

# Microhardness of the $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$ alloy system

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We report Vickers microhardness measurements on flux grown single crystals of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy system. Although sample dependent, the microhardness exhibits a clear concentration dependence: in general, it decreases with  $x$ . The lattice parameter as a function of  $x$  exhibits a similar behavior. For  $x < 0.5$ , where the lattice parameter is almost constant, the microhardness exhibits a weak enhancement. Similar concentration dependence of the lattice parameter, resistivity and microhardness allows us to conclude that the microhardness reflects the evolution of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy system towards more metallic character with increasing  $x$ . © 2005 Springer Science + Business Media, Inc.

## 1. Introduction

Recently Vickers microhardness measurements on the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  alloy system were reported. The spatial extent of f orbitals is reduced relative to d orbitals, and this fact enhances solubility, i.e., mixing with a non-magnetic ion in a wide concentration range. Therefore, it was expected that one could observe the influence of electronic structure on concentration dependence of hardness in such systems, what is rarely observed in metallic systems, although it is clear that electronic structure determines hardness of a material. Indeed, it was shown that the decrease of microhardness with increasing  $x$  in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  (see the inset to Fig. 1) is determined by the evolution of the electronic structure from semimetallic towards more metallic character [1].

In the light of this result, we seek to find some another alloy system which could approve the conclusions we have performed in Ref. [1] There exists correlation between hardness and electronic structure in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ , although, it is commonly accepted that moving and pinning of dislocations are responsible for hardness in metallic systems. There were some indications that  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  could be such a metallic alloy system. The main parameter which governs the evolution from a semimetallic system towards a more metallic one in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  is the lattice parameter. Both alloy systems crystallize in

the same crystallographic structure: the cubic C15b ( $\text{MgCu}_4\text{Sn}$  type) space group. The lattice parameter of  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  in general decreases with  $x$ , as it does in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ . This fact suggested us that  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  could be also a good candidate system where one could observe an influence of electronic structure on plastic mechanical properties, i.e., on microhardness.

The magnetic and electronic properties of  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  were thoroughly investigated and it was found that this alloy system is a valence fluctuating one up to  $x = 0.2$  in which the valence transition temperature increases with  $x$  [2–6 and references therein].

## 2. Experimental techniques

The single crystal samples of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy systems were prepared by a flux method [2]. For the microhardness measurements the samples were mounted into 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 a  $136^\circ$  diamond pyramid indenter. The crystallographic planes  $\{100\}$  were available for microhardness investigation in

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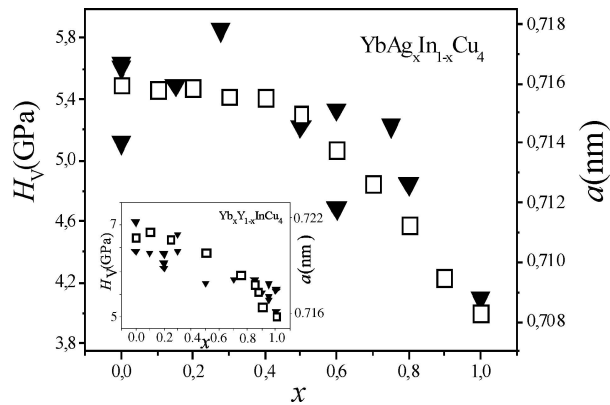


Figure 1 The microhardness (triangles) and lattice parameter (squares) of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy system. Inset: The microhardness (triangles) and lattice parameter (squares) of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  alloy system.

the as-grown-crystals. However, because of the process of polishing of surface, we were not able to control precisely the plane of indentation and a considerably tilting might be present. This fact might be one of the reasons of the relatively large dispersion of the results for a given concentration. Nevertheless, it seems clear that this dispersion does not hide the general concentration dependence of the microhardness in  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  systems. Moreover, it was shown recently in numerous similar novel alloy systems containing U and/or Ce that the expected general concentration dependence of microhardness was obtained in spite the fact that the samples were polycrystals [7].

Initial microhardness measurements revealed that microhardness data depended on the load applied on the indenter, but for loads exceeding 0.981 N, the microhardness was nearly independent of load. Therefore, 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 microhardness tests was constant and equal to  $500\times$ . 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. The indents, which differed much from a quadratic form, were not taken into account.

Our future investigations will be devoted to determine the microhardness as a function of crystal orientation.

Resistivity measurements were performed by standard four-point-technique. The dc-current used was 10 mA. The distance between voltage loads were hold fixed for all samples, 2 mm. However, the other dimensions depended of particular sample. Estimated error in determination of geometrical factor was within 10%.

### 3. Results and discussion

In Fig. 1, we display results of microhardness measurements of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy system (triangles) together with lattice parameter data (squares). Each point on the graph represents measurements on a particular sample. The samples for a given concentration were from the same batch. In order to obtain the average value of microhardness, twenty indentations were made. Standard deviation was about 10% of the corresponding mean value.

According to the Mott-Nabarro theory, hardness of a binary alloy system with atoms of different sizes should have a maximum at about  $x = 0.50$  [5]. Our results show clearly that this is not obeyed in  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$ . According to a Rydberg study, one can correlate hardness with the reciprocal of the size of the atoms for some pure elements [6]. This conclusion and the underlying experimental results follow from the view that hardness is proportional to cohesive forces. In our case hardness is roughly proportional to lattice parameter (and not to its reciprocal) and, therefore, our results cannot be explained by this ansatz. Intuitively, hardness is proportional to density of a material. Again, our results are not in accordance with this expectation. Our system is less dense for lower  $x$  not only because the lattice parameter increases with decreasing  $x$ , but also because In has a lower mass than Ag. We note that very similar behaviours of the measured physical quantities are seen for the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  alloy system (see Fig. 1 and the inset). Similar issues were discussed in reference 1.

Unfortunately, the electronic structure of  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  is not discussed in the same level of detail in references 2–5 as for the case of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ . However, the gross dependence of the lattice constant,  $a$ , (i.e., the decrease of  $a$  with  $x$ ) could indicate an evolution of the alloy system towards more metallic character with increasing  $x$ . This conclusion is consistent with the theoretical calculations of the electronic structure of  $\text{LuInCu}_4$  and  $\text{YbInCu}_4$  [8] and the related discussion in reference 9. In fact, the resistivity data at room temperature of  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  are rather similar to  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  in which the room temperature resistivity decreases with  $x$  (Fig. 2). Such a dependence of the resistivity with  $x$  was one of the signs that led to the conclusion that  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  evolves from a system with semimetallic characteristics towards more metallic behaviour [9]. Although the room-temperature resistivity of  $\text{YbInCu}_4$ ,  $150 \mu\Omega\text{cm}$ , is considerably lower than that of  $\text{YInCu}_4$ ,  $354 \mu\Omega\text{cm}$ , it is still large compared to metals with wide conduction band. Thus, it follows that in  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$ , as in  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ , metallic characteristics of the

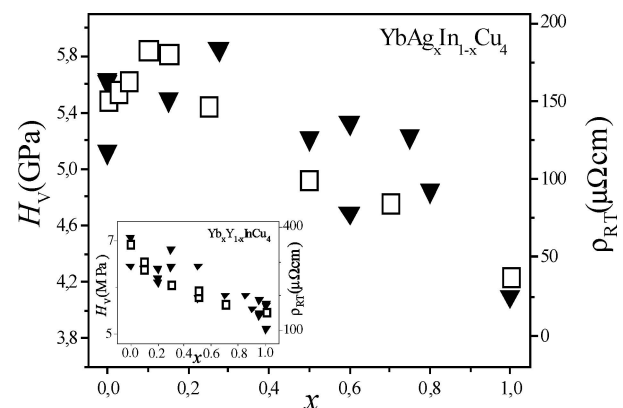


Figure 2 The microhardness (triangles) and room temperature resistivity (squares) of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy system. Inset: The microhardness (triangles) and room temperature resistivity (squares) of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  alloy system. By these graphs we do not want to suggest that there is a direct connection between microhardness and resistivity, but their concentration dependence may be due to the same origin.

alloy system increase with increasing  $x$ . Therefore we conclude, as in the case of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$ , that the microhardness of  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  decreases with  $x$  because the system evolves towards more metallic character with  $x$ . Namely, it is known that insulators and semimetals are generally harder than metals.

Nevertheless, it seems strange that hardness decreases with decreasing lattice parameter because band overlap increases in this case. To explain this point one has to have in mind, first, the process of the Vickers measurements of hardness. A Vickers prism penetrates in a material pushing crystallographic planes apart and these planes then glide one over another. Therefore, Vickers microhardness scales with shear modulus and not with bulk modulus. Second, it can be shown that an s-orbital electronic state is unstable against shear strain in contrast to electronic states that are oriented in space. Both of these facts were used in the explanation of the microhardness of  $\text{TiC}_x\text{N}_{1-x}$  [10]. According to the band calculations for  $\text{LuInCu}_4$  and  $\text{YbInCu}_4$  [8] and the investigations of  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  [9], it is clear that overlapping and filling of s bands increase with decreasing lattice parameter, i.e., with increasing  $x$ . These processes affect the shear modulus in two ways: first by increasing the number of s electrons and, as a consequence, decreasing the number of d- and, perhaps, f-electrons that are also present at the Fermi level according to calculations. It appears that the same explanation for the concentration dependence of the microhardness can be applied to the case of the  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy system.

#### 4. Conclusion

Direct connections between microhardness, i.e., mechanical properties in the plastic regime, and electronic

properties are very difficult to identify especially in metallic systems and are rarely studied. Nevertheless, the concentration dependence of microhardness in single crystals of the  $\text{Yb}_x\text{Y}_{1-x}\text{InCu}_4$  and  $\text{YbAg}_x\text{In}_{1-x}\text{Cu}_4$  alloy systems appears to reflect just such a connection. The microhardness decreases as the system evolves from semimetallic towards more metallic character.

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