ALIGNMENT AND ORIENTATION OF ATOMIC OUTER SHELLS INDUCED BY ELECTRON AND ION IMPACT: SOME RECENT DEVELOPMENTS AND REMAINING PROBLEMS

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Alignment and orientation of atoms in collision experiments with planar symmetry have now been studied for about 15 years and close to 500 papers have been produced, mainly devoted to S+P excitation. Despite the large variety of electron-atom, ion-atom and atom-atom collision systems considered, a unified framework for description of these phenomena is now emerging. This framework is a generalization of the original ideas of Macek and Jaecks and is based on consideration of symmetries, conservation laws, etc. The key parameters are directly related to the shape and dynamics of the charge cloud of the excited electron as well as to experimental observables. A brief review is given of this framework, and some current problems and prospects for the future are discussed.

1. INTRODUCTION

The field of alignment and orientation in atomic collisions is devoted to the study of the shape and dynamics of the electronic charge clouds excited in a collision process. Obviously this kind of information, most effectively obtained from experiments with planar symmetry, provides a much more severe test of our understanding of the excitation mechanisms than determination of, say, a probability or a cross section for excitation. As will be detailed below, in favorable cases a complete determination of the quantum-mechanical state of the system may be obtained, thereby providing a so-called "perfect scattering experiment," the most fundamental level at which experiment and theory can be compared. The shape and dynamics are expressible in dimensionless parameters based on relative measurements, thereby eliminating the otherwise often serious problem of accurate determination of absolute units. Furthermore, they may be very sensitive to details in the theoretical description in cases where the cross sections show only minor variations.

At previous ICPEAC's several symposia (1,2) review papers (3,4) and progress reports (5), and a large number of contributed papers have dealt with various particular aspects of this field. The Data Center of the Joint Institute for Laboratory Astrophysics is currently undertaking a critical review of this whole flourishing field, restricted to excitation of outer shells of atoms in planar scattering experiments using unpolarized beams. During this review several parallel lines of thought within the otherwise traditionally separated fields of electron-atom and atom-atom collisions became evident. In

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particular a unified framework, or language for description of these phenomena, is developing though several "local dialects" still exist and probably will persist, partly due to differences in nature of the underlying physics for specific problems. Below we shall first try to summarize this framework, mainly concentrating on $S\rightarrow P$ excitation to which about 95% of the literature devoted. Then some selected current problems will be discussed within this framework and conclusions drawn concerning areas where future efforts might be most fruitfully concentrated. The discussion will be restricted to excitation of states decaying by photon emission, though most of the ideas can be easily modified to include electron emission as well, cf. Niehaus (6).

2. FRAMEWORK

2.1. Coordinate frames, basis functions, symmetries and time scales

In an experiment, the collision plane is determined by e.g. the two directions of incoming and outgoing particles, thereby fixing the scattering angle. Excitation processes corresponding to this scattering angle are then studied by analysis of either the polarization properties or the angular distribution of the secondary photons emitted when the excited state decays, detected in coincidence with the scattered particle, or, in time-reversed experiments, analysis of the scattered particles as function of the polarization properties of the laser light used to prepare the target atoms prior to the collision. These procedures are detailed in recent reviews (7-9), so we shall just briefly outline the principles here.

Since the symmetry properties of the force(s) determining the excitation and time evolution of the charge cloud will be seen to play an essential role, we shall recall the symmetry properties of the three eigenstates corresponding to the P-jets, neglecting at first the effect of fine and hyperfine structure, which will be included later. The upper panel of Fig. 1 shows the angular parts of the charge clouds for the so-called "atomic physics basis" (\(|p_1\rangle, \langle p_2\rangle\), \(|p_0\rangle\), with \(|p_0\rangle\) labelled according to the magnetic quantum number M. Here we quantize along the axis perpendicular to the collision plane since this choice simplifies the subsequent mathematical description considerably. The lower panel of Fig. 1 shows the alternative "molecular physics basis" corresponding to real-valued wave functions \((\langle px\rangle, \langle py\rangle, \langle pz\rangle)\), describing p-orbitals along the three coordinate axes. The state \(|p_0\rangle = |p_z\rangle\) has negative reflection symmetry with respect to this plane, while the other ones have positive reflection symmetry.

Notation for circularly polarized light varies in the literature. We here use the definition of classical optics (10), which uses the term left hand-circularly (LHC) polarized light, if the electric vector is seen to rotate counter-clockwise when looking toward the light source, i.e. LHC-photons have positive helicity. Referring to Fig. 1, decay of the state \(|p_z\rangle\) will thus lead to emission of LHC-photons in the +z (RHC-photons in the -z) direction.

At this point it is useful to recall some important time scales:

(i) \(\tau_c\), the collision time, during which the excitation takes place; \(\tau_c = a/v\), where a is a characteristic interaction length and v the collision velocity.

(ii) \(\tau_{fs}\), the characteristic time for the fine structure; \(\tau_{fs} = 1/\Delta u_{fs}\), where \(\Delta u_{fs}\) is the fine structure splitting.

(iii) \(\tau_{hs}\), the characteristic time for the hyperfine structure; \(\tau_{hs} = 1/\Delta u_{hs}\), where \(\Delta u_{hs}\) is the hyperfine structure splitting.

(iv) \(\tau_{nat}\), the (natural) lifetime; \(\tau \sim 1/A\), where A is the decay probability.

(v) \(\tau_{obs}\), the observation time for an atom in the actual experimental setup.

Each specific situation requires consideration of these magnitudes. An atom that is initially in an S state has positive reflection symmetry.
The total wave function describing the collision event conserves its reflection symmetry with respect to the scattering plane. Often fine and hyperfine forces are so weak, they can be neglected during the collision so that the excitation is caused by Coulomb interaction only. In the simple case where the collision partner acts as a spinless, structureless particle, this implies that the spatial part of the wave function of the atom that is excited preserves its reflection symmetry during the collision. Thus excitation of \( |p_0\rangle \) is forbidden.

After the collision, the isolated, excited atom may develop further under the influence of the fine structure (and possibly hyperfine structure) force, which does not conserve reflection symmetry of the spatial part of the wave function, thereby allowing the charge cloud to change shape in time until the decay takes place. We shall treat these two regimes -- excitation and time development -- separately, and see how it is possible to reconstruct the nascent charge cloud, the object of interest to collision physics, from the actual observed radiation pattern and a knowledge of \( \tau_{fs}, T_{fs}, T_{nat} \) and \( T_{obs} \).

2.2. S-P excitation

2.2.1. The simplest case: Full coherence -- the Poincaré sphere

We shall first analyze the properties of the radiation pattern in the simplest case in which the excited P-level of the atom can be described by a state vector, i.e., its coordinates \( (a_x, a_y, a_z) \) in the basis of Fig. 1. Reflection symmetry conservation implies \( a_0 = 0 \). This situation may be encountered in, e.g., electron impact excitation of a He(nP) level.

Figure 2(a) shows an example of the angular part of the P-state electron density. Three coordinate frames are shown: (i) the collision frame \( (x^C, y^C, \)
$z^c$) with $z^c$ in the direction $k_{in}$ of the incident particles, $(x^c, z^c)$ is the collision plane, and $y^c$ is parallel to $k_{in} \times k_{out}$. $\theta_{CD}$ is the scattering angle; (iii) the natural frame $(x^0, y^0, z^0)$ with $x^0\parallel x^c$, $y^0\parallel y^c$, $z^0\parallel z^c$; finally, another convenient frame (iii) is the atomic frame $(x^a, y^a, z^a)$ obtained from the natural frame by rotation through an angle $\gamma$ around $z^a = z^0$ so that $x^a$ parallels the major symmetry axis of the charge cloud, which is also a symmetry axis for the radiation pattern. Assuming normalization, $a_1^2 + a_2^2 = 1$, then, apart from an arbitrary common phase factor, the wave function is completely characterized by the two parameters $(\gamma, L_z)$, where $\gamma$ is the alignment angle and $L_z = a_1^2 - a_2^2$ is the angular momentum, pointing along $z^a$. In the atomic frame the wave function takes the simple form

$$|\psi\rangle^a = z^{-1/2} (L_z)^{1/2} [p_{+1}]^a - (L_z)^{1/2} [p_{-1}]^a \) \) . (1)
Using polar coordinates \((\theta, \phi)\) in the natural frame, the angular part \(T\) of the charge cloud may be written as (omitting a \(3/4\pi\) normalization factor)

\[
T(\theta, \phi) = \frac{1}{2} \left[ 1 + \frac{P_k}{P} \cos 2(\theta - \gamma) \right] \sin^2 \theta
\]  

(2)

Figure 2(b) shows a cut through the charge cloud in the collision plane, given by

\[
T(\theta, \phi) = \frac{1}{2} \left[ 1 + \frac{P_k}{P} \cos 2(\theta - \gamma) \right]
\]  

(3)

\(P_k\), the linear polarization, is thus a width parameter, with \((1+P_k)/2\) and \((1-P_k)/2\) determining the major and minor axes of the charge cloud in the collision plane.

The angular correlation pattern of the photons emitted from the excited atoms and detected in the collision plane is given by \((\theta = \pi/2)\)

\[
1(\phi) = 1 - \frac{P_k}{P} \cos 2(\phi - \gamma)
\]  

(4)

This pattern, Fig. 2(c), is thus identical to the shape of the charge cloud, Eq. (3) and Fig. 2(b), rotated by 90°, as expected from the properties of electric dipole radiation.

Alternatively, \(\gamma\) may measure the polarization ellipse observed in the \(+z^0\) direction with a linear and circular polarizer, Fig. 2(d), or equivalently, the three Stokes parameters \((P_1, P_2, P_3)\) defined by

\[
\begin{align*}
P_1 & = 1(0^\circ) - 1(90^\circ) \\
1-P_2 & = 1(45^\circ) - 1(135^\circ) \\
1-P_3 & = 1(RHC) - 1(LHC)
\end{align*}
\]

where \(I\) is the total light intensity in the \(z^0\) direction, and \(I(\theta)\) is the light transmitted through an ideal linear polarizer tilted at an angle \(\theta\) with respect to \(z^0\) [sometimes one may find the alternative notation \((n_1, n_2, n_3) = (P_2, P_3, P_1)\)]. Evidently

\[
\begin{align*}
\gamma & = (P_1, P_2, P_3) = (P_k \cos 2\gamma, P_k \sin 2\gamma, -L_k)
\end{align*}
\]

(5)

with the linear polarization \(P_k = (P_1^2 + P_2^2 + P_3^2)^{1/2}\) being invariant under rotation around \(z^0\). Furthermore, the light emitted is fully coherent, such that the degree of polarization

\[
P = |\gamma|^2 = (P_1^2 + P_2^2 + P_3^2)^2
\]

(6)

in this case is unity, and \(P_k\) thus fully determined from \(L_k\).

The Stokes vector \((P_1, P_2, P_3)\) measured in the \(y^0 z^0\) direction is \((1, 0, 0)\) and adds no further information in this case.

A correlation experiment, Fig. 2(c), determines \((\gamma, P_k)\) while a coherence experiment, Fig. 2(d), determines \((\gamma, L_k)\). Thus, here, a coherence experiment is a "perfect scattering experiment" in the sense discussed in the introduction, while a correlation experiment only determines \(P_k\), leaving the sign of \(L_k\) undetermined. The parameters used in Fig. 2 are \(\gamma = 35^\circ, L_k = 0.8\) and \(P_k = 0.6\).

For later generalization we also state the density matrix \(\rho_{nm} = \rho_{nm}\) in the natural frame \((\theta = 1)\)

\[
\begin{align*}
\begin{bmatrix}
\rho_{11} & 0 & \rho_{1-1} \\
0 & \rho_{00} & 0 \\
\rho_{-11} & 0 & \rho_{-1-1}
\end{bmatrix} &= \frac{1}{2} \begin{bmatrix}
1-P_3 & 0 & -P_1^*P_2 \\
0 & 0 & 0 \\
-P_1^*P_2 & 0 & 1-P_3
\end{bmatrix}
\end{align*}
\]

(7)
A succinct way of summarizing this situation is by introducing the Poincare sphere (11), making use of the fact that the point \( P = (P_1, P_2, P_3) \) is located on a unit sphere, cf. Eq. (6). Figure 3 illustrates the polarization ellipse corresponding to various points on the sphere, with \( H(1,0,0) \) corresponding to \( |p_H \rangle \) or horizontal linear polarization in Fig. 2(d), \( V(-1,0,0) \) corresponding to \( |p_V \rangle \) or vertical linear polarization, the north pole to \( |p_+ \rangle \), and the south pole to \( |p_- \rangle \). Two opposite points on the sphere correspond to orthogonal states. In this language, a coherence experiment determines a point on the Poincare sphere, while a correlation experiment only determines its projection on the equatorial plane, \( P_1 + iP_2 = P \cos \theta \).}

2.2.2. Loss of full coherence

Where several processes which are in principle distinguishable contribute, the excitation may no longer be coherent. For instance, in the low energy electron impact excitation of a \( ^1H(n^2p) \) level, exchange effects lead to different amplitudes for singlet and triplet scattering, and if no spin analysis is performed before or after the collision the corresponding density matrix elements have to be added incoherently. This situation can no longer be represented by a wave function. Positive reflection symmetry is still conserved. The light emitted in the \( +z^n \) direction is no longer fully coherent so that \( P(\psi) \). All Eqs. (2)-(7) remain valid. However, \( P_2 \) and \( L_\| - P_3 \) are now independent parameters. Thus, in this case three parameters are needed to specify the situation completely, namely \( (\gamma, L_\|, P_2) \). A correlation experiment only determines shape \( (P_2) \) and alignment angle \( \gamma \), but gives no information about dynamics \( (L_\|) \), while coherence analysis still provides complete information.

2.2.3. The general case

In the general case the assumption for the atomic wave function of positive reflection symmetry only cannot be maintained. This may happen, for example, in electron impact excitation of \( P \) levels of the heavy rare gases for which spin-orbit effects are so strong that they play a role during the collision \( (F_{S^o - T}) \), at least at low energies. Equation (7) must then be replaced by

\[
\begin{align*}
\begin{pmatrix}
\rho_{00}^n & \rho_{01}^n & \rho_{0-1}^n & \rho_{10}^n & \rho_{11}^n & \rho_{1-1}^n
\end{pmatrix}
&= \left(1 + \frac{P_2}{P_1 + iP_2}\right) \begin{pmatrix}
1-P_3 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
+ \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} + \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}.
\end{align*}
\]
Equation (8) shows the decomposition of the density matrix into two parts, one having positive and one negative reflection symmetry. While Eq. (5) still holds, Eq. (2) is replaced by

$$\rho(\theta,\phi) = (1-\rho_{00}) \frac{1}{2} \left[ 1 + P e^{i [2(\phi - \gamma)]} \right] \sin^2 \theta + \rho_{00} \cos^2 \theta . \quad (9)$$

$\rho_{00}$ is thus a height parameter, the determination of which requires observation from a direction different from $z^0$. Here and below we abbreviate $\rho_{00} = \rho_{00}^0 = \rho_{00}^1$. Within the positive reflection symmetry we define explicitly $P e^{i} = (P_1^+ + P_2^+)/2$, $L_1^+ = -P_3$, and $P = (P_1^2 + P_2^2 + P_3^2)/2 = (P_1^2 + P_2^2 + P_3^2)/2$. In the $y^0$ direction the Stokes parameters are now $(P_4,0,0)$ with $P_4 < 1$. Then

$$\rho_{00} = \frac{(1+P_1)(1-P_4)}{(1-P_1)(1-P_4)} \quad (10)$$

The angular momentum is given by

$$L_\perp = -P_3(1-\rho_{00}) = L_\perp(1-\rho_{00}) . \quad (11)$$

Similarly, in a correlation experiment, determination of $\rho_{00}$ requires observations from at least two $\phi$ angles. The angular distribution of the total intensity is given by

$$I(\theta,\phi) = (1-\rho_{00}) \frac{1}{2} \left[ 1 + \cos^2 \theta - P \cos \theta \sin \theta \right] + \rho_{00} \sin^2 \theta . \quad (12)$$

Replacing Eq. (4), Equation (12) may be written as

$$I(\theta,\phi) = B(a)[1 - A(a) \cos \theta \sin \theta] . \quad (13)$$

where

$$A(a) = \frac{(1-\rho_{00}) \cdot P \sin^2 \theta}{(1+\rho_{00}) \cdot (1-3\rho_{00}) \cdot \cos^2 \theta} . \quad (14)$$

The value of $A$ in the scattering plane is

$$A(\theta,\phi) = \frac{1-\rho_{00}}{1+\rho_{00}} . \quad (15)$$

i.e., a smaller amplitude than the amplitude $P_\perp$ for the corresponding coherence analysis along $z^0$, see Eq. (9). Equation (14) also implies that

$$A(\pi,\phi) = P_\perp(1-\rho_{00})/(3-\rho_{00}) . \quad (16)$$

Defining the ratio $R = A(\pi/4)/A(\pi/2)$ one obtains

$$P_{00} = (3R-1)/(R+1) . \quad (17)$$

The general case is thus described by four parameters $(\gamma, L_\perp, P_\perp, \rho_{00})$ of which the three first can be determined from coherence analysis along $z^0$. Notice that one may still have full coherence within the positive reflection symmetry, $P_\perp < 1$.

The situation is summarized in Fig. 4, which shows the shapes of the charge clouds and below, in comparable scales, cuts along the principal axes in the atomic frame. In both cases (a) and (b) the alignment angle is $35^\circ$ and the width parameter $P_\perp = 0.6$, but the height parameter in (a) is $\rho_{00} = 0$, i.e., positive reflection symmetry, while $\rho_{00} = 1/3$ in (b). The angular momentum has no influence on the shape, and $P_\perp$ can be anywhere in the region $-0.8 < P_\perp < 0.8$, so that $P_\perp < 1$. The parametrization suggested above is a natural development of
the semiclassical model put forward by Macek and Jaecks in Sec. III of their fundamental paper (12).

2.2.4. Relations to the Blum-Paixão parameters

Blum, Paixão and collaborators (13) were the first to realize and formulate a parametrization for the general case. Here we summarize their mathematics and give relations to the parameters defined above. Again the atomic basis of Fig. 1 is used, however this time quantized along $z^C-x^N$. The density matrix may be decomposed into two components with positive and negative reflection symmetry with respect to the scattering plane, expressible in terms of the Stokes parameters in the following way (tr $\rho$=1)

$$
\begin{align*}
\begin{pmatrix}
\rho_{11}^C & \rho_{10}^C & \rho_{1-1}^C \\
\rho_{01}^C & \rho_{00}^C & \rho_{0-1}^C \\
\rho_{-11}^C & \rho_{-10}^C & \rho_{-1-1}^C
\end{pmatrix}
= & \begin{pmatrix}
\frac{1}{2} (\rho_{11}^C - \rho_{-11}^C) & \rho_{10}^C & -\frac{1}{2} (\rho_{11}^C + \rho_{-11}^C) \\
\rho_{10}^* & 1-2 \rho_{11}^C & -\rho_{10}^* \\
-\frac{1}{2} (\rho_{11}^C - \rho_{-11}^C) & -\rho_{10}^C & \frac{1}{2} (\rho_{11}^C - \rho_{-11}^C)
\end{pmatrix}
\end{align*}
$$
\[
\begin{align*}
&\frac{1}{2} \begin{pmatrix} 1 & 0 & 1 \\
0 & 0 & 0 \\
1 & 0 & 1 \\
\end{pmatrix} + \frac{1}{\sqrt{2}} \begin{pmatrix}
\frac{1}{2} (1-P_1) & \frac{1}{\sqrt{2}} (-P_2+ip_3) & -\frac{1}{2} (1-P_1) \\
\frac{1}{\sqrt{2}} (P_2+ip_3) & 1+p_1 & \frac{1}{\sqrt{2}} (P_2+ip_3) \\
-\frac{1}{2} (1-P_1) & \frac{1}{\sqrt{2}} (P_2+ip_3) & \frac{1}{2} (1-P_1) \\
\end{pmatrix} \\
&\frac{1}{2} \begin{pmatrix} 1 & 0 & 1 \\
0 & 0 & 0 \\
1 & 0 & 1 \\
\end{pmatrix}
\end{align*}
\]

where \( \rho_{11}^C + \rho_{11}^C = \rho_{00}^C \) is determined by \( P_1 \) and \( P_2 \); cf. Eq. (10). Following Blum and Paixão we now define

\[
\begin{align*}
\lambda &= \rho_{00}^C \\
\bar{\chi} &= \arg(\rho_{10}^C) = -\arg(\rho_{01}^C) \\
\cos \Delta &= |\rho_{10}^C|/\rho_{00} \rho_{11}^{C^{1/2}} = |\rho_{01}^C|/\rho_{00} \rho_{11}^{C^{1/2}} \\
\cos \varepsilon &= -\rho_{11}^C / \rho_{00}^C = -\rho_{11}^C / \rho_{11}^C \\
\end{align*}
\]

so that

\[
\begin{align*}
(1-\rho_{00}^C) \cdot P_1 &= \frac{1}{2} \{ \lambda (2 \cos \varepsilon) - (1+\cos \varepsilon) \} \\
(1-\rho_{00}^C) \cdot P_2 &= -2 \{ \lambda (1-\lambda) \}^{1/2} \cdot \cos \Delta \cdot \cos \bar{\chi} \\
(1-\rho_{00}^C) \cdot P_3 &= 2 (\lambda (1-\lambda))^{1/2} \cdot \cos \Delta \cdot \sin \bar{\chi}
\end{align*}
\]

where \( \rho_{00}^C = (1-\lambda)(1-\cos \varepsilon)/2 \). Here \( 0\leq\lambda \leq 1 \), \( 0\leq\bar{\chi} \leq 2\pi \), \( 0\leq\cos \varepsilon \leq 1 \), \( -1\leq\cos \Delta \leq 1 \). We may require \( \bar{\chi} \leq 2\pi \) since \( \sin \bar{\chi} \) has no physical meaning. Similarly \( \cos \varepsilon \leq 1 \) since \( \cos \varepsilon \) has no meaning. If \( \rho_{00}^C = 0 \) then \( \varepsilon = 0 \).

2.3. Time evolution due to internal forces. Depolarization

2.3.1. The effect of fine structure

When the collision is over, the isolated atom develops under the influence of internal forces until the optical decay. Again, restricting ourselves to an excited \( P \) state, we shall first analyze the simplest case of electron spin \( S=1/2 \), and see how the shape and dynamics of the charge cloud changes in time, resulting in a modification of the observed radiation compared to the unperturbed case, \( S=0 \).

Consider first the shape. Referring to Fig. 4 it is most conveniently analyzed in terms of the molecular basis in the atomic frame for which the relevant density matrix elements at the time of excitation \( t=0 \) are given by

\[
\begin{align*}
\rho_{xx}^a(0) &= \frac{1}{2} (1+p_1^a) \\
\rho_{yy}^a(0) &= \frac{1}{2} (1-p_1^a) \\
\rho_{zz}^a(0) &= 0
\end{align*}
\]
which directly measure the relative length, width and height of the charge cloud. The shape now develops in time according to [see, e.g., Ref. (14)]

\begin{align*}
\rho_{xx}(t) &= \rho_{xx}(0) \cdot G_2 + \frac{1}{3} (G_0 - G_2) \\
\rho_{yy}(t) &= \rho_{yy}(0) \cdot G_2 + \frac{1}{3} (G_0 - G_2) \\
\rho_{zz}(t) &= 0 \cdot G_2 + \frac{1}{3} (G_0 - G_2)
\end{align*}

where \( G_0 = G_0(t) = 1 \) and \( G_2 = G_2(t) = 1/3 \cdot [1 + 2 \cos(\omega_{fs} t)] \); \( \hbar \omega_{fs} \) is the energy splitting between the fine structure levels \( 2P_{3/2} \) and \( 2P_{1/2} \). Thus, as long as we assume \( \rho_{00}(0) = 0 \), the height varies in time as

\begin{equation}
\rho_{00}(t) = \rho_{zz}(t) = \frac{2}{9} [1 - \cos(\omega_{fs} t)]
\end{equation}

independent of \( \omega_{xy}(0) \) and \( \omega_{yy}(0) \). For the simple shape discussed previously in Fig. 2 and Fig. 4(a) having \( P_x = 0.6 \), Fig. 5(a) shows the (reversible) change in time of the shape, where the five situations correspond to \( \omega_{fs} t = 0, \pi/2, \pi, 3\pi/2, 2\pi \), respectively, showing the quantum-beat phenomenon well known in e.g. beam-foil spectroscopy. Since the symmetry axes stay fixed in time the plot has been made for \( \gamma = 0^\circ \).

Most collision experiments only monitor the time average of these beats, i.e. the limit \( \tau_{fs} \ll \tau_{obs} \), though some intermediate situations, displaying various aspects of this beat phenomenon, have been reported \((15, 16)\). Averaging over time yields a charge cloud corresponding to \( \omega_{fs} t = \pi/2 \) or \( 3\pi/2 \) (here we assume that \( \tau_{fs} \ll \tau_{nat} \), so that the pattern has time to develop before decay); i.e.,

![Figure 5](image-url)
\[<\rho_{xx}^a>_{t} = \frac{1}{3} \rho_{xx}^a(0) + \frac{2}{3} \frac{7}{18} (1 + \frac{3}{5} \rho_{xx}^a) \]
\[<\rho_{yy}^a>_{t} = \frac{1}{3} \rho_{yy}^a(0) + \frac{2}{3} \frac{7}{18} (1 - \frac{3}{5} \rho_{yy}^a) \]
\[<\rho_{zz}^a>_{t} = \frac{2}{9} \frac{7}{27} \rho_{zz}^a \]

thereby modifying the linear polarizations
\[P_{1/2}(S=1/2) = \frac{3}{2} \cdot P_{1/2}(S=0) \]

provided that the sum of the two fine structure components, $2P_{3/2} = 2S_{1/2}$ and $2P_{1/2} = 2S_{1/2}$, is monitored.

The time evolution of the angular momentum $L_{1}(t)$ is best analyzed in the atomic basis, where the initial conditions are
\[\rho_{11}^a(0) = \frac{1}{2} (1+L_{1}) \]
\[\rho_{-1,-1}^a(0) = \frac{1}{2} (1-L_{1}) \]
The time evolution is governed by (14)
\[\rho_{11}^a(t) = \frac{1}{2} L_{1} \cdot \rho_{11} + \frac{1}{2} (\rho_{11} + \frac{1}{2} \rho_{zz}) \]
\[\rho_{-1,-1}^a(t) = -\frac{1}{2} L_{1} \cdot \rho_{-1,-1} + \frac{1}{2} (\rho_{-1,-1} + \frac{1}{2} \rho_{zz}) \]
with $G_{1} = G_{1}(t) = 1/9 \cdot [7 \pm 2 \cos(\omega_{eg} t)]$. Thus the spin-orbit coupling causes the angular momentum component along $z^{a}$ to vary in time as
\[L_{1}(t) = L_{1} \cdot \rho_{11}^a + \rho_{-1,-1}^a = L_{1} \cdot \frac{1}{9} [7 \pm 2 \cos(\omega_{eg} t)] \]  

The photon intensity $I_{j}(t)$ in the $z$ direction varies in time as
\[I_{j}(t) = \rho_{11} + \rho_{-1,-1} = \frac{1}{9} [7 \pm 2 \cos(\omega_{eg} t)] \]
in agreement with Eq. (18). Equations (19) and (20) show that $L_{1}(t)$ and $I_{j}(t)$ vary with the same amplitude, implying that $P_{3} = (\rho_{-1,-1}-\rho_{11})/2 \rho_{11}$ stays constant in time and is thus not affected by the fine structure.

So, a measurement of the Stokes parameters for a $2P_{3/2}2S_{1/2}$ transition allows reconstruction of the shape and dynamics of the charge cloud created in the collision by multiplying the linear polarizations by a factor $7/9$, while $P_{3}$ is unchanged. The average intensity $I_{j} = <I_{j}(t)> = 7/9$ is reduced because of the change in photon angular distribution.

Table 1 summarizes the (average) intensity and the depolarization factors $c_{1}$ for circular and $c_{2}$ for linear polarization in the $z^{a}$ direction for the cases $S=1/2$ and $S=1$, including also the individual fine structure components. General formulas may be found in (17).

2.3.2. The effect of hyperfine structure

In analogy to the fine structure effect, the presence of nuclear spin will cause further oscillatory behavior. We shall not present the details in general but refer to the literature (14) and here just give as an example the effect of further adding a nuclear spin $I=3/2$. The effect on the (time averaged) circular polarization is a reduction to almost half the size, while the linear polarization is reduced by almost a factor of eight, cf. Table 1.
TABLE 1

<table>
<thead>
<tr>
<th>Transition</th>
<th>$l_1$</th>
<th>$c_1$</th>
<th>$c_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 = 0$:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$1^p + 1^s$</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$2^p + 2^s$</td>
<td>7/9</td>
<td>1</td>
<td>3/7</td>
</tr>
<tr>
<td>$3^p + 2^s$</td>
<td>2/9</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>$3^p + 3^s$</td>
<td>5/9</td>
<td>1</td>
<td>3/5</td>
</tr>
<tr>
<td>$3^p + 3^s$</td>
<td>41/54</td>
<td>27/41</td>
<td>15/41</td>
</tr>
<tr>
<td>$3^p + 3^s$</td>
<td>2/27</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$3^p + 3^s$</td>
<td>1/4</td>
<td>1/3</td>
<td>1/3</td>
</tr>
<tr>
<td>$3^p + 3^s$</td>
<td>47/108</td>
<td>45/47</td>
<td>21/47</td>
</tr>
<tr>
<td>$1 = 3/2$:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$2^p + 2^s$</td>
<td>209/300</td>
<td>325/627</td>
<td>27/209</td>
</tr>
</tbody>
</table>

Figure 5 illustrates the effect on the (time-averaged) shape by subsequently adding an electron spin $S=1/2$, (b) + (c), and a nuclear spin $I=3/2$, (c) + (d), leading to an almost isotropic charge distribution. This situation is close to the case of the $^7$Li($^2$p) state, where, however, effects due to a finite lifetime, etc., also show up (16).

Fine and hyperfine structure may thus cause a severe reduction in measured anisotropy compared to the nascent charge cloud, provided of course that they have time to develop.

2.4. S=0 excitation

S=0 excitation has so far only been studied in a few cases. A D state has five substates of which, in the natural frame, the atomic basis states with $N=0, 0,-2$ have positive reflection symmetry with respect to the scattering plane, while $N=1, -1$ yield negative reflection symmetry. In the simple case where the excitation is fully coherent and the system possesses positive reflection symmetry, three amplitudes $(a_p, a_0, a_{-2})$ thus come into play. The normalization condition $|a|^2=1$ and an arbitrary phase factor leave four real parameters to be determined. The dipole radiation pattern for a subsequent N-p decay is completely determined by the four parameters $P_1, P_2, P_3, P_4$ introduced above. One might think that a coherence analysis giving these four Stokes parameters determines the D state completely. However, this turns out not to be the case (18). Though characteristic parameters for the shape and dynamics of the charge cloud, like $\gamma$, $l_1$, $P_2$ and the relative height, still given by Eq. (10), can be evaluated, analysis shows that in general two D-states exist having identical dipole radiation patterns, one charge cloud being the mirror of the other one in the $(x^0, z^0)$ plane. Here $x^0$ is still the symmetry axis of the radiation pattern, but the charge cloud does not exhibit reflection symmetry with respect to this plane (18). Application of external fields, which break the symmetry even further and influence the time development of the charge cloud during the time from excitation to decay, is necessary in order to distinguish between the two possibilities (19).

3. ORIENTATION EFFECTS

We shall now address some central, not yet resolved problems of current interest, which may conveniently be discussed within the framework presented above. Our aim is to attempt to understand orientation effects, a feature unique for planar scattering experiments.
3.1. Orientation in electron-atom collisions

3.1.1. He excitation

Starting out with the simplest case, excitation of the He(21P) level, a great number of theoretical and experimental investigators have studied this process for electron impact energies around 80 eV. The situation is schematically summarized in Fig. 6.

For scattering angles $\theta_{\text{col}}=0^\circ$ and $180^\circ$ we have $L_2=0$ and $\gamma=0^\circ$ for symmetry reasons. For small scattering angles $L_2>0$, and $\gamma$ follows the direction of momentum transfer, as predicted by the First Born Approximation (FBA), while $L_1$ (FBA)$=0$. $L_1$ goes through a maximum where the state is almost circular, and from about there also the alignment angle differs greatly from the FBA prediction. $L_1$ then decreases and goes through zero at some intermediate angles $\theta_{\text{col}}$, beyond which the sense of rotation of the electron changes. The shape approaches again a circular state, now with a negative angular momentum, which subsequently is reduced in size when $\theta_{\text{col}}$ approaches $180^\circ$.

Generally speaking, there is now reasonable agreement between theory and experiment up to the angle $\theta_{\text{col}}=0^\circ$, while the situation beyond this point is still unclear, with disagreements among the various theories and among the experimental results. And the qualitative physics behind the behavior shown in Fig. 6 still needs to be clarified. One suggestion is that at small angles the scattering is dominated by attractive polarization forces, leading to $L_2>0$, while at larger angles repulsive electron-electron forces dominate, hence $L_2<0$. However, this argument alone cannot be the whole story, because calculations for positron scattering give $L_2<0$ at small angles, although the polarization forces are still attractive (20). Analysis in terms of Born series shows that, in general, for small $n$ the leading term of $L_2$ is proportional to $q^4$ and a product of a first and second order term, while at larger angles a term proportional to $q^4$ and a product of two second order terms become important. $q$ being the projectile charge (20).

Going toward lower energies where the de Broglie wavelength begins to exceed the size of the He atom, the situation is even more unclear, and whether the picture outlined in Fig. 6 can be maintained (21) is still being debated. Fortunately, agreement between theory and experiment is now very good at energies very close to threshold, though a typical data point here takes about 1000 hours to measure (22).

One problem on the experimental side is that mainly the photon pattern of the He I 1S-21P 504 A line, being in the vacuum ultraviolet (VUV) region, has been studied with correlation techniques, by which the sign of $L_2$ cannot be directly determined. We refer to Ref. 23 for a detailed analysis of the situation, and will here just sketch an illuminating way of analyzing the problem making use of the Poincaré sphere introduced above. Figure 7 shows a recent calculation of Madison (24) for He(21P) excitation at 80 eV impact energy, illustrating quantitatively the behavior summarized in Fig. 6.
Crosses mark 5° steps in $\theta_{\text{col}}$. The curve starts out at $\vec{p}=(1,0,0)$ for $\theta_{\text{col}}=0^\circ$ and then moves westward, slightly below the equator, with increasing $\theta$. In contrast, FBA predicts a curve restricted to the equator since $L_1(\text{FBA})=0$. Then it makes a loop passing near the south pole at the angle $\theta_{\text{min}}$, then a rapid movement in the northern direction, crossing the equator at $L_1=65^\circ$. After an approach to the north pole at $\theta_{\text{min}}$ it returns to the starting point at $\theta=180^\circ$. Why a study of angular momentum sign changes is particularly difficult using correlation analysis is now evident when we bear in mind that this technique only determines the projection $P_1=|P_2|P_3$ of $\vec{p}_2$ onto the equatorial plane, as shown in Fig. 8 for the curve of Fig. 7. Crosses correspond to $L_1>0$, i.e. the southern hemisphere, circles to $L_1<0$. The lower part of Fig. 7 shows that when $L_1$ changes from 0.5 $\hbar$ to -0.5 $\hbar$, corresponding to the region from $30^\circ$ southern to $30^\circ$ northern latitude and $85^\circ < \theta_{\text{col}} < 95^\circ$, the longitude $2\varphi$ stays almost constant, so the projection, Fig. 8, varies little.

Thus despite the fact that $\vec{p}$ moves rapidly, the angular correlation pattern is almost stationary, as further illustrated in Fig. 9. This figure, symmetric with respect to the equator and based on Eqs. (3), (4) and (6), shows the change in shape of the charge cloud, and thereby the correlation pattern when moving from the north pole to the south pole on the Poincare sphere. Some numerical examples are given in Table 2, highlighting the difficulty of studying $L_1$ sign changes based on measurement of $P_2$.

Trapping of resonance radiation in the target gas will tend to reduce the anisotropy, and thereby $P_3$, but leaves the angle $\gamma$ unchanged. This effect might be difficult to exclude on the ~1% level which is required (25). So, though being difficult, circular polarization measurements in the VUV range at carefully selected angles would be highly desirable, and much larger error bars can be tolerated if one just wants to determine the sign unambiguously (26).

Another way of exploring this problem is to study the He(3pg) level instead. This level has two decay modes, one leading to VUV emission (537 Å) and one in the visible (5016 Å), see Fig. 10. So, here systematic comparison of correlation analysis in the VUV and coherence analysis in the visible (27) serves as a good consistency check.

3.1.2. He, Ar, Kr, Xe excitation

Much less work has been done for the heavier rare gases, so little in fact that no picture like Fig. 8 for He can be drawn. Some studies of the resonance levels He(3s,3s’), Ar(4s,4s’), Kr(5s,5s’)) and Xe(6s,6s’) have been
TABLE 2

<table>
<thead>
<tr>
<th>$L_\perp$</th>
<th>$P_E$</th>
<th>Length</th>
<th>Width</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>1.000</td>
<td>1.000</td>
<td>0.000</td>
</tr>
<tr>
<td>±0.1</td>
<td>0.995</td>
<td>0.9975</td>
<td>0.0025</td>
</tr>
<tr>
<td>±0.2</td>
<td>0.980</td>
<td>0.990</td>
<td>0.010</td>
</tr>
<tr>
<td>±0.5</td>
<td>0.866</td>
<td>0.933</td>
<td>0.067</td>
</tr>
</tbody>
</table>
performed, but more systematic work is needed. This series of levels shows the effect at low energies of spin-orbit coupling during the collision, leading to charge clouds where $\rho_{pp}$ becomes significant. Also here UV circular polarization measurements are very desirable to see, e.g., whether the small-angle behavior is similar to that for He, or the physics is different. This has not been done yet, but there are reasons to believe that at small scattering angles $L_2$ might have the opposite sign of the He(nP) case, since for He a p electron is created, while for the heavy rare gases one creates a p hole.

3.2. Orientation in heavy atom collisions

Turning now to the topic of heavy atom collisions we shall again try to speculate on some directions of research which fruitfully could be investigated in the future, being well aware that this might be a risky business.

Excitation in heavy atom collisions is known to take place in essentially two ways: (i) at short distances through radial and rotational couplings at potential curve crossings (or approaches) of the transient quasi-molecule, and (ii) at large distances excitation may take place via direct transitions, and with appreciable probabilities at velocities where the Massey criterion or "the maximum rule" is fulfilled: $\Delta \alpha/\Delta v = v$, $\Delta \epsilon$ being the excitation energy, $a$ the effective interaction length, and $v$ the collision velocity (28). Also the orientation may vary in sign for mechanism (i), depending on details of the molecular curves (18,29) there seems to be a preferred direction of rotation over a large velocity range for mechanism (ii), which we shall now briefly discuss.

3.2.1. Direct excitation: A propensity rule for orientation?

Large impact parameter direct excitation of the resonance 3p-5p transitions of alkali or alkali-like atoms in collisions with rare gases shows that $L_2$ consistently has a large negative value in the region of maximum excitation where the Massey criterion is fulfilled (28,30). To analyze this problem further we model the alkali-rare gas system as a quasi-one electron system, i.e., effectively a three-body system consisting of the alkali core, the valence electron, and the rare gas atom, with the electronic excitation induced by the electron-rare gas interaction $V$ for which various simple models have been proposed (28). If we select as basis the set of three states consisting of the ground state $\vert s \rangle$ and the two atomic basis states $\vert p_\uparrow \rangle$ and $\vert p_\downarrow \rangle$ of Fig. 1, all having positive reflection symmetry, the following close-coupling equations are obtained for the amplitudes $a = (a_s,a_\uparrow,a_\downarrow)$:

$$\frac{iv}{dx} \frac{\partial a}{\partial x} = \alpha \cdot a$$  \hspace{1cm} (21)

with $\alpha$ being the position along the trajectory, $h$ the impact parameter, and $R = (z^2 + b^2)^{1/2}$ the internuclear distance

$$a = F_{sp} (R) \exp \left( \frac{i \Delta E}{\hbar v} - s \right) \begin{pmatrix} 0 & c.c. & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} + F_{-}(R) \begin{pmatrix} 0 & 0 & c.c. \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \left( \begin{array}{c} 0 \\ 0 \\ 0 \end{array} \right) + \exp \left( \frac{i \Delta E}{\hbar v} + s \right) \begin{pmatrix} 0 & 0 & c.c. \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \left( \begin{array}{c} 0 \\ 0 \\ 0 \end{array} \right)$$  \hspace{1cm} (22)

Here $\Delta E$ is the $x$-dependent $S-P$ energy difference, and $\alpha$ the rotation angle of the internuclear axis (31). The valence electron potential $V$ only enters through the common factors $F_{sp}(R)$, describing the $S-3p$ coupling, and $F_{-}(R)$, describing the $|p_\uparrow \rangle \leftrightarrow |p_\downarrow \rangle$ coupling, respectively equal to the matrix element $\langle s \vert V \rangle$ and $\langle p \vert V \rangle$, referred to the body-fixed system. The interior of the matrices depend on the dynamics only, allowing us to state general conclusions. We can now easily see what happens. Starting the system in the ground state, i.e. $(1,0,0)$, one gets in first order:

$$a_{-1} = \frac{i}{2\nu} \int F_{sp} \exp \left( \frac{i \Delta E}{\hbar v} - s \right) dx$$  \hspace{1cm} (23a)
\[ a_{i_1} = -\frac{1}{i} \int F_{sp} \cdot \exp \left( i \frac{\Delta E}{\hbar v} + a \right) \, dx \]  \hspace{1cm} (23b)

The angle \( \alpha \) changes by \( \pi \) during the collision. Thus, if the velocity is such that the change in phase over the effective interaction length \( a, \Delta Ea/\hbar v = \pi \), then \( a_{i_1} \) has a stationary phase and its maximum value is obtained. For \( a_{i_1} \), however, the total phase changes by \( 2\pi \), giving a near cancellation of the integral, so that \( a_{i_1} \) is small. Thus a propensity rule (32,33) for circular polarization seems to hold in the region of maximum excitation. To illustrate the development of the circular state along the trajectory, Fig. II shows a calculation for the Na-He system (31) at an energy \( E_{CM} = 2 \) keV and impact parameter \( b = 2.2 \) a.u. using for \( V \) a Baylis-type (28) potential. This system is particularly interesting since a study of the time-inverse process, scattering of He on laser-excited, oriented sodium atoms may be feasible (34): Eqs. (21) and (22) predict that in the usual geometry the \( |p_{i_1}\) state should be easily deexcited, while the \( |p_{i_1}\) state should be quite stable.

![Graphs showing S-P Excitation Probability and Angular Momentum vs Position along trajectory](image)

**Figure 11.**
These ideas can be further generalized, admittedly now becoming even more speculative: Inspection of Eqs. (23) shows that if \(a \neq 0\) then the roles of \(a_{1}\) and \(a_{2}\) will be interchanged. Thus the situation of Fig. 12(a) might be possible: Excitation from an \([s]\) state will lead to a \([p_{1}]\) state, deexcitation to \([p_{2}]\). Furthermore, Fig. 12(b) shows another configuration that might be accessible: An atom in a \([p_{2}]\) state is easy to deexcite (to the \(S\) state) but hard to excite (to the \(D\) state). Changing the sense of rotation, i.e., to a \([p_{1}]\) state, will now give an atom unwilling to get deexcited (to the \(S\) state) but readily excited (to \(D\)). In other words, if an atom in the \([p_{1}]\) state is hit on its left side [Fig. 2(a)] it preferably gets deexcited, while hitting its right side triggers excitation.

To what extent such experiments, in which the dynamics of the active electron is strictly controlled, are feasible, the future will show.

4. CONCLUSIONS

We have suggested a common framework for description of alignment and orientation of atoms excited in planar scattering experiments, quantizing along the axis perpendicular to the scattering plane. Analysis of symmetry properties of the atomic states and the forces responsible for the evolution of the charge cloud suggest a parametrization of the shape (i.e., length, width, and height) and dynamics of the excited charge cloud, which is directly related to experimental observables as well as to the atomic scattering amplitudes and density matrix elements.

Considering the often very extensive investments of time and equipment needed for these investigations, one must carefully consider which systems are worth understanding. Obviously, it is worthwhile to see if we can understand the simplest and/or most fundamental systems at this level of accuracy.

Concentrating on orientation effects — unique for planar scattering geometry — we conclude that electron impact excitation of He is now quite well understood at small and intermediate scattering angles while the experimental and theoretical situation still needs to be improved at large angles. For the heavier rare gases, much remains to be done before a consistent picture emerges. Circular polarization measurements in the VUV would be particularly useful.

For heavy atom collisions a few simple systems have now been understood in the molecular regime. For direct excitation a propensity rule has been suggested, but it needs further experimental attention.

In this short overview we have omitted discussion of the now rapidly developing field of application of spin-polarized particles, which promises much information about spin-dependent forces (35) in the future. Also experiments involving coherent superposition of states with opposite parity, using hydrogen targets for photon emission (36) and, e.g., helium for electron emission (3) are interesting new developments.
ACKNOWLEDGEMENTS

During the preparation of this review the authors acknowledge stimulating discussions, collaborations, correspondence, and communication of results prior to publication on some of the topics presented here with M. Barat, A. Bühning, J. P. M. Beljers, D. Burns, D. Dowek, H. Fritsch, H. G. M. Heidemann, C. J. Joachain, H. Kleinpoppen, D. Madison, S. E. Nielsen, F. da Paixão, M. J. Roberts, H. Schmidt, and V. Sidis. Two of us (NA, IVH) are grateful for the facilities and financial support provided by JILA and the NBS office of Standard Reference Data to conduct this study. Two of us acknowledge travel grants from the Danish Natural Science Research Council and NATO (NA) as well as from the Deutsche Forschungsgemeinschaft (IVH).

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