Use of Intensity Correlations to Determine the Phase of a Scattering Amplitude*

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A new technique is described for observing scattered particles in a scattering experiment. This involves a measurement of the correlated counting rates of two detectors and is based on an idea proposed by Hanbury-Brown and Twiss for astronomical observations. With this technique it is possible, for example, to explicitly measure the phase of a scattering amplitude.

I. INTRODUCTION

MUCH of the understanding of phenomena on a microscopic scale is derived from an analysis of scattering experiments. In most of these, however, the wave properties of matter play little or no role. The elaborate techniques of classical optics, capable in principle of providing useful information, have received little attention for x-ray and particle beams. This is not related to the well-known fundamental limitations on measurements of fields at different space-time points; practical experimental methods are far from achieving these limits. It is, of course, the very short de Broglie wavelength of most particle beams which has prevented straightforward application of conventional interferometric techniques of making correlated space-time measurements. For the same reason the development of extended coherent sources (other than radio frequency and optical masers) has been slow.

We shall show that both of these difficulties associated with very short de Broglie wavelengths may, for a class of interesting experiments, be avoided by a technique for measuring *intensity correlations* at different space-time points. The fact that such measurements are feasible and that they provide useful geometrical information was pointed out in a remarkable paper by Hanbury-Brown and Twiss.[1,2] They showed that observation of the intensity correlations of light (or radio waves) from a star in separated receivers could be used in place of the amplitude correlations of the classical Michelson interferometric method to determine stellar diameters. The technique is a passive one, utilizing the radiation from a spontaneously radiating incoherent source. The principal virtue of the method is that it completely obviates the necessity for maintaining precise phase coherence between the two detectors. The most serious drawback to their technique is the requirement of rather intense illumination of the receivers. For this reason, and probably also because of rather widespread misunderstanding of the principles involved, the range of applications was limited.

We shall discuss here an extension of the Brown-Twiss idea to a new class of problems. The intensity difficulties are circumvented in our application by dealing with irradiated targets rather than with natural emitters. (This is not feasible for the case considered by Brown and Twiss.) Furthermore and most important, their requirement of incoherence of the radiation *from* the target will be shown to be unnecessary. [This does not imply, however, a need for the source (or sources) which illuminate the target to be coherent, as we shall see.] The essentially classical wave interference analysis of Brown and Twiss will be replaced by a quantum mechanical description of particle scattering. There are two reasons for doing this: first, even when the classical picture is valid (as it is for intense electromagnetic radiation), the quantum-mechanical treatment is simpler and completely unambiguous; and second, the same techniques apply to the scattering of particles obeying Fermi-Dirac statistics for which no classical wave theory exists.

The possibility of applying interferometry to microscopic systems is an exciting one. We suggest that one of the more important applications of this new technique will be to a solution of the so-called *phase problem* in the analysis of crystalline and molecular structure by x-ray scattering. Our treatment of the theory will be quite general, but we shall discuss x-ray scattering in the greatest detail.

The theoretical basis for the technique being proposed will be briefly described in Sec. II; the general form of the intensity correlation for an oversimplified situation is given and its relevance for the phase problem is indicated. The remainder of the paper is devoted to obtaining again the results of Sec. II using real sources and detectors so that counting rates and backgrounds may be evaluated. The kinematics of the ele-

* Supported in part by grants from the Air Force Office of Scientific Research, the Atomic Energy Commission, and the National Science Foundation.

[1] R. Hanbury-Brown and R. Q. Twiss, Phil. Mag. 45, 663 (1954); Proc. Roy. Soc. (London) 242A, 300 (1957); 243A, 291 (1957).

[2] E. M. Purcell, Nature 178, 1449 (1956).

mentary scattering interaction is described in Sec. III with attention to the construction of wave packets for the beam particles. The characteristics of the detectors are considered in Sec. IV. In Sec. V the theory of the intensity correlation is developed in detail. The particularly interesting case of intensity correlations when the particles scattered by the elementary scatterers in the target are coherent is discussed in Sec. VI. This is the intensity counterpart of the usual x-ray diffraction experiment and is of greatest importance for the phase problem. In Sec. VII an estimate is made of counting rates and backgrounds. Finally, in Sec. VIII the particular problems posed by a crystalline target are analyzed.

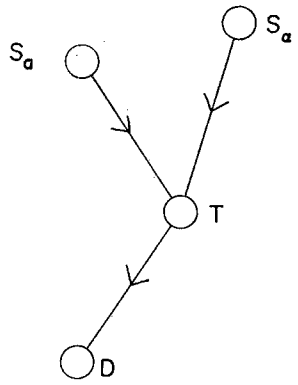

FIG. 2. Illumination of a target with two sources.

II. GENERAL DESCRIPTION OF INTENSITY CORRELATION EXPERIMENTS

In a conventional scattering experiment a source S of beam particles is used to irradiate a target T, as in Fig. 1. Those beam particles scattered through some angle corresponding to a momentum transfer Δk are detected by a detector D. From the counting rate, source intensity and geometry, the differential scattering cross section $\sigma(\Delta k)$ may be determined. In terms of the *scattering amplitude* $\mathfrak{F}(\Delta k)$, the cross section is given by

$$\sigma(\Delta k)=|\mathfrak{F}(\Delta k)|^2. \tag{2.1}$$

It is generally believed that the determination of $\mathfrak{F}(\Delta k)$ would provide all meaningful information concerning the interaction of a beam particle with one of the target particles. Frequently one assumes that the elementary scattering interaction is known and one is interested in determining the geometrical structure of the target. In either case it is $\mathfrak{F}(\Delta k)$ rather than the experimentally measured quantity $|\mathfrak{F}(\Delta k)|$ that is needed; one must have the phase as well as the magnitude of $\mathfrak{F}$. This troublesome problem arises in many contexts, ranging from chemistry to elementary particle physics. One of the most acute of these is the determination of molecular structure (especially of large organic molecules) by x-ray scattering, where the phase problem has received much attention.[3]

In an Appendix we discuss briefly the x-ray phase problem as one of analytic continuation of $\mathfrak{F}$ as a function of complex Δk. It is shown that an integral equation may be written for, say, the real part of $\mathfrak{F}$ which has a *finite* number of *discrete* solutions rather than a unique one. The classification of the ambiguities inherent in the phase problem in this way is interesting and may even be useful in conjunction with other more conventional approaches.[3] It doesn't really solve the problem, however.

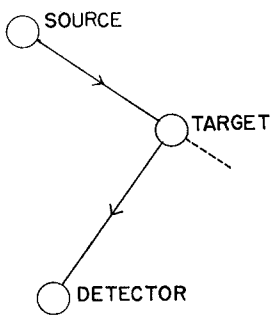

FIG. 1. Conventional scattering experiment.

Mathematical trickery or the elaborate computational schemes widely used in practical x-ray diffraction structure analysis are no substitutes for a direct experimental determination of the phase of the scattering amplitude. Such a determination appears to be feasible through the measurement of intensity correlations, as we shall now show.

To get a feeling for what is involved, consider a modification of the usual scattering experiment of Fig. 1 in which the target T is irradiated by two sources S_α and S_a (see Fig. 2). Particles from these sources scattered by the target into the detector will have undergone momentum transfers $\Delta\mathbf{k}_\alpha$ and $\Delta\mathbf{k}_a$ respectively. If it can be arranged that these scattered particles interfere coherently in the detector, the counting rate which is proportional to $|\mathfrak{F}(\Delta\mathbf{k}_\alpha)+\mathfrak{F}(\Delta\mathbf{k}_a)|^2$ has a term

$$\mathfrak{F}(\Delta\mathbf{k}_\alpha)\mathfrak{F}^*(\Delta\mathbf{k}_a)+\mathfrak{F}^*(\Delta\mathbf{k}_\alpha)\mathfrak{F}(\Delta\mathbf{k}_a). \tag{2.2}$$

By varying the positions of the sources S_α, S_a *independently*, we can cause $\Delta\mathbf{k}_\alpha$ and $\Delta\mathbf{k}_a$ to change independently and thus measure the phase of $\mathfrak{F}(\Delta\mathbf{k})$ to within a constant phase factor (which can be determined from other considerations).

This experiment will, of course, work only if the beam particles are coherently emitted by the two sources. Under ordinary experimental circumstances, particles emitted by two sources will have random phases and the term in the counting rate containing the interesting phase information, Eq. (2.2) will vanish.

By a simple modification of the experiment illustrated in Fig. 2, the lost phase information may be recovered. We replace the single detector D by two spatially separated ones, D_λ and D_l, as shown in Fig. 3. The in-

[3] See, for example, the survey by Encyclopedia of Physics.

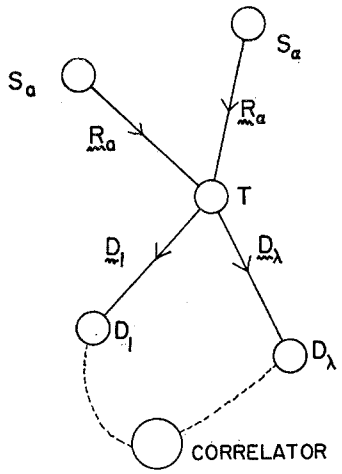

FIG. 3. Arrangement for measuring the correlated response of two detectors, from a target illuminated by two sources.

stantaneous counting rates of D_l and D_λ are multiplied together in a correlator which registers only if D_λ and D_l count in coincidence.[4] Since particles from either source may be scattered into either counter, there will be four scattering amplitudes in the correlator counting rate. We label these by the index pairs which designate source and detector; $\mathfrak{F}(\lambda,\alpha)$, $\mathfrak{F}(l,\alpha)$, $\mathfrak{F}(\lambda,a)$, and $\mathfrak{F}(l,a)$. It is clear, and we shall see it in detail shortly, that the correlator counting rate contains a term[5] proportional to

$$\mathfrak{F}(\lambda,\alpha)\mathfrak{F}^*(l,\alpha)\mathfrak{F}(l,a)\mathfrak{F}^*(\lambda,a)+\text{complex conjugate}. \quad (2.3)$$

By moving the sources and detectors independently of each other, the phase of the scattering amplitude $\mathfrak{F}$ may be determined.

The remarkable feature of this result is that there is a correlation yielding the phase information *in spite of the fact that the two sources S_α and S_a are entirely independent; they emit particles with completely random phases.* Similarly, there is no phase requirement on the detectors. We imagine only that the detector voltage outputs, proportional to the counting rates, are multiplied together in a correlator. The reason that the random phases of the beam particle wave function do not lead to a vanishing of the correlation expressed by Eq. (2.3) is that each index, λ, l, α, and a occurs twice—once in an $\mathfrak{F}$ and once in an $\mathfrak{F}^*$.

The fundamental principle involved here is the quantum mechanical indistinguishability of the two possible ways of getting a coincidence in the two counters. Needless to say, this is the basis of the original Brown-Twiss effect as was so clearly explained by Purcell.[2]

There are many possible variations of the experiment just described. For example, the two sources may be replaced by a single one; the two detectors replaced by one biased to count only coincidences of two or more particles at a time; more than two sources and/or detectors may be used.

Let us consider the two source–two detector experiment shown in Fig. 3 in more detail. We imagine that a given observation is conducted for a time T, during which a large number, n, of beam particles are emitted by both sources. The beam particles are each emitted at a time $t_j (j=1, 2, \cdots, n)$ during the interval T. The wave function describing the scattering of the jth particle is $\phi_j(\mathbf{x},t)$ where $\mathbf{x}(t)$ is the space (time) coordinate of the beam particle. A wave function $|\Phi\rangle$ for the entire scattering experiment is to be constructed from a symmetrized (antisymmetrized) product of the n ϕ_j's when the beam particles satisfy Bose-Einstein (Fermi-Dirac) statistics.

To characterize the counting rate, we introduce the beam particle flux operator $F(l)$ which describes the number of particles per unit area, per unit time at position l in detector D_l. For a detector of unit area which counts all of the particles striking it, the mean counting rate, $C_1(l)$, is given by

$$C_1(l)=\langle\Phi|F(l)|\Phi\rangle. \quad (2.4)$$

This is the average counting rate if the observation (repeated many times) is always over a time interval T, with the system in the *pure state* $|\Phi\rangle$. In practice, the same state $|\Phi\rangle$ is not appropriate for the separate observations. A statistical average over the times of emission, t_j, phases, and numbers n of particles must be performed. We designate this statistical average by a subscript "av." The mean counting rate for an actual experiment will be written as

$$C_1(l)=\langle\Phi|F(l)|\Phi\rangle_{\text{av}}. \quad (2.5)$$

If the counting rate at D_λ is also measured, but *not* multiplied in the correlator by the rate at D_l, its mean value will be

$$C_1(\lambda)=\langle\Phi|F(\lambda)|\Phi\rangle. \quad (2.6)$$

We have assumed that D_λ also has unit area and unit efficiency.

If we now attach the correlator to read the instantaneous outputs of the two detectors, its counting rate will be given by the average of the product of the two flux operators,

$$C_2(\lambda,l)=\langle\Phi|F(\lambda)F(l)|\Phi\rangle_{\text{av}}. \quad (2.7)$$

This is not necessarily equal to the product of $C_1(\lambda)C_1(l)$ and provides the basis for our proposed experiment. The physical origin of the effect lies in the fact that the average correlator reading represents the ensemble average, designated by $\langle\Phi|\cdots|\Phi\rangle_{\text{av}}$, of the instantaneous fluxes, while $C_1(\lambda)C_1(l)$ is the product of the ensemble averages of the two fluxes. The manner in which the ensemble averages are carried out is the critical factor.

Next we express the various counting rates in terms of the beam particle wave functions and ultimately in

[4] A "coincidence" is generally defined by the response-time characteristics of the circuitry of the detectors, as is described in Secs. IV and V.

[5] All other terms in the correlator counting rate may be eliminated by proper circuit design.

terms of the scattering amplitudes. The counting rates $C_1(\lambda)$ or $C_1(l)$ involve the average of a one-particle operator since $F(l)$ is proportional to the density of particles at $l, \Sigma_j\delta(l-\mathbf{x}_j)$. For an appropriately symmetrized product state $|\Phi\rangle$, $C_1(\lambda)$ takes the form

$$C_1(\lambda)=\langle\sum_{j=1}^{n}(\phi_j,F(\lambda)\phi_j)\rangle_{\rm av} \tag{2.8}$$

$$=\langle\sum_{j=1}^{n}v_j\phi_j^*(\lambda,t)\phi_j(\lambda,t)\rangle_{\rm av},$$

where v_j is the group velocity of the wave packet describing the jth particle, and $\langle\cdots\rangle_{\rm av}$ means that the purely statistical ensemble average is to be carried out as described above.

The expectation value encountered in the evaluation of $C_2(\lambda,l)$, Eq. (2.7) is a familiar one, involving a two-particle operator, essentially

$$\sum_{i=1}^{n}\delta(\lambda-\mathbf{x}_i)\sum_{j=1}^{n}\delta(l-\mathbf{x}_j).$$

Writing this out in detail, with the understanding that λ and l are spatially distinct, we find for $C_2(\lambda,l)$,

$$C_2(\lambda,l)=\langle\sum_{i\neq j=1}^{n}\{(\phi_j,F(\lambda)\phi_j)(\phi_i,F(l)\phi_i) \pm(\phi_j,F(\lambda)\phi_i)(\phi_i,F(l)\phi_j)\}\rangle_{\rm av} \tag{2.9}$$

$$=\langle\sum_{i\neq j=1}v_iv_j\{\phi_j^*(\lambda,t)\phi_j(\lambda,t)\phi_i^*(l,t)\phi_i(l,t) \pm\phi_j^*(\lambda,t)\phi_i(\lambda,t)\phi_i^*(l,t)\phi_j(l,t)\}\rangle.$$

We see that it is just by what is commonly referred to as the exchange term that $C_2(\lambda,l)$ and $C_1(\lambda)C_1(l)$ differ.

We may express the single counter rate $C_1(\lambda)$ or the correlated rate $C_2(\lambda,l)$ in terms of the scattering amplitudes $\mathfrak{F}$ by remarking that the wave function $\phi_j(\lambda,l)$ is proportional to

$$(e^{ikD_\lambda}/D_\lambda)F(\lambda,\alpha)\quad\text{or}\quad(e^{ikD_\lambda}/D_\lambda)F(\lambda,a),$$

where D_λ is the distance from target to detector λ, k is the wave number of the scattered particle, and α or a is to be used depending upon the source. Evidently, then, we see that $C_1(\lambda)$ as given by Eq. (2.8) is proportional to

$$|\mathfrak{F}(\lambda,\alpha)|^2+|\mathfrak{F}(\lambda,a)|^2, \tag{2.10}$$

for two sources of equal intensity. In a similar way we see that the first, or so-called direct term in $C_2(\lambda,l)$, Eq. (2.9), is proportional to the product of the expression (2.10) and one just like it with $\lambda\rightarrow l$. The exchange term is seen to be proportional to

$$\mathfrak{F}(\lambda,\alpha)\mathfrak{F}^*(l,\alpha)\mathfrak{F}(l,a)\mathfrak{F}^*(\lambda,a) +\text{complex conjugate}, \tag{2.11}$$

which is just the expression (2.3).

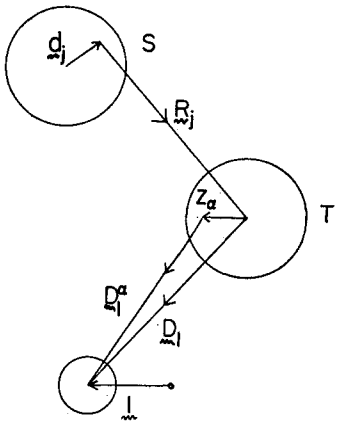

FIG. 4. The single scattering experiment.

In order to use the information contained in Eq. (2.11) we must study carefully the relative magnitude of all contributions to $C_2(\lambda,l)$, to learn whether the interesting one has a significant size. It is clear from the second form of Eq. (2.9) that for $\lambda\approx l$, the direct and exchange terms become equal in magnitude. When the detectors are moved apart, the exchange term oscillates in amplitude (because of the phases of the $\mathfrak{F}$'s), and this is what we want. In practice, the relative magnitude of the direct and exchange terms will be affected by the source and detector dimensions and by the electronic circuitry of the detectors and correlator.

The remainder of the paper will be devoted to investigating all of these questions in a systematic way.

III. DESCRIPTION OF THE SCATTERING PROCESS

Before giving a detailed analysis of the intensity correlation experiment, it is desirable to describe the fundamental scattering process more precisely. This may be done very generally in terms of only the scattering amplitude $\mathfrak{F}$, as introduced in the last section. Because of the specific applications that we have in mind, however, we shall limit the generality of our discussion to scattering by a "loosely bound" target [see the conditions (3.3) and (3.4) below]. This simplification is irrelevant for the observation of intensity correlations, but is useful for describing applications.

As was assumed in the last section, the beam particles may satisfy either Bose-Einstein or Fermi-Dirac statistics. They are emitted by a source S (which may be a composite system of several discrete sources). They are scattered by the target T and counted by the detector D_l, as in Fig. 4.[6] The target T is assumed to consist of N mutually bound particles, not necessarily identical. These bound particles are regarded as the elementary scatterers of the beam particles; their choice is largely a matter of convenience. For example, they might be the atoms forming organic molecules, which in turn are bound in a crystal lattice. (We defer to the next section a discussion of the detectors.)

[6] The second detector will be added later.

Each beam particle is assumed to have a spin S and an energy ϵ_p when its momentum is $\mathbf{p}$. A typical plane-wave function for a beam particle is written as χ_a, where "a" labels both the momentum and spin orientation. The N target particles are assigned spatial coordinates and spin variables $\mathbf{z}_\alpha$ and s_α ($\alpha=1, \cdots, N$), respectively. The wave function for the target system, prior to the scattering, is written as $g_0(\mathbf{z}_1,s_1; \mathbf{z}_2,s_2,\cdots\mathbf{z}_N,s_N)$, or more compactly as $g_0(\mathbf{z},s)$. The steady-state wave functions describing the scattering of a single-beam particle by the (entire) target are written in the conventional notation[7,8] as $\psi_a{}^+$. (The superscript "+" implies that asymptotically the wave function consists of the sum of the "initial state," $\chi_a g_0(\mathbf{z},s)$, together with *outgoing* scattered particles.) If the Hamiltonian for the system (beam particle plus target) is H, $\psi_a{}^+$ satisfies the Schrödinger equation (with the boundary conditions noted above),

$$H\psi_a{}^+=E_a\psi_a{}^+, \tag{3.1}$$

where

$$E_a=\epsilon_p+W_0, \tag{3.2}$$

with W_0 the energy of the target in the state g_0.

Since we shall ultimately be interested in exploring the geometrical structure of the target, it is desirable that the dynamics of the elementary scattering events be as simple as possible. Therefore, we make two assumptions concerning the scattering process. First, we assume the condition of *weak binding* obtains. By this, we mean that the ratio,

$$\frac{\text{kinetic energy of a beam particle}}{\text{magnitude of average binding energy of a single target particle}}\gg 1. \tag{3.3}$$

This condition simplifies the description of the scattering interaction at the target.[9] It is ordinarily satisfied with great accuracy by x-ray and electron scattering from chemically bound targets. The second condition that we require is that the ratio,

$$\frac{\text{scattering mean free path within } T}{\text{size of target } T}\gg 1. \tag{3.4}$$

This permits us to neglect multiple scattering of the beam particles within the target. In the language of ordinary x-ray theory, our condition (3.4) corresponds to the neglect of both primary and secondary extinction. This limitation is no more essential in principle for us than is it in the standard case.

For the process of interest to us, namely the correlation scattering experiment, it is important to consider the times of emission, scattering, and arrival at the detector of each beam particle. For this purpose we must construct wave packet states from the $\psi_a{}^+$ which permit a spatiotemporal localization of the particles (to within the limits, of course, set by the uncertainty principle and the experimental conditions). Using a symbol j ($j=1, 2, \cdots$) to designate an individual beam particle, we have[10]

$$\psi_{\bar{a}(j)}(t)=\int d^3p e^{-iE_at}A_j(\mathbf{p})\psi_{a(j)}{}^+, \tag{3.5}$$

where $A_j(\mathbf{p})$ is the amplitude (to be specified more precisely below) corresponding to an initial momentum $\mathbf{p}$ for the particle (recall that $E_a=\epsilon_p+W_0$), and $\psi_{a(j)}{}^+$ is a particular steady state $\psi_a{}^+$ for beam particle j. The amplitude $A_j(\mathbf{p})$ is assumed to satisfy the normalization condition,

$$\int d^3p\,|A_j(\mathbf{p})|^2=1. \tag{3.6}$$

It is convenient to limit ourselves to wave packets of particles of a rather well-defined energy ϵ_{κ_j} and momentum $\boldsymbol{\kappa}_j$ ($j=1, 2, \cdots$) in the sense that

$$A_j(\mathbf{p})\simeq 0, \quad \text{unless} \quad |\mathbf{p}-\boldsymbol{\kappa}_j|\ll|\boldsymbol{\kappa}_j|. \tag{3.7}$$

Finally, we shall choose the phases of the $\psi_{a(j)}{}^+$ in such a manner that if t_j is the time at which the particle j is emitted by the source, irrespective of the location on the source,

$$A_j(\mathbf{p})=e^{iE_at_j}A^j(\mathbf{p}), \tag{3.8}$$

where $A^j(\mathbf{p})$ is independent of t_j and of the coordinates of the emission point.

We require the wave function $\psi_{\bar{a}(j)}$ in the vicinity of the detectors, which in practice will evidently be placed at a distance large compared to $\hbar/\kappa_j$ from the target. Consequently, only the asymptotic form of $\psi_{a(j)}{}^+$ is needed for the evaluation of Eq. (3.5). This in turn may be given explicitly under our assumptions of weak binding, Eq. (3.3), and single scattering, Eq. (3.4). We define (see Fig. 4)

$$\mathbf{R}_j{}^\alpha\equiv\mathbf{R}_j+\mathbf{Z}_\alpha \tag{3.9}$$

as the vector from the source point $\mathbf{d}_j$ of beam particle j to the coordinate $\mathbf{z}_\alpha$ of the scattering particle α. Also, we define

$$\mathbf{D}_l{}^\alpha\equiv\mathbf{D}_l-\mathbf{Z}_\alpha \tag{3.10}$$

as the vector from the coordinate $\mathbf{z}_\alpha$ to a point $\boldsymbol{l}$ in the detector D_l. We may write, then, according to our

[7] M. Gell-Mann and M. L. Goldberger, Phys. Rev. **91**, 398 (1953).

[8] M. L. Goldberger and K. M. Watson, *Collision Theory* (John Wiley & Sons, Inc., New York, 1963), Chaps. III and V.

[9] A thorough discussion of the weak binding condition is given in Secs. 11.A and 11.B of Ref. 8.

[10] See Ref. 8, Chap. III, for a detailed description of the scattering of wave packets. See also E. Merzbacher, *Quantum Mechanics* (John Wiley & Son, Inc., New York, 1961).

assumptions,

$$\psi_{a(j)}{}^{+}(l) = \left\{ \chi_a + \sum_{\alpha=1}^{N} (2\pi)^{-3/2} \frac{e^{ipD_l{}^{\alpha}}}{D_l{}^{\alpha}} f_\alpha(\hat{D}_l{}^{\alpha}, \hat{p}) e^{ip\cdot \mathbf{R}_j{}^{\alpha}} \right\} g_0(z,s) \equiv \varphi_{a(j)}{}^{+}(l) g_0(\mathbf{z},s). \quad (3.11)$$

Here the quantity $f_\alpha(\hat{D}_l{}^{\alpha}, \hat{p})$ represents the scattering amplitude for scattering a beam particle by the target particle α, the momentum of the beam particle being parallel to $\hat{p}(\hat{D}_l{}^{\alpha})$ before (after) the scattering. The scattering amplitude f_α is in general one column of the scattering amplitude matrix in the spin space of the beam particle and of particle α. The quantity,

$$\sigma_\alpha = |f_\alpha|^2, \quad (3.12a)$$

where f_α is a given matrix element of the scattering amplitude,[11] is the differential scattering cross section for scattering between pure spin states of the beam particle on the target particle α. The cross section averaged over spin states is

$$\bar{\sigma}_\alpha = S|f_\alpha|^2, \quad (3.12b)$$

where S represents a sum over final and an average over initial spin states. The factor $\exp(i\mathbf{p}\cdot\mathbf{R}_j{}^{\alpha})$ in Eq. (3.11) appears as a result of our phase convention (3.8), since this accounts for the "travel time" from $\mathbf{d}_j$ to $\mathbf{z}_\alpha$. The sum over α in Eq. (3.11) corresponds, of course, to the adding of waves scattered from each of the target particles.

Strictly speaking, the expression we have written for $\psi_{a(j)}{}^{+}$ is correct only for what is called *quasielastic* scattering. This is scattering under conditions such that the momentum transfer to a target particle is not large compared to the range of momenta which it has in the initial state, g_0.[12] For larger momentum transfers we may still use Eq. (3.11) if we interpret the coordinates $\mathbf{z}_\alpha$ in g_0 to be "displaced coordinates" and appropriately shift the momentum of the scattered particle in accordance with energy conservation. For our eventual applications (to intensity measurements) we need not bother to do this. The reason is that the corrections are required only when the scattering is inelastic, or incoherent, in which case there is no interference among the scattered waves in Eq. (3.11). We shall see that this lack of interference is taken care of, for our purposes, automatically in Eq. (3.11).[13] We may simply use it as it stands provided the weak binding and single-scattering conditions are obtained.

To give the scattered wave in Eq. (3.5) an explicit form we introduce a wave packet amplitude $G^j(\mathbf{x})$ defined by

$$G^j(\mathbf{x}) = \int d^3k' A^j(\mathbf{k}'+\boldsymbol{\kappa}_j)[e^{i\mathbf{k}'\cdot\mathbf{x}}/(2\pi)^{3/2}]. \quad (3.13a)$$

We note that

$$\int d^3x |G^j(\mathbf{x})|^2 = 1 \quad (3.13b)$$

according to Eq. (3.6), so that $|G^j(\mathbf{x})|^2$ represents the density of beam particle j at the point $\mathbf{x}$. This function G^j is so chosen that it is different from zero only in the neighborhood of $\mathbf{x}=0$. To see the significance of G^j a little more clearly, consider the wave function of a free-beam particle chosen to be located near a point $\mathbf{R}$ at time $t=t_j$ as it evolves in time in the absence of a scatterer. Its wave function would be

$$\begin{aligned}\psi^j(\mathbf{x}) &= \int \frac{d^3p}{(2\pi)^{3/2}} A^j(\mathbf{p}) \exp[-i\epsilon(p)(t-t_j)+i\mathbf{p}\cdot(\mathbf{x}-\mathbf{R})] \\ &\cong \exp[-i\epsilon(\kappa_j)(t-t_j)+i\boldsymbol{\kappa}_j\cdot(\mathbf{x}-\mathbf{R})] \\ &\quad \times \int \frac{d^3k}{(2\pi)^{3/2}} A^j(\mathbf{k}+\boldsymbol{\kappa}_j) \exp[ik\cdot(\mathbf{x}-\mathbf{R}-\mathbf{v}_j(t-t_j))] \\ &= \exp[-i\epsilon(\kappa_j)(t-t_j)+i\boldsymbol{\kappa}_j\cdot(\mathbf{x}-\mathbf{R})] \\ &\quad \times G(\mathbf{x}-\mathbf{R}-\mathbf{v}_j(t-t_j)),\end{aligned}$$

where $\mathbf{v}_j \equiv \nabla_{\kappa_j}\epsilon_{\kappa_j}$ is the group velocity (which for x rays is just the velocity of light). We have neglected the spreading of the wave packet here, which is legitimate under most experimentally interesting conditions.[14]

Using the fact that, in forming the wave packet state described by Eq. (3.5), $p \cong \kappa_j + (\mathbf{p}-\boldsymbol{\kappa}_j)\cdot\hat{\kappa}_j$, together with our phase condition (3.8), we find (for a point $\boldsymbol{l}$ not in the path of the incident beam)

$$\psi_{\bar{a}(j)} = \sum_{\alpha=1}^{N} \frac{e^{i\kappa_j D_l{}^{\alpha}}}{D_l{}^{\alpha}} e^{i\boldsymbol{\kappa}_j\cdot\mathbf{R}_j{}^{\alpha}} f_\alpha(\hat{D}_l{}^{\alpha}, \hat{\kappa}_j) \times G^j\{\hat{\kappa}_j[D_l{}^{\alpha} - v_j(t-t_j)] + \mathbf{R}_j{}^{\alpha}\} \times g_0 e^{-iE_{\bar{a}(j)}(t-t_j)}, \quad (3.14a)$$

where

$$E_{\bar{a}(j)} = W_0 + \epsilon_{\kappa_j}$$

and

$$\mathbf{v}_j = \nabla_{\kappa_j}\epsilon_{\kappa_j} = \hat{\kappa}_j d\epsilon_{\kappa_j}/d\kappa_j,$$

as above. We have again assumed that spreading of the wave packet may be neglected and that the time t is so

[11] We use the same symbol for both the matrix f_α and a single element of this matrix. The distinction will be clear from the context in which f_α is used.

[12] See Secs. 11.A and 11.B of Ref. 5 for an extended discussion of this point.

[13] A kinematic correction is required, however. This amounts to replacing Eq. (3.12) by the cross section for a free target particle α. If we simply replace σ_α in our final expressions by the cross section for a free target particle, we need not worry about this point. See Ref. 5, Sec. 11.B.

[14] The neglect of wave packet spreading is irrelevant for our final results, however.

large that the asymptotic form (3.11) of the wave packet may be used.

The source point $\mathbf{d}_j$ must clearly lie at that location on the source for which

$$\hat{R}_j{}^\alpha=\hat{\kappa}_j, \tag{3.15}$$

since the "direction of travel" must be parallel to the momentum vector of the particle. This permits us to rewrite Eq. (3.14a) as

$$\psi_{\bar{a}(j)}=\sum_{\alpha=1}^{N}\frac{e^{i\kappa_j D_l{}^\alpha}}{D_l{}^\alpha}e^{i\kappa_j R_j{}^\alpha}f_\alpha(\hat{D}_l{}^\alpha,\hat{R}_j{}^\alpha)\times G^j\{\hat{\kappa}_j[D_l{}^\alpha+R_j{}^\alpha-v_j(t-t_j)]\}\times g_0 e^{-iE_{\bar{a}(j)}(t-t_j)}. \tag{3.14b}$$

This expression vanishes unless

$$t\simeq t_j+1/v_j(D_l{}^\alpha+R_j{}^\alpha),$$

which specifies the time of arrival of the wave packet at the detector.

We have so far considered the scattering of only a single-beam particle. In order to discuss any process involving the interference of different beam particles, we must take into account the scattering of many particles. To do this we imagine that a given observation is carried out for a time interval T (large compared to the beam flight time from source to detector), during which time there are $n(\gg 1)$ beam particles emitted by the source. We assume that any cumulative change in the target (i.e., in the state g_0) due to previous scatterings may be neglected when we are studying any given scattering. Such an experimental constraint seems necessary if target structure is to be revealed by successive scattering events.[15]

In terms of the wave function $\varphi_{a(j)}{}^+$ introduced in connection with Eq. (3.11) we may define a new time-dependent wave packet, analogous to Eq. (3.5):

$$\Phi_{\bar{a}(j)}(\mathbf{r},t)=\int d^3p e^{-iE_a t}A_j(\mathbf{p})\varphi_{a(j)}{}^+(\mathbf{r}). \tag{3.16}$$

Here the coordinate $\boldsymbol{l}$ has been replaced by the general point $\mathbf{r}$. In the asymptotic region we find from Eq. (3.14b) [by simply striking out the factor $g_0(\mathbf{z},s)$]

$$\Phi_{\bar{a}(j)}(\mathbf{r},t)=\sum_{\alpha=1}^{N}\frac{e^{i\kappa_j D_r{}^\alpha}}{D_r{}^\alpha}e^{i\kappa_j R_j{}^\alpha}f_\alpha\times G^j(\hat{\kappa}_j[D_r{}^\alpha+R_j{}^\alpha-v_j(t-t_j)])\times e^{-iE_{\bar{a}(j)}(t-t_j)}. \tag{3.17}$$

We have again assumed that $\mathbf{r}$ does not lie in the path of the unscattered beam.

[15] We would not, for example, want the target to be melted or vaporized or have even the lattice structure changed during the course of the experiment.

The symmetrized product wave function for the scattering of the n beam particles is[16]

$$\Phi(r_1,\cdots r_n;t)=\frac{1}{(n!)^{1/2}}\sum_Q \epsilon_Q\Phi_{\bar{a}(1)}(\mathbf{r}_{Q_1},t)\times\Phi(\cdots)\cdots\Phi_{\bar{a}(n)}(\mathbf{r}_{Q_n},t). \tag{3.18}$$

Here Q represents a general permutation of the beam particle coordinates (now including spin variables) $\mathbf{r}$, so that under the permutation Q, $\mathbf{r}_1\rightarrow\mathbf{r}_{Q_1},\cdots,\mathbf{r}_n\rightarrow\mathbf{r}_{Q_n}$. The quantity ϵ_Q has the values

$$\begin{aligned}&\epsilon_Q=1 \text{ for Bose-Einstein statistics;}\\ &=-1 \text{ if } Q \text{ is an odd permutation of a standard order,}\\ &=+1 \text{ if } Q \text{ is an even permutation of a standard order, Fermi-Dirac.}\end{aligned} \tag{3.19}$$

The complete state vector describing the scattering experiment is then

$$|\Phi\rangle=\int d^3r_n\Phi(\mathbf{r}_1,\cdots,\mathbf{r}_n;t)\,|\,\mathbf{r}_1,\mathbf{r}_2\cdots\mathbf{r}_n\rangle g_0(\mathbf{z},s), \tag{3.20}$$

where $|\,\mathbf{r}_1\cdots\mathbf{r}_n\rangle$ is a basis vector for the n beam particles. The integral over the $\mathbf{r}$'s is to be understood as including a sum over spin variables.[17]

IV. DESCRIPTION OF THE DETECTORS

In this section we shall construct a counting rate operator for the detectors. It is clear that this must involve essentially the flux operator at the detector, but should properly take into account the geometric efficiency of the detector as well as any time-delay characteristics of its response. Since the detectors are generally far from the target (i.e., many wavelengths away) we are concerned only with the asymptotic form of the scattering wave functions. Since such functions may be represented as a superposition of plane-wave states, it is natural to introduce a complete set of such states $\chi_s(\mathbf{x})$ for the beam particles. The index s labels both the momentum k and the spin orientation of the particle. We introduce creation (annihilation) operators $a_s{}^\dagger(a_s)$ for these states satisfying the usual commutation relations[18]:

$$\begin{aligned}&[a_s,a_{s'}{}^\dagger]_\pm=\delta_{s,s'},\\ &[a_s,a_{s'}]_\pm=[a_s{}^\dagger,a_{s'}{}^\dagger]=0.\end{aligned} \tag{4.1}$$

In order to take into account the finite counter size, geometric efficiency, and response time characteristics, the simple flux operator introduced in Sec. II which was simply proportional to the particle density, Eq.

[16] The notation here is that of Sec. 4.B of Ref. 8.

[17] The use of space coordinates to represent asymptotic photon wave functions is described in Sec. 9.B of Ref. 5.

[18] Here $[A,B]_\pm=AB\pm BA$, where the plus (minus) sign is to be used for Fermi-Dirac (Bose-Einstein) statistics.

(2.5) must be generalized. This is most easily done in terms of "field operators" constructed from the operators (4.1). These are

$$\varphi(\mathbf{x})=\sum_s \gamma(k)a_s\chi_s(\mathbf{x}),$$
$$\varphi^\dagger(\mathbf{x})=(\varphi(\mathbf{x}))^\dagger. \tag{4.2}$$

If it were not for the presence of $\gamma(k)$, these would be the usual configuration space second-quantization operators satisfying the commutation relations

$$[\varphi(\mathbf{x}),\varphi^\dagger(\mathbf{x}')]_\pm=\delta(\mathbf{x}-\mathbf{x}'), \tag{4.2'}$$

such that operating on an n-particle state $|\mathbf{x}_1,\mathbf{x}_2,\cdots\mathbf{x}_n\rangle$

$$\varphi^\dagger(\mathbf{x})\varphi(\mathbf{x})|\mathbf{x}_1,\cdots,\mathbf{x}_n\rangle$$
$$=[\sum_{i=1}^{n}\delta(\mathbf{x}-\mathbf{x}_i)]|\mathbf{x}_1,\cdots,\mathbf{x}_n\rangle. \tag{4.2''}$$

The reader who is unfamiliar with the techniques of second quantization may retain his sanity (and understanding of the subsequent manipulations) by remembering Eq. (4.2).

The function $\gamma(k)$ will be chosen to take into account the efficiency of the counter; it is related to the actual counter efficiency $\Gamma(k)$ for particles of momentum k by

$$|\gamma(k)|^2=v(k)\Gamma(k), \tag{4.3}$$

where $v(k)$ is the velocity of a particle of momentum $\mathbf{k}$. If the counter were localized at $\mathbf{x}$ and had instantaneous response characteristics, the counting rate operator would be

$$R_0(\mathbf{x})\equiv\varphi^\dagger(\mathbf{x})\varphi(\mathbf{x}). \tag{4.4}$$

Were $\Gamma(k)=1$, this would be just the flux operator of Eq. (2.5).

Next we consider the bandwidth characteristics of the counter. To do this we must introduce a time-delayed counting rate operator and a transfer function which describes the manner in which instantaneous signals are transformed into actual recorded ones after a time delay inherent in the apparatus. The total kinetic energy operator for the n beam particles is

$$K=\sum_{\nu=1}^{n}K_\nu. \tag{4.5}$$

Here K_ν is the kinetic energy operator for the νth beam particle. If this particle has been absorbed, the corresponding K_ν is taken to vanish; otherwise for the one-particle plane-wave state $\chi_s(\mathbf{x}_\nu)$,

$$K_\nu\chi_s=\epsilon_k\chi_s, \tag{4.6}$$

where ϵ_k is the particle energy. We may now define the *time-delayed* counting rate operator $R(\mathbf{x},\tau)$ by

$$R(x,-\tau)\equiv e^{-iK\tau}R_0(\mathbf{x})e^{iK\tau},$$
$$=\varphi^\dagger(\mathbf{x},-\tau)\varphi(\mathbf{x},-\tau), \tag{4.7}$$

where

$$\varphi(\mathbf{x},-\tau)=e^{-iK\tau}\varphi(\mathbf{x})e^{iK\tau}=\sum_s\gamma(k)A_se^{i\epsilon_k\tau}\chi_s(\mathbf{x}),$$
$$\varphi^\dagger(\mathbf{x},-\tau)=[\varphi(x,-\tau)]^\dagger. \tag{4.8}$$

The operator $R(\mathbf{x},-\tau)$ evidently introduces a delay τ in registering each particle counted. A counter with finite bandwidth characteristics is then represented by a counting rate operator

$$R(\mathbf{x})\equiv\int_0^\infty d\tau L(\tau)R(\mathbf{x},-\tau), \tag{4.9}$$

where

$$L(\tau)=\int_0^\infty\frac{d\omega}{2\pi}e^{-i\omega\tau}B(\omega). \tag{4.10}$$

The quantity $B(\omega)$ gives the frequency characteristics of the counter "behind the square law detector."

Finally, the finite detector size may be taken into account by supposing that Eq. (4.9) describes the counting rate for an element of the counter (which might be an atom or molecule or small area element located at $\mathbf{x}=\boldsymbol{l}$). The complete counting rate operator for the detector D_l in Fig. 4 is obtained by summing these "elements:"

$$R_l(t)=\sum_{\boldsymbol{l}}R(\boldsymbol{l}). \tag{4.11}$$

The average counting rate $C_1(\boldsymbol{l})$ introduced in Sec. II, Eq. (2.5) becomes[19]

$$C_1(l)=\langle\Phi|R_l(t)|\Phi\rangle_{\rm av},$$
$$=\sum_l\int_0^\infty d\tau L(\tau)J'(\boldsymbol{l},t-\tau), \tag{4.12}$$

where

$$J'(\boldsymbol{l},t-\tau)=\langle\Phi|\phi^\dagger(\boldsymbol{l},-\tau)\phi(\boldsymbol{l},-\tau)|\Phi\rangle_{\rm av}. \tag{4.13}$$

The state $|\Phi\rangle$ was defined by Eq. (3.20).

The evaluation of J', using Eqs. (3.18) and (3.20) is relatively straightforward. It is helpful to mentally express the wave functions in $|\Phi\rangle$ in terms of a sum over the complete set of plane-wave states while carrying out the manipulations. The result is almost self-evident so we shall omit the details. We find, using Eq. (4.3),

$$J'(\boldsymbol{l},t)=\langle\sum_{j=1}^{n}\Gamma(\kappa_j)v_j(g_0,\Phi_{\bar{a}(j)}^\dagger(\boldsymbol{l},t)\Phi_{\bar{a}(j)}(\boldsymbol{l},t)g_0)\rangle_{\rm av}. \tag{4.14}$$

The asymptotic form, Eq. (3.17), may be used for the

[19] In doing this we make use of the ensemble average, denoted by "av," to introduce a phase randomization which will ensure the orthogonality of the various $\phi_{\bar{a}(j)}$'s.

scattering wave functions ϕ in Eq. (4.14) to give

$$J'(l,l)=\frac{1}{D_l^2}\Big\langle\sum_{j=1}^{n}\Gamma(\kappa_j)v_j\sum_{\alpha,\beta=1}^{N}\{g_0,\,G^j((\hat{\kappa}_j[D_l{}^\alpha+R_j{}^\alpha-v_j(t-t_j)])G^{j*}(\hat{\kappa}_j[D_l{}^\beta+R_j{}^\beta-v_j(t-t_j)])$$
$$\times\exp[i\kappa_j(D_l{}^\alpha-D_l{}^\beta+R_j{}^\alpha-R_j{}^\beta)]f_\beta{}^\dagger f_\alpha g_0\}\Big\rangle_{\rm av}. \quad (4.15)$$

We have assumed that the target is small enough that $[D_l{}^\alpha D_l{}^\beta]^{-1}$ may be replaced by $D_l{}^{-2}$; it is trivial to insert the exact factors at any time. Since the f_α's are column (spin) matrices, we write $f_\beta{}^\dagger f_\alpha$ to indicate a matrix product.

The simplest application of Eq. (4.15) is to the case of incoherent scattering. To avoid any possible misunderstanding, we remark that by incoherent scattering we mean that the waves scattered from two different elementary scatterers do not interfere. Thus all terms in the sum in Eq. (4.15) for which $\alpha\neq\beta$ drop out. If the energy spread of the beam is sufficiently narrow, the counter efficiency, $\Gamma(\kappa_j)$, may be taken to be a constant; Γ, and the differential cross-section $\bar{\sigma}_\alpha=\langle f_\alpha{}^\dagger f_\alpha\rangle$ [Eq. (3.12b)] removed from the sum over j. We find then that J' becomes

$$J'(l,l)=\frac{\Gamma}{D_l^2}\sum_\alpha\bar{\sigma}_\alpha\{\langle\sum_{j=1}^{n}v_j\{g_0,|G^j(\hat{\kappa}_j[D_l{}^\alpha+R_j{}^\alpha-v_j(t-t_j)])|^2 g_0\}\rangle_{\rm av}. \quad (4.16)$$

If the target is uniformly illuminated by the beam, we may remove $|G^j|^2$ from the wave function average and then set $(g_0,g_0)=1$. Now the flux incident upon the target F_T is given by

$$F_T=\langle\sum_{j=1}^{n}v_j|G^j(\hat{\kappa}_j[D_l{}^\alpha+R_j{}^\alpha-v_j(t-t_j)])|^2\rangle_{\rm av}; \quad (4.17)$$

this will be independent of time during the time interval T if the beam intensity is held constant. The flux of scattered beam particles at the detector, F_D, is given by

$$F_D=1/D_l^2(\Sigma_\alpha\bar{\sigma}_\alpha)F_T. \quad (4.18)$$

The factor Γ in Eq. (4.16) connects this flux with the actual detector efficiency. The counting rate as defined by Eq. (4.12) is[20]

$$C_1(l)=\sum_1\frac{\Gamma}{D_l^2}(\sum_\alpha\bar{\sigma}_\alpha)F_T\int_0^\infty d\tau L(\tau). \quad (4.19)$$

If we choose to interpret Γ as the efficiency per unit active area of the detector, we may set $\Sigma_l=\int d\Sigma(\boldsymbol{l})$ where $d\Sigma(\boldsymbol{l})$ is an area element of the detector surface. For a detector small enough that D_l and $\bar{\sigma}_\alpha$ are constant over the surface, we have

$$C_1(l)=(\Sigma_l)\Gamma B(0)F_D \quad (4.20)$$

where (Σ_l) is the detector area and $B(0)$ is the dc bandpass characteristic of the circuitry [see Eq. (4.10)].

The normal situation considered in x-ray diffraction theory is one in which the waves scattered from the different elementary scattering centers in the target are coherent. In this case, all of the terms in the sum over α,β in Eq. (4.15) must be kept. To discuss this case, we again assume that we may set $\Gamma(\kappa_j)=\Gamma$ = constant. We are led to consider the quantity 'F' defined by

$$'F'=\langle\sum_{j=1}^{n}v_jG^j((\hat{\kappa}_j[D_l{}^\alpha+R_j{}^\alpha-v_j(t-t_j)])$$
$$\times G^{j*}((\hat{\kappa}_j[D_l{}^\beta+R_j{}^\beta-v_j(t-t_j)])$$
$$\times\exp[i\kappa_j(D_l{}^\alpha-D_l{}^\beta+R_j{}^\alpha-R_j{}^\beta)]\rangle_{\rm av}. \quad (4.21)$$

This expression, which occurs in Eq. (4.15), reduces to F_T when $\alpha=\beta$.

Now we insert the Fourier representation for the wave packet amplitudes G^j, as given by Eq. (3.13), into Eq. (4.21),

$$G^j(\mathbf{x})=\int\frac{d^3k'}{(2\pi)^{3/2}}A^j(\mathbf{k}'+\boldsymbol{\kappa}_j)e^{i\mathbf{k}'\cdot\mathbf{x}}.$$

We obtain, on writing $k=\kappa_j+\mathbf{k}'\cdot\hat{\kappa}_j$, $p=\kappa_j+\mathbf{k}''\cdot\hat{\kappa}_j$,

$$'F'=\Big\langle\sum_{j=1}^{n}v_j\int\frac{d^3k}{(2\pi)^{3/2}}\int\frac{d^3p}{(2\pi)^{3/2}}A^j(\mathbf{k})A^{j*}(\mathbf{p})$$
$$\times\exp[ik(D_l{}^\alpha+R_j{}^\alpha)]\exp[i(\epsilon_p-\epsilon_k)(t-t_j)]$$
$$\times\exp[-ip(D_l{}^\beta+R_j{}^\beta)]\Big\rangle_{\rm av}. \quad (4.22)$$

In deriving this relation we have used the fact that the $A^j(\mathbf{k})$ are peaked around $\boldsymbol{\kappa}_j$ to replace $(p-k)v_j$ by $(\epsilon_p-\epsilon_k)$. It is convenient to introduce new wave packet amplitudes by

$$a^j(k)=\int k^2(d\Omega_{\rm k}A^j(\mathbf{k})/(2\pi)^{3/2}), \quad (4.23)$$

[20] We have supposed that in a practical experiment the response time of the detector will be very short compared to the observation time T.

in terms of which 'F' becomes

$$'F'=\left\langle\sum_{j=1}^{n} v_j\int dk\int dp a^j(k)a^{j*}(p)\times\exp[ik(D_l{}^\alpha+R_j{}^\alpha)]\exp[i(\epsilon_p-\epsilon_k)(t-t_j)]\times\exp[-ip(D_l{}^\beta+R_j{}^\beta)]\right\rangle_{\text{av}}. \quad (4.24)$$

As part of our agreement on the ensemble average, we must imagine that the emission times, t_j, will occur randomly during the interval T, the time duration of each observation in the ensemble. We may, therefore, insert an average

$$\frac{1}{T}\int_0^T dt_j\cdots, \quad (4.25)$$

within the summation in Eq. (4.16). In general, one will choose the observation time T to be such that v_jT is much greater than the jth packet. After all, we do want to observe the entire scattering process. We may consequently set

$$\frac{1}{T}\int_0^T dt_j\exp[i(\epsilon_p-\epsilon_k)t_j]=\frac{2\pi}{T}\delta(\epsilon_p-\epsilon_k). \quad (4.26)$$

Since $v_jdp\approx d\epsilon_p$ [because of the packet $a^j(p)$], we may rewrite Eq. (4.24) as

$$'F'=\left\langle\sum_{j=1}^{n}\int dk\frac{2\pi}{T}|a^j(k)|^2\exp[ikD_{\alpha\beta}{}^j]\right\rangle_{\text{av}}, \quad (4.27)$$

where

$$D_{\alpha\beta}{}^j=D_l{}^\alpha-D_l{}^\beta+R_j{}^\alpha-R_j{}^\beta.$$

Since we are considering an ensemble average, the sum over j in Eq. (4.19) is equivalent to an integral over the surface of the source and an integral over the energy spectrum of the beam particles. The first of these may be written as

$$\frac{1}{\Sigma_s}\int d\Sigma(j)$$

where Σ_s is the total source area, and $d\Sigma(j)$ is an element of surface in the neighborhood of the source point $\mathbf{d}j$ (see Fig. 4). If the surface of the source emits particles uniformly, we may introduce a normalized beam spectrum in the following way. A frequency $\omega(k)$ is defined by

$$\omega(k)=\epsilon_k/\hbar,$$

where ϵ_k is the beam particle energy. Now we use the fact that F_T, the incident flux, in the present notation becomes [setting $D_{\alpha\beta}{}^j=0$ in (4.27)]

$$F_T=\left\langle\sum_i\int dk\frac{2\pi}{T}|a^j(k)|^2\right\rangle_{\text{av}}=\int d\omega\left\langle\sum_i\int dk\delta[\omega-\omega(k)]\left[\frac{2\pi}{T}|a^j(k)|^2\right]\right\rangle_{\text{av}}, \quad (4.29)$$

where in the second line we have simply inserted a formal δ function. This leads us to define the beam spectral function $g(\omega)$ according to

$$g(\omega)=\left\langle\sum_i\int dk\delta[\omega-\omega(k)]\left[\frac{2\pi}{T}\frac{a^j(k)^2}{F_T}\right]\right\rangle_{\text{av}} \quad (4.30)$$

which has the property $\int d\omega g(\omega)=1$. [If the source is not uniform, $g(\omega)$ will be a function of the source point $\mathbf{d}_j$.] In terms of $g(\omega)$, 'F' Eq. (4.29) may be written as

$$'F'=\frac{F_T}{\Sigma_s}\int d\Sigma(j)\int_0^\infty d\omega g(\omega)\exp[ik(\omega)D_{\alpha\beta}{}^j], \quad (4.31)$$

where we have inverted the δ-function restriction $\omega=\omega(k)$ to express k as a function of ω.

On inserting (4.31) into Eq. (4.15), we obtain

$$J'(\mathbf{l},t)=\frac{F_T}{D_l{}^2}\frac{\Gamma}{\Sigma_s}\int d\Sigma(j)\int_0^\infty d\omega g(\omega)\{g_0,\sum_{\alpha,\beta=1}^{N} f_\beta{}^\dagger f_\alpha\exp[ik(\omega)(D_l{}^\alpha-D_l{}^\beta+R_j{}^\alpha-R_j{}^\beta)]g_0\}_{\text{av}}. \quad (4.32a)$$

In many cases of practical interest the f_α are independent of spin, or the spin dependence may be factored out of $f_\beta{}^\dagger f_\alpha$ (as is usually done for x-ray scattering). Then we may write (see Fig. 4)

$$J'(\mathbf{l},t)=\frac{F_T}{D_l{}^2}\frac{\Gamma}{\Sigma_s}\int d\Sigma(j)\int_0^\infty d\omega g(\omega)\{g_0,|\sum_{\alpha=1}^{N} f_\alpha\exp[ik(\omega)(D_l{}^\alpha-D_l+R_j{}^\alpha-R_j)]|^2g_0\}_{\text{av}}. \quad (4.32b)$$

Since this is independent of τ, the counting rate (4.12) is

$$C_1(l)=\int_0^\infty d\tau L(\tau)\sum_1\frac{F_T\Gamma}{D_l{}^2\Sigma_s}\int d\Sigma(j)\int_0^\infty d\omega g(\omega)\{g_0,|\sum_{\alpha=1}^{N} f_\alpha\exp[ik(\omega)(D_l{}^\alpha-D_l+R_j{}^\alpha-R_j)]|^2g_0\}_{\text{av}}. \quad (4.33)$$

For a small source, a small target, a small detector and a nearly monochromatic beam [so $k(\omega)\approx\bar{k}$, a constant] this reduces to

$$C_1(l)=\frac{B(0)\Gamma_D F_T}{D_l^2} \times\{g_0,|\sum_{\alpha=1}^{N} f_\alpha \exp[i\bar{k}(\hat{R}_j-\hat{D}_l)\cdot\mathbf{z}_\alpha]|^2 g_0\}_{\rm av}. \tag{4.34}$$

Here we have absorbed the detector area in the constant Γ_D. The quantity

$$|\sum_{\alpha=1}^{N} f_\alpha \exp[i\bar{k}(\hat{R}_j-\hat{D}_l)\cdot\mathbf{z}_\alpha]|^2$$

is just the usual x-ray form factor. It is usually customary in discussing x-ray scattering to rewrite the average in Eq. (4.34) in the *approximate* form,

$$C_1(l)=\frac{B(0)\Gamma_D F_T}{D_l^2} \times|\sum_{\alpha=1}^{N} f_\alpha\{g_0,\exp[i\bar{k}(\hat{R}_j-\hat{D}_l)\cdot\mathbf{z}_\alpha]g_0\}_{\rm av}|^2. \tag{4.35}$$

This form may be used if the scattering is *elastic* and in some other cases.[21]

For later reference we note the commutation relations for the field operators (4.8),

$$[\varphi(\mathbf{x},-\tau),\varphi(\mathbf{x}',t-\tau)]_\pm =[\varphi^\dagger(\mathbf{x},-\tau),\varphi^\dagger(\mathbf{x}',t-\tau)]_\pm=0 \tag{4.36}$$

$$[\varphi(\mathbf{x},-\tau),\varphi^\dagger(\mathbf{x}',t-\tau)]_\pm =\sum {}_s\Gamma(k)v(k)\chi_s(\mathbf{x})\chi_s{}^*(\mathbf{x}')\exp(i\epsilon_k t).$$

V. THE MEASUREMENT OF INTENSITY CORRELATIONS

We are now ready to study in detail the correlated counting rate for the two detectors D_λ and D_l, as described in Sec. II. For simplicity, we assume that the two counters are identical and that they are represented by the counting rate operators R_λ and R_l; these are the operators introduced in Eq. (4.11). The geometry of the experiment of interest is shown in Fig. 5. The source (which may be a composite of two or more sources) and target are those described in Secs. III and IV. The vectors $\boldsymbol{\lambda}$ and $\boldsymbol{l}$ designate arbitrary points in D_λ and D_l. Vectors from the target point $\mathbf{z}_\alpha$ to $\boldsymbol{\lambda}$ and $\boldsymbol{l}$ are

$$\mathbf{D}_\lambda{}^\alpha=\mathbf{D}_\lambda-\mathbf{z}_\alpha,$$
$$\mathbf{D}_l{}^\alpha=\mathbf{D}_l-\mathbf{z}_\alpha,$$

just as in Eq. (3.10). The vector $\mathbf{R}_j{}^\alpha$ from source point

[21] We do not wish to go into this question further here. See, for example, Ref. 8, Chap. XI, for a discussion of this point.

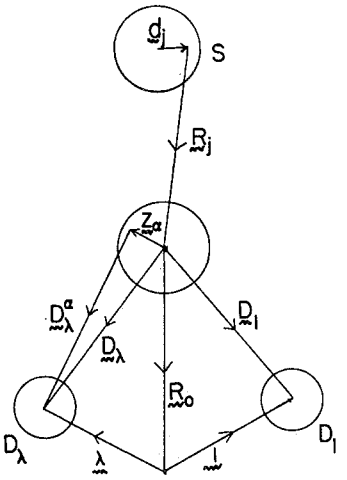

FIG. 5. Scattering with one source and two detectors.

$\mathbf{d}_j$ to the target point $\mathbf{z}_\alpha$ was introduced in Eq. (3.9). We shall again assume, as in Sec. IV, that the target is so small that

$$|\mathbf{D}_\lambda{}^\alpha-\mathbf{D}_\lambda|\ll D_\lambda,$$
$$|\mathbf{D}_l{}^\alpha-\mathbf{D}_l|\ll D_l, \tag{5.1}$$

although it is trivial to remove this restriction.

We shall imagine that the entire target is uniformly illuminated by the source (or sources). This source (or sources) is nothing special for our intensity correlation experiment; we cannot overemphasize this point. There are no coherence requirements or anything else. It need be only a conventional x-ray or other particle beam source. The state vector $|\Phi\rangle$ given by Eq. (3.20) continues to be appropriate.

The outputs of the two detectors D_λ and D_l are multiplied together; the product is the measured signal. We imagine that each observation is made for a time T and that an ensemble average of many such observations is taken. This is the same instruction we gave for the normal x-ray experiment in Sec. IV. The mean correlated counting rate for the experiment is

$$C_2=\langle(\langle\Phi|R_\lambda R_l|\Phi\rangle)\rangle_{\rm av} =\sum_\lambda\sum_l\int_0^\infty d\tau'\int_0^\infty dt' L(\tau')L(t')J_\alpha(\boldsymbol{\lambda},t-\tau';\boldsymbol{l},t-t'), \tag{5.2}$$

where

$$J_\alpha(\boldsymbol{\lambda},t-\tau';\boldsymbol{l},t-t') =\langle\{\langle\Phi|\phi^\dagger(\boldsymbol{\lambda},-\tau')\phi(\boldsymbol{\lambda},-\tau')\phi^\dagger(\boldsymbol{l},-t')\phi(\boldsymbol{l},-t')|\Phi\rangle\}\rangle_{\rm av}. \tag{5.3}$$

The expressions (5.2) and (5.3) describe the output of a system in which the counting rate at each detector is regarded as a classical quantity. If the treatment were fully quantum mechanical through the correlator, one might expect the operators in (5.3) to be "normally ordered," as in (5.7). The difference between these two orderings (which will shortly be shown to be small)

involves subtleties of the quantum-mechanical theory of measurement in which we would prefer not to involve ourselves here. The symbol $\langle\cdots\rangle_{\rm av}$ again denotes spin, thermal, and ensemble averages over repeated observations.

We find it convenient to rearrange Eq. (5.3) by writing

$$\varphi(\boldsymbol{\lambda},\tau)\varphi^\dagger(\boldsymbol{l},t)=[\varphi(\boldsymbol{\lambda},\tau),\varphi^\dagger(\boldsymbol{l},t)]_\pm+\epsilon\varphi^\dagger(\boldsymbol{l},t)\varphi(\boldsymbol{\lambda},\tau)\,, \quad (5.4)$$

where $\epsilon=+1(-1)$ for Bose-Einstein (Fermi-Dirac) beam particles. The commutator in (5.4) is not an operator; its value was given in Eq. (4.36). Using this relation, we may write J_α as follows:

$$J_\alpha(\boldsymbol{\lambda},t-\tau';\boldsymbol{l},t-t') =[\phi(\boldsymbol{\lambda},-\tau'),\phi(\boldsymbol{l},-t')]_\pm\times J'(\boldsymbol{\lambda},t-\tau';\boldsymbol{l},t-t') +J''(\boldsymbol{\lambda},t-\tau';\boldsymbol{l},t-t')\,, \quad (5.5)$$

where

$$J'(\boldsymbol{\lambda},t-\tau';\boldsymbol{l},t-t') =\langle\{\langle\Phi|\phi^\dagger(\boldsymbol{\lambda},-\tau')\phi(\boldsymbol{l},-t')|\Phi\rangle\}\rangle_{\rm av}\,, \quad (5.6)$$

and

$$J''(\boldsymbol{\lambda},t-\tau';\boldsymbol{l},t-t') =\langle\epsilon\{\langle\Phi|\phi^\dagger(\boldsymbol{\lambda},-\tau')\phi^\dagger(\boldsymbol{l}-t')\phi(\boldsymbol{\lambda},-\tau')\phi(\boldsymbol{l},-t')|\Phi\rangle\}\rangle_{\rm av}. \quad (5.7)$$

The first term in Eq. (5.5) is entirely nonclassical; it results from the fundamental quantum limitations on one's ability to make correlated measurements at separated space time points—just what we have in mind here. Were it not for the smearing in coordinate space introduced by our nonuniform counter efficiency as described by Eq. (4.21), the commutator in (5.4) would be nonzero only on the light cone of the points $\boldsymbol{\lambda}$, τ and $\boldsymbol{l}$, t, i.e., at $(\boldsymbol{\lambda}-\boldsymbol{l})^2-c^2(\tau-t)^2=0$. We shall find presently that the term involving J' is generally negligible in practice.

Following the arguments leading to the counting rate for a single detector as described in Sec. IV, in particular Eq. (4.14), we see that the present J' may be written as

$$J'(\boldsymbol{\lambda},\tau;\boldsymbol{l},t) =\langle\sum_{j=1}^{n} v_j\Gamma(\kappa_j)(g_0,\phi_{\bar a(j)}{}^*(\boldsymbol{\lambda},\tau)\phi_{\bar a(j)}(\boldsymbol{l},t)g_0)\rangle_{\rm av}. \quad (5.8)$$

It is straightforward to see that J'' may be put into the form

$$J''(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)=J_n(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)+J_c(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)\,, \quad (5.9)$$

where the direct term, J_n, is [as in Eq. (2.9)],

$$J_n(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)=\langle\sum_{i\neq j} v_iv_j\Gamma(\kappa_i)\Gamma(\kappa_j)\{g_0,[\phi_{\bar a(i)}{}^\dagger(\boldsymbol{\lambda},\tau)\phi_{\bar a(i)}(\boldsymbol{\lambda},\tau)\phi_{\bar a(j)}{}^\dagger(\boldsymbol{l},t)\phi_{\bar a(j)}(\boldsymbol{l},t)]g_0\}\rangle_{\rm av}\,, \quad (5.10)$$

and the exchange term, J_c, is

$$J_c(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)=\epsilon\langle\sum_{i\neq j} v_iv_j\Gamma(\kappa_i)\Gamma(\kappa_j)\{g_0,[\phi_{\bar a(i)}{}^\dagger(\boldsymbol{\lambda},\tau)\phi_{\bar a(i)}(\boldsymbol{l},t)\phi_{\bar a(j)}{}^\dagger(\boldsymbol{l},t)\phi_{\bar a(j)}(\boldsymbol{\lambda},\tau)]g_0\}\rangle_{\rm av}. \quad (5.11)$$

We make the earlier assumption that the target, T, is a macroscopic distance from the two detectors, so that we may use the now familiar asymptotic form for the $\Phi_{\bar a}$'s as given by Eq. (3.17). Furthermore we replace the factors $(D_l{}^\alpha)^{-1}$ by $D_l{}^{-1}$ etc., in the wave functions.[22]

Carrying out the substitution of Eqs. (3.17) for the wave functions, we obtain

$$D_l{}^2D_\lambda{}^2J_n(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)=\langle\sum_{i\neq j=1}^{n} v_iv_j\Gamma(\kappa_i)\Gamma(\kappa_j)\sum_{\alpha,\beta,\gamma,\delta=1}^{N}\{g_0,G^i(\hat\kappa_i[D_\lambda{}^\alpha+R_i{}^\alpha-v_i(\tau-t_i)])\} \times G^{i*}(\hat\kappa_i[D_\lambda{}^\delta+R_i{}^\delta-v_i(\tau-t_i)])G^j(\hat\kappa_j[D_l{}^\gamma+R_j{}^\gamma-v_j(t-t_j)])G^{j*}(\hat\kappa_j[D_l{}^\beta+R_j{}^\beta-v_j(t-t_j)])f_\delta{}^\dagger f_\alpha f_\beta{}^\dagger f_\gamma \times\exp(i\kappa_i[D_\lambda{}^\alpha-D_\lambda{}^\delta+R_i{}^\alpha-R_i{}^\delta])\exp(i\kappa_j[D_l{}^\gamma-D_l{}^\beta+R_j{}^\gamma-R_j{}^\beta])g_0\}\rangle_{\rm av}\,, \quad (5.12)$$

and

$$D_l{}^2D_\lambda{}^2J_c(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)=\epsilon\langle\sum_{i\neq j=1}^{n} v_iv_j\Gamma(\kappa_i)\Gamma(\kappa_j)[\exp-i(\epsilon_{\kappa j}-\epsilon_{\kappa i})(\tau-t)] \times\sum_{\alpha,\beta,\gamma,\delta=1}^{N}\{g_0,G^i(\hat\kappa_i[D_l{}^\alpha+R_i{}^\alpha-v_i(t-t_j)])G^{i*}(\hat\kappa_i[D_\lambda{}^\delta+R_i{}^\delta-v_i(\tau-t_i)]) \times G^j(\hat\kappa_j[D_\lambda{}^\gamma+R_j{}^\gamma-v_j(\tau-t_j)])G^{j*}(\hat\kappa_j[D_l{}^\beta+R_j{}^\beta-v_j(t-t_j)])f_\delta{}^\dagger f_\alpha f_\beta{}^\dagger f_\gamma \times\exp(i\kappa_i[D_l{}^\alpha-D_\lambda{}^\delta+R_i{}^\alpha-R_i{}^\delta])\exp(i\kappa_j[D_\lambda{}^\gamma-D_l{}^\beta+R_j{}^\gamma-R_j{}^\beta])g_0\}\rangle_{\rm av}. \quad (5.13)$$

[22] Here and in Eq. (5.26) are the only uses made in this section of the condition (5.1); if it is not legitimate, the correct factors are trivially provided, as described in Sec. IV.

We shall first discuss these expressions for the relatively simple case that the various waves scattered by different target particles do not interfere. This is *incoherent scattering*, which for a single detector was described by Eq. (4.19). It is worthwhile noting that the strictly incoherent scattering in the conventional case gives no information about the target structure [see Eq. (4.19)]. Only the coherent scattering leads to the intensity dependence on crystalline structure, etc., through the form factor of Eq. (4.34). This contrasts sharply with the intensity correlation experiment, in which the strictly incoherent radiation *does* give structure information—and, in fact in a somewhat more useful form than that obtained from conventional experiments. The intensity correlation experiment for coherent scattering will be described in Sec. VI.

There are several possibilities for experimental arrangements which will insure incoherent scattering. One common method is to choose the beam energy and scattering angle in such a way that there is sufficient momentum transfer to the target to cause it to undergo a transition to an excited state. In this case the various scattered waves will not interfere with one another [as was discussed in connection with the single detector counting rate, Eq. (4.16)]. Consequently, we have for the only nonvanishing terms in J_n and J_c, Eqs. (5.12) and (5.13) the index pairings

$$\alpha=\delta, \quad \beta=\gamma \quad \text{(Case } N\text{)} \tag{5.14a}$$

$$\alpha=\beta, \quad \gamma=\delta \quad \text{(Case } E\text{)}. \tag{5.14b}$$

(The other possible pairings such as $\alpha=\gamma, \beta=\delta$ do not contribute to the ensemble average—the phase factors cannot match properly.) The Case N corresponds to what might be called "normal pairing" in the sense that the scattered waves from beam particle "i" are matched; similarly those for beam particle "j" are matched. The Case E describes "exchange pairing" in which the scattered waves from two beam particles interfere. In this situation both beam particles i and j lead to equivalent excitations of target particles α and γ. The relative contributions of these two terms to the correlated counting depends on the experimental arrangements.

It may well be undesirable or not even feasible to ensure inelastic scatterings only, as described above. A particularly simple means of achieving incoherence, if it is desired, involves the use of an extended, or perhaps multiple, source for the beam. Alternatively one might imagine the source (S) to be moved during the course of the experiment. It is clear that by effectively varying the points of origin of the beam particles one may arrange that the ensemble average of the individual observations yields for the exponential factors in Eqs. (5.12) and (5.13)

$$\langle\exp(i[\kappa_i(R_i^{\alpha}-R_i^{\delta})+\kappa_j(R_j^{\gamma}-R_j^{\beta})])\rangle_{\text{av}} = \delta_{\alpha,\delta}\delta_{\beta,\gamma}. \tag{5.15}$$

This then corresponds to the Case N, Eq. (5.14a). The correlated counting rate in two small detectors is then measured as a function of counter separation.

Another possible experiment involves the use of a single large detector, so that D_l and D_λ represent the same counter; the counter must, of course, be biased to count only the coincidence of two scattered particles. (One could also use two large detectors.) In either case we would want a well defined beam energy, so that $\kappa_i=\kappa_j=\kappa$, and a pair of small sources with variable separation. The counting rate would be measured as a function of this separation. In this situation it is the variability of the target-to-detector distance that leads in the case of J_n [Eq. (5.12)] for the ensemble average to the result

$$\langle\exp(i\kappa[D_\lambda^{\alpha}-D_\lambda^{\delta}+D_l^{\gamma}-D_l^{\beta}])\rangle_{\text{av}}=\delta_{\alpha,\delta}\delta_{\beta,\gamma} \tag{5.16}$$

which is Case N. Similarly for J_c [Eq. (5.13)] we have

$$\langle\exp(i\kappa[D_l^{\alpha}-D_l^{\beta}+D_\lambda^{\gamma}-D_\lambda^{\delta}])\rangle_{\text{av}}=\delta_{\alpha,\beta}\delta_{\gamma,\delta}, \tag{5.17}$$

which is Case E.

There are evidently many possible experiments involving combinations of the conditions considered above. The symmetric role played by source and detector positions in the correlated counting rate is seen in Eqs. (5.12) and (5.13).

We write out in detail the forms taken by J_n and J_c for the two situations encountered in incoherent scattering: normal pairing, Case N, exchange pairing, and Case E.

For Case N [Eq. (5.14a)] we find for J_n the result,

$$J_n(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)=J'(\boldsymbol{\lambda},\tau)J'(\boldsymbol{l},t), \quad \text{(Case } N\text{)}, \tag{5.18}$$

where J' is precisely the quantity (4.13) encountered in the experiment with a single detector.[23] (This contribution to the correlated counting rate evidently contains no information concerning the target structure.) Again for Case N, we obtain for J_c

$$\begin{aligned} D_\lambda^2 D_l^2 J_c(\boldsymbol{\lambda},\tau;\boldsymbol{l},t)=\epsilon\Big\langle \sum_{i\neq j=1}^{n} v_i v_j \Gamma(\kappa_i)\Gamma(\kappa_j)\exp[-i(\tau-t)(\epsilon_{k_j}-\epsilon_{\kappa_i})] \\ \times\sum_{\alpha,\beta=1}^{N}\{g_0, G^i(\hat{\kappa}_i[D_l^{\alpha}+R_i^{\alpha}-v_i(t-t_i)])G^{i*}(\hat{\kappa}_i[D_\lambda^{\alpha}+R_i^{\alpha}-v_i(\tau-t_i)]) \\ \times G^j(\hat{\kappa}_j[D_\lambda^{\beta}+R_j^{\beta}-v_j(\tau-t_j)])G^{j*}(\hat{\kappa}_j[D_l^{\beta}+R_j^{\beta}-v_j(t-t_j)]) \\ \times(f_\alpha^\dagger f_\alpha)(f_\beta^\dagger f_\beta)\exp(i[\kappa_i(D_l^{\alpha}-D_\lambda^{\alpha})+\kappa_j(D_\lambda^{\beta}-D_l^{\beta})])g_0\}\Big\rangle_{\text{av}}. \quad \text{(Case } N\text{)} \end{aligned} \tag{5.19}$$

[23] Equation (5.18) involves an approximation in that we have replaced the average "$\langle\Phi|\cdots|\Phi\rangle_{\text{av}}$" by a product of such averages for each J' factor. Unless the target is so small that mutual excitations due to the two beam particles interfere at the time of scattering, this is a valid approximation.

If the experimental situation is such that Case E obtains, we find

$$D_\lambda^2 D_l^2 J_n(\boldsymbol{\lambda},\tau;\mathbf{l},t)=\Big\langle \sum_{i\neq j=1}^{n} v_i v_j \Gamma(\kappa_i)\Gamma(\kappa_j) \sum_{\alpha,\gamma=1}^{N} \{g_0, G^i(\hat{\kappa}_i[D_\lambda{}^\alpha+R_i{}^\alpha-v_i(\tau-t_i)])G^{i*}(\hat{\kappa}_i[D_\lambda{}^\gamma+R_i{}^\gamma-v_i(\tau-t_i)])$$
$$\times G^j(\hat{\kappa}_j[D_l{}^\gamma+R_j{}^\gamma-v_i(t-t_j)])G^{j*}(\hat{\kappa}_j[D_l{}^\alpha+R_j{}^\alpha-v_j(t-t_j)])$$
$$\times\exp(i\kappa_i[D_\lambda{}^\alpha-D_\lambda{}^\gamma+R_i{}^\alpha-R_i{}^\gamma])\,|f_\gamma{}^\dagger f_\alpha|^2\exp(i\kappa_j[D_l{}^\gamma-D_l{}^\alpha+R_j{}^\gamma-R_j{}^\alpha])g_0\}\Big\rangle_{\rm av},\quad \text{(Case } E\text{)}\quad (5.20)$$

and

$$D_\lambda^2 D_l^2 J_c(\boldsymbol{\lambda},\tau;\mathbf{l},t)=\epsilon\Big\langle \sum_{i\neq j=1}^{n} v_i v_j \Gamma(\kappa_i)\Gamma(\kappa_j)\exp[-i(\tau-t)(\epsilon_{\kappa j}-\epsilon_{\kappa i})]$$
$$\times \sum_{\alpha,\gamma=1}^{N} \{g_0,\, G^i(\hat{\kappa}_i[D_l{}^\alpha+R_i{}^\alpha-v_i(t-t_i)])G^{i*}(\hat{\kappa}_i[D_\lambda{}^\gamma+R_i{}^\gamma-v_i(\tau-t_i)])$$
$$\times G^j(\hat{\kappa}_j[D_\lambda{}^\gamma+R_j{}^\gamma-v_j(\tau-t_j)])G^{j*}(\hat{\kappa}_j[D_l{}^\alpha+R_j{}^\alpha-v_j(t-t_j)])\,|f_\gamma{}^\dagger f_\alpha|^2$$
$$\times\exp(i[(\kappa_i-\kappa_j)(D_l{}^\alpha-D_\lambda{}^\gamma)])\exp(i[\kappa_i(R_i{}^\alpha-R_i{}^\gamma)+\kappa_j(R_j{}^\gamma-R_j{}^\alpha)])g_0\}\Big\rangle_{\rm av}.\quad \text{(Case } E\text{)}\quad (5.21)$$

Let us now discuss the simpler Case N. We wish to cast it into a more useful form which expresses explicitly the target structure dependence of the correlated counting rate. The "normal term," J_n, given by Eq. (5.18), is seen to lead to a contribution which is simply proportional to the product of the counting rates of the individual counters. It contains no structure information and describes the result of an experiment done with "classical" beam particles having no wave properties. Thus J_n leads to the rate $C_{2,n}$ [see Eq. (5.2)] given by

$$C_{2,n}=\sum_\lambda\sum_l\int_0^\infty d\tau'\int_0^\infty dt' L(\tau')L(t')J_n(\boldsymbol{\lambda},\,t-\tau';\,\mathbf{l},\,t-t') \quad (5.22)$$
$$=C_1(l)C_1(\lambda),$$

where $C_1(l)$ and $C_1(\lambda)$ are defined by Eq. (4.19).

The simplification of J_c, Eq. (5.19), may be carried out following the arguments given in connection with the single detector [Eq. (4.21)]. We shall assume again that $\Gamma(\kappa_i)=\Gamma$, a constant, so that Eq. (5.19) contains two factors like

$$\text{"}F\text{"}\equiv\Big\langle\sum_{j=1}^{n} v_j G^j(\hat{\kappa}_j[D_\lambda{}^\beta+R_j{}^\beta-v_j(\tau-t_j)])$$
$$\times G^{j*}(\hat{\kappa}_j[D_l{}^\beta+R_j{}^\beta-v_j(t-t_j)])$$
$$\times\exp[i\kappa_j(D_\lambda{}^\beta-D_l{}^\beta)]\exp[-i(\tau-t)\epsilon_{\kappa j}]\Big\rangle_{\rm av}.\quad (5.23)$$

Actually, the terms corresponding to $i=j$ which result from the occurrence of two such "F" factors should not be there; if the number of beam particles is large, as it is in practice, this leads to negligible error. Inserting the Fourier representation of the wave packets as we did in connection with Eq. (4.22), we obtain,

$$\text{"}F\text{"}=\Big\langle\sum_{j=1}^{n} v_j\int\frac{d^3k}{(2\pi)^{3/2}}\int\frac{d^3p}{(2\pi)^{3/2}}A^j(\mathbf{k})A^{j*}(\mathbf{p})$$
$$\times\exp(ik[D_\lambda{}^\beta+R_j{}^\beta-v_j(-t_j)])$$
$$\times\exp(-ip[D_l{}^\beta+R_j{}^\beta-v_j(-t_j)])$$
$$\times\exp(-i[\epsilon_\kappa\tau-\epsilon_p t])\Big\rangle_{\rm av}.$$

Introducing as part of the ensemble average, the average over the emission times t_j as in Eq. (4.25), together with the Fourier amplitude a^j defined by Eq. (4.18), we are led to the result,

$$\text{"}F\text{"}=\Big\langle\sum_{j=1}^{n}\int dk\Big[\frac{2\pi}{T}|a^j(k)|^2\Big]$$
$$\times\exp(i[k(D_\lambda{}^\beta-D_l{}^\beta-\epsilon_k(\tau-t)])\Big\rangle_{\rm av}.\quad (5.24)$$

In terms of our spectral function (4.30) and the variables ω and $k(\omega)$ used in Eq. (4.31), we obtain

$$\text{"}F\text{"}=F_T\int_0^\infty d\omega g(\omega)$$
$$\times\exp(i[k(\omega)(D_\lambda{}^\beta-D_l{}^\beta)-\omega(\tau-t)]).\quad (5.25)$$

To complete the simplification of "F," we use the expansion of $D_\lambda{}^\beta=D_l{}^\beta$ based on the assumption that the distance R_0 (Fig. 5) is large compared to the detector dimensions, namely

$$D_\lambda{}^\beta-D_l{}^\beta=Y+\frac{\mathbf{z}_\beta\cdot(\mathbf{l}_\perp-\boldsymbol{\lambda}_\perp)}{R_0}+\cdots,\quad (5.26)$$

where Y is the value of $D_\lambda{}^\beta - D_l{}^\beta$ when $z_\beta = 0$; the subscript $\perp$ means the component of a vector perpendicular to the direction $\hat{R}_0$, from target to detector origin. When the spectrum $g(\omega)$ is reasonably narrow, we may introduce the wave number $\bar{k}$ at the center of g in some of the factors of "F." Thus we set

$$k(\omega)\mathbf{z}_\beta\cdot(\boldsymbol{l}_\perp-\boldsymbol{\lambda}_\perp)/R_0 \approx \bar{k}\mathbf{z}_\beta\cdot(\boldsymbol{l}_\perp-\boldsymbol{\lambda}_\perp)/R_0, \quad (5.27)$$

and "F" becomes

$$\text{"}F\text{"} = \exp\left[i\bar{k}\frac{\mathbf{z}_\beta\cdot(\boldsymbol{l}_\perp-\boldsymbol{\lambda}_\perp)}{R_0}\right]F_T\int_0^\infty d\omega g(\omega) \times\exp(i[k(\omega)\cdot Y-\omega(\tau-t)]). \quad (5.28)$$

In this case our expression for J_c, Eq. (5.19) simplifies to[24]

$$J_c(\boldsymbol{\lambda},\tau;\boldsymbol{l},t) = (\epsilon/R_0{}^4)\Gamma^2 F_T{}^2[\textstyle\sum_\alpha \bar{\sigma}_\alpha]^2|\eta|^2 \times\left|\int_0^\infty d\omega g(\omega)\exp(i[k(\omega)Y-\omega(\tau-t)])\right|^2, \quad (5.29)$$

where η is

$$\eta = \left(g_0,\sum_{\alpha=1}^N \bar{\sigma}_\alpha \exp\left[i\bar{k}\left(\frac{\mathbf{z}_\alpha\cdot(\boldsymbol{l}_\perp-\boldsymbol{\lambda}_\perp)}{R_0}\right)\right]g_0\right)\Big/\left(\sum_{\alpha=1}^N \bar{\sigma}_\alpha\right). \quad (5.30a)$$

In deriving Eq. (5.29) for J_c, we have used the assumption stated in Sec. III, that we may average g_0 independently for different noninteracting beam particles. In terms of a scattering density function $\mathcal{S}(z)$ defined by

$$\mathcal{S}(z) = (g_0,\sum_\alpha \sigma_\alpha\delta(z-z_\alpha)g_0)/(\Sigma\sigma_\alpha),$$

we may write η as

$$\eta = \int d^3z\mathcal{S}(\mathbf{z})\exp[i\bar{k}\mathbf{z}\cdot(\boldsymbol{l}_\perp-\boldsymbol{\lambda}_\perp)/R_0]. \quad (5.30b)$$

It is clear that structure information is contained in the quantity η. Its precise relation to the standard x-ray structure factor will be seen on comparison with Eq. (4.35). The scattering amplitude in Eq. (4.35) is here replaced by the scattering cross section.

We may now write the complete correlated counting rate, Eq. (5.2), for Case N, [aside from the term involving J' in Eq. (5.5) which we take up shortly] in the form

$$C_2 = \sum_l\sum_\lambda\left[\Gamma F_T\frac{\sum_\alpha\bar{\sigma}_\alpha}{R_0{}^2}\right]^2\int_0^\infty d\tau'\int_0^\infty dt' L(\tau')L(t')\left\{1+\epsilon|\eta|^2\left|\int_0^\infty d\omega g(\omega)\exp(i[k(\omega)Y+\omega(\tau'-t')])\right|^2\right\}. \quad (5.31)$$

For sufficiently small detectors, the sums over $\boldsymbol{l}$ and $\boldsymbol{\lambda}$ may be absorbed into the definition of Γ as was done in Eq. (4.34).

If the target is so large that the approximations made in the exponential in Eq. (5.25), as expressed by Eqs. (5.26) and (5.27), are not valid, there is no difficulty in modifying Eq. (5.31). The more complicated resulting expression does not seem to yield additional information.

In practice it will probably be desirable to filter out the dc component of the output of each detector. This corresponds to taking

$$\int_0^\infty d\tau L(\tau) = \int_{-\infty}^{+\infty} d\tau L(\tau) = B(0) = 0$$

[see Eq. (4.10)]. In this case Eq. (5.31) becomes simply

$$C_2 = \epsilon\sum_l\sum_\lambda\left[\Gamma F_T\frac{\sum_\alpha\sigma_\alpha}{R_0{}^2}\right]^2\left\{\int_0^\infty d\tau'\int_0^\infty dt' L(\tau')L(t')\left|\int_0^\infty d\omega g(\omega)\exp(i[k(\omega)Y+\omega(\tau'-t')])\right|^2\right\}|\eta|^2. \quad (5.32)$$

It is just the factor $|\eta|^2$ which will ordinarily be desired as a result of the experiment.

Experiments utilizing others of the conditions (5.14), (5.16), and (5.17) are evidently feasible and may be described in the same manner.

Before completing this section we shall discuss the contribution to C_2 from the hitherto neglected term involving J' in Eq. (5.5). From its definition in Eq. (5.8) and in terms of the quantities used in Eq. (5.25) we may write J' as

$$J'(\boldsymbol{\lambda},\tau';\boldsymbol{l},t') = \Gamma F_T\frac{\bar{\sigma}_\alpha}{R_0{}^2}\int_0^\infty d\omega g(\omega) \times\exp(-i[k(\omega)(D_\lambda{}^\alpha - D_l{}^\alpha)+\omega(\tau'-t')]). \quad (5.35)$$

Referring to the definition of the correlated counting rate, Eq. (5.2), the expression for the commutator of our operators φ, $\varphi^\dagger$ given by Eq. (4.44), we find for

[24] We have written the "av" of the product $f_\alpha{}^+ f_\alpha f_\beta{}^+ f_\beta$ which occurs in Eq. (5.19) as $\bar{\sigma}_\alpha\bar{\sigma}_\beta$. This is valid when spin correlations in the target are weak, or when all spin states are equally populated, etc. In general, the complete product may have to be evaluated with more care.

the contribution to C_2 from the term involving J' is

$$\delta C_2=\Gamma F_T\sum_\lambda\sum_l\int d\tau'\int dt'L(\tau')L(t')\{\sum_s v(k)\Gamma(k)\chi_s(\lambda)\chi_s^*(l)\exp[-i\epsilon_k(t'-\tau')]\}$$
$$\times\sum_{\alpha=1}^{N}\frac{\sigma_\alpha}{R_0^2}\int_0^\infty d\omega g(\omega)\exp\{-i[k(\omega)(D_\lambda^\alpha-D_l^\alpha)+\omega(\tau'-t')]\} \quad (5.34)$$
$$=\Gamma F_T\sum_\alpha\frac{\sigma_\alpha}{R_0^2}\sum_s\int_0^\infty d\omega B(\omega+\epsilon_k)B(-\omega-\epsilon_k)g(\omega)v(k)\Gamma(k)\chi_s(\lambda)\chi_s^*(l)\exp[ik(\omega)(D_\lambda^\alpha-D_l^\alpha)].$$

In the second form of δC_2 we have introduced the Fourier representation of the L's in terms of the frequency response function of the detectors as defined by Eq. (4.10). Now $g(\omega)$ and $\Gamma(k)$ are expected to be nonvanishing only for a range of frequencies near

$$\omega\approx\text{beam particle energy}/\hbar. \quad (5.35)$$

On the other hand, the low pass filter characteristic of the detector, $B(\omega)$, is presumedly chosen to be very small at this high "carrier frequency," (5.35). We conclude, therefore, that under almost any reasonable circumstances, δC_2 as given by Eq. (5.34), will be completely negligible.

VI. INTENSITY CORRELATIONS FOR COHERENT SCATTERING

We turn now to an analysis of the intensity correlation experiment for coherent scattering. The corresponding result for conventional scattering was obtained in Eq. (4.33) or Eq. (4.35). Coherent scattering is generally of greater interest than incoherent scattering for target structure determination; waves scattered from different parts of the target give quite direct information of the target geometry. It is the unusual characteristic of the intensity correlation experiment, as we have noted earlier, that incoherent scattering gives any geometric information. We shall find, however, it is the coherent scattering experiment that has the real potential for a direct solution of the phase determination problem.

We begin with the expressions (5.2) and (5.9) for the correlator counting rate, repeated here for convenience,

$$C_2=\sum_\lambda\sum_l\int_0^\infty d\tau'\int_0^\infty dt'L(\tau')L(t')$$
$$\times[J_n(\lambda,\,t-\tau';\,\mathbf{l},\,t-t')+J_c(\lambda,\,t-\tau';\,\mathbf{l},\,t-t')]; \quad (6.1)$$

J_n and J_c are given in Eqs. (5.12) and (5.13). To simplify the discussion, we now assume that the scattering amplitudes, f_α that appear there are either independent of spin orientation, or (as in x-ray scattering) that the spin dependence may be removed by averaging. It will also be convenient to assume that the variation with energy of the f_α may be neglected, either because the energy spectrum of the beam is rather narrow, or because the f_α are essentially constant, as in the usual x-ray energy regime. Finally, we shall again take the $\Gamma(\kappa_j)$ to be constant and equal to Γ for all beam energies.

Let us first consider J_n[Eq. (5.12)]. It is seen to contain two factors such as[25] [compare Eq. (4.21)],

$$F_n=\langle\sum_{j=1}^{n}v_jG^j(\hat{\kappa}_j[D_l^\gamma+R_j^\gamma-v_j(t'-t_j)])$$
$$\times G^{j*}(\hat{\kappa}_j[D_l^\beta+R_j^\beta-v_j(t'-t_j)])$$
$$\times\exp(i\kappa_j[D_l^\gamma-D_l^\beta+R_j^\gamma-R_j^\beta])\rangle_{\text{av}}. \quad (6.2)$$

On inserting the Fourier representation (3.13) for the G^j's and using the definition (4.23), we obtain

$$F_n=\Big\langle\sum_{j=1}^{n}v_j\int dkdpa^j(k)a^{j*}(p)$$
$$\times\exp(ik[D_l^\gamma+R_j^\gamma])\exp(-ip[D_l^\beta+R_j^\beta])$$
$$\times\exp[i(\epsilon_p-\epsilon_k)(t'-t_j)]\Big\rangle_{\text{av}}. \quad (6.3)$$

The ensemble average (4.25) leads then to

$$F_n=\Big\langle\sum_j\int dk\Big[\frac{2\pi}{T}|a^j(k)|^2\Big]\exp[ikD_{\gamma\beta}{}^j]\Big\rangle_{\text{av}} \quad (6.4)$$

where

$$D_{\gamma\beta}{}^j\equiv D_l^\gamma-D_l^\beta+R_j^\gamma-R_j^\beta.$$

Finally, the spectral function $g(\omega)$ and the integral over source area may be introduced, as in Eqs. (4.30) and (4.31), to give

$$F_n=\frac{F_T}{\Sigma_s}\int d\Sigma(j)\int_0^\infty d\omega g(\omega)\exp[ik(\omega)D_{\gamma\beta}{}^j]. \quad (6.5)$$

The expression (5.12) is seen to contain two such factors.

A scattering amplitude for the target may be defined as [see Fig. 5 for a description of the distances]

$$\mathfrak{F}_0(l,j;\omega)=\sum_{\gamma=1}^{N}f_\gamma(\exp ik(\omega)[D_l^\gamma-D_l+R_j^\gamma-R_j)). \quad (6.6)$$

[25] We here ignore, as in Sec. V, the restriction $i\neq j$ in the summations in Eqs. (5.12) and (5.13).

[This quantity will be recognized as that appearing in Eq. (4.32b).] The expression (5.12) contains four such factors. Putting together the various factors, we obtain

$$J_n(\lambda,\tau';l,t')=\frac{\Gamma^2F_T^2}{D_l^2D_\lambda^2}\left[\frac{1}{\Sigma_s}\int d\Sigma(j)\right]\left[\frac{1}{\Sigma_s}\int d\Sigma(i)\right]\left\{g_0,\left[\int_0^\infty d\omega g(\omega)\,|\mathfrak{F}_0(l,j;\omega)|^2\right]\left[\int_0^\infty d\omega' g(\omega')\,|\mathfrak{F}_0(\lambda,i;\omega')|^2\right]g_0\right\}_{\rm av}. \tag{6.7}$$

It is important to note that J_n is *independent* of τ' and t', just as in the incoherent case.

The expression for J_c [Eq. (5.13)] may be simplified in the same way. It is seen to contain two factors such as

$$F_c=\Big\langle\sum_{j=1}^{n}{}' v_jG^j(\hat{\kappa}_j[D_\lambda{}^\gamma+R_j{}^\gamma-v_j(\tau'-t_j)])G^{j*}(\hat{\kappa}_j[D_l{}^\beta+R_j{}^\beta-v_j(\tau'-t_j)])$$
$$\times\exp(i\kappa_j[D_\lambda{}^\gamma-D_l{}^\beta+R_j{}^\gamma-R_j{}^\beta])\exp[i\epsilon_{\kappa_j}(t'-\tau')]\Big\rangle. \tag{6.8}$$

On following the arguments leading to Eq. (6.5), we now find

$$F_c=\frac{F_T}{\Sigma_s}\int d\Sigma(j)\int_0^\infty d\omega g(\omega)\exp[ik(\omega)(D_\lambda{}^\gamma+R_j{}^\gamma-D_l{}^\beta-R_j{}^\beta)]\exp[+i\omega(t'-\tau')]. \tag{6.9}$$

The expression (5.13) contains also four scattering amplitudes, such as (6.6). Again putting together the pieces, we obtain

$$J_c(\lambda,\tau',l,t')=\epsilon\frac{\Gamma^2F_T^2}{D_l^2D_\lambda^2}\left[\frac{1}{\Sigma_s}\int d\Sigma(j)\right]\left[\frac{1}{\Sigma_s}\int d\Sigma(i)\right]\int_0^\infty d\omega g(\omega)\int_0^\infty d\omega' g(\omega')$$
$$\times\exp(i[k(\omega)(D_\lambda-D_l)+\omega(t'-\tau')])\times\exp(-i[k(\omega')(D_\lambda-D_l)+\omega'(t'-\tau')])$$
$$\times(g_0,\mathfrak{F}_0(\lambda,j;\omega)\mathfrak{F}_0{}^*(l,j;\omega)\mathfrak{F}_0(l,i;\omega')\mathfrak{F}_0{}^*(\lambda,i;\omega')g_0). \tag{6.10}$$

The correlator counting rate may now be obtained by inserting the expressions (6.7) and (6.10) into Eq. (6.1).

When feasible, it is ordinarily desirable to use a beam spectrum narrow enough that the ω-dependence of the $\mathfrak{F}$'s may be neglected as was assumed in obtaining Eq. (4.34). For a crystalline target which has a large number of unit cells, this assumption is unwarranted. As is well known, in ordinary x-ray scattering, the frequency dependence of $\mathfrak{F}$ is important and must be considered explicitly; it gives rise to what is sometimes called a Lorentz factor. We shall return to this case in Sec. VIII, but for the time being neglect the effect. Then,

$$\mathfrak{F}_0(l,j)\equiv\mathfrak{F}_0(l,j;\bar{\omega}), \tag{6.11}$$

where $\bar{\omega}$ is an appropriate mean frequency of the beam. Now Eqs. (6.7) and (6.10) simplify considerably:

$$J_n(\lambda,\tau';l,t')=(\Gamma^2F_T^2/D_l^2D_\lambda^2)(1/\Sigma_s)^2\int d\Sigma(j)\int d\Sigma(i)(g_0,|\mathfrak{F}_0(l,j)|^2|\mathfrak{F}_0(\lambda,i)|^2g_0)_{\rm av}. \tag{6.12}$$

$$J_c(\lambda,\tau';l,t')=(\epsilon)(\Gamma^2F_T^2/D_l^2D_\lambda^2)(1/\Sigma_s)^2\int d\Sigma(j)\int d\Sigma(i)\left|\int_0^\infty d\omega g(\omega)\exp(i[k(\omega)Y+\omega(t'-\tau')])\right|^2$$
$$\times(g_0,\mathfrak{F}_0(\lambda,j)\mathfrak{F}_0{}^*(l,j)\mathfrak{F}_0(l,i)\mathfrak{F}_0{}^*(\lambda,i)g_0)_{\rm av}. \tag{6.13}$$

Here $Y\equiv D_\lambda-D_l$ is the quantity introduced in Eq. (5.26). For an experiment performed with two small sources, we see that Eq. (6.13) contains just the expression (2.3), multiplied by factors appropriate for determining the actual counting rate.

It is often permissible,[26] as was described in connection with Eq. (4.35), to replace the wave function average of the product of $\mathfrak{F}$'s in Eqs. (6.12) and (6.13) by a product of averages of each $\mathfrak{F}$. In this case we may introduce the amplitudes

$$\mathfrak{F}(l,j)=\{g_0,\sum_{\gamma=1}^{N}f_\gamma\exp(ik(\bar{\omega})[D_l{}^\gamma-D_l+R_j{}^\gamma-R_j])g_0\}_{\rm av}, \tag{6.14}$$

into Eqs. (6.12) and (6.13) and drop the explicit average indicated by $(g_0,\cdots g_0)_{\rm av}$. For the remaining discussion of this section, we shall assume that this may be done.

[26] When the scattering is predominantly *elastic*, for example, this may be done.

The correlator counting rate (6.1) is now

$$C_2=\Gamma^2F_T^2\sum_\lambda\sum_l\frac{1}{D_l^2D_\lambda^2\Sigma_s^2}\int d\Sigma(i)\int d\Sigma(j)\Big\{(B(0))^2|\mathfrak{F}(l,j)|^2|\mathfrak{F}(\lambda,i)|^2+\epsilon\int_0^\infty d\tau'\int_0^\infty dt'L(\tau')L(t')$$
$$\times\Big|\int_0^\infty d\omega g(\omega)\exp(i[k(\omega)Y+\omega(\tau'-t')])\Big|^2\mathfrak{F}(\lambda,j)\mathfrak{F}^*(l,j)\mathfrak{F}(l,i)\mathfrak{F}^*(\lambda,i)\Big\}, \quad (6.15)$$

where $B(0)$ is the dc bandpass characteristic, as determined from Eq. (4.10). This equation should be compared with Eq. (5.31), obtained for incoherent scattering. (The geometric simplification of taking $D_l^{-2}D_\lambda^{-2}=R_0^{-4}$ has not been made here.) When the approximation described in connection with Eq. (6.14) cannot be made, $\mathfrak{F}_0$'s must be inserted in place of $\mathfrak{F}$'s in Eq. (6.15) and the entire right-hand side inserted within the $(g_0,\cdots g_0)_{\text{av}}$ average.

Equation (6.15) might have been derived directly in terms of the target scattering amplitude without the elaborate discussion given in Sec. III of scattering from a composite system and without the introduction of the restrictive assumptions (3.3) and (3.4).

To discuss a specific application of Eq. (6.15) let us assume that the circuits in the detectors are so chosen that $B(0)=0$. We shall also suppose that two "small" sources, S_a and S_α are used, as in Fig. 2 and that the detectors are "small." The assumption of "smallness" is interpreted to imply that the variation of the $\mathfrak{F}$'s over the surface of a given source of detector may be neglected, an assumption made also in obtaining Eq. (4.35). This must be reinvestigated for a large crystalline target. (See Sec. VIII.) In this case, and using the notation introduced in Eqs. (2.3) and (4.35), we may rewrite Eq. (6.15) in the form

$$C_2=M[|\mathfrak{F}(\lambda,a)|^2|\mathfrak{F}(l,a)|^2+|\mathfrak{F}(\lambda,\alpha)|^2|\mathfrak{F}(l,\alpha)|^2$$
$$+\mathfrak{F}(\lambda,\alpha)\mathfrak{F}^*(l,\alpha)\mathfrak{F}(l,a)\mathfrak{F}^*(\lambda,a)$$
$$+\mathfrak{F}(\lambda,a)\mathfrak{F}^*(l,a)\mathfrak{F}(l,\alpha)\mathfrak{F}^*(\lambda,\alpha)], \quad (6.16)$$

where

$$M=\epsilon\frac{\Gamma_D^2F_T^2}{D_l^2D_\lambda^2}\int_0^\infty d\tau'\int_0^\infty dt'L(\tau')L(t')$$
$$\times\Big|\int_0^\infty d\omega g(\omega)\exp(i[k(\omega)Y+\omega(\tau'-t')])\Big|^2. \quad (6.17)$$

The factor M is evidently of importance in determining the counting rate of the correlator. This will be described in Sec. VII. It contains, among other things, the important information about the requisite geometric stability of sources and detectors. In terms of the filter response time, $\Delta\tau_r$, defined in Sec. VII, we require only that $Y<C\Delta t_r$, which is of the order of feet. In the remainder of this section we shall suppose that M has been determined and that C_2 has been measured for all combinations of detector and source angles, and shall discuss the use of this information for the determination of the phases.

Let us first rewrite Eq. (6.14) in the form

$$\mathfrak{F}(l,a)=\{g_0,\sum_{\gamma=1}^N f_\gamma\exp[ik(\tilde{\omega})(\hat{R}_a-\hat{D}_l)\cdot\mathbf{z}_\gamma]g_0\}_{\text{av}}, \quad (6.18)$$

with corresponding expressions for the other three amplitudes appearing in Eq. (6.16). In obtaining Eq. (6.18), we have assumed that the target dimensions are small compared with R_a, D_l, etc. (see Fig. 3). Each of the four scattering amplitudes in Eq. (6.16) is seen to depend on the scattering geometry through its functional dependence on one of the vectors

$$\begin{aligned}\mathbf{g}_{la}&=\hat{R}_a-\hat{D}_l\\ \mathbf{g}_{l\alpha}&=\hat{R}_\alpha-\hat{D}_l\\ \mathbf{g}_{\lambda a}&=\hat{R}_a-\hat{D}_\lambda\\ \mathbf{g}_{\lambda\alpha}&=\hat{R}_\alpha-\hat{D}_\lambda.\end{aligned} \quad (6.19)$$

These vectors are not independent, but satisfy the single relation

$$\mathbf{g}_{la}-\mathbf{g}_{l\alpha}=\mathbf{g}_{\lambda a}-\mathbf{g}_{\lambda\alpha}. \quad (6.20)$$

Now, since the $\mathfrak{F}$'s are complex, we may write

$$\mathfrak{F}(l,a)=|\mathfrak{F}(l,a)|\exp[i\varphi(\mathbf{g}_{la})], \quad (6.21)$$

etc., where the phases φ are evidently real. The magnitudes $|\mathfrak{F}(l,a)|$ may be determined directly from a conventional experiment (for example, by recording the average counting rate of the detector D_l with only the source S_a present). Assuming that the $|\mathfrak{F}|$'s are known, we wish to determine the phases φ.

Having measured the magnitudes $|\mathfrak{F}|$ and the correlator counting rate (6.16), we may deduce the quantity $\cos\Gamma$, where

$$\Gamma=\varphi(\mathbf{g}_{la})-\varphi(\mathbf{g}_{l\alpha})+\varphi(\mathbf{g}_{\lambda\alpha})-\varphi(\mathbf{g}_{\lambda a}). \quad (6.22)$$

To determine the individual functions φ, we first require boundary conditions. A condition often assumed in x-ray scattering is that

$$\varphi(0)=\varphi(\mathbf{g}_{la})|_{\mathbf{g}_{la}=0}=0, \quad (6.23)$$

which requires in general that the f_γ's in Eq. (6.18) are real. In any event, we may always assume the condition (6.23) and the correct the final result for φ by adding to its deduced values the constant phase of $\sum_{\gamma=1}^N f_\gamma$, for forward scattering. We may, in addition

to the condition (6.23), assume that

$$\varphi'(0)\equiv\partial\varphi/\partial g_{l\alpha}|_{g_{l\alpha}=0}=0, \tag{6.24}$$

which merely specifies the origin of the coordinate system in the target.[27]

Now, let us suppose that Γ [Eq. (6.22)] has been measured, modulo (2π), for a complete array of values for $\mathbf{g}_{l\alpha}$, $\mathbf{g}_{l\alpha}$, etc. The discrete values of $\mathbf{g}_{l\alpha}$ will be represented by the sets of numbers $\mathbf{g}_1^{(1)}$, $\mathbf{g}_1^{(2)}$, $\cdots$, etc. Similarly, the values of $\mathbf{g}_{l\alpha}$ will be written as $\mathbf{g}_2^{(1)}$, $\mathbf{g}_2^{(2)}\cdots$, values of $\mathbf{g}_{\lambda,\alpha}$ as $\mathbf{g}_3^{(1)}$, $\mathbf{g}_3^{(2)}$, $\cdots$, and values of $\mathbf{g}_{\lambda,\alpha}$ as $\mathbf{g}_4^{(1)}$, $\mathbf{g}_4^{(2)}$, $\cdots$. The condition (6.20) implies that for any set ν $(\nu=1, 2, \cdots)$,

$$\mathbf{g}_4^{(\nu)}=\mathbf{g}_1^{(\nu)}-\mathbf{g}_2^{(\nu)}+\mathbf{g}_3^{(\nu)}. \tag{6.25}$$

The measured values of Γ may then be labeled as

$$\Gamma(\mathbf{g}_1^{(\nu)},\mathbf{g}_2^{(\nu)},\mathbf{g}_3^{(\nu)}) = \varphi(\mathbf{g}_1^{(\nu)})-\varphi(\mathbf{g}_2^{(\nu)})+\varphi(\mathbf{g}_3^{(\nu)})-\varphi(\mathbf{g}_4^{(\nu)}). \tag{6.26}$$

Let us next consider the difference of the Γ's for the two neighboring sets of values $\mathbf{g}_1^{(\nu+1)}$, $\mathbf{g}_2^{(\nu)}$, $\mathbf{g}_3^{(\nu)}$ and $\mathbf{g}_1^{(\nu)}$, $\mathbf{g}_2^{(\nu)}$, $\mathbf{g}_3^{(\nu)}$

$$\begin{aligned}\delta\Gamma(\mathbf{g}_1^{(\nu)},\mathbf{g}_2^{(\nu)},\mathbf{g}_3^{(\nu)})&\equiv\Gamma(\mathbf{g}_1^{(\nu+1)},\mathbf{g}_2^{(\nu)},\mathbf{g}_3^{(\nu)})\\ &-\Gamma(\mathbf{g}_1^{(\nu)},\mathbf{g}_2^{(\nu)},\mathbf{g}_3^{(\nu)})=[\varphi(\mathbf{g}_1^{(\nu+1)})-\varphi(\mathbf{g}_1^{(\nu)})]\\ &-[\varphi(\mathbf{g}_1^{(\nu+1)}+\mathbf{g}_3^{(\nu)}-\mathbf{g}_2^{(\nu)})-\varphi(\mathbf{g}_1^{(\nu)}+\mathbf{g}_3^{(\nu)}-\mathbf{g}_2^{(\nu)})].\end{aligned} \tag{6.27}$$

If $\delta\Gamma$ is now determined for an array of $\mathbf{g}$'s, such that $\mathbf{g}_1^{(\nu)}+\mathbf{g}_3^{(\nu)}-\mathbf{g}_2^{(\nu)}$ and $\mathbf{g}_1^{(\nu+1)}+\mathbf{g}_3^{(\nu)}-\mathbf{g}_2^{(\nu)}$ are each held constant, we may deduce the quantity,

$$\varphi(\mathbf{g}_1^{(\nu+1)})-\varphi(\mathbf{g}_1^{(\nu)})\equiv\delta\varphi(\nu), \tag{6.28}$$

to within a constant. This set of difference equations, and the boundary conditions (6.23) and (6.24), permit the determination of the phase $\varphi(g)$ as a function of $\mathbf{g}$.[28]

VII. COUNTING RATE AND FLUCTUATIONS

We have given expressions for the mean correlator counting rate in Secs. V and VI. The mean counting rate will of course be obtained only in the limit of a very long period of observation. For the practical design of an experiment it is necessary to estimate the fluctuations about the mean for an experiment of finite duration.

Let us first express the correlator counting rate C_2 [Eqs. (6.16) and (6.17)] in terms of several parameters characteristic of the experiment. The energy spread of the incident beam will be written as $\hbar\Delta\omega_B$, where

$$\frac{1}{\Delta\omega_B}\equiv\int d\omega[g(\omega)]^2. \tag{7.1}$$

Similarly, the response time of the counters $\Delta\tau_r$ is defined by

$$2\pi/\Delta\tau_r\equiv\int d\omega|B(\omega)|^2, \tag{7.2}$$

where $B(\omega)$ is defined by Eq. (4.10). We shall assume that

$$\Delta\tau_r\Delta\omega_B\gg1 \tag{7.3}$$

(although it would be desirable, if feasible, to have $\Delta\tau_r\Delta\omega_B\simeq1$). Then, using the condition (7.3) and Eq. (4.10), we have

$$\begin{aligned}\int_0^\infty d\tau'\int_0^\infty dt'L(\tau')L(t')&\\ \times\left|\int d\omega g(\omega)\exp(i[kY+\omega(\tau'-t')])\right|^2&\\ \approx\int dp|B(p)|^2\int d\omega|g(\omega)|^2=\frac{2\pi}{\Delta\tau_r\Delta\omega_B}.&\end{aligned} \tag{7.4}$$

Therefore, according to Eq. (6.16), assuming "small" detector and sources,

$$\begin{aligned}C_2=\frac{2\pi}{\Delta\tau_r\Delta\omega_B}\frac{\Gamma_D^2F_T^2}{D_l^2D_\lambda^2}&[\mathfrak{F}(l,a)\mathfrak{F}^*(\lambda,a)\mathfrak{F}(\lambda,\alpha)\mathfrak{F}^*(l,\alpha)+\text{c.c.}\\ &+|\mathfrak{F}(\lambda,a)|^2|\mathfrak{F}(l,a)|^2+|\mathfrak{F}(\lambda,\alpha)|^2|\mathfrak{F}(l,\alpha)|^2].\end{aligned} \tag{7.5}$$

To estimate the fluctuations in C_2, we shall continue to suppose that

$$\int_0^\infty d\tau L(\tau)=0, \tag{7.6}$$

which resulted in the elimination of the J_n term from C_2. We shall next drop the ensemble average, denoted by "$\langle\cdots\rangle_{\text{av}}$" from Eqs. (5.12) and (5.13). The resulting quantities,

$$\begin{aligned}J_n&=J_n(\boldsymbol{\lambda},\tau';\mathbf{l},t';t_1\cdots t_n;t)\\ J_c&=J_c(\boldsymbol{\lambda},\tau';\mathbf{l},t';t_1\cdots t_n;t),\end{aligned} \tag{7.7}$$

therefore depend parametrically on the number n of detected particles and the emission times $t_1\cdots t_n$. If the experiment is conducted for a time T, the mean counting rate for a pure state $|\Phi)$ is

$$\begin{aligned}C_2(t_1\cdots t_n;T)=\frac{1}{T}\int_0^T dt\int_0^\infty d\tau'\int_0^\infty dt'L(\tau')L(t')&\\ \times[J_n(\boldsymbol{\lambda},\tau';\mathbf{l},t';t_1\cdots t_n;t)&\\ +J_c(\boldsymbol{\lambda},\tau';\mathbf{l},t';t_1\cdots t_n;t)],&\end{aligned} \tag{7.8}$$

where the effect on counter size will be assumed to be absorbed in the factor Γ_D, as in Eq. (7.5). The mean counting rate (7.5) is the ensemble average of (7.8),

[27] That is, we may always add a constant vector $\mathbf{y}$ to all the $\mathbf{z}_\gamma$'s in Eq. (6.18), so chosen that condition (6.24) is valid.

[28] For application to scattering by a crystal our discussion has been somewhat schematic. We hope to return to a more detailed description in a later publication.

the average being made over the emission times $t_1 \cdots t_n$ and the number of counted particles n.

The fluctuations in C_2 may be estimated from the quantity

$$\delta C_2^2 \equiv \langle [C_2(t_1 \cdots t_n; T)]^2 \rangle_{\text{av}} - (C_2)^2. \tag{7.9}$$

Evaluation of this expression, assuming that the t_j's ($j=1, 2, \cdots n$) are independent random variables uniformly distributed over the interval $0 \leq t_j \leq T$, is straightforward but tedious. The leading term in (7.9) is determined by the fluctuation in $J_n \times J_n$—that is, the fluctuations getting past the dc filter [condition (7.6)].

Keeping only the $J_n \times J_n$ terms, then, we find

$$\delta C_2^2 = (M_n)^2 \{ [|\mathfrak{F}(l,a)|^2 + \mathfrak{F}(l,\alpha)|^2] \times [|\mathfrak{F}(\lambda,a)|^2 + |\mathfrak{F}(\lambda,\alpha)|^2] \}^2, \tag{7.10}$$

where

$$D_l^4 D_\lambda^4 (M_n)^2 \simeq \Gamma_D^4 \frac{T}{2\pi} \int d\omega |B(\omega)|^4 \left\langle \sum_{i=j=1}^{n} \int dk dl dk' dl' \left[\frac{2\pi}{T} |a^j(k)|^2\right]\left[\frac{2\pi}{T} |a^j(l)|^2\right] \times \left[\frac{2\pi}{T} |a^i(k')|^2\right]\left[\frac{2\pi}{T} |a^i(l')|^2\right] \right\rangle_{\text{av}}. \tag{7.11}$$

In obtaining this expression we have used the condition (7.3).

To give (7.11) a simple qualitative interpretation we shall replace

$$\left\langle \sum_{j=1}^{n} \left\{ \int dk \left[\frac{2\pi}{T} |a^j(k)|^2\right] \right\}^2 \right\rangle_{\text{av}}$$

by

$$\frac{1}{\bar{n}} F_T^2,$$

where $\bar{n}$ is the average number of particles counted during the interval T. Then the expression (7.11) becomes

$$(M_n)^2 = \frac{T}{2\pi \bar{n}^2} \left[\frac{\Gamma_D^2 F_T^2}{D_l^2 D_\lambda^2}\right]^2 \int d\omega |B(\omega)|^4. \tag{7.12}$$

Also, using Eq. (7.2), we set

$$\int d\omega |B(\omega)|^4 \simeq (2\pi/\Delta\tau_r), \tag{7.13}$$

as an "order of magnitude" relation.

Finally, on combining Eqs. (7.5), (7.10), (7.12), and (7.13), we obtain the "signal-to-noise" ratio for our experiment as

$$(\delta C_2^2)^{1/2}/C_2 \approx (\Delta\omega_B/\bar{n})(T\Delta\tau_r)^{1/2}, \tag{7.14}$$

ignoring the coefficients involving the scattering amplitude factors. Since $\bar{n}$ is proportional to T, the ratio (7.14) decreases as $T^{-1/2}$, as might be expected.

VIII. DISCUSSION OF AN EXPERIMENT

In this section we make a preliminary, though rather rudimentary effort to relate the formalism that has been developed to the real word. To be more specific, we direct our attention to crystalline substances. We assume that we are dealing with an ideal crystal. Some problems involved in extending the theory to mosaic crystals have not been solved, though we hope to return to them elsewhere. This limitation alone restricts our proposed experiment to excellent laboratory crystals, which will serve only to demonstrate feasibility of the techniques. Further, we have ignored primary extinction; this is a particularly poor approximation for ideal crystals since the absorption of the primary wave is very great for any allowed scattering geometry. To include the effect, we would have to superimpose on our already rather involved analysis the full structure of the dynamical theory of x-ray scattering. There seem to be no difficulties in principle, but we choose not to go into the matter here. Instead, we *simulate* the effects of extinction by assuming that our crystal is thin, and by adjusting its thickness to absorb the thickness of the absorption layer due to extinction in the crystal. Thus the angular and frequency width of the scattered radiation will be about those to be expected from honest extinction.

We turn to the experimental situation illustrated in Fig. 3, with two sources and two detectors. The correlator counting rate has been discussed in Sec. VI and we refer in particular to Eq. (6.15) [with $B(0)=0$, to cut out the uninteresting dc component]. Actually for our present purpose, it is useful to restore the frequency dependence of the scattering amplitudes, as in Eq. (6.10), so that (6.15) becomes

$$C_2 = \epsilon \Gamma^2 F_T^2 \sum_\lambda \sum_l \frac{1}{D_l^2 D_\lambda^2 \Sigma_s^2} \int d\Sigma(i) \int d\Sigma(j) \int_0^\infty d\tau' \int_0^\infty dt' L(\tau') L(t') \int_0^\infty d\omega g(\omega) \int_0^\infty d\omega' g(\omega') \times \exp(ik(\omega)Y + i\omega(\tau' - t')) \exp(-ik(\omega')Y - i\omega'(\tau' - t')) \mathfrak{F}(\lambda,j,\omega) \mathfrak{F}^*(l,j,\omega) \mathfrak{F}(l,i,\omega') \mathfrak{F}^*(\lambda,i,\omega'), \tag{8.1}$$

$$= \epsilon \Gamma^2 F_T^2 \sum_\lambda \sum_l \frac{1}{D_l^2 D_\lambda^2 \Sigma_s^2} \int d\Sigma(i) \int d\Sigma(j) \int_0^\infty d\omega g(\omega) \int_0^\infty d\omega' g(\omega') \times \exp(iY[k(\omega) - k(\omega')]) |B(\omega' - \omega)|^2 \mathfrak{F}(\lambda,j,\omega) \mathfrak{F}^*(l,j,\omega) \mathfrak{F}(l,i,\omega') \mathfrak{F}^*(\lambda,i,\omega'). \tag{8.2}$$

The last expression follows from Eq. (4.10). We shall not, at the moment, do the integrals over the sources; it is, however, worth bearing in mind that we are thinking of two small sources and that these integrals must be done. In contrast to the treatment described in Sec. VI, we cannot, in the real crystalline problem, disregard the very strong angular dependence of the $\mathfrak{F}$'s. Further, it should be noted that the interesting phase information is obtained only when $d\Sigma(i)$ and $d\Sigma(j)$ are on different sources [as in the last two terms of Eq. (6.16)].

Let us begin a discussion of the frequency ranges ω and ω' which contribute appreciably to C_2 in Eq. (8.2). For the x-ray case, the most restrictive factor will be $|B(\omega'-\omega)|^2$, since it will be difficult to design a filter network as broad as the width, even after Bragg reflection, of an x-ray line. If this is the case, we may define a resolving time of the filter as in Eq. (7.2) by

$$\frac{2\pi}{\Delta\tau_r}\equiv\int_0^\infty d\nu|B(\nu)|^2 \tag{8.3}$$

and obtain

$$C_2=\frac{2\pi\epsilon\Gamma^2F_T{}^2}{\Delta\tau_r\Sigma_s{}^2D^4}\sum_\lambda\sum_l\int d\Sigma(i)\int d\Sigma(j)\int_0^\infty d\omega|g(\omega)|^2\mathfrak{F}(\lambda,j,\omega)\mathfrak{F}^*(l,j,\omega)\mathfrak{F}(l,i,\omega)\mathfrak{F}^*(\lambda,i,\omega)\,. \tag{8.4}$$

We have also omitted from Eq. (7.4) the factor $\exp(iY[k(\omega)-k(\omega')])$. The resolving time τ_0 is unlikely to be smaller than about 10^{-9} sec so that the exponent will be of the order of $Y(\partial k/\partial\omega)/\tau_0\approx Y/v\tau_0$, where v is the group velocity of the medium involved, in our case, the velocity of light. If Y can be controlled to within a few centimeters, which should not be difficult, the exponent can be made very small. As an incidental and irrelevant consequence, we have set $D_l=D_\lambda$; it is trivial to remove this restriction.

It is in consideration of the scattering amplitudes $\mathfrak{F}$ that we encounter the problems associated with the dynamical theory of scattering. The geometrical constraints implied by Eq. (8.4) are associated with the fact that all four $\mathfrak{F}$'s must be nonvanishing at the *same* frequency (at least to within the bandwidths of the filters). It is easy to see that if the crystal were infinite in extent, this would be essentially impossible, since scattering would then occur only for changes in momentum precisely equal to a reciprocal lattice vector of the crystal. For the first two amplitudes in Eq. (8.4) corresponding to the source point j this is not a problem, as is shown in Fig. 6. There $\mathbf{k}_1$ is the incident photon wave-number vector and we are required by the Bragg condition to add it to a reciprocal lattice vector, so that the sum of the two has the same magnitude as $\mathbf{k}_1$. This is achieved by rotating the crystal (represented by its reciprocal lattice) around the tip of $\mathbf{k}_1$, until the terminus of the reciprocal lattice vector $\mathbf{g}_1$ lies on the sphere of reflection, as shown. This is, of course, the standard Bragg construction. There is one further degree of freedom to be exploited; namely a rotation of the crystal about the axis represented by $\mathbf{g}_1$. In this way, a second reciprocal lattice vector $\mathbf{g}_2$ may also be made to lie on the reflection sphere. Thus, unless the $\mathbf{g}$'s are too large, it is generally possible to achieve two reflections at the same time, at any frequency. Needless to say, the geometrical constraint is precise, and therefore unachievable. It is only to the extent that reflected lines have a finite width, for a finite crystal, that the experiment is possible.

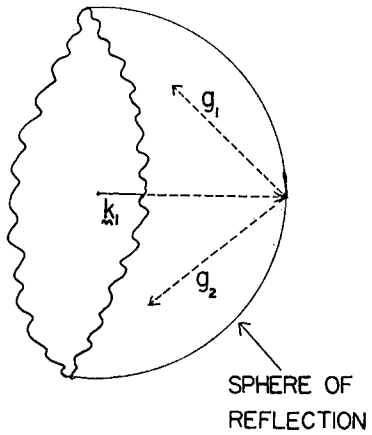

FIG. 6. Construction to illustrate the illumination of two Bragg reflections at once.

There is no simple cure to this geometrical problem analogous to the crystal rotation technique of ordinary x-ray diffraction. To put any given lattice vector $\mathbf{g}_1$ on the sphere of reflection, it is customary to rotate the crystal about some axis, allowing $\mathbf{g}_1$ to pass through the sphere from time to time. Each time it does, some photons are scattered and the accumulation of these produces the spot. The Lorentz factor measures, in effect the rate at which the terminus of $\mathbf{g}_1$ goes through the sphere. Although in our case, also, $\mathbf{g}_1$ and $\mathbf{g}_2$ could be made to go through the sphere by rotation, there is no way to assure that they go through at the same time, which is what we need for our experiment. There seems to be no alternative to painstaking alignment. If the angular width of the scattering from a good crystal is a few seconds of arc (due to primary extinction), then the alignment will have to be that good to achieve the required conditions. A 0.1 mm collimator at 10 m represents 2 sec of arc.

Consider now the remaining two $\mathfrak{F}$'s in Eq. (8.4) and ask whether they can be made nonvanishing with the now fully determined geometry. The only free variable is the direction of illumination from the second source, $\mathbf{k}_2$. We require that it make two Bragg reflections which will land on the detectors placed at the termini of $\mathbf{k}_1+\mathbf{g}_1$ and $\mathbf{k}_1+\mathbf{g}_2$. Thus, both $\mathbf{k}_1+\mathbf{g}_1-\mathbf{k}_2$ and $\mathbf{k}_1$

$+\mathbf{g}_2-\mathbf{k}_2$ must be reciprocal lattice vectors, say $\mathbf{g}_3$ and $\mathbf{g}_4$. Since $\mathbf{g}_4-\mathbf{g}_3=\mathbf{g}_1-\mathbf{g}_2$, which by virtue of the group property is itself a reciprocal lattice vector we see that it is sufficient if either one fulfills the requirement. It follows that $\mathbf{k}_2-\mathbf{k}_1$ must also be a vector in the reciprocal lattice of the crystal, in its given orientation. Since $\mathbf{k}_2$ must have the same magnitude as $\mathbf{k}_1$, and we have just laboriously given $\mathbf{k}_1$ its two Bragg reflections, this can only mean that $\mathbf{k}_2$ *is* either $\mathbf{k}_1+\mathbf{g}_1$ or $\mathbf{k}_1+\mathbf{g}_2$. In that case, one of the detectors lies in the forward direction for the second source, and no real scattering has occurred. It is not immediately clear that information cannot be obtained from this configuration, but the experimental problems are severe with a detector in the direct beam. We abandon this arrangement for the present.

The only other possibility is that $\mathbf{k}_2-\mathbf{k}_1$ is a reciprocal lattice vector other than $\mathbf{g}_1$ or $\mathbf{g}_2$, which in turn implies that *three* Bragg reflections must be possible for the first source. This does not in general happen because we have exhausted all of our geometrical degrees of freedom in achieving two scatterings. However, there remains one parameter, the frequency $\sim|\mathbf{k}_1|$.

If in Fig. 6, we consider the origin of $\mathbf{g}_1$ and $\mathbf{g}_2$, the termini of $\mathbf{g}_1$ and $\mathbf{g}_2$, and the terminus of some other reciprocal lattice vector $\mathbf{g}_3$ measured from the same origin, these four points determine a possible sphere of reflection. The vector $\mathbf{k}_1$ from the center of the sphere to the origin of $\mathbf{g}_1$, $\mathbf{g}_2$, and $\mathbf{g}_3$ is a possible source direction for the illumination of $\mathbf{k}_1+\mathbf{g}_1$, and $\mathbf{k}_1+g_2$. By our construction, $\mathbf{k}_1+\mathbf{g}_3$ will be a second, equally good, source direction. Thus all of our experimental requirements are fulfilled, but at the rather fearful price of having to work with a small rigidly determined part of the continuous spectrum; the crystal itself serves as the monochromator.

We shall not give a numerical estimate of the correlator counting rate (8.4). Nor shall we discuss the problem of experimental achievement of the required remarkable conjunction of circumstances. These questions will be taken up in a separate publication.

There are evidently a large variety of possible applications of the general theory presented here. For example, it may well be that electron scattering, rather than x-ray scattering, will be a preferable technique. The final calculations will have to be performed with a specific experiment in mind and not being experts in this field we may well be evaluating an experimentally uninteresting case.

APPENDIX

The question was raised in Sec. II of deducing the scattering amplitude $\mathfrak{F}(\Delta\mathbf{k})$ from a measurement of its magnitude $|\mathfrak{F}(\Delta\mathbf{k})|$. It was remarked that by a process of analytic continuation a discrete set of $\mathfrak{F}(\Delta\mathbf{k})$'s may be obtained, all having the same magnitude.

We shall discuss this problem only for x-ray scattering, limiting ourselves to the case that the scattering amplitudes for all electrons are equal.[29] Then the x-ray form factor is [see Eq. (4.35)]

$$\eta(\mathbf{g})=\int d^3z\rho(\mathbf{z})e^{i\mathbf{g}\cdot\mathbf{z}}, \tag{A1}$$

where the scattering amplitudes f_α have been factored out,

$$\mathbf{g}=\bar{k}(\hat{R}_j-\hat{D}_l), \tag{A2}$$

and $\rho(\mathbf{z})$ is the electron density in the target. [For the case of scattering from a crystal, we interpret $\rho(\mathbf{z})$ to be the electron density within the unit cell.]

Let us now write

$$\begin{aligned}\mathbf{g}&=g\hat{e},\\ u&\equiv\mathbf{z}\cdot\hat{e}\\ \mathbf{z}&\equiv\mathbf{y}+\hat{e}u.\end{aligned} \tag{A3}$$

Then, defining

$$k(u)\equiv\int d^2y\rho(\mathbf{z}); \tag{A4}$$

we have

$$\eta(g)=\int_{-\infty}^{+\infty}duk(u)e^{igu}, \tag{A5}$$

which (of course) depends parametrically on $\hat{e}$. We may also suppose that

$$k(u)=0, \text{ except for } 0\leq u\leq L, \tag{A6}$$

and observe that the real function $k(u)$ is bounded and non-negative; that is,

$$k(u)\geq 0. \tag{A7}$$

Therefore, Eq. (A5) is equivalent to

$$\eta(g)=\int_0^\infty duk(u)e^{igu}=\int_0^L du\,k(u)e^{igu}. \tag{A8}$$

It is apparent from Eq. (A8) that $\eta(g)$ is analytic, except for $\mathrm{Im}(g)=-\infty$, in the entire complex g plane. For $\mathrm{Im}(g)\to-\infty$,

$$|\eta(g)|\leq O(e^{L|\mathrm{Im}(g)|}). \tag{A9}$$

Thus, $\eta(g)$ is an integral function of order one.[30]

A coordinate translation

$$x=u-L \tag{A10}$$

permits us to define

$$\eta_L(g)\equiv e^{-iLg}\eta(g)=\int_{-\infty}^0 dxk'(x)e^{ixg}, \tag{A11}$$

where $k'(x)\equiv k(u)$, is an integral function of order one, analytic except at $\mathrm{Im}(g)=+\infty$.

[29] This assumption is not essential for our argument, but simplifies somewhat the discussion.

[30] See E. C. Titchmarsh, *Theory of Functions* (Oxford University Press, Oxford, 1939), 2nd ed., Chap. VIII.

The conventional scattering experiment [see Eq. (4.35)] provides a measurement of $|\eta(g)|$ for real g. To deduce the electron density $\rho(\mathbf{z})$, one must know $\eta(g)$ (for real g) as a function of the parameter $\hat{e}$. Dispersion relations for the real and imaginary parts of η may be deduced in various forms from Eq. (A8). A typical one of such relations is

$$\eta_i(g)=\frac{2g}{\pi}P\int_0^\infty \frac{dg'\eta_r(g')}{(g')^2-g^2}, \tag{A12}$$

where η_r and η_i are the real and imaginary parts of $\eta(g)$ (for real g), respectively. One might contemplate combining the measured value of

$$\eta_r^2(g)+\eta_i^2(g),$$

with Eq. (A12) to deduce η_r and η_i separately as functions of g. This would provide, then, a solution to the "phase problem," or the problem of deducing $\eta(g)$ from $|\eta(g)|$. As we shall now show, the solutions so obtained are unfortunately not unique.

Let us suppose that the zeros of $\eta(g)$ [Eq. (A8)] occur at the values of $g_1, g_2\cdots$ of g so $\eta(g_n)=0$ for $n=1, 2, \cdots$. Then $\eta(g)$ has the representation[31]

$$\eta(g)=ce^{ag}\prod_n[(1-(g/g_n))e^{g/g_n}], \tag{A13}$$

where c and a are constants and the product extends over all zeros g_n of η. We first observe that

$$\eta(0)=c \text{ is real.} \tag{A14}$$

We see, next, that Eq. (A8) implies that

$$\eta^*(-g^*)=\eta(g). \tag{A15}$$

From this relation and Eq. (A13) it follows that the zeros g_n either lie on the imaginary axis so $g_n=\pm i|g_n|$, or they are paired so that

$$g_n=-g_{\bar{n}}^* \tag{A16}$$

for a suitable pairing of indices n and $\bar{n}$. Equation (A15) also implies that the constant a is pure imaginary, so $a=id$ with d real

$$\eta(g)=ce^{idg}\prod_n[(1-(g/g_n))e^{g/g_n}]. \tag{A17}$$

To study the uniqueness of $\eta(g)$, let us suppose that $N(g)$ is a second function, satisfying the conditions that

$$|N(g)|=|\eta(g)|, \quad g \text{ real}, \tag{A18}$$

and that

$$N(g)=\int_0^L duK(u)e^{igu}, \tag{A19}$$

where $K(u)\geq 0$ and is bounded. Evidently, if an $N(g)\neq\eta(g)$ can be found, the solution to the phase problem (without other conditions) is not unique.

Let us suppose that the zeros of $N(g)$ are at the points $g=G_1, G_2, \cdots$. Then N has the representation, analogous to (A17),

$$N(g)=ce^{iDg}\prod_l[(1-(g/G_l))e^{g/G_l}], \tag{A20}$$

where the condition (A18) implies that the constant c in Eqs. (A17) and (A20) is the same.

Consider now the ratio

$$\begin{aligned}U(g)\equiv e^{i\varphi(g)}&=N(g)/\eta(g)\\&=e^{i(D-d)g}\prod_l[(1-(g/G_l))e^{g/G_l}]/\\&\qquad\prod_n[(1-(g/g_n))e^{g/g_n}].\end{aligned} \tag{A21}$$

It follows from Eq. (A18) that $|U(g)|=1$, or $\varphi(g)$ is real, for real g. Therefore, the function

$$F(g)\equiv U^*(g^*)-U^{-1}(g)$$

vanishes identically for g real. Since $F(g)=0$ on the real g axis, we conclude that $F(g)=0$ everywhere (except at the singularities of F) in the complex g plane. From this we conclude that

$$U^*(g^*)=1/U(g). \tag{A22}$$

Substitution of (A21) into both sides of (A22) gives

$$\frac{\prod_l\left[\left(1-\frac{g}{G_l^*}\right)e^{g/G_l^*}\right]\prod_n\left[\left(1-\frac{g}{g_n}\right)e^{g/g_n}\right]}{\prod_n\left[\left(1-\frac{g}{g_n^*}\right)e^{g/g_n^*}\right]\cdot\prod_l\left[\left(1-\frac{g}{G_l}\right)e^{g/G_l}\right]}=1. \tag{A23}$$

Now, $U(g)$ has zeros at all (and only at) G_l's and poles at all (and only at) g_n's (except for those G_l's which coincide with g's).[32] Similarly, $U^*(g^*)$ has zeros (poles) at the $G_l^*(g_n^*)$. On the other hand, $U^{-1}(g)$ has zeros [poles at $g_n(G_l)$]. It follows, then, from Eq. (A23) that all g_n's and G_l's may be paired and placed in either of two classes:

Class I: $g_n=G_l$ (with appropriate pairing of indices)

Class II: $g_n=G_l^*$ (with appropriate pairing of indices). (A24)

Expressed in words, the functions $N(g)$ and $\eta(g)$ may differ only by a *reflection* of some of their zero's with respect to the *real* g axis. We shall see below that the number of g_n's not on the real axis may be assumed to be finite. Thus, the most general form of a function

[31] See Ref. 30, Chap. VIII.

[32] As a practical matter, we may assume that no two g_n's, or two G_l's, are equal. Were it otherwise, we could imagine a small, unobservable change in $k(u)$, or $K(u)$, to shift the zeros apart.

$N(g)$, satisfying (A18), is

$$N(g)=e^{i\varphi(g)}\eta(g),$$
$$U(g)\equiv e^{i\varphi(g)}=\prod_j{}'\frac{(g-g_j^*)}{(g-g_j)}, \tag{A25}$$

where the product runs over any given set of the zeros g_j of η. If $K(u)$ and $k(u)$ are both to be real, associated pairs [Eq. (A16)] of zeros must be reflected together.

The inherent ambiguity in the phase problem is expressed by Eqs. (A24) and (A25).[33] That is, there are a finite, discrete set of functions $N(g)$, satisfying the condition that $|N(g)|=|\eta(g)|$ for g real. These functions differ by a reflection of zeros about the real g axis.[34]

To give a more explicit discussion, let us suppose that an attempt is made to represent $k(u)$ by a *finite* number of terms in a Fourier series

$$k(u)=\sum_{n=0}^{n_0}\left[a_n\sin\frac{2\pi nu}{L}+b_n\cos\frac{2\pi nu}{L}\right],\quad 0\leq u\leq L. \tag{A26}$$

Then, from Eq. (A8), we have

$$\eta(g)=\psi(g)[e^{igL}-1]/2i, \tag{A27}$$

where

$$\psi(g)=\sum_{n=-n_0}^{n_0}\frac{Q_n}{g-(2\pi n/L)}, \tag{A28}$$

and

$$\begin{aligned}&Q_{-n}=Q_n^*,\\ &Q_n=b_n+ia_n\quad(n>0),\\ &Q_0=b_0.\end{aligned} \tag{A29}$$

Let the zero's of $\psi(g)$ be at $g=g_1, g_2\cdots g_p$ (p finite). The remaining zeros of $\eta(g)$ lie on the *real* axis at $g=2\pi n/L$, n an integer and $|n|<n_0$. The most general form of $U(g)$ [Eq. (A25)] is then

$$[(g-g_j^*)/(g-g_j)][(g+g_j)/(g+g_j^*)], \tag{A30}$$

where product runs over a selected set of those $g_1\cdots g_p$ which are not on the real axis. [For any g_j on the imaginary axis, only one factor should appear in Eq. (A30).] The function $K(u)$ is [for $0<u<L$]

$$K(u)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}dgN(g)e^{-igu}$$
$$=-\frac{1}{4\pi i}\int_c dgU(g)\psi(g)e^{-igu}. \tag{A31}$$

Here c is a contour displaced slightly above the real axis at the points $g=2\pi n/L(|n|<n_0)$ and deformed in such a manner that all poles of $U(g)$ lie *above* the contour, as illustrated in Fig. 7.

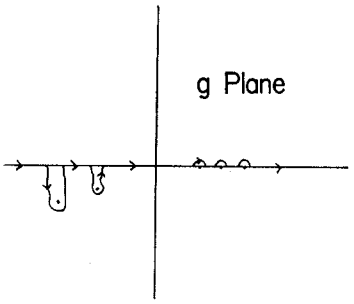

FIG. 7. The contour used in Eq. (A31).

Evaluation of Eq. (A31) gives

$$K(u)=\sum_{n=-n_0}^{n_0}\tfrac{1}{2}Q_nU\left(\frac{2\pi n}{L}\right)e^{-i2\pi nu/L}. \tag{A32}$$

On writing

$$U(2\pi n/L)\equiv e^{-i\varphi_n},\quad \varphi_{-n}=-\varphi_n, \tag{A33}$$

and using Eq. (A29), we find

$$K(u)=\sum_{n=0}^{n_0}[a_n\sin(2\pi nu/L+\varphi_n)$$
$$+b_n\cos(2\pi nu/L+\varphi_n)], \tag{A34}$$

which should be compared with Eq. (A26).

The inherent ambiguity in the "phase problem" is expressed by Eqs. (A26) and (A34). It must be noted, however, that the phases φ_n in Eq. (A34) are not arbitrary, but are members of a discrete set of phases which are *uniquely* determined by the function $k(u)$.

[33] It is hoped to return in a later publication to a discussion of further conditions to be imposed on the ambiguity in η.

[34] The reality condition on $k(u)$, expressed by Eq. (A16), implies that the zeros must be reflected in pairs, except for those which are purely imaginary.