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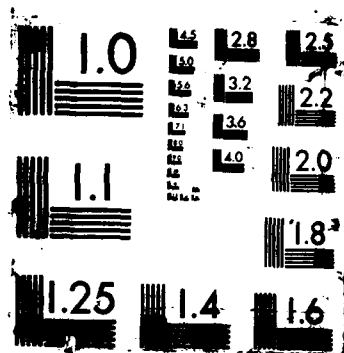
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NONMONOTONIC EVOLUTION AND THERMODYNAMIC  
TRENDS AT METAL/(Hg,Cd)Te INTERFACES: Yb/Hg<sub>0.78</sub>Cd<sub>0.22</sub>Te

D. J. Peterman

McDonnell Douglas Research Laboratories  
P. O. Box 516, St. Louis, Missouri 63166

A. Raisanen, S. Chang, P. Philip, A. Wall, and A. Franciosi  
Department of Chemical Engineering and Materials Science  
University of Minnesota, Minneapolis, Minnesota 55455

Abstract

The microscopic interaction of cleaved Hg<sub>0.78</sub>Cd<sub>0.22</sub>Te surfaces with the highly reactive rare-earth, Yb, was studied by use of photoemission spectroscopy and synchrotron radiation. We compare the results of this study with our previous investigation of Sm/(Hg,Cd)Te interface formation. Yb and Sm are similar in their initial stage of interface formation--rapid attenuation and near loss of the Hg emission is followed by its dramatic recovery. However, differences in valency between Sm and Yb develop during metal deposition and subsequently lead to differences in interface morphology. Calculated heats of solution are shown to correlate with the different behaviors observed during rare-earth/(Hg,Cd)Te interface formation.

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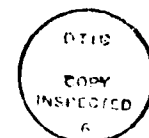
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| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number)<br><br>The microscopic interaction of cleaved Hg <sub>0.78</sub> Cd <sub>0.22</sub> Te surfaces with the highly reactive rare-earth Yb was studied by use of photoemission spectroscopy and synchrotron radiation. We compare the results with those of our previous investigation of Sm/(Hg,Cd)Te interface formation. Both overlayers exhibit a dramatic recovery of the Hg concentration at the interface after the first stage of metal-Te reaction. However, differences in valency between Sm and Yb develop during metal deposition and lead to differences in interface morphology. <i>(Kryger et al.)</i> |                       |                                                                                       |

## I. INTRODUCTION

Exploiting the electronic consequences of the Hg/Cd-ratio-controlled band gap of (Hg,Cd)Te alloys requires the use of electrical contacts. When metals are deposited on the alloy, however, considerable surface chemical disruption often occurs as the weak Hg-Te bonds give way to the formation of metal-Te bonds. To understand and perhaps learn to control the extent of this surface disruption, we have employed synchrotron radiation and photoemission to systematically study (Hg,Cd)Te interfaces with metals of varying chemical reactivity. In this paper we present experimental results on interface formation with the highly reactive metal Yb. We contrast these results with our previous study of Sm/(Hg,Cd)Te interfaces<sup>1</sup> and discuss the physical origin of the dramatically different interface-formation behaviors observed.

Bulk heats of metal-telluride formation correlate with certain aspects of metal/(Hg,Cd)Te interface formation. For example, in studies of Ag,<sup>1</sup> Cr,<sup>2,3</sup> and Sm,<sup>1</sup> we observed that the rate of surface-Cd depletion with metal deposition increases with the reactivity of the metal. Davis et al.<sup>4</sup> have reported similar observations in comparing Hg depletion with Ti and Al deposition. While bulk thermodynamics is not expected to rigorously apply to interface formation, apparent anomalies in correlations with heats of formation, evidenced by the persistent concentration of Cd in Pt overlayers<sup>5</sup> or the persistent Hg concentration in Sm overlayers,<sup>1</sup> are explained by considering heats of cation-metal



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alloying (heats of solution) calculated with the formalism of Miedema et al.<sup>6</sup> Yb and Sm have similar heats of metal telluride formation but their interface-formation behaviors are radically different. We show that these differences correlate with trends in the calculated heats of solution when the difference in rare-earth valence is considered.

## II. EXPERIMENTAL PROCEDURE

Experiments were conducted on oriented single crystals of commercially-obtained<sup>7</sup>  $\text{Hg}_{0.78}\text{Cd}_{0.22}\text{Te}$  which were cut in the form of rectangular posts with a (110) surface area of  $3 \times 3 \text{ mm}^2$ . The crystals were cleaved in the photoelectron spectrometer (pressure  $< 2 \times 10^{-8} \text{ Pa}$ ) and metal interfaces with (110) surfaces were prepared in situ by the sublimation and deposition of Yb. Metal depositions were measured with a quartz thickness monitor. Synchrotron radiation from the 1-GeV electron-storage ring at the Synchrotron Radiation Center of the University of Wisconsin-Madison was monochromatized with a grasshopper monochromator. Photons in the energy range of  $40 < h\nu < 150 \text{ eV}$  were used to obtain photoelectron energy distribution curves (EDCs) as a function of metal coverage. The photoelectron energies are referred to the Fermi level ( $E_F$ ), which was located by observing the Fermi level cut-off of photoemission from thick, freshly-evaporated Cr films. Interface evolution with metal coverage was studied by analyzing EDCs of the valence band, and the Hg 5d, Cd 4d, Te 4d,

Yb 4f, and Yb 5p core levels. The results presented below were obtained from four cleaves with deposited metal layer thicknesses,  $\theta$ , up to 2.0 nm. For the (Hg,Cd)Te (110) surface density of  $6.76 \times 10^{14}$  atoms/cm<sup>2</sup>, one monolayer of Yb corresponds to a Yb layer thickness of 0.28 nm.

### III. RESULTS

In Figs. 1 and 2 we highlight select EDCs to demonstrate that there are at least three distinct stages of interface formation which occur with Yb deposition. In the first stage, with  $\theta < 0.1$  nm, Fig. 1 indicates that the Hg and Cd d cores shift to higher binding energy, the Hg and Cd emissions rapidly attenuate, and the Hg/Cd ratio decreases. Representative EDCs showing Te 4d core emission as a function of  $\theta$  are shown in Fig. 2. Backgrounds have been subtracted from these curves by use of a fourth-degree-polynomial fitting procedure. In the first stage of interface formation, the Te 4d cores shift to lower binding energy and the Te surface concentration increases. The changes in the Hg, Cd, and Te concentrations in this stage are similar to those which we have reported for Sm in the 0- to 0.6-nm deposition range.<sup>1</sup> Furthermore, the changes in Hg, Cd, and Te intensities are qualitatively similar to those observed with deposition of other reactive metals such as Al,<sup>8</sup> Cr,<sup>2,3</sup> and Ti.<sup>4</sup>

The changes in Hg 5d and Te 4d photoemission intensity for  $0 \leq \theta \leq 2.0$  nm are summarized in Fig. 3. The Te 4d intensities

were obtained by computing the area under the background-subtracted curves. This procedure could not be used with the Hg 5d cores because of the overlapping Cd 4d emission features. Rather, the Hg 5d intensity was obtained by measuring the height of the well-resolved Hg 5d<sub>5/2</sub> peak above a linear background. While this method is somewhat inaccurate because of the simple background estimate and because (as Fig. 4 shows) the width of this peak also varies with  $\theta$ , inaccuracies will not be important to our present discussion. Quantitatively, we find that in the first stage of interface formation the rate of Hg depletion is greater with Yb deposition than with Cr,<sup>2</sup> or Al<sup>8</sup> deposition and is closest to that with Ti deposition,<sup>4</sup> a trend that correlates with bulk heats of telluride formation.<sup>9</sup>

In the second stage of interface formation, with  $0.1 < \theta < 0.3$  nm, Figs. 1 and 3 show that the Hg photoemission intensity increases with Yb deposition. This peculiar, nonmonotonic behavior was first observed with Sm interfaces in their second (and final) stage of interface formation.<sup>1</sup> The increase in surface Hg concentration with Yb deposition is accompanied by a rapid decrease in Te concentration as is shown in Fig. 3. Furthermore, the Hg and Te binding energies are radically shifted as is detailed in Fig. 4. Also shown in Fig. 4 is the evolution of the full width at half maximum of the Hg 5d<sub>5/2</sub> peak. The peak-width changes and the direction of the binding-energy shifts are consistent with the formation of dissociated Hg and Te. However, the shifted Hg 5d binding energy, 8.0 eV, is slightly greater than the tabulated value of 7.8 eV for metallic Hg,<sup>10</sup> and the

shifted Te binding energy is much higher at  $\theta = 0.3$  nm ( $\sim 40.8$  eV) than the tabulated value given<sup>10</sup> for elemental Te, 40.4 eV.

While the Hg core level shifts observed in this Yb deposition interval are similar to those observed with Sm depositions, the evolution of the Yb 4f cores indicates important differences between Yb and Sm. With Sm, the second (and final) stage of interface formation involved a transition to a mixed valent state, whereas Fig. 1 indicates that the Yb remains divalent at all coverages (trivalent Yb would be signaled by several spectral features between 5 and 10 eV, the strongest of which would be at 10.0 eV).<sup>11</sup>

Figure 5 provides a more detailed picture of the Yb 4f evolution. In the 0.1- to 0.3-nm deposition range the Yb 4f emission becomes metal-like, demonstrating the well-known bulk- and surface-shifted-cores with a final (at  $\theta = 0.5$  nm) bulk 4f<sub>7/2</sub> binding energy of 1.3 eV; this energy can be compared to the value reported for metallic Yb, 1.2 eV.<sup>12</sup> Furthermore, in Fig. 5, a Fermi level can be identified at a deposition of only 0.2 nm.

During the third stage of interface formation, with  $\theta > 0.3$  nm, the Hg and Te emissions are attenuated as metallic Yb covers the surface, as shown in Figs. 1-3. Again, this is in sharp contrast to the Sm interface where there was no third stage and Hg and Te photoemission persisted at high Sm depositions. Figures 2 and 4, show that in this stage of Yb deposition, while the Hg and Yb binding energies are constant, that of the Te 4d<sub>5/2</sub> core still shifts to a final value of about 41.2 eV.

#### IV. DISCUSSION

Trends observed in comparing the Yb and Sm results with each other and with those of other metals do correlate with some bulk thermodynamic properties. In the previous section we noted that, in the first stage of interface formation, rapid attenuation of the cation photoemission followed the trend observed with other metal interfaces; generally, the higher the heat of telluride formation the faster the cation emissions attenuate with initial metal deposition. The trends observed in the subsequent stages of interface formation with Sm and Yb can be understood in terms of another energy term--the heat of solution. Friedman, et al.<sup>5</sup> first noticed the relevance of this quantity to metal/(Hg,Cd)Te interface formation in their work with Pt. With Pt deposition, Cd dissociated and alloyed with the metal layer. At Pt depositions of 1.6 nm, a substantial Cd photoemission signal was still evident. With Sm, on the other hand, it was Hg that was still observed<sup>1</sup> after a deposition of 5 nm. An examination of the heats of solution given in Table I reveals some interesting correlations with this interface formation behavior. The values listed were computed using the parameters given in Ref. 13. (Parameters for  $\text{Sm}^{2+}$  were not available.) First, comparing Pt with Yb and Sm, we note that the energetics are such that the cation most likely to form a solution with the metal overlayer is Cd in the Pt case and Hg in the rare-earth cases. Second, the large difference in  $\Delta H_{\text{sol}}$  between  $\text{Yb}^{2+}$  and  $\text{Sm}^{3+}$  indicates that Hg is more likely to form a solution with the trivalent rare earth

than with the divalent rare earth. Thus, while the heat of telluride formation governs the first stage of interface formation with Sm and Yb, it is the heat of solution which governs the last stage.

Even though Hg does not go into solution with  $\text{Yb}^{2+}$  as it does with  $\text{Sm}^{3+}$ , the increase in the surface Hg concentration (in the second stage of interface formation with Yb) may still be related to the divalent heat of solution. We note that Yb and Sm interfaces have similar core level shifts which accompany the dramatic Hg recovery. We have used the values of  $\Delta H_{\text{sol}}$  given in Table I to calculate the  $5d_{5/2}$  binding energies of Hg in Yb and Sm following the procedure outlined by Steiner and Hufner.<sup>14</sup> In both cases the Hg core level should shift to a position within 0.1 eV of its metallic position and in both cases such huge shifts in that direction were observed as the Hg reappeared. For Yb, the calculated binding energy of 7.9 eV (0.07 eV higher in binding energy than for metallic Hg) is comparable to our measured value of 8.0 eV.

At this point it is difficult to determine whether or not the correlations that we have observed with heats of solution are more than fortuitous. There are many other factors that can be considered in understanding the chemistry of interface formation<sup>15</sup> and a great deal of experimental input will be needed to clearly identify systematics, especially for a semiconductor as complicated as (Hg,Cd)Te. In this respect we note that the evolution of the Yb 4f line shape, specifically the shift away

from the Fermi level with deposition (Fig. 5), is unlike that seen for the Yb interfaces with Si,<sup>16</sup> Ge,<sup>17</sup> or GaAs.<sup>18</sup>

## V. CONCLUSIONS

The Yb/(Hg,Cd)Te interface is highly reactive because of its large heat of telluride formation. The interface becomes metallic at a deposition of only 0.2 nm (~ one monolayer) and the evolution of the Yb 4f features is complete after a deposition of only 0.5 nm (~ two monolayers). This metallization leads to an exponential attenuation of the emissions from all semiconductor core levels, marking the first observation of a metallic layer on (Hg,Cd)Te which does not exhibit significant photoemission from one of the semiconductor's elements which is either segregated on or alloyed with the overlayer. The calculated heats of solution of Hg in  $\text{Sm}^{3+}$  and  $\text{Yb}^{2+}$  are significantly different and they likely explain the observed differences between the Yb- and Sm/(Hg,Cd)Te interface morphologies.

## ACKNOWLEDGMENTS

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## FIGURE CAPTIONS

Fig. 1 Representative photoelectron energy distribution curves (EDCs) as a function of Yb deposition,  $\theta$ . The Hg and Cd d cores initially shift to higher binding energy, their emission intensity rapidly decreases, and the surface Hg/Cd ratio decreases. For  $0.1 < \theta < 0.3$  nm the Hg 5d core levels abruptly shift to lower binding energy and their emission intensity begins to increase as a function of  $\theta$ . For  $\theta > 0.3$  nm the EDCs begin to show characteristic metallic Yb features as the Hg signal is attenuated. The curves have been offset for clarity.

Fig. 2 Representative EDCs as a function of  $\theta$ . The Te 4d cores initially shift to lower binding energy and their emission intensity increases. For  $\theta > 0.1$  nm the cores shift to higher binding energy and their emission intensity is rapidly attenuated. The EDCs have had a background subtracted and are offset for clarity.

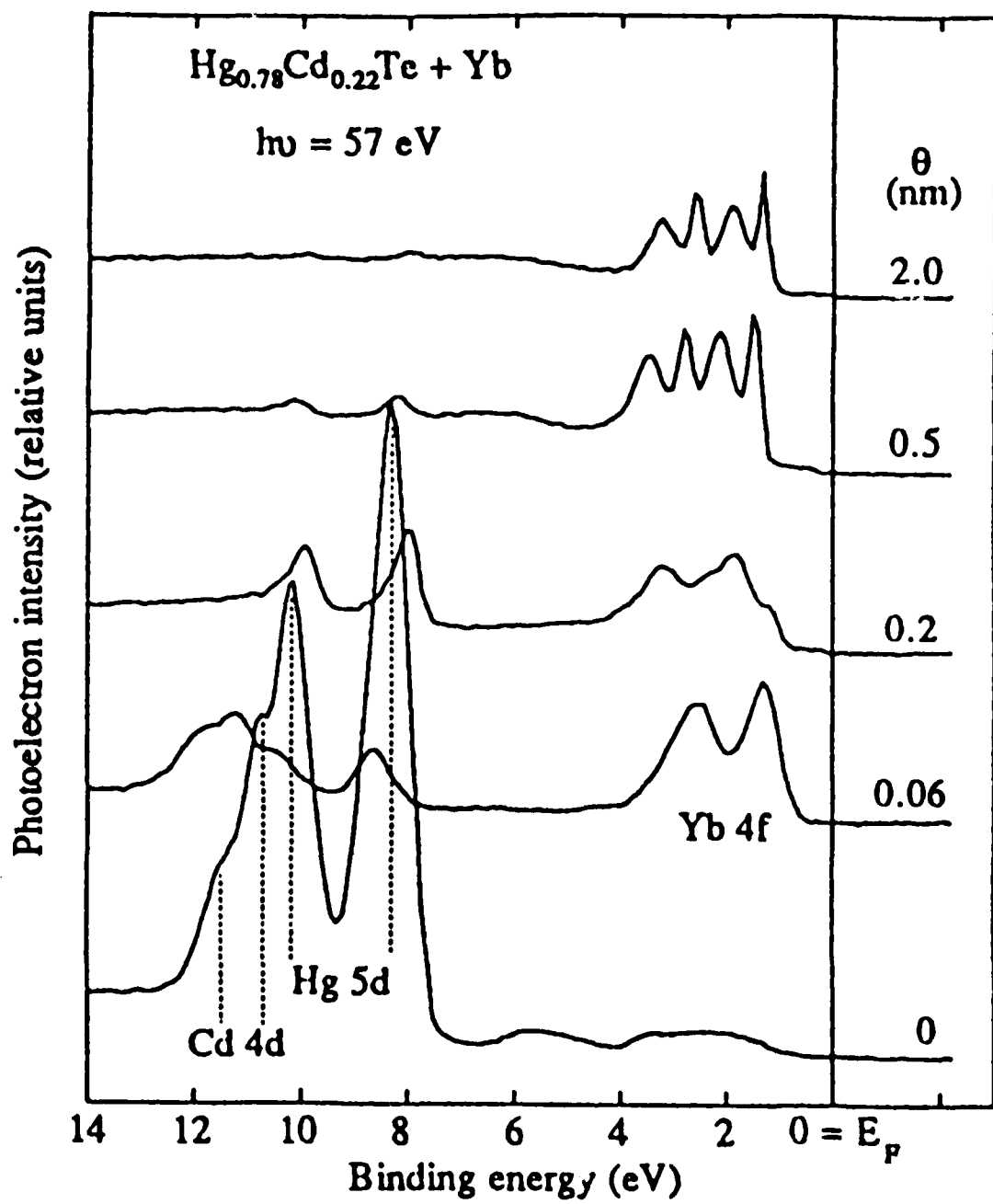
Fig. 3 Summary of the changes in emission intensities from the Hg 5d and Te 4d core levels as a function of  $\theta$ . (The trivial data points at  $\theta = 0$  have been omitted for clarity).

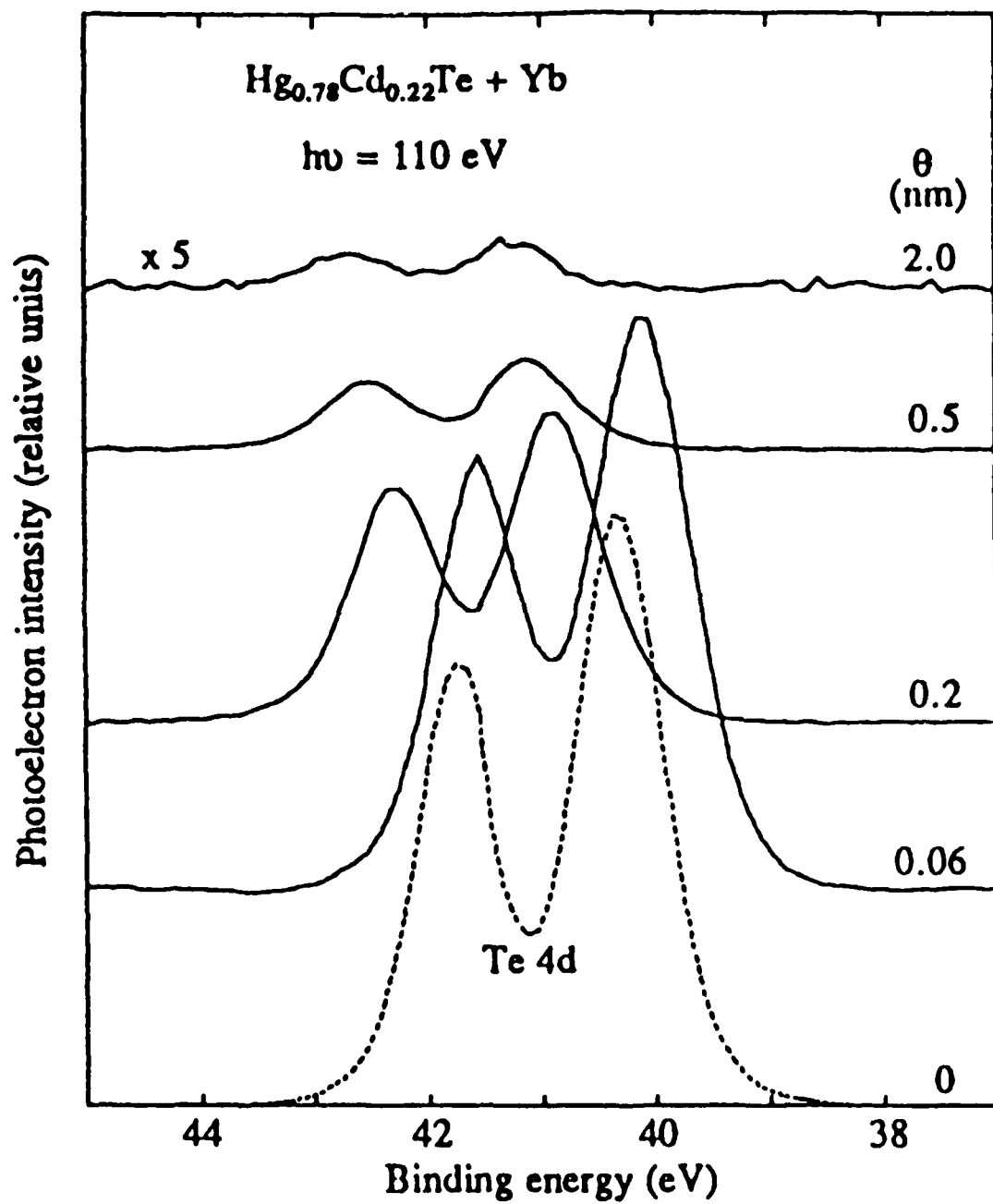
Fig. 4 Summary of binding energy shifts for (A) the Te 4d cores and (B) the Hg 5d cores as a function of  $\theta$ . Positive values of  $\Delta E_B$  denote a shift to higher binding energy. Lines have been added to guide the eye. Also in (B) is the evolution of the Hg 5d<sub>5/2</sub> peak width.

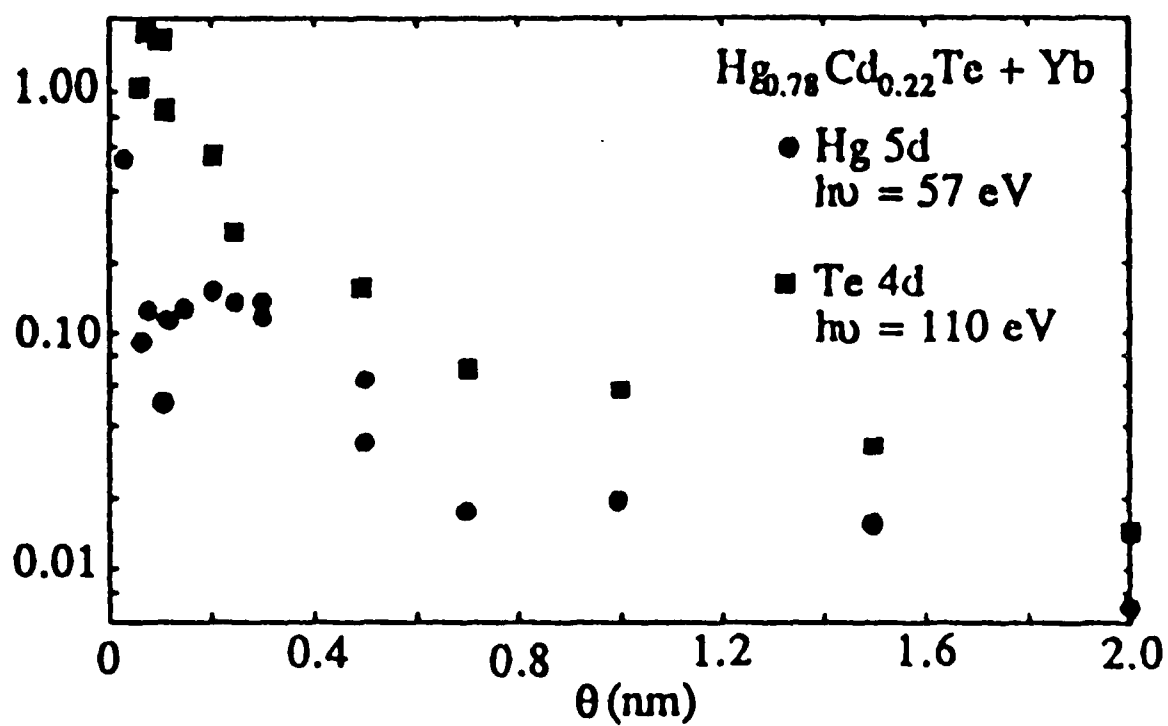
Fig. 5 Representative EDCs showing divalent Yb 4f emission as a function of  $\theta$ . For  $\theta > 0.5$  nm, the positions and line shapes of the indicated bulk- and surface-derived features are characteristic of Yb metal.

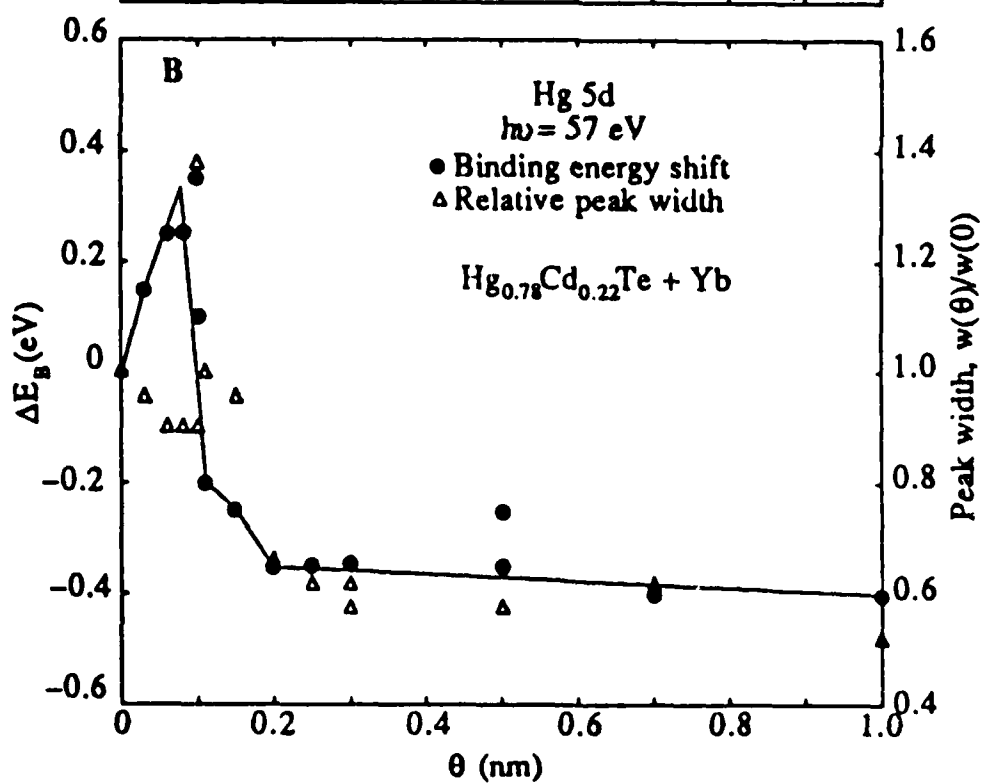
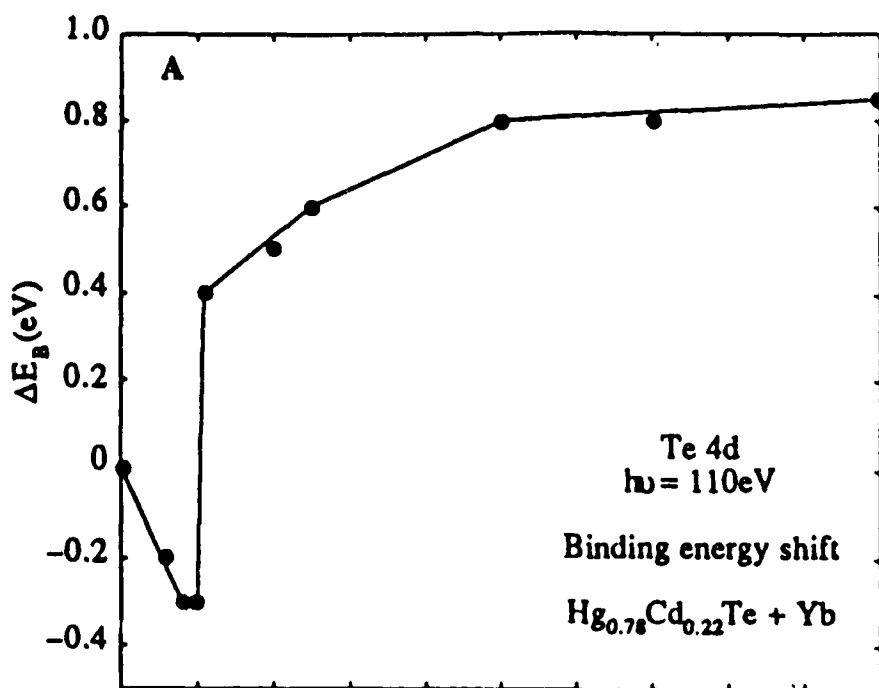
TABLE I. Heats of telluride formation,  $\Delta H_f$  (estimated in Ref. 9), of the stable tellurides listed in column 2 and heats of solution  $\Delta H_{sol}(C;M)$  (calculated following Ref. 13), of the cations (C) Hg and Cd in the metals (M) Pt, Yb and Sm. The rare-earth valences used in computing  $\Delta H_{sol}$  are given in column 1.

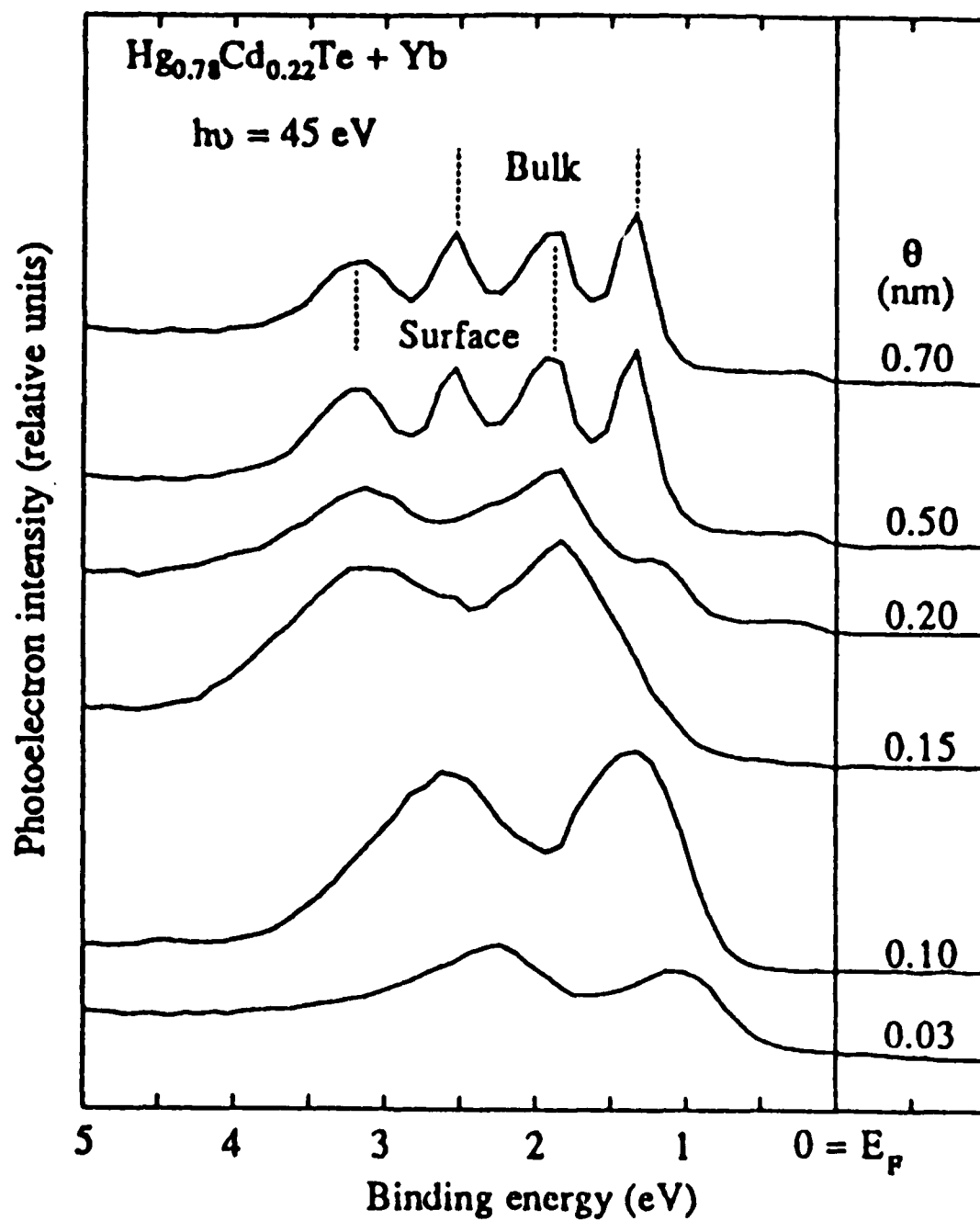
| Metal (M)        | Telluride                       | $\Delta H_f$<br>(kJ/mol Te) | $\Delta H_{sol}(Cd;M)$<br>(kJ/mol Cd) | $\Delta H_{sol}(Hg;M)$<br>(kJ/mol Hg) |
|------------------|---------------------------------|-----------------------------|---------------------------------------|---------------------------------------|
| Hg               | HgTe                            | -31.8                       | ---                                   | ---                                   |
| Cd               | CdTe                            | -101.                       | ---                                   | ---                                   |
| Pt               | PtTe                            | -41.9                       | -121.                                 | -79.6                                 |
| Yb <sup>2+</sup> | YbTe                            | -314.                       | -74.1                                 | -108.                                 |
| Yb <sup>3+</sup> |                                 |                             | -140.                                 | -172.                                 |
| Sm <sup>2+</sup> | SmTe                            | -310.                       |                                       |                                       |
| Sm <sup>3+</sup> | Sm <sub>2</sub> Te <sub>3</sub> | -265.                       | -141.                                 | -173.                                 |











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