Proc. Indian natn. Sci. Acad., **46**, A, No. 5, 1980, pp. 448–455.

I. PHYSICS

Electron Microscopy

CONTRAST IN SUPERVOLTAGE ELECTRON MICROSCOPY IN BRIGHT-FIELD IMAGING

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(*Received 23 February 1980; after revision 19 April 1980*)

The variation of the total scattering cross-section with the accelerating voltage of the electron beam in the range 100 KV to 1 MV has been given for various aperture angles of the objective lens and for amorphous films of five different elements giving rise to the scattering. The calculations are based on the single scattering theory of Lenz. A method has been described for the estimation of contrast in supervoltage microscopy of thin films of any element at any specified voltage and aperture angle, using the bright-field mode of operation. Conversely, the method can also be utilised for deducing the cross-section from the measured contrast of the electron microscopic image. A comparison has been made between the theoretically estimated cross-sections and those deduced experimentally.

Keywords : Supervoltage Microscopy; Electron Scattering; Image Contrast.

INTRODUCTION

IN the past few years, there has been a pronounced tendency to use higher operating voltages in electron microscopy, beyond the conventional 100 KV. Although the maximum operating voltage of the electron microscope attained so far is about 3 MV, the majority of these supervoltage microscopes are restricted to 1 MV (Fisher & Imura, 1978). With the ever increasing use of such instruments, it has become necessary to explore the characteristics of the scattered electron intensity and contrast in the supervoltage region, compared to that at 100 KV. This paper reports the variation of the total scattering cross-section based on Lenz single scattering theory (Lenz, 1954), for five different elements with atomic number varying from 6 to 79. A method has been described by which the expected contrast in the image of thin amorphous films of thickness (t) of any element may be estimated in bright-field electron microscopy, with electron accelerating voltages (ϕ) ranging from 100 KV to 1 MV and widely different aperture angles (α) varying between 0 and 30 mrad.

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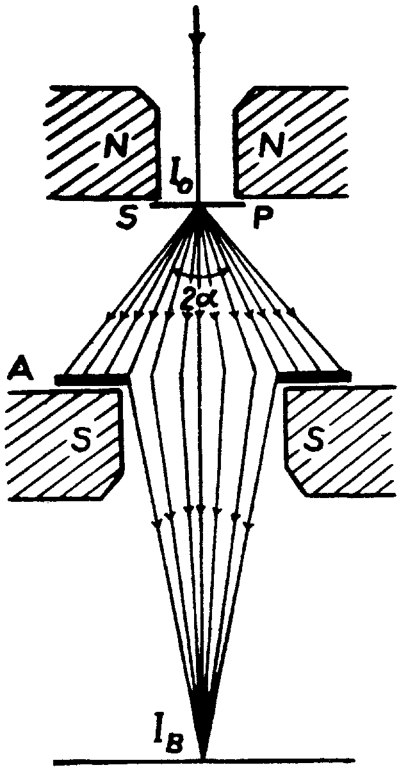

FIG. 1. Schematic diagram of an electron lens for bright-field microscopy showing the positions of the specimen and the aperture.

THEORETICAL CONSIDERATIONS

Fig. 1 is a schematic representation of an electron lens where an aperture A subtends an angle 2α at the specimen SP. A beam of high voltage electrons of intensity I_0 is incident on the specimen. While passing through the specimen these are scattered in all directions. In bright-field electron microscopy, out of these scattered electrons only a small fraction is allowed by the aperture A to form the image. If the intensity on the image plane is I_B, then I_B consists of completely unscattered electrons as well as of those which have suffered neither elastic nor inelastic collision beyond the angle α. The fractional transmission (T) is given by

$$T = I_B/I_0 = \exp\left[-(N_a/A)\rho t(\sigma_e + \sigma_i)\right] = \exp(-nt\sigma) \qquad \text{...(1)}$$

where σ_e, σ_i and σ refer to elastic, inelastic and total scattering cross-sections (cm^2/atom) for scattering between α and π. The quantity $n = (N_a/A)\rho$ signifies the number of atoms/cm^3 of the specimen. N_a is the Avogadro's number. A, ρ and t are the atomic weight, density (gm/cm^3) and thickness (cm) of the specimen respectively.

In actual experimental work, a specimen composed of an element (X) is placed over a thin carbon substrate. In this case one has to compare the relative darkening in the images of the substrate alone and of the specimen plus substrate. If $I_c - I_{xc}$ denote the intensities transmitted through the carbon substrate and substrate plus specimen respectively, it is easy to show that the contrast is given by

$$g = (I_c - I_{xc})/I_c = 1 - \exp(-nt\sigma)_X$$
$$\simeq (nt\sigma)_X \qquad \ldots(2)$$

This will be true when $t << (1/n\sigma)$.
If n is expressed in 10^{20} per cm^3, σ in 10^{-20} cm^2 per atom, t in AU and g in per cent, then the percentage contrast per angstrom is given by

$$g/t = (n\sigma)_X \times 10^{-6} \qquad \ldots(3)$$

Total Cross-Section σ for any ϕ, α and Z

Both σ_e and σ_i vary in complicated and different ways with the atomic number Z, the beam voltage ϕ and the aperture angle α. These have been discussed in detail in a previous paper (Bhattacharya & Das Gupta, 1978). The present paper is concerned with the total cross-section σ for any ϕ, α and Z. Table I shows the

Table I

The total scattering cross-sections (σ) of electrons of energy ϕ MV for scattering beyond an aperture angle α mrad. Specimen thin amorphous films of C, Al, Cu, Ag and Au.

ϕ (MV)	α (mrad)	σ in 10^{-20} cm^2 per atom				
		C (6)	Al (13)	Cu (29)	Ag (47)	Au (79)
0.1	0	359.2	619.7	1118	1734	2916
	1	193.1	367.4	807.2	1367	2501
	5	118.2	268.1	673.8	1208	2308
	10	82.51	214.1	592.0	1103	2170
	20	44.68	140.6	452.7	903.2	1879
	30	26.24	92.80	335.5	711.4	1565
0.3	0	193.3	327.5	581.4	893.2	1488
	1	81.88	164.5	377.9	653.0	1213
	5	42.82	109.6	299.7	556.3	1091
	10	23.89	73.74	233.6	462.5	955.8
	20	9.23	34.49	132.9	292.7	669.3
	30	4.60	18.36	78.06	183.3	450.0
0.5	0	162.5	272.9	480.7	734.9	1219
	1	60.21	125.2	295.2	516.1	966.5
	5	27.44	76.18	221.3	421.3	842.6
	10	12.95	44.03	152.7	316.8	682.1
	20	4.29	17.03	70.91	164.7	398.8
	30	2.04	8.45	37.61	91.84	236.5
0.75	0	149.3	248.9	435.9	663.7	1096
	1	48.94	105.3	254.6	449.5	848.6
	5	18.92	56.90	176.1	345.1	706.5
	10	7.66	28.05	105.5	229.0	515.4
	20	2.30	9.44	41.31	99.51	251.9
	30	1.06	4.49	20.56	51.34	136.3
1.0	0	144.2	239.3	417.1	633.0	1043
	1	42.83	94.78	234.0	416.7	791.8
	5	14.16	45.33	148.4	298.3	625.2
	10	5.15	19.71	78.38	176.2	411.4
	20	1.46	6.11	27.46	67.55	175.8
	30	0.667	2.84	13.20	33.35	90.03

estimated values of σ of electrons of energy ϕ MV, for scattering beyond an aperture angle α mrad, while passing through films of C, Al, Cu, Ag and Au. The computations are based on the single scattering theory of Lenz (1954). It will be seen from this table that the total cross-section varies widely with Z, α and ϕ. For α ranging between 1 and 30 mrad and ϕ from 0.1 to 1 MV σ changes for C from 0.67 to 193×10^{-20} cm^2, whereas for Au the corresponding variation ranges between 90 and 2500×10^{-20} cm^2. For other elements the variation is within this range.

Figs. 2 and 3 show the variation of σ with α for the two extreme elements C(6) and Au(79) for five discrete values of ϕ. For any value of ϕ there is a rapid decrease in σ from 0 to 1 millirad. Thereafter the decrease is slowed down. The difference in σ between 0.1 and 0.3 MV is large, while this is reduced as ϕ increases further. This is true for all the elements. In the case of C, σ decreases smoothly with α for all values of ϕ. The smooth variation of σ with α continues upto the element Cu, but thereafter the variation is not smooth. For Au, at 0.1 MV σ decreases almost linearly with α between 3 and 30 mrads; but for higher voltages the decrease is somewhat irregular.

Experimental work on the measurement of scattering cross-sections in supervoltage microscopy has been carried out in different laboratories of the world with different specific values of α, ϕ and Z (say α_1, ϕ_1 and Z_1). In order to compare such experimental data with the theoretical estimates given in this paper, one has therefore to find out the theoretical value of $\sigma(\alpha_1, \phi_1, Z_1)$. The following procedure may be used for this purpose.

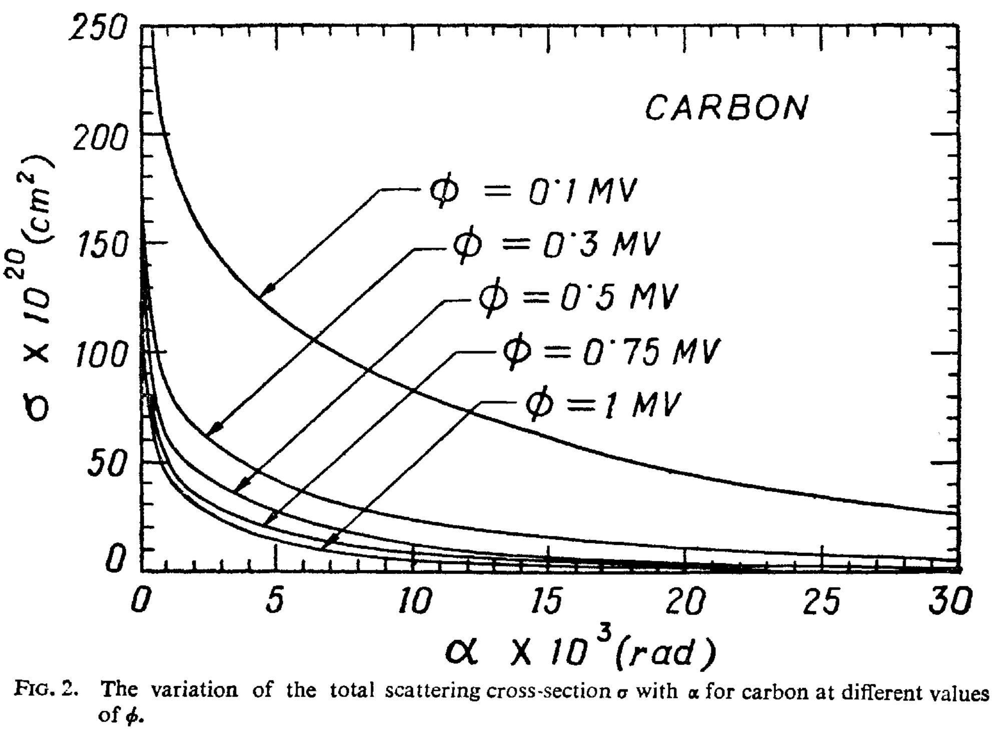

FIG. 2. The variation of the total scattering cross-section σ with α for carbon at different values of ϕ.

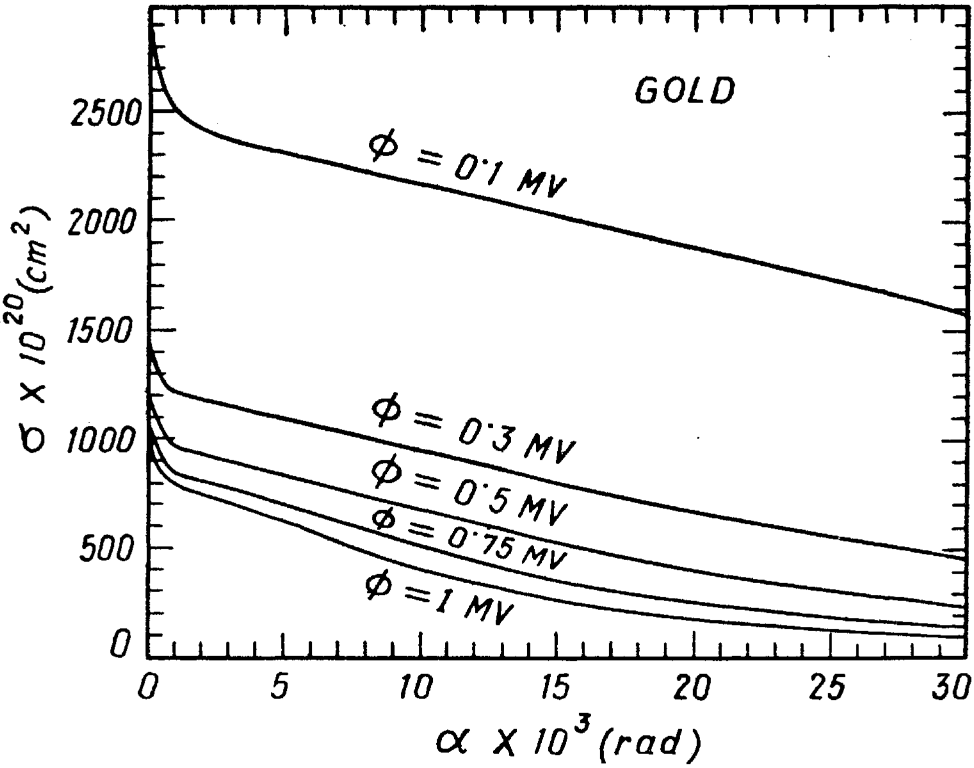

FIG. 3. The variation of total scattering cross-section σ with α for gold at different values of ϕ.

With the help of Table I, one can prepare curves similar to Figs. 2 and 3 for the other three elements. From these curves one can find σ for α_1 for the five given voltages and the five elements. A graph may now be drawn for $\sigma(\alpha_1)$ as a function of ϕ for the five elements. This plot gives $\sigma(\alpha_1, \phi_1)$ for the five widely different values of Z. From a plot of $\sigma(\alpha_1, \phi_1)$ as a function of Z, one deduces immediately $\sigma(\alpha_1, \phi_1, Z_1)$.

CONTRAST AND SPECIMEN THICKNESS

The contrast (g) per unit thickness of an amorphous film of an element is given by the product of the number of atoms/cm^3 (n) of the element and the corresponding cross-section σ [eqn. (2)]. The values of n for the ten elements commonly used in electron microscopy are given in Table II. In Fig. 4 the contrast per unit thickness (g/t) has been plotted as a function of σ for different elements. With the help of this figure one can deduce the expected contrast for a given thickness when σ is known. It will be seen from Table I that the extreme values of σ for different elements range from 0.67 to 2916 $\times$ 10^{-20} cm^2/atom. In order to deduce g/t accurately from Fig. 4 for low values of σ, one may reduce both the abscissa and the ordinate scales by the same factor and read from the modified scales. Conversely,

TABLE II

Number of atoms/cm³ (n) for different elements, in units of 10^{20} *cm*$^{-3}$

C (6)	Cr (24)	Cu (29)	Ge (32)	Pd (46)	Ag (47)	W (74)	Pt (78)	Au (79)	U (92)
1137	833.3	846.4	439.7	679.2	586.2	632.2	662.5	589.5	482.0

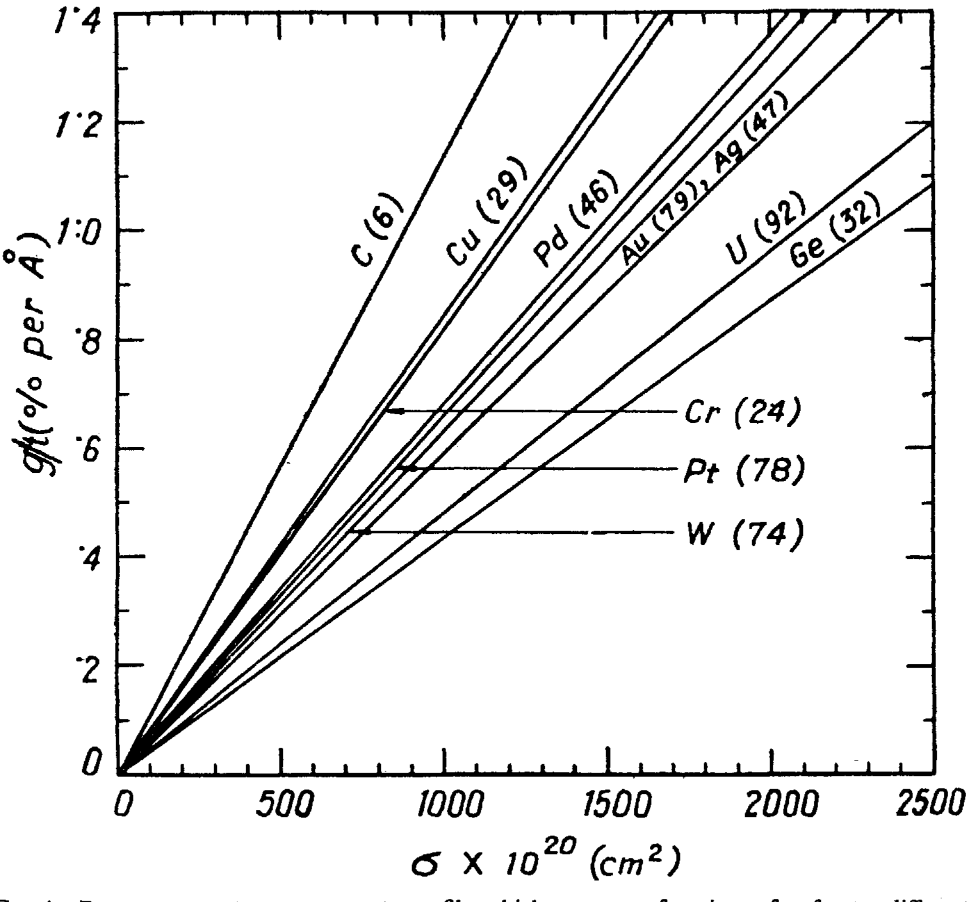

FIG. 4. Percentage contrast per angstrom film thickness as a function of σ for ten different elements.

if the contrast is measured one can immediately deduce the corresponding experimental value of the scattering cross-section.

As an example of the use of this curve the expected contrast for a gold film 5Å thick at 0.1 and 1 MV with $\alpha = 10$ mrad will be estimated. The cross-sections for Au for the above values of α and ϕ are 2170 and 411 $\times$ 10^{-20} cm² (Table I). Corresponding to these values of σ, the expected contrast for the Au-film will be 6.4 per cent and 1.2 per cent from Fig. 4. Under similar conditions of α and ϕ, the expected contrast for a 20Å diameter DNA molecule will be approximately 2 per cent and 0.1 per cent respectively, assuming that it consists entirely of carbon atoms. The molecule will thus be invisible. If the same molecule is shadowed with Au, it will be easily visible at 0.1 MV.

A few experimental values of σ in supervoltage microscopy will now be compared with the theoretical calculations given in the previous section. Arnal *et al.* (1977) and Martinez *et al.* (1979) deduced the experimental cross-section from a direct measurement of transmission (T) [eqn. (1)] with a Faraday cage. They plotted $\ln (1/T)$ against t and deduced the single scattering cross-section from the slope of the curve at $t \to 0$. On the other hand, some authors determined transmission for one or two definite values of thickness. σ obtained under such conditions may not exactly agree with the cross-section deduced from single scattering theory.

An idea of the order of agreement between the experimental and the theoretical cross-sections given in this paper may be obtained from Table III. It will be seen that there is a fair amount of agreement between the theoretical and experimental cross-sections in supervoltage microscopy. The better agreement between the theoretical and the experimental results at higher voltages may be explained as follows. The theoretical cross-sections given in this paper were deduced by using Born's approximation. This becomes more valid as the electron energy increases.

TABLE III

Comparison of experimental and theoretical cross-sections for gold. The cross-sections are in units of 10^{-20} cm^2

Author	α (mrad)	ϕ (MV)	σ(expt.)	σ(theo.)
Curtis *et al.* (1968)	1	0.3	944	1213
		0.5	782	967
		0.6	751	911
	5	0.3	839	1091
		0.5	700	843
		0.6	662	777
Arnal *et al.* (1977)	20	0.3	887	669
		0.5	436	399
		0.75	259	252
		1.0	179	176
	30	0.3	471	450
		0.5	231	237
		0.75	138	136
		1.0	95	90

No electron microscope operating at voltages above 100 KV is at present available in India; one has therefore to depend exclusively on foreign laboratories for experimental data in this field.

ACKNOWLEDGEMENTS

The authors are indebted to the University Grants Commission, New Delhi, for sponsoring this work. Thanks are also due to Professor V. E. Cosslett, F. R. S., Cavendish Laboratory, Cambridge, U. K., for some valuable comments.

References

Arnal, F., Balladore, J. L., Soum, G., and Verdier, P. (1977) Calculation of the cross sections of electron interaction with matter. *Ultramicroscopy*, **2**, 305–310.

Bhattacharya, D. K., and Das Gupta, N. N. (1978) Electron scattering in high voltage electron microscopy. *Ultramicroscopy*, **3**, 29–37.

Curtis, G. H., Cosslett, V. E., and Ferrier, R. P. (1968) Electron transmission through carbon and gold films at high voltages. *Proc. 4th European Regional Conf. Electron Microscopy, Rome*, **1**, 61–62.

Fisher, R. M., and Imura, T. (1978) New applications and extensions of the unique advantages of HVEM for physical and materials research. *Ultramicroscopy*, **3**, 3–18.

Lenz, F. (1954) Scattering of medium energy electrons at very small angles. *Z. Naturforsch.*, **9a**, 185–204.

Martinez, J. P., Balladore, J. L., and Trinquier, J. (1979) Determination of partial and total scattering cross-sections for high energy electrons in the range 1.2–3 MeV. *Ultramicroscopy*, **4**, 211–219.