

LOW-ENERGY ELECTRON-IMPACT SPECTROSCOPY OF C₆₀
BUCKMINSTERFULLERENE MOLECULE

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Abstract

The methods of electron-impact spectroscopy were utilized to obtain the first low-energy, high-resolution energy-loss spectra of gas phase pure C_{60} and $C_{60} + C_{70}$ mixture buckminsterfullerene molecules. The impact energies and scattering angles ranged from 8 to 100 eV and 0° to 90° , respectively. Broad spectral features in the energy-loss spectra have been assigned to overlapping electronic transitions based on the identification of features in the photoabsorption spectrum of C_{60} molecules in n-hexane solution by Leach et al. (1992 *Chem. Phys.* 160, 451). Pure vibrational excitation features were also observed. The relative scattering intensities of these spectral features represent the corresponding relative cross sections.

1. Introduction

The C_{60} (buckminsterfullerene or buckyball) molecule was discovered in 1985 (Kroto et al., 1985) but it was only very recently (Krättschmer et al. 1990) that substantial amount of this compound could be manufactured for extensive studies. Here we concern ourselves with the electronic, energy levels of this molecule and the relative cross sections associated with various energy-loss features as obtained from low-energy electron scattering experiments.

Some information on the electronic energy level structure of C_{60} (and C_{70}) has been available from optical absorption measurements in various solutions (Ajie et al., 1990; Hare et al., 1991; Leach et al., 1992), and in the solid phase (Krättschmer et al., 1990; Shinohara et al., 1991; Lichtenberger et al., 1991; Hebard et al., 1991; Skumanich, 1991; Gasyna et al., 1991 and Weaver et al., 1991). The first gas phase absorption study was carried out by Heath et al. (1987). More recently, Haufler et al. (1991a) reported photon absorption spectra of C_{60} and C_{70} in supersonic molecular beam, Brady and Beiting (1992) measured the absolute photo absorption cross section for gas phase C_{60} . A number of investigations were aimed to gain information on the lowest electronic states of buckminsterfullerenes in various solutions (Arbogast et al., 1991; Hung and Grabowski, 1991; Terazima et al., 1991; Kajii et al., 1991; Ebbesen et al., 1991; Tanigaki et al., 1991; and Lee et al., 1991), and in supersonic beam (Haufler et al., 1991b). Luminescence of C_{60} (C_{70}) in solution, excited by photons, was studied by Arbogast and Foote (1991), Reber et al. (1991) and Sibley et al.,

(1992.) and in solid, excited by electron impact, by Jest et al. (1991) and by Weaver (1992). Photoionization measurements on buckyball were performed by Lichtenberger et al. (1991), Hertel et al., (1992) Yoo et al. (1992) and Ding et al. (1993). It should be mentioned here also that Zimmerman et al., (1991) determined ionization potentials for C_{50} , C_{60} and C_{70} by a Fourier-transform, ion-cyclotron-resonance, mass-spectrometric, charge-transfer bracketing technique.

in the area of electron-impact spectroscopy, the situation can be briefly summarized as follows. Energy-loss studies of solid C_{60} (C_{70}) at high impact energies (E_0) with low energy resolution have been carried out by Hansen et al. (1991; E_0 =high unspecified), Saito et al. (1991a and b; E_0 =200keV), Kuzuo et al. (1992; E_0 =60KeV) and Tong et al. (1991; E_0 = 206 eV) Kuzuo et al. also obtained an energy-loss spectrum with 0.14eV resolution. The first low-energy, high-resolution (about 10 meV for vibrational and about 40 meV for electronic state. excitations) electron reflection, energy-loss spectroscopy of solid C_{60} was reported by Geusterblum et al. (1991) and described in more detail by Lucas et al. (1992), see also Lucas (1992) and Geusterblum et al. (1992), They identified 11 of the 46 distinct pure vibrational excitations in their 3.7 eV impact-energy, energy-loss spectrum. In the energy-loss region (corresponding to the visible and UV optical regions) using 10 and 30 eV energy electrons, they clearly identified the lowest energy-loss feature. at 1.55 eV and strong features at 2.2, 3.7, 4.8, 6.3 and 7.6 eV energy losses. In addition, they observed 10 weak shoulders in this region of the energy-loss spectrum. They also utilized electrons with impact energies ranging from 70 to 150 eV to extend the energy-loss region up to 50eV.

At energy-losses larger than 7.6 eV, they found five weak, broad features and the strong plasmon excitation at 28 eV. Sohmen et al. (1992a and b) studied the energy-loss spectra of C_{60} and C_{70} (as well as of K-doped C_{60}) films using 170 keV electrons in a transmission arrangement with 0.13 eV energy resolution. They were mainly concerned with the plasmon excitations. The only gas-phase electron-impact energy-loss spectra published for buckyball so far are those obtained by Keller and Coplan (1992). They utilized 1 keV electrons and observed the scattering from a mixture of C_{60} and C_{70} beam at 1.5° and 10° scattering angles with an energy resolution of 0.6 eV. Energy-loss features in these spectra (2.5, 3.9, 4.9, 6.3, 7.5, 10, 13, 17 and 21.5 eV) correspond approximately to those found in optical and electron impact studies of solid buckyball. They converted the low-momentum-transfer spectrum to relative optical oscillator strength (absorption cross section) in the 1 to 28 eV (IR to 400 Å) region. Electron impact ionization cross sections for C_{60} were measured in the threshold to 50 eV impact energy region by Baba et al. (1992). Very recently, Trajmar et al. (1993) observed UV fluorescence induced by electron impact on C_{60} and C_{70} molecules in an experiment utilizing beam-beam arrangement. They assigned these emissions to transitions in the C_{60}^+ and C_{70}^+ ions.

Here we describe differential, electron-energy-loss measurements on pure C_{60} and mixed ($C_{60} + C_{70}$) thermal beams and present energy-loss spectra obtained at low impact energies (8 to 100 eV) with a resolution of about 0.08 eV at scattering angles (θ) ranging from 0° to 90° . From these spectra, we deduce information on the energy level structure, relative electron-impact excitation and optical absorption cross sections.

II. Experimental

in the present work we utilized low-energy, high-resolution electron scattering techniques and a beam-beam scattering arrangement. We studied both a mixture composed of approximately 66% C_{60} , 25% C_{70} and 3% each of C_{76} , C_{78} and C_{84} and pure C_{60} (purchased from Materials and Electrochemical Research Corporation). We will refer to the buckyball mixture as **BBM** in the present paper. This mixture was used in the preliminary studies for economical reason and because it was expected that the general features of the C_{60} and C_{70} energy-loss spectra would be similar. Support for this expectation came from comparison of spectra obtained for C_{60} , C_{70} and mixtures in earlier optical and electron-impact studies (in solutions and in the solid phase) and from gas phase electron energy-loss studies carried out by Keller and Coplan (Keller, 1992). In our final studies pure C_{60} sample was used.

The apparatus and experimental procedures have been described in detail earlier (Trajmar and Register, 1984). Electrostatic lens systems and hemispherical energy selectors were utilized to form a well collimated electron beam with the desired impact energy and resolution. The molecular beam was formed by heating a stainless steel crucible containing the sample and collimating the effusing beam. Normal operating temperature as measured at the surface of the crucible was about 500°C . At the beginning of the experiments sufficient beam intensities were obtained at around 300°C , but as time went on the temperature had to be raised to obtain the same beam intensity. The electron and target

beams crossed each other at 90° . Scattered electrons were detected over a solid angle of about 10^{-3}sr at fixed scattering angles and energy analyzed using electrostatic lenses and a pair of hemispherical analyzers. The scattered signal intensity as a function of energy loss constituted the energy-loss spectrum. Each spectrum was obtained by repetitive scanning utilizing pulse counting and multichannel scaling techniques. Typical energy resolution in these measurements were about 0.08eV . The impact energies and scattering angles are nominal but calibrations were carried out to make sure that these values are very close to the true values. The contact potential for the instrument was measured on the He elastic scattering resonance at 18.36eV and found to be about 0.3eV . The angular resolution was about $\pm 2^\circ$ and the true zero angle was checked both on the symmetry of inelastic scattering and the electron beam direction (as determined by using the detector as a Faraday cup). Deviations from the nominal angles were less than $\pm 1^\circ$.

111. Results and Discussion

We obtained spectra in the zero to 10eV energy-loss range at impact energies ranging from 8 to 100eV and scattering angles ranging from 0° to 90° . Typical energy-loss spectra are shown in Figs. 1 to 6. Fig. 1 shows an energy-loss spectrum at $E_0 = 20\text{eV}$, $\theta = 5^\circ$ taken in the early stage of evaporation of the BBM sample. It contains contamination features associated with Zn, CO and H_2O which conveniently serve to calibrate the energy-loss scale to an accuracy of better than 10meV . Subsequent spectra were taken after the contaminations were eliminated. The fullerene features are indicated by letters which correspond to the designations of Keller and Coplan (1992) and to our designations in the

figures and also in Table I which summarizes the energy-loss information obtained by various investigators. The Zn line in this spectrum came from our furnace structure, and was later eliminated. In the case of C_{60} samples the initial contamination was much less.

Fig. 2 shows spectra obtained at $E_0 = 100$ eV. The scattering angles, target species and feature designations are indicated. The general appearance of C_{60} and BBM spectra are very similar at this relatively high impact energy and these low scattering angles. There are, however, some differences which are associated with the presence of C_{70} in the BBM sample and indicate that the spectra of C_{60} and C_{70} are not identical. For example, we found that the energy-loss region 2.5 to 3.0 eV (between features b and c) which is a valley in the case of pure C_{60} is filled up in the BBM spectra. There is also a slight shift (~ 0.2 eV) to the lower energy-loss direction for the C_{60} spectra with respect to the BBM spectra for features b, c, e and f (but not for feature d). Feature f is well defined at 0° but it is weaker and less defined at higher angles. All these features should be associated with dipole allowed excitations which typically dominate low-momentum-transfer spectra. (See e.g. Rice et al., 1968 and Cartwright et al., 1977). There is a change in the relative intensities of the features with scattering angle indicating somewhat different angular dependence of the corresponding cross sections. Each feature corresponds to a large number of undistinguishable excitation processes. We will discuss their possible assignment below. The dominant features are at 2.2, 3.7, 4.8, 6.1 and 7.5 eV energy losses. These values are in good agreement (see Table I) with those obtained by Leach et al. (1992) and Lucas et al. (1992) indicating that solvent effects and the weak Van der Waals bonding in the solid have no significant effect on the electronic energy levels of the strongly bound C_{60} molecule.

We have also investigated the energy-loss region from 10eV up to 30eV but found no clearly recognizable fullerene feature. We particularly searched for the weak features at 10, 13, and 17 eV found by Keller and Coplan (1992) and Lucas et al. (1992). At very low angles and after many scans we observed features at around 10 and 13 eV which can be definitely assigned to electronic excitation in H₂O and N₂, respectively. At low angles, because of the long scattering path length, these background features can be observed even though our background pressure during the experiment was typically 10⁻⁶ Torr. We also looked for the collective excitation feature around 20 eV energy loss. It appears but is rather weak in our spectrum. This is somewhat surprising since the corresponding feature is quite well developed in the electron reflection spectrum of C₆₀ film at this impact energy range. (See Fig. 6 of Lucas et al., 1992).

Fig. 3 shows a collection of spectra obtained at E₀=20eV. No significant difference between the pure C₆₀ and BBM spectra was found here. The relative intensities of the spectral features change with angle and the sharp definition of the features gradually disappears with increasing angle due to the increased importance of contribution from optically forbidden excitations. This becomes especially apparent at 40° scattering angle (not shown). At this angle the scattering signal becomes weak and prevented us from collecting data at higher angles. The onset for inelastic scattering in these spectra is at 1.65 eV, and there is no clear indication for the lowest triplet state excitation found at around 1.5eV in solutions by indirect methods (Arbogast et al., 1991; Hauffler et al., 1991; Hung et al., 1991; Terazima et al., Tanigaki et al., 1991; Lee et al., 1992; Arbogast and Foote, 1991; Wasielewski et al., 1991; Sibley et al., 1992; and Kim and Lee, 1992). The 7.5 eV

energy-loss feature is not recognizable at this impact energy. At this impact energy our energy-loss spectra are practically identical with those of Lucas et al. (1992).

Fig. 4 shows energy-loss spectra obtained at 10 eV impact energy. These spectra are similar in general appearance to those obtained at $E_0 = 20$ eV. We also show the elastic scattering peak and clear indication for vibrational excitation in these spectra. (This matter will be addressed later.) There is no indication for the lowest triplet state excitation, although at the same impact energy a well defined peak is present at 1.55 eV in the electron reflection spectrum. There is a significant difference between the pure C_{60} and the BBM spectra indicating different energy-loss distributions for C_{60} and C_{70} . The reflection spectrum at this energy is more similar to our BBM spectra than to those of C_{60} . Figure 5 shows a spectrum obtained with 8 eV impact energy and 17° scattering angle. The optically allowed features are not sharply defined and contribution from spin and/or symmetry forbidden transitions are apparently present filling up the valleys found at higher impact energies. A weak feature now appears at 1.55 eV energy loss indicating excitation to the lowest triplet states. This excitation is not as clearly apparent as in the 10 eV spectrum of Lucas et al. (1992).

We can assign the observed energy-loss features to groups of electronic transitions based on the work of Leach et al. (1992). They measured the absorption spectrum of C_{60} in n-hexane solution with 10 Å resolution in the 1900 to 7000 Å region at room temperature and in n-hexane and 3-methylpentane solutions at 77°K with 2.5 Å resolution in the 3900 to 7000 Å region. Their assignments were based on theoretical results and single electron excitations (and of course apply to the spectrum of C_{60} in solution). A comparison of

solution, solid and gas phase spectra of fullerenes, however, reveals a great deal of similarities irrespective of the phase of the sample. Furthermore, no gas phase spectrum has been determined. Until such spectrum becomes available this is the best one can do.

The ground electronic state of C_{60} corresponds to the closed shell molecular orbital configuration: $a_g^2 t_{1u}^6 h_g^{10} t_{2u}^6 g_u^8 g_g^8 h_g^{10} h_u^{10}$ and is designated as 1^1A_g . The lowest two excited configurations and the corresponding terms are:

$$\begin{aligned} \dots h_g^{10} h_u^9 t_{1u} & \quad (T_{1g}, T_{2g}, G_g, H_g) \text{ (excitation to the LUMO)} \\ \dots h_g^{10} h_u^9 t_{1g} & \quad (T_{1u}, T_{2u}, G_u, H_u) \end{aligned}$$

in addition, excitations of the type (HOMO-1) \rightarrow LUMO corresponding to $h_g^9 h_u^{10} t_{1u}$ (T_{1u}, T_{2u}, G_u, H_u) are possible. Absorption and excitation to the bands of the lowest triplet states are indicated by the symbols α and β in the spectrum of I-etch et al., and fall in the less than 1.78 to 1.88 eV region. The only indications for these excitations in our energy-loss spectra are the very weak feature at 1.55 eV (indicated as t in Fig. 5) and the shoulder on the low-energy-loss side of feature b in the 8 eV impact energy spectrum. The absorption bands in the 2.00 to 3.00 eV region have been assigned to optically forbidden singlet-singlet transitions (indicated by $\gamma, \delta, \epsilon, \xi,$ and η , symbols by Leach et al). These transitions can be induced by the Herzberg-Teller effect and are influenced by Jahn-Teller vibronic interactions. In our energy-loss spectra, we find a broad peak but no structure in this region. The peak indicated as b in our low-momentum-transfer C_{60} spectra (which most closely corresponds to optical absorption) is at around 2.1 eV and most likely corresponds to the $J^1A_g \rightarrow 1^1T_g$ and $1^1A_g \rightarrow 1^1T_{2u}$ excitations. Above 3.00 eV, features corresponding to optically allowed transitions begin to appear. The scattering intensity in our spectra

gradually increases above 3.00 eV (due to transitions indicated by Leach et al. as A and B) and the first energy-loss peak is formed at around 3.7 eV (designated by c in our energy-loss spectra and by C in the absorption spectrum of Leach et al.). Leach et al. assigned this peak to the $1^1A_g \rightarrow h^1T_{1u}$ transition. The next peak, denoted as d in our spectra, appears at around 4.8 eV. Leach et al. assign the absorption at this energy to the $1^1A_g \rightarrow n^1T_{1u}$ ($n=3,4$ and 5) excitations and denote the corresponding absorption features as D and E. The broad feature appearing in our spectra at around 6.1 eV corresponds to the features F, G and H in the absorption spectrum and Leach et al. and they assigned them to the $1^1A_g \rightarrow n^1T_u$ ($n=7, 8$ and 9) excitations. The feature observed at around 6.5 eV in the energy-loss spectra of solid C_{60} (C_{70}) was assigned by Saito et al. (1991), Kuzuo et al. (1991), Gensterblum et al. (1991), Lucas et al. (1992) and Sohmen et al. (1992b) to the collective excitation of the valence electrons. Keller and Coplan (1992) made the same assignment for the energy-loss feature observed by them at 6.0 eV in their vapor phase energy-loss spectrum of mixed $C_{60}+C_{70}$ sample. It is likely that both single-electron and collective plasmon excitations contribute to the scattering signal in this energy-loss region.

Leach et al. did not cover the region corresponding to our 7.5 eV feature. Naturally, a large number of other spin and/or symmetry forbidden transitions can be present in our energy-loss spectra, but these are weak in spectra obtained at high impact energies and low scattering angles. There is clear indication, however, for their presence in our spectra taken at the lower impact energies (8 and 10 eV) and at higher ($\theta > 20^\circ$) scattering angles as discussed above. The energy-loss values corresponding to the prominent (optically allowed) transitions in our spectra are in good agreement with the values obtained from the

absorption spectrum of Leach et al. The energy-loss spectrum obtained by Keller and Coplan (1992) for a 80% C_{60} + 20% C_{70} mixture at $E_0 = 1 \text{ keV}$ and 1.50 scattering angle shows features indicated as b, c, d, e and f (both in their and our spectra). The energy-loss values for these features are in very good agreement with those obtained from our spectra, (see Table 1). As we mentioned above, we did not observe any other energy-loss feature attributable to buckyball above the 7.5 eV energy-loss feature in our spectra.

In Fig. 6 we show energy-loss spectra corresponding to elastic scattering and vibrational excitation for C_{60} . Weak, but definite energy-loss features appear at 0.17 and 0.36 eV. The leg of the elastic scattering feature is indicated by dashed line (based on the symmetry of this feature). The peak at 0.17 eV can be assigned on the basis of the work of Gensterblum et al. (1991). They found prominent peaks at 0.156 and 0.194 eV energy losses in the electron reflection spectrum of solid C_{60} using 3.7 eV primary electrons and 0.01 eV resolution. The 0.36 eV energy-loss feature is most likely associated with the C-H stretching mode excitation of some contaminant which is present in the sample,

The 100 eV, 0° energy-loss spectrum of C_{60} shown in Fig. 2 was converted to optical absorption cross sections utilizing the procedure previously described by Khakoo et al. (1990). We assume that at this energy and angle the optical limit has been approximately reached. In Fig. 7, we compare these cross sections with those obtained by Keller and Coplan (1992) from their 1 keV, 1.50 energy-loss spectrum and to the direct optical absorption measurements of Brady and Beiting (1992) and Leach et al. (1992). The data of Leach et al. were in relative units and we normalized them to our value at the 4.8 eV peak. The optical absorption cross section deduced from the present study (in the 1.5 to

7 eV or 265 to 1771 Å region) is in good agreement with those deduced by Keller and Coplan (1992) in the 1.5 to 5.0 eV region but their values are larger than ours in the 5 to 7 eV region. The agreement between the cross section measured directly by photoabsorption techniques by Brady and Beiting (1992) in the 1.5 to 4.2 eV region and our result is excellent.

in Fig. 8 the angular behavior of the differential cross section (DCS) for excitation of the 4.62 to 4.98 eV energy-loss region is shown. The DCS are strongly forward peaked and their value decreases by about three orders of magnitude, from 5° to 90°.

IV. Summary and Conclusions

Energy-loss spectra were obtained for pure C₆₀ and C₆₀+C₇₀ mixture of buckminsterfullerene molecules at impact energies and scattering angles ranging from 8 to 100 eV and 0 to 90°, respectively. This work represents the first low-energy, high-resolution electron scattering study of gas phase buckyballs. Broad inelastic scattering features found in these energy-loss spectra correspond to overlapping electronic transitions. Pure [vibrations] excitation features were also observed. The energy-loss values found here for the gas phase species correspond closely to those observed previously in the solid phase and in solutions of buckyballs, although some features observed previously are not present in our spectra. The general characters of energy-loss spectra obtained for pure C₆₀ and for the mixtures of C₆₀ and C₇₀ are very similar but there are apparent differences indicating that the spectra of C₆₀ and C₇₀ are not identical. The relative scattering intensities of the

spectral features measured here represent the relative values of the corresponding differential cross sections. The photoabsorption cross section derived from our 100 eV, 0° spectrum agrees within about 10% with those deduced by Keller and Coplan (1992) except in the 5 to 7 eV region where their values are considerably larger. The direct photoabsorption cross sections measured by Brady and Berting in the 1.5 to 4.2 eV region are in excellent agreement with our results.

Further investigations of this type, preferably with expansion cooled buckyball beams, would be needed to learn more details about the electron impact excitation processes and the absolute cross sections associated with them. It would be also desirable to extend the measurements to the 100 to few keV impact energy range to obtain information on the higher energy-loss features and on the collective excitation of the valence electrons corresponding to an energy-loss of about 20 eV. We plan to address these matters in our future studies,

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Table I. Summary of excitation energies (in eV) obtained recently from photo absorption]] and electron energy-loss measurements for C_{60}

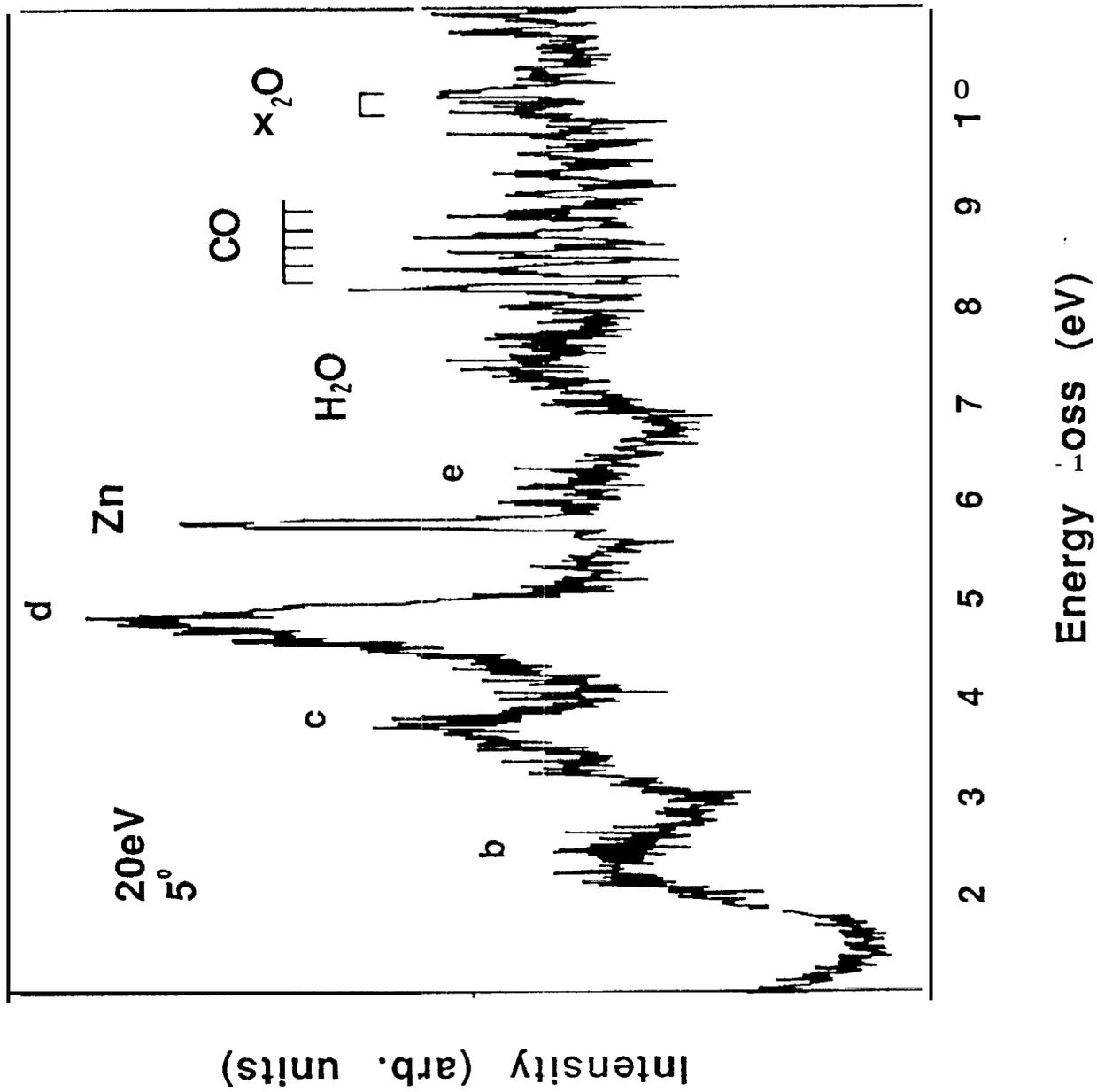
Optical — Solution	Electron scattering		Present	Designation ^d
	solid Lucas et al ^b	Gas phase Keller-Coplan ^c		
Leach et al ^a	1.55 (1.72) (1.92)			—
2.26(8)	2.20 (2.36) (2.58) (2.74) (3.0) (3.3) (3.4)	2.5	2.35	b
3.78(C)	3.7	3.9	3.72	c
4.83(E)	4.8 (5.5) (5.8)	4.9	4.82	d
5.88(G)	6.3 7.6 (9.4) (10.2) (12.8) (14.3) (17.2) 28	6.0 7.5 10 13 17	6.1 7.5	e f g h i

- a. Leach et al., 1992. Only the strong peaks are indicated with the corresponding designations of Leach et al.
- b. Lucas et al., 1992. Parenthesis indicates small shoulders on strong features.
- c. Keller and Coplan, 1992. The target beam contained both C_{60} and C_{70} .
- d. Designation used by Keller and Coplan and in our spectra.

Figure Captions

1. Energy-loss spectrum of **BBM** obtained at **20eV impact energy and 5° scattering angle**. Contamination features corresponding to the **Zn** resonance excitation, to vibrational bands of the **CO A¹ Π state** excitation and several electronic state excitations in **H₂O** are indicated.
2. Energy-loss spectra obtained at **$E_0=100\text{eV}$** . The target species and scattering angles are indicated.
3. Same as Fig. 2 except **$E_0=20\text{eV}$** .
4. Same as Fig. 2 except **$E_0=10\text{eV}$** . In the top and bottom spectra the elastic scattering and vibrational excitation region are also shown.
5. Energy-loss spectrum obtained at **$E_0=8\text{eV}$ and $\theta=17^\circ$** . The onset of electronic state excitation is indicated.
6. Energy-loss spectra obtained at **$E_0=10\text{eV}$** showing the elastic scattering and pure vibrational excitation features for **C₆₀**.
7. Optical absorption cross sections: dots, present results obtained from our **$E_0=100\text{eV}$, $\theta=0^\circ$** energy-loss spectrum for pure **C₆₀**; crosses, the results of **Keller and Coplan (1992)** obtained from their **$E_0=1\text{keV}$, $\theta=1.5^\circ$** energy-loss spectrum for **C₆₀** and **C₇₀** mixture; heavy solid line, obtained by **Brandy and Beiting (1992)** from optical absorption measurement for gaseous **C₆₀** sample; and light solid line, obtained by **Leach et al. (1992)** from optical absorption measurement for **C₆₀** in **n-hexane** solution.

Fig. 1



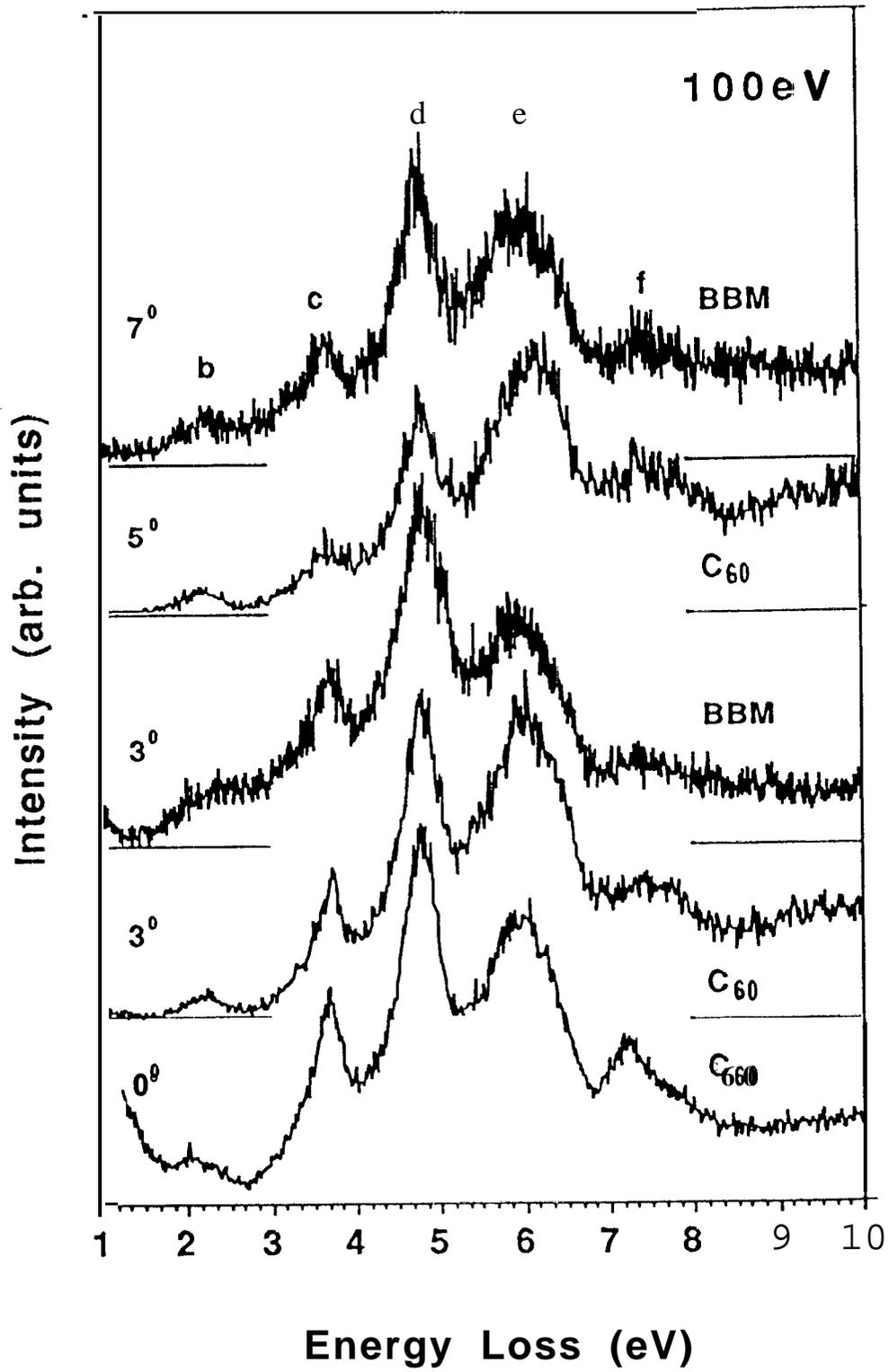


Fig. 2

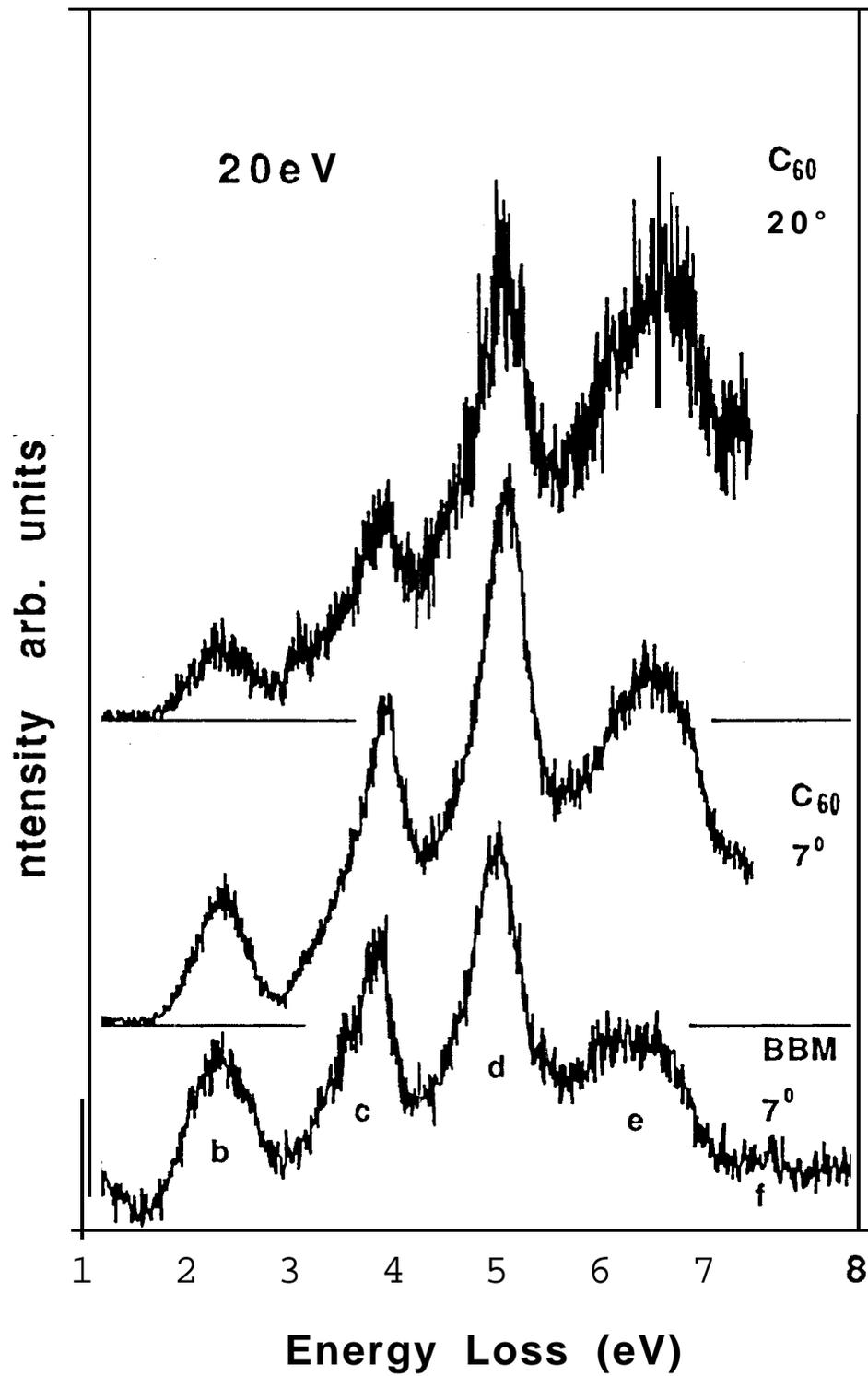


Fig 3

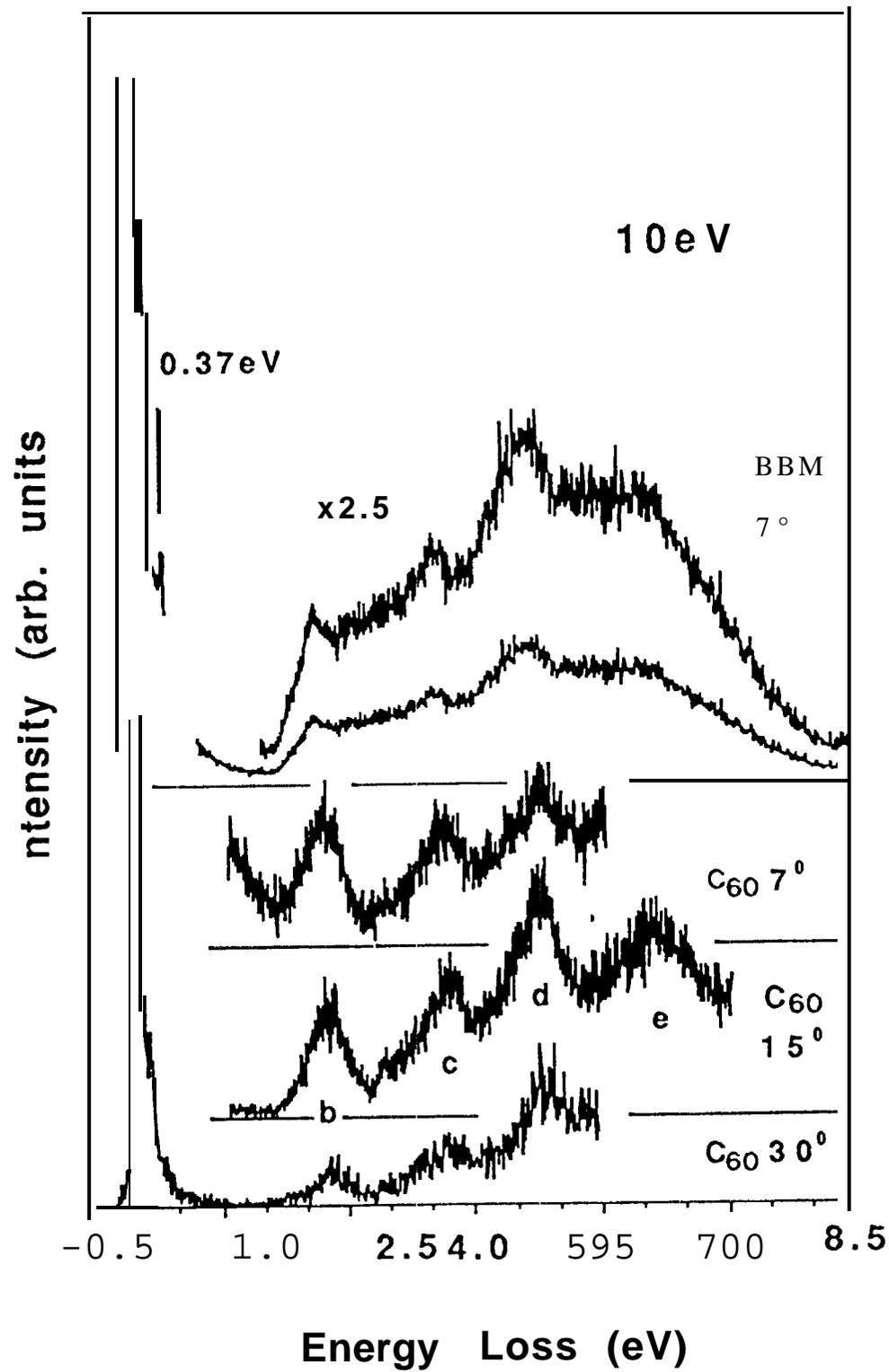
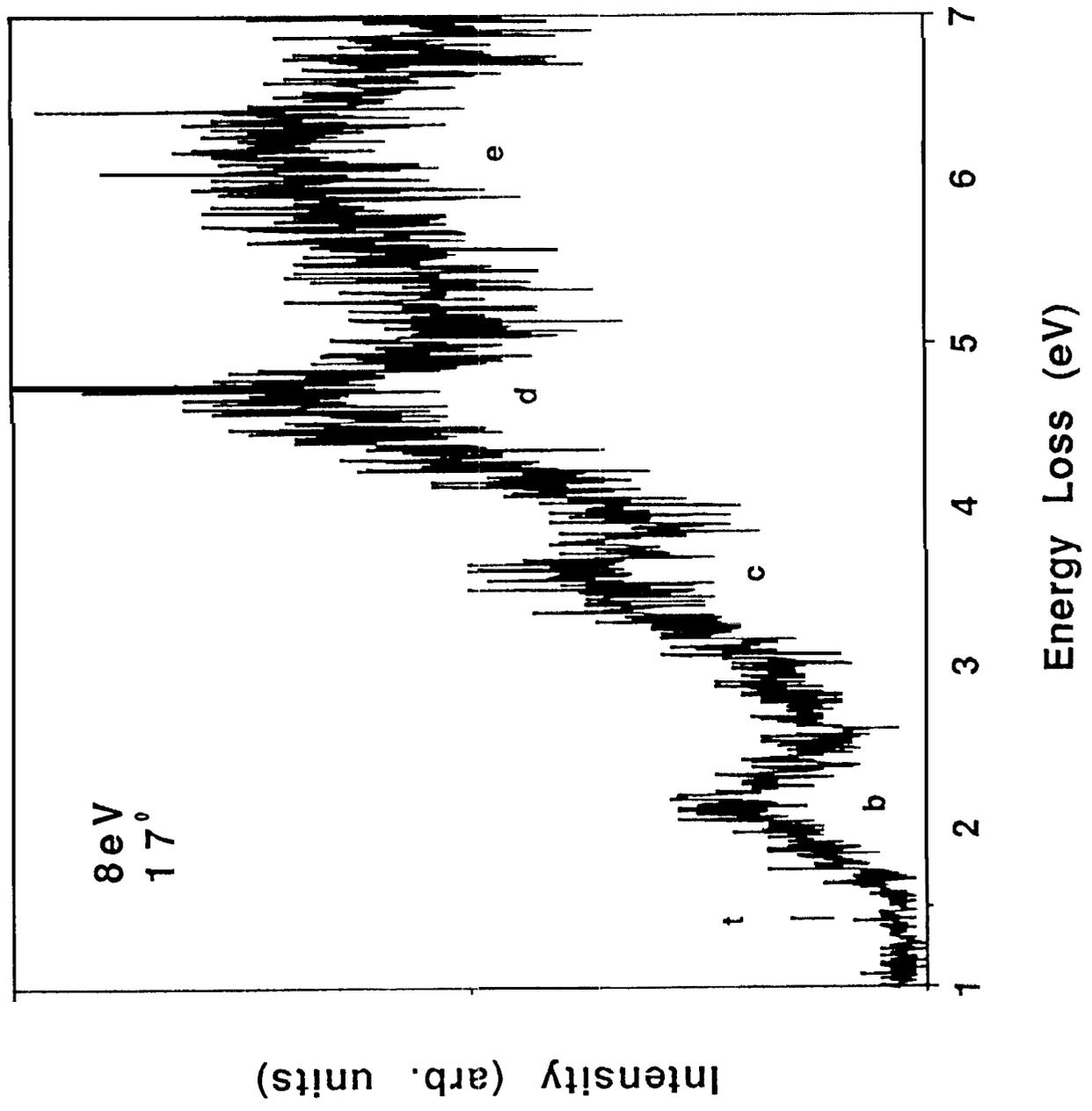


Fig. 4

Fig. 5



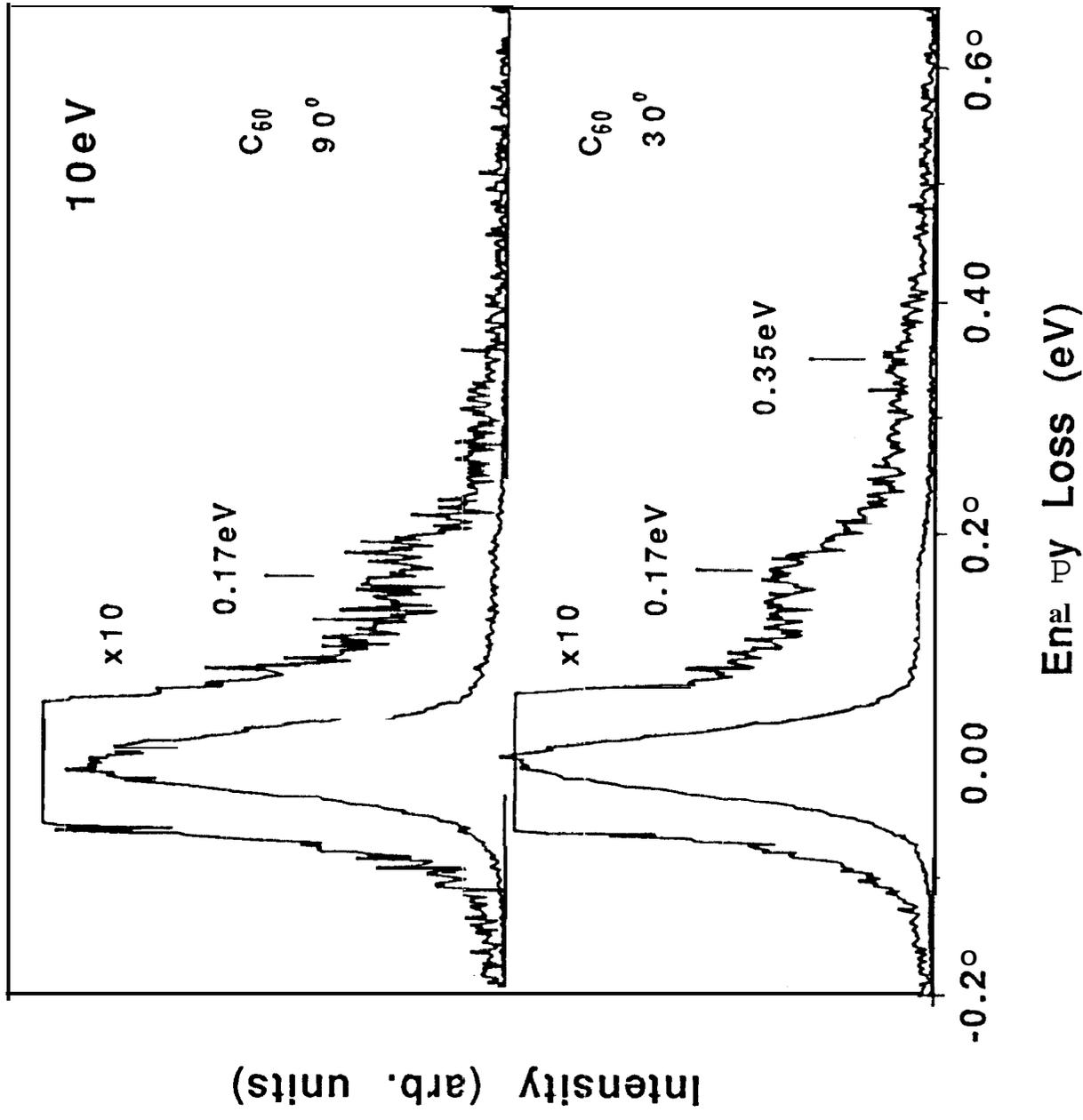
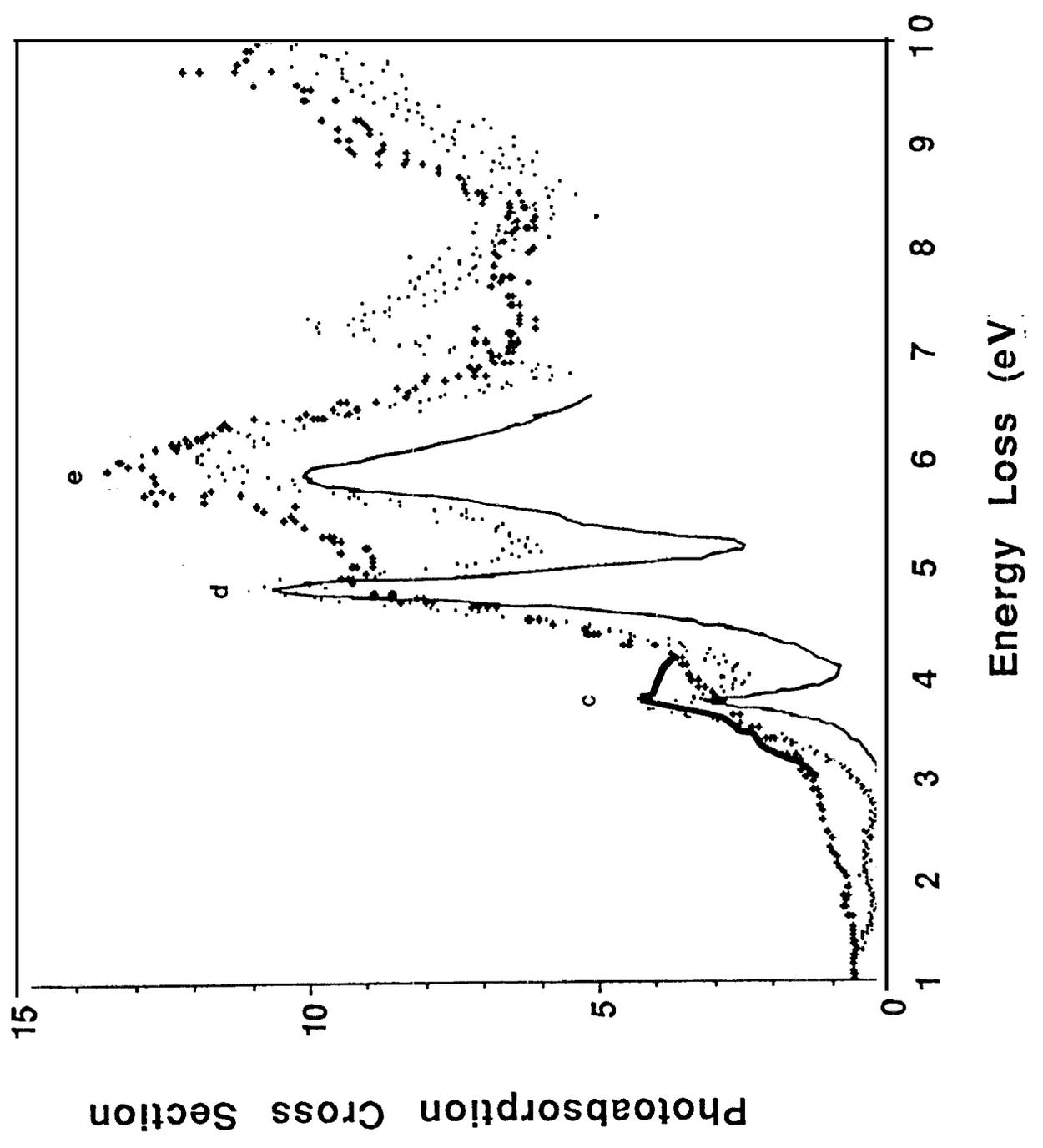


Fig. 6

Fig. 7



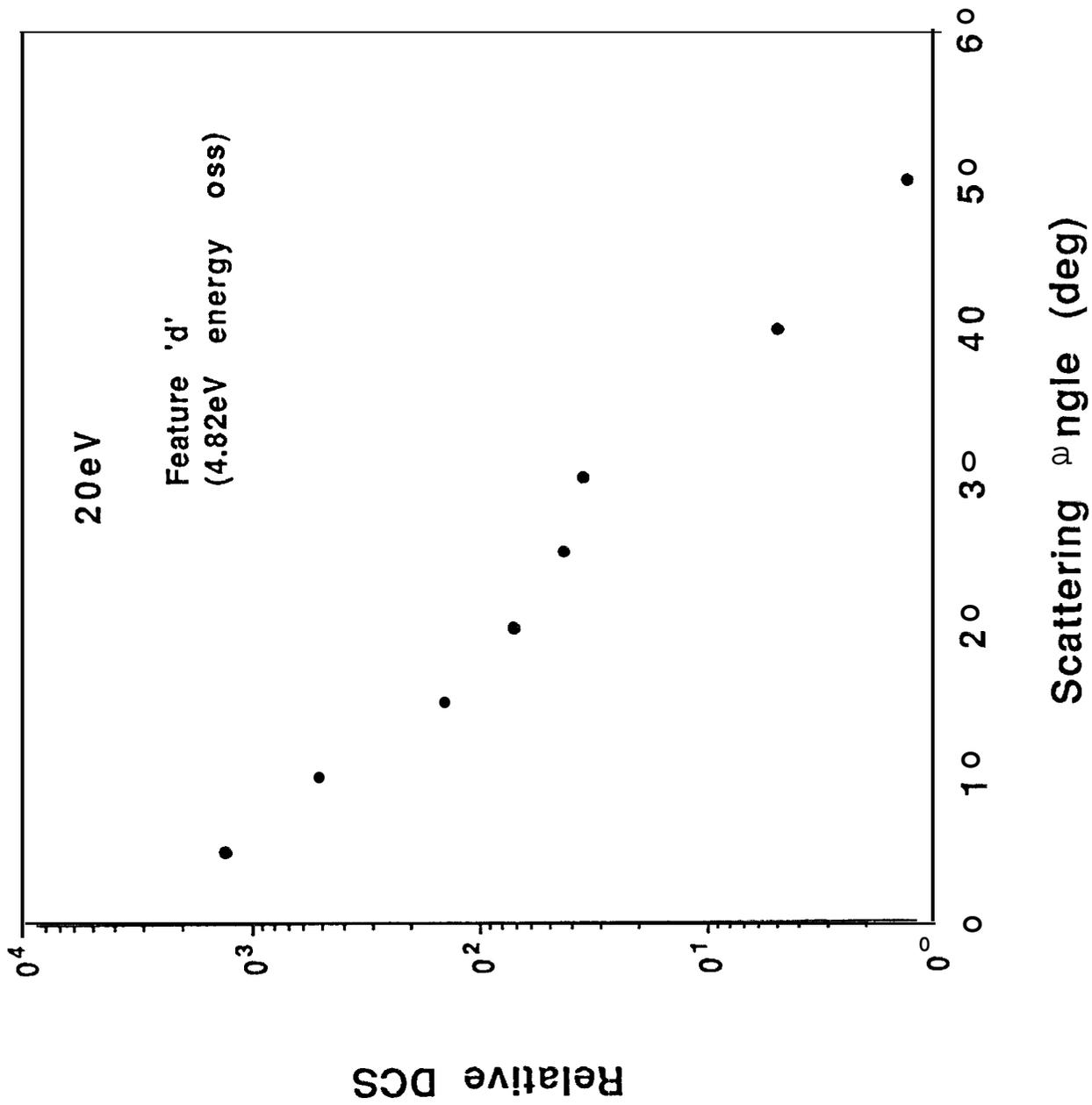


Fig. 8