

Reprinted from: ELECTRON AND PHOTON INTERACTIONS WITH ATOMS
(1976)

Edited by H. Kleinpoppen and M.R.C. McDowell
Book available from: Plenum Publishing Corporation
227 West 17th Street, New York, N.Y. 10011

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Spatial Asymmetries in Atomic Collisions

H. G. BERRY, L. J. CURTIS, D. G. ELLIS,
AND R. M. SCHECTMAN

Anisotropic excitation of atoms is discussed in terms of the subsequently emitted radiation. Examples are given of radiation from beam-foil excited atoms: (a) quantum beats between levels of the same parity are observed from aligned atoms; (b) quantum beats between levels of opposite parity are observed from excited atoms with net electric dipole moments; and (c) circularly polarized light is observed from atoms oriented in a tilted-foil geometry which does not have cylindrical symmetry.

A system of atoms which has been excited isotropically is statistically populated among the different magnetic substates of orbital angular momentum. That is, the atoms are not oriented or aligned and, consequently, the light emitted in the decay of an excited state is isotropic and unpolarized. Departures from isotropy or spherical symmetry can be represented in terms of the alignment and orientation of the atoms. We shall discuss how the atomic alignment and orientation can be measured in terms of the linear and circular polarization of the emitted radiation.

The first measurements of the breakdown of spherical symmetry in excited atoms through observations of the polarization of emitted light involved excitation by beams of electrons or photons or other atoms and ions. In most such cases, cylindrical symmetry was retained. The beam axis can be defined as the traditional z axis for orbital angular momenta of the excited atoms. Then the cross sections for different absolute values of angular

H. G. BERRY • Department of Physics, The University of Chicago, Chicago, Illinois 60637, U.S.A. L. J. CURTIS, D. G. ELLIS, and R. M. SCHECTMAN • Department of Physics and Astronomy, The University of Toledo, Toledo, Ohio 43606, U.S.A.

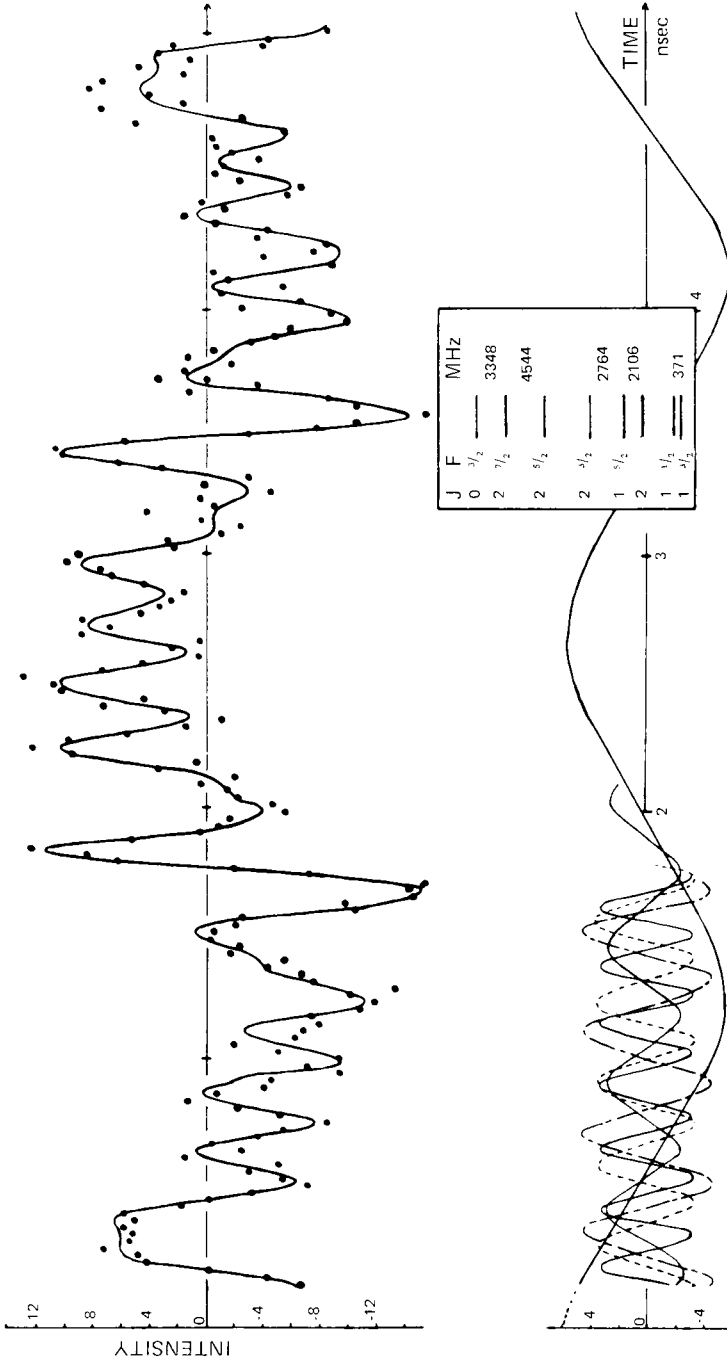


Figure 1. Modulations in the decay $4p(^3P)$, $^7\text{Li II}$. The five contributing frequencies and the energy levels are shown below.

momentum projections with respect to this axis, $\sigma(|m_L|)$, are not necessarily equal. Their departures from equality have been set in terms of the fractional linear polarization of the light emitted during decay of the excited state (Percival and Seaton, 1958) for interactions which are independent of nuclear and electronic spins. The departure from spherical symmetry is observed as a net electric dipole along or perpendicular to the beam axis (depending on whether low or high $|m_L|$ are populated). Thus, the alignment, a single parameter, can be measured from either two total intensity measurements at two angles or the linear polarization fraction at one angle.

In beam-foil spectroscopy, the added factor of good time resolution of 10^{-11} sec or better after the impulsive excitation (10^{-14} sec) allows observation of excited states which are not energy eigenstates of the atomic Hamiltonian. The fractional linear polarization, observed in a particular direction, is then modulated in time with the frequencies corresponding to the energy differences of the coherently superposed energy eigenstates. Figure 1 is an example of such a time-resolved decay (Berry *et al.*, 1974b), and the theory of these modulations under conditions of cylindrical symmetry is well

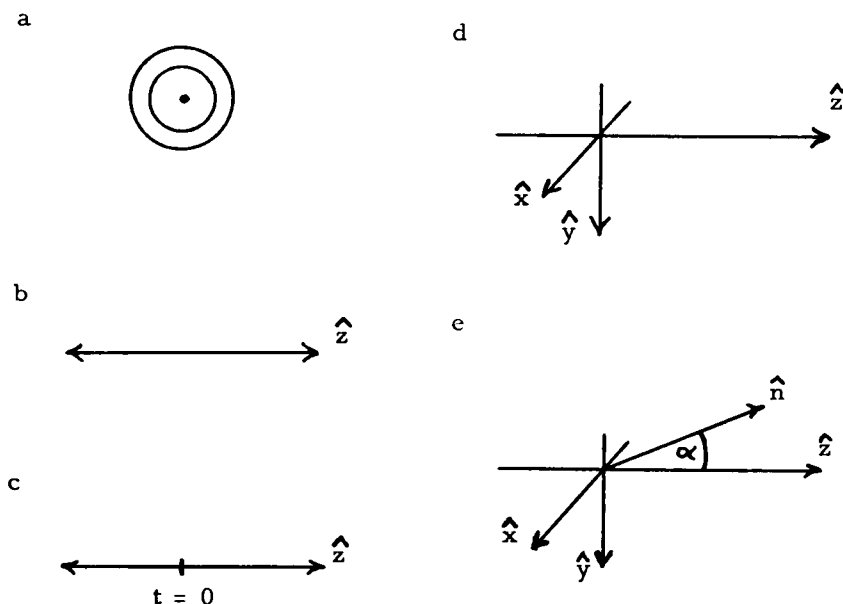
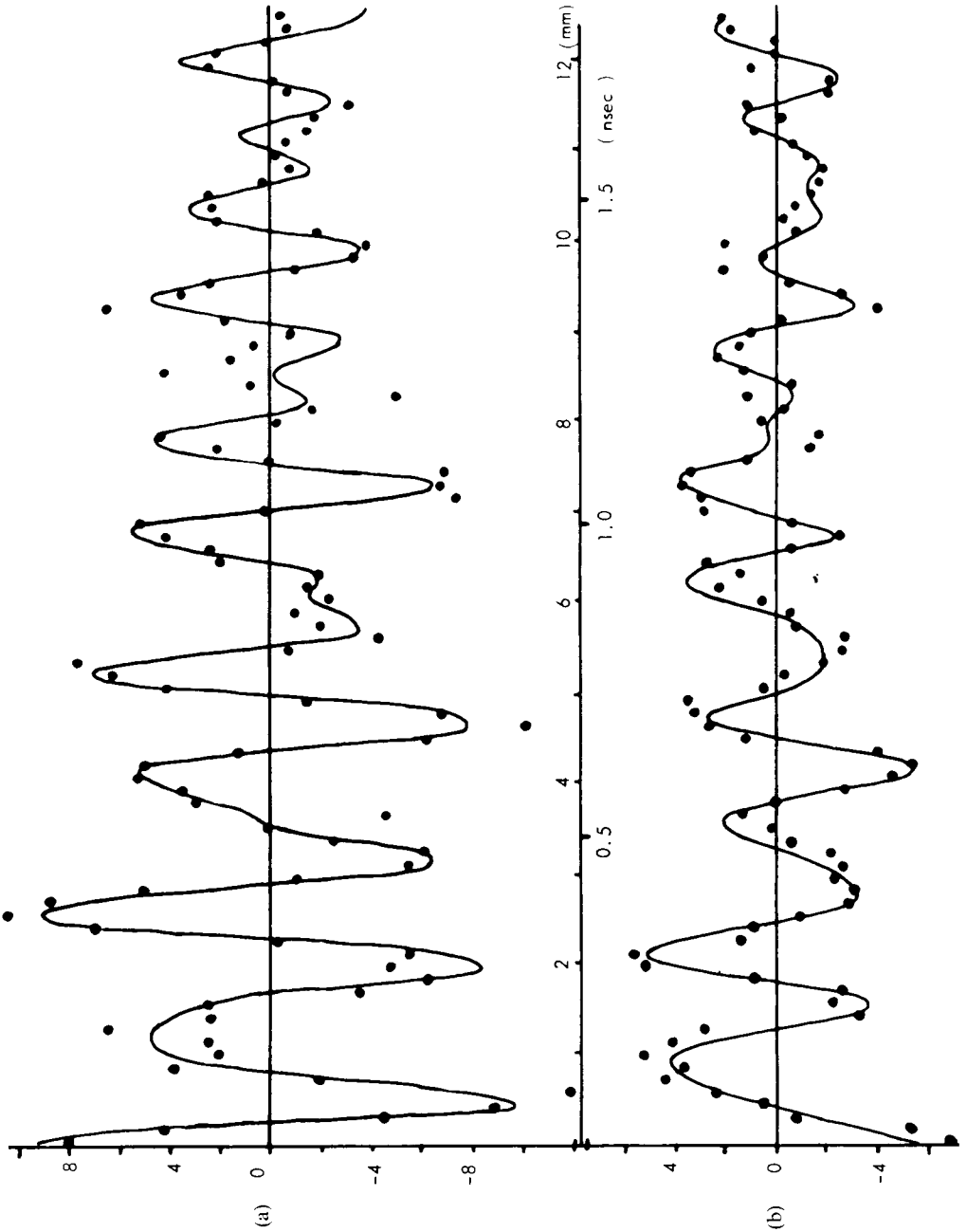


Figure 2. Schematic representation of increasing source asymmetry: (a) spherical symmetry, (b) cylindrical symmetry about z axis, (c) time-resolved cylindrical symmetry, (d) reflection asymmetry in xy plane, and (e) orientation axis $\hat{n} \times \hat{z}$.



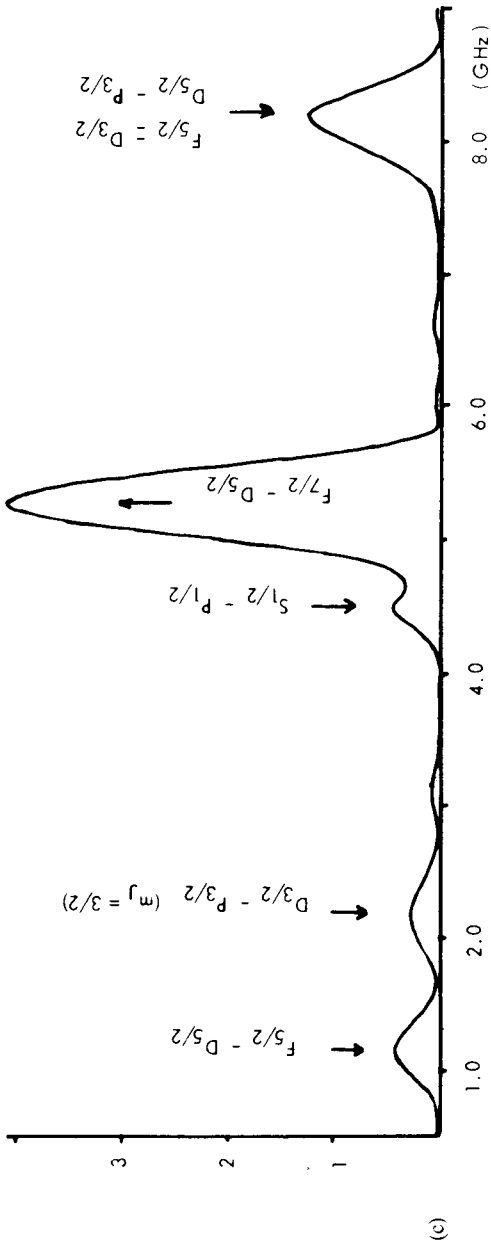


Figure 3. Modulations in the decay of $\text{He}^+(n=4)$ in electric fields of ± 465 V/cm (a, b), and the Fourier transform of the difference curve (c). The modulation frequencies correspond to $m_J = \frac{1}{2}$ Stark-shifted energy separations, except where noted.

developed (Macek and Jaecks, 1971; Berry *et al.*, 1972) in terms of the density operators of the excited system.

In Figure 2, we indicate schematically the gradual breakage of excitation symmetry. The pure cylindrical symmetry of 2b has a definite time $t = 0$ imposed for the beam-foil excitation in 2c. This could equivalently be any time-resolved beam excitation by electrons or photons, for example. We next look for breakage of the cylindrical symmetry through reflection asymmetry in the xy plane perpendicular to the beam z axis, as in 2d. The $+z$ and $-z$ directions are then distinguishable, and a coherent superposition of states of opposite parity with respect to reflection in the xy plane can be formed. Eck (1973) suggested such $2s, 2p$ coherence in beam-foil excited Lyman α , which has since been verified (Sellin *et al.*, 1973; Gaupp *et al.*, 1974).

We have observed mixed-parity coherence between many states of different orbital angular momentum in Balmer α of hydrogen ($n = 3$, SPD) and in 4686 Å of He⁺ ($n = 4$, SPDF). An external electric field is used to mix the atomic eigenstates of opposite parity which are part of the coherent superposition so that time-dependent intensity modulations can be observed in the photon decay, the frequencies being proportional to the Stark-shifted energy differences. If a coherent superposition occurs, the amplitudes and phases of these modulations will vary with the external field direction. The difference between the field parallel and antiparallel to the beam direction will give twice the amplitude of the modulations due to the excitation coherence of the mixed-parity states, as shown in Figure 3. We find that coherence is exhibited between essentially all possible mixed-parity combinations and is a rather general attribute of beam-foil collisions, which thus produce states both of nondefinite energy and nondefinite parity.

We have shown (Berry *et al.*, 1974a) that the beam-foil interaction is a function of the foil surface direction. Thus, there occurs a further breaking of the cylindrical symmetry as shown in Figure 2e when the foil normal n is tilted relative to the beam axis z . The alignment and orientation of such a collision which retains only reflection symmetry in the yz plane can be described by four parameters: three for the alignment and one for the orientation. Two of many possible such sets of four parameters are suggested: in spherical tensor notation the three second-rank components, $\rho_0^2, \rho_1^2, \rho_2^2$, describe the alignment and the first-rank component, ρ_1^1 , describes the orientation (Berry *et al.*, 1974a). These are proportional respectively to the parameters $A_0^s, A_{1+}^s, A_{2+}^s$, and O_{1-}^c of Fano and Macek (1973). Ellis (1973) has discussed the general case for time-resolved impulsive excitation of atoms.

The polarization state of the emitted light is a measure of the source anisotropy. The four Stokes parameters describe the polarization state and, in the viewing geometry shown in Figure 4, are related to the alignment and

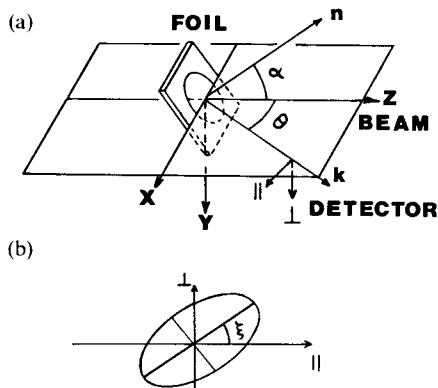


Figure 4. Beam and viewing geometry. The foil normal n is tilted at an angle α to the beam axis z .

orientation parameters by

$$\begin{aligned}
 I &= I_0 \left[2 + \left(\frac{3}{10}\right)^{1/2} \rho_0^2 (2 - 3 \sin^2 \theta) + \left(\frac{9}{2}\right)^{1/2} \rho_2^2 \sin^2 \theta \right] \\
 M &= I_0 \left[-3 \left(\frac{3}{10}\right)^{1/2} \rho_0^2 \sin^2 \theta - \left(\frac{9}{2}\right)^{1/2} \rho_2^2 (1 + \cos^2 \theta) \right] \\
 C &= I_0 \cdot \frac{6}{5} \sqrt{5} i \rho_1^2 \sin \theta, \quad S = I_0 2 \sqrt{3} \rho_1^2 \sin \theta
 \end{aligned} \tag{1}$$

where I_0 is a normalization constant. Thus, the polarization state of the emitted light completely determines the alignment and orientation of the excited state. However, we must note that the Stokes parameters are insensitive to tensor components ρ_q^k with $k > 2$, and hence excitation amplitudes are completely determined only for S and P states. In Eq. (1), M/I is the standard fractional linear polarization $(I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp})$, which is proportional to the alignment ρ_0^2 for the cylindrical case only (when $\rho_1^2 = \rho_2^2 = 0$), while S is the fractional circular polarization.

We have observed the ${}^4\text{He } 2s({}^1S) - 3p({}^1P)$ transition at 5016 Å excited in the beam-foil geometry Figure 4 and measured its Stokes parameters to obtain the linear polarization fraction M/I and the circular polarization fraction S/I . We find that $S > 0$, which corresponds classically to the emitting electron preferentially orbiting clockwise, as viewed in Figure 4. The foil-tilt angle dependence was measured at 130-keV beam energy and is quite well fitted to a $\sin \alpha$ dependence as shown in Figure 5a. This suggests that the atom is set spinning by a torque fixed in direction relative to the foil surface. The very different energy dependences of the ‘‘alignment,’’ Figure 5b, and the orientation, Figure 5c, suggest that the interaction mechanisms for the

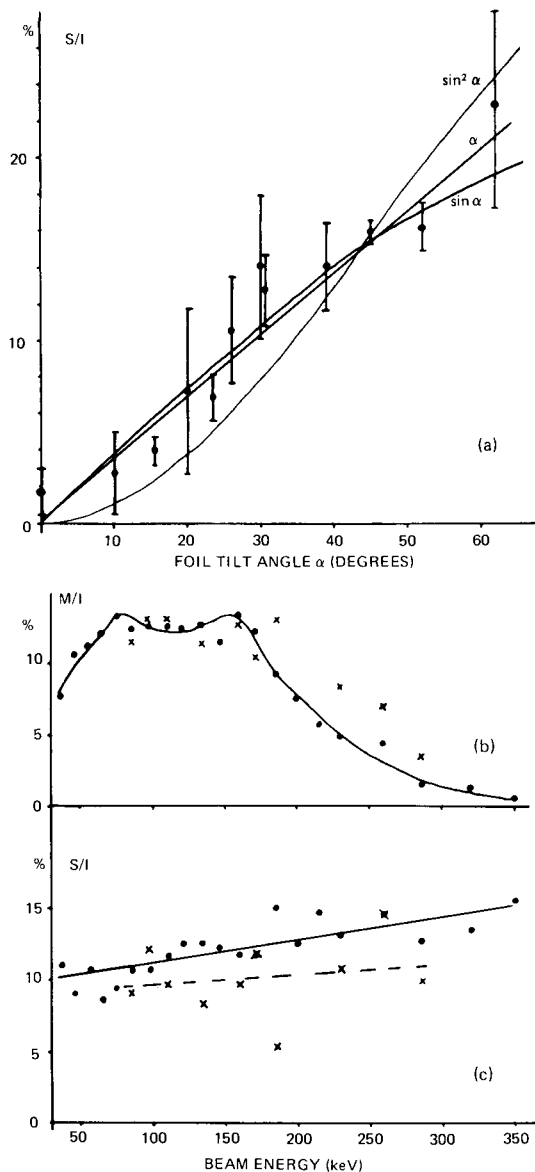


Figure 5. Fractional polarizations of the $5016 \text{ \AA } 2s(^1S)-3p(^1P)^4$ He I transition: (a) Circular polarization as a function of foil tilt angle α ; (b, c) Linear and circular polarization fractions respectively, as functions of beam energy for a carbon foil (\bullet) and an aluminum foil (\times).

two are at least partially independent. However, an initial Born approximation calculation of asymmetric charge capture at the foil surface predicts no orientation (Band, 1974).

The excitation amplitudes are straightforwardly derived from the Stokes parameters, as detailed elsewhere (Berry *et al.*, 1974a). In conclusion, we note that the asymmetric surface interaction has induced a relatively large orientation of the angular momentum of the atom. It would be interesting to compare the radiation produced in the analogous experiments of nonperpendicular crossed beams of atoms and electrons or photons or other atoms.

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