Extracting dynamics from collision data. I. Analysis of integral angular momentum

John Bohn* and U. Fano

Department of Physics and James Franck Institute, The University of Chicago, Chicago, Illinois 60637

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Collision data in forms currently available from experimental sources are reduced by quadratures and algebraic transformations to dynamical parameters. The reduction hinges on suitable normalizations and utilization of sum rules and removes the influence of experimental geometries. The dynamical parameters, labeled by angular momenta restricted to a modest range, appear more suited to comparison with theory than the original experimental data. Our analytical developments are simplified by certain restrictions, whose removal is discussed in a final section.

I. INTRODUCTION

The problem of extracting dynamical information from collision experiments has been formulated and discussed extensively in a series of papers by Lee,[1] without arriving at an explicit solution. Formulas of Ref. 1 express collision data as bilinear combinations of scattering matrix elements $S^{\dagger}\times S$ with algebraic coefficients that represent the joint influence of angular momenta and other geometric factors. No procedure has been developed previously to disentangle individual elements of such bilinear expressions from an adequate set of measured data, except for a conference report on part of the present work.[2]

We introduce here three remarks.

(1) The algebraic expression of collision data transforms a set of elements of $S^{\dagger}\times S$ into the set of collision data; the matrix U of this transformation is unitary, provided both sets are properly normalized.

(2) Each of the normalized sets of collision data and of $S^{\dagger}\times S$ elements may be interpreted as the set of components of a unit vector, related to the other by U.

(3) The relevant collision data are constructed as products of polarization parameters of the initial and final states of the target by the following procedure: each of these complete sets of parameters is evaluated for targets prepared in a specific pure initial state; their product is then *averaged* over a complete set of alternative initial states. The total set of measurements will form a "complete experiment."

Remark (2) is critical for disentangling single elements of the S matrix from their bilinear products $S^{\dagger}\times S$. Unit normalization of the set of elements of $S^{\dagger}\times S$, viewed as components of a *vector,* implies that the square of the *matrix* $S^{\dagger}\times S$ has unit trace. It follows that each set of matrix elements $\{S_{\alpha\beta}\}$ or $\{S^{\dagger}_{\gamma\delta}\}$ represents the components of the eigenvector corresponding to the single nonzero (unit) eigenvalue of the matrix $S^{\dagger}\times S$. Each element $S_{\alpha\beta}$ (or $S^{\dagger}_{\gamma\delta}$) can thus be extracted from the bilinear set $\{S^{\dagger}_{\gamma\delta}S_{\alpha\beta}\}$.

These remarks will be developed in Sec. III. Section II will instead show how to extract the S matrix elements from collision data for targets that are initially in a ^1S state without resorting to the full machinery of Sec. III. Section II stands thus also independent of Ref. 1. Its application to the extensive data[3] on the process

$$e+\mathrm{He}(1s^2\,{}^1S)\rightarrow e'+\mathrm{He}(1s2p\,{}^1P) \tag{1}$$

is reported separately.[4] Section III will instead relate to Sec. II of Ref. 1(c) and to general aspects of Ref. 1(a). The remainder of Ref. 1(c) and other parts of Ref. 1 that stress symmetries will bear only on applications, which are not presented here.

Successful unraveling of scattering amplitudes $S_{\alpha\beta}$ from collision data will also shift the focus of dependence on the scattering angle. Reference 1 focused on the observables' dependence on this angle and therefore expanded the observables in harmonic series. The harmonic analysis will be performed here on probability *amplitudes* already unraveled from the observables, thus bypassing interferences in their angular dependence. Interferences in the linkage of projectile and target parameters are also bypassed.

Certain restrictions are implied for simplicity by the formulations of Secs. II and III, as in Ref. 1: spins of electrons and nuclei and exchange effects are not considered. Only pure states of projectile and target are considered. Reactive collisions are excluded. In addition, effects of parity and of angular momentum conservation are not dealt with explicitly, in contrast to Ref. 1. Prospects for lifting these restrictions are presented in the final Sec. IV.

II. SPECIAL CASE: INITIALLY SPHERICAL TARGETS

A. Target polarization formulas

Experimental studies of electron collisions with atoms have dealt extensively with He and alkaline earth targets, initially in their ^1S ground states. Inelastic collisions have largest cross sections for excitation to ^1P levels, but we find it instructive here to consider excitations to ^1L levels with an unspecified value of L. We disregard, however, for simplicity, the incident electron's spin, which is realistic for low-Z targets.

Following general practice[3] we characterize the collision effect at the outset by a set of probability amplitudes $\{a_M(\theta)\}$ for target excitation to each of its degenerate states $-L \le M \le L$ of a specific ${}^1L$ level. The index M represents the angular momentum component L_z with the z axis along the incidence ("collision frame"). Each amplitude a_M depends on the scattering angle θ. Symmetry under reflection through the scattering plane requires that[3]

$$a_{-M}(\theta)=(-1)^M a_M(\theta) \; ; \tag{2}$$

accordingly we need consider only $M \ge 0$.

This representation of a collision effect by probability amplitudes $a_M(\theta)$ replaces the use of S matrices in Ref. 1. Recall that rows of a scattering matrix correspond usually to *all* final states attainable by a collision with given energy, whereas we deal here only with a subset of such states. Parametrizing elements of the subset by the probability amplitudes $a_M(\theta)$ affords the appropriate normalization. Specifically, we assume they are normalized to unity at each scattering angle:[3]

$$\sum_M |a_M(\theta)|^2=1 \; . \tag{3}$$

The collision data describe the polarization of the target's final state, in addition to its excitation cross section. Following Ref. 5 we represent the polarization by the set of mean values of multipole operators T_Q^K whose matrix elements consist of Wigner coefficients

$$\langle LM'|T_Q^K|LM\rangle=(-1)^{L-M}\langle LM',L\,-M|KQ\rangle \; , \qquad 0\le K\le 2L, \quad -K\le Q\le K \; . \tag{4}$$

These coefficients pertain to the addition of the angular momenta of a ket $|LM\rangle$ and a bra $\langle LM'|$. The set of operators $\{T_Q^K\}$ is a special case of the complete sets of orthonormal operators $\{U_i\}$ in Ref. 6, regarded as vectors in a Hilbert space (the "Liouville representation"). Specifically, there are $(2L+1)^2$ operators T_Q^K, as many as there are elements of the matrix $\langle LM'|O|LM\rangle$ of an operator O, and the T_Q^K satisfy the orthonormality condition

$$\mathrm{Tr}(T_{Q'}^{K'\dagger}T_Q^K)=\delta_{KK'}\delta_{QQ'} \; . \tag{5}$$

The mean value of the operator T_Q^K for the target state with probability amplitudes $a_M(\theta)$ will be indicated by

$$\langle T_Q^K\rangle_\theta=\sum_{M',M} a_{M'}^*(\theta)\langle LM'|T_Q^K|LM\rangle a_M(\theta) \; . \tag{6}$$

Applying the normalization (3) and orthonormality of the Wigner coefficients in (4) normalizes the set of mean values,

$$\sum_{K,Q}\langle T_Q^{K\dagger}\rangle_\theta\langle T_Q^K\rangle_\theta=\left[\sum_M |a_M(\theta)|^2\right]^2=1 \; . \tag{7}$$

This normalization characterizes $\{\langle T_Q^K\rangle_\theta\}$ as the set of components of a unit vector, which in the Liouville representation represents the state of the target.

The operator T_0^0 deserves special mention because it is proportional to the identity operator, $\langle LM'|T_0^0|LM\rangle=(2L+1)^{-1/2}\delta_{MM'}$. Consequently its mean value is a constant independent of the target state,

$$\langle T_0^0\rangle_\theta=(2L+1)^{-1/2}\sum_M |a_M(\theta)|^2=(2L+1)^{-1/2} \; . \tag{8}$$

This means that vectors representing pure states are confined to a hyperplane of dimension $(2L+1)^2-1$ in the Liouville representation. We will return to this point in Sec. III B.

The collision data themselves, namely, the expectation values of the operators T_Q^K, correlated with the detection of an electron scattered through an angle θ with cross section $d\sigma(\theta)/d\Omega$ for excitation of the ${}^1L$ level, are then represented by the set

$$\left\{\langle T_Q^K\rangle_\theta \frac{d\sigma(\theta)}{d\Omega}\right\} . \tag{9}$$

The cross section itself is, however, irrelevant to our purpose of extracting dynamical parameters for a specific excitation, being factored out of the normalization (3). Dynamics will be extracted here from *ratios* of measurements. Accordingly, the term "collision data" will indicate hereafter the values of polarization parameters of the final target state $\{\langle T_Q^K\rangle_\theta\}$.

[Our use of $(2L+1)^2$ target polarization parameters contrasts with the smaller number of parameters, $4L$, that suffice to identify a "pure state" of the target. The vector of the Liouville representation with components $\langle T_Q^K\rangle_\theta$ is accordingly restricted to a $4L$-dimensional manifold. Its $(2L+1)^2-1$ components with $K\neq 0$ would, however, be linearly independent for a general ("mixed") state, to be touched upon in item (e) of Sec. IV.]

B. Extracting the probability amplitudes

Measurement of the entire set $\{\langle T_Q^K\rangle_\theta\}$ provides information on the $a_M(\theta)$ through the relation reciprocal to (6), namely,

$$a_{M'}^*(\theta)a_M(\theta)=\sum_{K,Q}\langle T_Q^K\rangle_\theta\langle KQ|LM',L\,-M\rangle(-1)^{L-M} \; . \tag{10}$$

The squared modulus of each amplitude $a_M(\theta)$ is found by setting $M'=M$, in which case the Wigner coefficient vanishes for $Q\neq 0$,

$$|a_M(\theta)|^2=\sum_K \langle T_0^K\rangle\langle K0|LM,L\,-M\rangle(-1)^{L-M} \; . \tag{11}$$

Since the coefficients on the right of (11) vanish for odd values of K, owing to symmetry under sign reversal of M, we see that $L+1$ nonzero values of $\langle T_0^K\rangle_\theta$ determine the $L+1$ different values of $|a_M(\theta)|^2$. Noting that even values of K correspond to electric charge multipoles of the electron density in the excitation of the ${}^1L$ level, Eq. (11) specifies that the *axially symmetric* components of these multipoles determine the squared amplitudes $|a_M(\theta)|^2$.

We are thus led to represent the $a_M(\theta)$ through their moduli and phases,

$$a_M(\theta)=|a_M(\theta)|e^{i\phi_M(\theta)} \; . \tag{12}$$

The phase *differences* $\phi_M(\theta)-\phi_{M+1}(\theta)$ are determined by setting $Q=1$ in (10),

$$a^*_{M+1}(\theta)a_M(\theta)=|a_{M+1}(\theta)||a_M(\theta)| \times e^{i[\phi_M(\theta)-\phi_{M+1}(\theta)]} \times \sum_K \langle T_1^K\rangle_\theta \langle K1|LM+1,L-M\rangle \times(-1)^{L-M} . \quad (13)$$

We have thus completed our immediate task of determining the $a_M(\theta)$ to within a common phase factor to be set by convention. This task has utilized only the values of $\langle T_Q^K\rangle_\theta$ with $Q=0$ and 1. Measurements of the values for $Q>1$ may, however, be required to verify the completeness relation (7). Specifically, a sufficient number of $\langle T_Q^K\rangle_\theta$, or of equivalent linearly independent parameters, need to be measured to exhaust the sum in (7).

C. Dynamical parameters

The Introduction to Ref. 1(a) has stressed the desirability of comparing experimental and theoretical data in the form of dynamical parameters free from geometrical or incidental aspects of experiments (see also Sec. 7.10.1 and p. 248 of Ref. 7). These parameters should thus depend on the magnitudes rather than on the directions of angular momenta and on the profiles of the functions $a_M(\theta)$ rather than on the selection of specific scattering angles. We should accordingly replace the parameters $a_M(\theta)$ with a new set independent of both the scattering angle θ and the magnetic quantum number M. The dependence on θ will be removed by expansion in spherical harmonics, while the dependence on M indices will be factored out in the form of Wigner coefficients.

We begin by indicating explicitly the dependence of $a_M(\theta)$ on initial and final quantum numbers of the target and on the initial and final directions of the projectile,

$$a_M(\theta)=\langle LM,\hat{\mathbf{p}}_b|g|00,\hat{\mathbf{p}}_a\rangle, \quad \cos\theta=\hat{\mathbf{p}}_b\cdot\hat{\mathbf{p}}_a . \quad (14)$$

Here (a,b) label the initial and final states of the projectile, as in Ref. 1, and g replaces the amplitude a to avoid confusion. The dependence on the directions is expanded into spherical harmonics using the relevant elements of plane-wave expansions [Eq. (4.7) of Ref. 7]

$$|\hat{\mathbf{p}}_a\rangle=\sum_{l_a,m_a} i^{l_a} Y_{l_a m_a}(\hat{\mathbf{p}}_a) , \qquad \langle\hat{\mathbf{p}}_b|=\sum_{l_b,m_b} i^{-l_b} Y^*_{l_b m_b}(\hat{\mathbf{p}}_b) . \quad (15)$$

Having set the z axis parallel to $\hat{\mathbf{p}}_a$ and setting now the x axis in the scattering plane, we have

$$Y_{l_a m_a}(\hat{\mathbf{p}}_a)=\left[\frac{2l_a+1}{4\pi}\right]^{1/2}\delta_{m_a 0} , \qquad Y^*_{l_b m_b}(\hat{\mathbf{p}}_b)=Y^*_{l_b m_b}(\theta,0) . \quad (15')$$

Equation (14) expands thus into

$$a_M(\theta)=\sum_{l_a,l_b,m_b} i^{l_a-l_b}\left[\frac{2l_a+1}{4\pi}\right]^{1/2} Y^*_{l_b m_b}(\theta,0) \times\langle LM,l_b m_b|g|00,l_a 0\rangle . \quad (16)$$

The desired dynamical parameter G emerges now by factoring out of g its dependence on magnetic quantum numbers, setting

$$\langle LM,l_b m_b|g|00,l_a0\rangle\equiv\langle L0|G|l_b l_a\rangle\langle LM|l_a0,l_b-m_b\rangle \times(-1)^{l_b-m_b} , \quad (17)$$

which requires m_b+M to vanish. Entering (17) into (16), we find that G is related to the probability amplitudes by

$$a_M(\theta)=\sum_{l_a,l_b} i^{l_a-l_b}\left[\frac{2l_a+1}{4\pi}\right]^{1/2} Y^*_{l_b -M}(\theta,0) \times\langle L0|G|l_b l_a\rangle\langle LM|l_a0,l_bM\rangle(-1)^{l_b+M} . \quad (18)$$

The orthonormality of the spherical harmonics allows us to single out a particular value of l_b':

$$2\pi\int_{-1}^{1} d(\cos\theta)Y_{l_b' -M}(\theta,0)a_M(\theta)=\sum_{l_a} i^{l_a-l_b'}\left[\frac{2l_a+1}{4\pi}\right]^{1/2}\langle L0|G|l_b'l_a\rangle\langle LM|l_a0,l_b'M\rangle(-1)^{l_b'+M} . \quad (19)$$

Now we appeal once again to the orthogonality of the Wigner coefficients, using the modified relation

$$\sum_M \langle l_a'0,l_b'M|LM\rangle\langle LM|l_a0,l_b'M\rangle=\left[\frac{2L+1}{2l_a+1}\right]\delta_{l_a'l_a} , \quad (20)$$

which can be derived from the formulas in Chap. 1 of Ref. 9. Using (20), our final result for G, after removing the primes for simplicity of notation, becomes

$$\langle L0|G|l_b l_a\rangle=\left[\frac{2l_a+1}{2L+1}\right] i^{l_b-l_a}\left[\frac{2l_a+1}{4\pi}\right]^{-1/2}\sum_M\left[2\pi\int_{-1}^{1} d(\cos\theta)Y_{l_b -M}(\theta,0)a_M(\theta)\right](-1)^{l_b-m_b}\langle l_a0,l_b-m_b|LM\rangle . \quad (21)$$

where m indicates the reduced mass of projectile and target, as discussed further in Ref. 4.

Equation (21) identifies a very large set of dynamical parameters G because the magnitudes of the orbital momenta (l_a, l_b) range to infinity. The difference $|l_a - l_b|$ is restricted to $\le L$ by the triangular condition on (l_a, l_b, L) but the sum $l_a + l_b$ is unrestricted. The magnitude of G converges to zero as $l_a + l_b$ increases but a very large set of G values remains nevertheless significant. On the other hand, the small values of G with large $l_a + l_b$ need not be extracted from experiments, being provided dependably by the perturbative Born approximation. A procedure for systematic application of this approach has been developed recently[8] and is applied and evaluated in Ref. 4.

Note here that the dynamical parameter G is defined by (21) in terms of the probability amplitude a_M for the target transition $0 \to M$. This amplitude is proportional in turn to the corresponding transition amplitude T'_M, solution of a Lippman-Schwinger equation $T'_M = [V + V(E - H + i\epsilon)^{-1} T']_M$, according to

$$a_M = -\left[\frac{4\pi\hbar^2}{2m}\right]\left[\frac{k_i}{k_f}\frac{d\sigma}{d\Omega}\right]^{-1/2} T'_M \ . \tag{21'}$$

III. INITIALLY NONSPHERICAL TARGETS

A. Target polarization formulas

Whereas Sec. II labeled a base set of target states by indices (L, M), we consider now initial states $|L_A M_A\rangle$ and final states $\langle L_B M_B|$, in accordance with Ref. 1. We still consider targets in singlet states and disregard any projectile spin, for simplicity. Also, we restrict ourselves to "parity favored" transitions, i.e., those in which $L_A + L_B + j_t$ and $l_a + l_b + j_t$ in Eq. (22) are both even.

Characterization of the collision effect on the target by a set of probability amplitudes $a_M(\theta)$ no longer suffices here. Equation (14) associates each $a_M(\theta)$ to a transition of the target from an initial state $|00\rangle$ to a final state $\langle LM|$. The more general probability amplitudes $\langle L_B M_B, \hat{\mathbf{p}}_b | g | L_A M_A, \hat{\mathbf{p}}_a\rangle$, which bear on the present case, pertain to the probability that the target undergoes the transition $|L_A M_A\rangle \to \langle L_B M_B|$ when the projectile is scattered through the angle $\theta = \arccos(\hat{\mathbf{p}}_b \cdot \hat{\mathbf{p}}_a)$. Following again Ref. 1, we characterize this transition by the angular momentum $\mathbf{j}_t = \mathbf{L}_B - \mathbf{L}_A^*$ transferred to the target. The probability amplitudes transform accordingly into a new set $\{a_{j_t m_t}(\theta)\}$ defined by

$$a_{j_t m_t}(\theta) \equiv \sum_{M_A, M_B} \langle j_t m_t | L_A - M_A, L_B M_B\rangle (-1)^{L_A - M_A} \times \langle L_B M_B, \hat{\mathbf{p}}_b | g | L_A M_A, \hat{\mathbf{p}}_a\rangle \ . \tag{22}$$

The $a_{j_t m_t}(\theta)$ have a normalization parallel to (3):

$$\sum_{j_t, m_t} |a_{j_t m_t}(\theta)|^2 = \sum_{M_A, M_B} |\langle L_B M_B, \hat{\mathbf{p}}_b | g | L_A M_A, \hat{\mathbf{p}}_a\rangle|^2 = 1 \ , \tag{23}$$

reflecting the fact that the total probability of the target making a transition from $\mathbf{L}_A$ to $\mathbf{L}_B$ is set to unity.

As in Sec. II, the polarization of the target's final state is given by a set of mean values of tensorial operators $\{\langle T^{K_B}_{Q_B}\rangle_\theta\}$ with matrices and normalizations analogous to (4) and (5). Here, however, we must take into account the nontrivial initial state, indicated by a density operator ρ_A, and consider the mean values $\{\langle T^{K_B}_{Q_B}\rangle_{\rho_A \theta}\}$ conditional on this initial state. Initial and final target states are related by the probability amplitudes in the usual way, analogous to (10) of Ref. 1(c),

$$\begin{aligned}\langle T^{K_B}_{Q_B}\rangle_{\rho_A \theta} &= \mathrm{Tr}[g(\theta)\rho_A g^\dagger(\theta) T^{K_B}_{Q_B}] \\ &= \sum_{M_A, M'_A, M_B, M'_B} \langle L_B M_B, \hat{\mathbf{p}}_b | g | L_A M_A, \hat{\mathbf{p}}_a\rangle \langle L_A M_A | \rho_A | L_A M'_A\rangle \\ &\quad \times \langle L_A M'_A, \hat{\mathbf{p}}_a | g^\dagger | L_B M'_B, \hat{\mathbf{p}}_b\rangle \langle L_B M'_B | T^{K_B}_{Q_B} | L_B M_B\rangle \ .\end{aligned} \tag{24}$$

We now expand the density operator ρ_A into a set of tensorial operators[6]

$$\rho_A = \sum_{K_A, Q_A} \langle T^{K_A}_{Q_A}\rangle_{\rho_A} T^{K_A\dagger}_{Q_A} \ , \tag{25}$$

where the Hermitian conjugation of $T^{K_A\dagger}_{Q_A}$ accords with the minus signs of $\mathbf{L}_A$ in the relation $\mathbf{j}_t = \mathbf{L}_B - \mathbf{L}_A^*$ and of M_A in (22). Using (4), the definitions (22) and (25), and rearranging, Eq. (24) becomes

$$\langle T^{K_B}_{Q_B}\rangle_{\rho_A\theta}=\sum_{K_A,Q_A}\langle T^{K_A}_{Q_A}\rangle_{\rho_A}\sum_{j_t',m_t',j_t,m_t}a^*_{j_t'm_t'}(\theta)a_{j_tm_t}(\theta)$$
$$\times\sum_{M_A,M_A',M_B,M_B'}(-1)^{L_B-M_B'}\langle L_AM_A',L_B-M_B'|j_t'-m_t'\rangle(-1)^{j_t'-m_t'}$$
$$\times(-1)^{L_A-M_A}\langle L_A-M_A,L_BM_B|j_tm_t\rangle$$
$$\times(-1)^{L_A-M_A'}\langle L_A-M_A',L_AM_A|K_A-Q_A\rangle(-1)^{K_A-Q_A}$$
$$\times(-1)^{L_B-M_B}\langle L_BM_B',L_B-M_B|K_BQ_B\rangle\ . \tag{26}$$

The final sum on the right of (26) transforms according to Eq. (3.21) of Ref. 9 into

$$\sum_{K_t,Q_t}(-1)^{j_t-m_t}\langle j_t'm_t',j_t-m_t|K_tQ_t\rangle\langle K_tQ_t|K_A-Q_A,K_BQ_B\rangle(-1)^{K_A-Q_A}$$
$$\times\langle (L_AL_B)j_t',(L_AL_B)j_t|(L_AL_A)K_A,(L_BL_B)K_B\rangle^{(K_t)}\ . \tag{27}$$

This transformation has the effect of reducing the direct product of operators $\{T^{K_A\dagger}_{Q_A}\times T^{K_B}_{Q_B}\}$ in the $|L_AM_A,L_BM_B\rangle$ representation to a set of operators $\{T^{K_t}_{Q_t}\}$ in the $|j_tm_t\rangle$ representation.[5] The coefficient on the far right of (27), which as an invariant does not depend on Q_t, is expressed in terms of a 9-j symbol in Eq. (3.9) of Ref. 9. It also appears in Eq. (14) of Ref. 1(c), where a similar reduction takes place. Hereafter, we shall suppress the indices L_A and L_B in this coefficient, writing it as $\langle j_t'j_t|K_AK_B\rangle^{(K_t)}$.

Now we write expressions for "mean values" of the $T^{K_t}_{Q_t}$, which correspond to the state multipole moments transferred to the target, by analogy with (6),

$$\langle T^{K_t}_{Q_t}\rangle_\theta=\sum_{j_t',m_t',j_t,m_t}a^*_{j_t'm_t'}(\theta)\langle j_t'm_t'|T^{K_t}_{Q_t}|j_tm_t\rangle$$
$$\times a_{j_tm_t}(\theta)\langle j_t'j_t|K_AK_B\rangle^{(K_t)}\ . \tag{28}$$

The initial and final state polarization data are therefore related through the multipole moments $\langle T^{K_t}_{Q_t}\rangle_\theta$ by

$$\langle T^{K_B}_{Q_B}\rangle_{\rho_A\theta}=\sum_{K_A,Q_A}\left[\sum_{K_t,Q_t}\langle T^{K_t}_{Q_t}\rangle_\theta\times\langle K_tQ_t|K_A-Q_A,K_BQ_B\rangle\times(-1)^{K_A-Q_A}\right]\langle T^{K_A}_{Q_A}\rangle_{\rho_A}\ . \tag{29}$$

B. Extracting probability amplitudes

Whereas Sec. II B extracted amplitudes by direct analytic inversion of their relationship to the polarization of the final target state, the relation of the $a_{j_tm_t}(\theta)$ to measured $\langle T^{K_B}_{Q_B}\rangle_{\rho_A\theta}$ resolves into *two* steps, namely (28) and (29). Our first step of inversion should therefore disentangle the initial state parameters $\langle T^{K_A}_{Q_A}\rangle_{\rho_A}$ from the right of (29), thus recasting the collision data into the directly invertible form (28).

To this end we denote the quantity in large parentheses in (29) by $F(K_A,Q_A;K_B,Q_B)_\theta$, and note that it is explicitly independent of the initial state ρ_A. Next we choose a complete orthonormal basis $\{\rho^j_A\}$ consisting of $(2L_A+1)^2$ density operators. Likewise, we choose an ordering of the basis set $\{T^{K_A}_{Q_A}\}$ and denote the resulting set by $\{U_i\}$, $1\le i\le(2L_A+1)^2$; by convention, we set $U_1=T^0_0$. Then for each *fixed* pair (K_B,Q_B), (29) reads

$$\langle T^{K_B}_{Q_B}\rangle_{\rho^j_A\theta}=\sum_i F(i;K_B,Q_B)_\theta\langle U_i\rangle_{\rho^j_A}\ . \tag{29'}$$

As j varies, Eqs. (29′) describe a linear system in the unknowns F with matrix $V_{ij}=\langle U_i\rangle_{\rho^j_A}$. But V_{ij} is the matrix of the transformation which carries each orthonormal basis vector U_i onto the corresponding orthonormal basis vector ρ^j_A, and is therefore *unitary*. Equations (29) and (29′) can thus be inverted analytically:

$$F(i;K_B,Q_B)_\theta=\sum_j\langle U_i^\dagger\rangle_{\rho^j_A}\langle T^{K_B}_{Q_B}\rangle_{\rho^j_A\theta}\ , \tag{30}$$

$$\sum_{K_t,Q_t}\langle T^{K_t}_{Q_t}\rangle_\theta\langle K_tQ_t|K_A-Q_A,K_BQ_B\rangle(-1)^{K_A-Q_A}=\sum_j\langle T^{K_A\dagger}_{Q_A}\rangle_{\rho^j_A}\langle T^{K_B}_{Q_B}\rangle_{\rho^j_A\theta}\ . \tag{30'}$$

The multipole moments are therefore given by

$$\langle T_{Q_t}^{K_t} \rangle_\theta = \sum_{Q_A,Q_B} \left[\sum_j \langle T_{Q_A}^{K_A \dagger} \rangle_{\rho_A^j} \langle T_{Q_B}^{K_B} \rangle_{\rho_A^j \theta} \right] \times (-1)^{K_A - Q_A} \langle K_A - Q_A, K_B Q_B | K_t Q_t \rangle \,, \tag{31}$$

requiring a *sum* over a *complete set* of initial states.

Equation (31) provides the desired set $\{\langle T_{Q_t}^{K_t} \rangle_\theta\}$ as a weighted sum over collision data $\langle T_{Q_B}^{K_B} \rangle_{\rho_A \theta}$. A geometrical aspect of the inversion of (29′) views the set $\{\langle U_i \rangle_{\rho_A^j}\}$ as the components of a vector representing the density operator ρ_A^j. Upon multiplication of both sides of (29′) by one component $\langle U_k^\dagger \rangle_{\rho_A^j}$ and summation over all values of j, ρ_A^j ranges symmetrically over the hyperplane defined by $U_1 = (2L_A+1)^{-1/2}$ in the Liouville representation. This causes the sums over dyadics $\sum_j \langle U_k^\dagger \rangle_{\rho_A^j} \langle U_i \rangle_{\rho_A^j}$ with $k \neq i$ to vanish, whereas $\sum_j \langle U_i^\dagger \rangle_{\rho_A^j} \langle U_i \rangle_{\rho_A^j}$ reduces to unity owing to (5).

Having thus seen that the measured values of the final target parameters $\langle T_{Q_B}^{K_B} \rangle_{\rho_A \theta}$, properly averaged over a complete set of ρ_A, yield the set of parameters $\langle T_{Q_t}^{K_t} \rangle_\theta$, we may enter the latter in (28) and extract the $a_{j_t m_t}(\theta)$ values by analytic inversion. The inverted equation, analogous to (10), is

$$a^*_{j_t' m_t'}(\theta) a_{j_t m_t}(\theta) = \sum_{K_t, Q_t} \left[\sum_{K_A, K_B} \langle T_{Q_t}^{K_t} \rangle_\theta \times \langle K_A K_B | j_t' j_t \rangle^{(K_t)} \right] \times \langle K_t Q_t | j_t' m_t', j_t - m_t \rangle \times (-1)^{j_t - m_t} \,. \tag{32}$$

Proceeding as in Sec. II B we begin extracting amplitudes by selecting the terms of (32) diagonal in (j_t, m_t), which determine the amplitude moduli,

$$|a_{j_t m_t}(\theta)|^2 = \sum_{K_t} \left[\sum_{K_A, K_B} \langle T_0^{K_t} \rangle_\theta \langle K_A K_B | j_t j_t \rangle^{(K_t)} \right] \times \langle K_t 0 | j_t m_t, j_t - m_t \rangle (-1)^{j_t - m_t} \,. \tag{33}$$

The next step, parallel to (12), introduces the phase of each amplitude

$$a_{j_t m_t}(\theta) = |a_{j_t m_t}(\theta)| \exp[i \phi_{j_t m_t}(\theta)] \,, \tag{34}$$

and uses the terms with $j_t' = j_t$ but $m_t' = m_t - 1$ and $Q_t = 1$,

$$|a_{j_t m_t - 1}(\theta)| |a_{j_t m_t}(\theta)| \exp\{i[\phi_{j_t m_t}(\theta) - \phi_{j_t m_t - 1}(\theta)]\} = \sum_{K_t} \left[\sum_{K_A, K_B} \langle T_1^{K_t} \rangle_\theta \langle K_A K_B | j_t j_t \rangle^{(K_t)} \right] \langle K_t 1 | j_t m_t - 1, j_t m_t \rangle (-1)^{j_t - m_t} \,. \tag{35}$$

The third and final step determines the phase differences between the amplitudes with any pair of different (j_t, j_t') but with $m_t' = m_t$,

$$|a_{j_t' m_t}(\theta)| |a_{j_t m_t}(\theta)| \exp\{i[\phi_{j_t m_t}(\theta) - \phi_{j_t' m_t}(\theta)]\} = \sum_{K_t} \left[\sum_{K_A, K_B} \langle T_0^{K_t} \rangle_\theta \langle K_A K_B | j_t' j_t \rangle^{(K_t)} \right] \langle K 0 | j_t' m_t, j_t - m_t \rangle (-1)^{j_t - m_t} \,. \tag{36}$$

Application of this formula for a *single* value of m_t, say $m_t = 0$, together with recursive application of (35), determines all the phases to within a single phase to be set by convention for all $a_{j_t m_t}(\theta)$.

We review now the circumstances that have afforded the determination of the amplitudes $a_{j_t m_t}(\theta)$ from collision data, particularly the role of the Introduction's "remark (2)." Let us indicate for this purpose the right-hand side of (32) as a matrix $\tau_{j_t' m_t' j_t m_t}$, which corresponds to the $S^\dagger \times S$ of Ref. 1. The matrix so defined is readily seen to satisfy both $\mathrm{Tr}(\tau) = 1$ and $\mathrm{Tr}(\tau^2) = 1$, owing to (a) the orthonormality of all Wigner coefficients, (b) the orthonormality of $\langle K_A K_B | j_t' j_t \rangle^{(K_t)}$, and (c) the completeness of the operator sets $\{T_{Q_A}^{K_A}\}$ and $\{T_{Q_B}^{K_B}\}$ as represented by (5). The condition $\mathrm{Tr}(\tau) = \mathrm{Tr}(\tau^2) = 1$ guarantees that τ has a single nonzero eigenvalue equal to 1, just as the condition $\mathrm{Tr}(\rho) = \mathrm{Tr}(\rho^2) = 1$ guarantees that the density operator ρ represents a pure state. Also, the set of amplitudes $\{a_{j_t m_t}\}$ represents the eigenvector for τ that belongs to its unit eigenvalue.

Two further and essential circumstances, however, underlie the expression (31) of τ in terms of collision data $\{\langle T_{Q_B}^{K_B} \rangle_{\rho_A \theta}\}$. Each element of this complete set must have been measured, *and* it must have been measured for each initial state of the set $\{\rho^j_A\}$, whose normalization to $\mathrm{Tr}[\sum_j(\rho^j_A)^2]=1$ requires each state to be pure. This requirement goes beyond the usual sense of the term "complete experiment," in which only a single set of collision data are measured. Failure of either condition would cause $\mathrm{Tr}(\tau^2)$, as determined from collision data on the left of (32), to fall short of unity, with consequences to be described in item (e) of Sec. IV.

C. Dynamical parameters

The extraction of dynamical parameters proceeds now as in Sec. II C, except that we must first determine the correct partial wave expansion of the $a_{j_t m_t}(\theta)$. To do this, we expand the probability amplitudes $\langle L_B M_B,\hat{\mathbf{p}}_b|g|L_A M_A,\hat{\mathbf{p}}_a\rangle$ by analogy with (16),

$$\langle L_B M_B,\hat{\mathbf{p}}_b|g|L_A M_A,\hat{\mathbf{p}}_a\rangle=\sum_{l_a,l_b,m_b} i^{l_a-l_b}\left[\frac{2l_a+1}{4\pi}\right]^{1/2} Y^*_{l_b m_b}(\theta,0)\langle L_B M_B,l_b m_b|g|L_A M_A,l_a 0\rangle\ . \tag{37}$$

The matrix element on the right of (37) can, in turn, be expanded into products of Wigner coefficients and rotationally invariant factors depending on the total angular momentum $\mathbf{J}=\mathbf{L}_A+\boldsymbol{l}_a=\mathbf{L}_B+\boldsymbol{l}_b$ of the target plus projectile complex:

$$\langle L_B M_B,l_b m_b|g|L_A M_A,l_a 0\rangle=\sum_{J,M}\langle L_B M_B,l_b m_b|JM\rangle\langle L_B l_b|G(J)|L_A l_a\rangle\langle JM|L_A M_A,l_a 0\rangle\ . \tag{38}$$

A recoupling similar to that in Eqs. (3)–(6) of Ref. 1(c) translates (38) from the total angular momentum representation to the angular momentum transfer representation,

$$\begin{aligned}\langle L_B M_B,l_b m_b|g|L_A M_A,l_a 0\rangle=\sum_{j_t,m_t}&(-1)^{L_A-M_A}\langle L_A\,-M_A,L_B M_B|j_t m_t\rangle\langle L_B L_A|G(j_t)|l_b l_a\rangle\\&\times\langle j_t m_t|l_a 0,l_b\,-m_b\rangle(-1)^{l_b-m_b}\ .\end{aligned} \tag{39}$$

The same transformation also serves to define the dynamical parameters G,

$$\langle L_B L_A|G(j_t)|l_b l_a\rangle\equiv\sum_J\langle L_B l_b|G(J)|L_A l_a\rangle(2J+1)\begin{Bmatrix}L_A & l_a & J\\ l_b & L_B & j_t\end{Bmatrix}. \tag{40}$$

[Note that the phase factors in (39) and (40) are different from those in Eqs. (5) and (6) of Ref. 1(c). Our concern here, as throughout this paper, is to emphasize that writing a ket in bra notation involves a contragredient transformation, e.g., "$|L_A M_A\rangle$" and "$(-1)^{L_A-M_A}\langle L_A-M_A|$" both denote ket states.]

Substituting (39) back into (37) and rearranging yields

$$\begin{aligned}\langle L_B M_B,\hat{\mathbf{p}}_b|g|L_A M_A,\hat{\mathbf{p}}_a\rangle=\sum_{j_t,m_t}&(-1)^{L_A-M_A}\langle L_A\,-M_A,L_B M_B|j_t m_t\rangle\\&\times\left[\sum_{l_a,l_b,m_b} i^{l_a-l_b}\left[\frac{2l_a+1}{4\pi}\right]^{1/2} Y^*_{l_b m_b}(\theta,0)\langle j_t m_t|l_a 0,l_b\,-m_b\rangle\right.\\&\left.\times(-1)^{l_b-m_b}\langle L_B L_A|G(j_t)|l_b l_a\rangle\right]\ .\end{aligned} \tag{41}$$

Comparison of this relation with the inverse of (22) identifies the quantity in square brackets in (41) as $a_{j_t m_t}(\theta)$; indeed, this relation motivated the definition of $a_{j_t m_t}(\theta)$ in (22). Having made this identification, the inversion which gives the dynamical parameters G in terms of $a_{j_t m_t}(\theta)$, paralleling (18)–(21), is immediate:

$$\begin{aligned}\langle L_B L_A|G(j_t)|l_b l_a\rangle=&\left[\frac{2l_a+1}{2j_t+1}\right]i^{l_b-l_a}\left[\frac{2l_a+1}{4\pi}\right]^{-1/2}\\&\times\sum_{m_b,m_t}\left[2\pi\int_{-1}^{1}d(\cos\theta)Y_{l_b\,-m_b}(\theta,0)a_{j_t m_t}(\theta)\right](-1)^{l_b-m_b}\langle l_a 0,l_b\,-m_b|j_t m_t\rangle\ .\end{aligned} \tag{42}$$

In the event that $L_A=0$, we have $L_B=j_t=L$, and (42) reduces to (21), as we would expect.

For purposes of comparison with theory, recall that calculations exploit the rotational invariance of the Hamiltonian by dealing with eigenstates of the squared total angular momentum, $|\mathbf{J}|^2$. This paper, however, has treated the target and projectile as separate entities, and has therefore presented the dynamical parameters G as functions of the angular momentum transfer $\mathbf{j}_t$. The advantage of the latter treatment lies in the limited range of the quantum number j_t, $|L_A-L_B|\le j_t\le L_A+L_B$, as opposed to the unlimited range of J.[1] Our method, then, need only extract a few $G(j_t)$ from collision data, as opposed to many $G(J)$. [Values of $G(j_t)$ for large l_a and l_b are again to be found using the Born approximation, as discussed in Sec. II C.] The $G(J)$ are, nevertheless, obtained from the inverse relation to (40), namely,

$$\langle L_B l_b|G(J)|L_A l_a\rangle=\sum_{j_t}\langle L_B L_A|G(j_t)|l_b l_a\rangle(2j_t+1)\times\begin{Bmatrix} L_A & l_a & J\\ l_b & L_B & j_t\end{Bmatrix}. \tag{43}$$

IV. COMMENTS ON EXTENSIONS

We discuss here the several restrictions on the treatments of Secs. II and III that were listed in Sec. I, as an introduction to their removal in future works.

(a) *Targets with half-integer angular momentum.* Replacement of the integral angular momentum indices (L,L_A,L_B) with alternative notations (J,J_A,J_B), whose values may be integral or half-integral, affects none of the equations pertaining to the target amplitude parameters alone. The J indices are understood here to be either all integral or all half-integral, whereby j_t and m_t are integers. Half-integer values of J_B-J_A occur only upon change in the number of target constituents, the subject of item (h) below.

(b) *Projectiles with spin.* The indication of incidence and scattering directions by $(\hat{\mathbf{p}}_a,\hat{\mathbf{p}}_b)$ is complemented in this case by spin indices (m_{sa},m_{sb}). The harmonic functions $Y_{lm}(\theta,\phi)$ in (15) and thereafter, are then replaced by the transformation functions of Euler angles $D^j_{mm'}(\phi,\theta,\psi)$, properly normalized. Laying $\hat{z}$ along the incidence and $\hat{x}$ in the scattering plane sets ϕ and ψ to zero.

(c) *Parity under coordinate inversion.* Conservation of parity in the transition of the projectile plus target system from its initial to its final state is a feature of atom-molecular processes. The parity of projectile states is analyzed by harmonic expansion. The parity of the whole system is then represented by the parity of l_a+L_A and of l_b+L_B in the present notation. Analysis of collisions in terms of the angular momentum transfer j_t classifies them as "parity favored (unfavored)" when $L_A+L_B+j_t$ and $l_a+l_b+j_t$ are even (odd) (see, e.g., Sec. 7.10.3 of Ref. 7). Processes involving only single particle transitions from one to another eigenstate of orbital momentum are parity favored. (Reference 1 was tacitly restricted to these cases.) The mechanism of parity unfavored transitions involves a vector product, a spin flip, or analogous operations, which exclude, e.g., projectile scattering at $\theta\to0°$ (Ref. 10). Effects of parity unfavoredness may combine with the permutation symmetries of Wigner coefficients to restrict the magnitudes of polarization parameters.

(d) *Exchange effects.* The identity of electrons or of nucleons requires analysis of each collision process into alternative symmetry classes, except for the cases of closed shells or analogous features to which this paper has been restricted. The analysis of projectile-target systems into such symmetry classes (see, e.g., Sec. 7.2 of Ref. 7) leads the amplitude parameters a, g, and G of Secs. II and III to be labeled by additional invariant symmetry indices, typically by the total spin S label for electron-atom collisions.

(e) *"Mixed" states.* The treatment of Secs. II and III hinges on maximum information being provided on the initial and final states of both projectile and target. This condition is embodied in the unit normalization of the target polarization parameters and in the assumption that any projectile spin remains unaffected by the collision. Failure of this condition forces on the matrix τ in Sec. III B the condition $\mathrm{Tr}(\tau^2)<1$, in which case τ no longer factors as a dyadic product of eigenvectors $a^*_{j_t'm_t'}a_{j_tm_t}$. The matrix τ can still be represented as an incoherent superposition of eigenvector dyadics $\sum_n p_n(a^{(n)}_{j_t'm_t'})^*a^{(n)}_{j_tm_t}$, whose p_n are nonzero eigenvalues of τ, but the information embodied in τ fails to determine the *phase differences* of the eigenvectors $a^{(n)}_{j_tm_t}$, thus restricting the knowledge of dynamical parameters.

(f) *General scattering partners.* Even though the immediate motivation of this paper stemmed from electron-atom collisions such as (1), its development appears readily adaptable to any collision of electrons, ions, atoms, or molecules which preserves the integrity of projectile and target.

(g) *Nonstationary states.* We have dealt here explicitly with collisions that excite a target from one to another stationary state. Excitations to states with a fine structure, or to quasidegenerate levels, often do not afford energy resolution of individual stationary states; the final state is then nonstationary and its polarization displays quantum beats.[11] Extension of the treatment of Sec. III to allow for nonstationary excitations is favored by the flexibility, e.g., of Eq. (27), where pairs of identical L_B indices could be replaced by unequal pairs (L_B,L_B') to represent interference effects between alternative final states. Procedures of Ref. 11 extract from quantum beat observations the *initial* values of polarization parameters $\langle T^{K_B}_{Q_B}\rangle_{\rho_A\theta}$; these are *independent* of fine structure quantum numbers that become relevant later.

(h) *Reactive collisions.* Collisions involving transfer of constituent particles between projectile and target proceed through the formation and fragmentation of a "complex," a process that exceeds the scope of this paper.

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*Present address: Department of Mathematics, Yale University, New Haven, CT 06520.

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