

Article



## Generation of Ultrafast Optical Pulses via Molecular Modulation in Ambient Air

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The determination of an optical pass length.



**Figure S1.** The optical path variation by rotating a pair of BK7 glass plates.  $\alpha$  – the angle of incidence,  $\theta$  – thickness of a glass plate,  $Z_1$  – the angle of refraction,  $Z_2$  – considerable pass in air,  $Z_3$  – optical path length inside a glass plate,  $n_M$  – refractive index of BK7 glass,  $n_a$  – refractive index of air.

As shown in the manuscript, we used the Sellmeier equation to estimate the refractive index and apply Snell's law to calculate refraction angles and the optical path length for each individual sideband  $E_q$ . Figure S1 shows the refracted optical pass through two BK7 glass plates. With the rotation of BK7 glass plates the beams are refracted at different angles depending on the wavelength. Hence, the optical path length is different for each wavelength. The equations below show the angle and the optical length variation. In Figure S1,  $Z_3$  can be shown as:

$$Z_{3} = \frac{Z_{1}}{\cos\left(\sin^{-1}\left(\frac{n_{a} \times \sin(\alpha)}{n_{M}}\right)\right)}$$

Additionally, from Snell's law:

$$\theta = \sin^{-1}\left(\frac{n_a \sin(\alpha)}{n_M}\right)$$

The relative delay in the glass plate can be expressed as  $Z_3 - Z_1$ . Additionally, one needs to compensate for the delay with respect to air:  $Z_2 = Z_3 \times cos(\theta - \alpha) - Z_1$ .

From these calculations, the optical pass length variation can be calculated:

$$\Delta Z_q = n_M(\lambda_q) \times (Z_3 - Z_1) - n_a \times (Z_2)$$

In this case, the variation of the individual sideband phases can be expressed as:

$$\Delta \phi_q = \frac{2\pi \Delta Z_q}{\lambda_q} = \frac{2\pi}{\lambda_q} \left( n_M(\lambda_q) \times (Z_3 - Z_1) - n_a \times (Z_2) \right) = \frac{2\pi Z_1(n_M(\lambda_q) - n_a)}{\lambda_q} \left( \frac{1 - \cos(\theta(\lambda_q) - \alpha)}{\cos(\theta(\lambda_q))} - 1 \right)$$

 $\lambda_q$  is corresponding to the wavelength of the experimentally measured spectra shown in Figure 2. Then, we used the obtained sideband phase in conjunction with Equation (1) to plot the theoretical curves in Figure 3.