CHIRALEX: theory

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(Dated: January 10, 2022)

I. INTRODUCTION

For gaining direct access into the physics in the molecular frame, it is imperative to maximally confine at least one of the molecule's axes along a laboratory-fixed direction. Experiments utilizing high-harmonic generation (HHG) [? ? ? ? ? ? ?],

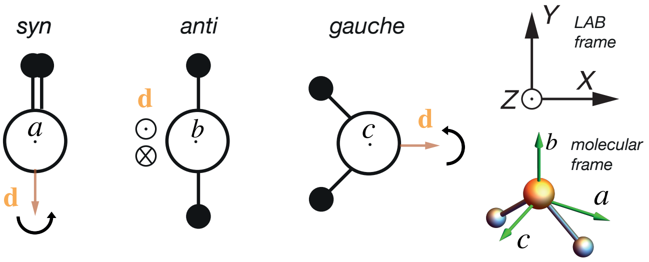

FIG. 1. Schematic view of an asymmetric top molecule rotating about its a-, b- and c- principal inertia axis. From the laboratory frame perspective the molecules are in *syn*, *anti* and *gauche* geometry. We assume that the probe pulse propagates along the laboratory Z-direction, whereas the molecule-confining pulse is restricted to the XY plane. d denotes the dipole moment vector.

$$\langle \cos^2\phi\rangle_{\psi(t)} = \frac{1}{16\pi^2}\sum_{J,J'=J_{min}}^{J_{max}} c_J^* c_{J'}\sqrt{(2J+1)(2J'+1)} \times \int_0^{2\pi} d\phi \cos^2\phi\, e^{-i\phi\Delta J}\sum_{K,K'=0}^{J,J'} a_K^{J,h_J,\tau *} a_{K'}^{J',h'_J,\tau'} \times$$
$$\times \int d\Omega \left[e^{-i\Delta_-K\chi} d^{J'}_{J'K'} d^J_{JK} + e^{i\Delta_-K\chi} d^{J'}_{J',-K'} d^J_{J,-K} + (-1)^\tau \left(e^{i\Delta_+K\chi} d^{J'}_{J',-K'} d^J_{J,K} + e^{-i\Delta_+K\chi} d^{J'}_{J',K'} d^J_{J,-K}\right)\right]$$

where $\Delta J = J'-J$, $\Delta_\pm K = K'\pm K$ and $d\Omega = \sin\theta d\theta d\chi$. Integrals over the ϕ angle impose rigorous selection rules on total angular momentum coupled by the $\cos^2\phi$ operator:

$$\int_0^{2\pi} d\phi \cos^2\phi\, e^{-i\phi\Delta J} = \begin{cases} 2\frac{\sin(\pi\Delta J)}{\Delta J}\frac{\Delta J^2-2}{\Delta J^2-4}e^{-i\pi\Delta J}, & \text{if } \Delta J \neq 2 \\ \frac{\pi}{2}, & \text{if } \Delta J = 2 \end{cases} \tag{1}$$

we note that the integral given in (1) is non-zero only if $\Delta J = 2$. The integral over the χ Euler angle can also be carried out analytically:

$$\int_0^{2\pi} d\phi e^{-i\phi\Delta_\pm K} = \begin{cases} -\frac{i}{\Delta_\pm K}(1-e^{-i2\pi\Delta_\pm K}), & \text{if } \Delta_\pm K \neq 0 \\ 2\pi, & \text{if } \Delta_\pm K = 0 \end{cases} \tag{2}$$

whereas appropriate integrals over the azimuthal Euler angle θ are generally denoted as:

$$b_{JJ'KK'} = \int_0^\pi d\theta \sin\theta d^J_{J,K}(\theta) d^{J'}_{J',K'}(\theta) \tag{3}$$

Equation (2) restricts contributions to the alignment cosine from states with the same K quantum number. After some algebra one finds explicit expression for the alignment cosine $\langle\cos^2\phi\rangle_{\psi(t)}$:

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$$\langle\cos^2\phi\rangle_{\psi(t)} = \frac{1}{8}\sum_{J=J_{min}}^{J_{max}-2}|c_J||c_{J'}|\sqrt{(2J+1)(2J+5)}\times\Bigg[2b_{JJ+200}(1+(-1)^\tau)Re(a_0^{J,h_J,\tau *}a_0^{J+2,h_{J+2},\tau})+$$
$$+\sum_{K\neq 0}^{J}Re(a_K^{J,h_J,\tau *}a_K^{J+2,h_{J+2},\tau})(b_{JJ+2KK}+b_{JJ+2-K-K})\Bigg]\cos(\omega_{J+2J}t+\Delta\phi^0_{J+2J})$$

If we assume that only states with $K = J$ contribute significantly to the wavepacket, we arrive at the equation (**??**) from the main text, where we note that $Re(a_J^{J,h_J,\tau *}a_{J+2}^{J+2,h_{J+2},\tau}) \approx 1$, $d^J_{JJ}(\theta) = \cos^{2J}\frac{\theta}{2}$, $d^J_{J,-J}(\theta) = \sin^{2J}\frac{\theta}{2}$ and $b_{JJ+2JJ} = b_{JJ+2-J-J} = \frac{2}{2J+3}$.