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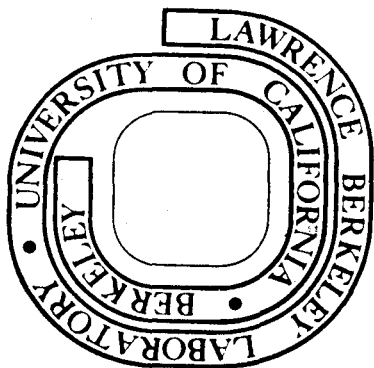
B. E. Mills and D. A. Shirley

January 1977

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X-RAY PHOTOEMISSION SPECTRA OF THE 2s VALENCE ORBITALS IN CYCLIC ALKANES
IN RELATION TO VALENCE BANDS OF AMORPHOUS GROUP IV AND V ELEMENTS*

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January 1977

ABSTRACT

The crystalline elements Ge, Si, As, Sb, and Bi display a characteristic two-peak structure in the x-ray photoelectron spectra of their s-like valence bands. In the corresponding amorphous materials the gap between these two bands is filled in. It has been suggested that this is a consequence of topological disorder; i.e., of the existence of odd-membered rings (especially 5-membered rings) in the amorphous cases, whereas the crystalline solids contain only 6-membered rings. To test this hypothesis we have studied tetrahedrally-bonded systems of different ring size in the gas-phase cyclic alkanes. The XPS spectrum of cyclohexane shows a striking resemblance to that of diamond. The other gas-phase spectra can be combined according to different ring-structure

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models. When this is done, we find that 5- and 6-membered rings alone are not sufficient to explain the "filling-in effect", but that 5-, 6-, and 7-membered rings are. The topological disorder model of bonding in amorphous semiconductors therefore appears valid. More generally, this work establishes further links between molecular orbital theory and bonding in covalent solids.

I. Introduction

The valence-band x-ray photoelectron spectra of amorphous Ge, Si, As, Sb, and Bi display certain differences from the spectra of their crystals.^{1,2} One of the more striking differences, illustrated for Ge, Si, and As in Fig. 1, is that peaks 1 and 2 in the cleaved crystal spectra are replaced by a broad peak in the amorphous case. These peaks all arise from bands built primarily from atomic s-like orbitals. The bands could differ in the crystalline and amorphous cases because of long-range disorder, short-range disorder, or a combination of both. We have studied model systems to help decide among these alternatives.

At standard conditions Ge and Si have the diamond structure. The atoms are tetrahedrally bonded to form connected chair-form six-membered rings. The group V elements have an A-7 crystal structure, also composed of six-membered rings. Of course, Ge and Si have been more intensely studied as amorphous materials. It is known that they can be prepared in the amorphous form with approximately the same density as in the crystalline form.³ Radial distribution function measurements of amorphous Ge, for example, give a first-neighbor (bond-length) distance which is nearly constant (± 0.04 Å), whereas there is a rather large variation in second-neighbor distance of ± 0.25 Å, indicating bond-angle variations. Furthermore, the third-neighbor peak is not well defined.⁴ This suggests short-range disorder rather than a microcrystalline structure or any amorphous cluster models in which angles do not change. The requirements of fixed bond length and angular variation can be met by a continuous random network model, which can be infinite at the expense of local distortion. This type of structure

does not require discontinuities as would micro-crystals or clusters, and can have a density as high or nearly as high as the crystalline phase, in agreement with experiments on defect-free samples.

An interesting structural feature of the continuous random network model is that it can be constructed to have non-six-membered rings. Polk⁵ noted that there were 20% five-membered rings in his model. It has been suggested that odd-membered rings could have a significant effect on the s-like valence band spectra of amorphous materials. This follows because these elements are covalently bonded, with localized orbitals. The s-levels, which are basically atomic, are sensitive to their local environment rather than to the crystal as a whole. Joannopoulos and Cohen⁶ have suggested a model for local disorder which is amenable to band-structure computations. This is the ST-12 high-pressure structure of Ge III⁷ and ice III,⁸ with 12 atoms per simple tetragonal unit cell. Eight of the atoms form two spirals with other atoms connecting the spirals. There is very little bond-length variation, but the bond angles vary from 91 to 120 degrees in Ge III (a much larger variation than in amorphous samples). In addition, the structure contains 5-, 6-, 7-, and 8-membered rings. Calculations on Ge III indicate a "filling-up" of the gap between the two s-like valence bands, as is observed in the amorphous valence-band spectra.

In order to definitively ascribe the change in valence-band spectra to ring size, one would like to be able to study covalent tetrahedral compounds of various ring sizes. Unfortunately, a crystal lattice of, for example, symmetrical interconnecting five-membered rings is impossible. However, a series of covalent tetrahedral

molecules of every ring size exists--the cyclic alkanes. We have used these hydrocarbons as model compounds because of their availability and because of the cross-section enhancement of the spectral region of interest in x-ray photoemission. Through the use of the cyclic alkanes we have confirmed the hypothesis that the s-like valence band spectrum reflects the local ring sizes in tetrahedrally-bonded solids. The spectra are described in Section II. They are compared with theory in Section III, and the connection with amorphous semiconductors is discussed in Section IV.

II. The Spectra

Photoelectron spectra were taken on the 50 cm. radius Berkeley magnetic spectrometer^{9,10} at a sample pressure of about 100 Torr. Cyclopentane, cyclohexane, cycloheptane, and cyclooctane spectra were taken with Mg K α x-rays (Fig. 2) at energy increments of 0.16 eV. All compounds were also run using Al K α x-rays at energy increments of 0.18 eV: The spectra were identical within experimental error.

At first glance it is not obvious how the spectra in Fig. 1 are related to those in Fig. 2. The energy scale, for example, is quite different. Part of the reason for this is that spectra of solids are referenced to the Fermi level. The spectra of gases are plotted relative to the vacuum energy level, and first ionization potentials in hydrocarbons are about 10 eV. In addition, the total width of the valence region in the carbon compounds is considerably greater than for the other species. This is generally true for first-row elements.

X-ray photoelectron spectra of first row elements also show intensity modulation caused by the fact that the 2s wavefunction has a radial node while the 2p does not. As a result, the greater overlap of a 2s orbital with the wavefunction of the departing photoelectron (de Broglie wavelength = 0.35 Å) yields a cross-section more than an order of magnitude greater than that of a 2p orbital.¹¹ For this same reason, H 1s levels have too little intensity to be seen. This strong cross-section effect is absent in second- and subsequent-row elements, where both types of orbitals have radial nodes. The p-like levels in Fig. 2, between 10 and 17 eV, are barely visible, whereas the (s-like) levels of interest are well-defined. A similar effect is present in solids.¹²

Let us summarize. The lack of suitable solid and gaseous materials precludes our using model ring compounds of the same element as a given amorphous semiconductor. This drawback is, however, far outweighed by the enhanced intensity of the carbon 2s atomic orbitals in x-ray photoemission and the ready availability of cycloalkanes.

To establish the value of cycloalkanes as model compounds for the solid structures in question, it now remains to show that cyclohexane is a good model compound for the s valence bands of diamond. Spectra of the valence bands of cyclohexane and diamond appear in Fig. 3, with the energy scale of diamond¹¹ shifted to correspond roughly with the cyclohexane spectrum. Also shown is the spectrum of $C(CH_3)_4$ (neopentane), to illustrate that simply having tetrahedral carbon bonds is not sufficient for simulating the diamond bands. If each data point in the cyclohexane spectrum is broadened by 0.8 eV (to reflect the

interaction of the rings in the solid), the resulting spectrum, shown at the top of Fig. 4, is almost identical to the diamond spectrum.

III. Theoretical

To understand the spectra of 2s orbital energies in the ring compounds, we have performed calculations at several levels of sophistication. We can use (and gain) the most physical insight at the simplest level, using the Hückel approximation in a qualitative way. At this level the eigenvalue problem for the σ orbitals describing C-C bonds is formally identical to that for the π system of planar cyclic aromatics, despite several obvious differences between the two problems.¹³ The easiest way to show this is to consider the problem of forming an N-membered ring of C-H₂ moieties, by σ bonding, using a two-fold degenerate orbital on each. This leads with the usual approximations to a secular determinant with elements

$$D_{ij} = (\alpha - E)\delta_{ij} + \beta \delta_{i,j\pm 1}$$

where α and β are the usual Hückel integrals and the indices are taken modulo N (i.e., $D_{1N} = D_{N1} = \beta$) for $i, j = 1, 2, \dots, N$. The roots are

$$E_n = \alpha + 2\beta \cos \frac{2n\pi}{N}, \quad n = 0, 1, 2, \dots, N-1$$

where α is the 2s energy of an isolated carbon atom and β is the interaction between neighboring C 2s orbitals. Both α and β are intrinsically negative. Each of the ring systems has one deepest singly-degenerate α level followed by e levels. In the case of even-membered

rings, the last level is again singly degenerate. For a 6-membered ring this gives two singly-degenerate levels at $E = \alpha + 2\beta$ and $\alpha - 2\beta$. Between them are two doubly-degenerate (e) levels at $E = \alpha + \beta$ and $\alpha - \beta$. The two e levels are separated by 2β , while the other levels are separated from the e levels by β . This leads to a gap centered at α . We interpret this gap as the big dip between the two large peaks in the cyclohexane spectrum, and (more qualitatively) as the dip in the s-band spectra of diamond and crystalline Si and Ge. Similarly there are gaps centered at -0.5α , 0.4α and -1.12α , and $\pm 0.7\alpha$ for 5-, 7-, and 8-membered rings, respectively. We note that the deepest level in each case has an energy of $\alpha + 2\beta$ while the least-bound orbital has an energy no higher than $\alpha - 2\beta$. Thus all the orbitals fall within the bounds of the energy region spanned by the 6-membered ring orbitals, and the width of the "valence band" is not increased as ring size is varied. This approach allows us to understand the gross features of our spectra.

To gain a more quantitative understanding of the spectra, more realistic calculations were performed employing more accurate geometries.¹⁴ First, the CNDO/2 semi-empirical method was used, yielding the results shown in Table I along with those from other computational methods. The CNDO/2 method is known to give exaggerated energy eigenvalues, but we thought that the level ordering would be useful and that the CNDO/2 wavefunctions, which tend to be much better than the energies, could be used to derive relative photoemission peak intensities, using the Gelius model.¹⁵ This is not the case. For example, in cyclopentane a p-type a_2'' level (not shown) is inserted between the e_2' and the e_1'

and in cyclohexane the a_{2u} and e_g are reversed. The intensities are also poor. There is too much mixing of C 2s and C 2p to be consistent with experiment.

We have available the Sichel and Whitehead CNDO method,¹⁶ which is known to give better energy agreement, and have tried it on this problem. Although it did not mix the s and p orbitals quite as strongly as CNDO/2, the orbital characters are quite strange. The deepest level in the odd-membered rings is an e level, not the a level that would be necessary to agree both with experiment and with our intuitive notion that the most stable level is the totally symmetric one. Indeed, the ordering seems to be almost precisely the reverse of that in the ab initio GAUSS-70 calculation,¹⁷ to be discussed below.

Disappointed in the CNDO methods, we tried extended-Hückel (EH), a semi-empirical method which is optimized for carbon compounds. The level ordering here is in perfect agreement with the ab initio results and the energies are quite good. Again, however, we see that the wavefunctions of the less deeply bound s-like levels are so strongly mixed with p and H 1s character that their intensities are not sufficient to reproduce the spectra.

It was necessary to use ab initio GAUSS-70 calculations to get a reasonable set of intensity ratios. These are shown in Table I.

IV. Discussion

Now that cyclohexane has been shown to be a good model for the s-like states of diamond, and assuming diamond to be a good model for the crystalline phases of interest, we can construct "spectra" for

hypothetical tetrahedrally-bonded amorphous carbon according to different structural schemes and note the effect on the valence-band structure. To do this we normalize the spectra of the ring compounds and then add the individual spectra together with weighting factors appropriate to the ring statistics of a given amorphous-lattice model. If we then broaden these "spectra" by 0.8 eV, we get something which can be directly compared to the "crystalline spectrum" in Fig. 4a.

Using Polk's model, for which he gave only the relative numbers of 5- and 6-membered rings, we get the "spectrum" in Fig. 4b. The minimum in the spectrum, which is still quite pronounced, is simply shifted toward lower binding energy. Indeed the dip merely shifts for any ratio of 5- to 6-membered ring. This can be seen in Fig. 4c, where we show the result of considering only these ring sizes in the ST-12 structure, which has twice as many 5- as 6-membered rings. The result is the same if only 6- and 7-membered rings are considered, except that in this case the minimum shifts to higher binding energy.

However, if we consider 5-, 6-, and 7-membered rings, which in ST-12 appear in the ratio 2:1:2, we get the curve shown in Fig. 4d. The two s-like peaks have not broadened significantly, nor has the splitting changed noticeably. The gap between them has simply been filled with new levels. To proceed one step further we can consider 8-membered rings. In ST-12 there are twice as many 8-membered rings as all the smaller ones combined. Including all these, one gets Fig. 4e. These 8-membered rings do not seem necessary to describe the spectra, but including them does not hurt.

The question of "short-circuiting" should be mentioned. This arises when a bridge connects two points in a ring. If a bridge contains fewer atoms than would be required to complete the original ring, the ring may be said to be "short-circuited" by a smaller ring. This is the reason Polk gave for not reporting rings larger than 6. If we apply this criterion to ST-12, however, we would have to say there are only 5-membered rings. This approach might not give a representative picture of the local environment.

In conclusion, we have employed cyclic alkanes as model compounds to test the hypothesis that amorphous semiconductors possess topological disorder, yielding odd-membered rings and filling in the gap between the s-band peaks. Spectral synthesis showed that 5, 6, and 7-membered rings are necessary to explain the XPS spectra observed in amorphous materials. This strongly supports the topological disorder model. Of more general interest from the viewpoint of solid-state chemistry, this paper provides an example in which rather detailed molecular-orbital ideas are used to explain a fairly subtle point about the valence-band densities of states of covalent solids.

ACKNOWLEDGEMENT

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- (14) The cyclopentane geometry was taken from W. J. Adams, H. J. Geise, and L. S. Bartell, *J. Amer. Chem. Soc.* 92, 5013 (1970). The cyclohexane geometry was given by O. Bastian, L. Fernholt, H. M. Seit, H. Kambara, and K. Kuchighu, *J. Molec. Struct.* 18, 163 (1973), and the C-C-H angle from N. V. Alekseev and A. I. Kitaigorodskij, *Z. Struct. Khim.* 4, 163 (1963). The cycloheptane geometry was provided by D. Bocian, private communication. Cyclooctane was calculated using tetrahedral angles, tub form, and distances from smaller rings.
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Table I. Binding Energies^a and Intensities^b of the s-Like Molecular Orbitals in Some Cyclic Alkanes: Theory and Experiment

LEVEL ^c	THEORY								EXPERIMENT	
	EH ^d		CNDO		SW/CNDO ^e		GAUSS 70 ^f		E _B	I/I ₀
	E _B	I/I ₀	E _B	I/I ₀	E _B	I/I ₀	E _B	I/I ₀		
cyclopentane										
a ₁ '	26.3	1.0	49.4	1.0	20.0	0.7	28.8	1.0	25.5(2)	1.0
e ₂ '	23.0	1.4	36.4	1.5	22.4	1.7	24.9	2.2	22.1(1)	2.0(1)
e ₁ '	19.0	0.7	27.1	0.8	24.8	2.0	19.7	1.9	18.4(1)	1.4(1)
cyclohexane										
a _{1g}	26.4	1.0	50.8	1.0	20.0	0.7	29.1	1.0	25.5(2)	1.0
e _u	23.8	1.6	38.7	1.7	21.8	1.6	26.0	2.2	23.2(1)	
e _g	19.9	0.9	29.7	1.0	24.3	1.9	21.1	2.1	19.7(1)	0.7(1)
a _{2u}	18.9	0.2	30.3	0.2	25.1	1.0	19.0	0.6	18.9(3) ^g	
cycloheptane ^h										
a ₁ '	26.4	1.0	49.1	1.0	20.1	0.6	28.7	1.0	25.1(3) ^g	1.0
1e ₂ '	24.4	1.7	40.7	1.8	21.5	1.6	26.6	2.1	23.7(1)	
e ₁ '	21.0	1.0	32.8	1.2	23.8	1.9	22.7	2.1	20.6(1)	0.6(2)
2e ₂ '	18.8	0.5	28.8	0.3	25.1	2.0	19.4	1.6	18.2(1)	0.5(1)
cyclooctane ⁱ										
a ₁	26.7	1.0	54.7	1.0	20.3	0.7	29.6	1.0	25.7(2) ^g	1.0
1e	24.5	1.7	41.8	1.8	21.4	1.5	27.0	2.4	24.4(1)	
2e	21.9	1.2	34.9	1.3	23.4	1.8	23.9	2.3	22.1(1)	0.6(2)
3e	19.8	0.7	29.3	1.0	24.8	2.0	21.1	1.9	19.7(1)	0.7(1)
a ₂	17.7	0.3	28.9	0.1	25.3	1.0	18.0	1.1	18.8(2) ^g	

(continued....)

Table I (continued)

- a) In eV.
- b) Relative to the deepest level. Where in SW/CNDO the deepest level is predicted to be an e level it is set to be $I_o = 2.0$. Calculations are based on net populations with a $\sigma(\text{C}2s) : \sigma(\text{C}2p) : \sigma(\text{H}1s)$ cross section ratio of 13 : 1 : 0. For these levels the exact ratios used are not important as long as $\sigma(\text{C}2s) \gg \sigma(\text{C}2p)$. For the experimental results, the deepest resolvable band is set to $I_o = 1.0$. See ref. 15.
- c) The level assignments for cyclopentane and cyclohexane are based on the planar (D_{5h}) and chair (D_{3d}) geometries¹⁴ used respectively. The assignments for cycloheptane and cyclooctane are discussed in notes h and i .
- d) Extended Hückel.
- e) See ref. 16.
- f) See ref. 17.
- g) Where a levels are very closed to e levels, it was necessary to fix their area ratios to those from GAUSS 70 when performing a non-linear least squares fitting program. This enabled good estimates of the energy separation between the a and e levels.
- h) Unlike cyclopentane, which can be considered flat with 108° C-C-C angles, planar cycloheptane would have 128.6° angles and considerable strain. The molecule actually pseudorotates among several forms. The lowest energy form is a twisted chair,¹⁴ which was used for the calculations and has no symmetry. The molecular orbitals can, however, be grouped according to energy and atomic orbital composition into an a and three e levels and assigned as if the molecule were D_{7h} , as shown.
- i) Cyclooctane calculations were performed using the tub geometry, D_{2d} . The b_2 and b_1 levels, which have the same atomic orbital character, are reported as $2e$. Because the tub form forces four sets of hydrogens within 1.4 Å of each other, the a_2 level is ~ 1 eV lower E_B than expected. The intensities, however, are quite insensitive to the precise geometry as long as the C-C distances and angles are constant.

FIGURE CAPTIONS

- Fig. 1. Cleaved-crystal and amorphous-film photoelectron spectra of silicon, germanium, and arsenic taken with monochromatized Al $K\alpha$ x-rays (1486.6 eV). Binding energy scale is relative to the Fermi level. Peaks 1 and 2 are s-like. See refs. 1 and 2.
- Fig. 2. Gas-phase photoelectron spectra of cyclopentane, cyclohexane, cycloheptane, and cyclooctane taken with Mg $K\alpha$ x-rays. The same spectra taken using Al $K\alpha$ x-rays look identical to these.
- Fig. 3. A comparison of the XPS spectra of cyclohexane, diamond, and neopentane $[C(CH_3)_4]$. The energy axis of the diamond spectrum has been shifted to roughly align the peaks with those in cyclohexane. This procedure is necessary because gas and solid phase spectra are referenced differently and thus only energy differences can be compared accurately.
- Fig. 4. The spectrum of cyclohexane (see Fig. 3) broadened by 0.8 eV and spectra simulating different ring systems (also broadened by 0.08 eV). See text.

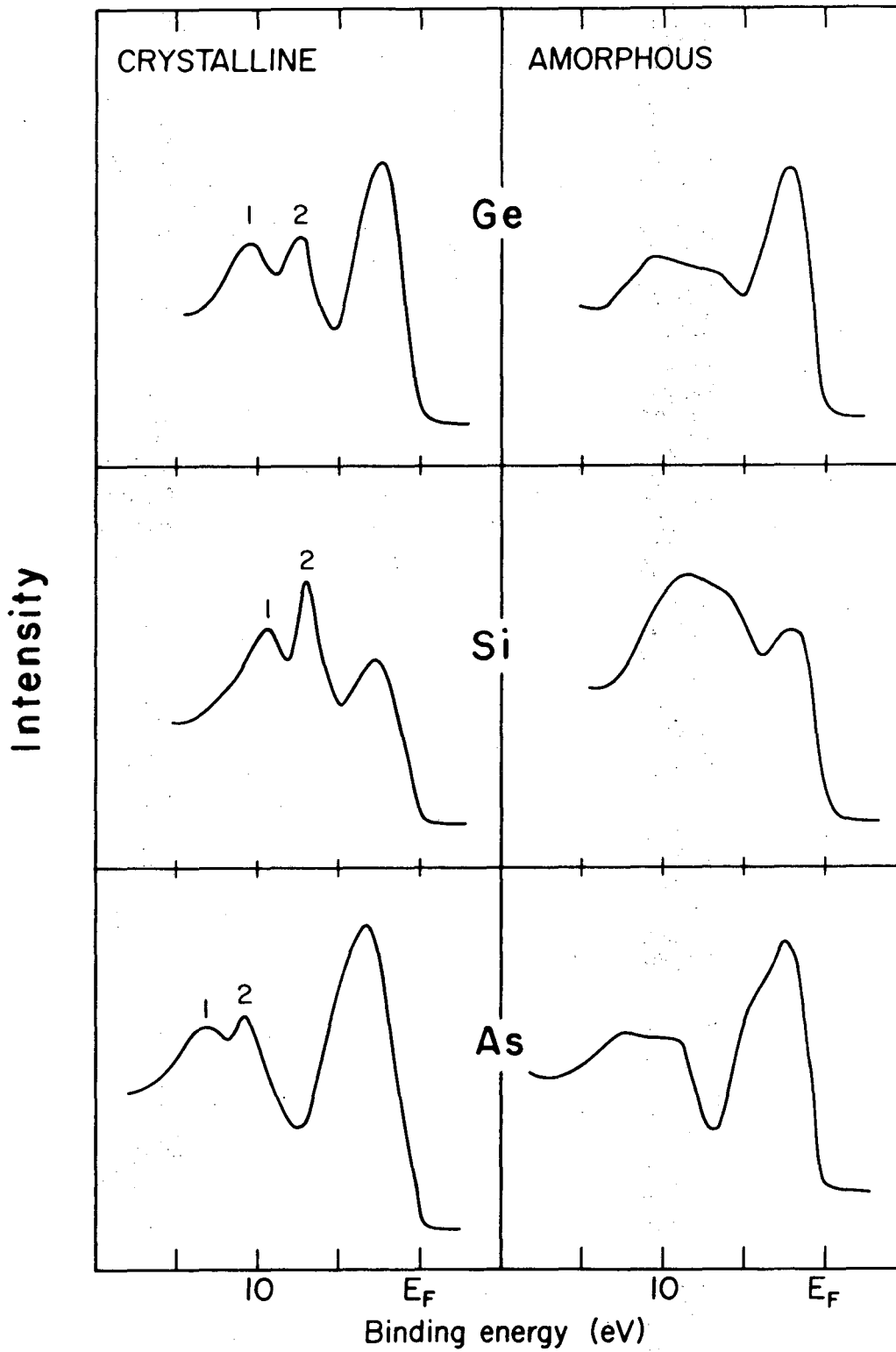


Fig. 1

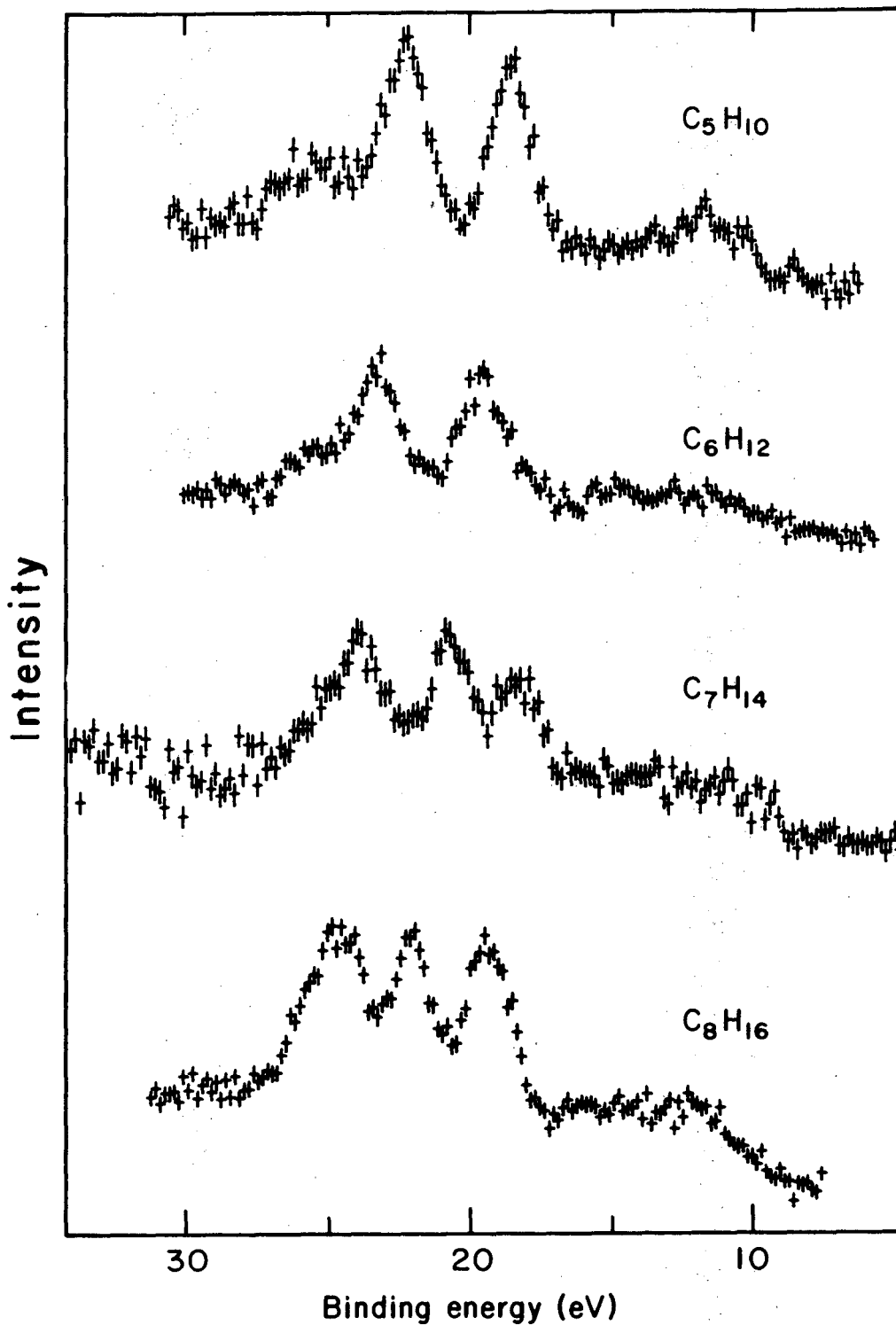


Fig. 2

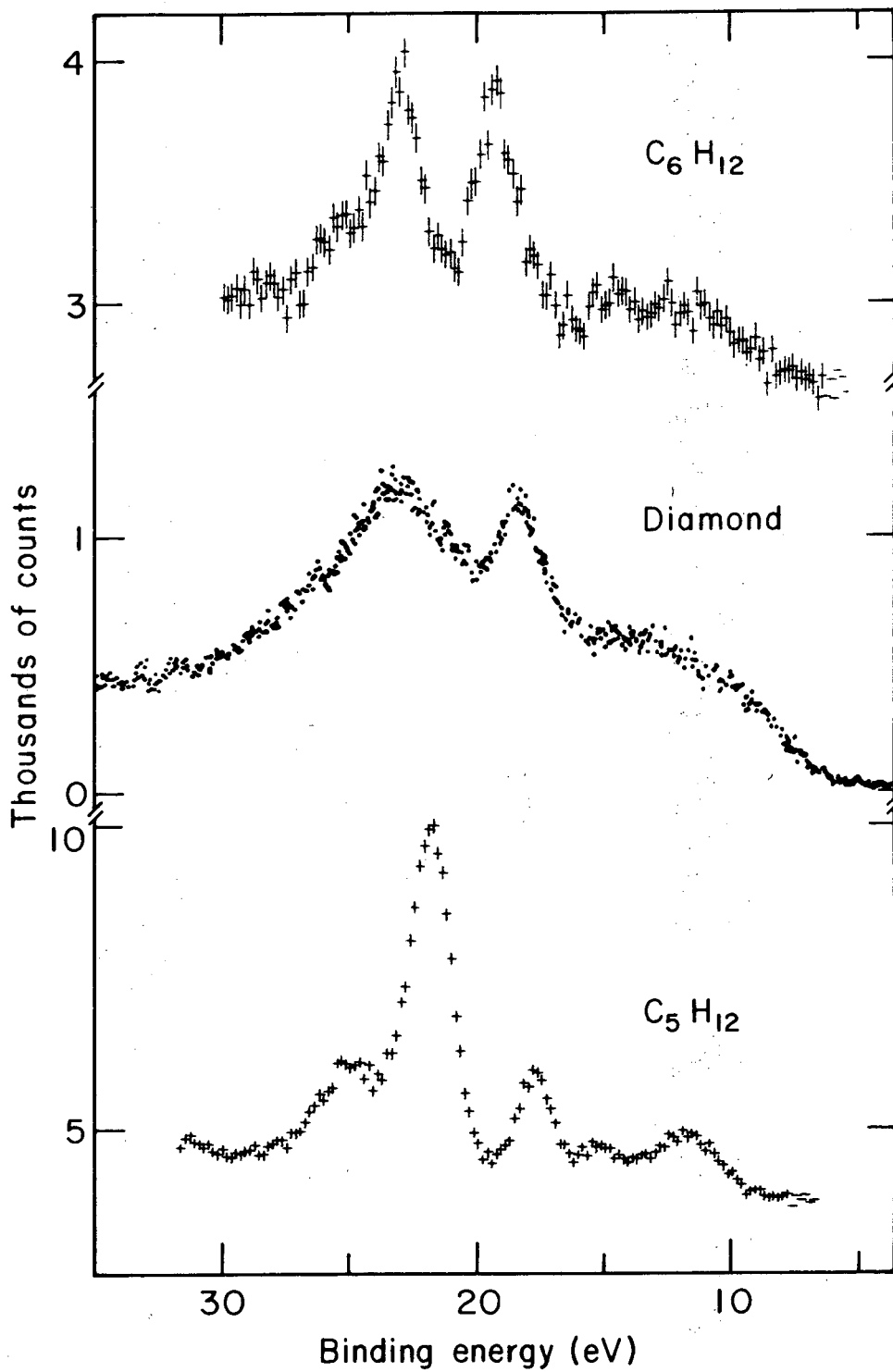


Fig. 3

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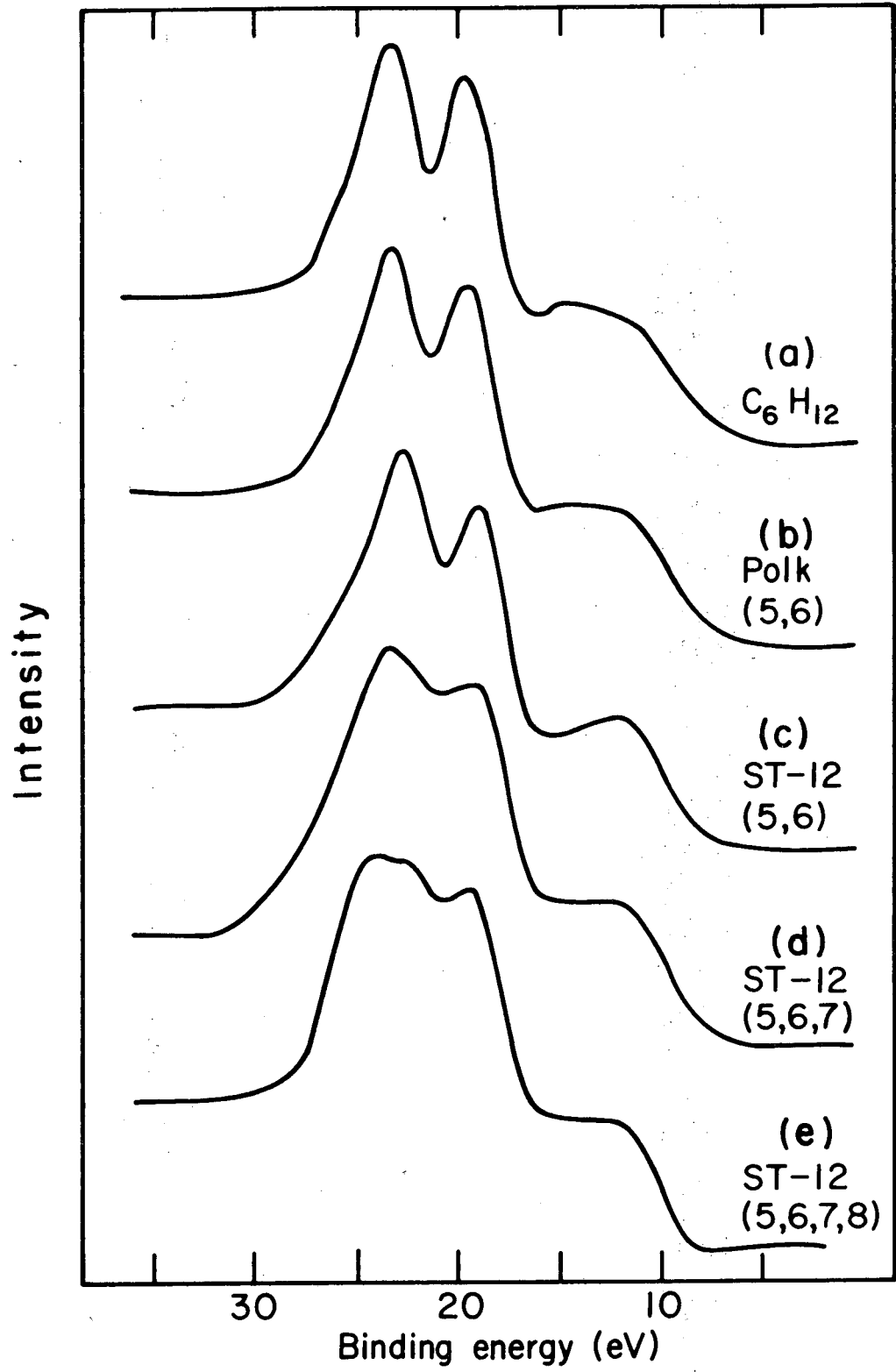


Fig. 4

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