Title

PART I: INTENSITY CORRELATIONS IN ELASTIC SCATTERING EXPERIMENTS. PART II: THEORY OF QUANTUM BEAT AND LEVEL CROSSING EXPERIMENTS UTILIZING ELECTRONIC EXCITATION

Author

Kelly, Robert L.

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Ernest O. Lawrence Radiation Laboratory

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Contents

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PART II: THEORY OF QUANTUM BEAT AND LEVEL CROSSING EXPERIMENTS UTILIZING ELECTRONIC EXCITATION*

Robert L. Kelly

Department of Physics and Lawrence Radiation Laboratory
University of California
Berkeley, California

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ABSTRACT OF PART I

Measurements of space-time correlations in the intensity of a beam of scattered particles yield information about the scatterer. Goldberger and Watson have developed a theory of such measurements appropriate to the case of nearly elastic scattering; in this paper we extend their theory to the case of inelastic scattering. The main result of our work is that under certain experimental conditions the correlated counting rate in an inelastic scattering experiment is of a form equivalent to the correlated counting rate for an incoherent beam of particles emitted by a source. This result applies, in particular, to Raman or Brillouin scattering of light by phonons in an extended target. However, estimates of the signal to noise ratio for this case indicate that it may not be possible to observe intensity correlations in Raman or Brillouin lines using presently available light sources.

ABSTRACT OF PART II

The coherent excitation of several atomic status by inelastic electron scattering and their subsequent radiative decay is considered. General expressions for the photon counting rate in a quantum beat experiment, and the total number of photons counted in a level crossing experiment are derived. The general results are used to calculate the phase of the oscillatory part of the photon counting rate in the Hadeishi-Nierenberg quantum beat experiment.

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* A slightly different version of this work has been published previously. R. L. Kelly, Phys. Rev. 147, 376 (1966).

PART I

I. INTRODUCTION

A quantum mechanical theory of intensity correlation experiments has recently been developed by Goldberger, Lewis, and Watson.[1-5] The experiments considered by these authors involve the detection of correlations between measurements of the intensity of a beam of particles made at two different space-time points. Intensity correlation measurements on a beam of scattered particles give information about the target. Goldberger and Watson[3] (hereafter referred to as GW) have developed a theory of such experiments appropriate to the case of nearly elastic scattering. In this paper we consider correlations in the intensity of a beam which has been inelastically scattered by a many-particle target.

The results of GW for intensity correlations in a beam which is emitted by a radiating source can be reproduced by the following heuristic "derivation." For definiteness we consider the case of a beam of photons. The amplitude for emission of a photon with wave vector k at a point $\underline{s}_1$ in the source at time t_1 and subsequent detection at a point $\underline{Y}_a$ at time t_a is,

$$A(k,\underline{s}_1,t_1;\underline{Y}_a,t_a) = A(k)\frac{\exp\{i[k|\underline{Y}_a - \underline{s}_1| - c\,k(t_a - t_1)]\}}{|\underline{Y}_a - \underline{s}_1|} .$$

The amplitude for emission of two photons of wave vector k at points $\underline{s}_1$ and $\underline{s}_2$ at times t_1 and t_2 and their subsequent detection at points $\underline{Y}_a$ and $\underline{Y}_b$ at times t_a and t_b is then

$$A(k, \underline{s}_1, t_1, \underline{s}_2, t_2;\ \underline{Y}_a, t_a, \underline{Y}_b, t_b)$$

$$= A(k, \underline{s}_1, t_1;\ \underline{Y}_a, t_a)\, A(k, \underline{s}_2, t_2;\ \underline{Y}_b, t_b)$$

$$+ A(k, \underline{s}_2, t_2;\ \underline{Y}_a, t_a)\, A(k, \underline{s}_1, t_1;\ \underline{Y}_b, t_b)$$

where we have symmetrized the amplitude in accordance with Bose-Einstein statistics. The probability of such a double measurement is,

$$P(k, \underline{s}_1, t_1, \underline{s}_2, t_2;\ \underline{Y}_a, t_a, \underline{Y}_b, t_b)$$

$$= |A(k, \underline{s}_1, t_1, \underline{s}_2, t_2;\ \underline{Y}_a, t_a, \underline{Y}_b, t_b)|^2$$

$$= 2\ \frac{|A(k)|^4}{Y_a^{\,2}\, Y_b^{\,2}} + \Big\{ A^*(k, \underline{s}_1, t_1;\ \underline{Y}_a, t_a)\, A(k, \underline{s}_1, t_1;\ \underline{Y}_b, t_b)$$

$$A^*(k, \underline{s}_2, t_2;\ \underline{Y}_b, t_b)\, A(k, \underline{s}_2, t_2;\ \underline{Y}_a, t_a) + \text{complex conjugate} \Big\}$$

where we have put $|\underline{Y}_a - \underline{s}| \doteq Y_a$ and $|\underline{Y}_b - \underline{s}| \doteq Y_b$ in the denominator. The total probability of observing two photons of wave vector k at $\underline{Y}_a, t_a$ and $\underline{Y}_b, t_b$ is obtained by integrating $\underline{s}_1$ and $\underline{s}_2$ over the source volume, V_s.

$$P(k;\underline{Y}_a, t_a, \underline{Y}_b, t_b)$$

$$= (\text{const.}) \int_{V_s} \frac{d^3s_1 \, d^3s_2}{V_s^{\,2}} \, P(k, \underline{s}_1, t_1, \underline{s}_2, t_2; \underline{Y}_a, t_a, \underline{Y}_b, t_b)$$

$$= (\text{const.}) \frac{2|A(k)|^4}{Y_a^{\,2} \, Y_b^{\,2}} \left(1 + \left| \int_{V_s} \frac{d^3s}{V_s} \right.\right.$$

$$\left.\left. \exp\{i[k(|\underline{Y}_b - \underline{s}| - |\underline{Y}_a - \underline{s}| - c\,k\,(t_b - t_a))]\} \right|^2 \right).$$

Finally, the probability of making such a measurement on a photon of arbitrary energy is obtained by integrating over the emission spectrum $|A(k)|^2$.

$$P(\underline{Y}_a, t_a, \underline{Y}_b, t_b) = \int dk \, \rho(k) \, P(k;\underline{Y}_a, t_a, \underline{Y}_b, t_b)$$

where $\rho(k)$ is an appropriate spectral weight function.

Two important points must be mentioned about this result. Intensity correlation effects are seen to arise from interference between the amplitude $A(k, \underline{s}, t; \underline{Y}_a, t_a)$ and $A(k, \underline{s}, t; \underline{Y}_b, t_b)$. We have squared the amplitude $A(k, \underline{s}_1, t_1, \underline{s}_2, t_2; \underline{Y}_a, t_a, \underline{Y}_b, t_b)$ before integrating over $\underline{s}_1$ and $\underline{s}_2$ rather than integrating first and then squaring the total amplitude to find $P(k;\underline{Y}_a, t_a, \underline{Y}_b, t_b)$. For this reason there is no interference between $A(k, \underline{s}_1, t_1; \underline{Y}, t)$ and $A(k, \underline{s}_2, t_2; \underline{Y}, t)$ where $\underline{s}_1 \neq \underline{s}_2$. This is a valid procedure if the source is large compared to any characteristic correlation lengths in it, so that different parts of the source radiate incoherently. Similarly, there is no interference between amplitude for emission of photons of different wave vector. This is valid if the beam is temporally incoherent, i.e., the emission spectrum is time independent.

The primary result of the present paper is that when double measurements are made on a beam of scattered light (or massive particles) the form of $P(\underline{Y}_a, t_a, \underline{Y}_b, t_b)$ is entirely similar to the above expression for the case of emitted light (or the appropriate expression for massive particles). The only difference is that the source volume is replaced by the target volume and that $|A(k)|^2$ represents the scattered spectrum at a certain scattering angle rather than an emission spectrum. There are two important restrictions on the types of targets we consider. The first is that the target is small enough so that multiple scattering may be neglected. The second is that a particular form of the impulse approximation is valid. We write our results in a way which allows the usual kind of impulse approximation (neglect of the binding of particles in the target) to be applied, but no use is made of this except when we compare our results with those of GW. The quite unrestrictive form of the impulse approximation that is used may be described as follows. It is assumed that there exists a correlation length, ℓ_c, such that two parts of the target separated by a distance much larger than ℓ_c scatter incoherently. The target is then imagined to be partitioned into a number of volume elements each of which is large compared to ℓ_c^3 but small enough to satisfy the conditions for Fraunhofer scattering (Eqs. (39) and (51)). It is then assumed that the impulse approximation may be applied to each of these volume elements, i.e., the binding of a particular volume element to the rest of the target is neglected when calculating the scattered intensity arising from that volume element. This is pointed out in the remarks following Eq. (30) and in the paragraph preceeding Eq. (54).

There are certain experimental conditions that must be fulfilled in order for our result to be valid. The most important of these is that the angular separation of $\underline{Y}_a$ and $\underline{Y}_b$ must satisfy a certain criterion of smallness. Unlike light from an isotropically radiating source, scattered light will in general have an energy spectrum which depends on the angle of observation. We will require the angle between $\underline{Y}_a$ and $\underline{Y}_b$ to be small enough so that both are characterized by the same energy spectrum. We do not investigate the potentially interesting case of wide angle intensity correlations in which $\underline{Y}_a$ and $\underline{Y}_b$ do not satisfy this criterion.

In Section II we give a preliminary description of the type of scattering experiment under consideration. The characteristics of the source of incident particles, the incident beam, and the target are discussed. The scattering cross-section of the target is written in a form which is useful for later applications. In Section III we derive the correlated counting rate for a beam of inelastically scattered particles, and compare it with the result of GW for the case of almost elastic scattering. Applications to Raman and Brillouin scattering are briefly considered.

In the main body of this paper we do not always give credit to GW when we use their techniques; this would require too many footnotes. However, our work is, to a large extent, based on that of GW, and much of what is said here is also contained in their paper. We attempt to stick to their notation as closely as possible.

II. PRELIMINARY DESCRIPTION OF EXPERIMENT

We consider a scattering experiment in which an incoherent incident beam is directed onto a many particle target. The wave function of the beam at points between the target and the source is,

$$\Phi_{in} = \mathcal{S} \prod_{i=1}^{N} \phi_i(\underline{x}_i, t) \, . \tag{1}$$

Here $\mathcal{S}$ is the projection operator onto symmetrized or anti-symmetrized states, and N is the total number of particles emitted by the source during the experiment. The wave functions of the individual particles are taken to be of the form $(\hbar = 1)$,

$$\begin{aligned} \phi_i(\underline{x}_i, t) &= 0, \; t < t_i \\ \phi_i(\underline{x}_i, t) &= \int dq \, a_i(q) \, \frac{\exp\{i[q|\underline{x}_i - \underline{d}_i| - \epsilon(q)(t - t_i)]\}}{|\underline{x}_i - \underline{d}_i|} \, u_i(s_i); \; t > t_i \end{aligned} \tag{2}$$

where

$$\begin{aligned} \epsilon(q) &= q^2/2m \quad \text{(non-relativistic particles)} \\ \epsilon(q) &= (c^2q^2 + m^2 c^4)^{1/2} \quad \text{(relativistic particles)} \\ \epsilon(q) &= c\, q \quad \text{(photons)} \, . \end{aligned} \tag{3}$$

and $u_i(s_i)$ is the spin wave function. The dependence of $\phi_i(\underline{x}_i, t)$ on s_i will be left implicit. $\phi_i(\underline{x}_i, t > t_i)$ is normalized to unity, and wave functions with different indices are assumed orthogonal. ϕ_i describes a particle which is emitted in an outgoing s-wave wave packet at the time t_i and location $\underline{d}_i$. These parameters, t_i and $\underline{d}_i$, are introduced phenomenologically in order to represent the incoherence of the beam. Their physical interpretation depends on the type of source under consideration. For a thermal source of massive particles, for example, they should be interpreted as the time and place of the last collision suffered by a particle before leaving the source.

The emission times lie within the range,

$$T_1 < t_i < T_1 + T_0 \quad . \tag{4}$$

That is, the source is turned on at time T_1 and turned off at time $T_1 + T_0$. T_0 is a macroscopic time (the duration of the experiment). In order to take collimation of the source into account we restrict $\underline{d}_i$ to lie in that part of the source that can "see" the target through the collimation apparatus. From now on when we speak of the "target" we will mean that part of the actual target which is intercepted by the actual incident beam. Thus the part of $\phi_i(\underline{x}_i, t)$ which lies outside the incident beam does not interact with the "target" and is not scattered so we need not take explicit account of the fact that this part of ϕ_i is removed by the collimating apparatus. Energy resolution in the incident beam will be accounted for by defining an energy spectrum in terms of the wave packet amplitudes $a_i(q)$.

We will choose an origin of coordinates which lies in the target and assume that the average density of the incident beam is uniform over the target. This average density is,

$$n = N \left\langle \sum_{s_i} |\phi_i(0,t)|^2 \right\rangle \tag{5}$$

where $T_1 < t < T_1 + T_0$ and the brackets indicate that averages over $a_i(q)$, $\underline{d}_i$, and t_i are to be taken. We will assume that the characteristics $a_i(q)$, $u_i(s)$, $\underline{d}_i$, and t_i of each wave packet are statistically independent of each other and of the characteristics of other wave packets. Further, we assume that the magnitude d_i and the direction $\hat{d}_i$ of $\underline{d}_i$ are statistically independent. Finally, we assume all incident wave packets to be statistically equivalent. Substituting Eq. (2) into Eq. (5) we obtain,

$$n = N \int dp\, dq \;\langle a_i^*(p)\, a_i(q)\rangle \left\langle \frac{e^{i(p-q)d_i}}{d_i^{\,2}} \right\rangle \left\langle e^{-i[\epsilon(q)-\epsilon(p)](t-t_i)} \right\rangle \tag{6}$$

where

$$\langle a_i^*(p) a_i(q)\rangle = \frac{1}{N} \sum_{i=1}^{N} a_i^*(p)\, a_i(q) \tag{7}$$

$$\left\langle \frac{e^{i(p-q)d_i}}{d_i^{\,2}} \right\rangle = \frac{1}{N} \sum_{i=1}^{N} \frac{e^{i(q-p)d_i}}{d_i^{\,2}} \tag{8}$$

$$\left\langle e^{-i[\epsilon(q)-\epsilon(p)](t-t_i)} \right\rangle = \frac{1}{N} \sum_{i=1}^{N} e^{-i[\epsilon(q)-\epsilon(p)](t-t_i)} \, \theta(t - t_i) \qquad (9)$$

$$\theta(t - t_i) = \begin{cases} 1, & t_i < t \\ 0, & t_i > t \end{cases} . \qquad (10)$$

The step function in Eq. (9) takes account of the fact that $\phi_i(\underline{x}_i, t)$ vanishes if $t < t_i$. If the distribution of emission times is uniform (9) may be replaced by,

$$\left\langle e^{-i[\epsilon(q)-\epsilon(p)](t-t_i)} \right\rangle = \frac{1}{T_0} \int_{T_1}^{t} dt' \; e^{-i[\epsilon(q)-\epsilon(p)](t-t')} . \qquad (11)$$

During most of the experiment $t - T_1$ will be much larger than any other relevant quantity with the dimensions of time. We utilize this fact by taking the average of Eq. (11) in the limit that $t - T_1 \to \infty$.

$$\left\langle e^{-i[\epsilon(q)-\epsilon(p)](t-t_i)} \right\rangle = \frac{1}{T_0} \lim_{t-T_1 \to \infty} \int_{T_1}^{t} dt' \; e^{-i[\epsilon(q)-\epsilon(p)](t-t')}$$

$$= \frac{\pi}{T_0} \, \delta[\epsilon(q) - \epsilon(p)] + \frac{i}{T_0} \, \frac{P}{\epsilon(p) - \epsilon(q)} . \qquad (12)$$

For $p \neq q$ the quantity $a_i^*(p) \, a_i(q)$ will have a non-zero phase, and this phase will have a random dependence on i. Thus we expect $\langle a_i^*(p) \, a_i(q) \rangle$ to vanish for $p \neq q$ so that the principle part in (12) may be neglected. This will be done here and in the future.

n now becomes,

$$n = \pi R \left\langle \frac{1}{d^2} \right\rangle \int d\, q \quad \frac{\langle |a(q)|^2 \rangle}{v(q)} \tag{13}$$

where

$$v(q) = \frac{d\epsilon(q)}{dq} \tag{14}$$

and

$$R = \frac{N}{T_0} \tag{15}$$

is the average rate at which particles are emitted from the source. In a similar way the incident current at the target is found to be

$$\underset{\sim}{\mathcal{J}} = \pi R \langle -\hat{d} \rangle \left\langle \frac{1}{d^2} \right\rangle \int dq \, \langle |a(q)|^2 \rangle \quad . \tag{16}$$

We now turn our attention to the average energy and momentum of the beam particles. The average energy is,

$$\langle \epsilon \rangle = \left\langle \left(\phi_i(\underline{x}_i, t) \; |K_i| \; \phi_i(\underline{x}_i, t) \right) \right\rangle \tag{17}$$
$$= \left\langle 4\pi \int dx' dp \; dq \; a_i^*(p) a_i(q) \epsilon(q) \; \exp \left\{ i \left[(q - p)x' - [\epsilon(q) - \epsilon(p)](t - t_i) \right] \right\} \right\rangle$$

where K_i is the Hamiltonian of the $\underline{i}$th beam particle and $x' = |\underline{x}_i - \underline{d}_i|$. The integral over x' yields,

$$\int_0^\infty dx' \; e^{i(q-p)x'} = \pi \, \delta(q - p) + i \; \frac{P}{q - p} \quad . \tag{18}$$

The contribution to $\langle\epsilon\rangle$ of the principal part in (18) vanishes,

$$4\pi i\int dp\, dq\, \langle |a(q)|^2\rangle\, \epsilon(q)\, \frac{P}{q-p}\, \frac{\pi}{T_0}\, \delta[\epsilon(p)-\epsilon(q)] = 0 \qquad (19)$$

so

$$\begin{aligned}\langle\epsilon\rangle &= \left\langle 4\pi^2\int dp\, dq\, a_i^*(p)\, a_i(q)\, \epsilon(q)\, \delta(p-q)\, e^{-i[\epsilon(q)-\epsilon(p)](t-t_i)}\right\rangle \\ &= 4\pi^2\int dq\, \langle |a(q)|^2\rangle\, \epsilon(q)\ .\end{aligned} \qquad (20)$$

Similarly the average momentum and the normalization integral are,

$$\langle q\rangle = 4\pi^2\int dq\, \langle |a(q)|^2\rangle\, q \qquad (21)$$

$$1 = \left\langle\Big(\phi_i(\underline{x}_i,t)\ \Big|\ \phi_i(\underline{x}_i,t)\Big)\right\rangle = 4\pi^2\int dq\, \langle |a(q)|^2\rangle\ . \qquad (22)$$

We note that Eqs. (13), (16), and (22) may be combined to give,

$$\underline{\gamma} = R\langle -\hat{d}\rangle\left\langle\frac{1}{4\pi d^2}\right\rangle \qquad (23)$$

$$n = \gamma\,\langle\tfrac{1}{v}\rangle \qquad (24)$$

where

$$\langle\tfrac{1}{v}\rangle = 4\pi^2\int dq\, \frac{\langle |a(q)|^2\rangle}{v(q)}\ . \qquad (25)$$

It is convenient to define a normalized energy spectrum for the beam as,

$$g[\epsilon(q)]\, d\epsilon(q) \;=\; 4\pi^2 \,\langle |a(q)|^2 \rangle\, dq \tag{26}$$

then

$$\langle \epsilon \rangle \;=\; \int d\epsilon\; g(\epsilon)\, d\epsilon \tag{27}$$

$$\langle q \rangle \;=\; \int d\epsilon(q)\; g[\epsilon(q)]\; q \quad . \tag{28}$$

We are now ready to consider scattering of the incident particles. We begin by considering the scattering of a single particle in the wave packet state $\phi_1(\underline{x}_1, t)$. The target Hamiltonian will be denoted by h, and its eigenstates and eigenvalues by g_n and W_n, respectively.

$$h\, g_n \;=\; W_n\, g_n \quad . \tag{29}$$

We will assume that the impulse approximation is valid so that the T-matrix for the particle-target interaction may be written as

$$T_1 \;=\; \sum_{\alpha} T_{\alpha 1} \tag{30}$$

where $T_{\alpha 1}$ is the T-matrix for the interaction of the incident particle and the $\underline{\alpha th}$ particle in the target. We allow the "particles" to which the impulse approximation applies to be composite, e.g., $T_{\alpha 1}$ might refer to the $\underline{\alpha th}$ atom in a gas or the $\underline{\alpha th}$ unit cell in a molecular crystal. Actually, there is no loss of generality in using (30) because we can always consider the target itself to be a single composite particle in which case (30) becomes simply $T_1 = T_{11}$.

It will be convenient to relocate the time origin by defining,

$$t' = t - t_c \qquad . \qquad t'_1 = t_1 - t_c \tag{31}$$

where t_c is a time at which $\phi_1(\underline{x}_1,t)$ overlaps the target. We take the initial target state to be g_m so that the wave function prior to scattering is,

$$\Phi_1(t) = e^{-iht}\,\phi_1(\underline{x}_1,t)\,g_m = e^{-iW_m t_c}\int dq\, a_1(q)\, e^{i\epsilon(q)t'_1}\,\chi_{in}(t') \tag{32}$$

$$\chi_{in}(t') = \frac{e^{i[q|\underline{x}_1-\underline{d}_1|-\epsilon(q)t']}}{|\underline{x}_1 - \underline{d}_1|}\; u_1(s_1)\, e^{-iW_m t'}\, g_m \quad . \tag{33}$$

The complete solution of the Schroedinger equation corresponding to the initial wave function $\Phi_1(t)$ is obtained by replacing χ_{in} in Eq. (32) by,[6]

$$\chi(t') = \chi_{in}(t') + \chi_{sc}(t') \tag{34}$$

where

$$\chi_{sc}(t') = \frac{e^{-i[\epsilon(q)+W_m]t'}}{W_m + \epsilon(q) + i\eta - h - K_1}\; T\,\chi_{in}\,(0) \tag{35}$$

$$= e^{-i[\epsilon(q)+W_m]t'} \sum_n g_n$$

$$\times \left(g_n \,\middle|\, \frac{1}{W_m + \epsilon(q) + i\eta - W_n - K_1} \sum_\alpha T_{\alpha 1}\, \frac{e^{iq|\underline{x}_1-\underline{d}_1|}}{|\underline{x}_1 - \underline{d}_1|}\; u_1(s_1) \,\middle|\, g_m \right) .$$

We will assume that the distance from the source to the target is large compared to the size of the target so that we may put,

$$\frac{1}{|\underline{x}_1 - \underline{d}_1|} = \frac{1}{d_1} . \tag{36}$$

The exponent in Eq. (35) may be expanded as,

$$q|\underline{x}_1 - \underline{d}_1| = q\, d_1 + \underline{q} \cdot \underline{x}_1 + \mathcal{O}(q\, \ell_c^{\,2}/d_1) \tag{37}$$

where

$$\underline{q} = - q\, \hat{\underline{d}}_1 \tag{38}$$

and ℓ_c is a typical linear dimension of the target $\left(T_{\alpha 1}\ f(\underline{x}_1)\right.$ vanishes unless $\underline{x}_1$ is in the target$\left.\right)$. It will be assumed that

$$\frac{\langle q \rangle\, \ell_c^{\,2}}{\langle d \rangle} \ll 1 \tag{39}$$

and the last term in Eq. (37) will be dropped. This is a rather restrictive approximation, and we will have to qualify it later.

In order to evaluate $\chi_{sc}(t')$ we Fourier analyze g_m with respect to the center-of-mass position vector of the αth target particle.

$$g_m = \int \frac{d^3k_\alpha}{(2\pi)^{3/2}}\, e^{i\underline{k}_\alpha \cdot \underline{r}_\alpha}\, g_m(\underline{k}_\alpha)$$

$$g_m(\underline{k}_\alpha) = \int \frac{d^3r_\alpha}{(2\pi)^{3/2}}\, e^{-i\underline{k}_\alpha \cdot \underline{r}_\alpha}\, g_m . \tag{40}$$

Using Eq. (40) and the approximations (36) and (39) we may write,

$$T_{\alpha 1}\frac{e^{iq|\underline{x}_1-\underline{d}_1|}}{|\underline{x}_1-\underline{d}_1|}u_1(s_1)\,g_m = \frac{e^{iqd_1}}{d_1}\int\frac{d^3k_\alpha}{(2\pi)^{3/2}}\,T_{\alpha 1}\,e^{i(\underline{q}\cdot\underline{x}_1+\underline{k}_\alpha\cdot\underline{r}_\alpha)}$$

$$\times\, u_1(s_1)\,g_m(\underline{k}_\alpha) \qquad (41)$$

$$= \frac{e^{iqd_1}}{d_1}\int\frac{d^3k_\alpha d^3q'\,d^3k'_\alpha}{(2\pi)^{3/2}}\,e^{i(q'\cdot\underline{x}_1+\underline{k}'_\alpha\cdot\underline{r}_\alpha)}\,(\underline{q}',\underline{k}'_\alpha|T_{\alpha 1}|\,\underline{q},\underline{k}_\alpha)$$

$$\times\, u_1(s_1)\,g_m(\underline{k}_\alpha)$$

where

$$(\underline{q}',\underline{k}'_\alpha\,|T_{\alpha 1}|\,\underline{q},\underline{k}_\alpha) = \int\frac{d^3x_1d^3r_\alpha}{(2\pi)^6}\,e^{-i(\underline{q}'\cdot\underline{x}_1+\underline{k}'_\alpha\cdot\underline{r}_\alpha)}\,T_{\alpha 1}\,e^{i(\underline{q}\cdot\underline{x}_1+\underline{k}_\alpha\cdot\underline{r}_\alpha)} \qquad (42)$$

$$= \delta(\underline{q}'+\underline{k}'_\alpha-\underline{q}-\underline{k}_\alpha)\,(\underline{q}',\underline{k}'_\alpha|T_{\alpha 1}{}^0|\underline{q},\underline{k}_\alpha)$$

and $T_{\alpha 1}{}^0$ is the momentum shell sub-matrix of $T_{\alpha 1}$. Equation (41) may now be formally evaluated as,

$$T_{\alpha 1}\frac{e^{iq|\underline{x}_1-\underline{d}_1|}}{|\underline{x}_1-\underline{d}_1|}u_1(s_1)\,g_m \qquad (43)$$

$$= \frac{e^{iqd_1}}{d_1}\,e^{i\underline{q}\cdot\underline{r}_\alpha}\int\frac{d^3k_\alpha\,d^3q'}{(2\pi)^{3/2}}\,e^{i\underline{q}'\cdot(\underline{x}_1-\underline{r}_\alpha)}\,(\underline{q}',\underline{k}_\alpha+\underline{q}-\underline{q}'|T_{\alpha 1}{}^0|\underline{q},\underline{k}_\alpha)$$

$$\times\, u_1(s_1)\,e^{i\underline{k}_\alpha\cdot\underline{r}_\alpha}\,g_m(\underline{k}_\alpha)$$

$$= \frac{e^{iqd_1}}{d_1}\,e^{i\underline{q}\cdot\underline{r}_\alpha}\int d^3\underline{q}'\,e^{i\underline{q}'\cdot(\underline{x}_1-\underline{r}_\alpha)}\,(\underline{q}',-i\nabla_\alpha+\underline{q}-\underline{q}'|T_{\alpha 1}{}^0|\underline{q},-i\nabla_\alpha)u_1(s_1)g_m$$

where $(\underline{q}' - i\nabla_\alpha + \underline{q} - \underline{q}' \,|T^0_{\alpha 1}|\, \underline{q}, - i\nabla_\alpha)$ is the same function of $- i\nabla_\alpha$ that $(\underline{q}', \underline{k}_\alpha + \underline{q} - \underline{q}' \,|T^0_{\alpha 1}|\, \underline{q}, \underline{k}_\alpha)$ is of $\underline{k}_\alpha$.

Putting (43) into (35) we obtain,

$$\chi_{sc}(t') = e^{-i[\epsilon(q)+W_m]t'} \sum_n g_n \left(g_n \middle| \sum_\alpha \frac{e^{iqd_1} e^{i\underline{q}\cdot\underline{r}_\alpha}}{d_1} \right.$$

$$\left. \int d^3\underline{q}' \; \frac{e^{i\underline{q}'\cdot(\underline{x}_1-\underline{r}_\alpha)} (\underline{q}', -i\nabla_\alpha + \underline{q} - \underline{q}' \,|T^0_{\alpha 1}|\, \underline{q}, -i\nabla_\alpha)}{W_m + \epsilon(q) + i\eta - W_n - \epsilon(q')} \; u_1(s_1) \middle| g_m \right) . \tag{44}$$

The integral in Eq. (44) is of a familiar form and its asymptotic value is

$$\int d^3\underline{q}' \; \frac{e^{i\underline{q}'\cdot(\underline{x}_1-\underline{r}_\alpha)} (\underline{q}' -i\nabla_\alpha + \underline{q} - \underline{q}' \,|T^0_{\alpha 1}|\, \underline{q}, - i\nabla_\alpha)}{W_m + \epsilon(q) + i\eta - W_n - \epsilon(q')}$$

$$\xrightarrow[|\underline{x}_1-\underline{r}_\alpha|\to\infty]{} \begin{cases} -(2\pi)^2 \dfrac{q_{nm}}{v(q_{nm})} \; \dfrac{e^{iq_{nm}|\underline{x}_1-\underline{r}_\alpha|}}{|\underline{x}_1 - \underline{r}_\alpha|} \; (\underline{q}_{nm}, -i\nabla_\alpha + \underline{q} - \underline{q}_{nm} |T^0_{\alpha 1}| \underline{q}, -i\nabla_\alpha) ; \epsilon(q_{nm}) > 0 \\ 0 \; ; \; \epsilon(q_{nm}) < 0 \end{cases} \tag{45}$$

where

$$\epsilon(q_{nm}) = \epsilon(q) + W_m - W_n \tag{46}$$

$$\underline{q}_{nm} = q_{nm}(\underline{x}_1 - \underline{r}_\alpha) \, |\underline{x}_1 - \underline{r}_\alpha|^{-1} . \tag{47}$$

It will be assumed here that the target to detector distance is large enough so that the following approximations may be made in (45),

$$\underline{q}_{nm} \doteq q_{nm} \hat{x}_1 \tag{48}$$

$$\frac{1}{|\underline{x}_1 - \underline{r}_\alpha|} \doteq \frac{1}{x_1} \tag{49}$$

$$e^{iq_{nm}|\underline{x}_1 - \underline{r}_\alpha|} \doteq e^{iq_{nm}x_1} \; e^{-i\underline{q}_{nm}\cdot\underline{r}_\alpha} \quad . \tag{50}$$

Approximation (50) requires that a condition analogous to (39) be satisfied,

$$\frac{\langle q_{nm}\rangle \; \ell_c^{\,2}}{\langle x_1\rangle} \ll 1 \quad . \tag{51}$$

Where $\langle q_{nm}\rangle$ is a typical final momentum and $\langle x_1\rangle$ is a typical target to detector distance.

Using (44), (45), and our subsequent approximations the scattered wave function at the detector(s) may now be written as,

$$\begin{aligned} \Psi_1(t) &= e^{-iW_m t_c} \int dq \;\; a_1(q) \; e^{i\epsilon(q)t'_1} \;\; \chi_{sc}(t') \\ &= e^{-iht} \int dq \; a_1(q) \; \sum_n{}' \, g_n \; \frac{1}{x_1 d_1} \;\; \exp\Big\{ i\Big[qd_1 + \epsilon(q)t_1 \\ &\qquad + q_{nm}\, x_1 - \epsilon(q_{nm})t \Big]\Big\} \; \Big(g_n \big| A_1(\underline{q}_{nm}, \underline{q}) \big| \; g_m\Big) \end{aligned} \tag{52}$$

where the sum $\sum_n{}'$ includes only those states for which $\epsilon(q_{nm})$ is positive and,

$$A_1(\underline{k},\underline{q}) = -(2\pi)^2 \frac{k}{v(k)} \sum_{\alpha} e^{-i(\underline{k}-\underline{q})\cdot\underline{r}_\alpha}$$

$$\times(\underline{k}, -i\nabla_\alpha + \underline{q} - \underline{k} \,|T^0_{\alpha 1}|\, \underline{q}, -i\nabla_\alpha)\, u_1(s_1) \quad . \tag{53}$$

We recall that $(\underline{k},\underline{k}_\alpha + \underline{q} - \underline{k}\,|T^0_{\alpha 1}|\,\underline{q}, \underline{k}_\alpha)$ does not operate on $\underline{x}_1$ or $\underline{r}_\alpha$, but that it is still an operator on the incident particle's spin and on the internal degrees of freedom of the αth target particle. $(\underline{k},-i\nabla_\alpha + \underline{q} - \underline{k}\,|T^0_{\alpha 1}|\,\underline{q}, -i\nabla_\alpha)$ also operates on $\underline{r}_\alpha$, of course.

We digress for a moment to consider the conditions (39) and (51). These are actually too restrictive, and ℓ_c may be thought of as an appropriate correlation length in the target rather than the size of the target. The target may then be thought of as being made up of a number of essentially independent parts each of volume ℓ_t^3 where $\ell_t \gg \ell_c$ and $\ell_t^2 \ll \langle d\rangle/\langle q\rangle$, $\ell_t^2 \ll \langle x_1\rangle/\langle q_{nm}\rangle$. The scattering from one of these parts will be incoherent with the scattering from any other part, and the scattered intensities (rather than amplitudes) from the different parts may be added to find the total intensity. The calculations given here are appropriate to the part located at the (arbitrarily chosen) origin; knowing the scattered intensity from this part one may find the total intensity by the methods of geometrical optics.

It is useful to write (52) in the form,

$$\Psi_1(t) = e^{-iht}\, \psi_1(\underline{x}_1, t)\, g_m \tag{54}$$

where $\psi_1(\underline{x}_1, t)$ is an operator whose matrix elements are given by (52),

$$\psi_1(\underline{x}_1, t) = \int dq\, a_1(q)\, \psi_1(q, \underline{x}_1, t)$$

$$\left(g_n \middle| \psi_1(q, \underline{x}_1, t) \middle| g_m\right) = \frac{1}{x_1 d_1} \exp\left\{ i \left[q d_1 + \epsilon(q)\, t_1 \right.\right. \tag{55}$$

$$\left.\left. + q_{nm}\, x_1 - \epsilon(q_{nm}) t \right]\right\} \left(g_n \middle| A_1(\underline{q}_{nm}, \underline{q}) \middle| g_m\right);\ \epsilon(q_{nm}) > 0 \;=\; 0;\ \epsilon(q_{nm}) < 0 .$$

We shall deal here with the case of a low density incident beam. The density is assumed to be low enough so that the excitations of the target due to one incident particle do not appreciably affect the properties of the target "seen" by another incident particle. In this case the wave function for the scattered particles and the target may be written as,

$$\Psi_{sc}(t) = e^{-iht}\, \mathcal{S} \prod_i \psi_i(\underline{x}_i, t)\, g_0 \tag{56}$$

where the $\psi_i(\underline{x}_i, t)$ are to be treated as commuting operators. Only wave packets for which $t_i < t$ are to be kept in the product in (56). g_0 is the initial target state, and it must be averaged over an ensemble after expectation values have been taken.

To close this preliminary section we will use expression (52) to calculate the scattering cross-section of the target. The average current of scattered particles far from the target is,

$$\mathcal{J}_{sc}(\underline{r}) = N \left\langle \mathrm{Re} \left(\Psi_1(t) | \delta(\underline{x}_1 - \underline{r}) \, \underline{v}_1 | \Psi_1(t) \right) \right\rangle$$

$$= N \left\langle \mathrm{Re} \int d^3x_1 \sum_{s_1} \left(g_0 | \psi_1^{\dagger}(\underline{x}_1, t) \, \delta(\underline{x}_1 - \underline{r}) \, \underline{v}_1 \, \psi_1(\underline{x}_1, t) | g_0 \right) \right\rangle \tag{57}$$

where $\underline{v}_1$ is the velocity operator of the incident particle. Inserting a complete set of target states and writing out the matrix elements of ψ_1 explicitly we obtain,

$$\mathcal{J}_{sc}(\underline{r}) = N \Bigg\langle \mathrm{Re} \int d^3x_1 \, \delta(\underline{x}_1 - \underline{r}) \sum_{s_1} \int dq' \, dq \, a_1^{*}(q') \, a_1(q)$$

$$\sum_n{}' \frac{1}{d_1^2 x_1^2} \, v(q_{n0}) \, \hat{x}_1 \, \exp \Big\{ i \Big[(q - q') \, d_1 + [\epsilon(q) - \epsilon(q')] t_1$$

$$+ (q_{n0} - q'_{n0}) \, x_1 - [\epsilon(q_{n0}) - \epsilon(q'_{n0})] t \Big] \Big\} \tag{58}$$

$$\times \left(g_0 | A_1^{+}(\underline{q}'_{n0}, \underline{q}') | g_n \right) \left(g_n | A_1(q_{n0}, \underline{q}) | g_0 \right) \Bigg\rangle .$$

We carry out the average over emission times,

$$\left\langle e^{i[\epsilon(q) - \epsilon(q')] t_1} \right\rangle = \frac{\pi}{T_0} \, \delta[\epsilon(q) - \epsilon(q')] \tag{59}$$

and use (23) to obtain,

$$\mathcal{J}_{sc}(\underline{r}) = \frac{\mathcal{G}\,\hat{\underline{r}}}{r^2} \left\langle \sum_{s_1} \sum_n{}' \frac{v(q_{n0})}{v(q)} \left| \left(g_n | A_1(q_{n0} \, \hat{\underline{r}}, \underline{q}) | g_0 \right) \right|^2 \right\rangle . \tag{60}$$

In Eq. (60) the brackets include an average over the incident beam spectrum,

$$\langle f(q)\rangle = \int d\epsilon(q)\, g[\epsilon(q)]\, f(q) \quad . \tag{61}$$

The differential scattering cross-section is,

$$\left\langle\frac{d\sigma}{d\Omega}\right\rangle = \frac{r^2|\mathcal{J}_{sc}(\underline{r})|}{\mathcal{J}} = \left\langle \sum_{s_1} \sum_n{}' \frac{v(q_{n0})}{v(q)} \left|\left(g_n|A_1(q_{nm}\hat{r},\underline{q})|g_0\right)\right|^2 \right\rangle$$
$$= \int_0^\infty d\epsilon \left\langle \frac{d^2\sigma}{d\Omega\, d\epsilon}\right\rangle \tag{62}$$

where

$$\left\langle\frac{d^2\sigma}{d\Omega\, d\epsilon}\right\rangle = \left\langle \frac{v(k)}{v(q)} \sum_{n,s_1} \delta[\epsilon - \epsilon(q_{n0})] \left|\left(g_n|A_1(q_{nm}\hat{r},\ \underline{q})|g_0\right)\right|^2 \right\rangle . \tag{63}$$

Here k is defined by,

$$\epsilon = \epsilon(k) \ . \tag{64}$$

If the δ-function in (63) is written as,

$$\delta[\epsilon - \epsilon(q_{n0})] = \int \frac{dt}{2\pi}\, e^{i[\epsilon-\epsilon(q)-W_0 + W_n]t} \quad , \tag{65}$$

and the factor $e^{i(W_n-W_0)t}$ is used to form a Heisenberg operator, we may write the energy-dependent cross-section as,

$$\left\langle \frac{d^2\sigma}{d\Omega\, d\epsilon} \right\rangle = \left\langle \frac{v(k)}{v(q)} \int \frac{dt}{2\pi} \; e^{i[\epsilon-\epsilon(q)]t} \sum_{s_1} (g_0 | A_1^{\dagger}(k\hat{r},\underline{q}) A_1(k\hat{r},\underline{q},t) | g_0) \right\rangle \tag{66}$$

where

$$A_1(k\hat{r},\underline{q},t) = e^{iht} \; A_1(k\hat{r},\underline{q}) \; e^{-iht} \; . \tag{67}$$

Expressions of this general type for $\langle d^2\sigma / d\Omega\, d\epsilon \rangle$ are familiar in the theory of scattering by many-particle systems.[7,8] To obtain useful results it is usually necessary (and quite valid) to make a number of approximations in $T_{\alpha 1}$. For example, Van Hove's[7] expression for the neutron scattering cross-section of a many-particle target may be obtained from (66) by putting $T_{\alpha 1}$ equal to the Fermi pseudo-potential,

$$\begin{aligned} T_{\alpha 1} &= \frac{2\pi}{m} \; a_{\alpha 1} \; \delta(\underline{x}_1 - \underline{r}_\alpha) \\ (\underline{k}, - i \nabla_\alpha + \underline{q} - \underline{k} \, | T_{\alpha 1}{}^0 | \, \underline{q}, - i \nabla_\alpha) &= a_{\alpha 1} / (2\pi)^2 \, m \end{aligned} \tag{68}$$

where $a_{\alpha 1}$ is the spin-dependent scattering length of the $\underline{\alpha th}$ nucleus in the target and m is the neutron mass.

III. INTENSITY CORRELATIONS

In order to describe intensity correlations we will use the GW counting operators. We first consider the case of observations made at a single detector called detector a. It will be represented by the operator,

$$J_a = \sum_{i=1}^{N} j_{a,i} \qquad (69)$$

$$j_{a,i} = \int_a d^3x_a \int_a d\epsilon_a \, \gamma_a(\underline{x}_a, \epsilon_a) \, \delta(\underline{x}_a - \underline{x}_i) \, \delta(\epsilon_a - K_i) \, . \qquad (70)$$

The integrals in $j_{a,i}$ extend over the active volume of counter a and over the range of scattered particle energies accepted by counter a. This operator is slightly different from the GW operator in that we include the possibility of a finite range of acceptable final energies. (The range of final energies will usually be determined by some sort of energy filter in front of the actual detector.) $\gamma_a(\underline{x}_a, \epsilon_a)$ depends on the sensitivity and calibration of the detector. For the sake of simplicity we will take γ_a to be a constant,

$$j_{a,i} = \gamma_a \int_a d^3x_a \, d\epsilon_a \, \delta(\underline{x}_a - \underline{x}_i) \, \delta(\epsilon_a - K_i) \, . \qquad (71)$$

Except for a numerical factor J_a represents the number of particles in counter a at a given instant. In an actual measurement this number can not be observed because of the finite resolving time of electronic apparatus. Thus, if

$$\langle J_a(t)\rangle = \langle\left(\Psi_{sc}(t)\right| J_a \left|\Psi_{sc}(t)\right)\rangle \tag{73}$$

is the average (averaged over $a_i(q)$, $u_i(s)$, t_i, $\underline{d}_i$, and g_0) expectation value of J_a at time t only the quantity

$$\langle G_a(T)\rangle = \int_{-\infty}^{\infty} dt \; L_a(T - t) \langle J_a(t)\rangle \tag{74}$$

can be observed. Here L_a is the response function of the electronic recording apparatus. It is convenient to define a frequency characteristic for the recording apparatus as,

$$B_a(\omega) = \int_{-\infty}^{\infty} d\tau \; L_a(\tau) \; e^{i\omega\tau} \; . \tag{75}$$

If the intensity of scattered particles is steady (so that $\langle J_a(t)\rangle$ does not depend on t) $\langle G_a\rangle$ becomes,

$$\langle G_a\rangle = B_a(0) \langle J_a\rangle \; . \tag{76}$$

We will take the detector to be so calibrated that $T_0\langle J_a\rangle$ is the total number of particles detected at a during the experiment. $\langle J_a\rangle$ may then be interpreted as the average counting rate at a and $\langle G_a\rangle$ as an amplified counting rate.

If there is never more than one particle at a time in the detector Eq. (73) for $\langle J_a(t)\rangle$ reduces to

$$\langle J_a(t)\rangle = N \left\langle \left(\Psi_1(t)\right| j_{a,i} \left| \Psi_1(t)\right)\right\rangle . \tag{77}$$

The evaluation of $\langle J_a(t)\rangle$ is similar to the evaluation $\mathcal{J}_{sc}(\underline{r})$ given in the previous section. It is found that $\langle J_a(t)\rangle$ is independent of t and is given by,

$$\langle J_a\rangle = \mathcal{J}\gamma_a \int_a \frac{d^3x_a}{x_a^2} \int_a d\epsilon_a \frac{1}{v(k_a)} \left\langle \frac{d^2\sigma}{d\Omega_a\, d\epsilon_a} \right\rangle \tag{78}$$

where $\epsilon(k_a) = \epsilon_a$, $d\Omega_a$ is an element of solid angle in the direction of $\hat{x}_a$, and the cross-section is defined by Eq. (66). In (78) we will put,

$$\int_a \frac{d^3x_a}{x_a^2} \doteq w_a \int_a d\Omega_a \tag{79}$$

where w_a is a mean depth of penetration of scattered particles into the counter. $\langle J_a\rangle$ then becomes,

$$\langle J_a\rangle = \mathcal{J}\gamma_a\, w_a\, \sigma_a/v_a \tag{80}$$

where σ_a is the cross-section for scattering into a and v_a is an average value of $v(k_a)$. If the counter were 100% efficient the counting rate at a would be just $\mathcal{J}\sigma_a$. Thus our calibration requirements reduce to,

$$\gamma_a = \frac{v_a}{w_a}\eta_a \tag{81}$$

where η_a is the counter efficiency.

We next consider the case of observations made at two detectors, a and b. The output of detector a is sent through a delay line and then multiplied with the output of detector b. The output of the multipler is,

$$\langle G_{ba}(T,\tau_0)\rangle = \int_{-\infty}^{\infty} dt_b\, dt_a\, L_b(T+\tau_0-t_b)\, L_a(T-t_a)\, \langle J_{ba}(t_b,t_a)\rangle \tag{82}$$

where τ_0 is the delay time and

$$\langle J_{ba}(t_b,t_a)\rangle = \langle (\Psi_{sc}(t_a)|J_b(\tau)J_a|\Psi_{sc}(t_a))\rangle \tag{83}$$

$$J_b(\tau) = \sum_{i=1}^{N} e^{iK_i\tau}\, j_{b,i}\, e^{-iK_i\tau} \tag{84}$$

$$\tau = t_b - t_a \quad . \tag{85}$$

It is assumed that τ_0 is large compared to electronic response times so that τ is always positive. K_i is the Hamiltonian of the ith beam particle and $j_{b,i}$ is defined analogously to $j_{a,i}$. The correlated counting rate is defined as

$$N_{ba}(\tau_0) = \frac{1}{T_0}\int_{T_1}^{T_1+T_0} dT\, \langle G_{ba}(T,\tau_0)\rangle \quad . \tag{86}$$

Equations (82)-(86) also apply to the case in which a and b are actually a single detector. The use of the time dependent operator $J_b(\tau)\, J_a$ to describe intensity correlation measurements has been discussed in detail by GW.

In the case that no particle can be counted in both detectors (or twice in a single detector if a and b are the same) $\langle J_{ba}(t_b, t_a)\rangle$ reduces to,

$$\langle J_{ba}(t_b, t_a)\rangle = \tag{87}$$
$$\frac{N^2}{2} \left\langle\left\langle \Psi_{12}(t_a) \middle| e^{iK_2\tau}\, j_{b,2}\, e^{-iK_2\tau}\, j_{a,1} + e^{iK_1\tau}\, j_{b,1}\, e^{-iK_1\tau}\, j_{a,2} \middle| \Psi_{12}(t_a) \right\rangle\right\rangle$$

where

$$\Psi_{12}(t_a) = \frac{e^{-iht_a}}{\sqrt{2}} \left(\psi_1(\underline{x}_1, t_a)\, \psi_2(\underline{x}_2, t_a) \pm \psi_1(\underline{x}_2, t_a)\, \psi_2(\underline{x}_1, t_a)\right) g_0 \,. \tag{88}$$

The upper (lower) sign in (84) refers to an incident beam of Bosons (Fermions). Equation (55) for the matrix elements of ψ_1 shows that

$$e^{-iK_1\tau}\, \psi_1(\underline{x}_1, t_a) = \psi_1(\underline{x}_1, t_b) \,. \tag{89}$$

Using (89) and the statistical equivalence of the incident wave packets it is readily shown that,

$$\langle J_{ba}(t_b, t_a)\rangle = \langle J_d(t_b, t_a)\rangle \pm \langle J_{ex}(t_b, t_a)\rangle \tag{90}$$

where

$$\langle J_d(t_b,t_a)\rangle = N^2 \langle (\psi_2(\underline{x}_2,t_b)\psi_1(\underline{x}_1,t_a)g_0|j_{b,2}j_{a,1}|\psi_2(\underline{x}_2,t_b)\psi_1(\underline{x}_1,t_a)g_0)\rangle$$

$$\tag{91}$$

$$\langle J_{ex}(t_b,t_a)\rangle = N^2 \langle (\psi_1(\underline{x}_2,t_b)\psi_2(\underline{x}_1,t_a)g_0|j_{b,2}j_{a,1}|\psi_2(\underline{x}_2,t_b)\psi_1(\underline{x}_1,t_a)g_0)\rangle .$$

In the evaluation of $\langle J_{ba}(t_b,t_a)\rangle$ we will use an approximation due to GW which neglects correlations between scattered wave-packets originating from different incident wave-packets. This allows us to put,

$$\langle J_d(t_b,t_a)\rangle \doteq N^2\int \underline{d^3x_2}\ \underline{d^3x_1} \sum_{s_2 s_1} \langle (g_2|\psi_2{}^\dagger(\underline{x}_2,t_b)\ j_{b,2}\ \psi_2(\underline{x}_2,t_b)|g_2)\rangle$$

$$\langle (g_1|\psi_1{}^\dagger(\underline{x}_1,t_a)\ j_{a,1}\ \psi_1(\underline{x}_1,t_a)|g_1)\rangle \tag{92}$$

$$\langle J_{ex}(t_b,t_a)\rangle \doteq N^2\int \underline{d^3x_2}\ \underline{d^3x_1} \sum_{s_2 s_1} \langle (g_2|\psi_2{}^\dagger(\underline{x}_1,t_a) j_{b,2}\ \psi_2(\underline{x}_2,t_b)|g_2)\rangle$$

$$\langle (g_1|\psi_1{}^\dagger(\underline{x}_2,t_b)\ j_{a,1}\ \psi_1(\underline{x}_1,\ t_a)|g_1)\rangle . \tag{93}$$

Independent (but equivalent) ensemble averages over the initial target states g_2 and g_1 are to be taken in (92) and (93). Upon inspection of (77) it is seen that $\langle J_d(t_b,t_a)\rangle$ is independent of t_b and t_a and is simply

$$\langle J_d\rangle = \langle J_b\rangle \langle J_a\rangle . \tag{94}$$

$\langle J_{ex}\rangle$ is the more interesting term in $\langle J_{ba}\rangle$. The matrix elements in (93) are evaluated similarly to (57). It is found that,

$$N\langle (g_1|\psi_1^\dagger(\underline{x}_2,t_b)\ j_{a,1}\ \psi_1(\underline{x}_1,t_a)|g_1)\rangle$$

$$= \frac{\gamma_a \mathcal{g}}{x_2 x_1} \int_a d\epsilon_a \ e^{i[k_a(x_1-x_2)+\epsilon_a \tau]} \left\langle \frac{1}{v(q)} \sum_n \delta[\epsilon_a - \epsilon(q_{n1})] \right. \tag{95}$$

$$\left. (g_1|A_{12}^\dagger(k_a\hat{x}_2,\underline{q})|g_n)\ (g_n|A_1(k_a\ \hat{x}_1,\underline{q})|g_1) \right\rangle$$

when $\underline{x}_1$ is in the active volume of counter a . A_{12} is the same as A_1 except that $u_1(s_1)$ is replaced by $u_1(s_2)$. A similar result is found for the other factor in (93). $\langle J_{ex}(t_b,t_a)\rangle$ thus depends on t_b and t_a only through $\tau = t_b - t_a$ and is given by,

$$\langle J_{ex}(\tau)\rangle = \mathcal{g}^2\ \gamma_a\gamma_b \int_b \frac{d^3x_b}{x_b^2} \int_a \frac{d^3x_a}{x_a^2} \int_b d\epsilon_b \int_a d\epsilon_a\ \exp\{i[(k_b-k_a)(x_b-x_a)-(\epsilon_b-\epsilon_a)\tau]\}$$

$$\left\langle \frac{1}{v(p)v(q)} \sum_{nn'} \delta[\epsilon_b - \epsilon(p_{n'2})]\ \delta[\epsilon_a - \epsilon(q_{n1})] \right. \tag{96}$$

$$\sum_{s_2 s_1} (g_2|A_{21}^\dagger(\underline{k}_{ba},p)|g_{n'})\ (g_{n'}|A_2(\underline{k}_{bb},p)|g_2)$$

$$\left. (g_1|A_{12}^\dagger(\underline{k}_{ab},\ \underline{q})|g_n)\ (g_n|A_1(\underline{k}_{aa},\ \underline{q})|g_1) \right\rangle$$

where

$$\epsilon(k_a) = \epsilon_a,\ \epsilon(k_b) = \epsilon_b$$

$$\underline{q} = -q\,\hat{d}_1,\quad p = -p\,\hat{d}_2 \tag{97}$$

$$\underline{k}_{aa} = k_a\,\hat{x}_a,\ \underline{k}_{ab} = k_a\,\hat{x}_b,\ \underline{k}_{bb} = k_b\,\hat{x}_b,\ \underline{k}_{ba} = k_b\,\hat{x}_a\ .$$

The brackets in (96) indicate averages over $\hat{d}_1$, $\hat{d}_2$, u_1, u_2, g_1, g_2, and beam spectrum averages over both p and q,

$$\langle f(p,q)\rangle = \int d\epsilon(p)\, d\epsilon(q)\, g[\epsilon(p)]\, g[\epsilon(q)]\, f(p,q)\ . \tag{98}$$

We will assume an unpolarized incident beam so that those terms in the sum $\sum_{s_2 s_1}$ for which $s_2 \neq s_1$ vanish. We also assume that the remaining terms are independent of s_1 or s_2. Then we may replace A_{21} and A_{12} by A_2 and A_1, respectively, in (96) if we include a factor s^{-1} to account for overcounting. s is the number of terms in the sum $\sum_{s_1}$ or $\sum_{s_2}$. These assumptions are not at all necessary, but they simplify the notation considerably.

Under certain limiting conditions (96) reduces to the result obtained by GW for $\langle J_{ex}(\tau)\rangle$. First we take the exponential in (96) inside the brackets and assume that the scattering is nearly elastic so that we may put

$$\begin{aligned} k_a &\doteq q + \frac{dq}{d\epsilon(q)}\,[\epsilon_a - \epsilon(q)] = q + \frac{W_1 - W_n}{v(q)} \\ k_b &\doteq p + \frac{W_2 - W_{n'}}{v(p)}\ . \end{aligned} \tag{99}$$

The exponent then becomes

$$\begin{aligned} (k_b - k_a)(x_b - x_a) - (\epsilon_b - \epsilon_a)\,\tau &\doteq (p - q)(x_b - x_a) - [\epsilon(p) - \epsilon(q)]\tau \\ &+ (W_1 - W_n)\left(\tau - \frac{x_b - x_a}{v(q)}\right) + (W_{n'} - W_2)\left(\tau - \frac{x_b - x_a}{v(p)}\right)\ . \end{aligned} \tag{100}$$

We also put

$$\epsilon(p_{n'2}) = \epsilon(p),\ \epsilon(q_{n1}) = \epsilon(q) \tag{101}$$

in the δ-functions and assume that there is no energy selection at the counters. Finally we assume that $T_{\alpha 1}^{0}$ may be approximated by,

$$(\underline{q}', \underline{k}_\alpha + \underline{q} - \underline{q}' | T_{\alpha 1}^{0} | \underline{q}, \underline{k}_\alpha) = -(2\pi)^{-2} \frac{v(q)}{q} f_{\alpha 1}(\hat{q}') \tag{102}$$

where $f_{\alpha 1}(\hat{q}')$ is a scattering amplitude whose energy dependence is negligible over the incident beam spectrum. The terms in (100) involving $W_1 - W_n$ and $W_{n'} - W_2$ are used to form Heisenberg operators and (96) reduces to,

$$\langle J_{ex}(\tau)\rangle \doteq \frac{1}{s} \gamma^2 \gamma_a \gamma_b \int_b \frac{d^3x_b}{x_b^2} \int_a \frac{d^3x_a}{x_a^2} \Bigg| \Bigg\langle \frac{1}{v(q)} e^{i[q(x_b - x_a) - \epsilon(q)\tau]} \sum_{s_1} \Bigg(g_1 \Bigg| \sum_{\alpha,\beta} u_1^*(s_1) f_{\alpha 1}^{\dagger}(\hat{x}_a) e^{i(q\hat{x}_a - \underline{q})\cdot \underline{r}_\alpha} \exp\Bigg[-i(q\hat{x}_b - \underline{q})\cdot \underline{r}_\beta \Bigg(\tau - \frac{x_b - x_a}{v(q)}\Bigg)\Bigg] f_{\beta 1}(x_b)\, u_1(s_1) \Bigg| g_1 \Bigg)\Bigg\rangle\Bigg|^2 \tag{103}$$

which is equivalent to GW's result.

Returning to Eq. (96) we note that by expanding the δ-functions as in (65) $\langle J_{ex}(\tau)\rangle$ may be written as,

$$\langle J_{ex}(\tau)\rangle = \frac{1}{s} \gamma^2 \gamma_a \gamma_b \int_b \frac{d^3x_b}{x_b^2} \int_a \frac{d^3x_a}{x_a^2} \int_b d\epsilon_b \int_a d\epsilon_a \, e^{i[(k_b-k_a)(x_b-x_a)-(\epsilon_b-\epsilon_a)\tau]}$$

$$\left\langle \frac{1}{v(p)} \int \frac{dt}{2\pi} \, e^{i[\epsilon_b-\epsilon(p)]t} \sum_{s_2} \left(g_0 \, | A_2^\dagger(\underline{k}_{ba}, \underline{p}) \, A_2(\underline{k}_{bb}, \underline{p}, t) | g_0 \right) \right\rangle \tag{104}$$

$$\left\langle \frac{1}{v(q)} \int \frac{dt}{2\pi} \, e^{-i[\epsilon_a-\epsilon(q)]t} \sum_{s_1} \left(g_0 | A_1^\dagger(\underline{k}_{ab}, \underline{q}, t) \, A_1(\underline{k}_{aa}, \underline{q}) | g_0 \right) \right\rangle .$$

When the target is translated by an amount $\underline{R}$ the first and second matrix elements in (104) are multiplied by $\exp[ik_b\underline{R}\cdot(\hat{x}_b - \hat{x}_a)]$ and $\exp[ik_a\underline{R}\cdot(\hat{x}_a - \hat{x}_b)]$, respectively. Thus we expect these matrix elements to be peaked about $\hat{x}_b = \hat{x}_a$. Letting θ be the angular separation of $\hat{x}_b$ and $\hat{x}_a$ the matrix elements in (104) will be peaked functions of θ about $\theta = 0$, and the width of these peaks will be

$$(\Delta\theta)_b \sim (k_b \ell_{ts})^{-1}, \quad (\Delta\theta)_a \sim (k_a \ell_{ts})^{-1} \tag{105}$$

where ℓ_{ts} is a length associated with translation symmetry breaking in the target. At its largest ℓ_{ts} will be of order ℓ_t . In the remainder of this paper we will assume that

$$\theta_m \ll (k_b \ell_{ts})^{-1}, \ (k_a \ell_{ts})^{-1} \tag{106}$$

where θ_m is the maximum angular separation of $\hat{x}_b$ and $\hat{x}_a$. When (106) is satisfied the matrix elements in (104) may be evaluated at $\theta = 0$.

There may well be interesting and useful information to be obtained in a wide-angle intensity correlation experiment where (106) is not satisfied but we will not investigate that possibility here.

Comparing (104) and (66) it is seen that we now have,

$$\langle J_{ex}(\tau)\rangle = \frac{1}{s} \gamma^2 \gamma_a \gamma_b \Omega_a \Omega_b \int_b dx_b \int_a dx_a \int_b d\epsilon_b \int_a d\epsilon_a \frac{1}{v(k_b)v(k_a)}$$
$$\exp\{i[(k_b - k_a)(x_b - x_a) - (\epsilon_b - \epsilon_a)\tau]\} \left\langle \frac{d^2\sigma}{d\epsilon_b d\Omega_0} \right\rangle \left\langle \frac{d^2\sigma}{d\epsilon_a d\Omega_0} \right\rangle . \qquad (107)$$

The direction of the solid angle element $d\Omega_0$ must be in the general direction of the counters but is otherwise arbitrary. The integrals over solid angle in (104) have been carried out in (107),

$$\int_b \frac{d^3x_b}{x_b^2} = \Omega_b \int_b dx_b \; ; \int_a \frac{d^3x_a}{x_a^2} = \Omega_a \int_a dx_a . \qquad (108)$$

We will assume hereafter that

$$|k_b - k_a| \ll w_b, w_a \qquad (109)$$

so that $\underline{x}_b$ and $\underline{x}_a$ may be replaced by average values $\underline{Y}_b$ and $\underline{Y}_a$, and that the ranges of energy acceptance are narrow enough so that $v(k_b)$ and $v(k_a)$ can be replaced by average values v_b and v_a. Using (81) we then obtain,

$$\langle J_{ex}(\tau)\rangle = \frac{1}{s} \gamma^2 \eta_a \eta_b \Omega_a \Omega_b \int_b d\epsilon_b \int_a d\epsilon_a$$
$$\exp\{i[(k_b - k_a)(Y_b - Y_a) - (\epsilon_b - \epsilon_a)\tau]\} \left\langle \frac{d^2\sigma}{d\epsilon_b d\Omega_0} \right\rangle \left\langle \frac{d^2\sigma}{d\epsilon_a d\Omega_0} \right\rangle . \tag{110}$$

We must now return to the comments made in Section II concerning conditions (39) and (51). Equations (80), (94), and (110) give only that part of the counting rates that arises from scattering by a small volume of order ℓ_t^{3} about the origin and we must now scale these up in order to take scattering from the whole target into account. It is easy to see how $\langle J_a \rangle$ should be scaled up; σ_a is replaced by σ_a^T which is the cross-section for scattering into a by the whole target.

$$\langle J_a \rangle^T = \gamma \eta_a \sigma_a^T . \tag{111}$$

In the evaluation of (92) we treated ψ_1 and ψ_2 as waves which were both scattered from the same small region near the origin. In scaling up $\langle J_d \rangle$ we must allow ψ_1 and ψ_2 to be scattered independently from any region of the target. Thus,

$$\langle J_d \rangle^T = \langle J_a \rangle^T \langle J_b \rangle^T . \tag{112}$$

In Eq. (110) we will assume for the sake of simplicity that the solid angle subtended by the target at the detectors is small enough so that variations in $d\Omega_0$ over the target may be neglected. Then only variations in the exponential need be taken into account. It is clear from Eq. (95) that the scaled up version of Eq. (110) is,

$$\langle J_{ex}(\tau)\rangle^T = \frac{1}{s} \mathcal{G}^2 \eta_a \eta_b \Omega_a \Omega_b \int_b d\epsilon_b \int_a d\epsilon_a \int_t \frac{d^3 s_b}{V_t} \int_t \frac{d^3 s_a}{V_t}$$

$$\times \exp\{i[k_b(|\underline{Y}_b - \underline{s}_b| - |\underline{Y}_a - \underline{s}_b|) - k_a(|\underline{Y}_b - \underline{s}_a| - |\underline{Y}_b - \underline{s}_a|) - (\epsilon_b - \epsilon_a)\tau]\}$$

$$\times \left\langle \frac{d^2\sigma}{d\epsilon_b d\Omega_0} \right\rangle^T \left\langle \frac{d^2\sigma}{d\epsilon_a d\Omega_0} \right\rangle^T \tag{113}$$

where the integral $\int_t \frac{d^3 s}{}$ runs over the volume of the target, V_t is the volume of the target, and $\langle d^2\sigma / d\epsilon d\Omega\rangle^T$ is the differential cross-section of the whole target. Equation (113) becomes particularly simple when the same energy filter is used for both detectors,

$$\int_b d\epsilon_b = \int_a d\epsilon_a = \int_f d\epsilon \, . \tag{114}$$

In this case

$$\langle J_{ex}(\;)\rangle = \frac{1}{s} \mathcal{G}^2 \eta_a \eta_b \Omega_a \Omega_b \left| \int_f d\epsilon \int_t \frac{d^3 s}{V_t} \right.$$

$$\left. \times \exp\{i[k(|\underline{Y}_b - \underline{s}| - |\underline{Y}_a - \underline{s}|) - \epsilon\tau]\} \left\langle \frac{d^2\sigma}{d\epsilon\, d\Omega_0} \right\rangle^T \right|^2 . \tag{115}$$

Equation (115) is quite similar in form to the results obtained by GW for the correlated counting rate in an incoherent beam of particles emerging from a source.

Photon beams are more amenable to intensity correlation experiments than are beams of massive particles because it is easier to produce long wavelength photons. When specialized to the case of light scattering Eq. (115) becomes completely equivalent to the correlated counting rate for a radiating source considered by Goldberger, Lewis, and Watson[5] (hereafter referred to as GLW). To show this correspondence explicitly we define,

$$R_T \equiv \mathcal{Y} \left\langle \frac{d\sigma}{d\Omega_0} \right\rangle_f^T \equiv \mathcal{Y} \int_f d\epsilon \left\langle \frac{d^2\sigma}{d\epsilon\, d\Omega_0} \right\rangle^T \tag{116}$$

$$g_t(\epsilon) \equiv \left\langle \frac{d^2\sigma}{d\epsilon\, d\Omega_0} \right\rangle^T \Big/ \left\langle \frac{d\sigma}{d\Omega_0} \right\rangle_f^T \tag{117}$$

$$\begin{aligned} \int(a)\cdots &\equiv \left(\int_a \underline{d^3x_a}\, \gamma_a \right) \int_{-\infty}^{\infty} dt_a\, L_a(T - t_a)\cdots \\ &\doteq 4\pi\, Y_a^{\,2}\, \Omega_a\, \eta_a\, c \int_{-\infty}^{\infty} dt_a\, L_a(T - t_a)\cdots \end{aligned} \tag{118}$$

$$\begin{aligned} \int(b)\cdots &\equiv \left(\int_b d^3x_b\, \gamma_b \right) \int_{-\infty}^{\infty} dt_b\, L_b(T + \tau_0 - t_b)\cdots \\ &\doteq 4\pi\, Y_b^{\,2}\, \Omega_b\, \eta_b\, c \int_{-\infty}^{\infty} dt_b\, L_b(T + \tau_0 - t_b)\cdots \end{aligned} \tag{119}$$

Using Eqs. (94) and (115) we may then write Eq. (82) for the correlated counting rate as,

$$\langle G_{ba}(\tau_0) \rangle = \langle G_a \rangle \langle G_b \rangle + \frac{1}{2} \int(b) \int(a)\, |X(ba)|^2 \tag{120}$$

where

$$\chi(ba) = \frac{R_T}{4\pi c\ Y_a Y_b} \int_t \frac{d^3 s}{V_t} \int_f d\epsilon\ g_t(\epsilon)\ \exp\{i[k(|\underline{Y}_b - \underline{s}| - |\underline{Y}_a - \underline{s}|) - \epsilon(t_b - t_a)]\}. \tag{121}$$

This is equivalent to GLW's Eqs. (2.21) - (2.24) except for the fact that we have already made certain approximations which they do not make until later in their paper. Practical applications of these equations to intensity correlation experiments have been considered in detail by GLW.

Two interesting types of experiments to which (120) and (121) apply (when a little more care is taken with the spin correlations) are the resonant scattering of light by a gas of atoms and Brillouin or Raman scattering of light by phonons. The first experiment is physically quite similar to the experiments considered by GLW, and we will not discuss it here. In order to get useful results in an intensity correlation experiment it is desirable for the width of $B(\omega)$ to be larger than that of $g_t(\epsilon)$. For the fastest electronic equipment currently available $B(\omega)$ has a width of order 10^{10} sec^{-1} while Brillouin lines have widths of order 10^9- 10^{10} sec^{-1} and Raman lines are somewhat wider. Thus it appears that intensity correlations in Brillouin lines, at least, might be observable. However, it is necessary for $\mathcal{J}$ and T_0 to be quite large in order to obtain a good signal to noise ratio. For the case of fast electronic response times and a large target the signal to noise ratio given by GLW is,

$$\frac{S}{N} = \eta(T_0\ \Delta\tau)^{\frac{1}{2}} \left(\frac{\lambda^2 R_T}{4\pi\ \ell_T^{\ 2}} \right) \tag{122}$$

where λ is a typical photon wavelength, ℓ_T is a typical linear dimension of the target, and $\Delta\tau$ is the response time of the recording apparatus. Taking $\lambda \sim 5 \times 10^{-5}$ cm, $\Delta\tau \sim 10^{-10}$ sec, and[9] $\langle d\sigma/d\Omega \rangle_f^T \sim 10^{-6} \ell_T^2$ the signal to noise ratio becomes,

$$\frac{S}{N} \sim 10^{-21} \eta \mathcal{J} \sqrt{T_0} \tag{123}$$

where T_0 is in seconds and $\mathcal{J}$ is in photons/cm^2 sec. This is rather discouraging because the highest available photon currents from gas lasers are about 10^{17} photons/cm^2 sec which would require a counting time of order

$$T_0 \sim \frac{3 \text{ years}}{\eta} \tag{124}$$

for a signal to noise ratio of one. However, there are so many factors of ten floating around in this crude estimate that the experiment might actually prove to be feasable upon closer examination.

Intensity correlations in Raman scattering have also been studied by Fetter[10] using a different method. Fetter's results are less general than ours because he uses a specific dynamical model for the target and makes a number of approximations that we have avoided, e.g., he considers only one-phonon scattering. Also, his calculations are restricted to the case of a small target while our Eqs. (120) and (121) are valid for a target of arbitrary size and shape. The most important difference between Fetter's work and ours, however, is that his method does not bring out the simple and exact correspondence between the intensity correlations in a beam of scattered light and the intensity correlations in a light beam emitted by a radiating source.

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PART II

I. INTRODUCTION

Interference effects associated with the decay of coherently excited non-degenerate atomic states have been observed and studied for some time. Most of the interest in these effects has been concerned with the decay of radiatively excited states.[1] However, a number of recent experiments have demonstrated the possibility of observing such effects in the decay of electronically excited states.[2] In this paper we derive a general expression for the integrated photon counting rate in an experiment in which an arbitrary number of atomic states are coherently excited by electron impact and the resultant luminescence is detected by an arbitrary system of photon counters. The result is directly applicable to quantum beat and level crossing experiments.

A quantum beat experiment involves the coherent excitation of several (usually just two) non-degenerate but closely spaced atomic levels, and the detection of the subsequent decay radiation. The excitation is performed by a pulse of electrons or radiation which passes through the target (a gas of atoms) within a time Δt . If $\Delta t/\hbar$ is small compared to the inverse of the level spacing, $\hbar\omega$, between the excited levels it is found that the intensity of the decay radiation oscillates in time with frequency ω . In order to obtain good resolution of these oscillations it is clearly necessary that $\Delta t \ll \omega^{-1}$. Since the uncertainty in the energy of the incident beam, $\Delta\epsilon$, is at least $\hbar/\Delta t$ the criterion for good resolution is the same as the condition $\Delta\epsilon \gg \hbar\omega$ which must be satisfied if the incident beam is to be capable of inducing coherent excitation. The effect of the parameter $\omega(\Delta t)$ on the experimental resolution may be seen clearly by comparing the results of Dodd, Kaul, and Warrington[1] $[(\omega/2\pi)\,(\Delta t) \sim 10^{-1}]$ with those of Hadeishi and Nierenberg[2] $[(\omega/2\pi)\,(\Delta t) \sim 2 \times 10^{-2}]$. In a level crossing experiment the target atoms are excited by a continuous beam of electrons or radiation and the total intensity of decay radiation is measured. Interesting effects are observed if the separation of the excited levels is produced by an external field. Then ω can be varied by varying the field, and it is found that the intensity of the decay radiation has a resonance as a function of ω at $\omega = 0$. This effect was first observed by F. Colegrove, et al.[1]

The physical mechanism involved in both quantum beat and level crossing experiments are the same; only the manner in which the decay radiation is detected distinguishes them. Thus it should be possible to give a unified theoretical treatment of both types of experiment. In the present paper this is done by using a wave packet description of the incident electrons. This wave packet description allows us to impose the condition for coherent excitation, $\Delta\epsilon \gg \hbar\omega$, in a straight forward manner. We have chosen to treat the case of electronic excitation because there are already a number of treatments of radiative excitation in existence (e.g., Breit[1] and Franken[1]). Also, it is easier to make a clean physical separation between the excitation and decay processes in the case of electronic excitation.

The main result of this paper is Eq. (43) for the integrated photon counting rate in either a quantum beat or level crossing experiment. This expression brings out the physical connection between these two kinds of experiments. Similar expressions have been derived for the case of radiative excitation and these have been extrapolated in various ways to the case of electronic excitation (e.g., Aleksandrov[2]). However, it appears that there has been no previous derivation of the integrated photon counting rate using wave packet techniques.

The remainder of this paper is divided into three sections and an Appendix. In Section II we consider the excitation process and derive the scattered wave function for the electron-atom system. The decay process is described in Section III, and the expression for the counting rate is obtained. In Section IV we apply our general results to a calculation of the phase of the oscillatory part of the photon counting rate in the Hadeishi-Nierenberg experiment.[2] The effect of exchange scattering is considered in the Appendix.

II. THE EXCITATION PROCESS

The excitation and subsequent decay will be described in terms of a single incident electron and a single infinitely heavy atom* located at the origin. An actual experiment, of course, involves a beam of electrons incident on a many-atom target, and it may be that different scattering events are correlated. Our treatment is applicable only to situations in which individual scattering events are independent. It is perhaps worth noting that by choosing the atom to be initially at rest we are simply making a convenient choice of inertial frame and are not neglecting effects associated with finite atomic velocities. Recoil effects, on the other hand, are completely neglected.

The atomic Hamiltonian will be denoted by h , where h includes the interaction with an external magnetic field if one is present. The internal atomic variables will be denoted by ξ , and the eigenstates of h by $g_a(\xi)$.

$$hg_a(\xi) = W_a g_a(\xi) \quad . \tag{1}$$

For definiteness, we will assume that the atom has a non-degenerate ground state g_0 with energy W_0 , and that it is initially in this state. In order to avoid cumbersome notation the incident electron will be treated as if it were distinguishable from the atomic electrons.

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* We use the word "atom" for convenience. Our results apply equally well to atoms or molecules.

The modifications which are necessary for inclusion of exchange effects are given in the Appendix. The incident electron's kinetic energy operator is T_e , and if a magnetic field is present the interaction energy operator is $-\mu \underline{B} \cdot \underline{\sigma}$. The interaction of the electron's orbital angular momentum with the field will change its trajectory from a straight line to a path which obeys the equation of motion $d\underline{v}/dt = (e/m)\underline{v} \times \underline{B}$. It will be assumed that the Larmor radius is large enough so that this effect may be neglected. It is convenient to choose the positive z - direction in the direction of $\underline{B}$. Then the electron Hamiltonian is $T_e - \mu B \sigma_z$.

We will use the wave packet formalism of Goldberger and Watson[3] to describe the excitation process. The pre-collision wave packet for the electron-atom system is $(\hbar = 1)$,

$$X(t) = \int d^3p\ a(\underline{p} - \underline{q})\ e^{-iEt}\ \chi = e^{-iKt} g(\underline{r}) \overline{\chi} \tag{2a}$$

where

$$\chi = (2\pi)^{-3/2}\ e^{i\underline{p}\cdot\underline{r}}\ u_s g_0(\xi) \ , \tag{2b}$$

$$\overline{\chi} = (2\pi)^{-3/2}\ e^{i\underline{q}\cdot\underline{r}}\ u_s g_0(\xi) \ , \tag{2c}$$

$$E = p^2/2m - \mu B s + W_0 \tag{2d}$$

$$K = T_e - \mu B \sigma_z + h \tag{2e}$$

$$g(\underline{r}) = \int d^3p \; a(\underline{p}) \; e^{i\underline{p}\cdot\underline{r}} \quad . \qquad (2f)$$

In Eqs. (2) $\underline{q}$ is the mean incident momentum and u_s is the initial electron spin wave function with $\sigma_z u_s = s u_s$ where s is either $+1$ or -1. $g(\underline{r})$ is the spatial packeting factor, centered at $\underline{r} = 0$. Thus $X(t)$ is centered at $\underline{r} = 0$ when $t = 0$, i.e., the electron-atom collision occurs at $t = 0$. The complete time dependent solution of the Schrodinger equation corresponding to the precollision wave packet $X(t)$ is ,

$$\Psi(t) = \int d^3p \; a(\underline{p} - \underline{q}) \; e^{-iEt} \, \psi^+ \, , \qquad (3a)$$

where

$$\psi^+ = X + (2\pi)^{-3/2} \sum_{g_a} \sum_{s_a} g_a(\xi) \, u_{s_a} \int d^3p \; \frac{e^{i\underline{p}_a \cdot \underline{r}} \, T_a(\underline{p}_a,\underline{p})}{\Delta_a + i\eta - p_a^{\,2}/2m} \qquad (3b)$$

$$\Delta_a = E - (-\mu B s_a + W_a) \quad . \qquad (3c)$$

In (3b) ψ^+ has been expanded in the complete set of eigenstates of K .

$$K X_a = E_a X_a \quad , \qquad (4a)$$

$$X_a = (2\pi)^{-3/2} \, e^{i\underline{p}_a \cdot \underline{r}} \, u_{s_a} \, g_a(\xi) \quad , \qquad (4b)$$

$$E_a = p_a^2/2m - \mu B s_a + W_a \tag{4c}$$

$T_a(\underline{p}_a,\underline{p})$ is the matrix element of the scattering matrix for scattering from X to X_a. The asymptotic value of the integral in (3b), is

$$\lim_{r\to\infty} \int d^3p_a \frac{e^{i\underline{p}_a \cdot \underline{r}} \, T_a(\underline{p}_a,\underline{p})}{\Delta_a + i\eta - p_a^2/2m}$$

$$= \begin{cases} -\frac{(2\pi)^2 m}{r} \, e^{ip'_a r} \, T_a(p'_a \hat{\underline{r}},\underline{p}) \; ; & \Delta_a > 0 \\ \mathcal{O}\left(\frac{m}{r} \, e^{-r/d_a} \, T_a \right) \; ; & \Delta_a < 0 \end{cases} \tag{5a}$$

where

$$p'_a = (2m\Delta_a)^{\frac{1}{2}} \tag{5b}$$

$$d_a = (-2m\Delta_a)^{\frac{1}{2}} \tag{5c}$$

and $\hat{\underline{r}}$ is a unit vector in the direction of $\underline{r}$. Hereafter we will drop the prime on p'_a and denote it simply as p_a. Dropping terms with $\Delta_a < 0$ and defining the scattering amplitude,

$$f_a(\hat{\underline{r}},\underline{p}) = -(2\pi)^2 m \, T_a(p_a\hat{\underline{r}},\underline{p}) \tag{6}$$

the asymptotic expression for ψ^+ becomes

$$\psi^{+} = \chi + \sum_{g_a}{}' \sum_{s_a}{}' f_a(\hat{\underline{r}},\underline{p}) \frac{e^{ip_a r}}{(2\pi)^{3/2}\, r} u_{s_a} g_a(\xi) \ , \quad (7)$$

where the sum $\sum'_{g_a} \sum'_{s_a}$ includes only those $u_{s_a} g_a$ for which $\Delta_a > 0$.

The scattered wave packet at large distances is obtained by inserting (7) into (3a). In doing this we will assume that the entire incident wave packet lies far enough from any excitation threshold so that $f_a(\hat{\underline{r}},\underline{p})$ is slowly varying over the packet and may be removed from the integral and replaced by $f_a(\hat{\underline{r}},\underline{q})$. It will also be assumed that the distance from threshold is much greater than μB so that $\sum'_{s_a}$ may be replaced by $\sum_{s_a}$ and $\sum'_{g_a}$ may be interpreted as a sum over those atomic states for which $W_a < q^2/2m + W_0$. We will take $q^2/2m$ to be less than the ionization energy so that $\sum'_{g_a}$ includes only discrete states. The resulting asymptotic expression for the scattered wave function of the electron-atom system is,

$$\Psi_{sc}(t) = \Psi(t) - X(t) = \sum_{g_a}{}' \phi_a(\underline{r},t)\, e^{-iht} g_a(\xi) \ , \qquad (8a)$$

where

$$\phi_a(\underline{r},t) = \sum_{s_a} \frac{f_a(\hat{\underline{r}},\underline{q})}{(2\pi)^{3/2}\, r}\, e^{i\mu B s_a t}\, u_{s_a} A_a(\underline{r},t) \qquad (8b)$$

$$A_a(\underline{r},t) = \int d^3p\; a(\underline{p}-\underline{q})\, e^{i[p_a r - (p_a{}^2/2m)t]} \qquad (8c)$$

The modifications of Eqs. (8) which are necessary in order to include exchange scattering are discussed in the Appendix. It is shown there that one need only redefine f_a to be,

$$f_a(\hat{\underline{r}},\underline{q}) = -(2\pi)^2 m\{T_a(p_{\underline{a}}\,\hat{\underline{r}},\underline{q}) - ZT_a^{ex}(p_{\underline{a}}\,\hat{\underline{r}},\underline{q})\} \tag{9}$$

where Z is the number of electrons in the atom and T_a^{ex} is an exchange scattering matrix defined in the Appendix. Here $p_{\underline{a}}$ is p_a evaluated at $p = q$,

$$p_{\underline{a}} = (2m)^{\frac{1}{2}} \left\{\frac{q^2}{2m} - \mu Bs + W_0 - (-\mu Bs + W_a)\right\}^{\frac{1}{2}} \tag{10}$$

Hereafter f_a will be defined by Eq. (9) rather than Eq. (6).

We note in passing that if wave packet spreading is neglected A_a may be put in a form which displays the properties of the scattered electron packet quite clearly. To neglect wave packet spreading we drop terms of order $|\underline{p} - \underline{q}|^2$ in the exponent of (8c),

$$p_a r - \left(\frac{p_a^2}{2m}\right)t = p_{\underline{a}} r - \epsilon_{\underline{a}} t + \frac{\underline{q} \cdot (\underline{p} - \underline{q})}{p_{\underline{a}}} \left(r - \frac{p_{\underline{a}}}{m} t\right) \tag{11a}$$

$$+ \mathcal{O}(|\underline{p} - \underline{q}|^2)$$

where

$$\epsilon_{\underline{a}} = p_{\underline{a}}^2/2m \tag{11b}$$

Substituting (11a) into (8c) and using (2f) we obtain,

$$A_a(\underline{r},t) \approx e^{i(p_a r - \epsilon_a t)} \, g\left(\frac{q}{p_a} \left(r - \frac{p_a}{m} t \right) \right) \tag{12}$$

Thus A_a is an outgoing spherical packet which expands with velocity p_a/m and has radial thickness $(p_a/q)\Delta r$ where Δr is the linear dimension of the incident packet.

III. THE DECAY PROCESS

In order to describe the decay process we will replace h by $H_r = h + K_r + V_r$ in (8a). K_r is the Hamiltonian for the free electromagnetic field, and V_r is the interaction between the electromagnetic field and the atom. The physical reasoning behind this prescription is as follows. The excitation process takes place over a time of order $(m/q)(\Delta r) \approx (m/q)(\Delta q)^{-1} \equiv \Delta t$ where Δr is the spatial extent of $g(\underline{r})$ and Δq is the width of $a(\underline{p})$. The uncertainty in the energy of the incident wave packet is $q\Delta q/m = (\Delta t)^{-1}$. The lifetime of an excited atomic state is Γ^{-1} (say) and its line width is Γ. We will require that $\Delta t \ll \Gamma^{-1}$. This condition may be interpreted either as requiring an experimental situation in which a well resolved temporal behavior of the decay luminescence may be observed $(\Delta t \ll \Gamma^{-1})$ or as requiring a situation in which the uncertainty in the incident electron energy is much greater than the line width $(q\Delta q/m \gg \Gamma)$. During the excitation V_r has little effect and may be neglected in comparison with the electron-atom interaction, but after the atom is excited and the electron has moved away V_r becomes the dominant interaction and causes the atom to decay. Since the excitation is very fast compared to the decay we may describe the decay process by treating the excitation as an impulse at $t = 0$, and this is just what our prescription of replacing h by H_r does. The error associated with this approximation is an error of order Δt in t, e.g., if we obtain a decay law of the form $e^{-\Gamma t}$ it is in error by an amount $e^{-\Gamma(t\pm\Delta t)} - e^{-\Gamma t} \approx \pm \Gamma\Delta t e^{-\Gamma t}$. In order to describe the decay we must include the initial state of the radiation field in (8a); we will neglect background radiation and take this initial state to be the vacuum state. Thus the wave function describing the decay of the excited atom is,

$$\Psi_{dc}(t) = \sum_{g_a}^{\prime} \phi_a(\underline{r},t)\, e^{-iH_r t}\, g_a(\xi)\, |vac\rangle \qquad (13)$$

The decay channel which will be considered here is a single photon emission which leaves the atom in its ground state; it will be assumed that only this decay channel contributes appreciably to the observed decay luminescence. The emitted photons are to be observed by a photon counting apparatus which operates continuously from some negative time onwards. The integrated photon counting rate, i.e., the probability that a photon has been observed prior to time t $(t > 0)$ is the expectation value of the operator,

$$P = \sum_{\underline{k},\hat{\underline{e}}}^{P} |\underline{k},\hat{\underline{e}}\rangle\langle\underline{k},\hat{\underline{e}}| \qquad (14)$$

taken with respect to $\Psi_{dc}(t)$. In (14) $|\underline{k},\hat{\underline{e}}\rangle$ is a one photon eigenstate of K_r with wave vector $\underline{k}$ and polarization $\hat{\underline{e}}$. The photon field will be quantized in a box of volume V_0 so that K_r has discrete eigenstates. The sum $\sum_{\underline{k},\hat{\underline{e}}}^{P}$ includes all photons admitted by the counting apparatus, e.g., the range of k in $\sum_{\underline{k},\hat{\underline{e}}}^{P}$ may be determined by an optical filter, the directional range of $\hat{\underline{k}}$ by the size and location of a photon counter, and the choice of $\hat{\underline{e}}$ by a polarizer. In using (14) we have assumed for the sake of simplicity that the counting apparatus detects all photons in its range of admittance with equal sensitivity; if this were not an adequate approximation we could replace P by $\sum_{\underline{k},\hat{\underline{e}}}^{P} s(\underline{k},\hat{\underline{e}}) \; |\underline{k},\hat{\underline{e}}\rangle\langle\underline{k},\hat{\underline{e}}|$ where $s(\underline{k},\hat{\underline{e}})$ is a sensitivity function, e.g., the dependence of s on k might be determined by the shape of the pass band of an optical filter. The integrated photon counting rate is,

$$\langle P(t)\rangle \;=\; \langle\Psi_{dc}(t)|P|\Psi_{dc}(t)\rangle \;=\; \sum_{\underline{k},\hat{\underline{e}}}^{P} \sum_{g_a} |\langle g_a,\underline{k},\hat{\underline{e}}|\Psi_{dc}(t)\rangle|^2 \qquad (15)$$

where we have multiplied by unity in the form $\sum_{g_a} g_a g_a^{\dagger}$ and used the notation $g_a|\underline{k},\hat{\underline{e}}\rangle = |g_a,\underline{k},\hat{\underline{e}}\rangle$. By assumption a photon $|\underline{k},\hat{\underline{e}}\rangle$ which is included in $\sum_{\underline{k},\hat{\underline{e}}}^{P}$ can be emitted with appreciable probability only by decay of $\Psi_{dc}(t)$ to the ground state, so the only appreciable term in the sum $\sum_{g_a}$ in (15) is the g_0 term. Thus,

$$\begin{aligned}\langle P(t)\rangle &= \sum_{\underline{k},\hat{\underline{e}}}^{P} |\langle g_0,\underline{k},\hat{\underline{e}}|\Psi_{dc}(t)\rangle|^2 \\ &= \sum_{\underline{k},\hat{\underline{e}}}^{P} \Bigg[\sum_{g_a}{}' \langle\phi_a(t)|\phi_a(t)\rangle \; |\langle g_0,\underline{k},\hat{\underline{e}}| \; e^{-iH_r t} |g_a,\text{vac}\rangle|^2\end{aligned}$$

(Equation (16) Contd)

$$+ 2\,\mathrm{Re} \sum_{\substack{g_b, g_a \\ b > a}}' \langle \phi_b(t) | \phi_a(t) \rangle \langle g_0, \underline{k}, \underline{\hat{e}} | \, e^{-iH_r t} \, | g_b, \mathrm{vac} \rangle^* \times \langle g_0, \underline{k}, \underline{\hat{e}} | \, e^{-iH_r t} \, | g_a, \mathrm{vac} \rangle \, . \Bigg] \qquad (16)$$

In (16) the sum $\sum_{g_a}'$ may be restricted to those atomic states which can actually emit a detectable photon, i.e., those states for which $\langle g_0, \underline{k}, \underline{\hat{e}} | \, e^{-iH_r t} \, | g_a, \mathrm{vac} \rangle$ is appreciable when $|\underline{k}, \underline{\hat{e}}\rangle$ is included in $\sum_{\underline{k}, \underline{\hat{e}}}^{P}$. Hereafter we will consider the sum to be so restricted.

We will begin our evaluation of $\langle P(t) \rangle$ by evaluating the scalar product $\langle \phi_b(t) | \phi_a(t) \rangle$. This scalar product is time independent, but we will choose t to be much larger than $\left(\frac{m}{p_{-a}}\right)\left(\frac{p_{-a}}{q}\right) \Delta r$ so that we may use (8b). This gives,

$$\langle \phi_b | \phi_a \rangle = \sum_{s_b, s_a} F_{ba} \, \sigma'_{ba}(\underline{q}) \, \delta_{s_b s_a} \qquad (17a)$$

where

$$\sigma'_{ba}(\underline{q}) = \frac{(p_{-b}\, p_{-a})^{\frac{1}{2}}}{q} \int d\Omega_r \, f_b^* (\underline{\hat{r}}, \underline{q}) \, f_a(\underline{\hat{r}}, \underline{q}) \qquad (17b)$$

$$F_{ba} = \frac{1}{(2\pi)^3} \, \frac{q}{(p_{-b}\, p_{-a})^{\frac{1}{2}}} \int_0^\infty dr \, A_b^*(r,t) \, A_a(r,t) \qquad (17c)$$

Equations (17) apply both to the case $b \neq a$ and the case $b = a$. We now show that, under suitable conditions, F_{ba} reduces to the net incident electron flux at the target. For large t the electron wave packet is localized far from the origin and the lower limit of integration in (17c) may be replaced by $-\infty$. This results in the expression,

$$F_{ba} = \frac{1}{(2\pi)^2} \frac{q}{(p_{\underline{b}} p_{\underline{a}})^{\frac{1}{2}}} \int d^3p\, d^3p'\, a^*(\underline{p}-\underline{q})\, a(\underline{p}'-\underline{q})\, \delta(p_b - p_a) \qquad (18)$$

where p'_a is the same as p_a except that p is replaced by p'. We will let

$$p_{\parallel} = \hat{\underline{q}} \cdot \underline{p} \qquad (19)$$

be the independent variable in $\delta(p_b - p'_a)$. It is readily shown that

$$\delta(p_b - p'_a) = \frac{p'_a}{n} \left[\delta(p_{\parallel} - n) + \delta(p_{\parallel} + n)\right] \qquad (20a)$$

where

$$n = (p'^2 - p_{\perp}^2 + p_{\underline{a}}^2 - p_{\underline{b}}^2)^{\frac{1}{2}} \qquad (20b)$$

$$p_{\perp} = (p^2 - p_{\parallel}^2)^{\frac{1}{2}} \qquad (20c)$$

The integrand of (18) vanishes when $p_{\parallel} = -n$ so F_{ba} reduces to,

$$F_{ba} = \frac{1}{(2\pi)^2} \frac{q}{(p_{\underline{b}} p_{\underline{a}})^{\frac{1}{2}}} \int d^3p\, d^3p'\, a^*(\underline{p}-\underline{q})\, a(\underline{p}'-\underline{q})\, \frac{p_a'}{n}\, \delta(p_{\parallel} - n) \qquad (21)$$

We now introduce the variables ω and W,

$$\omega = \epsilon_{\bar{a}} - \epsilon_{\bar{b}} \tag{22a}$$

$$W = \tfrac{1}{2}(\epsilon_{\bar{a}} + \epsilon_{\bar{b}}) \tag{22b}$$

and expand in powers of ω. To first order in ω Eq. (21) may be rewritten as,

$$F_{ba} = \frac{1}{(2\pi)^2} \frac{q}{p_{\bar{a}}} \left(1 + \frac{\omega}{4W}\right) \int d^3p\, d^3p'\, a^*(\underline{p} - \underline{q})\, a(\underline{p}' - \underline{q})$$

$$\left(\frac{p'_a}{n_0}\right)\left(1 - \frac{m\omega}{n_0^{\,2}}\right)\delta\left(p_{\parallel} - n_0 - \frac{m\omega}{n_0}\right) \tag{23a}$$

where

$$n_0 = (p'^2 - p_\perp^{\,2})^{\frac{1}{2}} \tag{23b}$$

In order to set $\omega = 0$ in (23a) we must require that,

$$\frac{\omega}{W} \ll 1 \tag{24}$$

$$\frac{m\omega}{n_0^{\,2}} \approx \frac{\omega}{q^2/m} \ll 1 \tag{25}$$

$$\frac{m\omega}{n_0} \approx \frac{\omega}{q/m} \ll \Delta q \tag{26}$$

These inequalities are of course satisfied identically if $b = a$. Note that (25) is implied by (26) because $\Delta q \ll q$. If (24) and

(26) are satisfied we may put,

$$F_{ba} = \frac{1}{(2\pi)^2} \frac{q}{p_{\underline{a}}} \int d^3p\, d^3p'\, a^*(\underline{p} - \underline{q})\, a(\underline{p}' - \underline{q})\, \frac{p'_a}{n_0}\, \delta(p_{\parallel} - n_0) \tag{27}$$

The last step in the reduction of F_{ba} is to expand n_0 and p'_a in powers of Δq .

$$n_0 = p'_{\parallel} - \frac{p_{\perp}^2 - p'^2_{\perp}}{2q} + \mathcal{O}(\Delta q)^3 = p'_{\parallel} + \mathcal{O}\left(\frac{(\Delta q)^2}{q}\right) \tag{28}$$

$$\frac{p'_a}{n_0} = \frac{p_{\underline{a}}}{q}\left(1 + \frac{q(p - q)}{2p_{\underline{a}}^2} - \frac{p'_{\parallel} - q}{q} + \mathcal{O}(\Delta q)^2\right) \tag{29}$$

$$= \frac{p_{\underline{a}}}{q}\left[1 + \mathcal{O}\left(\frac{q\Delta q}{p_{\underline{a}}^2}\right) + \mathcal{O}\left(\frac{\Delta q}{q}\right)\right]$$

We see from (28) and (29) that F_{ba} reduces to,

$$F_{ba} = \frac{1}{(2\pi)^2} \int d^3p\, d^3p'\, a^*(\underline{p} - \underline{q})\, a(\underline{p}' - \underline{q})\, \delta(p_{\parallel} - p'_{\parallel}) \tag{30}$$

if

$$\frac{q\Delta q}{p_{\underline{a}}^2} \ll 1 \tag{31}$$

and $\Delta q \ll q$.

The right-hand-side of (30) is the net incident electron flux. The incident electron wave packet is,

$$\phi_{in}(\underline{r},t) = \frac{1}{(2\pi)^{3/2}} \int d^3p \; a(\underline{p} - \underline{q}) \; e^{i(\underline{p} \cdot \underline{r} - (p^2/2m)t)} \tag{32}$$

and the incident flux is

$$F = \int_{-\infty}^{\infty} dz |\phi_{in}(z\hat{\underline{q}},0)|^2 \tag{33}$$

which reduces immediately to (30).

The inequalities we have invoked all have simple physical interpretations. $\Delta q \ll q$ needs no comment. Inequality (31) is equivalent to our requirement that the entire incident wave packet must lie well above any excitation threshold. Inequality (31) (which must hold for all states g_a included in the sum $\sum'_{g_a}$) and inequality (26) taken together imply (24). Thus (26) is the only new condition necessary to reduce F_{ba} to F. The weaker condition $\omega \lesssim q\Delta q/m$ is necessary in order to observe any interference effects at all, i.e., in order that $\langle\phi_b|\phi_a\rangle \neq 0$. The stronger condition (26) ensures that the interference effects will not depend on the detailed structure of the incident wave packet. This is seen most clearly by writing (26) as,

$$\Delta t \ll \omega^{-1} \tag{34}$$

which is an obvious criterion for the observability of well resolved beats in a quantum beat experiment.

The scalar product $\langle \phi_b | \phi_a \rangle$ may now be written as,

$$\langle \phi_b | \phi_a \rangle = F \sigma_{ba}(\underline{q}) \tag{35a}$$

where

$$\sigma_{ba}(\underline{q}) = \sum_{s_b, s_a} \sigma'_{ba}(\underline{q}) \; \delta_{s_b, s_a} \; . \tag{35b}$$

For $b = a$,

$$\sigma_a(\underline{q}) \equiv \sigma_{aa}(\underline{q}) = \sum_{s_a} \frac{p_a}{q} \int d\Omega r \; | \, f_a(\hat{\underline{r}}, \underline{q}) |^2 \tag{36}$$

is the total cross section for excitation of state g_a by an electron of initial momentum $\underline{q}$ and spin projection $s/2$. σ_{ba} with $b \neq a$ may be interpreted roughly as the total cross-section for coherent excitation of states g_a and g_b . Note, however, that σ_{ba} is in general complex.

It remains to calculate the matrix elements of $e^{-iH_r t}$ which occur in $\langle P(t) \rangle$. This is done in Goldberger and Watson[4] and only the results will be presented here. We will consider only the case in which each state in the (restricted) sum $\sum'_{g_a}$ decays to the ground state through an allowed dipole transition and the dominant contributions to the line widths arise from dipole transitions.

In this case $(c = 1)$,

$$\langle g_0, \underline{k}, \hat{\underline{e}} | e^{-iH_r t} | g_a, \mathrm{vac} \rangle = -\left(\frac{2\pi e^2}{V_0 k} \right)^{\frac{1}{2}} \langle g_0 | \frac{\hat{\underline{e}} \cdot \underline{P}}{m} | g_a \rangle$$

$$\times e^{-i(W_0 + k)t} \frac{1 - \exp\{i[k - (W_a - W_0) + i\Gamma_a/2]t\}}{k - (W_a - W_0) + i\Gamma_a/2} \tag{37a}$$

where

$$\underline{P} = \sum_{j=1}^{Z} \underline{p}_j \tag{37b}$$

is the total momentum operator for all the electrons in the atom, and

$$\Gamma_a = \sum_{\substack{g_b \\ W_b < W_a}} \frac{e^2 (W_a - W_b)}{2\pi} \int d\Omega_k \sum_{\hat{\underline{e}}} | \langle g_b | \frac{\hat{\underline{e}} \cdot \underline{P}}{m} | g_a \rangle |^2 \tag{37c}$$

is the natural line width. Using (35a) and (37a) and changing the sum $\sum_{\underline{k}, \hat{\underline{e}}}^{P}$ in (16) to an integral, the non-interferent part of $\langle P(t) \rangle$ becomes,

$$\langle P(t) \rangle_n = F \sum_{g_a}{}' \sigma_a(\underline{q}) \int d\Omega_k \sum_{\hat{\underline{e}}}^{P} \frac{e^2}{(2\pi)^2} | \langle g_0 | \frac{\hat{\underline{e}} \cdot \underline{P}}{m} | g_a \rangle |^2$$

$$\times \int_P dkk \left| \frac{1 - \exp\{i[k - (W_a - W_0) + i\Gamma_a/2]t\}}{k - (W_a - W_0) + i\Gamma_a/2} \right|^2 \tag{38}$$

We will assume that the filter used in the experiment passes all radiation with a frequency in the vicinity of $W_a - W_0$ (for all g_a included in $\sum_{g_a}'$). By "vicinity" we mean a frequency range much wider than Γ_a. This allows us to replace $\int_P dkk$ by $(W_a - W_0)\int_{-\infty}^{\infty} dk$ in (38). The result of this replacement is,

$$\langle P(t)\rangle_n = F \sum_{g_a}' \sigma_a(\underline{q}) \frac{\Gamma_a{}^P}{\Gamma_a} (1 - e^{-\Gamma_a t}) \tag{39a}$$

where

$$\Gamma_a{}^P = \frac{e^2(W_a - W_0)}{2\pi} \int_P d\Omega_k \sum_{\hat{\underline{e}}}^{P} |\langle g_0|\frac{\hat{\underline{e}}\cdot\underline{P}}{m}| g_a\rangle|^2 . \tag{39b}$$

Using (35a) and (37a) the interferent part of $\langle P(t)\rangle$ becomes,

$$\langle P(t)\rangle_i = 2F \,\mathrm{Re} \sum_{\substack{g_b, g_a \\ b > a}}' \sigma_{ba}(\underline{q}) \int_P d\Omega_k \sum_{\hat{\underline{e}}}^{P} \frac{e^2}{(2\pi)^2} \langle g_0|\frac{\hat{\underline{e}}\cdot\underline{P}}{m}|g_b\rangle^*$$

$$\times \langle g_0|\frac{\hat{\underline{e}}\cdot\underline{P}}{m}|g_a\rangle \int_P dkk \left[\frac{1 - \exp\{i[k - (W_b - W_0) + i\Gamma_b/2]t\}}{k - (W_b - W_0) + i\Gamma_b/2}\right]^*$$

$$\times \left[\frac{1 - \exp\{i[k - (W_a - W_0) + i\Gamma_a/2]t\}}{k - (W_a - W_0) + i\Gamma_a/2}\right] \tag{40}$$

The integral over k in (40) may be put in the form,

$$\frac{1}{W_b - W_a + i(\Gamma_b + \Gamma_a)/2} \int_P dkk \left(\frac{1}{k - (W_b - W_0) - i\Gamma_b/2} - \frac{1}{k - (W_a - W_0) + i\Gamma_a/2} \right)$$

$$\times \left(1 - \exp\{i[k - (W_b - W_0) + i\Gamma_b/2]t\} \right)^*$$

$$\times \left(1 - \exp\{i[k - (W_a - W_0) + i\Gamma_a/2]t\} \right) \qquad (41)$$

The overall multiplicative factor of k in (41) will be replaced by $W_b - W_0$ when it multiplies $[k - (W_b - W_0) - i\Gamma_b/2]^{-1}$ and by $W_a - W_0$ when it multiplies $[k - (W_a - W_0) + i\Gamma_a/2]^{-1}$. With these replacements (40) becomes,

$$\langle P(t)\rangle_i = 2F \,\mathrm{Re} \sum_{\substack{g_b, g_a \\ b > a}}{}' \frac{i\sigma_{ba}(q)\, \Gamma_{ba}{}^P}{W_b - W_a + i(\Gamma_b + \Gamma_a)/2}$$

$$\times \left(1 - \exp\{[i(W_b - W_a) - \tfrac{1}{2}(\Gamma_b + \Gamma_a)]t\} \right) \qquad (42a)$$

where

$$\Gamma_{ba}{}^P = \frac{e^2[\frac{1}{2}(W_b + W_a) - W_0]}{2\pi} \int_P d\Omega_k \sum_{\hat{e}}^{P} \langle g_0 | \frac{\hat{e} \cdot P}{m} | g_b \rangle^*$$

$$\times \langle g_0 | \frac{\hat{e} \cdot P}{m} | g_a \rangle \qquad (42b)$$

Adding (39a) and (42a) one obtains the following expression for the total integrated photon counting rate.

$$\langle P(t)\rangle = F\left\{\sum_{g_a}{}' \sigma_a(\underline{q}) \frac{\Gamma_a^P}{\Gamma_a}\left(1 - e^{-\Gamma_a t}\right) - 2\,\mathrm{Im} \sum_{\substack{g_b, g_a \\ b > a}}{}' \frac{\sigma_{ba}(\underline{q})\,\Gamma_{ba}^P}{W_b - W_a + i(\Gamma_b + \Gamma_a)/2}\left(1 - \exp\{[i(W_b - W_a) - \tfrac{1}{2}(\Gamma_b + \Gamma_a)]t\}\right)\right\} \tag{43}$$

The differential counting rate observed in a quantum beat experiment is,

$$\frac{d\langle P(t)\rangle}{dt} = F\left\{\sum_{g_a}{}' \sigma_a(\underline{q})\,\Gamma_a^P\, e^{-\Gamma_a t} + 2\,\mathrm{Re} \sum_{\substack{g_b, g_a \\ b > a}} \sigma_{ba}(\underline{q})\,\Gamma_{ba}^P \exp\{[i(W_b - W_a) - \tfrac{1}{2}(\Gamma_b + \Gamma_a)]t\}\right\} \tag{44}$$

The total number of photons counted in a level crossing experiment is,

$$\langle P(\infty)\rangle = F\left\{\sum_{g_a}{}' \frac{\sigma_a(\underline{q})\,\Gamma_a^P}{\Gamma_a} - 2\,\mathrm{Im} \sum_{\substack{g_b, g_a \\ b > a}} \frac{\sigma_{ba}(\underline{q})\,\Gamma_{ba}^P}{W_b - W_a + i(\Gamma_b + \Gamma_a)/2}\right\} \tag{45}$$

The expression for the counting rate must be modified if there are several possible initial and final states. The cross-sections defined by (35b) depend on the initial electron spin orientation. For an unpolarized incident electron beam these should be replaced by their averages over initial spin orientations,

$$\bar{\sigma}_{ba}(\underline{q}) = \tfrac{1}{2} \sum_{s} \sigma_{ba}(\underline{q}) \qquad (46)$$

There may be more than one possible initial atomic state. We will denote the initial states by a Greek subscript, and let p_α be the probability that g_α is the initial state. Finally, there may be several possible final states which we will denote by a primed Greek subscript. The statement that $g_{\alpha'}$ is a possible final state means that when $\sum_{g_a} g_a\, g_a^\dagger$ is inserted into $\langle P(t) \rangle$ (as in (15)) the term $g_{\alpha'}\, g_{\alpha'}^\dagger$ gives an appreciable contribution. The generalized form of (43) for this situation is,

$$\langle P(t) \rangle = F \sum_{g_\alpha} p_\alpha \sum_{g_{\alpha'}} \Bigg\{ \Bigg[{\sum_{g_a}}' \bar{\sigma}_{a\alpha}(\underline{q}) \frac{\Gamma_{\alpha' a}^{P}}{\Gamma_a} \left(1 - e^{-\Gamma_a t}\right) - 2\,\mathrm{Im} \sum_{\substack{g_b, g_a \\ b > a}} \frac{\bar{\sigma}_{ba\alpha}(\underline{q})\, \Gamma_{\alpha' ba}^{P}}{W_b - W_a + i(\Gamma_b + \Gamma_a)/2} \times \left(1 - \exp\{[i(W_b - W_a) - \tfrac{1}{2}(\Gamma_b + \Gamma_a)]t\}\right) \Bigg] \Bigg\} \qquad (47)$$

$\bar{\sigma}_{a\alpha}$ and $\bar{\sigma}_{ba\alpha}$ are the same as $\bar{\sigma}_a$ and $\bar{\sigma}_{ba}$, respectively, except that the initial atomic state is g_α rather than g_0, and $\Gamma_{\alpha' a}^{P}$ and $\Gamma_{a'ba}$ are the same as Γ_a^{P} and Γ_{ab}^{P}, respectively, except that the final atomic state is g_α, rather than g_0.

In any given experiment there will be corrections to (43) (or (47)) arising from the finite extent of the target and the finite velocity of the atoms. The operator P , as given by (14), is not a strictly correct description of a photon counter, e.g., a counter measures the position of a photon, but does not measure the direction in which it is traveling. However, if it is known that the photon was emitted at the origin, then when the photon is counted it is also known that the portion of the photon wave packet intercepted by the counter consisted of plane waves whose wave vectors lay within the solid angle subtended by the counter at the origin. So P is an effective measurement operator valid for a particular atomic position and velocity. By performing a spatial translation and a velocity translation one can construct an operator valid for an atom at position $\underline{R}$ and moving with velocity $\underline{V}$. The counting rate is then,

$$\overline{\langle P(t)\rangle} = \int d^3R\; d^3V\; \rho(\underline{R})\; F(\underline{V})\; \langle P_{\underline{R},\underline{V}}(t)\rangle \qquad (48)$$

where $\rho(\underline{R})$ is the density of atoms in the part of the target intercepted by the beam and $F(\underline{V})$ is the atomic velocity distribution, both normalized to one. With respect to such corrections we only wish to note that the Doppler effect will be unimportant if the range of k in $\sum^{P}_{\underline{k},\hat{\underline{e}}}$ is sufficiently greater than the Doppler widths of the lines involved in the experiment.

F is to be interpreted experimentally as the net beam flux, and $\langle P(t)\rangle$ as the integrated counting rate per atom in the part of the target intercepted by the beam. In a level crossing experiment the total duration of the beam may be much greater than the value of Δt for individual electrons. The total number of counts is still given by (45) (or the modified version of (45) obtained from (47)) in this case since the increase in the number of counts with increasing beam duration is accounted for by the increase in F with increasing beam duration.

IV. THE HADEISHI-NIERENBERG EXPERIMENT

As a simple application of our general considerations we will calculate the phase of the interferent part of the photon counting rate in the Hadeishi-Nierenberg quantum beat experiment.[2,*] We will not consider corrections arising from finite target volume or finite atomic velocities. Equation (44) will be used as it stands except that σ_a and σ_{ba} will be replaced by $\bar{\sigma}_a$ and $\bar{\sigma}_{ba}$, respectively.

In the Hadeishi-Nierenberg experiment g_0 is the 5^1S_0 ground state of Cd. The atom is in a weak magnetic field (0.88 gauss) and beats are observed between the 5^3P_1, $M_J = \pm 1$ excited states. The experimental configuration is shown in Fig. 1(a). The incident beam is perpendicular to $\underline{B}$, and the emitted photons are intercepted by a counter which subtends a small solid angle about the z-axis. Denote the $M_J = +1$ (-1) excited states by $g_+(g_-)$. Let W_+ and W_- be the energies of g_+ and g_- when $\underline{B}$ is in the positive z-direction as in Fig. 1(a). Then the interferent part of the differential counting rate is,

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* Hadeishi and Nierenberg state that in their experiment the incident electron energy was close to threshold while we have required that the incident electron wave packet lies far from threshold. However, their criterion for an energy close to threshold was $\epsilon_{\underline{a}} \ll W_a$ which clearly does not conflict with our criterion (31). In fact, (31) was well satisfied in the Hadeishi-Nierenberg experiment (T. Hadeishi, private communication).

$$\frac{d\langle P(t)\rangle_i}{dt} = 2F \,\mathrm{Re}\, \bar{\sigma}_{-+}(\underline{q})\, \Gamma^{P}_{-+} \quad e^{i(W_- - W_+)t} \, e^{-\Gamma t} \tag{49}$$

where we have put $\Gamma_+ = \Gamma_- = \Gamma$. In Fig. 1(b) another experimental configuration is shown; it is obtained from Fig. 1(a) by 180° rotation about the x-axis. Clearly the counting rate for the situation pictured in Fig. 1(b) is the same as that for Fig. 1(a). Let us calculate the counting rate for Fig. 1(b) using (44). For a sufficiently weak field g_+ , g_- , $\bar{\sigma}_{-+}(\underline{q})$, and Γ^{P}_{-+} are essentially field independent. Further, Γ^{P}_{-+} is the same for both situations pictured in Fig. 1 since it is invariant under replacement of $\hat{\underline{k}}$ by $-\hat{\underline{k}}$. The energies of g_+ and g_- depend linearly on the field, and the effect of reversing the field is to replace $W_- - W_+$ by $W_+ - W_-$. Thus the counting rate calculated from Fig. 1(b) is,

$$\frac{d\langle P(t)\rangle_i}{dt} = 2F \,\mathrm{Re}\, \bar{\sigma}_{-+}(\underline{q})\, \Gamma^{P}_{-+} \quad e^{i(W_+ - W_-)t} \, e^{-\Gamma t} \tag{50}$$

Comparison of (49) and (50) shows that,

$$\mathrm{Im}\, \bar{\sigma}_{-+}(\underline{q})\, \Gamma^{P}_{-+} = 0 \tag{51}$$

and

$$\frac{d\langle P(t)\rangle_i}{dt} = 2F \, \bar{\sigma}_{-+}(\underline{q})\, \Gamma^{P}_{-+} \quad e^{-\Gamma t} \cos(W_+ - W_-)t \tag{52}$$

According to (52) the oscillations should start out at either a maximum or a minimum. Extrapolation of the data in Hadeishi and Nierenberg's Fig. 2 to $t = 0$ shows that the oscillations start at a maximum which indicates that $\bar{\sigma}_{-+}(\underline{q})\ \Gamma^{P}_{-+}$ is positive.

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APPENDIX: EFFECTS OF EXCHANGE SCATTERING

In this appendix we will justify the use of equation (9). This will be done in a rather brief manner since much of what is said here is also contained in Goldberger and Watson.[5] We first replace the initial unsymmetrized wave packet $X(t)$ by an anti-symmetrized wave packet $X_s(t)$. Since the atomic wave functions $g_a(\xi)$ are already anti-symmetrized we need only anti-symmetrize with respect to interchange of the incident electron and each of the atomic electrons.

$$X_s(t) = (Z + 1)^{\frac{1}{2}} SX(t) \tag{A1a}$$

where S is the projection operator onto anti-symmetrized states

$$S = (Z + 1)^{-1} \sum_{j=0}^{Z} \delta_j Q_j \quad . \tag{A1b}$$

Here Z is the number of electrons in the atom, $\delta_0 = 1$, $\delta_{j\neq 0} = -1$, $Q_0 = 1$, and $Q_{j\neq 0}$ interchanges the incident electron's variables and the jth atomic electron's variables. Q_j is Hermitian and unitary so adjoints and inverses will not be indicated explicitly. Let t_c be a large negative time at which the electron wave packet does not overlap the atom. Let $H = K + V$ where V is the electron-atom interaction. Then since all permutations commute* with H the symmetrized solution of the Schroedinger equation is,

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* This is not strictly true since K does not include the interaction of the incident electron's orbital angular momentum with $\underline{B}$. It is assumed that this is of no physical significance.

$$\Psi_s(t) = e^{-iH(t - t_c)} X_s(t_c) = (Z + 1)^{\frac{1}{2}} S e^{-iH(t - t_c)} X(t_c)$$

$$= X_s(t) + \int d^3p \; a(\underline{p} - \underline{q}) \; e^{-iEt} \psi_{s(sc)}{}^+ \qquad \text{(A2a)}$$

where

$$\psi_{s(sc)}{}^+ = (Z + 1)^{\frac{1}{2}} S(\psi^+ - X) = (Z + 1)^{-\frac{1}{2}} \sum_{j=0}^{Z} \delta_j Q_j \frac{1}{E + i\eta - K} TX \qquad \text{(A2b)}$$

We will expand $\tilde{\Psi}_{s(sc)}(t) \equiv \Psi_s(t) - X_s(t)$ in anti-symmetrized eigen-states of K .

$$\chi_{s(a)} = (Z + 1)^{\frac{1}{2}} S\chi_a \qquad \text{(A3)}$$

In general the states $\chi_{s(a)}$ do not give rise to a simple expansion of the identity, but it is readily verified that

$$\sum_a \chi_{s(a)} \langle \chi_{s(a)} | f_s \rangle = \sum_a \chi_{s(a)} \langle \chi_a | f \rangle = f_s \qquad \text{(A4a)}$$

whenever f_s is of the form

$$f_s = (Z + 1)^{\frac{1}{2}} Sf \qquad \text{(A4b)}$$

where f is an unsymmetrized wave function in which the incident electron is localized far from the origin and the atomic electrons are localized near the origin. The nature of the sum $\sum_a$ in (A4a) is defined by $\sum_a \chi_a \chi_a^\dagger = 1$; it is displayed explicitly in (3b). For sufficiently large positive times $\Psi_{s(sc)}(t)$ is of the form (A4b) (we assume the excited atom is not ionized) and admits the expansion (A4a). Before carrying out the expansion we will put $\psi_{s(sc)}{}^+$ in a more convenient form by using the operator identity

$$Q_j \frac{1}{E + i\eta - K} T = \frac{1}{E + i\eta - K_{j'}} T_{j'j} Q_j \qquad \text{(A5a)}$$

where j' is arbitrary and,

$$T_{j'j} = V_j + V_{j'} \frac{1}{E + i\eta - H} V_j \qquad \text{(A5b)}$$

$$V_j = Q_j V Q_j \qquad \text{(A5c)}$$

$$K_j = H - V_j \quad . \qquad \text{(A5d)}$$

This leads to the expression,

$$\langle \chi_{s(a)} | \psi_{s(sc)}{}^+ \rangle = \frac{(Z+1)^{-1}}{E + i\eta - E_a} \sum_{kj} \delta_k \delta_j \langle Q_k \chi_a | T_{kj} | Q_j \chi \rangle$$

$$= \left[\frac{1}{E + i\eta - E_a} \{ T_a(\underline{p}_a, \underline{p}) - Z T_a{}^{ex}(\underline{p}_a, \underline{p}) \} \right] \qquad \text{(A6a)}$$

where

$$T_a(\underline{p}_a,\underline{p}) = \langle \chi_a | T | \chi \rangle \tag{A6b}$$

$$T_a^{ex}(\underline{p}_a,\underline{p}) = \langle \chi_a | T_{0k} | \chi \rangle \tag{A6c}$$

with $k \neq 0$. $\Psi_{s(sc)}(t)$ is thus,

$$\Psi_{s(sc)}(t) = (Z+1)^{\frac{1}{2}}\, S \sum_a \frac{\chi_a}{E + i\eta - E_a} \{T_a(\underline{p}_a,\underline{p}) - T_a^{ex}(\underline{p}_a,\underline{p})\} \quad . \tag{A7}$$

Following the same line of development as in Sec. II this may be put in the form,

$$\Psi_{s(sc)}(t) = (Z+1)^{\frac{1}{2}}\, S\Psi_{sc}(t) \tag{A8}$$

where $\Psi_{sc}(t)$ is given by equations (8) with f_a defined by (9).

The decay process is now described by,

$$\Psi_{s(dc)}(t) = (Z+1)^{\frac{1}{2}}\, S\Psi_{dc}(t) \tag{A9}$$

which is obtained from (A8) by replacing h with H_r and including the initial state of the radiation field. In the same way that one verifies (A4a) it is readily verified that,

$$\langle P(t) \rangle \equiv \langle \Psi_{s(dc)}(t) | P | \Psi_{s(dc)}(t) \rangle = \langle \Psi_{dc}(t) | P | \Psi_{dc}(t) \rangle \tag{A10}$$

Thus the symmetrization in (A9) may be ignored for the purpose of calculating the photon counting rate.

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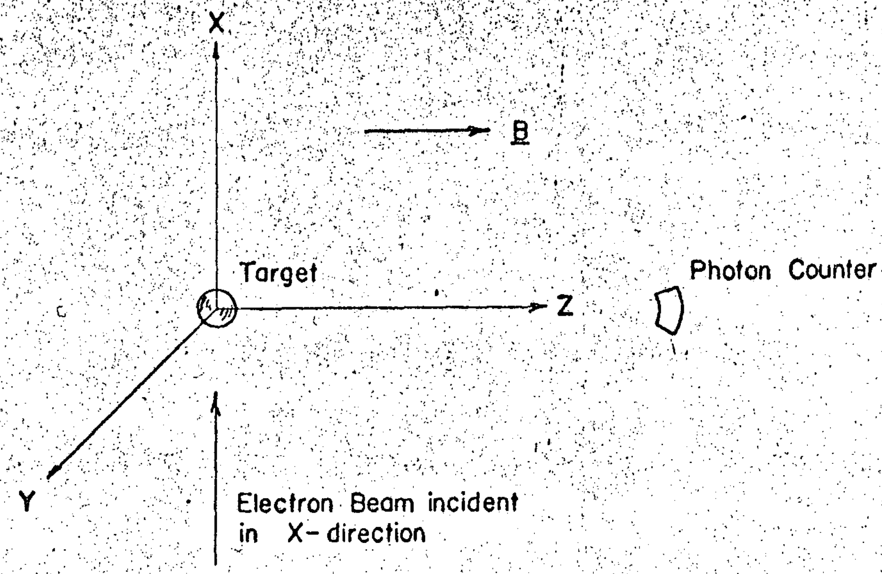
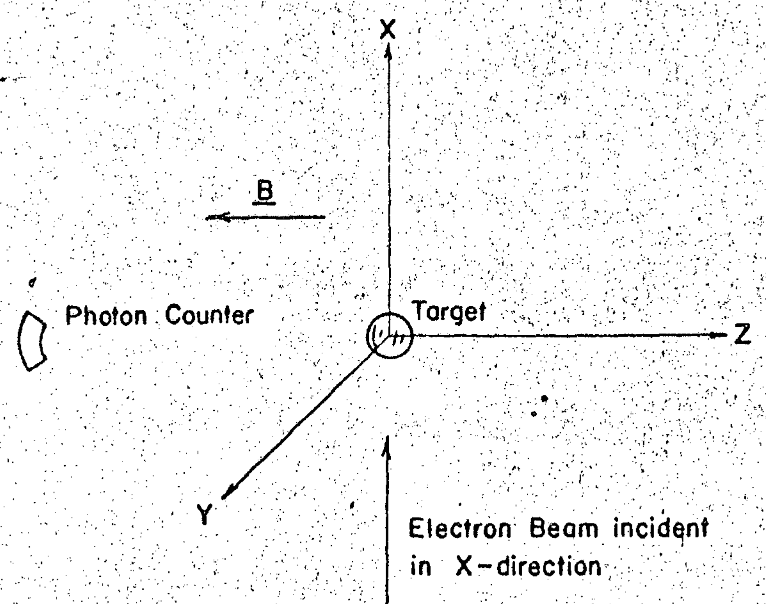
3. M. Goldberger and K. Watson, Collision Theory, John Wiley and Sons, New York, 1964. See particularly pp. 95-101.
4. Ref. 3, Chap. 8. Our Equation (37a) is a special case of Goldberger and Watson's Equation (8.119b), p. 451. See also the discussion of radiative decay on pp. 460-469.
5. Ref. 3, Chap. 4.

FIGURE CAPTION

Fig. 1. (a) Experimental configuration of the Hadeishi-Nierenberg experiment. (b) Experimental configuration obtained from the Hadeishi-Nierenberg configuration by a 180° rotation about the x-axis .


X
B
Target
Photon Counter
Z
Y
Electron Beam incident in X-direction

(a)


X
B
Photon Counter
Target
Z
Y
Electron Beam incident in X-direction

(b)