Normal modes calculation*

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I. NORMAL MODES CALCULATION

The force balance equation of two optically trapped spherical particles near to each other in a highly viscous medium (Reynold's number tends to zero) is,

$$-\gamma\dot{\mathbf{x}} - \mathbf{k}\mathbf{x} = \xi \tag{1}$$

Where γ is the many body friction tensor, $\mathbf{k}$ is the stiffness tensor for those two optical traps and ξ is the noise tensor of thermal origin which is correlated as $\left\langle \xi(t)\xi^{\mathbf{T}}(t')\right\rangle = 2K_B T\gamma\delta(t-t')$. The many body friction tensor is given by

$$\gamma = \begin{bmatrix} \gamma_{11} & \gamma_{12} \\ \gamma_{21} & \gamma_{22} \end{bmatrix}$$

where $\gamma_{12} = \gamma_{21}$ and for two spherical beads of same radius,$\gamma_{11} = \gamma_{22}$.The stiffness tensor

$$\mathbf{k} = \begin{bmatrix} k_1 & 0 \\ 0 & k_2 \end{bmatrix}$$

. If $\mathbf{S}$ be a similarity transformation matrix which diagonalizes γ then

$$\begin{aligned} -\mathbf{S}^{-1}\gamma_{\mathbf{D}}\mathbf{S}\dot{\mathbf{x}} - \mathbf{k}\mathbf{x} &= \xi \\ -\gamma_{\mathbf{D}}\mathbf{S}\dot{\mathbf{x}} - \mathbf{S}\mathbf{k}\mathbf{x} &= \mathbf{S}\xi \\ -\gamma_{\mathbf{D}}\dot{\mathbf{y}} - \mathbf{k}\mathbf{y} &= \mathbf{S}\xi \end{aligned} \tag{2}$$

where we used $\mathbf{S}\mathbf{x} = \mathbf{y}, \mathbf{S}^{-1}\gamma_{\mathbf{D}}\mathbf{S} = \gamma$ and $[\mathbf{S},\mathbf{k}] = 0$. The diagonalized friction tensor is given by

$$\gamma_{\mathbf{D}} = \begin{bmatrix} \lambda_1 & 0 \\ 0 & \lambda_2 \end{bmatrix} \tag{3}$$

$$\lambda_{1,2} = \left(\frac{\gamma_{11}+\gamma_{22}}{2}\right) \pm \frac{1}{2}\sqrt{(\gamma_{11}-\gamma_{22})^2 + 4\gamma_{12}\gamma_{21}}$$

and the similarity transformation matrix is

$$\begin{aligned} \mathbf{S} &= \begin{bmatrix} \frac{\lambda_1-\gamma_{22}}{\gamma_{21}} & \frac{\lambda_2-\gamma_{22}}{\gamma_{21}} \\ 1 & 1 \end{bmatrix} \\ &= \begin{bmatrix} -1 & 1 \\ 1 & 1 \end{bmatrix} \end{aligned}$$

Here we assumed $\gamma_{11} = \gamma_{22}$. So, from Eq 2 we get two equations of motions describing the normal modes of the movements of the two trapped beads as

$$-\lambda_1\dot{y_1} - k_1 y_1 = (\xi_2 - \xi_1) \tag{4}$$

$$-\lambda_2\dot{y_2} - k_2 y_2 = (\xi_1 + \xi_2) \tag{5}$$

From Eq 6 and 7 we get

$$y_1(\omega) = \frac{\xi_1(\omega) - \xi_2(\omega)}{i\omega\lambda_1 - k_1} \tag{6}$$

$$y_2(\omega) = \frac{\xi_1(\omega) + \xi_2(\omega)}{i\omega\lambda_2 - k_1} \tag{7}$$

Hence the corresponding power spectrums are given by

$$\begin{aligned} \langle y_1(\omega)y_1^*(\omega)\rangle &= \frac{\langle(\xi_1-\xi_2)(\xi_1-\xi_2)^*\rangle}{\omega^2\lambda_1^2 + k_1^2} \\ \langle y_1(\omega)y_1^*(\omega)\rangle &= \frac{4k_B T(\gamma_{11}-\gamma_{12})}{\omega^2\lambda_1^2 + k_1^2} \end{aligned} \tag{8}$$

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Similarly,

$$\langle y_2(\omega)y_2^*(\omega)\rangle = \frac{4k_BT(\gamma_{11}+\gamma_{12})}{\omega^2\lambda_2^2+k_2^2} \tag{9}$$

The auto correlations can be calculated by performing inverse Fourier transformations on the corresponding power spectrums. We get

$$\langle y_1(t)y_1(0)\rangle = \frac{2k_BT}{k_1}e^{-\frac{k_1}{\lambda_1}t} \tag{10}$$

$$\langle y_2(t)y_2(0)\rangle = \frac{2k_BT}{k_2}e^{-\frac{k_2}{\lambda_2}t} \tag{11}$$

Here, $\lambda_1 = (\gamma_{11}-\gamma_{12})$ and $\lambda_2 = (\gamma_{11}+\gamma_{12})$. Now, we can write

$$x_1 = a_1y_1 + a_2y_2 \tag{12}$$

$$x_2 = a_1y_1 - a_2y_2 \tag{13}$$

Where a_1, a_2 are constants.

$$\langle x_1(\omega)x_1^*(\omega)\rangle = \left\langle a_1^2y_1(\omega)y_1^*(\omega)\right\rangle + \langle a_1a_2y_1(\omega)y_2^*(\omega)\rangle + \langle a_2a_1y_2(\omega)y_1^*(\omega)\rangle + \left\langle a_2^2y_2(\omega)y_2^*(\omega)\right\rangle$$

Now,$\langle y_1(\omega)y_2^*(\omega)\rangle = 0$, if $\gamma_{11}=\gamma_{22}$ and $\gamma_{12}=\gamma_{21}$. So,

$$\langle x_1(\omega)x_1^*(\omega)\rangle = \left\langle a_1^2y_1(\omega)y_1^*(\omega)\right\rangle + \left\langle a_2^2y_2(\omega)y_2^*(\omega)\right\rangle$$

$$\langle x_1(0)x_1^*(t)\rangle = \left\langle a_1^2y_1(0)y_1^*(t)\right\rangle + \left\langle a_2^2y_2(0)y_2^*(t)\right\rangle$$

Hence,

$$\langle x_1(\omega)x_1^*(\omega)\rangle = \left\langle a_1^2y_1(\omega)y_1^*(\omega)\right\rangle + \left\langle a_2^2y_2(\omega)y_2^*(\omega)\right\rangle$$

$$\langle x_1(0)x_1^*(t)\rangle = 2k_BT\left[\frac{a_1^2}{k_1}e^{-\frac{k_1}{\lambda_1}t} + \frac{a_2^2}{k_2}e^{-\frac{k_2}{\lambda_2}t}\right]$$

Similarly,

$$\langle x_2(0)x_2^*(t)\rangle = 2k_BT\left[\frac{a_1^2}{k_1}e^{-\frac{k_1}{\lambda_1}t} + \frac{a_2^2}{k_2}e^{-\frac{k_2}{\lambda_2}t}\right]$$

A. Calculation for a_1, a_2

We initially assumed,

$$\mathbf{S}.\mathbf{x} = \mathbf{y}$$

$$\mathbf{x} = \mathbf{S}^{-1}\mathbf{y} \tag{14}$$

For $\lambda_1 = \gamma_{11}-\gamma_{22}$ and $\lambda_2 = \gamma_{11}+\gamma_{22}$

$$\mathbf{S} = \begin{bmatrix} -1 & 1 \\ 1 & 1 \end{bmatrix}$$

$$\mathbf{S}^{-1} = -\frac{1}{2}\begin{bmatrix} 1 & -1 \\ -1 & -1 \end{bmatrix} \tag{15}$$

Using this and Eq. 14 we'll get

$$x_1 = \frac{1}{2}(-y_1+y_2) \tag{16}$$

$$x_2 = \frac{1}{2}(y_2+y_2) \tag{17}$$

Now we took

$$x_1 = a_1y_1 + a_2y_2 \tag{18}$$

$$x_2 = a_1y_1 - a_2y_2 \tag{19}$$

Comparing Eqs 16, 22, 24, 19 we get $a_1 = -\frac{1}{2}$ and $a_2 = \frac{1}{2}$.So,

$$\langle x_1(0)x_1^*(t)\rangle = \langle x_1(0)x_1^*(t)\rangle = \frac{k_BT}{2}\left[\frac{1}{k_1}e^{-\frac{k_1}{\lambda_1}t} + \frac{1}{k_2}e^{-\frac{k_2}{\lambda_2}t}\right] \tag{20}$$

On the other way, If we take $\lambda_1 = \gamma_{11}+\gamma_{22}$ and $\lambda_2 = \gamma_{11}-\gamma_{22}$ then

$$\mathbf{S} = \begin{bmatrix} 1 & -1 \\ 1 & 1 \end{bmatrix}$$

$$\mathbf{S}^{-1} = \frac{1}{2}\begin{bmatrix} 1 & 1 \\ -1 & 1 \end{bmatrix} \tag{21}$$

and we'll get

$$\langle y_1(t)y_1(0)\rangle = \frac{2k_BT}{k_1}\frac{\lambda_2}{\lambda_1}e^{-\frac{k_1}{\lambda_1}t} \tag{22}$$

$$\langle y_2(t)y_2(0)\rangle = \frac{2k_BT}{k_2}\frac{\lambda_1}{\lambda_2}e^{-\frac{k_2}{\lambda_2}t} \tag{23}$$

and

$$\langle x_1(0)x_1^*(t)\rangle = \langle x_1(0)x_1^*(t)\rangle = \frac{k_BT}{2}\left[\frac{1}{k_1}\frac{\lambda_2}{\lambda_1}e^{-\frac{k_1}{\lambda_1}t} + \frac{1}{k_2}\frac{\lambda_1}{\lambda_2}e^{-\frac{k_2}{\lambda_2}t}\right] \tag{24}$$

II. EXPERIMENT

We know k_1, k_2 experimentally and also know that $\lambda_1 = \gamma_{11}-\gamma_{12}, \lambda_1 = \gamma_{11}+\gamma_{12}$.If

$$\gamma = \mu^{-1}$$

then,

$$\begin{bmatrix} \gamma_{11} & \gamma_{12} \\ \gamma_{21} & \gamma_{22} \end{bmatrix} = \frac{1}{\mu_{11}^2-\mu_{12}^2}\begin{bmatrix} \mu_{22} & -\mu_{12} \\ -\mu_{21} & \mu_{11} \end{bmatrix} \tag{25}$$

$$\lambda_1 = \frac{\mu_{11}+\mu_{12}}{\mu_{11}^2-\mu_{12}^2} = \frac{1}{\mu_{11}-\mu_{12}} \tag{26}$$

$$\lambda_1 = \frac{\mu_{11}-\mu_{12}}{\mu_{11}^2-\mu_{12}^2} = \frac{1}{\mu_{11}+\mu_{12}} \tag{27}$$

So, the first time constant should match with $\frac{k_1}{\lambda_1}$ and the second time constant should match with $\frac{k_2}{\lambda_2}$.

A. Matching

If $\lambda_1 = \gamma_{11}+\gamma_{12} = \frac{1}{\mu_1+\mu_{12}}$, $\lambda_2 = \gamma_{11}-\gamma_{12} = \frac{1}{\mu_1-\mu_{12}}$ and if $k_1 = 17\mu N/m$ and $k_2 = 3\mu N/m$ then we are getting

Theory	Experiment
$\tau_1 = 0.0011s$	$\tau_1 = 0.0010s$
$\tau_2 = 0.0192s$	$\tau_1 = 0.016s$
$A_1 = 3.75 \times 10^{-4} \mu m^2$	$A_1 = 1.475 \times 10^{-4} \mu m^2$
$A_2 = 2.24 \times 10^{-4} \mu m^2$	$A_2 = 0.5 \times 10^{-4} \mu m^2$

Note : *We should divide the auto co-variance amplitudes by total number of data points (40000 here) to get ACF amplitudes and have to match them with the theoretical amplitudes which we can get from Eq. 24. Please cross check.*

III. CONCLUSIONS

Amplitudes are not matching!!! What is the problem?? Is here any mistake in the theoretical calculation or we are analyzing the data in the wrong way? Or we got some other effect?

In that prl paper of mainer they told : " In the experimentally obtained autocorrelation functions we also see a second exponential with a different time constant, both with and without a second bead present. This second time constant is typically an order of magnitude longer than t_x, and the corresponding amplitude is about 20% of the principal exponential. We attribute this second time constant to the motion of the bead along the weaker z axis of the trap, which couples to a small degree into the detector signal." So, we have to prove that it is not z coupling.