

Understanding Centrifuges

Group Meeting

Cameron, Sören

October 22, 2025

Outline

- 1 Motivation and Theory
- 2 Frequency vs Time
- 3 Measurements

Outline

1 Motivation and Theory

2 Frequency vs Time

3 Measurements

Rotating molecules - Classical Treatment

Assume low initial temperature (which is the case for dopant molecules in He nanodroplets). Consider $\vec{E} = \hat{e}E_0\cos\omega_0 t$ and for simplicity a linear molecule with nonzero polarizability, α only along its axis, and moment of inertia I . Averaging over a period ($2\pi/\omega_0$) of the carrier oscillations, the molecule sees an angle dependent potential well

$$U(\theta) = -\frac{1}{4}E_0^2\alpha\cos^2\theta =: U_0\cos^2\theta. \quad (1)$$

See Board for drawing.

$$|\vec{\tau}| = \frac{\partial U}{\partial \theta} = U_0\sin 2\theta \implies \varepsilon_{\text{mol}} = \frac{|\vec{\tau}|}{I} \approx \frac{2U_0}{\pi I}. \quad (2)$$

Rotating molecules - Classical Treatment

Assume low initial temperature (which is the case for dopant molecules in He nanodroplets). Consider $\vec{E} = \hat{e}E_0\cos\omega_0 t$ and for simplicity a linear molecule with nonzero polarizability, α only along its axis, and moment of inertia I . Averaging over a period ($2\pi/\omega_0$) of the carrier oscillations, the molecule sees an angle dependent potential well

$$U(\theta) = -\frac{1}{4}E_0^2\alpha\cos^2\theta =: U_0\cos^2\theta. \quad (1)$$

See Board for drawing.

$$|\vec{\tau}| = \frac{\partial U}{\partial \theta} = U_0\sin 2\theta \implies \varepsilon_{\text{mol}} = \frac{|\vec{\tau}|}{I} \approx \frac{2U_0}{\pi I}. \quad (2)$$

If \hat{e} were to rotate with acceleration ε_{pol} , then it seems natural that trapping should occur when

Adiabaticity condition

$$\varepsilon_{\text{pol}} < \varepsilon_{\text{mol}} = \frac{2U_0}{\pi I}. \quad (3)$$

Smaller ε_{pol} becomes relevant when trying to rotate molecules with larger effective moments of inertia, such as molecules embedded in superfluid He. Centrifugal distortion can also increase the effective I by a few orders of magnitude.

Basic Theory of Centrifuge

Consider two oppositely circularly polarized waves that have shifted carrier angular frequencies of $\pm\Omega(t)$.

$$\vec{E}_+ = \frac{E_0}{2} \{ \hat{x} \cos[(\omega_0 + \Omega(t))t] + \hat{y} \sin[(\omega_0 + \Omega(t))t] \} \quad (4)$$

$$\vec{E}_- = \frac{E_0}{2} \{ \hat{x} \cos[(\omega_0 - \Omega(t))t] - \hat{y} \sin[(\omega_0 - \Omega(t))t] \} \quad (5)$$

Superimposing them gives a wave whose polarization vector rotates not at the carrier angular frequency but at $\Omega(t)$. The resultant wave is the optical centrifuge,

$$\vec{E}_{\text{CFG}} = \vec{E}_+ + \vec{E}_- = E_0 \cos(\omega_0 t) [\hat{x} \cos(\Omega(t)t) + \hat{y} \sin(\Omega(t)t)] \quad (6)$$

We call $\Omega(t)$ the (instantaneous) rotational angular frequency of the centrifuge.

Basic Theory of Centrifuge

Consider two oppositely circularly polarized waves that have shifted carrier angular frequencies of $\pm\Omega(t)$.

$$\vec{E}_+ = \frac{E_0}{2} \{ \hat{x} \cos[(\omega_0 + \Omega(t))t] + \hat{y} \sin[(\omega_0 + \Omega(t))t] \} \quad (4)$$

$$\vec{E}_- = \frac{E_0}{2} \{ \hat{x} \cos[(\omega_0 - \Omega(t))t] - \hat{y} \sin[(\omega_0 - \Omega(t))t] \} \quad (5)$$

Superimposing them gives a wave whose polarization vector rotates not at the carrier angular frequency but at $\Omega(t)$. The resultant wave is the optical centrifuge,

$$\vec{E}_{\text{CFG}} = \vec{E}_+ + \vec{E}_- = E_0 \cos(\omega_0 t) [\hat{x} \cos(\Omega(t)t) + \hat{y} \sin(\Omega(t)t)] \quad (6)$$

We call $\Omega(t)$ the (instantaneous) rotational angular frequency of the centrifuge.

- ① $\dot{\Omega} = 2\beta t$, $\beta > 0$ is the conventional centrifuge \implies accelerates the rotation of molecules (when $\dot{\Omega}$ not too large).
- ② $\dot{\Omega} = 0$ is the "constant frequency centrifuge" \implies molecular rotation follows this constant frequency.

Visualizing the centrifuge

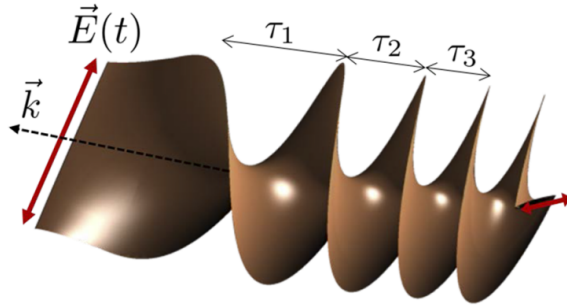


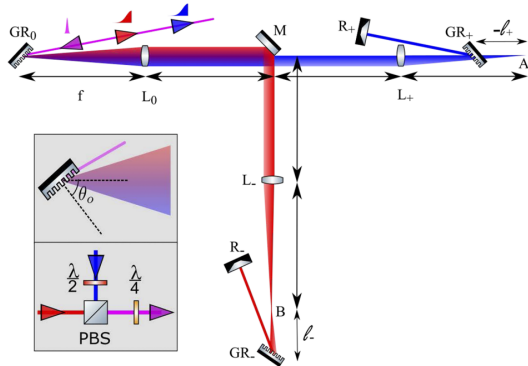
Figure: A depiction of the spatial and temporal behaviour of the conventional centrifuge propagating in the direction of the wavevector \vec{k} . Borrowed from MacPhail-Bartley et al. 2020.

When do the molecules follow the centrifuge?

The adiabaticity condition from earlier becomes

$$\frac{U_0}{\pi I \beta} = \frac{1}{4\pi} \times \frac{E_0^2}{\beta} \times \frac{\Delta\alpha}{I} > 1 \quad (7)$$

The slow centrifuge (sCFG)



Taking the limit of large stretching for the Gaussian envelope, the chirp applied by the grating pairs to the blue (+) and red (-) arms respectively is

Figure: θ_0 is the angle between the input beam and the normal vector to the grating grooves (the grating orientation angle). Borrowed from MacPhail-Bartley et al. 2020.

The slow centrifuge (sCFG)

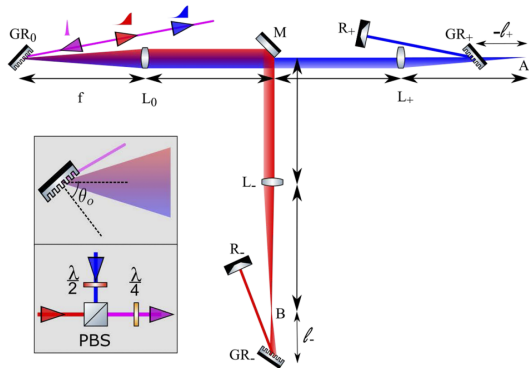


Figure: θ_0 is the angle between the input beam and the normal vector to the grating grooves (the grating orientation angle). Borrowed from MacPhail-Bartley et al. 2020.

Taking the limit of large stretching for the Gaussian envelope, the chirp applied by the grating pairs to the blue (+) and red (-) arms respectively is

$$\beta_{\pm} = -\frac{d^2\omega_0^3 \cos^2\theta_0}{16\pi^2 l_{\pm} c} \quad (8)$$

Note: GR_+ is at an effective distance l_+ from GR_0 which is negative!

Recall the centrifuge angular rotation frequency formula $\Omega(t) = 2\beta t$. Then using a $\lambda/2$ waveplate on the blue arm, followed by combination on the PBS and traversal of a $\lambda/4$ waveplate results in

$$\dot{\Omega} = 2\beta = \beta_+ - \beta_- \geq 0 \quad (9)$$

Limitations of sCFG

$$\beta_{\pm} = -\frac{d^2\omega_0^3\cos^2\theta_0}{16\pi^2 l_{\pm} c}$$

sCFG has the following hard constraints:

- ① Groove density $1/d$ is limited by manufacturing.
- ② $\theta_0 \lesssim 80^\circ$.
- ③ $l_+ < f$.

Choosing the appropriate parameters and building the setup such as to minimize the centrifuge acceleration resulted in $\beta = 0.017 \text{ rad/ps}^2$. This is too high for dopant molecules in superfluid Helium because the terminal frequency is well beyond the centrifugal wall!

The ultraslow centrifuge (usCFG)

Idea: Get a smaller $\dot{\Omega}$ from a smaller difference $\beta = (\beta_+ - \beta_-)/2$. But instead of actively applying equal and opposite chirps to two arms as in sCFG, we do the following.

- 1 *Delay Arm:* use the nominal (positive) chirp, β_0 from the **uncompressed** pulses after CPA.
- 2 *Grating Arm:* use a double pass grating pair compressor to introduce a **small additional chirp** $\Delta\beta < 0$.

Now, $\beta = (\beta_G - \beta_0)/2 = (\beta_0 + \Delta\beta - \beta_0)/2 = \Delta\beta/2$. If the delay arm has a controllable Δt delay with respect to the grating arm, then we can tune the terminal frequency of the centrifuge independently of $\Delta\beta$.

The ultraslow centrifuge (usCFG)

Idea: Get a smaller $\dot{\Omega}$ from a smaller difference $\beta = (\beta_+ - \beta_-)/2$. But instead of actively applying equal and opposite chirps to two arms as in sCFG, we do the following.

- 1 *Delay Arm:* use the nominal (positive) chirp, β_0 from the **uncompressed** pulses after CPA.
- 2 *Grating Arm:* use a double pass grating pair compressor to introduce a **small additional chirp** $\Delta\beta < 0$.

Now, $\beta = (\beta_G - \beta_0)/2 = (\beta_0 + \Delta\beta - \beta_0)/2 = \Delta\beta/2$. If the delay arm has a controllable Δt delay with respect to the grating arm, then we can tune the terminal frequency of the centrifuge independently of $\Delta\beta$.

Let b be the perpendicular distance between the parallel gratings. See Board for rest of equations.

Optical schematic of usCFG

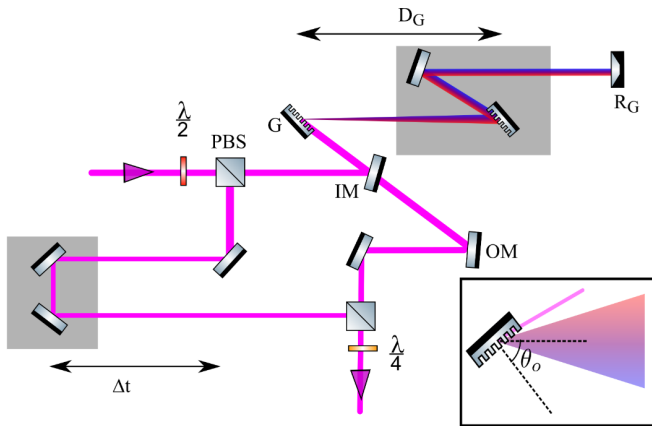


Figure: The grey boxes represent translation stages. This is a diagram in progress made by Kevin for usCFG draft. $D_G := b$ is the separation of gratings. The $\lambda/2$ plate at the input is only for power control. θ_0 is not taken to scale.

Outline

1 Motivation and Theory

2 Frequency vs Time

3 Measurements

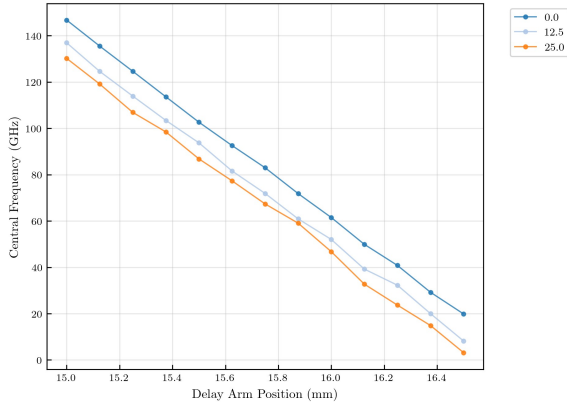
- Show animation for centrifuge

Outline

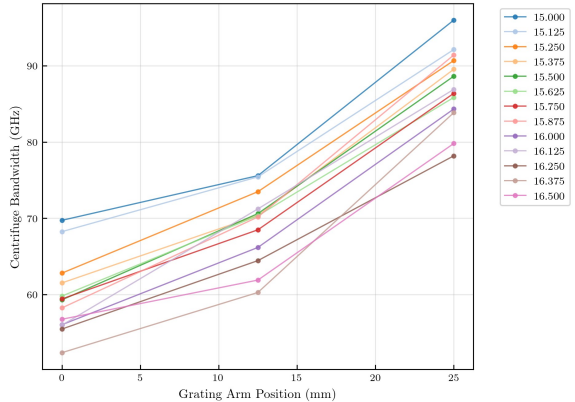
- 1 Motivation and Theory
- 2 Frequency vs Time
- 3 Measurements**

Arm Position Measurements

All Sets: Central Frequency vs Delay Arm Position



All Sets: Bandwidth vs Grating Arm Position



NO-Dimer (Central Frequency 20 GHz)

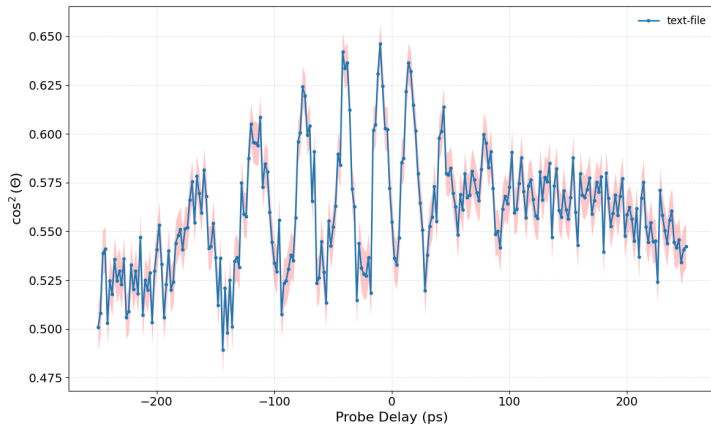
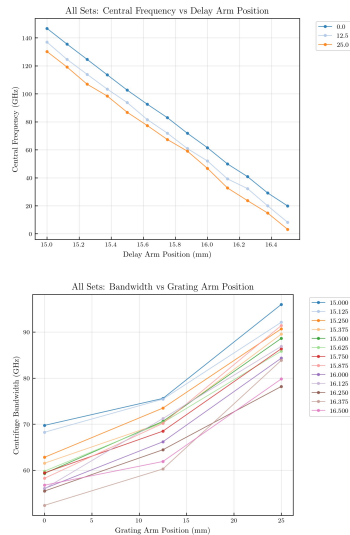


Figure: 30 bar / 18 K NO-Dimer; 2.9 mJ + 660 μ J; GA = 0 mm, DA = 16.375 mm



NO-Dimer (Central Frequency ≈ 0 GHz)

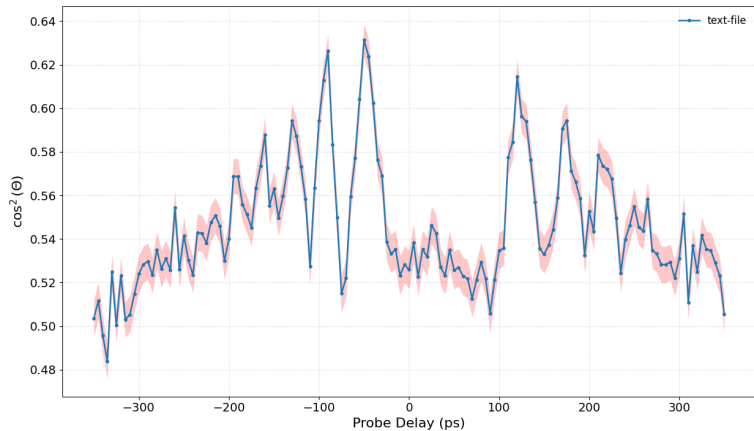
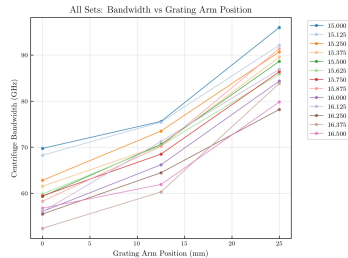
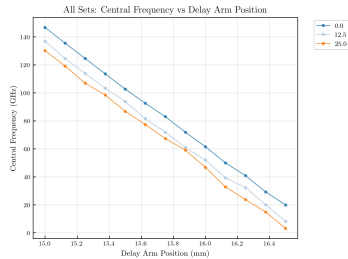


Figure: 30 bar / 18 K NO-Dimer; 2.9 mJ + 660 μ J; GA = 0 mm, DA = 16.8 mm



Questions?

Thank you for your attention!