

This expression is a generalization of the one describing the MFPAD for circularly polarized light,⁴⁶ where $t_1(\gamma)$ and $t_2(\gamma)$ average to zero and $s_3 = -1$ for LHC polarization.

Considering eqn (2) several conclusions are drawn, which will be used in the following sections:

(i) For any (unknown) polarization of the ionizing light, the four $F_{00}(\theta_e)$, $F_{20}(\theta_e)$, $F_{21}(\theta_e)$, $F_{22}(\theta_e)$ functions can be determined from the Fourier analysis of the $I(\theta_e, \phi_e, \chi)$ measured distribution, ignoring the (γ) dependence, therefore providing the MFPADs for any orientation (χ) of the molecular axis relative to the axis of linearly polarized light.⁵⁰ Their (l, λ) partial wave expansion in a Legendre polynomial basis, where l is the angular momentum of the electron in a one center description of the scattering process and the quantum number λ is its projection on the molecular axis, gives access to the complex transition dipole moments describing the parallel and perpendicular PI transitions, and the amplitude of their relative phases. Knowing the sign of these relative phases requires the determination of the $F_{11}(\theta_e)$ function, which appears as the mixed product $s_3 \times F_{11}(\theta_e)$ in the MFPAD expression (eqn (2)), as discussed below. Access to these complex transition dipole moments is of high value for the advanced study of the photoemission dynamics.

(ii) The s_1 and s_2 Stokes parameters describing the linear component of the polarization are extracted by fitting the $I(\chi, \gamma)$ angular distribution of the ion fragments after integration of the $I(\theta_e, \phi_e, \chi, \gamma)$ angular distribution over (θ_e) and (ϕ_e) :

$$I(\chi, \gamma) = C \left\{ P_0^0(\cos \chi) \left[1 + \frac{\beta}{2} t_1(\gamma) \right] - \frac{\beta}{2} P_2^0(\cos \chi) [1 + t_1(\gamma)] \right\} \quad (3)$$

The asymmetry parameter of the ion fragment distribution β is obtained from the (χ) dependence after integration over (γ) , while the Fourier analysis in (γ) can be performed in the projections on $P_0^0(\cos \chi)$ and $P_2^0(\cos \chi)$, providing two ways for extracting s_1 and s_2 , as illustrated in Fig. 1(b)–(e):

$$\begin{aligned} \text{Proj}_{00}(\gamma) &= C \left\{ 1 + \frac{\beta}{2} s_1 \cos(2\gamma) - \frac{\beta}{2} s_2 \sin(2\gamma) \right\} \text{ and} \\ \text{Proj}_{20}(\gamma) &= C' \{ 1 + s_1 \cos(2\gamma) - s_2 \sin(2\gamma) \} \end{aligned} \quad (4)$$

We note that a similar form describes the angular distribution of the photoelectrons in the FF, involving the asymmetry parameter of the electron distribution β_e .

(iii) As for the $F_{11}(\theta_e)$ function which characterizes the circular dichroism in the MF, and the s_3 Stokes parameter which features the helicity of the ionizing radiation, they appear as the product $s_3 \times F_{11}(\theta_e)$ in the expression of the MFPAD $I(\theta_e, \phi_e, \chi, \gamma)$. This implies that determination of $F_{11}(\theta_e)$ requires an independent measurement with a known s_3 helicity, or a calculation. *Vice versa*, if $F_{11}(\theta_e)$ is known independently, the s_3 Stokes parameter is determined from the MFPAD. It is worth noticing that the MF circular dichroism constitutes the dephasing element of molecular polarimetry. It is also featured by the dimensionless CDAD (circular dichroism in electron angular distribution) parameter, proportional to $F_{11}(\theta_e)$, which characterizes the MF left–right emission asymmetry in the