurements the samples were mounted onto an epoxy resin holder to facilitate handling during a standard polishing procedure and hardness measurements. The microhardness testing was performed at room temperature using a standard E. Leitz (Wetzlar, Germany) Miniload II apparatus supplied with the 136 ° diamond pyramid indenter. The crystallographic planes {100} were available for microhardness investigation on the as-grown crystals. However, because of the process of polishing the surface, we were not able to control precisely the plane of indentation and a considerable tilting might be present. This fact certainly is one of the reasons for relatively large dispersion of the measured data for a given concentration. Nevertheless, it seems clear that this dispersion does not hide the general concentration dependence of the microhardness in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  systems. Moreover, it was shown recently in numerous similar novel alloy systems containing U and Ce, that the expected general concentration dependence of microhardness was obtained in spite the fact that the samples were poly-crystals.<sup>1</sup>

Initial experiments revealed that microhardness data depended on the load applied on the indenter, but for loads

exceeding 0.981 N microhardness was nearly independent of load. Therefore, in the present work the microhardness measurements were performed with load of 0.981 N, so called HV 0.1. The loading time was 10 s. The magnification used in the micro-hardness tests was constant and equal to 500 X. Average value of microhardness is obtained from twenty indentations on each examined sample and the calculated standard deviation was mainly about 10 % of the mean value. Some samples were brittle, especially for lower  $x$  of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  alloy systems. For these concentrations the length of the indentation diagonal was less reproducible because of cracking around indentations. The indents, which differed much from a quadratic form, have not been taken into consideration.

## RESULTS

The results of the microhardness measurements are presented in Figure 1. Each point in the figure corresponds to a different sample. The error bars, which were approximately 10 %, are omitted for clarity.

In order to connect the microhardness data, *i.e.* mechanical properties with other physical characteristics of the investigated systems, first, we plot the microhardness data along with the lattice parameter data, Figure 2.

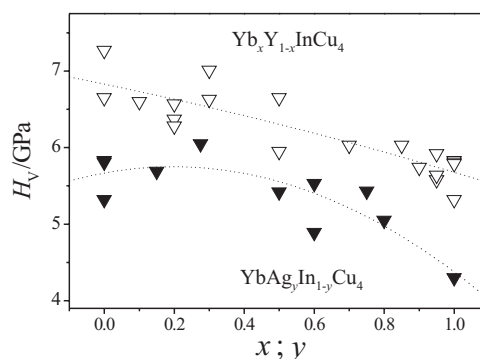


Figure 1. Variation of microhardness of the series of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  (open triangles) and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  (closed triangles) examined systems. The curves drawn in figure are estimated using the quadratic least square fit to the experimental data.

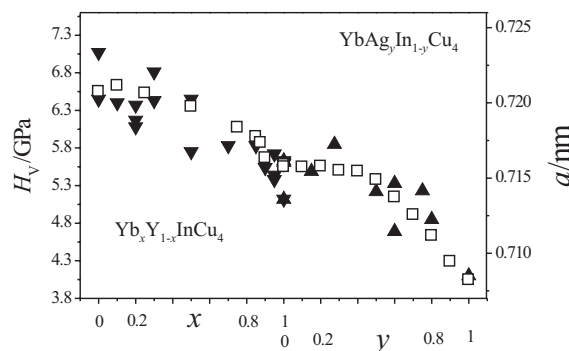


Figure 2. Variation of the lattice parameter of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  single crystals (squares) combined with the microhardness data (triangles).

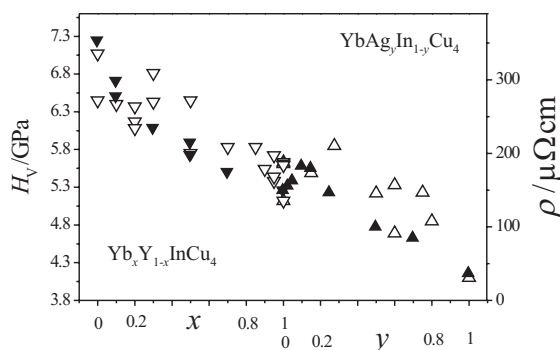


Figure 3. Variation of the room temperature resistivity of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  single crystals (closed triangles) together with the microhardness data (open triangles).

Some relation between these properties might be expected and there can be found in literature many discussions connecting these physical quantities. We also plot the microhardness data, Figure 3, along with the resistivity data. Such a possible connection is seldom seen. However, as we shall see in discussion, we believe that there is an indirect connection between these properties in our systems investigated. Besides, we plot the data in Figure 2 and in Figure 3 in a manner which reveal a property that connects both systems and the both properties, what we shall discuss latter: we plot the data by plotting the systems side by side.

## DISCUSSION

From Figure 1, one can see that there is clear difference between the microhardness of these two systems: the microhardness of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  is certainly larger than the microhardness of  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$ . This is an interesting result. Namely, it is known that there is very difficult to resolve a difference between microhardness of metallic systems. The sample dependency of Vickers microhardness usually exceeds the difference between metallic systems. Also, one may conclude that, although there is certain sample dependency of the data, the Vickers microhardness exhibits clear concentration dependence and it increases with decreasing concentration parameters  $x$  or  $y$ . Such dependence is not expected. In what follows, we shall discuss the obtained results and compare them with the known theories and with the similar results found in literature trying to give an explanation of this concentration dependence.

As one might expect, the microhardness values of the examined systems are rather high, which is in agreement with Mott-Nabarro theory, because these systems contain various kinds of elements with the different atomic radii.<sup>19</sup> Although there is, perhaps, no great sense to compare these values with the microhardness of pure metals, just in order to see that these microhardness values are rather high, we give here some data for the comparison: 0.44 GPa of Cu, 1.08 GPa of Y and 1.7 GPa of Ir.<sup>21</sup> On

the other hand, the microhardness of the diamond is 88 GPa.

According to the Mott-Nabarro theory, hardness of a binary alloy system with atoms of the different sizes should have a maximum at about 50%. Our results show clearly that Yb and Y or Ag are not equivalent and that they do not behave like hard spheres.<sup>20</sup> According to a Rydberg study, one can correlate hardness with the reciprocal of the size of the atoms for pure elements.<sup>22</sup> This conclusion comes out, in principle, from the view that hardness is proportional to cohesive forces. In our case hardness is roughly proportional to lattice parameter (and not its reciprocal), Figure 2. Therefore, our results cannot be explained using this view. Intuitively, hardness is proportional to the density of a material. Again, our results are not in accordance with this expectation. Our system is less dense for lower  $x$ , or  $y$  not only because the lattice parameter increases with decreasing  $x$ , or  $y$ , but also because Y is lighter than Yb and, also, In is lighter than Ag.

In a study of ultra-rapid-quenched 3d transition elements in Al, it was shown that microhardness is proportional to the deviation of lattice parameter from Vegard's law.<sup>23</sup> At present these results we understand within the following picture. The deviations from Vegard's law might be an indication that the nominal concentration of transition metal atoms did not substitute for Al in the lattice. Such transition element atoms, which are not included into substitution positions, form usually precipitates in the sample. The small concentration of precipitates cannot be observed by the Debye-Scherrer method, which was used for the examination of the crystal structure and for the determination of lattice parameters of these alloys. It is known that precipitates are rather effective in pinning dislocations, producing considerable enhancement of microhardness. At first glance, a similar effect might be indicated in the present investigation. A nonlinear behavior of the lattice parameter is observed in both investigated systems. There is a faint maximum at about  $x = 0.1$ , and there is a plateau even up to  $y = 0.5$  inconsistent with Vegard's law expectation. However, as it is shown in Refs. 17 and 19, here these deviations are intrinsic ones and result from the homogenous mixing of the  $\text{Yb}^{3+}$  and  $\text{Yb}^{2+}$  ions in the investigated systems. Knowing the above mentioned fact and the fact that there is the difference of the ionic volumes of  $\text{Yb}^{3+}$  and  $\text{Yb}^{2+}$ , we can speculate using the Mott-Nabarro theory again. The theory suggests than an enhancement of microhardness at lower concentrations. We shall point this fact below, but, certainly, the Mott-Nabarro approach cannot explain the difference in the microhardness among  $\text{YInCu}_4$ ,  $\text{YbInCu}_4$  and  $\text{YbAgCu}_4$ .

Recently, a comprehensive investigation of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  system has been reported.<sup>15</sup> One of the conclusions coming from this study is that the system evolves from semi-metallic towards more metallic character with

increasing  $x$ . Band calculations for  $\text{LuInCu}_4$  indicate that the Fermi surface consists of a hole and an electron band with the Fermi energies  $\varepsilon_F^h = 0.192$  eV and  $\varepsilon_F^e = 0.592$  eV around W and X symmetry points respectively.<sup>24</sup> With increasing lattice parameter, bands overlap and Fermi energies decrease. In fact, electrical resistivity data do suggest that the Fermi energy for  $\text{YInCu}_4$  might be of the order of room temperature. On the other hand, with decreasing lattice parameter bands overlap increase and the bandwidth of  $\text{YbInCu}_4$  has been estimated to be about 5000 K. In accordance with this picture of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  system, we believe that the microhardness data of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  systems might be explained along the same line. It is known that insulators and semimetals are harder than metals. For example, in the Group IV elements: C, Si, Ge, Sn, Pb, Bi, where metallic character increases with increase in atomic number, microhardness decreases: 78.5, 12.1, 5.4, 0.061, 0.031, 0.171 GPa, respectively (we note that Bi is again a semimetal due to its »nonmetallic« crystal structure).<sup>21</sup> The question is now whether the same explanation for the concentration dependence of the microhardness of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  is valid in the case of  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$ . Unfortunately, there is no similar investigation of particular electronic structure of  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  as for  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ . However, the gross dependence of the lattice constant,  $a$ , *i.e.*, the decrease of  $a$  with  $y$  indicates evolution of the alloy system towards more metallic character (Figure 2). Further, the data of the resistivity at room temperature could indicate indirectly the gross characteristics of the electronic structure. Likewise, in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ , the room temperature resistivity decreases with  $y$  (Figure 3). Such dependence of the resistivity was one of the signs, which led us to conclusion that the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  alloy system evolves from a system with semimetallic characteristics towards more metallic ones.<sup>15</sup> Although the resistivity of  $\text{YbInCu}_4$ , 150  $\mu\Omega$  cm, is considerably lower than the one of  $\text{YInCu}_4$ , 354  $\mu\Omega$  cm, it is still large compared to metals with wide conduction band. Thus, it follows that in  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$ , like in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ , metallic characteristics of the alloy system increase with increasing concentration parameters  $x$ , or  $y$ . Therefore we conclude that, as in the case of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ , the microhardness of  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  decreases with increasing  $y$ , since the system evolves towards more metallic character with increasing  $y$ .

A close inspection of microhardness data indicates a faint maximum for lower  $x$ , or  $y$ , or, at least, at these concentrations, there is something larger difference among the values of microhardness for different samples. The lattice parameter and the resistivity show some peculiarities, as well. We ascribe all these behaviors to an enhanced mixing of the  $\text{Yb}^{3+}$  and  $\text{Yb}^{2+}$  at these concentrations.

Finally, we should discuss why we are able to connect the electronic properties with the Vickers micro-

hardness of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  systems. It is known that, especially in metallic systems, the hardness is determined by the dislocation movement through microstructural obstacles and, thus, these processes are responsible for mechanical properties in plastic regime of metallic systems. There is no reason in the synthesis that would produce more dislocations for a particular alloy concentration in our investigated systems, and we assume that the number of dislocations is essentially uniform throughout the series. This number of dislocations presumably depends more on the extrinsic details of a particular sample and not on the concentration. We already mentioned we do not think that the concentration of an atom (explicated in the parameters  $x$  or  $y$ ) is important as the number of pinning obstacles regardless the pinning energy strength. If this was important we would get a clear maximum around  $x = 0.5$  or  $y = 0.5$  according to the Mott-Nabarro theory. Rather, the variation of concentration influences the change of the electronic structure. In our picture the dislocation movement depends on the strength of the connection between the ions in the alloys which is determined by the band structure. Further, the solubility exists in the whole concentration range and we do not expect that there exist obstacles like, *e.g.* precipitates in investigated systems.

Nevertheless, it seems surprising that the hardness decreases with decreasing lattice parameter because the band overlap increases in this case. To explain this point one must take into account the process of Vickers measurements of hardness. Vickers prism penetrates in a material and pushes crystallographic planes apart, and these planes then glide one over another. Therefore, Vickers microhardness scales with shear modulus and not with bulk modulus. It is known that s-electronic state is unstable against shear strain, in contrast to electronic states that are oriented in space, d- and f-state. Both facts were used in the explanation of the microhardness behavior of the  $\text{TiC}_x\text{N}_{1-x}$  system.<sup>25</sup> According to the band calculations for  $\text{LuInCu}_4$  and  $\text{YbInCu}_4$ <sup>24</sup> and the investigations of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ ,<sup>15</sup> it comes out that overlapping of bands and filling the s-band by electrons may explain the decrease of lattice parameter, which is equivalently to increase of  $x$ . As a consequence, the shear modulus is dependent: on the increase of number of s electrons and also, with the decrease of number of d- and, perhaps, f-electrons, that are also present at the Fermi level, according to calculations.<sup>24</sup> It seems that the same explanation for the concentration dependence of the microhardness one can apply in the case of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  system.

## CONCLUSIONS

Direct connections between microhardness, *i.e.*, mechanical properties in the plastic regime, and electronic properties, especially in metallic systems, are very diffi-

cult to identify and are rarely studied. Nevertheless, the concentration dependence of microhardness in the series of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  single crystals appears to reflect just such a connection, in particular, a decreasing microhardness as the system evolves from semi-metallic towards more metallic character.

The following conclusions can be drawn so far:

(i) Single crystal samples of the series of alloys:  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$ , for  $x$ , or  $y$  from the interval ( $0 \leq x$ ;  $y \leq 1$ ), were successfully prepared by the flux methods, and microhardness measurements were performed upon them.

(ii) The results presented in Figure 1 evidence discernable difference of the microhardness of the examined alloy series that is outside the experimental errors.

(iii) We ascribed the observed concentration dependence of the Vickers microhardness of the investigated systems to the change in the electron structure of these systems. With decreasing lattice parameter (Figure 2) the systems evolve from semimetallic towards more metallic character (Figure 3), and therefore, the microhardness decreases with increasing  $x$  or  $y$ .

(iv) It is demonstrated again that valuable empirical information can be obtained by means of microhardness measurements, but for the sake of its proper interpretation one must turn to other techniques by which the changes in structure and microstructure can be revealed and correlate with the microhardness behavior.

*Acknowledgement.* – The financial support of the Ministry of Science, Education and Sport of the Republic of Croatia is acknowledged. The authors express their sincere thanks to Dr B. Gumhalter for reading the manuscript.

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**SAŽETAK****Utjecaj elektronske strukture na plastična svojstva jediničnih kristala  
 $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  i  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  sustava****Mirko Stubičar, Miroslav Očko, John-Louis Sarrao, Nada Stubičar i Željko Šimek**

Izloženi su i objašnjeni rezultati mjerenja mikrotvrdoće Vickersovom metodom na dvije serije jediničnih kristala  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  i  $\text{YbAg}_y\text{In}_{1-y}\text{Cu}_4$  sustava s vrijednostima koncentracijskih parametara  $x$  i  $y$  iz intervala  $0 \leq x, y \leq 1$ . Uzorci su bili pripremljeni »flux« metodom. Za obje serije uzoraka mikrotvrdoća se smanjuje porastom vrijednosti  $x$  ili  $y$ . Sličnu ovisnost pokazuju i izmjereni parametri rešetke, kao i vrijednosti električne otpornosti. Opažene prethodne ovisnosti objašnjavamo promjenom naravi veze (iz polumetalnoga u karakter bliži metalnome), a uzrokovane su promjenom elektronske strukture sustava.