

Scattering and Diffraction

CHAPTER PREVIEW

The electron is a low-mass, negatively charged particle. As such, it can easily be deflected by passing close to other electrons or the positive nucleus of an atom. These Coulomb (electrostatic) interactions cause electron scattering, which is the process that makes TEM feasible. We will also discuss how the wave nature of the electron gives rise to diffraction effects. What we can already say is that if the electrons weren't scattered, there would be no mechanism to create TEM images or DPs and no source of spectroscopic data. So it is essential to understand both the particle approach and the wave approach to electron scattering in order to be able to interpret all the information that comes from a TEM. Electron scattering from materials is a reasonably complex area of physics, but it isn't necessary to develop a detailed comprehension of scattering theory to be a competent microscopist.

We start by defining some terminology that recurs throughout the book and then we introduce a few fundamental ideas that have to be grasped. These fundamental ideas can be summarized in the answers to four questions.

- What is the probability that an electron will be scattered when it passes near an atom?
- If the electron is scattered, what is the angle through which it is deviated?
- What is the average distance an electron travels between scattering events?
- Does the scattering event cause the electron to lose energy or not?

The answer to the first question concerning the probability of scattering is embodied in the idea of a cross section. The angle of scattering (usually determined through the differential cross section) is also important because it allows you as the TEM operator to control which electrons form the image and therefore what information is contained in the image. We will develop this point much further when we talk about image contrast in Part 3 of the book. The third question requires defining the mean-free path, an important concept given that we use thin specimens. The answer to the fourth question requires distinguishing elastic and inelastic scattering. The former constitutes most of the useful information in DPs obtained in the TEM, discussed in Part 2, while the latter is the source of X-rays and other spectroscopic signals discussed in Part 4. The distinction between electrons that lose energy and those that don't is important enough that we devote the next two chapters to each kind of electron and expand on the basic ideas introduced here.

2.1 WHY ARE WE INTERESTED IN ELECTRON SCATTERING?

We need to know about electron scattering because it is fundamental to all electron microscopy (not just TEM). You know well that your eyes cannot see any object unless it interacts with visible light in some way, for example through reflection or refraction, which are two forms of scattering (e.g., we can't see a light beam unless it is scattered by dust within it or it hits a surface). Similarly, we cannot see anything in EM images unless

the specimen interacts with and scatters the electrons in some way. Thus, any non-scattering object is invisible and we will come across situations where 'invisibility' is an important criterion in TEM images. In the TEM we are usually most interested in those electrons that do not deviate far from the incident-electron direction. This is because the TEM is constructed to gather these electrons primarily and they also give us the information we seek about the internal structure and chemistry of the specimen. Other forms of scattering, such as electrons which are scattered through large angles (e.g., backscattered

electrons) and electrons ejected from the specimen (such as secondary electrons) are also of interest and we will not totally neglect them (although they are of much greater interest in the SEM where they provide atomic number contrast and surface-sensitive, topographical images, respectively).

WAVE AND PARTICLE

The electron is treated in two different ways throughout this book: in electron scattering it is a succession of particles, while in electron diffraction it is treated by wave theory. The analogy to X-rays or visible light would be to compare a beam of photons and an electromagnetic wave. However, you must always remember that electrons are charged particles and that Coulomb forces are very strong.

In this chapter we introduce the fundamental ideas of electron scattering; then, in the next two chapters, we discuss the two principal forms of scattering, namely, elastic and inelastic. Both forms are useful to us, but you'll see that the latter has the unfortunate side effect of being responsible for specimen damage and ultimately limits what we can do with a TEM.

To give you some feel for the importance of electron scattering, it is worth illustrating at this stage the basic principles of the TEM. You will see in due course that in a TEM we illuminate a thin specimen with a broad beam of electrons in which the intensity is uniform over the illuminated area.

We will often refer to incident and scattered electrons as beams of electrons, because we are dealing with many electrons, not an individual electron; these electrons are usually confined to well-defined paths in the microscope. So the electrons that hit the specimen are often called the incident beam and those scattered by the specimen are called scattered (or sometimes specifically, diffracted) beams. Electrons coming through a thin specimen are separated into those that suffer no angular deviation and those scattered through measurable angles. We call the undeviated electrons the 'direct beam' (in contrast to most texts that describe this as the 'transmitted beam' despite the fact that all electrons coming through the specimen have been 'transmitted'). As the electrons travel through the specimen they are either scattered by a variety of processes or they may remain unaffected. The end result, however, is that a non-uniform distribution of electrons emerges from the exit surface of the specimen, as shown schematically in Figure 2.1. It is this non-uniform distribution that contains all the structural, chemical, and other information about our specimen. So everything we learn about our specimen using TEM can be attributed to some form of electron scattering.

DIRECT BEAM

The beam that comes through the specimen, but remains parallel to the direction of the incident electrons is a very important beam, which we will term the **direct beam**.

We'll see in Chapter 9 that the TEM is constructed to display this non-uniform distribution of electrons in two different ways. First the *spatial distribution* (Figure 2.1A) of scattering can be observed as contrast in *images* of the specimen, and the *angular distribution* of scattering (Figure 2.1B) can be viewed in the form of scattering patterns, usually called *diffraction patterns*. A simple (and fundamental) operational step in the TEM is to use a restricting aperture, or an electron detector, of a size such that it only selects electrons that have suffered more or less than a certain angular deviation. So you as the operator have the ability to choose which electrons you want to use and thus you control what information

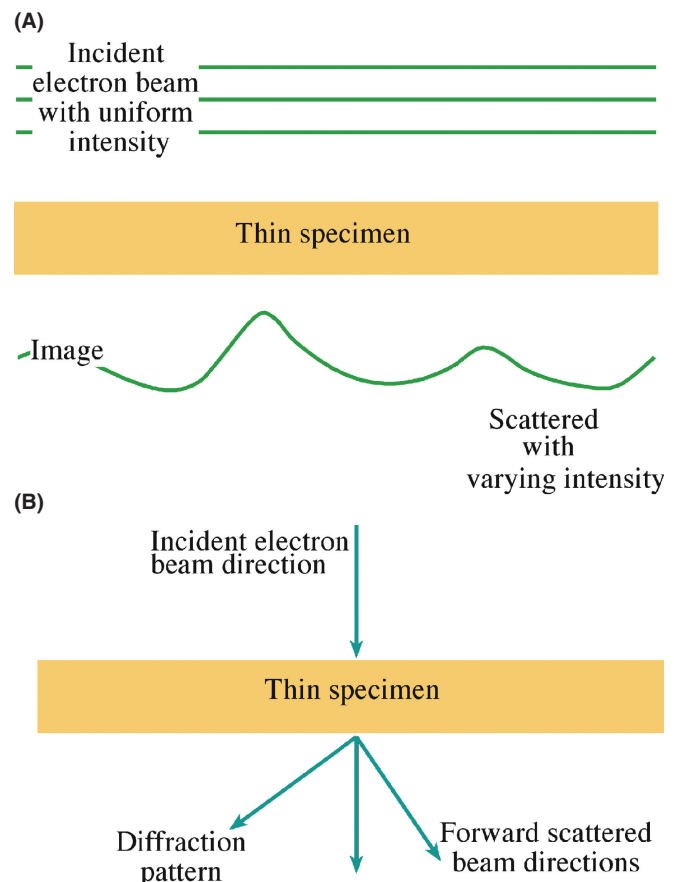


FIGURE 2.1. (A) A uniform intensity of electrons, represented by the horizontal lines, falls on a thin specimen. Scattering within the specimen changes both the spatial and angular distributions of the emerging electrons. The spatial distribution (intensity) is indicated by the wavy line. (B) The change in angular distribution is shown by an incident beam of electrons being transformed into several forward-scattered beams.

will be present in the image. Therefore, to comprehend these images, you have to understand what causes electrons to scatter in the first place. The same is true for DPs since you can also control (to a lesser extent) the angular-scattering distribution, e.g., by tilting your specimen.

We devote the whole of Part 2 to diffraction phenomena and Part 3 to images. Lastly, Part 4 deals with ways in which we use inelastic scattering for analytical electron microscopy (AEM) to study, e.g., the chemistry and the bonding of the atoms in our specimen.

2.2 TERMINOLOGY OF SCATTERING AND DIFFRACTION

Electron-scattering phenomena can be grouped in different ways. We've already used the most important terms: *elastic* and *inelastic* scattering. These terms, respectively, describe scattering that results in no loss of energy or in some measurable loss of energy (usually very small with respect to the beam energy). In either case, we can consider the beam electrons and specimen atoms as particles, and scattering of the incident electrons by the atoms in the specimen can often be approximated to something like billiard balls colliding. The billiard-ball analogy will be good through Section 2.7 after which we'll be talking about waves.

ELECTRON SCATTERING

This theme permeates the whole text and connects ALL aspects of TEM.

However, we can also separate scattered electrons into *coherent* and *incoherent*, which refers, of course, to their wave nature. These distinctions are related since elastically scattered electrons are usually coherent and inelastic electrons are usually incoherent (note the modifier 'usually'). Let's assume that the incident electron waves are coherent, that is, they are essentially in step (in phase) with one another and of a fixed wavelength, governed by the accelerating voltage. You'll see that this isn't a bad assumption in most circumstances. Then, coherently scattered electrons are those that remain in step and incoherently scattered electrons have no phase relationship, after interacting with the specimen.

The nature of the scattering can result in different angular distributions. Scattering can be either *forward scattering* or *back scattering* (usually written as one word) wherein the terms refer to the angle of scattering with respect to the incident beam and a specimen that is normal to that beam. (Note: you will sometimes see the

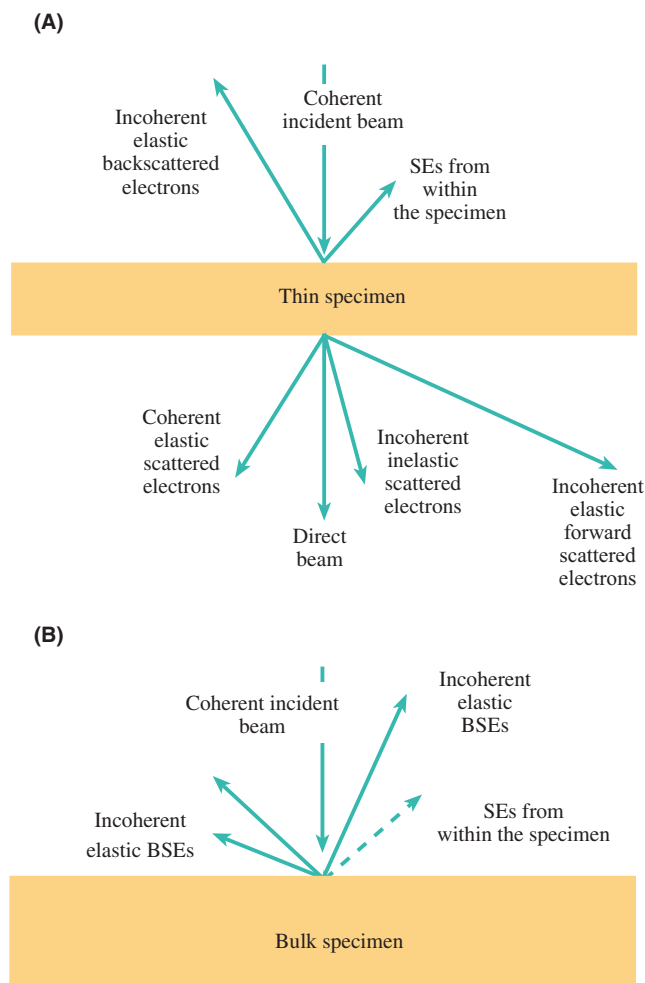


FIGURE 2.2. Different kinds of electron scattering from (A) a thin specimen and (B) a bulk specimen: a thin specimen permits electrons to be scattered in both the forward and back directions while a bulk specimen only backscatters the incident-beam electrons.

term 'forward scattering' used in another sense.) If an electron is scattered through $< 90^\circ$, then it is forward scattered and $> 90^\circ$ it is backscattered. These various terms are related by the following general principles, summarized in Figure 2.2.

- Elastic scattering is usually coherent, if the specimen is thin and crystalline (think in terms of waves).
- Elastic scattering usually occurs at relatively low angles ($1\text{--}10^\circ$), i.e., it is strongly peaked in the forward direction (waves).
- At higher angles ($> \sim 10^\circ$) elastic scattering becomes more incoherent (now think of particles).
- Inelastic scattering is almost always incoherent and is very low angle ($< 1^\circ$) scattering (think particles).
- As the specimen gets thicker, fewer electrons are forward scattered and more are backscattered. Incoherent, backscattered electrons are the only remnants of the incident beam that emerge from bulk, non-transparent specimens (think particles).

The notion that electrons can be scattered through different angles is related to the fact that an electron can also be scattered more than once. Generally, the more scattering events, the greater the angle of scattering (although sometimes a second scattering event can redirect the electron back into the direct beam, so it appears to have undergone no scattering).

The simplest scattering process is *single scattering* and we often approximate all scattering within a TEM specimen to this process (i.e., an electron either undergoes a single-scattering event or it suffers no scattering). We'll see that this can be a very reasonable assumption if the specimen is very thin (something you can control). If the electron is scattered more than once, we use the term *plural scattering* and if it is scattered >20 times, we say *multiple scattering*. It is generally safe to assume that, unless you have a particularly thick specimen (through which you probably can't see anything anyhow), multiple scattering will not occur in the TEM. The greater the number of scattering events, the more difficult it is to predict what will happen to the electron and the more difficult it is to interpret the images, DPs, and spectra that we gather. So, once again, we emphasize the importance of the 'thinner is better' criterion, i.e., if you create thin enough specimens so that the single-scattering assumption is plausible, your TEM research will be easier.

Diffraction is a very special form of elastic scattering and the terminology used can be confusing. Collins' Dictionary defines *diffraction* as 'a deviation in the direction of a wave at the edge of an obstacle in its path' while *scattering* is defined as 'the process in which particles, atoms, etc., are deflected as a result of collision.' The word scatter can also be a noun denoting the act of scattering. So scattering might best apply to particles and diffraction to waves; both terms thus apply to electrons! You should also note that the term diffraction is not limited to Bragg diffraction which we'll emphasize in TEM; it refers to *any* interaction involving a wave, but many texts are not consistent in this respect.

DEFINE DIFFRACTION

An interaction between a wave of any kind and an object of any kind (Taylor 1987).

In the TEM we utilize the electrons that go through a specimen; it is important to note that such electrons are not simply 'transmitted' in the sense of visible light through window glass. Electrons are scattered mainly in the forward direction, i.e., parallel to the incident beam direction (and we've already noted the confusion between 'direct' and 'transmitted'). We'll tell you in a short while what fraction of the electrons are forward scattered and how this varies with the thickness of the specimen and atomic number of the 'target' atom. This

scattering is a direct consequence of the fact that there is such a strong interaction between electrons and matter.

Forward scattering includes the direct beam, most elastic scattering, diffraction, particularly Bragg diffraction (see Chapter 3), refraction, and inelastic scattering (see Chapter 4). Because of forward scattering through our thin specimen, we see a DP or an image on the viewing screen, and detect an X-ray spectrum or an electron energy-loss spectrum outside the TEM column. But don't neglect backscattering; it is an important imaging mode in the SEM.

FORWARD SCATTERING

The cause of most of the signals used in the TEM.

When physicists consider the theory of electron interactions within a solid, they usually consider scattering of electrons by a single, isolated atom, then progress to agglomerations of atoms, first in amorphous solids and then in crystalline solids and we'll follow a similar path.

2.3 THE ANGLE OF SCATTERING

When an electron encounters a single, isolated atom it can be scattered in several ways which we will cover in the next two chapters. For the time being, let's imagine simply that, as shown in Figure 2.3, the electron is scattered through an angle θ (radians) into some solid angle ω , measured in steradians (sr). We have to define this angle first because you'll see that it plays an important role in the subsequent discussion of cross sections.

SEMI-ANGLE

Note that the scattering angle θ is in fact a semi-angle, not a total angle of scattering. Henceforth, whenever we say "scattering angle" we mean "scattering semi-angle."

Often we assume that θ is small enough such that $\sin \theta \approx \tan \theta \approx \theta$. When θ is this small, it is often convenient to use milliradians or mrad; 1 mrad is 0.0573° , 10 mrad is $\sim 0.5^\circ$.

SMALL ANGLE

A convenient upper limit is <10 mrad.

The characteristics of the scattering event are controlled by factors such as the incident-electron energy and the atomic number/weight of the scattering atom. When we consider a specimen rather than a single

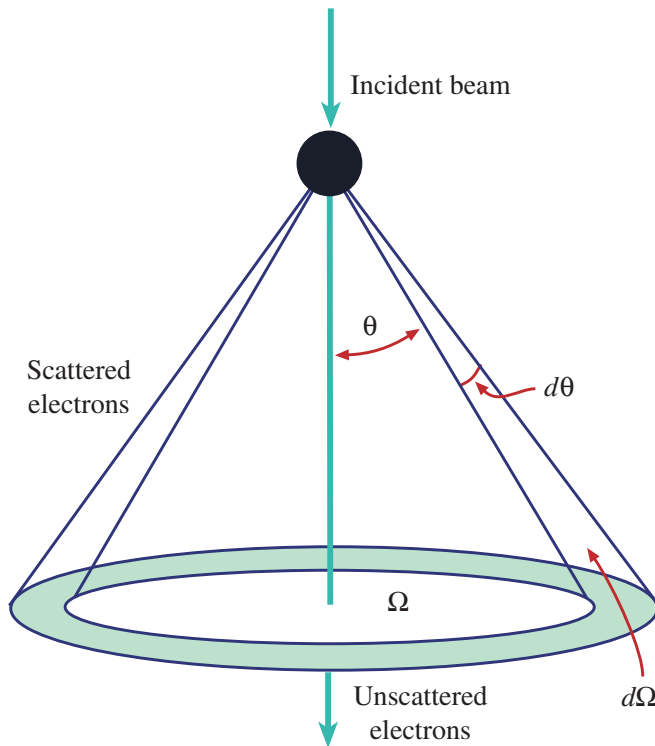


FIGURE 2.3. Electron scattering by a single isolated atom. The electrons are scattered through an angle θ and the total solid angle of scattering is Ω . An incremental increase in scattering angle $d\theta$ gives an incremental increase in a solid angle $d\Omega$, which is the basis for determining the differential scattering cross section.

atom, factors such as the thickness, density, crystallinity, and angle of the specimen to the incident beam also become important. To understand these variables, we need to examine the physics of scattering in more detail. Of necessity, we'll be rather brief and often imprecise since we're trying to condense much of Mott and Massey's substantial and classic textbook into just a few pages.

2.4 THE INTERACTION CROSS SECTION AND ITS DIFFERENTIAL

The chance of a particular electron undergoing any kind of interaction with an atom is determined by an interaction *cross section*. The concept of a cross section is well described by the following analogy given by Rudolf Peierls (Rhodes 1986)

"If I throw a ball at a glass window one square foot in area, there may be one chance in ten that the window will break and nine chances in ten that the ball will just bounce. In the physicist's language this particular window, for a ball thrown in this particular way, has a disintegration (inelastic!)

cross section of 0.1 square feet and an elastic cross section of 0.9 square feet."

So each possible interaction has a different cross section which depends on the energy of the particle, in our case the beam energy. The cross section (for which we'll use the Greek letter σ) has units of area (not square feet as used in Peierls' analogy, but a tiny fraction of the area of an atom termed a 'barn'). One barn is 10^{-28} m^2 (that's $(10^{-5} \text{ nm})^2$) and the name arises because of the perverse sense of humor of some of the early atomic physicists who considered that this unimaginably small area is 'as big as a barn door.' The cross section does *not* represent a physical area but, when divided by the actual area of the atom, it represents a *probability* that a scattering event will occur.

2.4.A Scattering from an Isolated Atom

First of all we'll consider the scattering cross section for a single isolated atom, then extend the concept to a specimen with many atoms. We'll use a generalized form to start with in this chapter and then break down the concept of a total cross section into cross sections for individual processes such as elastic scattering and the various inelastic processes in the next two chapters.

SCATTERING PROBABILITY

The larger the cross section, the better the chances of scattering.

Following Heidenreich (1964), we can define the cross section (an area) in terms of the *effective radius* of a single, isolated atom, r

$$\sigma_{\text{atom}} = \pi r^2 \quad (2.1)$$

where r has a different value for each scattering process as we'll see in the next chapter. What interests us in the TEM is whether or not the scattering process deviates the incident-beam electrons outside a particular scattering angle θ such that, e.g., they do not go through the aperture in the lens or they miss the electron detector. So we have to know the *differential cross section* ($d\sigma/d\Omega$) which describes the angular distribution of scattering from an atom. As shown in Figure 2.3 electrons are scattered through an angle θ into a solid angle Ω and there is a simple geometrical relationship between θ and Ω

$$\Omega = 2\pi(1 - \cos \theta) \quad (2.2)$$

and therefore

$$d\Omega = 2\pi \sin \theta d\theta \quad (2.3)$$

So the differential scattering cross section for a single, isolated atom can be written as

$$\frac{d\sigma}{d\Omega} = \frac{1}{2\pi \sin \theta} \frac{d\sigma}{d\theta} \quad (2.4)$$

Now, we can calculate σ_{atom} for scattering into all angles *greater than* θ by integrating equation 2.4 from θ to π

$$\sigma_{\text{atom}} = \int_{\theta}^{\pi} d\sigma = 2\pi \int_{\theta}^{\pi} \frac{d\sigma}{d\Omega} \sin \theta d\theta \quad (2.5)$$

The limits of the integration are governed by the fact that the values of θ can vary from 0 to π , depending on the specific type of scattering. If we work out the integral we find that σ decreases as θ increases (which makes physical sense). Since $d\sigma/d\Omega$ is often what is measured experimentally (but not in the TEM), equation 2.5 gives us an easy way to determine the cross section for a given atom (σ_{atom}) for all values of θ , by working out the integral of $(d\sigma/d\Omega)\sin \theta$ from 0 to π .

INTEGRATE

If we integrate from 0 to θ then we determine the cross section for scattering into all angles less than θ which is in fact more relevant to the TEM situation.

2.4.B Scattering from the Specimen

So let's move on from the cross section for a single isolated atom (with units of area) and consider that the specimen contains N atoms/unit volume. Therefore, we can define the total cross section for scattering from the specimen (in units of m^{-1}) as

$$\sigma_{\text{total}} = N\sigma_{\text{atom}} \quad (2.6)$$

Since $N = N_0\rho/A$ where N_0 is Avogadro's number (in units of atoms mol^{-1}), A is the atomic weight of the scattering atoms in the specimen (kg mol^{-1}) which has density ρ (kg m^{-3}), we can write

$$\sigma_{\text{total}} = N\sigma_{\text{atom}} = \frac{N_0\sigma_{\text{atom}}\rho}{A} \quad (2.7)$$

Thus, σ_{total} is the number of scattering events per unit distance that the electron travels through the specimen. If the specimen has thickness t , then the probability of scattering from the specimen is given by

$$\sigma_{\text{total}} t = \frac{N_0\sigma_{\text{atom}}(\rho t)}{A} \quad (2.8)$$

Here we've gathered together the product of ρ and t because this is called the mass thickness of the specimen (e.g., doubling ρ produces the same effect as doubling t) and we'll come across this term again when we discuss image contrast and also X-ray absorption. Equation 2.8 is an important expression, since it contains all the variables that affect the scattering probability from a real specimen. We'll use it again when we consider how certain kinds of image contrast arise in the TEM.

So we can now appreciate, through a few (rather simplified) equations, the relationship between the physics of electron scattering and the information we collect in the TEM.

Expressions for the cross section become more complicated as they are modified to give better approximations for the scattering in a real specimen, as we'll see in the next couple of chapters. However, the more complex equations don't alter the basic scattering behavior predicted by the simple equations we've just given.

2.4.C Some Numbers

Because of all the variables that affect σ_{atom} and σ_{total} , it is only possible to give a ballpark (barnyard?) value for the cross section. For TEM electron energies (100–400 keV), the elastic cross section is almost always the dominant component of the total scattering. If you look ahead to Figure 3.3, typical small-angle, elastic cross sections for transition metals bombarded by 100-keV electrons are $\sim 10^{-22} \text{ m}^2$. This is a good number to remember for typical elastic scattering. Inelastic cross sections are generally smaller and range from $\sim 10^{-22} \text{ m}^2$ down to 10^{-26} m^2 depending on the specific type of scattering and the material. Going back to equation 2.1, a typical scattering radius (r) is $\sim 10^{-11} \text{ m}$ or $\sim 0.01 \text{ nm}$, which might seem a bit small (about a tenth of an atomic radius) but, since the scattering is localized to the inner or core shells which are closer to the nucleus or to a particular electron-electron interaction, perhaps this isn't such a bad estimate, given all the caveats.

2.5 THE MEAN FREE PATH

Instead of using an area to describe the interaction we can use a length since the distance an electron travels between interactions with atoms is clearly going to be important when we are using thin specimens. This new parameter is then the average distance that the electron travels between scattering events. This distance is important because, if we know what it is, we can work out how thin we have to make our specimen, so plural scattering is not significant, thus making it easier to interpret our images and spectroscopic data in terms of single-scattering theory. The term σ_{total} can be expressed as the inverse

of the mean free path, λ . Because the dimensions of σ_{total} are m^{-1} (you can check this using equations 2.1 and 2.7) there is a simple expression for the mean free path λ which has units of length

$$\lambda = \frac{1}{\sigma_{\text{total}}} = \frac{A}{N_0 \sigma_{\text{atom}} \rho} \quad (2.9)$$

Typical values of λ for scattering at TEM voltages are of the order of tens of nm, so single-scattering approximations imply specimen thicknesses of this order. It is, unfortunately, conventional to use λ to denote the mean free path; it is *not* the wavelength of the electron. From equation 2.9 we can define a probability of scattering p as the electron travels through a specimen thickness t

$$p = \frac{1}{\lambda} = \frac{N_0 \sigma_{\text{atom}} (\rho t)}{A} \quad (2.10)$$

which is $(\sigma_{\text{total}} t)$ from equation 2.8.

Although computational resources are constantly improving, our knowledge of the values of σ , λ , and θ is imprecise at best, particularly at the 100–400 keV beam energies used in TEMs. Cross sections and mean free paths for particular scattering events may only be known within a factor of two, but we can often measure θ very precisely in the TEM. We can combine all our knowledge of scattering in Monte Carlo simulations to predict the electron paths as a beam is scattered through a thin foil.

Monte Carlo Simulation

So called because random numbers are used in the computer programs; the outcome is always predicted by statistics!

The first Monte Carlo calculation was developed by two of the United States' foremost mathematicians, John von Neumann and Stanley Ulam, at Los Alamos in the late 1940s. Ulam actually rolled dice and made hand (!) calculations to determine the paths of neutrons through deuterium and tritium which proved that Teller's design for the 'Super' (H-bomb) was not feasible (Rhodes 1995). Monte Carlo methods are used more often in SEM imaging and X-ray calculations (see, e.g., NIST's Web site (URL #1), Joy 1995, Goldstein et al. 2003) but they have a role in TEM in determining the expected spatial resolution of X-ray analysis as we'll discuss in Chapter 36. Figure 2.4 shows Monte Carlo simulations of electron paths through thin foils of Cu and Au.

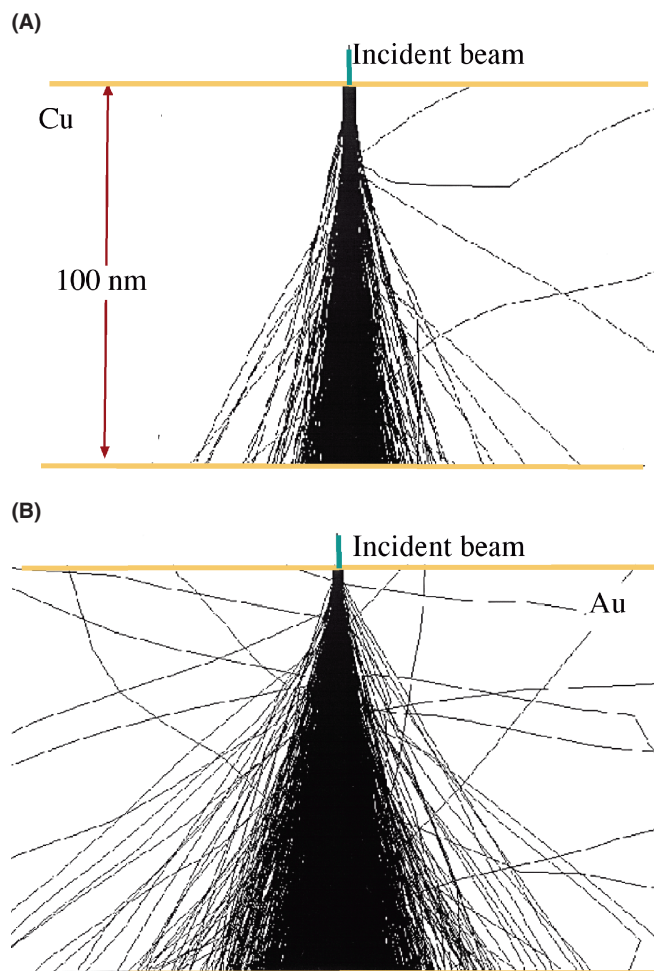


FIGURE 2.4. Monte Carlo simulation of the paths followed by 10^3 100-keV electrons as they pass through thin foils of (A) Cu and (B) Au. Notice the increase in scattering angle with atomic number and the small number of electrons that are scattered through $>90^\circ$.

2.6 HOW WE USE SCATTERING IN THE TEM

So why have we made you go through all this math? Because when you select electrons that have been scattered through a certain angle (choosing a θ), you are changing the effective scattering cross section (σ_θ), because the scattering strength generally decreases as the angle of scattering increases. Therefore, there will generally be less scattering at higher angles, which explains why we said at the start of the chapter that we are mainly interested in forward scattering in the TEM. Most scattered electrons are contained within $\pm 5^\circ$ of the direct beam.

300 VERSUS 100 kV

Total σ decreases as E_0 increases; electron scattering at 300 kV will be less than at 100 kV. Higher-density regions of your specimen scatter more than lower-density regions. The target becomes smaller as the bullets become faster!

You also have control of the scattering cross section in other ways. First, the accelerating voltage, which determines the electron energy E_0 (eV), will affect the cross section as implied in equation 2.3 (specifically for elastic scattering). In fact, for all forms of scattering, the total cross section decreases as E_0 increases. Therefore, intermediate- and higher-voltage TEMs will result in *less* electron scattering than typical 100-kV instruments and, as we'll see in Chapter 4, this has important implications for electron-beam damage in delicate specimens, such as polymers. Second, and more intuitively, you can choose specimens with different densities. Denser materials scatter more strongly, so you have to make them thinner to keep the single-scattering approximation valid.

We shall see in the next two chapters that the effect of the atomic number of the atom is more important in elastic than inelastic scattering and, as Z increases, elastic scattering dominates. This behavior helps when we consider ways to enhance scattering (and therefore contrast) in low- Z materials such as polymers and biological tissue.

2.7 COMPARISON TO X-RAY DIFFRACTION

There is a very good reason why electrons are used in microscopy: they have a 'suitable interaction' with matter. Most descriptions of the interaction of electrons with matter are based on scattering. You will come across such topics as kinematical scattering and dynamical scattering in addition to elastic and inelastic scattering, and we will use the formalism of a scattering factor to describe the process mathematically. It is this scattering process that varies with the structure or composition of the specimen, permitting us ultimately to image a microstructure, record a DP, or collect a spectrum. We'll see in the next chapter that scattering factors are used when we consider electrons as waves and their diffraction as a specific form of scattering.

So now it's time to move from billiard balls to waves. Historically, it was diffraction that provided most of the crystallographic information we have about materials, and the majority of those studies used X-rays. This is partly why X-ray diffraction is so well documented in the scientific literature. A good understanding of X-ray diffraction helps considerably in understanding electron diffraction; however, the primary processes by which electrons are scattered are very different to the processes by which X-rays are scattered. Electron scattering is much more complex.

X-rays are scattered by the *electrons* in a material through an interaction between the negatively charged electrons and the electromagnetic field of the incoming X-rays. The electrons in the specimen

respond to the applied field of the X-ray flux, oscillating with the period of the X-ray beam. These accelerated charged particles then emit their own electromagnetic field, identical in wavelength and phase to the incident X-rays. The resultant field, which propagates radially from every scattering source, is called the scattered wave.

ELECTRONS VERSUS X-RAYS

Electrons are scattered much more strongly than X-rays.

Electrons are scattered by *both* the electrons and the nuclei in a material; the incoming negatively charged electrons interact with the local electromagnetic fields of the specimen. The incoming electrons are therefore directly scattered by the specimen; it is not a field-to-field exchange as occurs for the case of X-rays.

2.8 FRAUNHOFER AND FRESNEL DIFFRACTION

Diffraction of visible light is well understood, so we should carry over as much of the analysis as possible. Optics is a venerable discipline with a history of several hundred years and what we're trying to do here is condense the principal messages from classic texts such as Hecht (2003) into a few pages. So, as with electron scattering, we'll be making a few simplifications. If you have any experience with diffraction of visible light you will have encountered Fraunhofer and Fresnel diffraction.

- Fraunhofer diffraction occurs when a flat wavefront interacts with an object. Since a wave emitted by a point becomes planar at large distances, this is known as far-field diffraction.
- Fresnel diffraction occurs when it's not Fraunhofer. This case is also known as near-field diffraction.

We will see later that electron-diffraction patterns correspond closely to the Fraunhofer case while we 'see' the effects of Fresnel diffraction in our images.

In TEM we will find both forms of diffraction. We will briefly go through the Huygens' explanation of how a wave propagates, then consider Fraunhofer diffraction from two slits (Young's slits) and then extend this process to many slits. So why discuss these topics now? There are two reasons for reviewing this analysis

- It reminds us that coherent interference is purely a matter of physical optics.
- We can introduce the concept of phasor diagrams which we'll use in later chapters.

Huygens explained the propagation of any wavefront by imagining that each point on the wavefront itself acts as a new source for a spherical wavelet. The wavelets interfere with one another to give the new wavefront and the process is repeated.

2.9 DIFFRACTION OF LIGHT FROM SLITS AND HOLES

In this section we will very briefly review the topic known as physical or geometric optics as it relates to diffraction. Much of what we know about diffraction of electron waves has been carried over from the understanding of the diffraction of visible light and X-rays. There are textbooks on this topic if you don't vaguely remember it from high school.

Two slits (the Young's slits experiment)

We start with diffraction caused by a wavefront incident on a pair of very narrow slits. We then select just two of the Huygens wavelets; these wavelets then must have the same phase at the slits. As they propagate past the slits, their phases differ depending on the position of the detector. The important term is the path difference $L = d \sin \theta$ as shown in Figure 2.5. The two wavelets propagating in direction \mathbf{r} have a path difference of L and a phase difference of $2\pi L/\lambda$, i.e., $2\pi d \sin \theta/\lambda$. If d and λ are such that this phase difference is actually a multiple of 2π (so $d \sin \theta/\lambda = \text{an integer}, n$) then the rays are again in phase and their amplitudes add. The condition for this additive interference is thus that $d \sin \theta = n\lambda$. Therefore, there is an inverse relationship between d and θ for a given d ; as d decreases, $\sin \theta$ increases. If we think of each wavelet as having an amplitude and a phase we can represent each by a vector—a phasor. When the phasors are parallel to one another (in phase) they add; when they are antiparallel, they cancel (since they have equal lengths). A phasor diagram is a way to plot the amplitude and phase of the total scattered wave; in other words, when we add the amplitudes of beams we must take into account their phase.

THE INVERSE RELATIONSHIP

$\theta \propto d^{-1}$ solely due to the positions of the slits. We'll come across an identical relationship when we talk about electron diffraction.

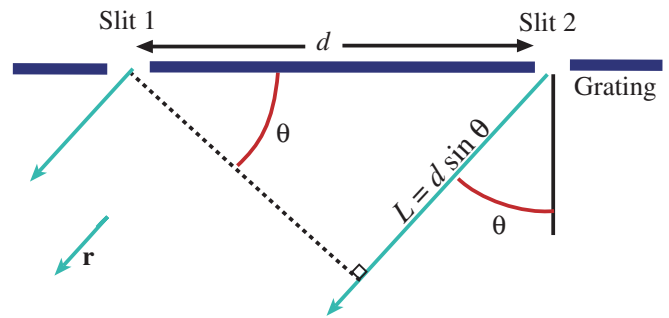


FIGURE 2.5. An incident plane wave is scattered by two slits, distance d apart. The scattered waves are in phase when the path difference $d \sin \theta$ is $n\lambda$.

Many slits (the diffraction grating)

When we extend this analysis to more than two slits we see a similar result but with added subsidiary peaks. The origin of the subsidiary peaks can best be illustrated by considering a series of phasor diagrams. (We'll find similar diagrams useful when we discuss TEM images in Chapter 27.) We'll examine the case of five slits. Each of the polyhedra in Figure 2.6 represents a different value of θ . When θ is zero, the five rays are all in phase and we simply add all of the amplitudes (the phasor vectors are aligned); as θ increases the rays become out of phase but the phasors can still add to give a large resultant vector but can also add to give zero. For example, when θ is exactly 72° ($360^\circ/5$ for 5 slits) the phasor diagram is a closed pentagon (shown in the figure) and the resultant amplitude is zero. This process repeats at 144° ($2 \times 360^\circ/5$) and 216° ($3 \times 360^\circ/5$). In between these values at 108°

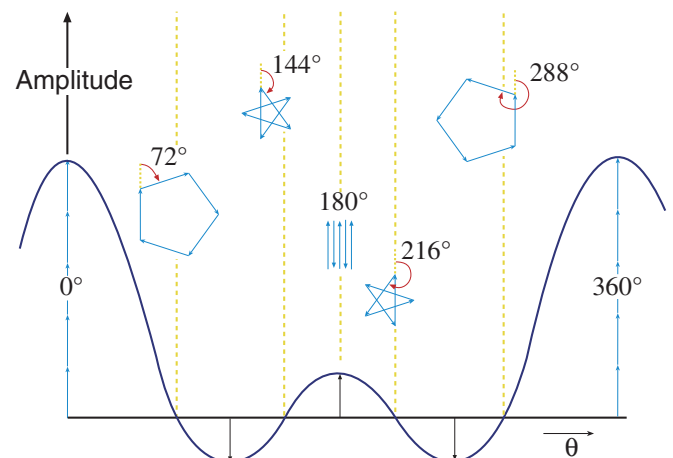


FIGURE 2.6. A phasor diagram showing how the total amplitude produced by summing five waves produced by five slits varies with the phase angle θ between the different waves. The individual phasors from each of the five slits sum to create a total amplitude of zero at $\theta = 72^\circ, 144^\circ, 216^\circ$, and 288° , large positive amplitudes at $\theta = 0^\circ$ and 360° , a single-phasor negative amplitude at $\theta = 108^\circ$ and 252° , and a single-phasor positive amplitude at $\theta = 180^\circ$. Remember the intensity is governed by the square of the amplitude, so positive and negative values both contribute to diffracted intensity.

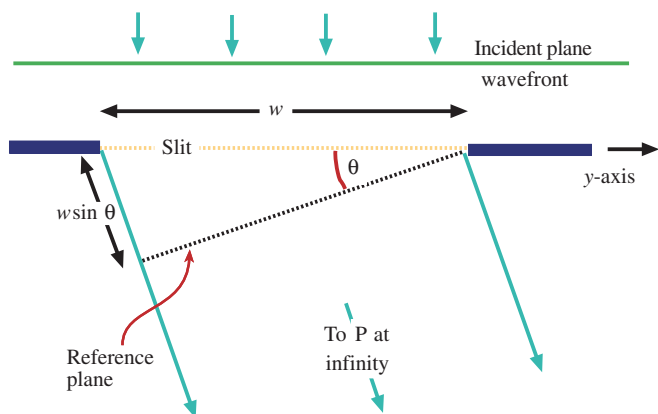


FIGURE 2.7. Geometry for the scattering from an individual slit.

($1.5 \times 360^\circ/5$) we produce a local maximum in amplitude which is repeated at 180° ($2.5 \times 360^\circ/5$). If we plot the amplitude as a function of θ , we produce the curve with a series of subsidiary maxima shown in Figure 2.6. From this figure you see that the amplitude is a strong function of θ and you'll learn in the next chapter that the electron intensity (which is what we see in images and DPs) is proportional to the square of the amplitude (so negative amplitudes don't mean anything) and the scattered electron intensity is, therefore, a similarly strong function of θ .

A single wide slit

What happens if we allow the slit to have some width as shown in Figure 2.7? Now the rays from *within* a single slit will interfere with each other. We can think of the single slit as being many adjacent slits of width δw . Imagine dividing the one slit into 11 slits of width $\delta w/11$. This one slit would then produce a phasor diagram as shown in Figure 2.8; if we make δw increasingly small,

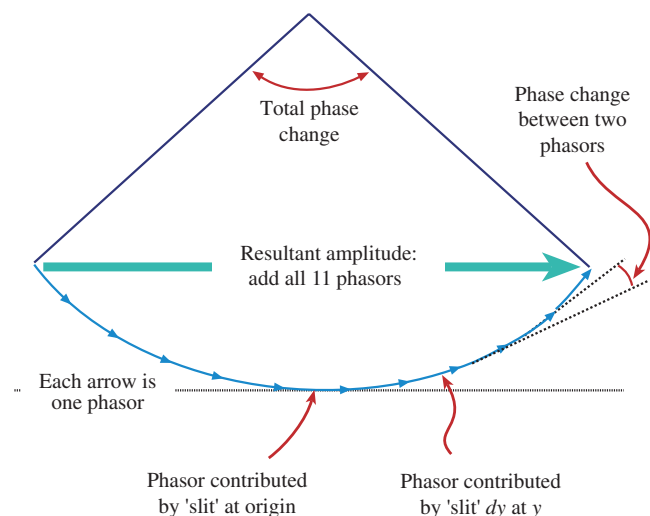


FIGURE 2.8. How the phasors from within an individual slit can be added to give the total phasor for the slit shown in Figure 2.7.

the phasor diagram becomes a curve: instead of having Figure 2.8, we have Figure 2.9 (for several different values of θ). If you do the full analysis you'd find that the amplitude from a single slit varies as $A = A_0 \phi^{-1} \sin \phi$ where ϕ is the phase $\pi w \sin \theta / \lambda$ for a slit of width w (which reminds you of the analysis from Figure 2.5). For one slit, we would see a zero in the phasor diagram when $\phi = \pm n\pi$. If we plot the intensity (rather than A) we obtain the Airy plot shown in Figure 2.10.

THE AIRY DISK

The disk of radius $r = 1.22\lambda/D$ is named after Airy and is one of the fundamental limits on the achievable resolution in TEM, as we will discuss in Chapter 6. If we introduce *any* aperture into *any* microscope we will limit the ultimate resolution of the instrument.

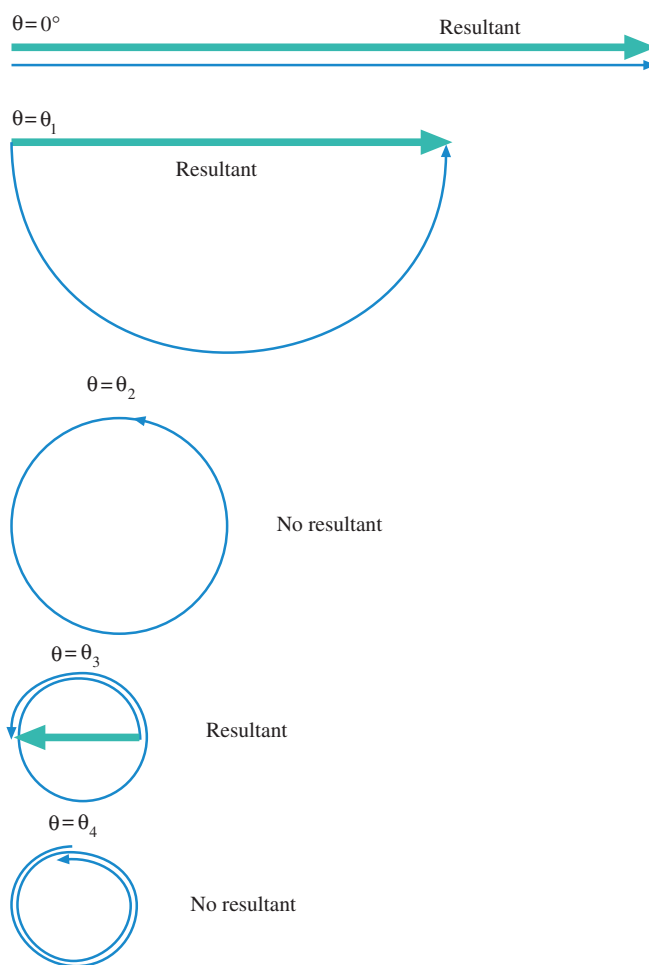


FIGURE 2.9. How a single slit can produce a beam which has zero amplitude for certain values of θ in Figure 2.7. The circles are directly comparable to the polyhedra in Figure 2.6. The total length of the phasor increments (from each dy) is the same in each figure.

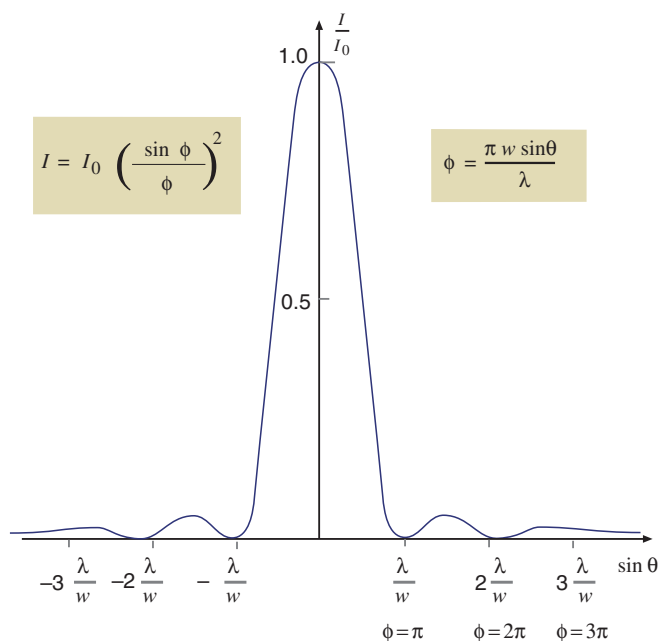


FIGURE 2.10. The plot of the resulting intensity for scattering from the slit shown in Figure 2.7; this is known as the Fraunhofer DP from a single slit; w is the slit width defined in Figure 2.7.

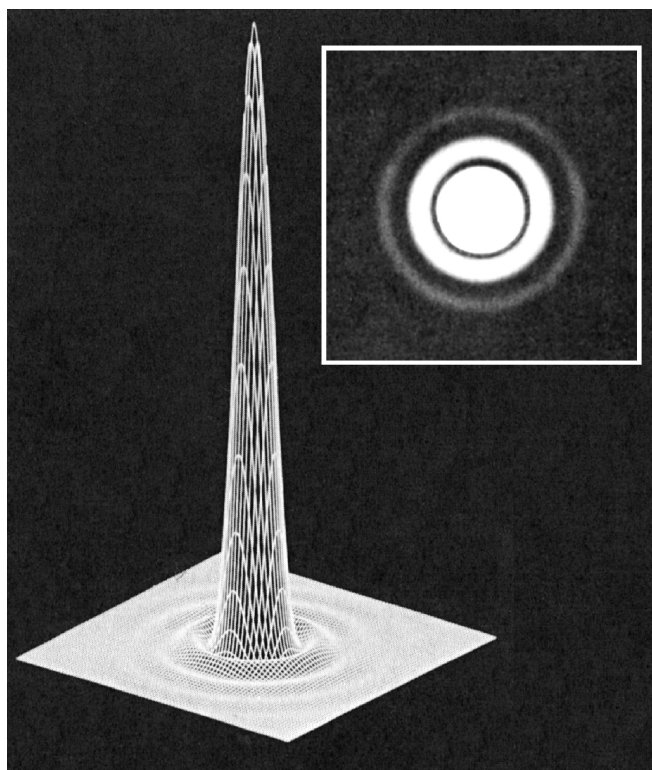


FIGURE 2.11. The visible-light intensity produced by a 0.5-mm-diameter circular aperture and the observed Airy rings (inset). The width of the central intense region is $1.22\lambda/D$.

Scattering from a circular hole

Now the real purpose of the exercise: without going into the detailed math we can replace the slit of width w ,

by a circular hole or aperture of diameter D . The resulting peak width in the plot of amplitude versus θ then has a maximum at $1.22\lambda/D$ as shown in Figure 2.11, which is a 3D representation of Figure 2.10 (but the third dimension is I , not I/I_0).

Because of the circular symmetry of the aperture, the calculation needed to obtain the number 1.22 involves the use of Bessel functions which you can find in texts on physical optics, a few of which we reference at the end of the chapter.

As the diameter of the aperture, D , decreases, the minimum resolvable spacing, r , increases (i.e., the resolving power gets worse). This expression for the Airy disk diameter also shows that as λ decreases, r decreases (so decreasing λ by increasing the accelerating voltage of the TEM will improve resolution).

Why is this relevant to TEM?

The important point about this analysis for TEM is that we'll see the same relationship in several later chapters. In those chapters, we will replace the slits by an aperture or we'll replace the hole by an atom or by your specimen. In other words, this analysis of diffraction from slits and holes is just geometry applied to optics—it's geometric optics.

2.10 CONSTRUCTIVE INTERFERENCE

To expand on this point, consider an infinite plane wave described by the usual characteristics of amplitude and phase. We can describe the wave function ψ by the standard expression

$$\psi = \psi_0 \exp[i\phi] \quad (2.11)$$

where ψ_0 is the amplitude and ϕ the phase of the wave. The phase depends on position x , such that if the path length changes by one wavelength λ , the phase difference is 2π . Stated another way, the phase difference $\Delta\phi$ between any two monochromatic (same wavelength) waves is related to the path difference Δx they must travel in going from the source to the detector. The relationship is

$$\Delta\phi = \frac{2\pi}{\lambda} \Delta x \quad (2.12)$$

This phenomenon of constructive interference is precisely what we discussed in Figure 2.6. Constructive interference between waves relies on the fact that the amplitudes of the waves add when you take account of the phase. If all waves scattered by all of the atoms in the specimen are to interfere constructively, they must all differ in phase by integral multiples of 2π . Clearly this condition requires that the path differences

traveled by all of the waves be integer multiples of the wavelength of the incident wave. We can ensure this by requiring that the scattering centers be periodically spaced; fortunately this can happen for all crystals. So the mathematical description of constructive interference is simplified (as we'll see in Part 2 of this text). The point here is that this analysis was carried out for X-rays and was not changed for electrons since it does not depend on the scattering mechanism, only on the geometry.

2.11 A WORD ABOUT ANGLES

Since angles (remember we mean semi-angles) are so important in the TEM (you can control some of them and the specimen controls others) we want to try to be consistent in our terminology.

- We can control the angle of incidence of electrons on the specimen and we will define the angle of incidence as α , as shown in Figure 2.12.
- In the TEM we use apertures or detectors to collect a certain fraction of the scattered electrons and we will define any angle of collection as β .
- We will define all scattering angles controlled by the specimen as θ . This may be a specific angle, such as twice the Bragg angle (where $\theta = 2\theta_B$) (see Section 11.4) or a general scattering angle θ . So θ is the scattering semi-angle for diffraction even though it is $2\theta_B$!

In fact the only angle of interest in TEM which is not given as a semi-angle is the solid angle of collection of X-rays by the XEDS detector (see Chapter 32) which is such a miserably small fraction of the total solid angle of X-ray generation (4π sr) that it is traditionally given in terms of the full collection angle!

2.12 ELECTRON-DIFFRACTION PATTERNS

We've mentioned a couple of times that the TEM is uniquely suited to take advantage of electron scattering because it can form a picture (DP) of the distribution of scattered electrons, which we'll discuss in Part 2 in much more detail. To understand fully how a DP is formed in the TEM, you need to go to Chapter 6 to see how electron lenses work and then to Chapter 9 to find how we combine several lenses to create the TEM imaging system. But before we take you through these concepts it is worth just showing a few of the many kinds of DPs that can be formed in the TEM. At this stage, all you have to do is imagine that a photographic film is placed directly after the thin specimen and that electrons scattered by the specimen as in Figure 2.1B impinge

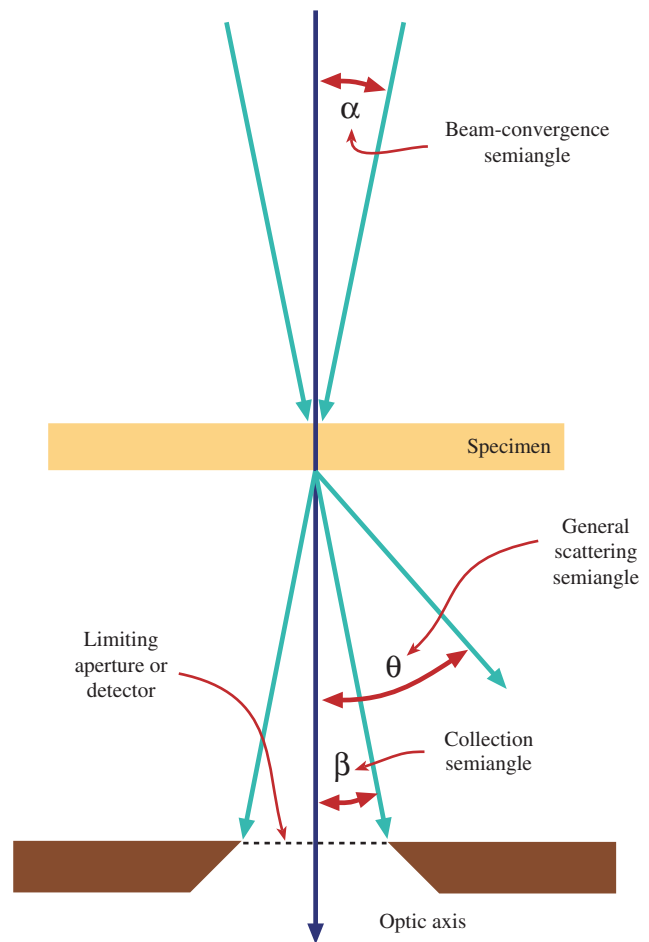


FIGURE 2.12. Definition of the major angles (i.e., semi-angles) in TEM. Any incidence/convergence angle of the beam is α ; any collection angle is β and general scattering angles are θ . All the angles are measured from the optic axis, an imaginary line along the length of the TEM column.

directly on the film. Under these circumstances, the greater the angle of scatter, the further off center the electron hits the film.

ON THE 'FILM'

Thus in a DP, distances on the film correspond to angles of scatter at the specimen.

Even using this simple description, however, you can comprehend some of the basic features of DPs. Figure 2.13 is a montage of several kinds of DPs, all of which are routinely obtainable in a TEM. You can see that several points we've already made about scattering are intuitively obvious in the patterns. First, most of the intensity is in the direct beam, in the center of the pattern, which means that most electrons appear to travel straight through the specimen. Second, the scattered intensity decreases with increasing θ (increasing distance from the direct beam), which reflects the

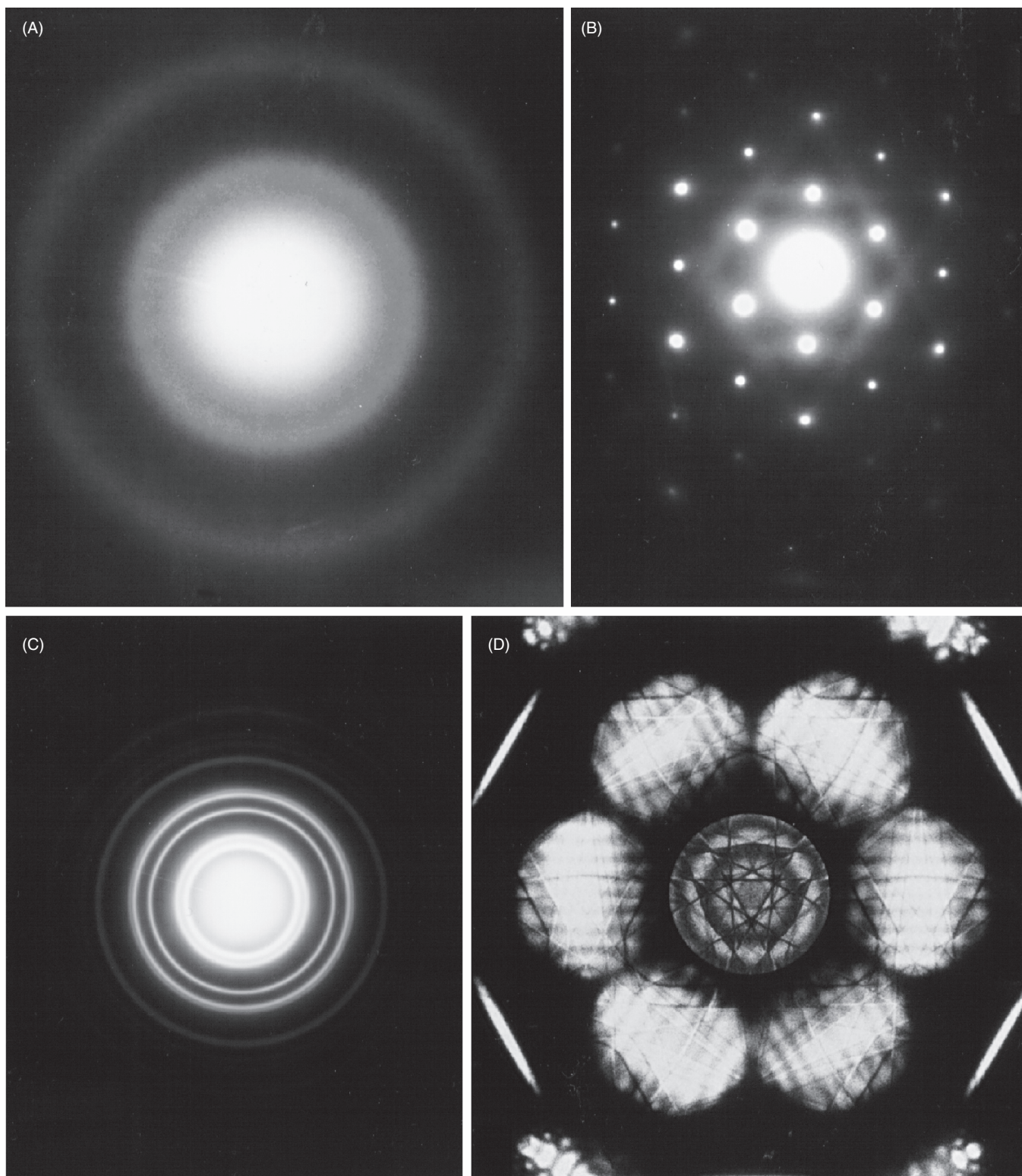


FIGURE 2.13. Several kinds of DPs obtained from a range of materials in a conventional 100-kV TEM: (A) amorphous carbon, (B) an Al single crystal, (C) polycrystalline Au, (D) Si illuminated with a convergent beam of electrons. In all cases the direct beam of electrons is responsible for the bright intensity at the center of the pattern and the scattered beams account for the spots or rings that appear around the direct beam.

decrease in the scattering cross section with increasing θ . Third, the scattering intensity varies strongly with the structure of the specimen. You'll see much more of this in Part II.

ANGLE OF SCATTER AND DISTANCES IN DPs

This relationship is different to the usual interpretation of images in which distances correspond to distances in the specimen, but it is critical to our understanding of diffraction patterns.

So far, we've only considered the amplitude/intensity of the electron wave and neglected the phase. When a wave is scattered, it will change its phase

with respect to the incident wave. This is because a wave cannot change direction and remain in step with a wave that is not scattered. The phase of the scattered wave is most important in the specific topic of phase-contrast images, which have until recently been the principal form of high-resolution, atomic-level images such as shown back in Figure 1.2. We'll also come across the importance of the phase of the scattered wave when we consider the intensity of diffracted electron beams and the intensity in diffraction-contrast images. But at this stage all you need to know is that the electrons in the beam are in phase when they hit the specimen and the process of scattering, in any form, results in a loss of phase between the scattered and direct beams.

CHAPTER SUMMARY

Remember that electrons are strongly scattered because they are charged particles. This is the big difference compared to X-rays. Electrons are scattered by the electron cloud and by the nucleus of an atom. Remember X-rays are only scattered by the electron cloud. (In case you are physics oriented, a quantum-mechanical calculation does give the same distribution as the classic calculation for the Coulomb force.)

We have defined four important parameters in this chapter:

σ_{atom}	the scattering cross section of one atom
σ_{total}	the number of scattering events per unit distance traveled in the specimen
$d\sigma/d\Omega$	the differential scattering cross section of one atom
λ	the mean free path of (average distance traveled by) an electron between scattering events

Finally, a note on grammar! Should we discuss electron *scatter* or electron *scattering*? Electrons are scattered and we observe the results of this scattering (a gerund) but in fact we see the scatter (noun) of the electrons, which can be measured. However, if you've been observant you'll have noticed that we have always used *scattering* to denote the effect. Our practice is also consistent with the popular usage, which goes back to the early work of Bragg and others.

SCATTERING AND CROSS SECTIONS

Born, M and Wolf, E 1999 *Principle of Optics* 7th (yes, 7th!) Ed. Cambridge University Press New York.

Perhaps *the* optics textbook in terms of classical treatments and number of editions.

Heidenreich, RD 1964 *Fundamentals of Transmission Electron Microscope* Interscience Publisher New York NY.

Jones 1992 gives a succinct introduction to scattering and Newbury (1986) gives a clear exposition on the units of cross sections. If you want to see a fuller description, read Wang (1995). If you're a glutton for punishment, the classic text is by Mott and Massey (1965) as we've already mentioned. You should realize that we've introduced you to some of the giants of electron optics, e.g., Airy, Fresnel, and Fraunhofer, who never knew about electron waves.

Jones, IP 1992 *Chemical Microanalysis Using Electron Beams* The Institute of Materials London.

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Wang, ZL 1995 *Elastic and Inelastic Scattering in Electron Diffraction and Imaging* Plenum Press New York. An in-depth treatment of scattering using a much more rigorous mathematical approach than in this chapter.

OPTICS

We should have references to some of the founders of optics here, especially Abbe, Airy, Fraunhofer, and Fresnel, but we'll leave you to chase those up in the optics texts.

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SOME MICROANALYSIS AND MORE

Goldstein, JI, Newbury, DE, Joy, DC, Lyman, CE, Echlin, P, Lifshin, E, Sawyer, LC and Michael, JR 2003 *Scanning Electron Microscopy and X-ray Microanalysis* 3rd Ed. Kluwer New York.

Joy, DC 1995 *Monte Carlo Modeling for Electron Microscopy and Microanalysis* Oxford University Press New York.

Rhodes, R 1986 *The Making of the Atomic Bomb* Simon and Schuster New York. See p 282.

Rhodes, R 1995 *Dark Sun: The Making of the Hydrogen Bomb* Simon and Schuster New York. See p 423. These books are well worth reading because of both the historical and the scientific content.

URLs

1) <http://www.cstl.nist.gov/div837/837.02/epq/index.html>

SELF-ASSESSMENT QUESTIONS

- Q2.1 What is a cross section and in what units is it measured?
- Q2.2 Distinguish between total, atomic, and differential cross sections.
- Q2.3 Why are we interested in variations in the scattering intensity and the angular distribution of electron scattering?
- Q2.4 What is the mean free path of an electron?
- Q2.5 What do we mean by the term *electron beams* and why do we ask this question?
- Q2.6 How is the direct beam different from or similar to the scattered beams?
- Q2.7 Distinguish scatter and scattering.
- Q2.8 What's the difference between forward scattering and backscattering?
- Q2.9 What distinguishes elastic and inelastic scatterings?
- Q2.10 Distinguish between coherent and incoherent scattering.
- Q2.11 Describe what distinguishes diffraction from other kinds of scattering.
- Q2.12 Distinguish between Fraunhofer and Fresnel diffractions.
- Q2.13 Distinguish the angles α , β , θ , and Ω .
- Q2.14 List the different ways a specimen can scatter electrons.
- Q2.15 How many different ways can you control the scattering processes in the TEM?
- Q2.16 How can you select electrons that have suffered a specific kind of scattering?
- Q2.17 What's the fundamental difference between electron scattering and X-ray scattering?
- Q2.18 What is a phasor diagram?
- Q2.19 Why would you want to draw a phasor diagram in TEM?
- Q2.20 How small is a small angle in the TEM and why are scattering angles in the TEM usually this small?

TEXT-SPECIFIC QUESTIONS

- T2.1 Write down concise definitions of coherent, incoherent, elastic, and inelastic as we use them and link these definitions to the information in Figure 2.2.
- T2.2 Explain in a paragraph the relationship between scattering cross section and atomic scattering factor, mentioning the important factors that influence them.
- T2.3 Explain the link between the information in Figures 1.3 and 2.1.
- T2.4 Distinguish the scattering angles θ and Ω in Figure 2.3 and the information that can be gathered within them. Relate these angles to the relevant angles in a TEM described in Figure 2.12.
- T2.5 Sketch the intensity projected onto a photographic plate or viewing screen from the scattering produced by the Cu and Au specimens in Figure 2.4. The result does not look like the intensity in either a typical TEM image or DP shown in many figures throughout the book. Explain why this is so.

- T2.6 Why are the backscattered electrons so few in number in Figure 2.4A and B and why do they all scatter to the one side in Figure 2.4A?
- T2.7 Draw equivalent diagrams to Figure 2.5 for (a) 2 slits $d/2$ apart; (b) 2 slits $2d$ apart; (c) 5 slits d apart. What does this tell you about the effect on the scattering distribution of both the number and the spacing of the scattering centers?
- T2.8 Draw a phasor diagram like Figure 2.6, but for three slits only.
- T2.9 What is the relationship between Figure 2.10 and Figure 2.11?
- T2.10 Make a copy of Figure 2.13. Cut out two circular holes with diameters ~ 5 and ~ 40 mm in another sheet of paper corresponding to different collection angles (β) in Figure 2.12. Superimpose the smaller circular hole on the different patterns in different positions to simulate the selection of electrons for forming images in a TEM. Note how easy it is to select electrons scattered in specific directions, but also note how many electrons are excluded when you do this. (a) What does this tell you about the advantages and disadvantages of a small selection aperture (or small detector)? Now superimpose the larger hole and note how many more electrons can be selected. (b) What does this tell you about the advantages and disadvantages of a large selection aperture (or large detector)?



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