TEMPERATURE DEPENDENT CORE-LEVEL PHOTOEMISSION STUDY OF UNiSn*

Ji-Yong So, S.-J. Oh

Department of Physics, Seoul National University, Seoul 151-742, Korea

E.-J. Cho

Department of Physics, Chonnam National University, Kwang-Ju 500-757, Korea

J.-G. Park

Department of Physics, SungKyunKwan University, Suwon 440-746, Korea

and K.A. McEwen

Department of Physics and Astronomy, University College London
London WC1E 6BT, UK

(Received July 10, 2002)

UNiSn undergoes an anomalous phase transition at $T_N = 47\,K$, at which temperature it transforms from an antiferromagnetic metal to a paramagnetic semiconductor with an energy gap $\simeq 70\,meV$. In order to investigate how the electronic structure of UNiSn changes as it crosses the transition temperature, we have used the X ray photoemission spectroscopy (XPS) technique from 20 to 70 K. According to the XPS studies, the U $4f$ core levels are almost temperature independent while the Ni $2p$ core levels and the satellite structure display a weak anomaly at T_N.

PACS numbers: 79.60.–i, 71.20.Eh, 71.27.+a

1. Introduction

UNiSn shows an anomalous phase transition at $T_N = 47\,K$, below which it is an antiferromagnetic metal while it is a paramagnetic semiconductor at higher temperatures [1]. Resistivity measurement shows an activation-type T-dependence with an estimated energy gap of $\Delta \sim 70\,meV$ [1]. It is generally accepted that the inverse metal-insulator transition is closely

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

related to the nature of U $5f$ electrons so it is very important to investigate experimentally the electronic structure of UNiSn [2]. However, there are very few studies so far of electronic structures including U $5f$ except for a very recent work [3], where studies have been made of the temperature dependence of the valence band electronic structure. In this study, we have concentrated on the temperature dependence of the core levels of U and Ni.

2. Experiments

We have measured X ray photoemission spectroscopy (XPS) of Ni $2p$ and U $4f$ to investigate the electronic structure from 20 to 70 K. We used a bulk sample prepared as described elsewhere [3]. All our experiments were made after scrapping at 20 K in ultra high vacuum with a base pressure better than 1.0×10^{-10} torr. We used Mg Kα photon source ($\hbar\omega = 1253.6$ eV). The total experimental resolution is about 0.8 eV at the measured energy range.

3. Data and results

Fig. 1 shows XPS spectra of UNiSn taken at 20 and 70 K. As can be seen in Fig. 1(a), Ni $2p_{3/2}$ spectrum is composed of one main peak and one satellite peak. The 6 eV satellite of Ni $2p$ indicates the importance of many body effects in the Ni electronic structures as often observed in other Ni compounds and pure Ni metal. In order to subtract the background off

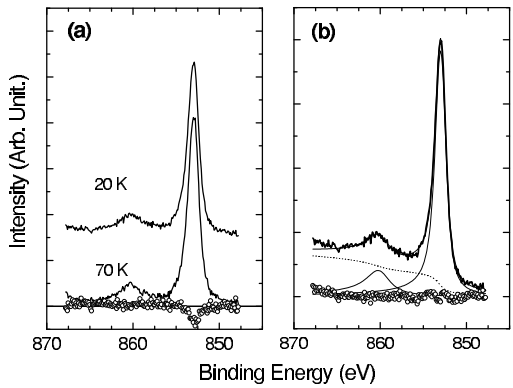

Fig. 1. (a) Ni $2p_{3/2}$ main and satellite peaks taken at 20 and 70 K after background subtraction using Tougaard's method. The 20 K data are shifted upwards for better presentation. Two spectra are normalised by the total area of the main and satellite peaks. Symbols at the bottom of the figure are for the difference curve between the two data. (b) Curve fitting results of the XPS spectrum of Ni $2p_{3/2}$ at 20 K: experimental results (thick line), fitting results (thin line), background (dashed line) following Tougaard's method (see the text) and the difference (symbols) between the experimental and fitting results.

TABLE I

Fitting results of XPS spectrum, where the first two columns labelled by r is for the ratio of satellite and main peak between 20 and 70 K and the next two columns labelled by Γ for the Lorentzian width of main and satellite (in parenthesis) peaks in eV units.

	r (20 K)	r (70 K)	Γ(20 K)	Γ (70 K)
Shirley	0.35	0.29	0.976 (2.62)	0.96 (1.55)
Tougaard	0.26	0.20	0.99 (2.81)	0.98 (2.17)

the data, we used two usual methods: Shirley procedure [5] and Tougaard procedure [6]. To fit the Ni $2p_{3/2}$ peak, we used a conventional line shape [4] and the summary of the fitting results is given in Table I. What is noticeable is that in both cases the main peak intensity becomes larger relative to the satellite in the semiconducting phase than in the metallic phase. At the same time, the width of the satellite peak decreases as it enters into the semiconducting phase. This temperature dependence of Ni $2p_{3/2}$ peak, although weak itself, makes a marked difference with that of U $4f$ data shown in Fig. 2. As one can see in the figure of U $4f$ peak, there is hardly any difference between the two data taken at 20 and 70 K unlike the Ni $2p_{3/2}$ peak.

As regards the satellite peak of Ni $2p_{3/2}$ peak, we note that previous UPS experiment by Kang *et al.* [3] showed a small 6 eV satellite of Ni $3d$. Satellite structures of the $2p$ core levels or the $3d$ valence band are usually considered to be due to the many body effects of Ni $3d$ electrons assisted by surrounding conduction and ligand electrons. Following the usual assignment, the main and satellite structures of Ni $2p$ peaks can be ascribed to $3d^{10}$ and $3d^9\underline{L}$, where $\underline{L}$ means a ligand hole. That we have observed stronger temperature

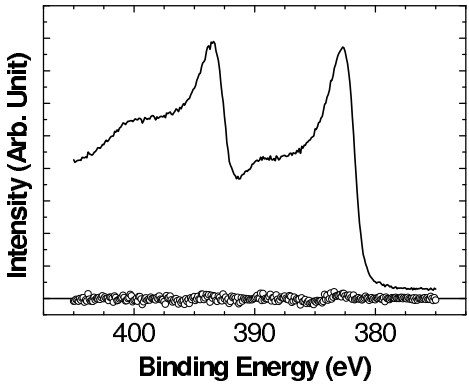

Fig. 2. It shows XPS spectrum of U $4f$ measured at 20 K (thick line) and the difference curve (symbols) between the data taken at 20 K and 70 K.

dependence in the Ni $2p$ peaks than in the U $4f$ structures indicates that the Ni electronic structure might undergo a more significant change as it becomes an antiferromagnetic metal below T_N. It then implies that Ni plays a more important role in the phase transition than originally assumed. Closely related to this point, we note that both our studies of the valence band (not shown here) as well as Kang's [3], which seems to reflect mainly U $5f$ bands, failed to show any considerable difference above and below the phase transition.

However, what our results show is that as far as the electronic structure is concerned the main change occurs in the Ni $3d$ band at the transition temperature and U $5f$ band plays a relatively minor role. On the other hand, inelastic neutron scattering measurements [7] show that the magnetic scattering of U $5f$ electrons indeed exhibits a considerable change at the transition temperature. Therefore, our data together with earlier UPS studies as well as inelastic neutron scattering results suggest an very interesting scenario for the phase transition of UNiSn that it is probably driven by the electronic degree of freedom of Ni $3d$ bands which is coupled to the spin degree of freedom of U $5f$ electrons. This then means that the most important hybridisation is between Ni $3d$ bands and U $5f$ electrons. We note that our conclusion drawn here contradicts the predictions of recent band calculations using the LDA+U method [8]. In order to further test our conclusion, we need to study the temperature dependence of the Ni $3d$ bands using Ni $3p \rightarrow 3d$ resonant photoemission spectroscopy with higher resolution.

4. Conclusion

We have measured XPS data of UNiSn below and above T_N. Unlike the predictions of the LDA+U band calculations, we found that a major change at T_N occurs to the Ni electronic structure, not the U $5f$ bands. Our data together with other UPS and inelastic neutron scattering data indicates that the main driving force of the phase transition is more likely to be the hybridisation between Ni $3d$ band and U $5f$ electrons.

REFERENCES

[1] H. Fujii *et al.*, *J. Phys. Soc. Jpn.* **58**, 2495 (1989).
[2] Y. Aoki *et al.*, *Phys. Rev.* **B47**, 15060 (1987).
[3] J.-S. Kang *et al.*, *Phys. Rev.* **B64**, 85101 (2001).
[4] G.D. Mahan, *Phys. Rev.* **B11**, 4814 (1975).
[5] D.A. Shirley, *Phys. Rev.* **B5**, 4709 (1972).
[6] S. Tougaard, *Surface and Interface Analysis II*, p. 453, 1988.
[7] K.A. McEwen *et al.*, *Physica B* **281-282**, 600 (2000).
[8] P.M. Oppeneer *et al.*, *Phys. Rev.* **B54**, R3706 (1996).