Addressing Coulomb’s singularity, nanoparticle recoil and Johnson’s noise

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Abstract. Recent experiments prompt rethinking of the basics of elastic and inelastic electron scattering in electron microscopy. Standard approximations of elastic scattering largely based on Bragg’s law seem less clearly relevant when individual columns or even single atoms are probed. Phase shift analysis of atomic scattering can provide some checks and insights. The dielectric theory of aloof beam interactions is severely tested by observations of nanoparticle recoil but may be capable of explaining de-coherence effects induced by thermal fluctuations.

1. Introductory challenges
Progress in electron microscopy seems to follow a helical trajectory. Improved instrumentation and pioneering observations provide fresh opportunities and challenges. These may expose the need for radically new thinking or more simply a reworking of earlier ideas that have fallen out of fashion. Bragg reflection concepts, highly relevant in the days of diffraction contrast [1], underpinned the standard approximations for elastic scattering, including diffuse scattering. They reached a high point of Baroque splendour in high resolution phase contrast, where images of crystal defects, simulated by periodic continuation of an artificially very large unit cell involve many hundred diffracted beams [2]. Although it continues to use Bragg concepts such as Bloch waves for image interpretation, scanning transmission electron microscopy (STEM) using high angle dark field mode in reality increasingly provides a quite discordant example where a single column or even a single atom is illuminated in surroundings that are largely irrelevant [3]. Had we started from here it seems unlikely that we would have tackled the problem by imposing some artificial periodicity and then applied Bragg reflection theory! The standard phase shift theory of electron scattering in an atom, recently explored for this situation [4] and further developed in section 2 below, deals more convincingly with the Coulomb singularity in the atomic potential and thus provides a useful check on Bragg methods. Although we may need to use over 200 phase shifts at 100 keV, this is still not excessive compared with the number of Bragg reflections noted above.

Easily accessible and relatively intense plasmon losses in free electron metals like Al attracted attention in electron microscopy about 50 years ago and were used for microanalysis that still looks impressive today. Dielectric excitation theory was then systematically developed for the more general interpretation of energy loss spectra in this valence region including excitation by both penetrating and aloof beams. Then for a period, the interest of microscopists in valence losses evaporated when the more directly interpretable core loss region became available. Recent access to the 1-2eV region including the surface losses of Au and Ag nanostructures has however arrived in a
fresh turn of the helix. The startling resurgence that has occurred may merit, as described in section 3 below, the description “plasmonic euphoria”.

Instrumentation is now being developed to push electron spectroscopy down towards 10 meV and below where thermal excitation must be considered. Energy gains as well as energy losses will then be observed. Much higher energy loss and gain probabilities can be generated in the visible region in conjunction with pulsed laser illumination. Events linked to energy losses or gains far too small to be measured can also be potentially important in causing electron beam de-coherence and loss of image contrast. In section 4 below the application of dielectric excitation theory to these problems and the effect of thermal excitation on bremsstrahlung emission in this region is investigated.

2. Exact computation of atomic exit waves

2.1. Partial wave theory atomic scattering

An exact solution for the scattered wave emerging from an atom illuminated by an incident plane wave \( \exp[ikz] = \exp[ikr\cos\theta] \) is in principle provided in terms of spherical Bessel functions \( j_l(kr) \) and \( n_l(kr) \) by the long established partial wave theory of electron scattering [5].

\[
\psi_{\text{scatt}}(r, \theta) = \sum_l \{ \exp[2i\delta_l] - 1 \} \left( l + \frac{1}{2} \right) i^l (j_l(kr) + in_l(kr)) P_l(\cos \theta) \tag{1}
\]

This equation gives directly the scattered wave contribution to the exit wave just outside the atom and rigorously describes its free space propagation between there and the far-field asymptotic region where the exact atomic scattering factor \( f(\theta) \) emerges.

\[
f(\theta) = (2ik)^{-1} \sum_l (2l + 1) \{ \exp[2i\delta_l] - 1 \} P_l(\cos \theta) \tag{2}
\]

Using the atomic potential \( V(r) = 2mU(r)/\hbar^2 \), procedures have been developed for accurate computation of the partial wave shifts \( \delta_l \) (which are real) but good approximations can be obtained from WKB methods or from the Born integral

\[
\delta_l^{\text{Born}} = \int_0^\infty kr^2 j_l^2(kr)U(r)dr \tag{3}
\]

When the phase shifts given by eqn. (3) are small and are substituted in eqn. (2) we get a connection to the Born atomic scattering factor generally used in electron microscopy

\[
f_b(\theta) = k^{-1} \sum_l (2l + 1) \delta_l^{\text{Born}} P_l(\cos \theta) = \frac{1}{4\pi} \int \exp[iK.r'] U(r')dr' \tag{4}
\]

Here as usual \( K = 2k \sin(\theta/2) = 4\pi \sin(\theta/2) / \lambda \). The exact scattering factor \( f(\theta) \) is complex and therefore significantly different than \( f_b(\theta) \) which is real although at 100 keV their magnitudes are quite similar [4].

2.2. Exit wave computations and checks on Bragg scattering at a Coulomb singularity

The partial wave scattering theory just outlined has recently been used to study the scattering of 100keV electrons in different atoms [4] though mostly with extremely small exit radii of only 0.7Å. With a larger exit sphere of radius \( r_e = 1.5A \), typical of ionic radii as well as half the near neighbour spacing in many solids, the number of phase shifts required at 100 keV can be \( kr_e \approx 250 \). For \( r > r_e \) the potential was treated as constant as is done in the muffin tin method of solid state physics. The magnitude and phase of the exit wave was computed by adding to the incident wave the scattered wave from eqn. (2) and evaluating the result at points \( \rho = \sqrt{x^2 + y^2} \) in the exit plane \((x,y, z = r_e)\). In this computation the Born phase shifts given by eqn. (3) were used since they agreed quite closely with values given by the WKB method. Writing the exit wave as \( \psi_e = \exp[ikz]/F\exp[i\eta] \), figs 1 and 2
display for a 100 keV electron the modulus $F$ and phase $\eta$ as a function of distance $\rho$ from the projected centre of an atom of Au. The focusing effect of the atom in raising the wave magnitude at small $\rho$ as well as the increasing phase shift is evident. Phase grating theory might roughly explain the phase shift but would predict constant unit amplitude which evidently holds approximately only for $\rho > 0.3\AA$. The small oscillations in this region may be due to ringing at the edge of the muffin tin but could also arise through interference between trajectories converging from opposite sides of the atom.

Figs 1 and 2 show the magnitude $F$ and phase $\eta$ of the exit wave in the exit plane $z_e = 1.5\AA$ as a function of distance $\rho$ in angstroms A from the projected centre of the atom. Light blue denotes the exact computation. The computations based on $f_B(\theta)$ with full phase shifts and reduced phase shifts modulo $\pi$ are shown in red and green respectively. The dark blue curve is an exact computation for a smoothed but still spherically symmetrical potential defined by low order Fourier components. For comparison with the exact result (light blue), three other approximations are shown in figs. 1 and 2 starting in red with the Born exit wave computed using the approximation $\langle \exp[2i\delta] - 1 \rangle \approx 2i\delta$ strictly valid only for small phase shifts. This is the exit wave that would be obtained by using the Born (single scattering) expression of eqn. (4) in the far field and correctly propagating back to the exit plane. It can be seen that this approximation considerably over-estimates the focusing effect. It agrees more closely at larger $\rho$, although the small oscillations there seem to be in antiphase with the exact theory. The next approximation shown in green is akin to the pseudo-potential method used in band theory where advantage is taken to re-express the phase shifts modulo $\pi$, which leaves the exact result of eqns. (1) and (2) unchanged but results in a reduced value for the far-field $f_B$ which can be used to propagate back to the exit plane. In the case considered each of the first five phase shifts could be reduced by $\pi$. This approach evidently agrees more closely with the exact results and gets the small oscillations in the right phase. The small changes resulting in $f_B(\theta)$ at small $\theta$ values might cause complications however in some EM applications.

More interesting is the third approximation (dark blue) which follows the usual practice of considering only the low order-Fourier components of the scattering potential that might for instance lie in the zero-order Laue zone of a crystal. The phase shifts for such a smoothed, but still spherically symmetric, potential were constructed from eqn. (4) using the orthogonal property of the $P_l$ functions and restricting $f_B(\theta)$ to the small angle region $\cos \theta > 0.98$. That the result appears to agree remarkably well both in modulus and phase with the exact computation indicates that essential forward scattering features of the Coulomb singularity are not violated by smoothing the potential in this way. It should be emphasized however that this smoothing is not equivalent to making any of the usual projection approximations either for the potential or for the scattering. It could be interesting to check the effect of these approximations by comparing the above results with those of slice computations taking several steps within the atom to describe the wave spreading effect that is handled automatically in the partial wave theory.
3. Nanoparticle recoil and plasmonic euphoria

The possible effect on small supported clusters of uncontrolled energy and momentum transfers from the electron beam has been a long-standing worry in electron microscopy. In their pioneering work in STEM development, Crewe and colleagues made cine film recordings of the motion of small clusters as well as of single atoms of U and other heavy elements on carbon supports [6]. Their careful analysis suggested however that in most cases this was purely thermal motion and was not induced by the electron beam. Similarly cluster recoil effects in the fast electron wind that might be expected from transfers of linear momentum in Bragg reflection have never been reported though some indirect evidence for torque-induced reorientation was found [7].

On the other hand, kaleidoscopic structural changes were observed more than two decades ago in video studies of metallic nanoclusters under HREM imaging conditions [8]. Beam heating effects were ruled out [9] and these changes seem possibly driven by much rarer events than the momentum transfer in a Bragg reflection or the energy transfer through plasmon excitation and decay. Rather recently however with the much higher and more localized beam currents available in STEM, nanoparticle attraction and recoil induced by placing the probe close to, but just outside, the particle have been observed [9]. This recoil is hard, though perhaps just possible [10], to understand with the usual theory of dielectric excitation by an aloof beam where the transverse forces are usually rather small and nearly always attractive. Coulomb forces due to temporary charging of the nanoparticle could be greater but would also tend to be attractive.

Figs 3 and 4 indicate however that it may be wise to cast aside plasmonic euphoria and think outside the narrow, sub 5eV spectral window. Substantial valence excitation takes place at energies higher than the sharp 2eV plasmon loss and could for instance result in the ejection of a secondary electron generating much more recoil momentum than is available from photon emission following surface plasmon radiative decay. All these cases call for a more careful treatment of momentum transfer involving the fast electron, the excited atom or cluster and any photon emitted or electron that is ejected [11]. It is also hard to understand the mechanism for the recently observed transfer of angular momentum to a nanocluster from a focused vortex beam. For non-spherical clusters elastic scattering has recently been suggested as an explanation [12] and could perhaps be consistent with the already mentioned detection of Bragg torque momentum transfer. The precise nature of the binding and frictional forces between the nanoparticle and the substrate in different cases is of course a potentially large unknown factor that has been addressed so far mainly by atomic force microscopy.
4. Thermal and laser excitation effects in inelastic scattering and de-coherence

4.1. Energy loss and gain events

Although a rigorous theory of thermal fluctuations in dielectric excitation has been developed [14], the effects are most easily understood, at least on an *ad hoc*, basis for harmonic oscillator excitations such as phonons or plasmons. These excitations have a ladder of states \( n \) with energy levels \((n + \frac{1}{2}) \hbar \omega \) and equilibrium thermal occupation probability \( p(n) = \exp[-n\hbar \omega / kT]/(1- \exp[-\hbar \omega / kT]) \). For radiative transitions with \( \Delta n = 1 \) from a state \( n \), the relative probabilities are \( n+1 \) and \( n \) respectively (corresponding to energy loss and energy gain events for a fast electron that stimulates them). When the stimulation is from equilibrium thermal radiation, the difference between these two rates is balanced by the spontaneous decay process \( \Delta n = -1 \). Summing the two rates indicates that the effect of temperature brings a multiplying factor \( F_T = (2n+1)p(n) = \coth(\hbar \omega / kT) \) to the total event probability calculated at \( T = 0 \). For \( \hbar \omega \ll kT \), \( F_T = 2kT/\hbar \omega \).

Gains as well as losses were first detected many years ago by pioneering transmission electron spectroscopy using thin films of LiF [15]. Creation and annihilation events were stimulated by the electron beam in a thermally excited population of optical phonons of energy \( \hbar \omega = 0.05 \text{ eV} \). The film thickness systematically influenced the precise energies of the loss peaks as well as the ratio \( R \) of the areas under the loss and gain peaks (which ranges between 5 and 15). Analysis of the results reveals quite good agreement between the observed values of \( R \) and the above theory for stimulated transitions of an oscillator which predicts \( R = (\Sigma(n+1)p(n))/\Sigma p(n) = 1 + 1/\Sigma p(n) = \exp[\hbar \omega / kT] \). Possibly because the spontaneous decay process is too slow in comparison with the passage time of the fast electron, it does not seem to make a detectable contribution to the energy gain.

A radically different situation arises in recent experiments using photon-induced near-field electron microscopy (PINEM) [16, 17]. Here, near nanostructures subjected to high intensity pulsed laser excitation, loss spectra are collected in a synchronously pulsed alook electron beam. Energy and momentum conservation excludes any interaction between the fast electron and the light waves in the far-field region but the observed cascade of energy losses and gains of equal magnitude is well explained through its interaction with the near-field region of the optical pulse emitted by the nanostructure [17-18]. In these experiments the electron is simply acting as a highly localized detector of the nanostructure’s response to the pulsed laser field. Because of the enormous intensity of the laser pulse, the contribution of energy loss or gain processes stimulated by the electron itself is by comparison negligible as can be seen from the loss spectra collected when the photon and electron pulses are not in synchrony. It may nevertheless be profitable to try to bridge the gulf between these photon-stimulated and electron-stimulated response situations.

In most cases, the pulsed photon excitation is delivered off resonance to a single mode. If the photon energy \( \hbar \omega \) were tuned to the resonance peak of the mode to drive it to a sufficiently high energy level \( n \), the electron-stimulated loss and gain peaks linked to \( n+1 \) and \( n \) respectively might be detectable. Such an investigation would be much easier for a planar or cylindrical structure parallel to the electron beam since the electron can then interact only with evanescent waves at \( q_z > \omega/v \) and these cannot be generated by the specimen in response to the laser pulse. In more general nanostructures, emitted waves stimulated by the electron are not cleanly separated from those generated in response to the laser pulse and the two effects must be added. It has even been conjectured that interference effects could be exploited between electron and laser stimulation [19]. To observe such effects however, rather than simple addition of intensities, a sufficiently precise phase correlation between the laser field and the electron wave function would have to be obtained and kept constant throughout the many pulses needed to acquire a spectrum.

4.2 De-coherence processes

Preservation of coherence is vital in the specimen and post specimen region in conventional electron microscopy and in the pre-specimen region in STEM. Various mechanisms of de-coherence were investigated recently [20] for aloof beam interactions where the dielectric excitation theory seemed to
offer an explanation for experimental observations [21] in holography. These cases usually involve interference between electron paths separated by some distance $d_y$ as they travel with the same impact parameter close to a dielectric surface. Evanescent waves arising from the dielectric response cause inelastic scattering and there will be immediate loss of coherence when the interaction affects the two paths with significantly different phases i.e. for tangential wave vectors $q_y > 1/d_y$ leading to a change in their relative phase of the paths after the scattering. Figure 5 schematically indicates the situation.

Fig. 5. Schematic drawing for de-coherence events suffered when an aloof beam travels across a conducting plate in two parallel interference paths (red) and is then focused by a lens to form an image with interference fringes. Interaction with the evanescent waves $(q_y, \omega/v)$ stimulated in the plate can cause complete destruction of any phase difference between paths separated by $d_y > 1/q_y$. At much smaller evanescent wave vectors $q_y$, the phase relation is preserved in the blue paths after scattering but the fringes they later produce will be shifted due to the angle of scattering $q_y/k$.

Recent experiments have [22] have demonstrated de-coherence caused by a random walk accumulation of very small-angle scattering events. The effect was studied with 200keV electrons in an unusual 50cm long, 5mm diameter tube and was found to be proportional to temperature. So far no strong dependence on wall material has been reported but de-coherence increases with distance from the tube axis (and with proximity to the wall). A quantitative explanation was offered in terms of very low frequency magnetic fields emitted by current loops in Johnson noise fluctuations.

Some further insight into this interesting phenomenon should in principle be available by extending the aloof beam dielectric excitation theory already applied to de-coherence in holography. Given the differential expression for the single photon event probability $P(x_0, \omega, q_y)$ that this theory provides in terms of $q_y$ momentum transfer, we can directly calculate the mean square deflection angle $\langle \theta^2_y \rangle$.

$$\frac{d^2P(x_0, \omega, q_y)}{d\omega dq_y} = \frac{e^2 L}{2\pi^2 \varepsilon_0 \hbar v^2} \text{Im} \left\{ -\frac{1}{v+\nu_0\varepsilon} + \frac{\beta^2}{v+\nu_0} \right\} \exp[-2\nu_0 x_0]$$  \hspace{1cm} (5)

$$\langle \theta^2_y \rangle = \frac{1}{k^2} (q_y^2) = \int_{-\infty}^{\infty} q_y^2 dq_y \int_{0}^{\infty} d^2P(x_0, \omega, q_y) dq_y d\omega$$  \hspace{1cm} (6)

In eqn. (5), where $\beta = v/c$, $\nu_0^2 = [q_y^2 + (\omega/v)^2 - (\omega/c)^2]$ and $\nu^2 = [q_y^2 + (\omega/v)^2 - \omega(\omega/c)^2]$, the relativistic form [22] has been used since this includes magnetic forces. For the observed 1mm impact parameters $x_0$, the exponential factor restricts attention to the low frequency range $\omega/2\pi < 10^{10}$Hz. The dielectric response can then still be described by the Drude expression $\varepsilon(\omega) = 1 - \omega^2/\omega(\omega+i\gamma)$ taking
the damping constant $\gamma = \varepsilon_0 \omega_p^2 / \sigma(T)$ where $\sigma(T)$ is the DC conductivity at temperature $T$. Even for high conductivity metals, $\gamma > \omega$ at these low frequencies and so $\sigma(\omega) \approx 1 - \omega^2 / \omega^2 + i \sigma(T) / \varepsilon_0 \omega$. The non-relativistic approximation to eqn. (5) then yields an event probability proportional to $1 / \sigma(T)$ which above the Debye temperature is proportional to $T$ due to scattering of carriers by the increased population of thermal phonons. Some of the thermal enhancement effects discussed in 4.1 may thus be included even if losses and gains are not properly distinguished. Initial investigations indicate however that the second (purely relativistic) term in eqn. (5), with a less simple dependence on conductivity, may actually make a bigger contribution. On the basis of work in progress [14] it seems possible that the relativistic theory of aloof beam interactions may be able to account for the Johnson noise observations. It should be noted however that scattering in the $x$ direction normal to the plate is more complex since we can now expect a non-zero mean deflection $<\theta_x>$ together with a fluctuation $<(\theta_x - <\theta_x>)^2 = <\theta_x^2> - <\theta_x>^2$. For electrons traversing a tube and depending on their distance from the wall, there will also be a mean deflection in the radial direction acting like a weak diverging lens with high aberration.

4.3 Touching the warm void

The thermal multiplication factor $F_T$ can cause significant increases in the probability of very low energy bremsstrahlung emission by an electron accelerated in some external potential. At $T = 0$ this process can be viewed as a spontaneous decay between the no longer free quantum states of the electron in that potential. For instance a fast electron propagating through a crystal and described by Bloch waves can lose energy through bremsstrahlung in a process that can be conveniently described as an inelastic interband transition coupled to the emission of a photon [1]. Such coupling, which would be forbidden in free space by energy-momentum conservation, is made possible by the non-free electron form of the electron’s $E(k)$ relation in the crystal. For Bloch waves involving only reflections from the zero order Laue zone, we can equally well consider the bremsstrahlung as channeling radiation generated by the undulating motion of the electron between the atomic planes. Similarly, in a uniform magnetic field $B$, the electron energy states are harmonic oscillator functions with energy levels $E_n = (n+\frac{1}{2})\hbar \omega$ and yield spontaneous decay emission at the Larmor frequency $\omega_L = eB/m$ matching the period for the circular orbit $T_L = 2\pi / \omega_L$.

From the classical theory of bremsstrahlung [24] the radiated energy loss rate $dE_{rad}/dt$ is related to the electron’s acceleration $a(t)$.

$$E_{rad} = \frac{e^2}{6\pi \varepsilon_0 c^3} \int_{-\infty}^{+\infty} [a(t)]^2 dt \frac{e^2}{12\pi^2 \varepsilon_0 c^3} \int_{-\infty}^{+\infty} [a(\omega)]^2 d\omega$$  (7)

Here through Parseval’s theorem, the energy loss can also be expressed as an integral over $[a(\omega)]^2$ where $a(\omega)$ is the Fourier transform of $a(t)$. This step provides the spectrum of energy losses. Assuming that these losses are entirely due to single photon emission the loss probability is

$$P_{rad} = \frac{e^2}{6\pi^2 \varepsilon_0 \hbar c^3} \int_{0}^{+\infty} \frac{1}{\omega} [a(\omega)]^2 d\omega$$  (8)

These equations were used [20] to compute event probabilities $P_{rad}$ for simple trajectories in electron optics including a single $2\pi$, 10 cm radius turn of a 200 keV electron in a mandolin or similar magnetic system. With $a(t) = v^2 / r$ we find $P_{rad} = 0.03$ for the emission of a photon of energy $\hbar \omega$ $\approx 10^6$eV. With the thermal magnification factor $F_T = 2kT / \hbar \omega$ (arising now from the occupation probabilities $p(n)$ of the photon far-field modes rather than from the Larmor oscillator modes) the expected number of events at room temperature is increased to about 1500. Since these events will be divided equally between energy losses and gains, we can estimate $4 \times 10^{-4}$ eV for the rms energy.
broadening. Such broadening is too small to be detected but the events might conceivably result in some de-coherence depending on where the bending system is placed in the column.

Unlike the example just considered, the episode of acceleration in most trajectories results in a change in the magnitude or direction of the velocity and eqn. (8) then exhibits a logarithmic divergence at low \( \omega \) known as the infrared catastrophe. Inclusion of the thermal magnification factor raises this low frequency divergence in \( P_{\text{rad}} \) to \( \omega^{-1} \) and creates a \( \ln(\omega) \) divergence in \( E_{\text{rad}} \). In practice it should be possible to avoid any problems by introducing an appropriate low energy or momentum transfer cut off.

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