Spin-Dependent Orientation Propensities Revealed in Polarized-Electron–Polarized-Photon Coincidence Studies

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The density matrix parametrization of collisionally excited atomic ensembles is generalized to account for a projectile spin. The elements are related to the "generalized Stokes parameters" of Andersen and Bartschat [J. Phys. B **27**, 3189 (1994)], determined in scattered-projectile–polarized-photon coincidence experiments after impact excitation by spin-polarized electrons. The well-established orientation propensity rule for unpolarized electron beam experiments is *not* valid for spin-resolved collisions.

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The traditional object of electronic and atomic collision studies is a total or differential cross section. Usually, these observables are the result of averages over key variables, such as impact parameter, magnetic quantum numbers, or electron spin. The averaging, however, may partly or completely obscure the collision dynamics responsible for the process, and makes comparison between theory and experiment less valuable. For many fundamental processes the focus of investigation has, therefore, increasingly been concentrated on identifying dimensionless quantities which can be derived from relative intensity measurements. The ultimate goal, first clearly formulated by Bederson [1], is the *perfect scattering experiment* which determines all the quantum mechanical, complex scattering amplitudes.

The study of such quantities, termed alignment and orientation parameters, has by now reached a very high level of sophistication. Although the goal of *perfect* has presently been achieved in a few cases only, this approach has dramatically increased our understanding of the collision dynamics for a broad range of collision processes, such as charge transfer in energetic collisions involving singly [2] or multiply [3] charged ions, and thermal collisions involving atoms with low [4] or high [5] level of excitation. Ever since the pioneering discussion by Kohmoto and Fano [6], a parameter of particular interest has been the orientation which describes the sense of circulation of the active electron around the atomic core. Propensity rules for orientation have been formulated and discussed for heavy particle [7] and electron [8] impact excitation. Recent efforts [9] have concentrated on exploring the generality of these rules [10,11].

For electron impact excitation at small scattering angles, a general observation is that the orientation vector, i.e., the transferred electronic orbital angular momentum $\mathbf{L}_\perp$ points in the direction of $\mathbf{k}_{\rm in} \times \mathbf{k}_{\rm out}$ where $\mathbf{k}_{\rm in}$ and $\mathbf{k}_{\rm out}$ are the linear momenta of the incoming and outgoing electron, respectively [12]. The development of efficient methods to produce beams of spin-polarized electrons unraveled spin-dependent alignment and orientation parameters. For light atoms, such studies were pioneered at NIST [13]. They showed that the propensity also holds true for spin-resolved orientations in sodium.

We report spin-resolved studies of heavy atom excitation which reveal that this propensity may be violated. In this case the analysis is complicated due to the many independent scattering amplitudes [14,15]. Introducing so-called "generalized Stokes parameters," two of us [16] showed how the description of the excitation process in terms of scattering amplitudes defined in the "natural coordinate" system (where the z axis is taken perpendicular to the scattering plane, and the incident beam direction defines the x axis) enables the solution of the nonlinear equations for determination of these amplitudes.

Below we provide what has been, to date, the missing link between the "generalized Stokes" parameters, the density matrix, and the spin-dependent coherence parameters that describe the excited atomic ensemble. The general structure of the new density matrix decomposition is valid well beyond our special case of interest, electron impact excitation of mercury. The concept of "generalized Stokes" parameters can be transferred directly to any experimental situation where two (or more) spin or light polarizations are prepared or analyzed.

For the case of unpolarized incident electrons, it is well known [12] how the set of (relative) parameters $(L_\perp^+, \gamma, P_\ell^+, h)$, representing the angular momentum transfer, the alignment angle, the degree of linear polarization, and the height of the charge cloud, can be determined in scattered-electron–polarized-photon coincidence experiments by measuring the set of Stokes parameters (P_1, P_2, P_3) with a photon detector *perpendicular* to the scattering plane, and the linear polarization P_4 with a photon detector *in* the scattering plane.

The generalization of the density matrix representation given in [12] to the case of polarized electron beams

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is a pair of density matrices, one for spin-up and one for spin-down electron impact excitation where "up" (↑) and "down" (↓) correspond to the initial spin projection with respect to the scattering plane. Here we restrict the discussion to a spin polarization perpendicular to the scattering plane and decompose the density matrix for unpolarized (subscript "u") beam excitation as

$$\rho_u = \sigma_u \frac{(1-h)}{2} \begin{pmatrix} 1+L_\perp^+ & 0 & -P_\ell^+ e^{2i\gamma} \\ 0 & \frac{2h}{1-h} & 0 \\ -P_\ell^+ e^{-2i\gamma} & 0 & 1-L_\perp^+ \end{pmatrix} = w^\uparrow \rho^\uparrow + w^\downarrow \rho^\downarrow \quad (1)$$

with

$$\frac{\rho^\uparrow}{\sigma_u} = \frac{(1-h^\uparrow)}{2} \begin{pmatrix} 1+L_\perp^{+\uparrow} & 0 & -P_\ell^{+\uparrow} e^{2i\gamma^\uparrow} \\ 0 & \frac{2h^\uparrow}{1-h^\uparrow} & 0 \\ -P_\ell^{+\uparrow} e^{-2i\gamma^\uparrow} & 0 & 1-L_\perp^{+\uparrow} \end{pmatrix}, \quad (2)$$

and similarly for $\rho^\downarrow$, where

$$\sigma_u = (\sigma^\uparrow + \sigma^\downarrow)/2 = (w^\uparrow + w^\downarrow)\sigma_u \quad (3)$$

is the cross section for unpolarized electron scattering.

The following relationships hold:

$$L_\perp^{+\uparrow,\downarrow} = -P_3^{\uparrow,\downarrow}, \quad (4)$$

$$P_\ell^{+\uparrow,\downarrow} e^{2i\gamma^{\uparrow,\downarrow}} = P_1^{\uparrow,\downarrow} + iP_2^{\uparrow,\downarrow}, \quad (5)$$

$$(1-h)L_\perp^+ = w^\uparrow(1-h^\uparrow)L_\perp^{+\uparrow} + w^\downarrow(1-h^\downarrow)L_\perp^{+\downarrow}, \quad (6)$$

$$(1-h)P_\ell^+ e^{2i\gamma} = w^\uparrow(1-h^\uparrow)P_\ell^{+\uparrow} e^{2i\gamma^\uparrow} + w^\downarrow(1-h^\downarrow)P_\ell^{+\downarrow} e^{2i\gamma^\downarrow}, \quad (7)$$

$$h = w^\uparrow h^\uparrow + w^\downarrow h^\downarrow. \quad (8)$$

Consequently, the maximum set of *nine* dimensionless independent parameters that can be extracted from the radiation pattern is given by

$$(L_\perp^{+\uparrow}, L_\perp^{+\downarrow}, \gamma^\uparrow, \gamma^\downarrow, P_\ell^{+\uparrow}, P_\ell^{+\downarrow}, h^\uparrow, h^\downarrow, w^\uparrow), \quad (9)$$

i.e., spin-resolved angular momentum transfers, alignment angles, degrees of linear polarization, and height parameters, as well as a probability parameter ($w^\uparrow$) that determines the relative importance of spin-up and spin-down scattering. The total degrees of polarization $P^{+\uparrow,\downarrow} = |\mathbf{P}^{+\uparrow,\downarrow}|$ must be unity for initial target states with total electronic angular momentum $J_0 = 0$, provided depolarization effects due to fine- and hyperfine-structure interaction in the target can be neglected. In such a case of *pure* initial states, the two relationships

$$(L_\perp^{+\uparrow,\downarrow})^2 + (P_\ell^{+\uparrow,\downarrow})^2 = 1 \quad (10)$$

reduce the number of independent parameters to *seven*.

A determination of the parameter set (10) does *not* correspond to a *perfect experiment*, since three relative phases of the scattering amplitudes remain unknown [16]. Their determination requires additional experimental setups discussed in a forthcoming review [17]. Here we concentrate on the above set, which forms the spin-resolved analog to the parameter set for unpolarized beams. These parameters can be extracted from a measurement of the "generalized Stokes parameter matrix" whose elements $(Q_{ij}^{\hat{\mathbf{n}}})_{\mathbf{P}}$ are defined as follows (see Fig. 1): With a photon detector placed in the $\hat{\mathbf{n}}$ direction, four light intensities are measured for orthogonal positions of the light polarization analyzers and electron beam polarizations $\pm\mathbf{P}$. Following [16], we define for $(0°, 90°)$:

$$I_u^{\hat{\mathbf{n}}}(Q_{11}^{\hat{\mathbf{n}}})_{\mathbf{P}} \equiv I_{\mathbf{P}}^{\hat{\mathbf{n}}}(0°) + I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(0°) - I_{\mathbf{P}}^{\hat{\mathbf{n}}}(90°) - I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(90°), \quad (11)$$

$$I_u^{\hat{\mathbf{n}}}(Q_{12}^{\hat{\mathbf{n}}})_{\mathbf{P}} \equiv I_{\mathbf{P}}^{\hat{\mathbf{n}}}(0°) - I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(0°) - I_{\mathbf{P}}^{\hat{\mathbf{n}}}(90°) + I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(90°), \quad (12)$$

$$I_u^{\hat{\mathbf{n}}}(Q_{13}^{\hat{\mathbf{n}}})_{\mathbf{P}} \equiv I_{\mathbf{P}}^{\hat{\mathbf{n}}}(0°) - I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(0°) + I_{\mathbf{P}}^{\hat{\mathbf{n}}}(90°) - I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(90°), \quad (13)$$

where

$$I_u^{\hat{\mathbf{n}}} \equiv I_{\mathbf{P}}^{\hat{\mathbf{n}}}(0°) + I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(0°) + I_{\mathbf{P}}^{\hat{\mathbf{n}}}(90°) + I_{-\mathbf{P}}^{\hat{\mathbf{n}}}(90°). \quad (14)$$

Similarly, we define $(Q_{2j}^{\hat{\mathbf{n}}})_{\mathbf{P}}$ and $(Q_{3j}^{\hat{\mathbf{n}}})_{\mathbf{P}}$, $j = \{1, 2, 3\}$, by replacing $(0°, 90°)$ with $(45°, 135°)$ and, for circular polarization analysis, (σ^-, σ^+). Such "generalized Stokes" parameters can be defined for any optical transition excited by spin-polarized beams.

For photon detection perpendicular to the scattering plane ($\hat{\mathbf{n}} = \hat{\mathbf{z}}$), the generalized Stokes matrix can be expressed in terms of the density matrix parameters as

$$I_u^{\hat{\mathbf{z}}}\left(Q_{ij}^{\hat{\mathbf{z}}}\right)_{P_z} = \frac{3}{2} \begin{pmatrix} w^\uparrow(1-h^\uparrow)P_1^\uparrow + w^\downarrow(1-h^\downarrow)P_1^\downarrow; & w^\uparrow(1-h^\uparrow)P_1^\uparrow - w^\downarrow(1-h^\downarrow)P_1^\downarrow; & \frac{2}{3}[w^\uparrow(1-3h^\uparrow) - w^\downarrow(1-3h^\downarrow)] \\ w^\uparrow(1-h^\uparrow)P_2^\uparrow + w^\downarrow(1-h^\downarrow)P_2^\downarrow; & w^\uparrow(1-h^\uparrow)P_2^\uparrow - w^\downarrow(1-h^\downarrow)P_2^\downarrow; & \frac{2}{3}[w^\uparrow(1-3h^\uparrow) - w^\downarrow(1-3h^\downarrow)] \\ w^\uparrow(1-h^\uparrow)P_3^\uparrow + w^\downarrow(1-h^\downarrow)P_3^\downarrow; & w^\uparrow(1-h^\uparrow)P_3^\uparrow - w^\downarrow(1-h^\downarrow)P_3^\downarrow; & \frac{2}{3}[w^\uparrow(1-3h^\uparrow) - w^\downarrow(1-3h^\downarrow)] \end{pmatrix}, \quad (15)$$

with $I_u^{\hat{\mathbf{z}}} = \frac{3}{2}(1-h)$. Similar expressions hold for observation directions $\hat{\mathbf{n}} = \hat{\mathbf{y}}, \hat{\mathbf{x}}$ in the scattering plane [17].

The first column $(Q_{i1}^{\hat{\mathbf{z}}})_{P_z}$ is the standard Stokes vector (P_1, P_2, P_3) for unpolarized incident electrons. The third

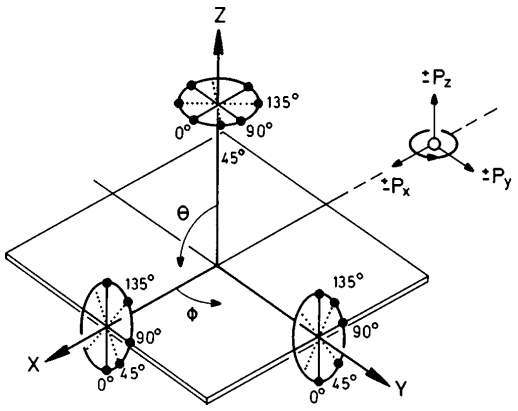

FIG. 1. Frame definition for generalized Stokes parameters.

column $(Q^{\hat{\mathbf{z}}}_{i3})_{P_z}$ corresponds to an "optical asymmetry" which compares light intensities measured with spin-up and spin-down electrons, independent of the light analyzer setting. Combining the first two columns according to

$$C_i^{\uparrow,\downarrow} = [(Q^{\hat{\mathbf{z}}}_{i1})_{P_z} \pm (Q^{\hat{\mathbf{z}}}_{i2})_{P_z}]/2, \tag{16}$$

one finds $w^{\uparrow,\downarrow}(1 - h^{\uparrow,\downarrow})\mathbf{P}^{\uparrow,\downarrow}/(1 - h)$ where $\mathbf{P}^{\uparrow,\downarrow} = (P_1^{\uparrow,\downarrow}, P_2^{\uparrow,\downarrow}, P_3^{\uparrow,\downarrow})$ are the spin-resolved Stokes vectors.

Spin-dependent alignment angles are obtained from

$$2\gamma^{\uparrow,\downarrow} = \arg(C_1^{\uparrow,\downarrow} + i\, C_2^{\uparrow,\downarrow}), \tag{17}$$

and angular momentum transfers from

$$L_\perp^{+\uparrow,\downarrow} = -C_3^{\uparrow,\downarrow}. \tag{18}$$

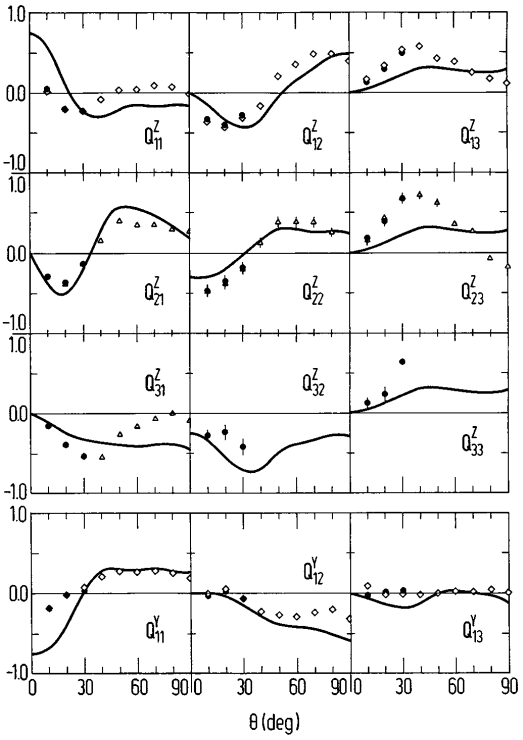

FIG. 2. Generalized Stokes parameters for electron impact excitation of $Hg(6s6p)^3P_1$ at 8 eV. See text for symbols.

The lengths of the two vectors $\mathbf{C}^{\uparrow,\downarrow}$ are the quantities $w^{\uparrow,\downarrow}(1 - h^{\uparrow,\downarrow})/(1 - h)$. Additional measurement of $(Q^{\hat{\mathbf{y}}}_{11})_{P_z} = -P_4$ [16] determines the height parameter h and subsequently the set $(w^\uparrow, h^\uparrow, h^\downarrow)$.

We have used experimental data from [18] (diamonds), [19] (triangles), and this work (circles) to extract the nonzero "generalized Stokes" parameter matrix elements for the $\hat{\mathbf{z}}$ and $\hat{\mathbf{y}}$ directions for electron impact excitation of the $(6s^2)^1S_0 \rightarrow (6s6p)^3P_1$ transition in Hg. Results for an incident electron energy of 8 eV are shown in Fig. 2, together with theoretical results based on a five-state Breit-Pauli R-matrix calculation [20], which is still the benchmark theory for this collision system. The agreement between experiment and theory is satisfactory, keeping in mind the complexity of the collision problem and the level of detail in the comparison. The experimental data for $(Q^{\hat{\mathbf{z}}}_{13})_{P_z}$, $(Q^{\hat{\mathbf{z}}}_{23})_{P_z}$, and $(Q^{\hat{\mathbf{z}}}_{33})_{P_z}$, which should be identical within the error bars, are not completely consistent, although a tendency toward the same result is recognized.

The corresponding spin-averaged and the new spin-resolved coherence parameters are presented in Fig. 3. We corrected for hyperfine-structure depolarization effects by first extracting state multipoles from the measured light polarizations and then recalculating the unperturbed radiation pattern. The vertical lines in Fig. 3 represent the scatter due to alternative ways of extracting the set (10), which is overdetermined by the 12 parameters of Fig. 2. Details are given in [17].

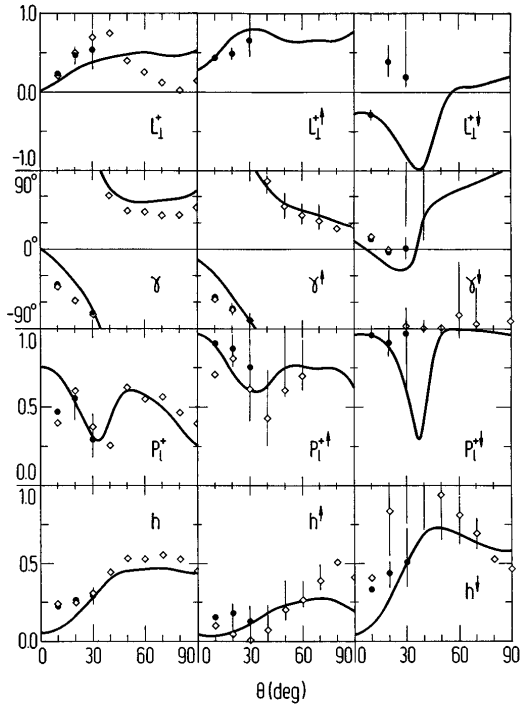

FIG. 3. Spin-averaged and spin-resolved coherence parameters for electron impact excitation of $Hg(6s6p)^3P_1$ at 8 eV. The error bars on the experimental data from [18,19] (◇) and the present work (●) reflect statistics and consistency tests.

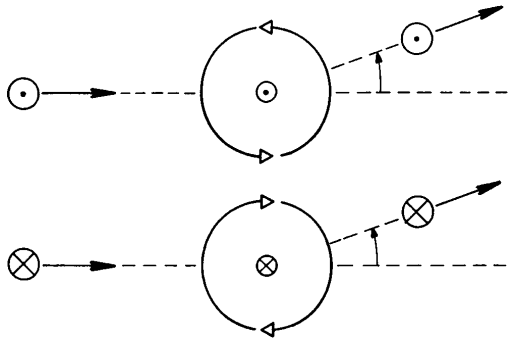

FIG. 4. Illustration of angular momentum transfer by spin-"up" (top) and spin-"down" (bottom) electrons for electron impact excitation of $Hg(6s6p)^3P_1$ at small scattering angles for an incident electron energy of 8 eV. The upper process is more likely to occur than the lower one.

The coherence parameters are very different for the two spin directions. Note that at $\theta = 10°$, $L_\perp^{+\uparrow} > 0$ while $L_\perp^{+\downarrow} < 0$. The positive value of $L_\perp^{+\uparrow}$ agrees with well-established propensity rules [12], while a *negative* value of $L_\perp^{+\downarrow}$ may seem surprising. However, both $L_\perp^{+\uparrow}$ and $L_\perp^{+\downarrow}$ may be nonzero for forward scattering, but $L_\perp^{+\uparrow}(0°) = -L_\perp^{+\downarrow}(0°)$ by symmetry requirements. The corresponding spin-resolved version of the classic Kohmoto-Fano diagram [6] is shown in Fig. 4.

The spin-resolved alignment angles $\gamma^\uparrow$ and $\gamma^\downarrow$ show no similarities, with the directions of the two major axes often perpendicular to each other [note that $\gamma^\uparrow(0°) = -\gamma^\downarrow(0°) \neq 0$]. Finally, there is a large difference between the height parameters $h^\uparrow$ and $h^\downarrow$, with the theory predicting a maximum value of $h^\downarrow \approx 75\%$ near a scattering angle of 40° while $h^\uparrow \approx 25\%$. Spin flips are thus very likely for spin-"down" electrons, but those spin-"down" electrons whose spin is not flipped tend to transfer a *negative* angular momentum to the atom.

In conclusion, we introduce a new spin-resolved density matrix parametrization to describe electron impact excitation by spin-polarized electron beams, with an explicit prescription to determine the spin-resolved parameters that generalize the well-known set for unpolarized beams. For polarized electron impact excitation of $Hg(6s6p)^3P_1$ at 8 eV, a large spin-flip probability for spin-"down" electrons was found, and opposite values for the orientation in the two spin channels. Consequently, the well-established propensity rule of positive values for the orientation parameter at small scattering angles holds only for the *spin-averaged* parameter. We hope that these results will stimulate further work in this area, to elucidate the physics of the collision process and to further explore the limits of applicability of propensity rules.

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