Inelastic scattering and holography

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Dedicated to Professor Harald Rose on the occasion of his 65th birthday

Abstract

The controversy about whether or not an inelastically scattered electron wave can still interfere with a reference wave is solved by treating the whole problem rigorously and describing electron, source and object in one Hamiltonian. It turns out that, in principle, interference can occur between an inelastically scattered wave and a reference wave from the incident beam spectrum provided the energy difference is smaller than about $10^{-15}$ eV. However, it is argued that the density of states in source object and electron wave is much too small to make this effect observable.

1. Introduction

Recently, it has been a matter of controversy whether inelastically scattered electrons can yield interference fringes so as to obtain holograms. Basically there are two schools of thought. In the first, it is argued that electron holography is a perfect energy filter [1–3]. The argument can be traced back to the “which way” reasoning of the two-slit experiment which is a basis for quantum mechanics [4]. In the case of off-axis holography, the electron beam is split into a reference wave passing through the vacuum and an object wave passing through the object (Fig. 1). If the interaction with the object is inelastic, the state of the object is changed. From observing this change it can be concluded that the electron is passed through the object and hence, according to Feynman, the interference with the reference wave is destroyed. A similar result can be obtained by reasoning that a fixed change in the energy of the object wave causes the holographic fringes to move at constant speed, so that the fringe contrast will vanish when recording the hologram [14]. For the fringes to be sufficiently stationary during a recording time of 1 s the maximal allowed energy change can be estimated at $10^{-15}$ eV. It should be noted that when also the reference wave passes through the object, both waves can excite the same object state. In that case the energy difference between both waves remains exactly zero and interference can occur. This has been experimentally demonstrated for plasmon excitations [5] and recently even the width of the interaction area could be estimated [6].

The above reasoning is similar to that in Ref. [7] where it is argued that for coherent illumination of
an object only contributions from waves resulting in the same final state of the object will be added up coherently whereas contributions leading to different states will add up incoherently. Another school of thought has been promoted by Cowley [8] who, along the lines of the pioneering work of Gabor [9], argues that interference between an inelastically scattered electron wave and a reference wave is still possible provided the energy loss in the object is smaller than the energy spread of the incident beams. The original reasoning in Ref. [8] which was based on optical arguments (Van Cittert–Zernicke theorem) was later confirmed in Ref. [10] by a quantum-mechanical approach using time-dependent perturbation theory to describe the inelastic process.

In discussions about coherence (and wave mechanics in general) it is always dangerous to rely on intuitive arguments (exchange of energy, time of interaction, etc.). Also, optical arguments may not necessarily be valid in quantum mechanics.

For this reason we will start in this work from the most general approach, which is inspired by the treatment of inelastic electron diffraction in crystals by Yoshioka in 1957 [11]. The principles of our approach have already been described briefly in Ref. [12]. Energy exchanges are always described quantum-mechanically by an Hamiltonian. Therefore, we can only investigate the balance between energy exchanges between electron, object, and source properly if they are all described within one global Hamiltonian.

2. Theory

2.1. General framework

Consider a global system consisting of an electron, with position vector \( r \), an object with particle vectors \( r_i \), and a source with particles at \( r_s \). In case the system is isolated, the wave function describing this system obeys the Schrödinger equation

\[
\frac{i}{\hbar} \frac{\partial \Psi(r, r_i, r_s, t)}{\partial t} = H(r, r_i, r_s) \Psi(r, r_i, r_s, t) \tag{1}
\]

with \( \Psi(r, r_i, r_s, t) \) being the wave function of the system and \( H(r, r_i, r_s) \) the Hamiltonian of the system.

Since the system is isolated, the Hamiltonian is independent of time. We can now expand the wave function in eigenfunctions of the Hamiltonian since these form an orthonormal basis (if no degeneracy occurs), i.e.

\[
\Psi(r, r_i, r_s, t) = \sum_E C_E(t) \psi_E(r, r_i, r_s) \tag{2}
\]

with

\[
H(r, r_i, r_s, t) = \sum_E \psi_E^*(r, r_i, r_s) E \psi_E(r, r_i, r_s). \tag{3}
\]

\( E \) is the energy of the system. The time dependence of \( \psi \) is in the coefficients \( C_E(t) \). Substituting Eq. (2) into Eq. (1) then yields (in shorthand notation)

\[
\frac{i}{\hbar} \sum_E \frac{dC_E(t)}{dt} \psi_E = \sum_E C_E(t) \psi_E \tag{4}
\]

and using the orthonormality of the \( \psi_E \)

\[
\frac{dC_E(t)}{dt} = EC_E(t) \tag{5}
\]

or

\[
C_E(t) = C_E(0)e^{E^t/\hbar} \tag{6}
\]
so that Eq. (2) becomes

$$\psi(r, r_1, r_2, t) = \sum_E C_E(0)e^{-iEt/h}\psi_E(r, r_1, r_2).$$  \hfill (7)

The coefficients can be found from

$$C_E(0) = \langle \psi_E | \Psi(0) \rangle.$$  \hfill (8)

The total wave function is then an interference between the eigenstates with different energies. Note that this is a consequence of the fact that the system is isolated and the energy is not known. The probability density to find particles at respective positions \((r, r_1, r_2)\) and time \(t\) is given by

$$p(r, r_1, r_2, t) = |\psi(r, r_1, r_2, t)|^2$$  \hfill (9)

or using Eq. (7)

$$p(r, r_1, r_2, t) = \sum_{E,E'} C_E(0)C_{E'}(0)e^{-i(E-E')t/h}$$

$$\psi_E(r, r_1, r_2)\psi_{E'}(r, r_1, r_2).$$  \hfill (10)

In practice, one only detects the electrons, i.e. one detects the probability \(p(r)\) that electrons hit the position \(r\) where \(r\) can be in the place of the image, the hologram, the diffraction pattern, etc. This probability is

$$p(r) = \int p(r, r_1, r_2) \, dr_1 \, dr_2 \, dt,$$  \hfill (11)

where one integrates over all possible object states, source states and the integration time of the experiment. We now have from Eq. (10)

$$p(r) = \sum_{E,E'} C_E(0)C_{E'}(0)\text{sinc}\left(\frac{(E-E')T}{h}\right) p_{E,E}(r)$$  \hfill (12)

with

$$p_{E,E}(r) = \int \psi_E(r, r_1, r_2)|\psi_{E'}(r, r_1, r_2)| \, dr_1 \, dr_2,$$  \hfill (13)

where the sinc function is defined as

$$\text{sinc}(x) = \frac{\sin \pi x}{\pi x}.$$  \hfill (14)

For simplicity we have assumed that the experiment took place at time \(t = 0\) over a time interval \(T\), i.e. between \(-T/2\) and \(T/2\) (generalisation is straightforward). The sinc function is a peaked function centered around the origin with a peak width of the order of \(|x| \approx 1\), i.e. interference of states with different energies is significant when the energy difference is smaller than

$$|E - E'| < \frac{\hbar}{T}.$$  \hfill (15)

For a typical recording time of \(1\) s the sinc function makes that the energy difference should be of the order of

$$|E - E'| \lesssim 10^{-15} \text{ eV}.$$  \hfill (16)

Integration over an infinite time would give an exact energy conservation. An energy difference of \(10^{-15} \text{ eV}\) is much less than, e.g. the energy of the lowest wave vector acoustical phonon and the distance between energy levels in the source so that, for all practical purposes, energy conservation holds (see discussion).

In case all energies in Eq. (12) have differences larger than this value one has

$$p(r) \approx T \sum_E |C_E(0)|^2 \int |\psi_E(r, r_1, r_2)|^2 \, dr_1 \, dr_2$$  \hfill (17)

**Note:** In case the system is in thermal equilibrium with the environment, it is in the so-called mixed state. Then Eq. (17) \(|C_E(0)|^2\) still holds with the Boltzmann factor of the energy \(E\).

Let us now calculate \(\psi_E\) explicitly from Eq. (3):

$$H(r, r_1, r_2)\psi_E(r_1, r_2) = E\psi_E(r_1, r_2),$$  \hfill (18)

where the Hamiltonian can be written without loss of generality as

$$H(r, r_1, r_2) = H_s(r) + H_{S,C}(r_1, r_2) + H_{im}(r, r_1, r_2).$$  \hfill (19)

\(H_s\) is the Hamiltonian of the free electron

$$H_s(r) = -\frac{\hbar^2}{2m} \Delta,$$  \hfill (20)

and \(\Delta\) is the Laplacian operator, describing the kinetic energy of the free electron.

For convenience, we have considered the source and the object (e.g. crystal) as one system which is described by the Hamiltonian \(H_{S,C}(r_1, r_2)\). The interaction of the electron with this system is described by \(H_{im}(r, r_1, r_2)\).
We can now, without any loss of generality, expand the wave function in eigenstates of the Hamiltonian $H_{S,C}$ since these form a complete (and orthogonal) basis for all functions of $(r_i, r_s)$, i.e.

$$
\psi_E(r, r_i, r_s) = \sum_m \phi_m^E |a_m^{S,C}(r_i, r_s)\rangle,
$$

(21)

where the eigenstates satisfy

$$
H_{S,C}(r_i, r_s)|a_m^{S,C}(r_i, r_s)\rangle = E_m^{S,C} |a_m^{S,C}(r_i, r_s)\rangle.
$$

(22)

From Eq. (13) we have now

$$
p_{E,E}(r) = \sum_{nm} \phi_n^E(r) \phi_m^{E'}(r) \langle a_m^{S,C}(r_i, r_s) | a_n^{S,C}(r_i, r_s) \rangle
$$

(23)

where the bracket product implicitly contains integration over the variables $(r_i, r_s)$ which in view of the orthogonality of the eigenstates

$$
\langle a_m | a_n \rangle = \delta_{nm}
$$

(24)

yields

$$
p_{E,E}(r) = \sum_{nm} \phi_n^E(r) \phi_m^{E'}(r)
$$

(25)

and for the total recorded intensity from Eq. (12)

$$
p(r) = T \sum_{E,E'} C_E(0) C_{E'}^*(0) \mu \frac{(E - E')T}{\hbar}
$$

$$
\times \sum_n \phi_n^E(r) \phi_n^{E'}(r).
$$

(26)

(27)

It is also convenient to rewrite Eq. (26) as

$$
p(r) = T \sum_E |C_E(0)|^2 \sum_n |\phi_n^E(r)|^2
$$

$$
+ T \sum_{E \neq E'} C_E(0) C_{E'}^*(0) \mu \frac{(E - E')T}{\hbar}
$$

$$
\times \sum_n \phi_n^E(r) \phi_n^{E'}(r).
$$

(28)

We will now focus on the computation of $\phi_n^E(r)$. By substituting Eq. (21) into Eq. (18) using Eq. (20) we find

$$
[H_e(r, r_i, r_s) + H_{S,C}(r_i, r_s) + H_{int}(r, r_i, r_s)]
$$

$$
\times \sum_n \phi_n^E(r) |a_n^{S,C}(r_i, r_s)\rangle
$$

$$
= E \sum_n \phi_n^E(r) |a_n^{S,C}(r_i, r_s)\rangle
$$

(29)

multiplying with $|a_m^{S,C}(r_i, r_s)\rangle$ and integrating over $r_i, r_s$ yields the orthogonality relation and using Eq. (22)

$$
H_e(r) \phi_m^E(r) + E_m^{S,C} \phi_m^E(r) + \sum_{n} z_{mn}(r) \phi_m(r) = E \phi_m^E(r)
$$

(30)

with

$$
z_{nm}^{S,C} = \langle a_m^{S,C}(r_i, r_s) | H_{int}(r, r_i, r_s) | a_n^{S,C}(r_i, r_s) \rangle
$$

(31)

which also implicitly contains integration over all $r_i, r_s$.

The interpretation of Eq. (30) is simple. $H_e$ represents the kinetic energy of the electron. The term $\sum_n z_{mn}(r) \phi_m(r)$ is the potential energy of the electron in the field of the $S, C$ system (source and object). $z_{mn}(r)$ is the matrix element for the transition of the electron from state $\phi_n$ to state $\phi_m$ by interaction with the $S, C$ system. Due to this interaction the $S, C$ system changes from state $|a_n\rangle$ to state$|a_m\rangle$. $E - E_m^{S,C}$ expresses the total energy minus the energy of the $S, C$ system, i.e. the net energy $E$ of the electron

$$
E - E_m^{S,C} = E_i.
$$

(32)

Hence, Eq. (30) describes all possible elastic and inelastic interactions of the electron with the $S, C$ system. In practice, we now have to compute $\phi_m(r)$ using Eq. (30) and substitute it into Eq. (27) so as to obtain the measured electron probability density.

Note that, in case of off-axis holography, the reference wave, which does not interact with the object is automatically included in the elastic wave so that the first term in Eq. (27) also includes the elastic holographic fringes. Possible inelastic interference is included in the second term of Eq. (27).

If we make the reasonable assumption that the object and source are mutually independent (i.e. they do not interact) we can show that (see the appendix)

$$
|a_n^{S,C}(r_i, r_s)\rangle |a_p^e(r_i)\rangle |a_q^e(r_s)\rangle.
$$

(33)
This means that the state \( n \) of the \( S, C \) system can be labelled by the state \( p \) of the object and the state \( q \) of the source. For the energy we then have

\[
E_n^{S,C} = E_p^C + E_q^S
\]

and for the interaction potential

\[
z_{nm}^{S,C}(r) = z_{r,p}^C(r)\delta_{s,q} + z_{s,q}^S(r)\delta_{r,p}
\]

with

\[
m = r, s
\]
\[
n = p, q.
\]

2.2. Interpretation

Eqs. (27) and (30) are the main results of this work. Their interpretation is as follows:

For convenience we will assume that object and source are independent but the discussion is more generally valid.

The electron wave function is \( \phi_n^E(r) \) when the object is in the state \( r \) with energy \( E_r^C \) and the source in the state \( s \) with energy \( E_s^S \). The total energy is \( E \) and the net electron energy is \( E - E_r^C - E_s^S \). The electron can change state from \( p, s \) to \( r, s \) by inelastic interaction with the object and from \( r, q \) to \( r, s \) by inelastic interaction with the source.

The interaction functions \( z_{r,p}^C(r) \) and \( z_{s,p}^S(r) \) are confined to the object and source, respectively, and do not overlap. In case the energy of the object is not changed \( \phi_n^E \) describes the elastic the elastic wave which, in case of electron holography, contains both the reference wave and the elastic object wave.

The first term in Eq. (27) describes the intensity at position \( r \) in the detector plane resulting from interference of all waves with exactly the same energy \( E \). This term describes as well the contrast of an hologram, or the contrast of an (elastic) HREM image. Even interference between inelastic waves is included in this term provided they have exactly the same energy \( E \). This happens for instance when in off-axis holography both the reference wave and the object wave pass through the object and generate the same inelastic event (e.g. plasmon). In this way, interference can be observed even in inelastically filtered images [5] and even the “cross-section” of a plasmon can be estimated [6].

Let us now address the question whether a change in energy in the source can be balanced by an energy change in the object so as to restore the coherence (interference).

In the second term of Eq. (27) such interference between states of different total energies is possible. The interference is only significant if the difference in energy balance should be smaller than that given by Eq. (15), i.e.

\[
|E - E'| < \frac{\hbar}{t}
\]

i.e. of the order of \( 10^{-15} \) eV. The importance of this contribution is then given by \( C_E \) and \( C_{E'} \) i.e. by the density of states within the energy interval. Note that in the case of thermal equilibrium, i.e. when the total system is in the mixed state, Eq. (17) holds, i.e. only the first term in Eq. (27) remains and interference would not be present. Inelastic holography may thus provide an interesting experiment about this issue.

3. Discussion

In principle, interference between inelastically scattered electrons even with different energies is possible provided the energy difference is smaller than \( h/t \). However, it is clear from Eq. (27) that the ratio of inelastic/elastic contribution to the interference fringes is of the order

\[
R \approx \sum_{E \neq E'}\frac{C_{E}(0)C_{E'}(0)}{\sum_{E}|C_E(0)|^2}
\]

with \( |C_E(0)|^2 = p(E) \) the probability that a state with this energy occurs in the spectrum. Note that, since the wave function \( \phi_n^E \) is different for different values of \( E \), many terms in Eq. (27) may cancel out due to destructive interference so that Eq. (38) is in fact an upper limit.

We will now try to estimate the order of magnitude of \( R \). Remember that we have from Eqs. (32) and (34) for the total energy

\[
E = E_n^S + E_p^C + E_e^S
\]

It is clear that for a finite object the object states are discrete and for a finite source the source states are
also discrete. Hence the energies $E_p^S$ and $E_q^C$ are quantised. The electron energy has a probability which, due to fluctuation in the accelerating voltage, is spread around its average value (e.g. 300 keV). However, contrary to what one would expect intuitively, also the electron energy $E_e$ is quantised. Indeed, Eq. (30) has to be solved between two boundary conditions. In the case of a reference wave we have extra boundaries at entrance and exit faces of the object. Now the confinement of the electron wave into the finite length between these boundaries causes the possible “free” electron waves to be discrete and hence the energy $E_e$ quantises. The energy difference between two successive states can be estimated from an argument put forward in Ref. [13] and discussed in Ref. [10].

Denoting by $L$ the length of the microscope (distance source–detector) and by $\lambda$ the electron wavelength, one has

$$\Delta E_e^S = \frac{\lambda}{2L}.$$  \hspace{1cm} (40)

Using typical values $L = 1$ m, $\lambda = 2\times10^{-12}$ m, and $E = 3\times10^5$ eV one has $\Delta E_e^S = 3\times10^{-7}$ eV.

The lowest energy changes in the object are caused by electron–phonon interactions. The lowest energy levels in the object are then those corresponding with low-vector acoustic phonons from which it can be estimated that the smallest energy differences are of the order of $\Delta E_e^C \approx 10^{-3}$ eV.

The energy difference of the source states depends on the effective size of the source and of the emission process. For instance, if the electron is extracted by a field, the different states of the source correspond with the electronic states in the conduction band. If the effective size of the source is small, the electrons are confined to a small volume and hence, following the same argument as above, have discrete states. The energy difference between these states is then inversely related to the effective size of the emitting tip. For a small source (as required for high coherence) such an energy difference could be of the order of $\Delta E_e^S \approx 10^{-3}$ eV. Now the total state of the whole system (source, object and electron) can be labelled by three quantum numbers, say $p, q, u$ with a total energy $E$

$$E = p\Delta E_e^S + q\Delta E_e^C + u\Delta E_e.$$  \hspace{1cm} (41)

Let us now estimate the probability that one can find states which differ less in energy than $\hbar/\ell$ and hence will yield significant interference. A lower limit for the smallest energy difference between different states is given by $\Delta E_e = 3\times10^{-7}$ eV since this value is much smaller than $\Delta E_e^S$ and $\Delta E_e^C$. Now, the ratio inelastic/elastic interference (38) is equal to the probability that a state finds a neighbouring state within the energy window of $10^{-15}$ eV so that in this case

$$R = \frac{3\times10^{-7}}{10^{-15}} = 3\times10^{-8}.$$  \hspace{1cm} (42)

We can rephrase this in terms of the hypothetical holographic experiment discussed in Ref. [10] and sketched in Fig. 1. Hence the energy spectra of the source (i.e. of the reference waves and the elastic waves) have an average energy $E_0$ and energy spread as shown in Fig. 2. Due to a single inelastic process with energy loss $\delta E$ the whole spectrum is shifted over $\delta E$ (Fig. 2). As argued in Ref. [10] interference is possible in the overlap area but as shown in this work this can only happen between states in the overlap area that differ by less than $10^{-15}$ eV in energy which is far below the average energy separation of the quantised states.

We can thus conclude that although inelastic interference is possible the effect is so small that it cannot be observed experimentally since this would
require an ideal energy filter with about $10^{-3}$ eV energy resolution (to eliminate elastic interference) and an unreasonably large recording time. Hence, holography is still de facto a perfect energy filter.

Appendix A

If the object and the source are independent of each other, i.e. they do not interact, we have for the Hamiltonian

$$H_{S,C}(r_i, r_s) = H_C(r_i) + H_S(r_s), \quad (A.1)$$

i.e. only the particles $r_i$ can interact mutually as well as the particles $r_s$, but no interaction is present between particles $r_i$ and $r_s$, respectively. Let us now call the eigenstates of both Hamiltonians

$$H_C(r)|a_p^c(r_i)\rangle = E_p^c|a_p^c(r_i)\rangle, \quad (A.2)$$

$$H_S(r_s)|a_q^s(r_s)\rangle = E_q^s|a_q^s(r_s)\rangle, \quad (A.3)$$

$$H_{S,C}(r_i, r_s)|a_p^c(r_i)\rangle|a_q^s(r_s)\rangle = (E_p^c + E_q^s)|a_p^c(r_i)\rangle|a_q^s(r_s)\rangle, \quad (A.4)$$

i.e. the eigenstates of $H_{S,C}$ are now products of eigenstates of $H_C$ and $H_S$ (this is generally a known result in quantum mechanics).

We can thus write

$$|a_p^S(r_i, r_s)\rangle = |a_p^c(r_i)\rangle|a_q^s(r_s)\rangle \quad (A.5)$$

and

$$E_n^S = E_p^c + E_q^s \quad (A.6)$$

the state $n$ of the $S, C$ system can thus be labelled by the state $p$ of the object and the state $q$ of the source, i.e.

$$n = (p, q). \quad (A.7)$$

Hence, the electron state too can be relabelled as

$$\phi_n^E(r) = \phi_{p,q}^E(r). \quad (A.8)$$

For the matrix element $z_{mn}^{S,C}$ we have

$$n = (p, q),$$

$$m = (r, s)$$

calling

$$z_{mn}^{S,C} = \langle a_m^{S,C}(r_1, r_2)|H_{S,C}(r, r_1, r_s)|a_n^{S,C}(r_1, r_s)\rangle \quad (A.10)$$

$$= \langle a_m^{S,C}(r_1, r_2)|H_{S,C}(r, r_1, r_s)|a_n^{S,C}(r_1, r_s)\rangle \quad (A.11)$$

$$= \langle z_{r_1}^c(r_1)|\langle a_n^{S,C}(r_2)|H_{S,C}(r, r_1, r_s)|a_n^{S,C}(r_1)\rangle\rangle. \quad (A.12)$$

Since source and object are assumed to be independent we can now write

$$H_{S,C}(r, r_1, r_s) = H_C(r, r_1) + H_S(r, r_s), \quad (A.13)$$

i.e. all interactions are described between electron and object $(r, r_1)$, electron and source $(r, r_s)$ but not between object and source $(r_1, r_s)$.

Substituting Eq. (A.13) into Eq. (A.12) then yields

$$z_{mn}^{S,C} = \langle a_m^{S,C}(r_1)\rangle\langle a_n^{S,C}(r_2)|H_C(r, r_1)$$

$$+ H_S(r, r_s)|a_n^{S,C}(r_1)\rangle\rangle. \quad (A.14)$$

we now have the orthogonality relations

$$|a_r^c(r_1)\rangle|a_r^c(r_1)\rangle = \delta_{rp}, \quad (A.15)$$

$$|a_r^s(r_2)\rangle|a_r^s(r_2)\rangle = \delta_{sq} \quad (A.16)$$

calling further

$$z_{r,p}^c(r) = \langle a_r^c(r)|H_C(r, r_1)|a_r^c(r_1)\rangle, \quad (A.17)$$

$$z_{s,q}^s(r) = \langle a_r^s(r)|H_S(r, r_s)|a_r^s(r_1)\rangle. \quad (A.18)$$

We then obtain from Eq. (A.14)

$$z_{mn}^{S,C} = z_{r,p}^c(r)\delta_{sq} + z_{s,q}^s(r)\delta_{rp}. \quad (A.19)$$

References

