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On the Determination of the Phases of Electromagnetic
Scattering Amplitudes from Experimental Data[†]

MASTER

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ABSTRACT

The scattering of electromagnetic waves by systems of unknown charge distribution is considered, with the aim of establishing whether the phase of the scattered radiation (relative to incident one) can be computed theoretically from experimentally obtainable data, such as the intensity and the polarization of the scattered wave. The problem is to determine the phases of the scattering amplitude tensor from these data, and we show that this can be done by employing the unitarity relation of classical electromagnetic scattering theory. In particular, the following results are established, subject to certain restrictions on the magnitude of the intensity data:

(i) Provided the incident beam is unpolarized and the target is spherically symmetrical, knowledge of the differential cross section and the polarization of the scattered radiation is sufficient to determine uniquely, through the unitarity relation, the phases of the scattering amplitude tensor.

(ii) For a general scatterer, and a fully circularly-polarized incident beam, knowledge of the differential cross section and the ellipticity of the outgoing radiation is sufficient to completely determine the phases of the scattering amplitude.

(iii) It is shown that in the above cases the phases can be calculated symmetrically by converging iterative procedures. Conditions for convergence are established, and bounds are provided on the errors associated with the approximants obtained after an arbitrary (but finite) number of iterations.

Similar results are then established within the framework of the quantum-electrodynamical theory of light scattering.

In purpose to test these proposed methods for phase determination, we apply them to the scattering of light by very simple systems. These include the free electron, the H-atom, and a macroscopic sphere of constant refraction index. The results obtained are very accurate. Using extremely crude approximations we also apply the unitarity method to a molecular system to get an order of magnitude estimate of the phases.

Finally, we conclude by discussing the possibility of applying the above results to the phase problem in x-ray scattering from amorphous and crystalline matter, and in light scattering from small macroscopic objects.

I. INTRODUCTION

The scattering amplitude pertaining to the scattering of an electromagnetic wave by a material species can provide a great deal of information on the structural properties of the scatterer. It is well known, for instance, that in x-ray crystallography knowledge of the relevant scattering amplitude, referred to in this context as the "structure factor," is sufficient to calculate the electronic charge distribution of the crystal, hence to determine completely the structure of the latter.¹ Also in studies on small macroscopic objects, such as colloidal particles, one can deduce the magnitudes of structural parameters, such as the size of the particles, from the values of the scattering amplitude.² Unfortunately, what is generally measured in light scattering and x-ray diffraction experiments are not scattering amplitudes, but intensities or differential cross sections. For the scattering of a scalar wave, the scattering amplitude is a complex-valued function, and the differential cross section equals the square of its absolute value.³ In the case of vector waves, such as electromagnetic waves, the scattering amplitude can be expressed as a tensor, whose components are complex-valued.^{4,5} (In applications of electromagnetic scattering theory to x-ray diffraction, it is nevertheless possible and accepted to employ a scattering amplitude function, rather than a tensor⁶.) Although in electromagnetic scattering the dependence of the cross section on the elements of the tensor amplitude is somewhat complicated,⁷ it can be expressed as a quadratic form in the absolute values of the tensor components. Thus, in the scattering of both scalar and vector waves, the observable quantity depends only on the magnitude of the scattering amplitude function or tensor elements and not

on the phase or phases. The fact that intensity data do not provide explicit information on the phase of the scattering amplitude leads to the well-known phase problem of scattering and diffraction theories; that is, the phases must be obtained by intelligent guessing, from theoretical considerations, or from experimental data other than straightforward intensity measurements.⁸ There is an enormous literature on this subject, and many methods for treating the problem have been developed, especially in the study of x-ray diffraction. In spite of the great progress in this field, a general solution to the phase problem that can be implemented effectively has not been found.

To illustrate the nature of the phase problem, we briefly consider its role in x-ray diffraction. The determination of crystal structures by x-ray diffraction is based, tacitly at least, on the fundamental result that with the exception of certain limited types of structures (homometric mates), a necessary and sufficient condition for a crystal-structure model to be correct is that it provides calculated intensities which match those observed experimentally for the real crystal.^{9,10} The implication of this theorem is that the phases are specified by the intensity, since the structure determines both the magnitude and the phase of the scattering amplitude. In view of this one-to-one correspondence between structure and intensity, it is possible to attempt to find the structure by trial and error methods with the aim of fitting the intensity data, or alternatively to try to construct the phase from the intensity data by more systematic so-called "direct methods."^{9,10} Although both of these approaches have been brilliantly successful in

many applications, they do suffer from limitations which have led to a continuation of the search for alternative methods.

In this paper, we formulate a general solution to the problem of phase determination in electromagnetic scattering by the use of invariance or conservation laws associated with the Maxwell dynamics of electromagnetic waves. That conservation laws pertaining to a dynamical theory can in fact be employed in determining the phases of amplitudes has been demonstrated already by Newton¹¹ and by Gerber and Karplus¹² for a problem closely related to the present one. In the context of quantum-mechanical scattering theory, these authors have shown that for the scattering of a particle by a potential, a knowledge of the differential cross section for fixed energy and for all the angles makes possible the determination of the phase of the corresponding scattering amplitude by means of the unitarity relation,^{11,12} which the scattering amplitude must satisfy. The unitarity relation follows from the conservation of total particle flux in any scattering process and reflects the resulting unitarity of the S-matrix in quantum scattering theory. A corresponding phase determination is possible also for many-channel processes.^{11,12}

In electromagnetic scattering theory¹³ there exists a unitarity relation as a mathematical manifestation of energy flux conservation in processes where absorption is not involved. We propose to show in this paper that the unitarity relation is sufficient to determine the phases of the scattering amplitude when the intensity and polarization state of the scattered radiation are known from experiment, given that the incident beam has a specified polarization state.

In Sec. II some basic quantities in electromagnetic scattering are defined and a certain important relation between them is introduced

(e.g., the expression for the cross section in terms of the elements of the scattering amplitude tensor). The unitarity relation for the scattering amplitude tensor is given in Sec. III in terms of a set of coupled nonlinear integral equations for the phases of the amplitude. We prove immediately that the kernels of the integral operators appearing in these equations can be constructed from the differential cross section and from the polarization of the scattered radiation in the following two cases:

- (i) The incident beam is fully circularly polarized; the scatterer is non-absorbing, but there are no other restrictions on its properties.
- (ii) The incident beam is unpolarized; the scatterer is non-absorbing and has full rotational-reflectional symmetry.

To demonstrate that the phases of the scattering amplitude can be obtained from experimental data by the unitarity relation under the above conditions, we must show that the integral equations are sufficient to determine the phases, and are not merely a weak restriction that non-physical solutions can satisfy as well. Also, we must demonstrate that the phases can be constructed from these equations by a feasible mathematical procedure. To that effect we prove that the solution of the set of coupled equations is unique and that it can be obtained by an iterative procedure which converges uniformly as a function of the scattering angle, subject to a restriction on the magnitude of an integral involving the cross section. Section IV presents a description of some approximation schemes for solving the unitarity equations. Sequences of upper and lower bounds on the sines of the phases are provided and it is shown that these sequences converge from above and from below, respectively, to the true values. Also, an approximation valid when the phases are small

is suggested and it is shown that, within this approximation, the dependence of the phase on the kernel of the integral equation is considerably simpler than in the general case. In Sec. V a unitarity theorem of quantum electrodynamics is employed to treat the quantum-electrodynamical theory of photon collisions with many-electron systems. Results similar to those for the classical case are obtained when elastic scattering dominates so that other processes can be neglected. It is shown that phase determination is possible in quantum electrodynamics even when inelastic processes have to be taken into account with the restriction that collisions involving a change in the number of photons are of negligible magnitude. However, this generalization of the previous result probably has little practical importance, since it involves the assumption that the differential cross sections for all the inelastic processes are known.

In Sec. VI we provide some elementary illustrations of the phase determination method. Since the scattering problem for the free electron can be solved almost exactly, the unitarity theorem can be checked for this case and shown to provide the phase to a very high order of accuracy. The breakdown of the unitarity theorem for the ordinary, approximate theory of Rayleigh scattering is discussed and it is demonstrated that this breakdown is a consequence of the usual neglect of the back-reaction of the radiation on the electron. The theory is applied to the scattering of an electromagnetic wave by a Lorentz-type model for the hydrogen atom. We verify also that the unitarity theorem gives the true phase for the solvable problem of scattering by a sphere with constant dielectric coefficient.

We conclude in Sec. VII with a discussion of the possibilities and the difficulties of applying the unitarity method to phase problems of practical interest. Future papers will present results of applications to model systems.

II. SOME RELATIONS OF ELECTROMAGNETIC SCATTERING THEORY

In the present section we outline briefly the definitions of certain basic quantities that appear in the classical theory of electromagnetic scattering and recall the relations between them that are of importance for the subsequent development. Our purpose here is to introduce the necessary notation as well as to summarize the essential results; the development follows the excellent discussion in Newton.⁴

For monochromatic radiation, scattered by an object, the electric field $\underline{E}_{\underline{k}', \underline{k}}(\underline{r})$ at distance $\underline{r}$ (whose magnitude is $r = |\underline{r}|$) from the center of the scatterer obeys Maxwell equations and satisfies an asymptotic boundary condition of the form

$$\underline{E}_{\underline{k}', \underline{k}}(\underline{r}) \underset{r \rightarrow \infty}{\rightarrow} \underline{E}_0 e^{i \underline{k} \cdot \underline{r}} + \frac{\underline{E}_s(\underline{k}', \underline{k})}{r} e^{i k r} \quad (1)$$

The subscripts $\underline{k}, \underline{k}'$ of $\underline{E}$ (the explicit dependence of the latter upon $\underline{k}, \underline{k}'$ will be suppressed below, for simplicity of notation), refer to the wave vectors of the incident radiation and of the scattered radiation at the point of observation, respectively. We suppose that the scattering is coherent and that no absorption occurs; thus, we have $|\underline{k}| = |\underline{k}'| = k$. The scattering amplitude tensor $\underline{A}$, which connects the amplitude of the scattered vector field, $\underline{E}_s$ with the vector amplitude $\underline{E}_0$ of the incident

field is defined by the relation

$$\underline{E}_s = \underline{A} \cdot \underline{E}_o, \quad \underline{A} = \underline{\tilde{A}}(\underline{k}', \underline{k}) \quad (2)$$

It is convenient to represent $\underline{E}_s$ and $\underline{E}_o$ in suitably chosen bases. If $\hat{\underline{k}}$ is a unit vector in the direction of $\underline{k}$, we define two unit vectors, $\underline{e}_{||}^o$ and $\underline{e}_{\perp}^o$ for a given $\underline{k}'$, such that $\underline{k} = \underline{e}_{||}^o \times \underline{e}_{\perp}^o$, $\underline{e}_{||}^o$ is parallel to the so-called scattering plane determined by $\underline{k}$ and $\underline{k}'$, and $\underline{e}_{\perp}^o$ is perpendicular to it. Since at each point in space the field is perpendicular to the direction of propagation at that point, the scalar product $\underline{E}_o \cdot \hat{\underline{k}}$ vanishes and we can write

$$\underline{E}_o = \xi_{||}^o \underline{e}_{||}^o + \xi_{\perp}^o \underline{e}_{\perp}^o \quad (3)$$

The unit vectors $\xi_{||}^o$ and $\xi_{\perp}^o$ will be referred to as the linear polarization components of the incident electric field, and $(\underline{e}_{||}^o, \underline{e}_{\perp}^o)$ will be called the linear polarization vector basis for $\underline{E}_o$. Corresponding definitions can be made for the scattered electric field $\underline{E}_s$, whose direction of propagation is $\underline{k}'$, a unit vector in the direction of $\underline{k}'$; thus

$$\underline{E}_s = \xi_{||}^s \underline{e}_{||}^s + \xi_{\perp}^s \underline{e}_{\perp}^s \quad ; \quad \underline{k}' = \underline{e}_{||}^s \times \underline{e}_{\perp}^s \quad (4)$$

The notations of parallel and perpendicular are again with respect to the scattering plane.

Using the representations [Eqs. (3) and (4)], we can write Eq. (2) in a two-component notation⁴

$$\begin{pmatrix} \mathcal{E}_{||}^s \\ \mathcal{E}_{\perp}^s \end{pmatrix} = \begin{pmatrix} A_{11}^L & A_{12}^L \\ A_{21}^L & A_{22}^L \end{pmatrix} \begin{pmatrix} \mathcal{E}_{||}^o \\ \mathcal{E}_{\perp}^o \end{pmatrix}; \quad A_{ij}^L = A_{ij}^L(\tilde{k}', \tilde{k}) \quad (5)$$

where the superscript indicates that the tensor A^L is the scattering amplitude in the linear polarization "representation". It must be noted that, although Eq. (5) is clearly equivalent to Eq. (2), it is not a representation of Eq. (2) in the strict mathematical sense of this term, since different bases are used to represent $\underline{E}_o$ and $\underline{E}_s$ in Eq. (5). There is another "representation," the circular polarization representation that is of equal importance in practical applications. We define the (complex) unit vectors

$$\underline{\xi}_1^o = \frac{1}{\sqrt{2}} (\underline{e}_{\perp}^o - i \underline{e}_{||}^o) \quad , \quad (\underline{\xi}_1^o)^* = \underline{\xi}_2^o \quad (6)$$

and analogously define the unit vectors $\underline{\xi}_1^s, \underline{\xi}_2^s$. The incident and scattered field can now be written

$$\underline{E}_o = \mathcal{E}_r^o \underline{\xi}_1^o + \mathcal{E}_{\ell}^o \underline{\xi}_2^o \quad ; \quad \underline{E}_s = \mathcal{E}_r^s \underline{\xi}_1^s + \mathcal{E}_{\ell}^s \underline{\xi}_2^s \quad (7)$$

where $\mathcal{E}_r^o, \mathcal{E}_{\ell}^o$ and $\mathcal{E}_r^s, \mathcal{E}_{\ell}^s$ are the circular polarization components of $\underline{E}_o$ and $\underline{E}_s$, respectively. The equation corresponding to Eq. (5) in the new representation is

$$\begin{pmatrix} \mathcal{E}_r^s \\ \mathcal{E}_{\ell}^s \end{pmatrix} = \begin{pmatrix} A_{11}^C & A_{12}^C \\ A_{21}^C & A_{22}^C \end{pmatrix} \begin{pmatrix} \mathcal{E}_r^o \\ \mathcal{E}_{\ell}^o \end{pmatrix} \quad (8)$$

where $\tilde{A}^C$ is the scattering amplitude tensor in the circular polarization representation. Using the definitions of $\underline{e}_i^O$, $\underline{e}_i^S$, $\underline{\xi}_i^O$, $\underline{\xi}_i^S$ ($i = 1, 2$), together with Eqs. (5) and (8), one can show that⁴

$$\tilde{A}^C = \tilde{B}^{(CL)} \tilde{A}^L \tilde{B}^{(CL)\dagger}, \quad \tilde{B}^{(CL)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix} \quad (9)$$

that is, Eq. (9) is the transformation law from the components of the scattering amplitude in the linear polarization representation, A_{ij}^L , to the components of the scattering amplitude in the circular polarization representations, A_{ij}^C .

It is sometimes useful to describe the incident and the scattered radiation by a set of parameters, called the Stokes parameters.⁴ For the incident beam in the linear polarization representation, the Stokes parameters are

$$\begin{aligned} I_o &= |\mathcal{E}_{||}^O|^2 + |\mathcal{E}_{\perp}^O|^2, & Q_o &= |\mathcal{E}_{||}^O|^2 - |\mathcal{E}_{\perp}^O|^2 \\ U_o &= -2\text{Re}[\mathcal{E}_{||}^O (\mathcal{E}_{\perp}^O)^*], & V_o &= -2\text{Im}[\mathcal{E}_{||}^O (\mathcal{E}_{\perp}^O)^*] \end{aligned} \quad (10)$$

where

$$I_o^2 = Q_o^2 + U_o^2 + V_o^2$$

i.e., the four parameters are not independent (but see below). Analogous definitions are made for the scattered beam. Alternatively, the Stokes parameters can be expressed in terms of the circular polarized components.⁴

The first Stokes parameter I_o (I_s) is an extremely important quantity--the intensity of incident (scattered) beam, a readily measurable observable.

In terms of I_o and I_s , differential cross section scattering is

$$\frac{d\sigma}{d\Omega} = \frac{I_s}{I_o} \quad (11)$$

It corresponds to the scattered flux per unit solid angle normalized to unit incident flux. To analyze the relation between differential cross section, which is an observable, and the elements of the scattering amplitude we introduce two additional quantities defined in terms of the Stokes parameters; for the incident radiation, they are

$$\tan 2\beta_o = V_o (U_o^2 + Q_o^2)^{-1/2} \quad (12)$$

$$\tan 2\chi_o = \frac{U_o}{Q_o} ; \quad (13)$$

β_s and χ_s are defined analogously. For an elliptically polarized plane wave, the ellipticity is specified by $\tan \beta$; that is, $\tan \beta$ equals the ratio of the ellipse minor axis to the major axis. It is thus a quantitative measure of the polarization of the wave (e.g., if $\tan \beta = 0$ or $\pm\infty$, the wave is linearly polarized; if $|\tan \beta| = 1$ it is circularly polarized). For such a general elliptically polarized wave, the angle χ measures the orientation of the polarization ellipse; i.e., the angle by which its major axis is rotated with regard to the vector $\underline{e}_1^o$.⁴

The Stokes parameters are not sufficient to uniquely specify a monochromatic elliptically polarized wave, since the phase is not determined by them. For such a wave propagating in the z -direction, we can write the expression

$$\underline{E}(z,t) = 2I^{1/2} [\hat{u}_1 \sin \beta \cos(\omega t - kx + \alpha) + \hat{u}_2 \cos \beta \sin(\omega t - kz + \alpha)]$$

where α is the phase and $\hat{u}_1$ and $\hat{u}_2$ are unit vectors directed along the principal axes of the ellipse. Because the phase α is not determined by the Stokes parameters, they can be used for the description of an incoherent beam.⁴ An incoherent wave can be formed by the supervision of many coherent wavelets with random phases and arbitrary polarizations. If $I^{(n)}$, $Q^{(n)}$, $U^{(n)}$, $V^{(n)}$ are the Stokes parameters of the n -th wavelet, we define the Stokes parameters for the incoherent wave by the sums

$$I \equiv \sum_n I^{(n)}, Q \equiv \sum_n Q^{(n)}, U \equiv \sum_n U^{(n)}, V \equiv \sum_n V^{(n)} \quad (14)$$

It is important to note that I in Eq. (14) is the intensity for the incoherent wave,⁴ because of the cancellation resulting from the assumed random choice of phases. The inequality $I^2 \geq Q^2 + U^2 + V^2$ obtains now, and the Stokes parameters of Eq. (14) are independent. Thus, for incoherent radiation, it is appropriate to define the degree of polarization P_o of the incident wave by the relation

$$P_o = \frac{(Q_o^2 + U_o^2 + V_o^2)^{1/2}}{I_o} \quad (15)$$

a corresponding definition holds for the polarization P_s of the scattered beam.⁴

It is now possible to write down (without proof) the expression for the differential cross section in terms of the A_{ij}^L or A_{ij}^C , and the parameters defined above;⁷ we have

$$\begin{aligned}
\frac{d\sigma}{d\Omega} = & 1/2 (|A_{11}^L|^2 + |A_{12}^L|^2 + |A_{21}^L|^2 + |A_{22}^L|^2) \\
& + 1/2 P_o \cos 2\chi_o \cos 2\beta_o (|A_{12}^L|^2 - |A_{11}^L|^2 - |A_{21}^L|^2 + |A_{22}^L|^2) \\
& + P_o \sin 2\chi_o \cos 2\beta_o \operatorname{Re}(A_{12}^L A_{11}^{L*} + A_{22}^L A_{21}^{L*}) \\
& + P_o \sin 2\beta_o \operatorname{Im}(A_{12}^L A_{11}^{L*} + A_{22}^L A_{21}^{L*})
\end{aligned} \tag{16}$$

An expression for the cross section in terms of the A_{ij}^C 's can be derived from Eq. (16) by using the transformation law [Eq. (9)]. It should be stressed that in Eq. (16), in addition to the cross section on the left-hand side, the parameters P_o , $\sin 2\chi_o$, $\cos 2\beta_o$ of the incident beam can, in principle, also be measured. $\sin 2\chi_o$ is usually much harder to obtain than the two other parameters.

If the incident beam is unpolarized, the degree of polarization of the scattered wave is⁴

$$P_s^2 = 1 - \frac{|\det(\tilde{A})|^2}{\left(\frac{d\sigma}{d\Omega}\right)^2} \tag{17}$$

The equation for the angle of ellipticity of the scattered wave is⁴

$$\sin 2\beta_s = \frac{\operatorname{Im}(A_{21}^L A_{11}^{L*} + A_{22}^L A_{12}^{L*})}{\left[\left(\frac{d\sigma}{d\Omega}\right)^2 - |\det(\tilde{A})|^2\right]} \tag{18}$$

III. THE UNITARITY THEOREM AND THE DETERMINATION OF THE PHASES

In the scattering of an electromagnetic wave by a non-absorbing object, the energy flux of the outgoing wave must equal that of the incoming wave. When this conservation law is combined with the time-reversal invariance which the scattering process must obey as well because of the form of Maxwell's equation, a non-trivial restriction on the scattering amplitude arises; that is, the scattering amplitude must satisfy the unitarity relation or the generalized optical theorem.¹⁴ The unitarity relation is most easily obtained from two properties--unitarity and reciprocity--possessed by the electromagnetic scattering matrix (S-matrix), which reflect the energy flux conservation and the time-reversal invariance.^{14,15} A clear and simple derivation of these properties of the S-matrix and the subsequent deduction of the unitarity relation for a spherically symmetric scatterer can be found in Newton;¹⁴ the generalization to a scatterer of arbitrary symmetry is given by Saxon.^{14,15}

The unitarity relation has the form

$$\text{Im} \tilde{A}(\underline{k}', \underline{k}) = \frac{k}{4\pi} \int d\Omega_{\underline{k}''} \tilde{A}(\underline{k}', \underline{k}'') \tilde{A}^*(\underline{k}'', \underline{k}) \quad (19)$$

In the above equation the integration on the right-hand side is over all directions of the vector $\underline{k}''$. Since there is no absorption, the lengths of the vectors $\underline{k}$, $\underline{k}'$, $\underline{k}''$ are equal ($|\underline{k}''| = |\underline{k}'| = |\underline{k}| = k$). As $\tilde{A}$ is a matrix, Eq. (19) is a set of coupled equations involving the elements of this matrix. In the linear polarization representation, Eq. (19) has the form

$$\begin{aligned} \text{Im} A_{ji}^L(\underline{k}', \underline{k}) &= \frac{k}{4\pi} \sum_{\ell=1}^2 \int d\Omega_{k''} A_{j\ell}^L(\underline{k}', \underline{k}'') A_{\ell i}^{L*}(\underline{k}'', \underline{k}) \\ &= \frac{k}{4\pi} \sum_{\ell=1}^2 \int d\Omega_{k''} A_{\ell j}^L(\underline{k}'', \underline{k}') A_{\ell i}^{L*}(\underline{k}'', \underline{k}), \quad j, i=1, 2 \end{aligned} \quad (20)$$

Corresponding equations hold for scattering amplitude components A_{ji}^C , as can be verified by applying Eq. (9) to Eq. (20). For simplicity of notation, we drop the subscript k'' of $d\Omega_{k''}$ and the superscript L or C of A_{ji}^L and A_{ji}^C , except when confusion between linear and circular polarization representations can arise.

We introduce the symbols d_{ji} and α_{ji} to denote, respectively, the magnitude and the phase of A_{ji} ,

$$A_{ji}(\underline{k}', \underline{k}) = d_{ji}(\underline{k}', \underline{k}) e^{i \alpha_{ji}(\underline{k}', \underline{k})} \quad (21)$$

Substituting Eq. (21) in Eq. (20), we obtain

$$\begin{aligned} d_{ji}(\underline{k}', \underline{k}) \sin \alpha_{ji}(\underline{k}', \underline{k}) &= \frac{k}{4\pi} \sum_{\ell=1}^2 \int d\Omega d_{\ell j}(\underline{k}'', \underline{k}') d_{\ell i}(\underline{k}'', \underline{k}) \\ &\quad \cos[\alpha_{\ell j}(\underline{k}'', \underline{k}') - \alpha_{\ell i}(\underline{k}'', \underline{k})] \quad j, i=1, 2 \end{aligned} \quad (22)$$

If we define

$$L_{ji}^{\ell}(\underline{k}'', \underline{k}', \underline{k}) = \frac{k}{4\pi} \frac{d_{\ell j}(\underline{k}'', \underline{k}') d_{\ell i}(\underline{k}'', \underline{k})}{d_{ji}(\underline{k}', \underline{k})}, \quad (23)$$

Eq. (22) can be written in the form

$$\sin \alpha_{ji}(\underline{k}', \underline{k}) = \sum_{\ell=1}^2 \int d\Omega L_{ji}^{\ell}(\underline{k}'', \underline{k}', \underline{k}) \cos[\alpha_{\ell j}(\underline{k}'', \underline{k}') - \alpha_{\ell i}(\underline{k}'', \underline{k})] \quad j, i=1, 2 \quad (24)$$

Equations (24) are the basic set of coupled integral equations of our method to determine the phases α_{ji} of the scattering amplitude tensor. To demonstrate that they can serve this purpose, we must show first that the kernels L_{ji}^{ℓ} for all the directions of the vectors $\underline{k}''$ and $\underline{k}'$ can be computed from experimental data. Even then it is still possible a priori that Eq. (24) is only a necessary restriction on the phases that is not sufficient to eliminate non-physical solutions. Thus, it is essential to demonstrate that the solutions of Eq. (24) are unique and, therefore, equal the true physical phases, or at most involve trivial ambiguities that do not effect the applications of the calculated phases. Finally, we need to show that there exist mathematical procedures for solving Eq. (24).

In what follows, we show that the above requirements for phase determination by Eq. (24) are satisfied under two types of experimental situations. For convenience we phrase the results in terms of two theorems. The first is for scattering of an unpolarized beam by a nonabsorbing object with complete rotation-reflection symmetry and the second is for scattering of a circularly polarized beam by an arbitrary nonabsorbing object. These two situations are not the only ones for which Eq. (24) can be employed. However, they do represent realistic experiments for which relatively simple results are obtained.

Theorem 1:

We consider a non-absorbing scatterer with complete rotational-reflectional symmetry and confine our attention to scattering experiments in which an unpolarized incident beam is employed. For this case, the linear polarization representation is most convenient. It follows that:

a) The kernels $L_{ji}^{\ell}(\underline{k}'', \underline{k}', \underline{k})$ can all be calculated if the differential cross section and the degree of polarization of the scattered wave are known. The kernels have the form

$$L_{ji}^{\ell}(\underline{k}'', \underline{k}', \underline{k}) = \delta_{ji} \delta_{j\ell} \delta_{i\ell} \frac{k}{4\pi} \frac{[1+(-)^{j+1}P_s(\theta')]^{1/2} \left[\frac{d\sigma(\theta')}{d\Omega}\right]^{1/2} [1+(-1)^{j+1}P_s(\theta'')]^{1/2} \left[\frac{d\sigma(\theta'')}{d\Omega}\right]^{1/2}}{[1+(-1)^{j+1}P_s(\theta)]^{1/2} \left[\frac{d\sigma(\theta)}{d\Omega}\right]^{1/2}}$$

$$j, i, \ell = 1, 2. \quad (25)$$

In the above equation the angle θ is the angle between the initial wave vector $\underline{k}$ and the final wave vector $\underline{k}'$; i.e., $\frac{d\sigma(\theta)}{d\Omega} \equiv \frac{d\sigma(\underline{k}', \underline{k})}{d\Omega}$ and $P_s(\theta) \equiv P_s(\underline{k}', \underline{k})$. Correspondingly, the angles θ' and θ'' denote the scattering directions $\underline{k}$ to $\underline{k}''$ and $\underline{k}'$ to $\underline{k}''$

b) For $j = 1, 2$, the R_j constants are chosen such that

$$R_j > \int d\Omega L_{jj}^j(\underline{k}'', \underline{k}', \underline{k}) \equiv S_j(\underline{k}', \underline{k}) \quad (26)$$

The solutions of Eq. (24) for $\sin\alpha_{jj}(\underline{k}', \underline{k})$ are unique if the R_j satisfy the condition

$$2R_j \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) < 1 \quad \text{for } j = 1, 2. \quad (27)$$

c) Subject to the above condition [Eq. (27)], the iteration sequence defined by

$$\sin\alpha_{jj}^{(N+1)}(\underline{k}', \underline{k}) = \int L_{jj}^j(\underline{k}'', \underline{k}', \underline{k}) \sin\alpha_{jj}^{(N)}(\underline{k}'', \underline{k}') \sin\alpha_{jj}^{(N)}(\underline{k}'', \underline{k}) d\Omega$$

$$+ \int L_{jj}^j(\underline{k}'', \underline{k}', \underline{k}) \cos\alpha_{jj}^{(N)}(\underline{k}'', \underline{k}') \cos\alpha_{jj}^{(N)}(\underline{k}'', \underline{k}) d\Omega \quad (28)$$

with $\sin \alpha_{jj}^{(0)}(\underline{k}', \underline{k}) \equiv 0$ and $\cos \alpha_{jj}^{(N)}(\underline{k}', \underline{k}) \equiv +\sqrt{1 - \sin^2 \alpha_{jj}^{(N)}}$ is valid for an arbitrary number of steps and converges to the true solution of Eq. (24); that is,

$$\sin \alpha_{jj}^{(N)}(\underline{k}', \underline{k}) \xrightarrow{N \rightarrow \infty} \sin \alpha_{jj}(\underline{k}', \underline{k}) \quad (29)$$

The convergence in Eq. (29) is uniform as a function of the orientation of $\underline{k}'$ and $\underline{k}$.

Of the four phases $\alpha_{11}, \alpha_{22}, \alpha_{21}, \alpha_{12}$, the latter two can be set equal to zero [$\sin \alpha_{ji} = 0, j \neq i$] since

$$A_{ji}^L(\underline{k}', \underline{k}) = 0 \text{ for } j \neq i. \quad (30)$$

where the subscript L here reminds the reader that Eq. (30) is valid only for the linear polarization representation which is being used in Theorem 1.

In short, there are only two non-zero phases (α_{11}, α_{22}) under the conditions of the theorem and the sines of these non-trivial phases can be obtained by the iterative procedure [Eq. (28)] which converges to the unique, physical solutions of Eq. (24).

Theorem 2:

We consider a non-absorbing, but otherwise arbitrary scatterer. If the differential cross section and the ellipticity of the scattered wave are measured for the scattering of both right- and left-hand circularly polarized radiation, it follows that:

a) The magnitudes $d_{ji}(\underline{k}', \underline{k})$ of the scattering amplitude components $A_{ji}^C(\underline{k}', \underline{k})$ can be calculated for all directions of $\underline{k}'$ from the experimental data. They are

$$d_{11}(\underline{k}', \underline{k}) = \left(\frac{d\sigma(\theta)}{d\Omega}\right)^{1/2} [1 + (\tan 2\beta_s + \{\cos 2\beta_s\}^{-1})^2]^{-1/2}$$

$$d_{22}(\underline{k}', \underline{k}) = \left(\frac{d\sigma(\theta)}{d\Omega}\right)^{1/2} [1 + (\tan 2\beta_s + \{\cos 2\beta_s\}^{-1})^2]^{-1/2} [\tan 2\beta_s + \{\cos 2\beta_s\}^{-1}] \quad (31b)$$

$$d_{21}(\underline{k}', \underline{k}) = \left(\frac{d\sigma(\theta)}{d\Omega}\right)^{1/2} [1 + (\tan 2\bar{\beta}_s + \{\cos 2\bar{\beta}_s\}^{-1})^2]^{-1/2} [\tan 2\bar{\beta}_s + \{\cos 2\bar{\beta}_s\}^{-1}] \quad (31c)$$

$$d_{22}(\underline{k}', \underline{k}) = \left(\frac{d\sigma(\theta)}{d\Omega}\right)^{1/2} [1 + (\tan 2\bar{\beta}_s + \{\cos 2\bar{\beta}_s\}^{-1})^2]^{-1/2} \quad (31d)$$

where θ is the $\underline{k}$ to $\underline{k}'$ scattering angle and β_s the ellipticity angle of the scattered wave as defined in Sec. II. In the above equations $\frac{d\sigma(\theta)}{d\Omega}$ and β_s denote respectively the cross section and ellipticity angle measured in the scattering of the right-handedly polarized beam, while $\frac{d\sigma(\theta)}{d\Omega}$ and $\bar{\beta}_s$ are the corresponding quantities pertaining to the experiment with the left-handedly polarized wave.

A knowledge of the magnitudes $d_{ji}(\underline{k}', \underline{k})$ for $j, i=1, 2$ for all the directions of $\underline{k}'$ and $\underline{k}$ is sufficient to determine the kernels $L_{ji}^{\ell}(\underline{k}'', \underline{k}', \underline{k})$ defined in Eq. (23).

b) For $j, i=1, 2$, the constants R_{ji} are chosen such that

$$R_{ji} > \sum_{\ell} \int d\Omega L_{ji}^{\ell}(\underline{k}'', \underline{k}', \underline{k}) \equiv S_{ji} \quad (32)$$

The solutions of Eq. (24) regarded as a set of coupled equations for $\sin \alpha_{ji}(\underline{k}', \underline{k})$ are unique if the R_{ji} satisfy the condition

$$J = 2R(1 + \frac{R}{\sqrt{1-R^2}}) < 1, \quad R \equiv \text{Max}_{ji}\{R_{ji}\} \quad (33)$$

c) Subject to the above condition [Eq. (33)], the iteration sequence defined by

$$\begin{aligned} \sin \alpha_{ji}^{(N+1)}(\underline{k}', \underline{k}) = & \sum_{\ell} \int d\Omega L_{ji}^{\ell}(\underline{k}'', \underline{k}', \underline{k}) [\cos \alpha_{\ell j}^{(N)}(\underline{k}'', \underline{k}') + \cos \alpha_{\ell i}^{(N)}(\underline{k}'', \underline{k}) \\ & + \sin \alpha_{\ell j}^{(N)}(\underline{k}'', \underline{k}) \sin \alpha_{\ell i}^{(N)}(\underline{k}'', \underline{k})] \end{aligned} \quad (34)$$

with $\sin \alpha_{ji}^{(0)}(\underline{k}', \underline{k}) = 0$ and $\cos \alpha_{ji}^{(N)}(\underline{k}', \underline{k}) \equiv +\sqrt{1 - \sin^2 \alpha_{ji}^{(N)}(\underline{k}', \underline{k})}$, converges to the true solution $\sin \alpha_{ji}$ of Eq. (24); that is,

$$\sin \alpha_{ji}^{(N)}(\underline{k}', \underline{k}) \xrightarrow{N \rightarrow \infty} \sin \alpha_{ji}(\underline{k}', \underline{k}). \quad (35)$$

The convergence in Eq. (35) is uniform as a function of orientation of $\underline{k}'$ and $\underline{k}$. The above theorem provides a general solution of the phase problem for experiments with circularly polarized incident beams. It is possible to prove a corresponding theorem for experiments with linearly polarized incident waves.

Proofs of the two essential theorems are given below and some illustrative applications are outlined in the subsequent sections.

III(1) Proof of Theorem 1

We present the proof of Theorem 1 here by considering each of the parts in the order given above.

a) For a scatterer having full rotational-reflectional symmetry, the scattering amplitude is diagonal in the linear polarization representation;¹⁶ that is,

$$A_{ji}^L(\underline{k}', \underline{k}) = 0 \text{ for } j \neq i. \quad (36)$$

For the scattering of an unpolarized incident beam, the polarization $P_0 = 0$. Substitution of this result and of Eq. (36) into expression Eq. (16) for the differential cross section yields the simple expression

$$\frac{d\sigma}{d\Omega} = 1/2 (|A_{11}^L|^2 + |A_{22}^L|^2) = 1/2 (d_{11}^2 + d_{22}^2) \quad (37)$$

The degree of polarization of the scattered wave given in Eq. (17) similarly reduces to

$$P_s = \pm \frac{|A_{11}^L|^2 - |A_{22}^L|^2}{|A_{11}^L|^2 + |A_{22}^L|^2} = \pm \frac{d_{11}^2 - d_{22}^2}{d_{11}^2 + d_{22}^2} \quad (38)$$

where the sign is chosen such that $P_s \geq 0$. Solving Eqs. (37) and (38) for d_{11}, d_{22} in terms of $\frac{d\sigma}{d\Omega}$ and P_s , we obtain

$$d_{jj}(\underline{k}', \underline{k}) = [1 + (-1)^{j+1} P_s]^{1/2} \left(\frac{d\sigma}{d\Omega} \right)^{1/2}, \quad j = 1, 2. \quad (39)$$

Substituting Eq. (39) into the definition [Eq. (23)] of L_{ji}^ℓ for the case $\ell=i=j$ gives

$$L_{jj}^j = \frac{k}{4\pi} \frac{[1 + (-1)^{j+1} P_s(\theta')]^{1/2} \left[\frac{d\sigma(\theta')}{d\Omega} \right]^{1/2} [1 + (-1)^{j+1} P_s(\theta'')]^{1/2} \left[\frac{d\sigma(\theta'')}{d\Omega} \right]^{1/2}}{[1 + (-1)^{j+1} P_s(\theta)]^{1/2} \left[\frac{d\sigma(\theta)}{d\Omega} \right]} \quad (40)$$

which is identical with Eq. (25) for $\ell=i=j$. Since $A_{ij}^\ell = 0$ ($i \neq j$), the coupled integral equations [Eq. (22)] for $\sin \alpha_{ji}$ reduce to a pair of uncoupled equations for $\sin \alpha_{ji}$ ($j=1,2$) of the form

$$\sin \alpha_{ji}(\underline{k}', \underline{k}) = \int d\Omega L_{jj}^j(\underline{k}'', \underline{k}', \underline{k}) \cos[\alpha_{ji}(\underline{k}'', \underline{k}') - \alpha_{ji}(\underline{k}'', \underline{k})] \quad j=1,2 \quad (41)$$

The phases $\sin \alpha_{ji} (j \neq i)$ are not determined by Eq. (41). However, since the nondiagonal elements of the scattering amplitude tensor are zero ($A_{ji}^L = 0, j \neq i$), the phases for these elements are of no physical interest and are, in fact, not well defined; correspondingly, $L_{ji}^L (j \neq i)$ also does not enter the problem and is not well defined. For purely formal arguments, we make the choice

$$\sin \alpha_{ji} = 0, L_{ji}^L = 0 \quad (j \neq i) \quad (42)$$

so that Eqs. (40) and (42) correspond to Eq. (25), which is the expression we set out to prove.

b) Since only the diagonal phases α_{jj} appear in the experiment to which Theorem 1 applies, we must show that $\sin \alpha_{jj} (j=1,2)$ are uniquely determined by Eq. (24) or (41). We follow the standard procedure and assume that there are two different solutions, $\sin \alpha_{jj}^A(\underline{k}', \underline{k})$ and $\sin \alpha_{jj}^B(\underline{k}', \underline{k})$ to Eq. (42). Subtracting the equation obeyed by $\sin \alpha_{jj}^A$ from that satisfied by $\sin \alpha_{jj}^B$, we obtain

$$\begin{aligned} \sin \alpha_{jj}^A(\theta) - \sin \alpha_{jj}^B(\theta) &\leq \int d\Omega L_{jj}^j |\cos \alpha_{jj}^A(\theta'') \cos \alpha_{jj}^A(\theta') - \cos \alpha_{jj}^B(\theta'') \cos \alpha_{jj}^B(\theta')| \\ &+ \int d\Omega L_{jj}^j |\sin \alpha_{jj}^A(\theta'') \sin \alpha_{jj}^A(\theta') - \sin \alpha_{jj}^B(\theta'') \sin \alpha_{jj}^B(\theta')| \end{aligned} \quad (43)$$

since $L_{jj}^j \geq 0$. We let θ_n denote the value of θ (or equivalently, the values of $\underline{k}'$ and $\underline{k}$) for which $|\sin \alpha_{jj}^A(\theta) - \sin \alpha_{jj}^B(\theta)|$ attains its largest value; that is,

$$|\sin \alpha_{jj}^A(\theta_n) - \sin \alpha_{jj}^B(\theta_n)| \equiv \max_{\theta} |\sin \alpha_{jj}^A(\theta) - \sin \alpha_{jj}^B(\theta)|$$

Similarly, we denote by θ_m the value of θ for which $|\tan 1/2[\alpha_{jj}^A(\theta) + \alpha_{jj}^B(\theta)]|$ attains its largest value. With these definitions and the identity

$$\frac{\cos \alpha - \cos \beta}{\sin \alpha - \sin \beta} = -\tan \left[\frac{1}{2}(\alpha + \beta) \right]$$

we have

$$\begin{aligned} & |\cos \alpha_{jj}^A(\theta') \cos \alpha_{jj}^A(\theta'') - \cos \alpha_{jj}^B(\theta') \cos \alpha_{jj}^B(\theta'')| = \\ & = |\cos \alpha_{jj}^A(\theta') [\cos \alpha_{jj}^A(\theta'') - \cos \alpha_{jj}^B(\theta'')] + \cos \alpha_{jj}^B(\theta'') [\cos \alpha_{jj}^A(\theta') - \cos \alpha_{jj}^B(\theta')]| \\ & \leq |\tan 1/2[\alpha_{jj}^A(\theta') + \alpha_{jj}^B(\theta')]| |\sin \alpha_{jj}^A(\theta') - \sin \alpha_{jj}^B(\theta')| + \\ & + |\tan 1/2[\alpha_{jj}^A(\theta'') + \alpha_{jj}^B(\theta'')]| |\sin \alpha_{jj}^A(\theta'') - \sin \alpha_{jj}^B(\theta'')| \\ & \leq 2 |\tan 1/2[\alpha_{jj}^A(\theta_m) + \alpha_{jj}^B(\theta_m)]| |\sin \alpha_{jj}^A(\theta_n) - \sin \alpha_{jj}^B(\theta_n)| \end{aligned} \quad (44)$$

Similarly, it can be shown that

$$|\sin \alpha_{jj}^A(\theta') \sin \alpha_{jj}^A(\theta'') - \sin \alpha_{jj}^B(\theta') \sin \alpha_{jj}^B(\theta'')| \leq 2 |\sin \alpha_{jj}^A(\theta_n) - \sin \alpha_{jj}^B(\theta_n)| \quad (45)$$

Equation (42) and the choice of R_j [Eq. (26)] imply that

$$|\sin \alpha_{jj}^A(\theta)|, |\sin \alpha_{jj}^B(\theta)| \leq \int d\Omega L_{jj}^j < R_j \quad (\text{for all } \theta) \quad (46)$$

from which it follows that

$$|\tan 1/2[\alpha_{jj}^A(\theta_m) + \alpha_{jj}^B(\theta_m)]| \leq \frac{R_j}{\sqrt{1-R_j^2}} \quad (47)$$

Combining Eqs. (43)-(47), we have

$$\begin{aligned}
 |\sin \alpha_{jj}^A(\theta) - \sin \alpha_{jj}^B(\theta)| &\leq 2 \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) \int d\Omega L_{jj}^j |\sin \alpha_{jj}^A(\theta_n) - \sin \alpha_{jj}^B(\theta_n)| \\
 &\leq 2R_j \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) |\sin \alpha_{jj}^A(\theta_n) - \sin \alpha_{jj}^B(\theta_n)|
 \end{aligned}
 \tag{48}$$

The inequality Eq. (48), by its derivation, holds for any value of θ on the left-hand side, in particular for $\theta = \theta_n$. Examining Eq. (48) for this case, we note that because of condition Eq. (27), $2R_j \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) < 1$, a contradiction would arise, except if

$|\sin \alpha_{jj}^A(\theta_n) - \sin \alpha_{jj}^B(\theta_n)| = 0$. Recalling the definition of θ_n given above, we can readily recognize that the last equation implies that $\sin \alpha_{jj}^A(\theta) = \sin \alpha_{jj}^B(\theta)$ for all θ . The uniqueness of the solution of the unitarity equation Eq. (24) has thus been demonstrated.

The above proof is essentially identical with that given by Karplus and Gerber¹² for the corresponding result valid in the quantum scattering theory of particles. The same is true for the proof, part c) of Theorem 1 (see below), which also has an analogue in quantum scattering theory.

c) To prove that the iteration sequence [Eq. (28)] converges to the true solution of the unitarity relation [Eq. (44)], we show first that the procedure is defined for an arbitrary number of steps. It is not obvious that this is the case, since substituting $\sin \alpha_{jj}^M$ on the right-hand side of Eq. (28) for some $N=M$ might yield a quantity of magnitude greater than 1 for $\sin \alpha_{jj}^{M+1}$, such a result would clearly stop the iteration procedure. To demonstrate that this cannot happen, we prove by induction the following lemma:

Lemma 1:

The quantity $\sin \alpha_{jj}^N$ obtained from Eq. (28) is $0 \leq \sin \alpha_{jj}^{(N)}(\underline{k}', \underline{k}) < 1$ for all values of $(\underline{k}', \underline{k})$ and for all N . Since $\sin \alpha_{jj}^{(0)} = 0$ by definition and $L_{jj}^j \geq 0$, $\sin \alpha_{jj}^{(1)} \geq 0$ from Eq. (28); further, from Eqs. (26) and (28), $\sin \alpha_{jj}^1 < 1$. Thus, Lemma 1 holds for $N=0,1$. We now suppose that the lemma is valid for $N=M$ and show that this implies that it is true also for $N=M+1$. By Eq. (28), by the definition of $\cos \alpha_{jj}^M$ ($\cos \alpha_{jj}^M = +\sqrt{1 - \sin^2 \alpha_{jj}^M}$) and by the assumption that $0 \leq \sin \alpha_{jj}^M < 1$, it follows readily that

$$\begin{aligned} 0 < \sin \alpha_{jj}^{M+1} &= \int d\Omega L_{jj}^j \sin \alpha_{jj}^M(\theta'') \sin \alpha_{jj}^M(\theta') + \int d\Omega L_{jj}^j \sqrt{1 - \sin^2 \alpha_{jj}^M(\theta')} \sqrt{1 - \sin^2 \alpha_{jj}^M(\theta'')} \\ &= \int d\Omega L_{jj}^j \cos[\alpha_{jj}^M(\theta'') - \alpha_{jj}^M(\theta')] \leq \int d\Omega L_{jj}^j < R_j < 1 \end{aligned} \quad (49)$$

Thus, the assumption that the lemma is true for $N=M$ implies its validity for $N=M+1$. By induction, the lemma holds for every integer N .

Since the iterative procedure is well defined for any number of steps, we can proceed now to prove its convergence. Subtracting the expression for $\sin \alpha_{jj}^N$ given by Eq. (28) from that for $\sin \alpha_{jj}^{N+1}$, we obtain

$$\begin{aligned} |\sin \alpha_{jj}^{N+1}(\theta) - \sin \alpha_{jj}^N(\theta)| &\leq \int d\Omega L_{jj}^j |\cos \alpha_{jj}^N(\theta'') \cos \alpha_{jj}^N(\theta') - \cos \alpha_{jj}^{N-1}(\theta'') \cos \alpha_{jj}^{N-1}(\theta')| \\ &\quad + \int d\Omega L_{jj}^j |\sin \alpha_{jj}^N(\theta'') \sin \alpha_{jj}^N(\theta') - \sin \alpha_{jj}^{N-1}(\theta'') \sin \alpha_{jj}^{N-1}(\theta')| \end{aligned} \quad (50)$$

This inequality has the same form as Eq. (43). Thus, the trigonometric manipulation used to transform Eq. (43) can be applied to Eq. (50) to yield

$$|\sin \alpha_{jj}^{N+1}(\theta) - \sin \alpha_{jj}^N(\theta)| \leq 2R_j \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) |\sin \alpha_{jj}^N(\theta_n) - \sin \alpha_{jj}^{N-1}(\theta_n)| \quad (\text{all } \theta) \quad (51)$$

where θ_n was defined earlier in this section. Since

$$|\sin \alpha_{jj}^1(\theta)| < R_j \quad (\text{all } \theta) \quad (52)$$

from Eq. (28) by the choice of R_j [Eq. (26)] and $\sin \alpha_{jj}^{(0)} = 0$, we can substitute into Eq. (51) and obtain the sequence of inequalities

$$\begin{aligned} |\sin \alpha_{jj}^2 - \sin \alpha_{jj}^1| &\leq 2 \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) R_j^2, \quad |\sin \alpha_{jj}^3(\theta) - \sin \alpha_{jj}^2(\theta)| < 4 \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right)^2 R_j^3, \\ |\sin \alpha_{jj}^{N+1}(\theta) - \sin \alpha_{jj}^N(\theta)| &\leq 2^N \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right)^N R_j^{N+1} \end{aligned} \quad (53)$$

The limit of the iteration sequence can be expressed in the form of a

$$\begin{aligned} \lim_{N \rightarrow \infty} \sin \alpha_{jj}^{N+1}(\theta) &= \sum_{N=0}^{\infty} \{\sin \alpha_{jj}^{N+1}(\theta) - \sin \alpha_{jj}^N(\theta)\} = \sin \alpha_{jj}^1 + [\sin \alpha_{jj}^2 - \sin \alpha_{jj}^1] \\ &+ [\sin \alpha_{jj}^3 - \sin \alpha_{jj}^2] + \dots + [\sin \alpha_{jj}^{N+1} - \sin \alpha_{jj}^N] + \dots \end{aligned} \quad (54)$$

so that the convergence of the iteration sequence can be proved by showing that the series given in Eq. (54) converges. Each term in this series is bounded by the corresponding term of the geometric series,

$$R_j \left[1 + 2R_j \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) + 4R_j^2 \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right)^2 + \dots + 2^N R_j^N \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right)^N + \dots\right] \quad (55)$$

which converges provided $2R_j(1 + \frac{R_j}{\sqrt{1-R_j^2}}) < 1$, which is the condition on R_j

assumed in Eq. (27). To demonstrate that the iteration sequence converges to the solution of Eq. (24), we denote the sum of the iteration series by $\sin\alpha'_{jj}(\theta)$; that is,

$$\sin\alpha'_{jj}(\theta) = \sum_{N=0}^{\infty} \{\sin\alpha_{jj}^{N+1}(\theta) - \sin\alpha_{jj}^N(\theta)\} \quad (56)$$

Since $0 \leq \sin\alpha_{jj}^N < 1$ for all N , $0 < \sin\alpha'_{jj}(\theta) < 1$ and we can define $\cos\alpha'_{jj}(\theta) = \frac{1}{\sqrt{1-\sin^2\alpha'_{jj}}}$. Introducing a function $\beta_j(\theta)$ by the relation

$$\beta_j(\theta) \equiv \int d\Omega L_{jj}^j \sin\alpha'_{jj}(\theta'') \sin\alpha'_{jj}(\theta') + \int d\Omega L_{jj}^j \cos\alpha'_{jj}(\theta'') \cos\alpha'_{jj}(\theta'), \quad (57)$$

subtracting Eq. (28) from Eq. (57) and using the same trigonometric manipulations employed previously to transform Eq. (43), we find

$$|\beta_j(\theta) - \sin\alpha_{jj}^{N+1}(\theta)| \leq 2R_j(1 + \frac{R_j}{\sqrt{1-R_j^2}}) |\sin\alpha'_{jj}(\theta_n) - \sin\alpha_{jj}^N(\theta_n)| \quad (58)$$

We note that the partial sum $\sum_{N=0}^M \{\sin\alpha_{jj}^{N+1}(\theta) - \sin\alpha_{jj}^N(\theta)\}$ of the series [Eq. (54)] equals $\sin\alpha_{jj}^{M+1}(\theta)$. Since the partial sums converge to $\sin\alpha'_{jj}(\theta)$, we can, for arbitrarily small $\epsilon < 0$, find an integer M such that for every $N > M$

$$|\sin\alpha'_{jj}(\theta_n) - \sin\alpha_{jj}^N(\theta_n)| < \epsilon \quad (\text{all } \theta)$$

From Eqs. (58) and (27), we have

$$|\beta_j(\theta) - \sin\alpha_{jj}^{N+1}(\theta)| \leq 2R_j \left(1 + \frac{R_j}{\sqrt{1-R_j^2}}\right) \quad \varepsilon < \varepsilon \quad (59)$$

Thus, $|\beta_j(\theta) - \sin\alpha_{jj}^N(\theta)|$ becomes arbitrarily small as $N \rightarrow \infty$ and

$$\beta_j(\theta) = \lim_{N \rightarrow \infty} \sin\alpha_{jj}^N(\theta) = \sin\alpha'_{jj}(\theta) \quad (60)$$

Consequently, the quantity $\sin\alpha'_{jj}(\theta)$, which is the sum of the iteration series by Eq. (56), satisfies Eq. (57), which is the unitarity relation [Eq. (24)]. This proves that $\sin\alpha'_{jj}(\theta)$ is the desired phase $\sin\alpha_{jj}(\theta)$.

Since R_j is a constant, the convergence of the series [Eq. (55)] and, consequently, also of the series [Eq. (54)] must be independent of the value of $(\underline{k}', \underline{k})$. Thus, the convergence is uniform in the directions $\underline{k}'$ and $\underline{k}$. This result completes the proof of Theorem 1.

III(ii). Proof of Theorem 2

We prove Theorem 2 here. Certain portions of the proof, which correspond closely to that of Theorem 1, are presented in a somewhat abbreviated form.

a) For right-handed, circularly polarized incident radiation, $P_0 = 1$, $\tan\beta_0 = +1$. Substituting these values into Eq. (16), transformed to the circular polarization representation by Eq. (9), we find that the differential cross section has the form

$$\frac{d\sigma}{d\Omega} = |A_{11}^C|^2 + |A_{21}^C|^2 = d_{11}^2 + d_{21}^2 \quad (61)$$

Equation (18) for the ellipticity angle β_s becomes⁷

$$2\tan 2\beta_s = \frac{|A_{11}^C|}{|A_{21}^C|} - \frac{|A_{21}^C|}{|A_{11}^C|} = \frac{d_{11}}{d_{21}} - \frac{d_{21}}{d_{11}} \quad (62)$$

To find expressions for d_{11} and d_{21} in terms of the measurable quantities $\frac{d\sigma}{d\Omega}$ and β_s , one solves Eqs. (61) and (62) as a system of two equations in the two unknowns d_{11} and d_{21} . Since the d_{ij} (for all i,j) are non-negative by definition, only one solution of these equations is acceptable; that is,

$$d_{11}(\underline{k}', \underline{k}) = \frac{d\sigma(\theta)}{d\Omega}^{1/2} [1 + (\tan 2\beta_s + \{\cos 2\beta_s\}^{-1})^2]^{-1/2} \quad (63a)$$

$$d_{12}(\underline{k}', \underline{k}) = \frac{d\sigma(\theta)}{d\Omega}^{1/2} [1 + (\tan 2\beta_s + \{\cos 2\beta_s\}^{-1})^2]^{-1/2} [\tan 2\beta_s + \{\cos 2\beta_s\}^{-1}] \quad (63b)$$

which are Eqs. (31a) and (31b), respectively, of Theorem 2(a). Similarly, from the cross section and ellipticity angle for left-handed, circularly polarized incident radiation, the expressions for d_{12} , d_{22} can be shown to be Eqs. (31c) and (31d), respectively. Since a knowledge of the d_{ij} ($i,j = 1,2$) is sufficient to construct the kernels L_{ji}^{ℓ} defined in Eq. (23), part (a) of Theorem 2 is verified.

b) The proof that the solution of Eq. (24) is unique, subject to the condition given in Eq. (33), is somewhat more involved than the demonstration of uniqueness in Theorem 1, where the unitarity equations [Eq. (24)] uncoupled to yield two relations [Eq. (41)], each of which involved just one of the phases $\alpha_{jj}(\underline{k}', \underline{k})$ ($j=1,2$). In the present case the full set of coupled equations [Eq. (24)] must be considered and the details of the

proof are technically more complicated. Nevertheless, the method employed for demonstrating uniqueness is fundamentally unchanged, so that we need give only a sketch of the proof.

If $\sin \alpha_{ji}^A(\underline{k}', \underline{k})$, $\sin \alpha_{ji}^B(\underline{k}', \underline{k})$ ($j, i=1, 2$) denote two sets of solutions to Eq. (24), we must show that they are equal provided Eq. (33) is satisfied. Subtracting Eq. (24) for $\sin \alpha_{ji}^A$ from Eq. (24) for $\sin \alpha_{ji}^B$, applying algebraic and ~~trigonometric~~ manipulations of the type employed in obtaining Eq. (48) from Eq. (43), and introducing the inequalities of Eqs. (32) and (33), we obtain

$$|\sin \alpha_{ji}^A(\theta) - \sin \alpha_{ji}^B(\theta)| \leq 2R(1 + \frac{R}{\sqrt{1-R^2}}) \{ |\sin \alpha_{rs}^A(\theta_n) - \sin \alpha_{rs}^B(\theta_n)| \} \text{ (all } \theta) \quad (64)$$

Here θ_n denotes the value of θ for which $|\sin \alpha_{ji}^A(\theta) - \sin \alpha_{ji}^B(\theta)|$ attains its largest value, so that there may be a different θ_n for each set of indices ji . The indices r, s are chosen such that

$$|\sin \alpha_{rs}^A(\theta_n) - \sin \alpha_{rs}^B(\theta_n)| = \max_{ji} \{ |\sin \alpha_{ji}^A(\theta_n) - \sin \alpha_{ji}^B(\theta_n)| \}; \quad (65)$$

that is, the quantity with indices rs is the maximum of all of the $|\sin \alpha_{ji}^A(\theta_n) - \sin \alpha_{ji}^B(\theta_n)|$.¹⁷ The inequality [Eq. (64)] holds for any pair of indices (j, i) , in particular for the indices r, s . Equation (64) implies that

$$|\sin \alpha_{rs}^A(\theta_n) - \sin \alpha_{rs}^B(\theta_n)| \leq 2R(1 + \frac{R}{\sqrt{1-R^2}}) |\sin \alpha_{rs}^A(\theta_n) - \sin \alpha_{rs}^B(\theta_n)| \quad (66)$$

Because of the condition on R [Eq. (33)], Eq. (66) involves a contradiction

unless $|\sin \alpha_{rs}^A(\theta_n) - \sin \alpha_{rs}^B(\theta_n)| = 0$. From the definition of θ_n and of (r,s) in Eq. (65), this result requires that

$$\sin \alpha_{ji}^A(\theta) = \sin \alpha_{ji}^B(\theta) \quad (\text{all } j,i \text{ and } \theta); \quad (67)$$

that is, the uniqueness requirement (part (b) of Theorem 2) is satisfied.

c) The proof of the convergence of the iterative scheme [Eq. (34)] is similar to that given for part (c) of Theorem 1, so that we give only an outline here. From the condition Eqs. (32) and (33), and the definitions $\sin \alpha_{jj}^{(0)} = 0$, $\cos \alpha_{ij}^{(N)} = \pm \sqrt{1 - \sin^2 \alpha_{ij}^{(N)}}$, it follows that the value of $\sin \alpha_{ji}^{N+1}$ obtained from Eq. (34) must obey the relation $0 < \sin \alpha_{ij}^{N+1} < 1$ (all N); thus, the iterative procedure is well defined for an arbitrary number of steps.

To demonstrate the convergence of the iteration procedure, we subtract Eq. (34) for $\sin \alpha_{ji}^N$ from that for $\sin \alpha_{ji}^{N+1}$, perform the appropriate algebraic and trigonometric manipulations, and obtain

$$|\sin \alpha_{ji}^{N+1}(\theta) - \sin \alpha_{ji}^N(\theta)| \leq 2R(1 + \frac{R}{\sqrt{1-R^2}}) \text{Max}_{g,l} |\sin \alpha_{gl}^N(\theta_n) - \sin \alpha_{gl}^{N-1}(\theta_n)| \quad (68)$$

valid for all θ and for all j,i . Proceeding now as we did after deriving Eq. (51) in part (c) of Theorem 1, we can use Eq. (68) to show that the terms of the series,

$$\lim_{N \rightarrow \infty} \sin \alpha_{ji}^N(\theta) = \sum_{N=0}^{\infty} \{\sin \alpha_{ji}^{N+1}(\theta) - \sin \alpha_{ji}^N(\theta)\} \quad (69)$$

are smaller in magnitude than the corresponding terms of the geometric series:

$$R[1+2R(1+\frac{R}{\sqrt{1-R^2}}) + \dots + 2^N R^N (1+\frac{R}{\sqrt{1-R^2}})^N + \dots] \quad (70)$$

Since the series [Eq. (70)] converges under condition Eq. (33), so does Eq. (69). It is straightforward to verify that the convergence of Eq. (69) is to the true solution $\sin \alpha_{ji}$ of Eq. (24). Since the quantity R is independent of θ , the convergence is uniform in θ (i.e., in the directions $\underline{k}', \underline{k}$). This concludes the proof of Theorem 2.

III(iii) Comments on Theorems 1 and 2

Some comments should be made on the results of Theorems 1 and 2, which are relevant to their application to the phase problem. Solution of the unitarity equation leads to a determination of the $\sin \alpha_{ji}(\underline{k}', \underline{k})$ rather than of the phases α_{ji} . Thus, it is not possible to distinguish between the phases α_{ji} and $(\alpha_{ji} + 2n\pi)$ where n is an integer. This ambiguity is unimportant, since the values of the scattering amplitude tensor elements A_{ji}^L or A_{ji}^C are the same for the phases $\alpha_{ji} + 2n\pi$ as for the phase α_{ji} .

Another ambiguity arises because using either of the values $+\sqrt{1-\sin^2 \alpha_{ji}(\underline{k}', \underline{k})}$ or $-\sqrt{1-\sin^2 \alpha_{ji}(\underline{k}', \underline{k})}$ for $\cos \alpha_{ji}(\underline{k}', \underline{k})$ on the right-hand side of Eq. (24), leaves the expression for $\sin \alpha_{ji}$ unchanged. In the iterative procedures [Eqs. (28) and (34)] we defined $\cos \alpha_{ji}^N(\underline{k}', \underline{k}) = +\sqrt{1-\sin^2 \alpha_{ji}^N}$ and obtained convergence to $\sin \alpha_{ji}$ as $N \rightarrow \infty$. We would have obtained the same result if we had chosen $\cos \alpha_{ji}^N = -\sqrt{1-\sin^2 \alpha_{ji}^N}$. It should be noted, however, that the sign of $\cos \alpha_{ji}(\underline{k}', \underline{k})$ is the same for all directions of $\underline{k}', \underline{k}$. Otherwise, we could find $\underline{k}', \underline{k}$ such that $\cos \alpha_{ji}(\underline{k}'', \underline{k}) \cos \alpha_{ji}(\underline{k}'', \underline{k}) < 0$, and it is not difficult to show that if this

were the case the iterative procedure [Eq. (34) or (28)], which satisfies $\cos\alpha_{ji}^N(\underline{k}'',\underline{k})\cos\alpha_{ji}^N(\underline{k}'',\underline{k}')>0$ (for all $\underline{k}',\underline{k}$ and N), could not converge uniformly to $\sin\alpha_{ji}$, as we have demonstrated it does. This ambiguity in the sign of $\cos\alpha_{ji}$, thus also in the sign of $\text{Re}A_{ji}$, is not very important, however. In the first place this amounts only to a two-fold ambiguity in the phase of A_{ji} , hence even in the absence of any additional information on the scatterer, the unitarity relation reduces the phase problem to a choice between two possibilities for each phase. In the second place, the type of ambiguity which arises is such that it should not be difficult to resolve it in practice by some additional information. Theoretically, it is possible to get strict uniqueness, provided the scattering results are known for all frequencies ω . The forward scattering amplitude satisfies the dispersion relation

$$\text{Re}A^L(\omega,0) = \frac{2}{\pi} P \int_0^\infty \frac{d\omega' \omega'}{(\omega')^2 - \omega^2} \text{Im}A^L(\omega,0) \quad (71)$$

where $A^L(\omega,0) \equiv A^L(\underline{k},\underline{k})$, $\omega=kc$. The sign of $\text{Re}A^L(\underline{k},\underline{k})$ can thus be readily determined. If the conditions under which $\cos\alpha_{ji}(\underline{k}',\underline{k})$ does not change its sign as $\underline{k}'$ varies over all directions are not satisfied, we still do not know the sign of $\text{Re}A^L(\underline{k}',\underline{k})$ for $\underline{k}' \neq \underline{k}$. It might be possible to obtain it from the requirement that $\text{Re}A^L(\underline{k}',\underline{k})$ be a continuous function in the direction of $\underline{k}'$, and thus join smoothly the value of $\text{Re}A^L(\underline{k}',\underline{k})$ as $\underline{k}' \rightarrow \underline{k}$.

IV. APPROXIMATIONS TO THE SOLUTIONS OF THE UNITARITY EQUATION

In the present section we describe briefly two approximation methods for treating the unitarity equation. Our aim is to illustrate the nature of the equations and to indicate possible simplifications that may be of use for practical phase determination problems. Since the methods introduced here are analogous to those discussed previously at some length in relation to quantum-mechanical scattering theory,¹² we do not give complete proofs of the results in this section; the interested reader is urged to refer to ref. 12 for details.

VI(i). Bounds on the Iteration Errors

In Sec. III, we developed an iteration scheme for one possible approximation systematically solving the unitarity equation. Since only a finite number of iterations can be performed in practice, it is important to have an estimate of the error at each stage of the iteration process. Such error estimates can be obtained for the conditions corresponding to Theorems 1 and 2.

Under the conditions of Theorem 1, the error associated with the iterant $\sin \alpha_{jj}^N(\underline{k}', \underline{k})$ of Eq. (28) is bounded by

$$|\sin \alpha_{jj}(\underline{k}', \underline{k}) - \sin \alpha_{jj}^N(\underline{k}', \underline{k})| \leq P_j \frac{J_j^N}{\sqrt{1-J_j}} \quad (72)$$

where R_j is defined by Eq. (26), and J_j is

$$J_j = 2R_j \left(1 + \frac{R_j}{\sqrt{1-R_j^2}} \right) \quad (73)$$

This result follows from Eqs. (53) and (54); that is,

$$\begin{aligned}
 |\sin \alpha_{jj} - \sin \alpha_{jj}^N| &= \sum_{M=0}^{\infty} \{ \sin \alpha_{jj}^{M+1} - \sin \alpha_{jj}^M \} - \sin \alpha_{jj}^N \\
 &\leq \sum_{M=N}^{\infty} |\sin \alpha_{jj}^{M+1} - \sin \alpha_{jj}^M| \leq \sum_{M=N}^{\infty} P_j \left[2P_j \left(1 + \frac{P_j}{\sqrt{1-P_j^2}} \right) \right]^M \\
 &= P_j \frac{J_j^N}{1-J_j}
 \end{aligned} \tag{74}$$

Correspondingly, under the conditions of Theorem 2, the error in the iterant $\sin \alpha_{ji}^N$ of Eq. (34) is bounded by

$$|\sin \alpha_{ji}(\underline{k}', \underline{k}) - \sin \alpha_{ji}^N(\underline{k}', \underline{k})| \leq \frac{PJ^N}{1-J} \tag{75}$$

where R obeys inequality [Eq. (32)], and J is defined by

$$J = 2R \left(1 + \frac{R}{\sqrt{1-R^2}} \right) \tag{76}$$

Since the proof of this result is very similar to that used for Eq. (72), we do not give it here.

IV(ii). Upper and Lower Bounds on $\sin \alpha_{ji}(\underline{k}', \underline{k})$

It is advantageous to have not only an estimate on the magnitude of the error corresponding to a given approximant, but also to know the sign of the error. This can be obtained from approximants that are either upper or lower bounds. We now introduce such approximants to the solutions of the unitarity equation.

We consider first a problem with the conditions of Theorem 1, but we suppose in addition that P_j of Eq. (26) satisfies the condition

$$2P_j \left(1 + \frac{1}{\sqrt{1-4P_j^2}}\right) < 1 \quad (77)$$

which is somewhat stronger than Eq. (27). The approximants defined by

$$\sin \alpha_{jj}^{N+1}(\theta) = \int d\Omega L_{jj}^j \cos \alpha_{jj}^N(\theta') \cos \alpha_{jj}^N(\theta'') + \int d\Omega L_{jj}^j \sin \alpha_{jj}^{N-1}(\theta'') \sin \alpha_{jj}^{N-1}(\theta') \quad (78)$$

with $\sin \alpha_{jj}^{(0)} \equiv 0$, $\sin \alpha_{jj}^{(1)}(\theta) \equiv \int L_{jj}^j d\Omega$, $\cos \alpha_{jj}^{(N)} \equiv +\sqrt{1 - \sin^2 \alpha_{jj}^{(N-1)}}$, form upper and lower bounds on $\sin \alpha_{jj}(\theta)$ for even and odd values of N , respectively; that is,

$$\sin \alpha_{jj}^{2M+1}(\theta) \geq \sin \alpha_{jj}(\theta) \geq \sin \alpha_{jj}^{2M}(\theta), \quad M = 0, 1, 2, \dots \quad (79)$$

Moreover, it can be shown that the approximants of odd and of even orders converge respectively from above and from below to the true solution $\sin \alpha_{jj}$.

Since the proof of Eq. (79) is exactly the same as that of the corresponding theorem in the quantum-mechanical case,¹² we mention here only the main steps involved. To establish Eq. (79) by induction, we show that it holds for $M=0$. From the definition of $\sin \alpha_{jj}^{(0)}$, $\sin \alpha_{jj}^{(1)}$, and the fact that $0 \leq \sin \alpha_j(\theta) \leq \int d\Omega L_{jj}^j$ demonstrated in Sec. III, we have

$$\sin \alpha_{jj}^{(0)} \leq \sin \alpha_{jj}(\theta) \leq \sin \alpha_{jj}^{(1)}$$

To verify that from the assumption that Eq. (79) is valid for $M=K$, it follows that Eq. (79) is true for $M=K+1$, and we could use Eqs. (77) and (78).

To prove that the approximants Eq. (78) converge to the correct solution of Eq. (24), we subtract the expression for the (N+1)-th approximant from that of the n-th, and show that the recursion relation

$$|\sin \alpha_{jj}^{N+1}(\theta) - \sin \alpha_{jj}^N(\theta)| \leq 2S_j [|\sin \alpha_{jj}^N(\theta_n) - \sin \alpha_{jj}^{N-1}(\theta_n)| + |\sin \alpha_{jj}^{N-1}(\theta_n) - \sin \alpha_{jj}^{N-2}(\theta_n)|] \quad (80)$$

is obtained, where θ_n is defined as in Sec. III and

$$S_j \equiv P_j \left[1 + \frac{P_j}{\sqrt{1-P_j^2}} \right]. \quad (81)$$

From Eqs. (80) and (77) and from the definitions of $\sin \alpha_{jj}^{(0)}$ and $\sin \alpha_{jj}^{(1)}$, it follows that

$$|\sin \alpha_{jj}^N(\theta) - \sin \alpha_{jj}^{N-1}(\theta)| \leq D_n \equiv a_N S_j^N + b_N S_j^{N-1} + c_N S_j^{N-2}, \quad (82)$$

where $a_0=0$, $a_N=2^{N-1}$ for $N \geq 1$, $b_0=b_1=0$, $b_N=(N-2)2^{N-2}$ for $N \geq 2$,

$$c_1=c_2=c_3=0, \quad c_N=(N^2-7N+12)2^{N-2}, \quad N \geq 4.$$

Applying well-known results from the theory of series to $\sum_{N=0}^{\infty} D_{N+1}$, it is possible to show that the latter converges.¹² Therefore, also the series $\lim_{N \rightarrow \infty} \sin \alpha_{jj}^N = \sum_{M=0}^{\infty} \{\sin \alpha_{jj}^{M+1} - \sin \alpha_{jj}^M\}$ must converge, and it is not difficult to verify that the value to which it converges is indeed a solution of Eq. (24).

If we suppose that the conditions of Theorem 2 are satisfied and that the quantity P of Eq. (33) obeys the inequality

$$2P(1 + \frac{2P}{\sqrt{1-4P^2}}) < 1 \quad (83)$$

The sequence of approximants defined by

$$\begin{aligned} \sin \alpha_{ji}^{N+1}(\underline{k}', \underline{k}) &= \sum_{\ell} f d \Omega_{ji}^{\ell} [\cos \alpha_{\ell j}^N(\underline{k}'', \underline{k}') \cos \alpha_{\ell i}^N(\underline{k}'', \underline{k}) \\ &+ \sin \alpha_{\ell j}^{N-1}(\underline{k}'', \underline{k}') \sin \alpha_{\ell i}^{N-1}(\underline{k}'', \underline{k})] \end{aligned} \quad (84)$$

with $\sin \alpha_{ji}^{(0)} \equiv 0$, $\sin \alpha_{ji}^{(1)} \equiv \sum_{\ell} f d \Omega_{ji}^{\ell}$, and $\cos \alpha_{ji}^N = \sqrt{1 - \sin^2 \alpha_{ji}^N}$ ($i, j=1, 2$), provide alternating upper and lower bounds on the true solutions of Eq. (24).

$$\sin \alpha_{ji}^{2N+1} \geq \sin \alpha_{ji} \geq \sin \alpha_{ji}^{2N} \quad (i, j=1, 2) \quad (85)$$

Moreover, the sequence of upper and lower bounds converge respectively from above and from below to the true solution; that is, $\lim_{N \rightarrow \infty} \sin \alpha_{ji}^N = \sin \alpha_{ji}$. The proof of this result is essentially that outlined for the previous theorem.

IV(111) The Small Phase Approximation

When the phases $\alpha_{j1}(\underline{k}', \underline{k})$ are small, say $|\alpha_{j1}(\underline{k}', \underline{k})| < \pi/6$, the unitarity equation can be simplified by making the obvious approximation $\sin \alpha_{j1} \approx \alpha_{j1}$, $\cos \alpha_{j1} \approx 1 - 1/2(\alpha_{j1})^2$. We refer to these approximations as the "Small Phase Approximation" (SPA). Because the small phase approximation has an analogue in the treatment of the quantum-mechanical unitarity equation, the results stated below are adapted from those given previously.¹²

If the conditions of Theorem 1 are satisfied, the approximate form of the unitarity equation is

$$\alpha_{jj}(\theta) = \int d\Omega L_{jj}^j [1 - 1/2\{\alpha_{jj}(\theta') - \alpha_{jj}(\theta'')\}^2] \quad (j=1,2) \quad (86)$$

Provided there exists a constant M such that

$$1/2 > M \geq \int L_{jj}^j d\Omega, \quad (87)$$

the solution of Eq. (86) can be shown to be unique; the condition given in Eq. (87) is a reasonable one for the present case since $0 \leq \alpha_{jj}(\theta) < 1$ for applicability of the SPA. Moreover, the solution of Eq. (86) can be obtained as the limit of the iteration sequence

$$\alpha_{jj}^{(N+1)}(\theta) = \int d\Omega L_{jj}^j [1 - 1/2\{\alpha_{jj}^{(N)}(\theta'') - \alpha_{jj}^{(N)}(\theta')\}^2] \quad (88)$$

with $\alpha_{jj}^{(0)} \equiv 0$.

The main value of the SPA is that it simplifies an analysis of the dependence of the approximants $\alpha_{jj}^{(N)}(\theta)$ on the cross section and polarization data. In fact, $\alpha_{jj}^{(N)}$ has the form of a convolution polynomial in the

quantity $d_{jj}(\theta)$ of Eq. (39). One can easily introduce upper and lower bounds to the SPA solution.¹² Similar results can also be established under the conditions of Theorem 2.

V. DETERMINATION OF PHASES IN THE SCATTERING OF PHOTONS

Although in the present article we are concerned primarily with the determination of phases in classical electromagnetic scattering, the unitarity equation method can be extended to the corresponding phase problem in quantum-electrodynamical photon scattering.

The state of the system prior to the collision (photon plus atom or molecule) is denoted by $|i\rangle = |\phi_i, \underline{k}_i, \underline{\epsilon}_i\rangle$, where ϕ_i stands for all the dynamical variables necessary to specify the state of the atom, $\underline{k}_i$ is the initial wavenumber for the photon and $\underline{\epsilon}_i$ is its polarization. If the collision does not lead to a change in the number of any of the particles involved (i.e., the photon is not absorbed, a second photon is not omitted, and electron-positron pair production does not occur), the final state can be written $|f\rangle = |\phi_f, \underline{k}_f, \underline{\epsilon}_f\rangle$. For final states in which the photon is absorbed we have simply $|f\rangle = |\phi_f\rangle$, while if photons are emitted so that there are n photons present in the final state, is $|f\rangle = |\phi_f, \underline{k}_f^{(1)}, \underline{\epsilon}_f^{(1)}; \dots; \underline{k}_f^{(n)}, \underline{\epsilon}_f^{(n)}\rangle$. If $\mathcal{H}$ be the Hamiltonian of the total system and $\mathcal{H}'$ is the interaction between the atom and the radiation field, the transition operator T is given by¹⁹

$$T = \mathcal{H}' + \mathcal{H}' \frac{1}{E - \mathcal{H} + i\epsilon} \mathcal{H}' \quad (89)$$

where E is the total energy of the system. The transition amplitude

$T_{i \rightarrow f}$ defined in terms of the matrix element of T between an initial and a final state is

$$\langle f | T | i \rangle = \delta(\underline{P}_i + \underline{k}_i - \sum_{j=1}^n \underline{k}_f^{(j)} - \underline{P}_f) T_{i \rightarrow f}, \quad (90)$$

where n is the number of photons in the final state, and $\underline{P}_i, \underline{P}_f$ denote respectively the initial and the final momenta of the atom. The δ -function in Eq. (90) arises from the conservation of total momentum in the collision. Since the measurable quantities in photon scattering experiments are the differential cross sections $\frac{d\sigma_{i \rightarrow f}}{d\Omega}$, which are proportional to $|T_{i \rightarrow f}|^2$,¹⁹ the absolute value of the transition amplitude $|T_{i \rightarrow f}|$ can be determined. As in the case of the classical scattering amplitude, it is desirable to know the phase of $T_{i \rightarrow f}$ as well, since this quantity is necessary for a complete determination of the interaction between the photon and the target atom.

The quantities $T_{i \rightarrow f}$ satisfy the quantum-electrodynamical unitarity relation²⁰

$$i(T_{f \rightarrow i} - T_{i \rightarrow f}^*) = \sum_n T_{n \rightarrow f}^* T_{n \rightarrow i} \quad (\text{all } i, f) \quad (91)$$

where the various $T_{n \rightarrow j}$ in Eq. (91) involve states $|n\rangle$ and $|j\rangle$ on the same energy shell. In the above relation, all the processes that are energetically allowed contribute; this includes absorption of the incident photon and, when possible, emission of a second photon and pair production. To simplify the discussion, we confine ourselves to cases in which the number of each type of particle is conserved. Each of the contributing $T_{n \rightarrow j}$ can be written in the form

$$T_{n \rightarrow j} \equiv k_n A_{nj}(k_j, \epsilon_j, k_i, \epsilon_i) \equiv -k_n d_{nj} e^{i\alpha_{nj}} \frac{1}{4\pi^2} \quad (92)$$

where $k_n = |k_n|$, and the subscripts nj of A_{nj} , d_{nj} , α_{nj} , refer to the initial and final states of the atomic system. Equation (92) defines the phases α_{nj} of T_{nj} . Substituting Eq. (92) in Eq. (91) we find that

$$d_{fi}(\theta) \sin \alpha_{fi}(\theta) = \frac{k_i}{4\pi} \sum_n d_{nf}(\theta'') d_{ni}(\theta') \cos[\alpha_{nf}(\theta'') - \alpha_{ni}(\theta')] \quad (93)$$

where $\theta \equiv (k_f, \epsilon_f; k_i, \epsilon_i)$; $\theta' \equiv (k_n, \epsilon_n; k_i, \epsilon_i)$; $\theta'' \equiv (k_n, \epsilon_n; k_f, \epsilon_f)$. The sum over n in Eq. (93), as in Eq. (91), is over all the internal states of the atomic system which are energetically accessible, over all possible directions of k_n , and over the two possible polarizations ϵ_n .

Equation (93) constitutes a set of coupled equations for the phases, since the d_{fi} can be determined from the experimental cross sections (which pertain to all possible initial and final polarization states). The number of coupled equations is $4N$, where N denotes the number of energetically accessible internal states of the atomic species. If the energy of the photon is smaller than the difference between the energy of the first excited atomic or molecular state and the ground state, the scattering is elastic and there are just four coupled equations for the unknowns $\alpha(k', \epsilon_j, k, \epsilon_i)$ with $i, j=1, 2$. This corresponds to the classical case we dealt with in the previous sections. No polarization data are needed here, since the cross sections themselves refer to transitions between definite initial and final polarization states.

Equation (93) can be solved by the methods of Secs. III and IV. In particular, when there exists a constant P such that

$$P < \frac{k_i}{4\pi} \sum_n \frac{d_{nf}(\theta'') d_{ni}(\theta')}{d_{fi}}$$

for all f, i and

$$2P(1 + \frac{P}{\sqrt{1-P^2}}) < 1$$

the solution of Eq. (93) is unique and can be obtained by iteration.

If two-photon final states were allowed, Eq. (93) would become a continuum of equations corresponding to all possible relative momenta of the two photons.

VI. APPLICATIONS AND TESTS OF THE UNITARITY METHOD

VI(i) The Optical Theorem.

In this section we consider some illustrative applications of the unitarity method. We first derive the optical theorem, which is a special case of the unitarity relation that can frequently be used to find the first estimate of the phase. Then we treat scattering by a free electron and by a bound electron. Although these two problems represent elementary applications, they are very important since their solution can serve as the basis for utilizing the unitarity method in x-ray diffraction. Finally, we consider the scattering by a sphere of constant dielectric material, which is of interest for studies of large systems by light scattering and related techniques.

To derive the optical theorem,²¹ we consider the unitarity relation in the form given in Eq. (22). The value of the phase α_{jj} in the forward

direction is obtained by setting $\underline{k}' = \underline{k}$, so that Eq. (22) reduces to

$$d_{jj}(\underline{k}, \underline{k}) \sin \alpha_{jj}(\underline{k}, \underline{k}) = \frac{k}{4\pi} \sum_{\ell=1}^2 \int d\Omega [d_{\ell j}(\underline{k}'', \underline{k})]^2 \quad (94)$$

From Eq. (61) for right-handed circularly polarized incident radiation we find

$$\sum_{\ell=1}^2 \int d\Omega [d_{\ell j}(\underline{k}'', \underline{k})]^2 = \int \frac{d\sigma}{d\Omega} d\Omega \equiv \sigma_j \quad (95)$$

where the quantity σ_j is by definition the total scattering cross section; the subscript j of σ_j denotes the direction in which the incident wave was polarized. It is very important to note that since Eq. (61) for $\frac{d\sigma}{d\Omega}$ applies only to cases where the incident wave is fully polarized, Eq. (95) is restricted to the scattering of a polarized beam; either a linearly or a circularly polarized incident beam is appropriate.

Combining Eqs. (94) and (95) we obtain the optical theorem for the scattering of a beam with polarization j ; that is,

$$d_{jj}(\underline{k}, \underline{k}) \sin \alpha_{jj}(\underline{k}, \underline{k}) = \frac{k}{4\pi} \sigma_j \quad (96)$$

A related result can be derived for the scattering of an unpolarized beam by an object with spherical symmetry. It was pointed out in Sec. III that for such a scatterer, the scattering amplitude tensor in the linear polarization representation is diagonal. It is possible also to show that in the forward direction, $A_{11}^L(\underline{k}, \underline{k}) = A_{22}^L(\underline{k}, \underline{k})$.¹⁶ Denoting by $d(\underline{k}, \underline{k})$ and $\sigma(\underline{k}, \underline{k})$, respectively, the magnitude and the phase of these quantities, we can prove the optical theorem for the scattering of an unpolarized

wave by a scatterer with complete rotation-reflection symmetry;²¹ that is,

$$d(\underline{k}, \underline{k}) \sin \alpha(\underline{k}, \underline{k}) = \frac{k}{4\pi} \sigma, \quad (97)$$

where $\sigma \equiv \int \frac{d\sigma}{d\Omega} d\Omega$ is the total scattering cross section.

In Sec. III it was stressed that the unitarity relation is subject to the assumption that there is no absorption or emission of energy by the scatterer. The optical theorems, which we have obtained as a special case of the unitarity relation, admit generalization to processes involving absorption as well as scattering. For an absorbing scatterer, the generalization of Eq. (96) has the form²¹

$$d_{jj}(\underline{k}, \underline{k}) \sin \alpha_{jj}(\underline{k}, \underline{k}) = \frac{k}{4\pi} \sigma_{\text{total}}^{(j)}, \quad \sigma_{\text{total}}^{(j)} \equiv \sigma_j + \sigma_j^{(a)} \quad (98)$$

This equation enables one, in principle, to correct results obtained by the unitarity equation method for an absorbing scatterer, at least in the forward direction and small-angle region.

VI(ii) Scattering by a Free Electron

We now apply the unitarity equation to the scattering of an electromagnetic wave by a free electron. This is an elementary problem, but it is not without interest because the scattering of high-frequency waves by molecular species and by macroscopic matter shows many features characteristic of scattering by free electrons. In particular, the polarization factor used for x-ray diffraction by crystals is exactly the same as the corresponding quantity for a classical free electron and, in many cases, entire crystals are treated as if they were composed of a set of coherently scattering free electrons.^{1,9}

The approximate result quoted in many texts^{1,9} for the Thomson scattering amplitude of a free electron does not satisfy the unitarity relation. Since the scattered field amplitude $\underline{E}_s$ (defined in Sec. II) is taken to be real valued for a real incident wave polarized along the $\underline{e}_\perp^0$ direction or the $\underline{e}_\parallel^0$ direction,^{1,9} the scattering amplitude tensor in the linear polarization representation, $\tilde{A}^L$, is real and $\sin\alpha_{ji}$ (all j,i) is zero. Thus, the general unitarity theorem and its special case, the optical theorem, are not satisfied by the approximate Thomson amplitude; that is, from the optical theorem Eq. (96) because the right-hand side of Eq. (96) is positive definite, $\sin\alpha_{jj}(\underline{k},\underline{k})$ must be positive definite.

To determine the corrected free electron scattering amplitude so that it does satisfy the unitary relation, we examine the method commonly used to derive the Thomson formula. In the simplest approach, one considers a real-valued x-polarized incident electric field, $\underline{E}_0$, which exerts a force of $\underline{E}_0 e$ on the electron. A knowledge of the resulting acceleration permits one to determine the emitted radiation that constitutes the scattered wave with a real field amplitude.

The violation of unitarity manifested by the above approximation can be removed by including in the calculation the effect of the reaction of the emitted radiation on the accelerated charge. The force on a charge e due to the reaction of the emitted radiation is $\frac{2}{3} \frac{e^2}{c} \ddot{\underline{x}}$ where $\ddot{\underline{x}} = \frac{d^3 \underline{x}}{dt^3}$.²³ Consequently, Newton's law of motion for the electron accelerated by the field $\underline{E}_0$, and experiencing the reaction of the radiated field, has the form²⁴

$$m \ddot{\underline{x}} = e \underline{E}_0(t) + \frac{2}{3} \frac{e^2}{c} \ddot{\underline{x}} \quad (99)$$

In this dynamical equation the explicit time dependence of the electric field is taken into account, in contrast to Sec. II where we gave a time-independent formulation of the scattering theory of waves. For the stationary fields with which we are concerned, the time dependence has the simple form

$$\underline{E}_0(t) = \underline{E}_0 e^{-i\omega t} \quad (100)$$

where ω is the frequency of the incoming radiation, and $\underline{E}_0$ is time independent; thus, the fields appearing in Sec. II are the time-independent factor of $\underline{E}(t)$. The solution of Eq. (99) is

$$\underline{x} = -\frac{e}{m\omega} \frac{1}{1+iq} \underline{E}_0 \cdot e^{-i\omega t}; \quad q = \frac{2}{3} \frac{e^2}{mc} \omega \quad (101)$$

The field of the emitted radiation, $\underline{E}_f(t)$, is given by^{9,23}

$$\begin{aligned} \underline{E}_f(t) &\equiv \frac{\underline{E}_s}{r} e^{i(kr-\omega t)} = \frac{e^2 \underline{x}}{rc^3} \sin\phi e^{i(kr-\omega t)} \\ &= -\frac{e^2 \sin\phi}{m(1+iq)rc^2} e^{i(kr-\omega t)} \underline{E}_0; \quad \underline{E}_s = -\frac{e^2 \sin\phi}{m(1+iq)c^2} \underline{E}_0 \end{aligned} \quad (102)$$

where r is the distance from the electron to the point at which the field is observed and ϕ is the angle between the direction of $\underline{r}$ and that of $\underline{E}_0$. Equation (102) enables us to calculate the scattering amplitude tensor $\underline{A}^L$, which is diagonal, since the electron is a spherically symmetric scatterer; that is,

$$\underline{A}^L = \begin{pmatrix} A_{11}^L & 0 \\ 0 & A_{22}^L \end{pmatrix}$$

If for a given scattering direction, the electric field $\underline{E}_0$ is polarized along $\underline{e}_{||}^0$ (i.e., in a direction parallel to the scattering plane) the angle $\phi = -\pi/2 - \theta$,⁹ where θ is the scattering angle, the angle between $\underline{k}'$ and $\underline{k}$. From Eq. (102) we have for this case

$$\underline{E}_s(\theta) = - \frac{e^2 \cos \theta}{m(1+iq)c^2} \underline{E}_0$$

Writing $\underline{E}_s$ and $\underline{E}_0$ in the two component notation of Sec. II, and using the fact that $\underline{A}^L$ is diagonal, we find with Eq. (5) that

$$A_{11}^L(\theta) = - \frac{e^2 \cos \theta}{m(1+iq)c^2} \quad (103)$$

If the incident field $\underline{E}_0$ is polarized along $\underline{e}_{\perp}^0$ (i.e., in a direction perpendicular to the scattering plane), we have $\sin \phi = 1$ and from Eq. (102)

$$\underline{E}_s(\theta) = - \frac{e^2}{m(1+iq)c^2} \cdot \underline{E}_0$$

Using this relation and the diagonal form of $\underline{A}^L$ in Eq. (5), we obtain

$$A_{22}^L(\theta) = - \frac{e^2}{m(1+iq)c^2} \quad (104)$$

For $\theta=0$, we have from Eqs. (103) and (104)

$$A_{11}(0) = A_{22}(0)$$

a result introduced previously for a scatterer having full rotational-reflectional symmetry (see Sec. V(i)).

Within the framework of the dynamical model employed here [Eqs. (99)-(104)], the cross section for scattering of an unpolarized wave by a free electron is given by²⁴

$$\frac{d\sigma}{d\Omega} = \frac{1}{2} \frac{e^4 (1 + \cos^2 \theta)}{m^2 c^4 (1 + q^2)}, \quad \sigma = \frac{8\pi}{3} \frac{e^2}{mc^2} \frac{1}{1 + q^2} \quad (105)$$

From Eq. (104) or Eq. (103) we find

$$d(0) \sin \alpha(0) = \text{Im} A(0) = \frac{e^2 q}{m(1 + q^2) c^2} = \frac{2}{3} \frac{e^4 \omega}{m^2 (1 + q^2) c^5} \quad (106)$$

Comparing Eqs. (105) and (106) and recalling that $k = \frac{\omega}{c}$, we find that the optical theorem [Eq. (97)] is satisfied exactly by the above model for the Thomson scattering amplitude.

It should be noted that the quantity q is often very small (e.g., it is $\sim 10^{-4}$ for a frequency ω in x-ray range). Thus, $\text{Im} A(0)$ is correspondingly small, as is evident from Eq. (106), and accurate cross sections can be calculated without taking the radiation reaction into account. Thus, the violation of the optical theorem by the standard Thomson formulation is "qualitative" error whose quantitative consequences may sometimes be unimportant.

To examine whether the general unitarity equation [Eq. (42)] is obeyed by the above model, we use the cross section [Eq. (105)] and the degree of polarization⁹ P_s ,

$$P_s(\theta) = \frac{\sin^2 \theta}{1 + \cos^2 \theta} \quad (107)$$

to calculate the phases of the scattering amplitude for comparison with known results. By substituting from Eqs. (105) and (107) into Eq. (25), we find that the kernel L_{22}^2 has the form

$$L_{22}^2 = \frac{1}{\sqrt{2}} \frac{e^2}{mc^2 \sqrt{1+q^2}} \frac{\cos\theta'' \cos\theta'}{\cos\theta} \quad (108)$$

We choose the vector $\underline{k}$ to lie along the z axis of a Cartesian coordinate system, and let ψ, ψ' denote the aximuthal angles of the vectors $\underline{k}', \underline{k}''$, respectively, in that system. The angles θ'', θ' and θ are then related by $\cos\theta'' = \cos\theta' \cos\theta + \sin\theta' \sin\theta \cos(\psi - \psi')$ and the volume element which appears in the unitarity relation $d\Omega_{\underline{k}''} \equiv d\Omega = \sin\theta' d\theta' d\psi'$. Since the optical theorem suggests that for scattering by a free electron the imaginary part of the scattering amplitude is extremely small compared with the real part, the phases of the scattering amplitude should be very small. Consequently, the lowest order iterant in Eq. (28) should give a satisfactory solution of Eq. (42); alternatively, the Small Phase Approximation could have been employed. From Eq. (28) for $N=0$ and Eq. (108), we obtain

$$\begin{aligned} \sin\alpha_{22}(\theta) &= \frac{e^2 k}{mc^2 \sqrt{1+q^2}} \frac{1}{4\pi} \frac{1}{\cos\theta} \int_0^{2\pi} \int_0^\pi \cos\theta'' \cos\theta' d(\cos\theta') d\psi' \\ &= \frac{2e^2 k}{3mc^2 \sqrt{1+q^2}} = \frac{2e^2 \omega}{3mc^3 \sqrt{1+q^2}} \end{aligned} \quad (109)$$

Comparison of this value for the phase with the result obtained from Eq. (104) for the tensor element A_{22} shows that the two are identical.

Corresponding agreement is found for $\sin \alpha_{11}$.

That the exact values of the phases are given by the above treatment, may appear suspicious because we did not solve the unitarity equation, but used only the first iterant. However, this is appropriate here since inspection of Eqs. (42) and (28) shows that when the phases are constants, as they are for this problem, the first iterant $\sin \alpha_{jj}^1$ equals the exact solution.

Although the present development was carried out for free electrons, its results are applicable to atomic electrons if their binding energy is very small compared with the frequency of the incoming radiation. A more satisfactory way of dealing with scattering by a bound electron is to introduce the harmonic oscillator model.²⁴ We do this in the following section.

It is important to note also that we have shown only that an approximate dynamical model for the free electron [Eqs. (99)-(109)] is consistent with the unitarity relation. The proper treatment of the classical dynamics of the problem requires inclusion of neglected relativistic corrections; that is, the equation of motion Eq. (99) is not Lorentz-invariant, the action of magnetic forces is considered, and Eq. (102) for the field emitted by the accelerated electron does not include retardation effects.²³ Thus, one must still consider which forces or effects are responsible for the unitarity property in more general electromagnetic scattering problems, and which approximations conserve this property.

Finally, we mention the corresponding quantum-electrodynamical problem of the scattering of photons by electrons. Covariant perturbation

theory in quantum electrodynamics should preserve unitarity to each order in perturbation theory.²⁵ To order of e^2 in perturbation theory²⁵, there is no imaginary part to the scattering amplitude, and the total cross section vanishes,²⁵ so that the optical theorem holds in a trivial sense. The total cross section is of order e^4 , and in this is the order in which there is an imaginary part to the elastic transition amplitude.²⁵ Unfortunately, there are very many possible processes for the photon-electron system, the computational aspects of the problem are involved, and attempts at accurate calculations testing the unitarity relation of Sec. V in a non-trivial way would lead us outside the scope of the paper.

VI(ii) Scattering by an Oscillating Electron

In the classical theory of the interaction of radiation and matter, extensive use was made of the Lorentz model, in which atomic electrons were regarded as bound by harmonic forces. The effects of an encounter between an electromagnetic wave packet and an atom were then calculated from the mutual interactions between the oscillating charges and the radiation. Since many predictions made by the Lorentz theory, including some on the scattering of light by atoms, are found to have the same form as those obtained from the quantum theory, it is of interest to test the unitarity relation for this model.

We consider an electric wave $\underline{E}(t) = \underline{E}_0 e^{-i\omega t}$ which impinges upon an electron performing harmonic oscillations with frequency ω_0 . This mechanical system is a Lorentz-type model of the hydrogen atom. Because the independent-particle assumption is a good approximation for wave scattering by electrons in an atom, the results are applicable also to the scattering by many-electron atoms. By analogy with Eq. (99) for a free

electron, the equation of motion in the present case can be written

$$m \ddot{\underline{x}} + \omega_o^2 \underline{x} - \frac{2e^2}{3c^3} \ddot{\underline{x}} = e \underline{E}_o e^{-i\omega t} \quad (110)$$

where, in addition to the elastic force and the force exerted by the incident wave, the reaction of the emitted radiation is taken into account. It should be noted that although the Lorentz electron is in general a three-dimensional oscillator, only the oscillation $\underline{x}$ along the direction of $\underline{E}_o$ contributes to the scattering, and it is this motion that we consider.²⁶ The solution of Eq. (110) is

$$\underline{x} = \frac{\frac{e}{m} \underline{E}_o}{\omega_o^2 - \omega^2 + i q \omega^2} \quad (111)$$

with q defined as in Eq. (101). With definitions corresponding to those employed in Eq. (102), we find that the scattered field amplitude $\underline{E}_s$ for the present case has the form

$$\underline{E}_s = \frac{e^2 \sin^2 \phi}{mc^2 (\omega_o^2 - \omega^2 + i q \omega^2)} \underline{E}_o \quad (112)$$

Since the scatterer has full rotational-reflectional symmetry, the tensor $\underline{A}^L$ is diagonal. Proceeding as in Sec. VI(ii), we consider $\underline{E}_o$ polarized along $e_{||}^o$ and then along $e_{\perp}^o$; the results for $\underline{A}^L$ are

$$A_{11}^L(\theta) = \frac{e^2 \cos^2 \theta}{mc^2 (\omega_o^2 - \omega^2 + i q \omega^2)} \quad (113)$$

and

$$A_{22}^L(\theta) = \frac{e^2}{mc^2 (\omega_o^2 - \omega^2 - i q \omega^2)} \quad (114)$$

To obtain the differential cross section and the degree of polarization of the scattered radiation, we substitute Eqs. (113) and (114), together with the fact that $P_0=0$, into Eqs. (16) and (17), respectively; we obtain

$$\frac{d\sigma}{d\Omega} = \frac{e^4 \omega^4 (\cos^2 \theta + 1)}{2m^2 c^4 [(\omega_0^2 - \omega^2)^2 + q^2 \omega^4]} \quad (115)$$

$$P_s(\theta) = \frac{\sin^2 \theta}{1 + \cos^2 \theta}; \quad (116)$$

the degree of polarization $P_s(\theta)$ is seen to be the same as in the free electron case, though the differential cross section is not.

To test whether the bound oscillator satisfies the unitarity equation, we substitute Eqs. (115) and (116) into Eq. (25) and find

$$L_{22}^2 = \frac{e^2 \omega^2}{mc^2 \sqrt{(\omega_0^2 - \omega^2)^2 + q^2 \omega^4}} \frac{\cos \theta'' \cos \theta'}{\cos \theta} \quad (117)$$

with the angles defined as before. To solve Eq. (42), we again evaluate only the first iterant of Eq. (28) to obtain

$$\begin{aligned} \sin \alpha_{22}^1(\theta) &= \frac{e^2 \omega^2 k}{mc^2 \sqrt{(\omega_0^2 - \omega^2)^2 + q^2 \omega^4}} \frac{1}{4\pi \cos \theta} \int_0^{2\pi} \int_0^\pi \cos \theta'' \cos \theta' \sin \theta' d\theta' d\phi' \\ &= \frac{2e^2 \omega^3}{3mc^3 \sqrt{(\omega_0^2 - \omega^2)^2 + q^2 \omega^4}} \end{aligned} \quad (118)$$

Since $\sin \alpha_{22}^1(\theta)$ is again independent of θ (as it was for the free electron), the first iterant of Eq. (28) is the true solution of Eq. (42) as can be

verified by direct substitution. Comparing Eq. (118) with the phase obtained for A_{11}^L from Eq. (113), we see that the two are identical; a corresponding result is found for A_{22}^L . Thus, the model corresponding to Eq. (110) for scattering by an oscillating electron obeys the unitarity relation.

The model we have employed in this section has a cross section [Eq. (115)] which differs from that usually given in the literature for scattering by an oscillating charge.²⁴ This cross section, for an unpolarized incident wave, has the form

$$\frac{d\sigma}{d\Omega} = \frac{e^4 \omega^4 (\cos^2 \theta + 1)}{2m^2 c^4 [(\omega_o^2 - \omega^2)^2 + \gamma^2 \omega^2]} , \text{ with } \gamma = \frac{2e^2 \omega_o^2}{3mc^3} \quad (119)$$

in which the second term in the denominator ($\gamma^2 \omega^2$) is different from the corresponding term in the denominator of Eq. (115). From a practical point of view, the difference between the two cross sections is not significant. Since γ and q are small quantities for all systems and conditions of physical interest,²⁷ we have $(\omega_o^2 - \omega^2)^2 \gg \gamma^2 \omega^2$, $q^2 \omega^4$ when ω is appreciably different from ω_o and the two expressions [Eqs. (119) and (115)] become identical; when $\omega \approx \omega_o$, we have $\gamma^2 \omega^2 \approx \gamma^2 \omega_o^2 = q^2 \omega_o^4 \approx q^2 \omega^4$ by the definitions of γ and q , and the cross section of Eq. (119) is again effectively the same as that of Eq. (115). However, because the cross section of Eq. (119) violates unitarity, it is appropriate to comment on the origins of the mathematical difference between the two formulae. The effect of emitted radiation reaction on the origins of the mathematical difference between the two formulae. The effect of emitted radiation

reaction on radiating oscillators is to cause a decay of the oscillations, if additional energy is not supplied from the outside.²⁴ It is advantageous to describe the decay as much as possible in terms of parameters intrinsic to the oscillating system and not involving variables (e.g., frequency) of the external forces. This motivates the standard approach in which one considers an oscillating electron of frequency ω_0 , in the absence of any external radiation. Because of its acceleration, the oscillator radiates, and its oscillations are damped by radiation reaction. For this isolated oscillator,

$$\frac{2}{3} \frac{e^2}{c} \ddot{\underline{x}} = -m\gamma \dot{\underline{x}}$$

where γ is given in Eq. (119);²⁴ thus, γ has the significance of a damping constant.²⁴ One then makes the assumption that the introduction of an external field, due to an incoming electric wave, does not alter the damping force $-\gamma \dot{\underline{x}}$, and only adds a driving force term to the equation of motion, which becomes²⁴

$$\ddot{\underline{x}} + \dot{\underline{x}} + \omega_0^2 \underline{x} = \frac{e}{m} \underline{E}_0 e^{-i\omega t} \quad (120)$$

in contrast to Eq. (110). The differential cross section [Eq. (119)], scattering amplitude tensor, and degree of polarization obtained from Eq. (120) violate the unitarity equation. Clearly, this can never occur for an exact treatment of electromagnetic scattering problems, where all the effects of the radiation (including radiation reaction on the scatterer) are taken into account, since we know that the unitarity theorem is valid. The trouble with the treatment based on Eq. (120) for scattering by an

oscillator, as opposed to that based on Eq. (110), is that the former model does not take proper account of the radiation reaction. As in Sec. VI(ii) we are led to the important conclusion that models which neglect or crudely approximate the contributions of radiation reaction violate unitarity even though they may yield accurate cross sections for the physical processes which they are supposed to describe.

VI(iii) Scattering by a Sphere of Constant Dielectric Material

The examples above are all microscopic systems characterized by an equation of motion which they obey and from which the scattered radiation emitted by the system can be computed. Many light scattering experiments are done on systems so large and complex that a detailed dynamical description of the motions of the particles constituting the scatterer is not possible. Consequently, the discussion of wave scattering by the object as a whole is best done in terms of macroscopic parameters, such as the dielectric coefficient ϵ and the magnetic permeability μ .²⁸ In this section, we apply the unitarity relation to such a macroscopic scatterer.

The problem of scattering by a sphere of constant dielectric material was solved by G. Mie, and modern descriptions of his work are given in standard references.^{4,28} The expression for the scattering cross section of unpolarized incident light by a non-conducting (non-absorbing) spherer is^{4,28,29}

$$\begin{aligned} \frac{d\sigma}{d\Omega} = \frac{1}{8k^2} [& \left| \sum_{J=1}^{\infty} (2J+1) \{ (e^{2i\delta_J^{(1)}} - 1) \tau_J + (e^{2i\delta_J^{(2)}} - 1) \pi_J \} \right|^2 \\ & + \left| \sum_{J=1}^{\infty} (2J+1) (e^{2i\delta_J^{(2)}} - 1) \tau_J + (e^{2i\delta_J^{(1)}} - 1) \pi_J \right|^2] \end{aligned} \quad (124)$$

In the above equation τ_J and π_J are functions of the scattering angle defined by

$$\pi_J(\cos\theta) \equiv \frac{1}{J(J+1)} \frac{dP_J(\cos\theta)}{d(\cos\theta)} \quad (125)$$

$$\tau_J(\cos\theta) \equiv \cos\theta \pi_J - \sin^2\theta \frac{d\pi_J}{d \cos\theta} \quad (126)$$

where P_J is the Legendre polynomial of order J . The quantities $\delta_J^{(1)}$ and $\delta_J^{(2)}$ (denoted, respectively, by α_J and β_J in refs. 4 and 29), are given in the literature explicitly in terms of expressions involving the values of spherical Bessel and Newmann functions at the points $x \equiv kR$ and $\bar{x} \equiv \sqrt{\mu\epsilon} x$, where k is the magnitude of the wave number, R is the radius of the scatterer, μ its permability, and ϵ its dielectric coefficient.⁴

For our purposes it suffices to point out that, for a non-conducting spherer, $\delta_J^{(1)}$ and $\delta_J^{(2)}$ are real valued.

On the basis of the Mie solution it is also possible to show that for an unpolarized incident beam, the polarization of the scattered wave is

$$P_s = \frac{\left| \sum_{J=1}^{\infty} \frac{1}{4k^2} (2J+1) \{ (e^{2i\delta_J^{(1)}} - 1) \tau_J + (e^{2i\delta_J^{(2)}} - 1) \pi_J \} \right|^2}{2 \left(\frac{d\sigma}{d\Omega} \right)}$$

$$- \frac{\left| \frac{1}{4k^2} \sum_J^{\infty} (2J+1) \{ (e^{2i\delta_J^{(2)}} - 1) \tau_J + (e^{2i\delta_J^{(1)}} - 1) \pi_J \} \right|^2}{2 \left(\frac{d\sigma}{d\Omega} \right)} \quad (127)$$

where $\left(\frac{d\sigma}{d\Omega} \right)$ is given by Eq. (124).

We now take the scattering amplitude tensor and verify that it obeys the unitarity relation in the form of Eq. (19). We consider the scattering amplitude tensor³⁰

$$\tilde{A}(\underline{k}', \underline{k}) = \frac{2\pi}{ik} \sum_{JM \ell n} Y_{JM}^{(n)}(\hat{k}') [e^{2i\delta_J^{(n)}} \delta_{\ell n} - \delta_{\ell n}] Y_{JM}^{(\ell)*}(\hat{k}) \quad (128)$$

The tensor is written in diadic notation; $\underline{k}', \underline{k}$ denote unit vectors in the direction of $\underline{k}', \underline{k}$, respectively, $Y_{JM}^{(n)}$ are the transverse vector spherical harmonics, and the indices n , go over the values 1, 2. By substituting Eq. (128) into Eqs. (16) and (17), it can be verified that $A(\underline{k}', \underline{k})$ yields the cross section [Eq. (126)] and the polarization [Eq. (127)]. To show that this tensor also satisfies the unitarity relation [Eq. (19)], we substitute Eq. (128) on the right-hand side of Eq. (19) and obtain the set of relations

$$\frac{k}{4\pi} \sum_{JM\ell n} \sum_{J'M'\ell'n'} \frac{4\pi^2}{k^2} [e^{2i\delta_J^{(n)}} \delta_{n\ell} - \delta_{n\ell}] [e^{-2i\delta_J^{(n')}} \delta_{n'\ell'} - \delta_{n'\ell'}] Y_{JM}^{(n)}(\hat{k}') [f d\Omega Y_{JM}^{(\ell)*}(\hat{k}'') \cdot Y_{J'M'}^{(n')*}(\hat{k}'')] Y_{J'M'}^{(\ell')}(\hat{k})$$

$$= \frac{\pi}{k} \sum_{JM\ell} \sum_{J'M'\ell'} [e^{2i\delta_J^{(\ell)}} - 1] [e^{-2i\delta_J^{(\ell')}} - 1] Y_{JM}^{(\ell)}(\hat{k}') [f d\Omega Y_{JM}^{(\ell)*}(\hat{k}'') \cdot Y_{J'-M'}^{(\ell')}(\hat{k}'')] (-1)^{J'+M'} Y_{J'M'}^{(\ell')}(\hat{k})$$

(129)

$$= \frac{\pi}{k} \sum_{JM\ell} \sum_{J'M'\ell'} [e^{2i\delta_J^{(\ell)}} - 1] [e^{-2i\delta_J^{(\ell')}} - 1] Y_{JM}^{(\ell)}(\hat{k}') \delta_{JJ'} \delta_{-M'M} \delta_{\ell\ell'} Y_{J'-M'}^{(\ell')*}(\hat{k})$$

$$= \frac{\pi}{k} \sum_{JM\ell} [2 - 2\cos 2\delta_J^{(\ell)}] Y_{JM}^{(\ell)}(\hat{k}') Y_{JM}^{(\ell)*}(\hat{k})$$

In deriving the first and second steps of Eq. (129), we employed the conjugation property of the vector harmonics (i.e., $Y_{JM}^{(\ell)*} = (-1)^{J+M} Y_{JM}^{(\ell)}$)³¹; in the second step, we used the orthogonality of the vector harmonics, and in the third step, the fact that summation over $JM\ell$, $J'M'\ell'$ is equivalent to summation over $JM\ell$, $J'-M'\ell'$ because M' goes from $-J'$ to $+J'$. Substituting from Eq. (128) into the left-hand side of Eq. (19), we have

$$\begin{aligned}
 \text{Im}\tilde{A} &= \frac{1}{2i} [\tilde{A} - \tilde{A}^*] = -\frac{\pi}{k} \sum_{JMn} Y_{JM}^{(n)}(\hat{k}') [e^{i2\delta_J^{(n)}} - 1] Y_{JM}^{(n)*}(\hat{k}) \\
 &\quad - \frac{\pi}{k} \sum_{JMn} Y_{JM}^{(n)*}(\hat{k}') [e^{-i2\delta_J^{(n)}} - 1] Y_{JM}^{(n)}(\hat{k}) = \\
 &= -\frac{\pi}{k} \sum_{JMn} Y_{JM}^{(n)}(\hat{k}') [e^{2i\delta_J^{(n)}} - 1] Y_{JM}^{(n)*}(\hat{k}) - \frac{\pi}{k} \sum_{JMn} Y_{J-M}^{(n)}(\hat{k}') [e^{-2i\delta_J^{(n)}} - 1] Y_{J-M}^{(n)*}(\hat{k}) \\
 &= \frac{\pi}{k} \sum_{JMn} Y_{JM}^{(n)}(\hat{k}') [2 - 2\cos 2\delta_J^{(n)}] Y_{JM}^{(n)}(\hat{k})
 \end{aligned} \tag{130}$$

In the last two steps of Eq. (130), the conjugation property of $Y_{JM}^{(n)}$, and the fact that summation over $-M$ can be replaced by summation over M , were employed. Comparison of the final expression in Eq. (129) with that in Eq. (130) shows the two are identical. Thus, the tensor $\tilde{A}$ given in Eq. (128), which corresponds to the cross section [Eq. (126)] and polarization [Eq. (127)], satisfies the unitarity relation [Eq. (19)]. What we have done here is to present the proof of the unitarity relation for a

special case, in which the scattering amplitude tensor is known. This demonstrates that the Mie solution for scattering by a sphere of constant dielectric material does satisfy unitarity. The computationally more difficult problem of using the unitarity relation to determine the phases from the scattering cross section and polarization is left to subsequent developments.

VII. DISCUSSION

The main result of this article is that knowledge of the cross section and polarization parameters of scattered radiation is sufficient to determine, under certain conditions on the magnitude of the above quantities, the sines of the phases of the scattering amplitude tensor, $\sin \alpha_{ji}$. The only ambiguity of physical relevance which is left then with regard to each phase α_{ji} is the sign of $\cos \alpha_{ji}$, and this ambiguity can be removed too, if the scattering data is known for all frequencies, by the use of dispersion relations.

The possibility of applying our results in practice depends on the answers, for each specific phase problem under consideration, to a number of questions:

a) Are the conditions of Theorem 1 or Theorem 2 satisfied by the data available? A positive answer here will, of course, by encouraging, since it will assure us of, say, convergence of the iterative scheme, the validity of the error bounds on the iterant that we provided, and also most important, of the uniqueness of the solutions. Nevertheless, even when the conditions of Theorems 1 and 2 are not obeyed, it does not necessarily follow that the unitarity method cannot be applied, only that it will be much less reliable and rigorous. The conditions imposed are sufficient, not necessary ones, and it is possible, for instance, that convergence obtains also under less restrictive requirements. (We have, in fact, a very trivial example demonstrating this, but we shall not bring it in this article.)

b) Is the accuracy of the data sufficient and its behavior convenient for practical solution of the unitarity equation? For application of

quadrature formulae necessary for the evaluation of the iterants of the unitarity relation, it is highly desirable that the data cross section be smooth and slowly varying function of the scattering angle; otherwise numerical integration becomes very difficult. This actually may be considerably worrying in the case of application to x-ray diffraction by crystals of high symmetry. The cross sections then have many sharp alternating maxima and minima.

c) There is finally the question of whether unitarity breaking may arise in the context of the application. Clearly the unitarity method will give as the phases of the true scattering amplitude, when the cross section and polarization data are correct. However, sometimes we are interested not in those phases as such, but in phases α'_{ji} of quantities which are identical with (or simply related to) the scattering amplitude, only when some approximations are introduced. In such cases we can find the above phases α'_{ji} by the unitarity method, only if the approximations mentioned do not break unitarity (at least do not break it "strongly" enough so as to introduce large errors). We have already seen in Secs. VI-i and VI-ii that there are approximations in scattering theory which give an almost correct value for the cross section, but break unitarity. For a further illustration of this point, consider the phase problem in x-ray diffraction. In this field the central aim is to find the so-called structure amplitude $F(\underline{k}', \underline{k})$, the Fourier transform of which gives the electron density distribution of the scattering object.^{1,9} Now it so happens that within the confines of the approximations employed in x-ray diffraction, one has for the scattering amplitude tensor of the scattering object $\tilde{A}(\underline{k}', \underline{k})$:

$$\tilde{A}(\underline{k}', \underline{k}) = F(\underline{k}', \underline{k}) \tilde{A}_{F.E.}(\underline{k}', \underline{k})$$

where $\tilde{A}_{F.E.}(\underline{k}', \underline{k})$ is the scattering amplitude for a classical free electron (see Sec. VI-1). (Note that the above relation is given in almost every textbook on x-ray diffraction,^{1,9} but in a different, though equivalent, form: They use in their expressions the amplitudes of the incoming and scattered waves, and do not employ explicitly the scattering amplitude tensor.) Thus we can use the unitarity method to find the phase of F in x-ray problems, but this only provided the approximation by which the above equation obtains do not violate unitarity. These approximations involve, for instance, neglecting the correlation in the scattering by the various electrons, the contributions of nuclear motions to the scattering, etc. We do not know yet from theory, and one may have to leave it for numerical calculations to decide, if these approximations (which give a cross section in agreement with experiment) break unitarity to an extent which could lead to serious errors in the phase determined.

In conclusion it can be said that the unitarity method seems an attractive tool for coping with the phase problem from a theoretical point of view, but its usefulness as a practical device can be decided only by numerical computations in specific applications.

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- 6. A scattering-amplitude function description of x-ray scattering is possible because the polarization states of the incoming and the outgoing waves are completely known, and are the same for the usual x-ray experiments. The incoming beam is unpolarized and the polarization state of the scattered radiation is determined by Thomson's polarization factor.¹ This is correct only within the approximations usually adopted in the analysis of x-ray diffraction; i.e., that
- 7. See ref. 4, p. 15-16.
- 8. An example of scattering experiments of a sophisticated type which do give information on phases are those based on the Hanbury-Brown and Twiss Effect. They involve simultaneous measurements with two

- beams and two detectors from the same target. See, for instance, R. Hanbury-Brown and R. Q. Twiss, *Phil. Mag.* 45, 663 (1954); *Proc. Roy. Soc. (London)* A242, 300 (1957); ref. 4, Sec. 4.3.
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 15. D. S. Saxson in "Symposium on Electromagnetic Wave Theory" (Report of Conference held June 1955 at the University of Michigan); *Institute of Radio Engineers Transactions* (New York), Vol. AP-4, 579 (1956).
 16. Ref. 4, p. 17.
 17. The phrasing of the proof is such that it supposes that there is a unique pair of indices r, t for which $\text{Max}_{jl} |\sin \alpha_{jl}^A(\theta_n) - \sin \beta_{jl}^B(\theta_n)|$ is attained. As can be verified by an inspection, this is not a necessary assumption and the result holds if two or more pairs of indices yield exactly the same magnitude.
 18. Ref. 4, p. 107.

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26. We assume implicitly that the electron oscillates with the same frequency ω_0 before it is perturbed by the incident field, for every chosen axis $\underline{x}$ (i.e., every possible direction of incident field $\underline{E}_0$). If the oscillations were not isotropic in this sense, the scatterer would not have the property of rotational-reflectional symmetry that we require subsequently.
27. Although q is proportional to ω , it remains negligibly small compared with 1, $q \ll 1$, for γ as large as the frequencies typical of nuclear γ rays! This is not true for the "harder" γ rays of cosmic radiation, but this is hardly a problem to be discussed in terms of classical models.
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31. Ref. 4, pp. 32-35.

$T + H_2$
($E - E_c = 0.2 \text{ eV}$)

$$(E - E_c = 0.2 \text{ eV})$$

Modified H_3 (#1)
($E - E_c = 0.05 \text{ eV}$)

Modified H_3 (#2)

H + HBr ($E_c = 0.0053\text{eV}$)

A + BC

$$E_c = 0.306 \text{ eV}; \quad E = E_c + 0.2 \text{ eV}; \quad M_A = M_B = M_C = 1$$