Unraveling the Molecular Dependence of Femtosecond Laser-induced Thermal Lens Spectroscopy in Fluids – Supplementary

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DERIVATIONS

S1. Probe beam propagation:

Under Fresnel diffraction approximation, amplitude of electric field at the center of the laser beam is given by[1]

$$U_{\mathrm{p}}\left(z_1+z_2,t\right)=\frac{j}{\lambda_{\mathrm{p}}z_2}e^{-j\frac{2\pi}{\lambda_{\mathrm{p}}}z_2}\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}U_{\mathrm{p}}\left(x,y,z_1\right)\exp\left(-j\frac{\pi}{\lambda_{\mathrm{p}}}\frac{x^2+y^2}{z_2}\right)dxdy \tag{1}$$

The amplitude of electric field at the output plane of the sample which is encountered with phase change $\Phi(x,y,t)$ is given as

$$U_{\mathrm{p}}\left(x,y,z_1\right)=\sqrt{\frac{2}{\pi}}\frac{1}{\omega_{1\mathrm{p}}}e^{-j\frac{2\pi}{\lambda_{\mathrm{p}}}z_1}\exp\left\{-j\left(\frac{\pi\left(x^2+y^2\right)}{\lambda_{\mathrm{p}}R_{1\mathrm{p}}}+\Phi\right)-\frac{\left(x^2+y^2\right)}{\omega_{1\mathrm{p}}^2}\right\} \tag{2}$$

Substituting Eq. (2) into Eq. (1) gives

$$U_{\mathrm{p}}\left(z_1+z_2,t\right)=C'\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}\exp\left\{-\left(1+jV\right)\frac{\left(x^2+y^2\right)}{\omega_{1\mathrm{p}}^2}\right\}e^{-j\Phi}dxdy \tag{3}$$

$$C'=\sqrt{\frac{2}{\pi}}\frac{1}{\omega_{1\mathrm{p}}}\frac{j}{\lambda_{\mathrm{p}}z_2}\exp\left\{-j\frac{2\pi}{\lambda_{\mathrm{p}}}\left(z_1+z_2\right)\right\} \tag{4}$$

$$V=\frac{\pi\omega_{1\mathrm{p}}^2}{\lambda_\mathrm{p}}\left(\frac{1}{R_{1\mathrm{p}}}+\frac{1}{z_2}\right) \tag{5}$$

Since the complex electric field amplitude of the TEM_{00} Gaussian laser beam at the entrance of a sample can be expressed as[1]

$$U_\mathrm{p}\left(x,y,z_1\right)=\sqrt{\frac{2}{\pi}}\frac{1}{\omega_{1\mathrm{p}}}e^{-jkz_1}\exp\left(-\frac{jk}{2q_1}\left(x^2+y^2\right)\right) \tag{6}$$

where, q_1 is the complex beam parameter which is defined as

$$\frac{1}{q_1}=\frac{1}{R_{1\mathrm{p}}}-\frac{j\lambda_\mathrm{p}}{\pi\omega_{1\mathrm{p}}^2} \tag{7}$$

$$\omega_{1\mathrm{p}}^2=\omega_{0\,\mathrm{p}}^2\left(1+\frac{z_1^2}{z_\mathrm{c}^2}\right) \tag{8}$$

$$\frac{1}{R_{1\mathrm{p}}}=\frac{z_1}{\left(z_1^2+z_\mathrm{c}^2\right)} \tag{9}$$

From Eq. (8) & (9), we get

$$V=\frac{z_1}{z_\mathrm{c}}+\frac{z_\mathrm{c}}{z_2}\left[1+\left(\frac{z_1}{z_\mathrm{c}}\right)^2\right] \tag{10}$$

The integration in Eq. (3) cannot be solved analytically. However, for the case of weak thermal lens effect (Φ<<1, phase shift is much less than 1, as is the case in Shen model[2]), we can write $\exp(-j\Phi)\approx 1-j\Phi$ [2,3]. Then Eq. (3) can be written as

$$U_\mathrm{p}\left(z_1+z_2,t\right)=C'\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}\exp\left\{-\left(1+jV\right)\frac{\left(x^2+y^2\right)}{\omega_{1\mathrm{p}}^2}\right\}(1-j\Phi)dxdy \tag{11}$$

or

$$U_{\mathrm{p}}(z_1+z_2,t)=C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-(1+jV)\frac{(x^2+y^2)}{\omega_{1\mathrm{p}}^2}\right\}dxdy \\ -C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}(j\Phi)\exp\left\{-(1+jV)\frac{(x^2+y^2)}{\omega_{1\mathrm{p}}^2}\right\}dxdy \tag{12}$$

We note that the phase change is change $\Phi(x,y,t)$ of the laser beam occurred due to variation in refractive index (n) is given by[2,3]

$$\Phi(x,y,t)=\frac{2\pi}{\lambda_p}l\left[n(x,y,t)-n(0,0,t)\right] \tag{13}$$

$$n(x,y,t)=n_0+\frac{\partial n}{\partial T}\Delta T(x,y,t) \tag{14}$$

$$\Phi(x,y,t)=\frac{2\pi}{\lambda_p}l\frac{dn}{dT}\left[\Delta T(x,y,t)-\Delta T(0,0,t)\right] \tag{15}$$

The absorbance and the beam divergence angle are small allowing the beam power and the beam radius to be taken as constant within the cell of path length of l.

The phase change Φ is obtained after substituting expression for temperature in Eq. (13) and defining $m=\left(\omega_{1\mathrm{p}}^2/\omega_{\mathrm{e}}^2\right)$, which is given as

$$\Phi=\frac{\theta}{t_{\mathrm{c}}}\int_0^t\frac{dt'}{1+2t'/t_{\mathrm{c}}}\left[\begin{array}{l}\left(1-\frac{2\left(v_xt'/\omega_{\mathrm{e}}\right)^2}{\left(1+2t'/t_{\mathrm{c}}\right)}\right)\left(1-\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{(x^2+y^2)}{\omega_{1\mathrm{p}}^2}\right\}\right)\\ -\frac{4xv_xt'/\omega_{\mathrm{e}}^2}{\left(1+2t'/t_{\mathrm{c}}\right)}\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{(x^2+y^2)}{\omega_{1\mathrm{p}}^2}\right\}\end{array}\right] \tag{16}$$

where, $\theta = -\dfrac{P_{\mathrm{e}}\alpha l \, dn/dt}{k\lambda_{\mathrm{p}}}$ and $t_{\mathrm{c}} = \dfrac{\omega_{\mathrm{e}}^2}{4D}$, θ relates to different physical properties of the sample that are responsible for the formation of thermal lens in such a way that its magnitude is directly related to the strength of the thermal lens.

For convenience, let us present Eq. (12) as

$$U_{\mathrm{p}}\left(z_1 + z_2, t\right) = F_a - F_b \tag{17}$$

such that

$$F_a = C' \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \exp\left\{-\left(1 + jV\right)\frac{\left(x^2 + y^2\right)}{\omega_{1\mathrm{p}}^2}\right\} dxdy \tag{18}$$

and

$$F_b = C' \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} (j\Phi) \exp\left\{-\left(1 + jV\right)\frac{\left(x^2 + y^2\right)}{\omega_{1\mathrm{p}}^2}\right\} dxdy \tag{19}$$

Solving for the F_a part, we get

$$F_a = C' \int_{-\infty}^{\infty} \exp\left\{-\left(1 + jV\right)\frac{x^2}{\omega_{1\mathrm{p}}^2}\right\} dx \int_{-\infty}^{\infty} \exp\left\{-\left(1 + jV\right)\frac{y^2}{\omega_{1\mathrm{p}}^2}\right\} dy \tag{20}$$

as $\mathrm{Re}\left(\dfrac{\left(1 + jV\right)}{\omega_{1\mathrm{p}}^2}\right) > 0$, we have

$$F_a = C'\left(\frac{\sqrt{\pi}}{\sqrt{\left(\left(1 + jV\right)/\omega_{1\mathrm{p}}^2\right)}}\right)\left(\frac{\sqrt{\pi}}{\sqrt{\left(\left(1 + jV\right)/\omega_{1\mathrm{p}}^2\right)}}\right) = C'\left(\frac{\pi\omega_{1\mathrm{p}}^2}{\left(1 + jV\right)}\right) = \frac{C}{1 + jV} \tag{21}$$

where, $C = \pi\omega_{1\mathrm{p}}^2 C'$

Eq. (19) can be solved as follows:

Substituting Eq. (16) into Eq. (19), we get

$$F_b = jC'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-(1+jV)\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\left(\frac{\theta}{t_{\mathrm{c}}}\int_0^t\frac{dt'}{1+2t'/t_{\mathrm{c}}}\left[\begin{array}{l}\left(1-\frac{2\left(v_x t'/\omega_{\mathrm{e}}\right)^2}{\left(1+2t'/t_{\mathrm{c}}\right)}\right)\left(1-\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\right)\\ -\frac{4xv_x t'/\omega_{\mathrm{e}}^2}{\left(1+2t'/t_{\mathrm{c}}\right)}\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\end{array}\right]\right)dxdy \quad (22)$$

$$F_b = j\frac{\theta}{t_{\mathrm{c}}}C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-(1+jV)\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\left(\begin{array}{l}\int_0^t\frac{dt'}{1+2t'/t_{\mathrm{c}}}\left(1-\frac{2\left(v_x t'/\omega_{\mathrm{e}}\right)^2}{\left(1+2t'/t_{\mathrm{c}}\right)}\right)\left(1-\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\right)\\ -\int_0^t\frac{4xv_x t'/\omega_{\mathrm{e}}^2}{\left(1+2t'/t_{\mathrm{c}}\right)^2}\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}dt'\end{array}\right)dxdy \quad (23)$$

Now, let

$$F_b = F_c + F_d \quad (24)$$

such that

$$F_c = j\frac{\theta}{t_{\mathrm{c}}}C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-(1+jV)\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\left(\int_0^t\frac{dt'}{1+2t'/t_{\mathrm{c}}}\left(1-\frac{2\left(v_x t'/\omega_{\mathrm{e}}\right)^2}{\left(1+2t'/t_{\mathrm{c}}\right)}\right)\left(1-\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\right)\right)dxdy \quad (25)$$

and

$$F_d = -j\frac{\theta}{t_{\mathrm{c}}}C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-(1+jV)\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\left(\int_0^t\frac{4xv_x t'/\omega_{\mathrm{e}}^2}{\left(1+2t'/t_{\mathrm{c}}\right)^2}\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}dt'\right)dxdy \quad (26)$$

This allows us to treat the two parts independently. Let us first solve the F_{c} part (Eq. (25)) and then use that knowledge to solve for F_{d} (Eq.(26)).

Eq. (25) can be solved as follows

$$F_c = j\frac{\theta}{t_{\mathrm{c}}}C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-(1+jV)\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\left(\int_0^t\frac{dt'}{1+2t'/t_{\mathrm{c}}}\left(1-\frac{2\left(v_x t'/\omega_{\mathrm{e}}\right)^2}{\left(1+2t'/t_{\mathrm{c}}\right)}\right)\left(1-\exp\left\{-\frac{2m}{\left(1+2t'/t_{\mathrm{c}}\right)}\frac{\left(x^2+y^2\right)}{\omega_{\mathrm{lp}}^2}\right\}\right)\right)dxdy \quad (27)$$

$$F_c = j\frac{\theta}{t_c}C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_0^t \frac{dt'}{1+2t'/t_c}\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left(1-\exp\left\{-\frac{2m}{\left(1+2t'/t_c\right)}\frac{\left(x^2+y^2\right)}{\omega_{1p}^2}\right\}\right)\exp\left\{-\left(1+jV\right)\frac{\left(x^2+y^2\right)}{\omega_{1p}^2}\right\}dxdy \quad (28)$$

$$F_c = j\frac{\theta}{t_c}C'\int_0^t\left(\frac{1}{1+2t'/t_c}\right)\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left[\begin{array}{l}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-\left(1+jV\right)\frac{\left(x^2+y^2\right)}{\omega_{1p}^2}\right\}dxdy \\ -\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left\{-\left(1+jV\right)\frac{\left(x^2+y^2\right)}{\omega_{1p}^2}\right\}\left(\exp\left\{-\frac{2m}{\left(1+2t'/t_c\right)}\frac{\left(x^2+y^2\right)}{\omega_{1p}^2}\right\}\right)dxdy\end{array}\right]dt' \quad (29)$$

Once again, let

$$F_c = F_{c1} + F_{c2} \quad (30)$$

such that

$$F_{c1} = jC'\frac{\theta}{t_c}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\left(\left(\exp\left\{-\left(1+jV\right)\frac{\left(x^2+y^2\right)}{\omega_{1p}^2}\right\}dxdy\right)\left(\int_0^t\frac{1}{1+2t'/t_c}\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)dt'\right)\right) \quad (31)$$

Using solution of Eq. * MERGEFORMAT (28) in above equation we get

$$F_{c1} = jC\frac{\theta}{t_c}\frac{1}{\left(1+jV\right)}\int_0^t\frac{1}{1+2t'/t_c}\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)dt' \quad (32)$$

and

$$F_{c2} = -jC'\frac{\theta}{t_c}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_0^t\left(\frac{dt'}{1+2t'/t_c}\right)\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left(\exp\left\{-\left(\left(1+jV\right)+\frac{2m}{\left(1+2t'/t_c\right)}\right)\frac{\left(x^2+y^2\right)}{\omega_{1p}^2}\right\}\right)dxdy \quad (33)$$

$$F_{c2} = -jC'\frac{\theta}{t_c}\int_0^t\left[\left(\frac{dt'}{1+2t'/t_c}\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\right)\left(\frac{\sqrt{\pi}\sqrt{\omega_{1p}^2}}{\sqrt{\left(1+jV\right)+\frac{2m}{\left(1+2t'/t_c\right)}}}\right)\left(\frac{\sqrt{\pi}\sqrt{\omega_{1p}^2}}{\sqrt{\left(1+jV\right)+\frac{2m}{\left(1+2t'/t_c\right)}}}\right)\right] \quad (34)$$

as $\mathrm{Re}\left(\frac{\left(1+jV\right)+\frac{2m}{\left(1+2t'/t_c\right)}}{\omega_{1p}^2}\right) > 0$

$$F_{c2} = -jC\frac{\theta}{t_c}\int_0^t \frac{\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)}{\left(1+2t'/t_c\right)\left(\left(1+jV\right)+\frac{2m}{\left(1+2t'/t_c\right)}\right)}dt' \tag{35}$$

$$F_{c2} = -jC\frac{\theta}{t_c}\frac{1}{\left(1+jV\right)}\int_0^t \frac{\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)}{\left(1+2t'/t_c\right)\left(1+\frac{2m}{\left(1+2t'/t_c\right)\left(1+jV\right)}\right)}dt' \tag{36}$$

With this simplification, we can get back to F_c in Eq. (30), as follows:

$$F_c = jC\frac{\theta}{t_c}\frac{1}{\left(1+jV\right)}\left(\int_0^t \frac{1}{1+2t'/t_c}\left(\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left[1-\frac{1}{\left(1+\frac{2m}{\left(1+2t'/t_c\right)\left(1+jV\right)}\right)}\right]\right)dt'\right) \tag{37}$$

$$F_c = jC\frac{\theta}{t_c}\frac{1}{\left(1+jV\right)}\left(\int_0^t \frac{1}{1+2t'/t_c}\left(\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left[1-\frac{1}{\left(\frac{\left(1+2t'/t_c\right)\left(1+jV\right)+2m}{\left(1+2t'/t_c\right)\left(1+jV\right)}\right)}\right]\right)dt'\right) \tag{38}$$

$$F_c = jC\frac{\theta}{t_c}\frac{1}{\left(1+jV\right)}\left(\int_0^t \frac{1}{1+2t'/t_c}\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left[1-\frac{\left(1+2t'/t_c\right)\left(1+jV\right)}{\left(1+2t'/t_c\right)\left(1+jV\right)+2m}\right]dt'\right) \tag{39}$$

$$F_c = j\frac{\theta}{t_c}\frac{C}{\left(1+jV\right)}\left(\int_0^t \frac{1}{1+2t'/t_c}\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left[\frac{2m}{\left(1+2t'/t_c\right)\left(1+jV\right)+2m}\right]dt'\right) \tag{40}$$

$$F_c = j\frac{\theta}{t_c}\frac{C}{\left(1+jV\right)}\left(\int_0^t \frac{1}{1+2t'/t_c}\left(1-\frac{2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)}\right)\left[\frac{1}{\left(1+2t'/t_c\right)\left(\frac{1+jV}{2m}\right)+1}\right]dt'\right) \tag{41}$$

$$F_c = j\frac{\theta}{t_c}\frac{C}{\left(1+jV\right)}\int_0^t \frac{\left(1+2t'/t_c\right)-2\left(v_x t'/\omega_e\right)^2}{\left(1+2t'/t_c\right)^3\left(\frac{1+jV}{2m}\right)+\left(1+2t'/t_c\right)^2}dt' \tag{42}$$

$$Let \quad a=\frac{2}{t_c}, \qquad b=\frac{1}{2m} \qquad f=\frac{V}{2m} \qquad p=2\left(v_x/\omega_e\right)^2 \tag{43}$$

$$F_c = j\frac{\theta}{t_c}\frac{C}{(1+jV)}\int_0^t \frac{1+at'-pt'^2}{(1+at')^3(b+jf)+(1+at')^2}dt' \quad (44)$$

$$\text{Now, } \int_0^t \frac{1+at'-pt'^2}{(1+at')^3(b+jf)+(1+at')^2}dt' = \frac{1}{2a^3}\left[\begin{array}{l} -2j\left(a^2+\frac{p(b+jf+1)^2}{b+jf}\right)\left(\begin{array}{l}\tan^{-1}\left(\frac{aft+f}{abt+b+1}\right)- \\ 2j\tan^{-1}\left(\frac{f}{b+1}\right)\end{array}\right) \\ -\left(a^2+\frac{p(b+jf+1)^2}{b+jf}\right)\ln\left\{\frac{(abt+b)^2+2b(at+1)+(aft+f)^2+1}{b^2+2b+f^2+1}\right\} \\ +\frac{2p}{at+1}-2p+2\left(a^2+p(b+jf+2)\right)\ln\{at+1\} \end{array}\right] \quad (45)$$

So,

$$F_c = j\frac{\theta}{t_c}\frac{C}{(1+jV)}\frac{1}{2a^3}\left[\begin{array}{l} -2j\left(a^2+\frac{p(b+jf+1)^2}{b+jf}\right)\tan^{-1}\left(\frac{\left(\frac{aft+f}{abt+b+1}\right)-\left(\frac{f}{b+1}\right)}{1+\left(\frac{aft+f}{abt+b+1}\right)\left(\frac{f}{b+1}\right)}\right) \\ -\left(a^2+\frac{p(b+jf+1)^2}{b+jf}\right)\ln\left\{\frac{(abt+b)^2+2b(at+1)+(aft+f)^2+1}{b^2+2b+f^2+1}\right\} \\ +\frac{2p}{at+1}-2p+2\left(a^2+\frac{p(b+jf+1)^2}{b+jf}-\frac{p}{b+jf}\right)\ln\{at+1\} \end{array}\right] \quad (46)$$

$$F_c = j\frac{\theta}{t_c}\frac{C}{(1+jV)}\frac{1}{2a^3}\left[\begin{array}{l} -\left(a^2+\frac{p(b+jf+1)^2}{b+jf}\right) \\ \left[2j\tan^{-1}\left(\frac{\left(\frac{aft+f}{abt+b+1}\right)-\left(\frac{f}{b+1}\right)}{1+\left(\frac{aft+f}{abt+b+1}\right)\left(\frac{f}{b+1}\right)}\right)+\ln\left\{\frac{(abt+b)^2+2b(at+1)+(aft+f)^2+1}{b^2+2b+f^2+1}\right\}\right] \\ +\frac{2p}{at+1}-2p+\left(a^2+\frac{p(b+jf+1)^2}{b+jf}\right)2\ln\{at+1\} \\ -\left(\frac{2p}{b+jf}\right)\ln\{at+1\} \end{array}\right] \quad (47)$$

$$F_c = j\frac{\theta}{t_c}\frac{C}{(1+jV)}\frac{1}{2a^3}\left[\begin{array}{l} -\left(a^2+\dfrac{p(b+jf+1)^2}{b+jf}\right) \\ \left[2j\tan^{-1}\left(\dfrac{\left(\dfrac{aft+f}{abt+b+1}\right)-\left(\dfrac{f}{b+1}\right)}{1+\left(\dfrac{aft+f}{abt+b+1}\right)\left(\dfrac{f}{b+1}\right)}\right)+\ln\left\{\dfrac{(abt+b)^2+2b(at+1)+(aft+f)^2+1}{(b^2+2b+f^2+1)(at+1)^2}\right\}\right] \\ +\dfrac{2p}{at+1}-2p-\left(\dfrac{2p}{b+jf}\right)\ln\{at+1\} \end{array}\right] \tag{48}$$

$$F_c = j\frac{\theta}{t_c}\frac{C}{(1+jV)}\frac{1}{2a}\left[\begin{array}{l} -\left(1+\dfrac{p(b+jf+1)^2}{a^2(b+jf)}\right) \\ \left[2j\tan^{-1}\left(\dfrac{\left(\dfrac{aft+f}{abt+b+1}\right)-\left(\dfrac{f}{b+1}\right)}{1+\left(\dfrac{aft+f}{abt+b+1}\right)\left(\dfrac{f}{b+1}\right)}\right)+\ln\left\{\dfrac{(abt+b)^2+2b(at+1)+(aft+f)^2+1}{(b^2+2b+f^2+1)(at+1)^2}\right\}\right] \\ -\dfrac{2p}{a^2}\left[\left(\dfrac{1}{b+jf}\right)\ln\{at+1\}+\left(1-\dfrac{1}{at+1}\right)\right] \end{array}\right] \tag{49}$$

where

$$\frac{2p}{a^2} = \frac{2\times 2(v_x/\omega_e)^2\times t_c^{\,2}}{4} = (v_x t_c/\omega_e)^2 = \left(\frac{v_x\omega_e}{4D}\right)^2 = P_E^2 \tag{50}$$

and

$$\frac{p(b+jf+1)^2}{a^2(b+jf)} = \frac{P_E^2}{2}\frac{(1+\mathrm{j}V+2m)^2}{2m(1+\mathrm{j}V)} \tag{51}$$

The factor $\frac{\omega_e v_x}{4D}$, is the well-known dimensionless parameter called the Peclet number. It is the ratio of convective to conductive heat transfer rates[4]. ω_e has been chosen as characteristics length as it is imposed by laser beam size itself. Thus,

$$\text{Peclet Number } (P_E) = \frac{\text{Rate of convection}}{\text{Rate of conduction}} = \frac{\omega_e v_x}{4D} \tag{52}$$

Now the logarithm part in Eq. * MERGEFORMAT (59) is simplified separately as follows:

$$\ln\left\{\frac{(abt+b)^2+2b(at+1)+(aft+f)^2+1}{(b^2+2b+f^2+1)(at+1)^2}\right\}=\ln\left\{\frac{b^2(at+1)^2+2b(at+1)+f^2(at+1)^2+1}{(b^2+2b+f^2+1)(at+1)^2}\right\}$$

$$=\ln\left\{\frac{b^2+f^2+\frac{2b}{(at+1)}+\frac{1}{(at+1)^2}}{(b^2+2b+f^2+1)}\right\}=\ln\left\{\frac{\left(b+\frac{1}{(at+1)}\right)^2+f^2}{(b+1)^2+f^2}\right\}=\ln\left\{\frac{\left(\frac{1}{2m}+\frac{1}{\left(1+2t/t_c\right)}\right)^2+\left(\frac{V}{2m}\right)^2}{\left(\frac{1}{2m}+1\right)^2+\left(\frac{V}{2m}\right)^2}\right\}$$

$$=\ln\left\{\frac{\left(1+\frac{2m}{\left(1+2t/t_c\right)}\right)^2+V^2}{(1+2m)^2+V^2}\right\} \tag{53}$$

Similarly, tangent inverse part in Eq. (49) is simplified as follows:

$$\tan^{-1}\left(\frac{\left(\frac{aft+f}{abt+b+1}\right)-\left(\frac{f}{b+1}\right)}{1+\left(\frac{aft+f}{abt+b+1}\right)\left(\frac{f}{b+1}\right)}\right)=\tan^{-1}\left(\frac{(aft+f)(b+1)-(f)(abt+b+1)}{(abt+b+1)(b+1)+(aft+f)(f)}\right)$$

$$=\tan^{-1}\left(\frac{f(at+1)}{(at+1)}\left[\frac{1-\frac{1}{at+1}}{\left(b+\frac{1}{(at+1)}\right)(b+1)+(f)^2}\right]\right)=\tan^{-1}\left(\left[\frac{f\left(\frac{at}{at+1}\right)}{\left(b+\frac{1}{(at+1)}\right)(b+1)+(f)^2}\right]\right)$$

$$=\tan^{-1}\left(\frac{\left(\frac{V}{2m}\right)\left(\frac{2t/t_c}{1+2t/t_c}\right)}{\left(\frac{1}{2m}+\frac{1}{(1+2t/t_c)}\right)\left(\frac{1}{2m}+1\right)+\left(\frac{V}{2m}\right)^2}\right)=\tan^{-1}\left(\frac{2mV\left(\frac{2t/t_c}{1+2t/t_c}\right)}{\left(1+\frac{2m}{(1+2t/t_c)}\right)(1+2m)+V^2}\right)$$

$$=\tan^{-1}\left(\frac{2mV}{\left(\frac{1+2t/t_c}{2t/t_c}\right)\left[\left(1+\frac{2m}{(1+2t/t_c)}\right)(1+2m)+V^2\right]}\right)=\tan^{-1}\left(\frac{2mV}{\left(\frac{t_c}{2t}\right)\left[(1+2m+V^2)(1+2t/t_c)+2m(1+2m)\right]}\right)$$

$$=\tan^{-1}\left(\frac{2mV}{(1+2m+V^2)+\left[(1+2m)^2+V^2\right]\left(\frac{t_c}{2t}\right)}\right) \tag{54}$$

Once these simplifications are attained, we can finally get F_c as:

$$F_c = j\frac{\theta}{4}\frac{C}{(1+jV)}\left[\begin{array}{l} -\left(1+\frac{P_{\mathrm{E}}^2}{2}\frac{(1+\mathrm{j}V+2m)^2}{2m(1+\mathrm{j}V)}\right)\left[2j\tan^{-1}\left(\frac{2mV}{(1+2m+V^2)+\left[(1+2m)^2+V^2\right]\left(\frac{t_c}{2t}\right)}\right)+\ln\left\{\frac{\left(1+\frac{2m}{\left(1+2t/t_c\right)}\right)^2+V^2}{(1+2m)^2+V^2}\right\}\right] \\ -P_{\mathrm{E}}^2\left[\left(\frac{2m}{(1+\mathrm{j}V)}\right)\ln\{2t/t_{\mathrm{c}}+1\}+\left(\frac{2t/t_{\mathrm{c}}}{1+2t/t_{\mathrm{c}}}\right)\right] \end{array}\right]$$
(55)

Now, let us get back to F_d in Eq. (26):

$$F_d = j\frac{\theta}{t_{\mathrm{c}}}C'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\left(\exp\left\{-(1+jV)\frac{(x^2+y^2)}{\omega_{\mathrm{lp}}^2}\right\}\right)\left(-\int_0^t\frac{4xv_xt'/\omega_{\mathrm{e}}^2}{(1+2t'/t_{\mathrm{c}})^2}\left(\exp\left\{-\frac{2m}{(1+2t'/t_{\mathrm{c}})}\frac{(x^2+y^2)}{\omega_{\mathrm{lp}}^2}\right\}\right)dt'\right)dxdy \tag{56}$$

$$F_d = -j\frac{\theta}{t_{\mathrm{c}}}C'\int_0^t\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\left(\frac{4xv_xt'/\omega_{\mathrm{e}}^2}{(1+2t'/t_{\mathrm{c}})^2}\left(\exp\left\{-(1+jV)\frac{(x^2+y^2)}{\omega_{\mathrm{lp}}^2}\right\}\exp\left\{-\frac{2m}{(1+2t'/t_{\mathrm{c}})}\frac{(x^2+y^2)}{\omega_{\mathrm{lp}}^2}\right\}\right)\right)dxdydt' = 0 \tag{57}$$

Thus, $F_b = F_c$, and $U_{\mathrm{p}}(z_1+z_2,t) = F_a - F_b$ becomes:

$$U_{\mathrm{p}}(z_1+z_2,t) = \frac{C}{(1+jV)}\left[1-\frac{j\theta}{4}\left[\begin{array}{l} -\left(1+\frac{P_{\mathrm{E}}^2}{2}\frac{(1+\mathrm{j}V+2m)^2}{2m(1+\mathrm{j}V)}\right)\left(\begin{array}{l} 2\mathrm{j}\tan^{-1}\left\{\frac{2mV}{\left[(1+2m)^2+V^2\right](t_{\mathrm{c}}/2t)+1+2m+V^2}\right\} \\ +\ln\left\{\frac{\left[1+2m/(1+2t/t_{\mathrm{c}})\right]^2+V^2}{(1+2m)^2+V^2}\right\} \end{array}\right) \\ -P_{\mathrm{E}}^2\left[\frac{2m}{(1+\mathrm{j}V)}\ln(1+2t/t_{\mathrm{c}})+\left(\frac{2t/t_{\mathrm{c}}}{1+2t/t_{\mathrm{c}}}\right)\right] \end{array}\right]\right] \tag{58}$$

Finally, the intensity at center of the laser beam $I(t)=\left|U_{\mathrm{p}}\left(z_1+z_2,t\right)\right|^2$ comes out to be:

$$S(t)=\frac{I(t)}{I(0)}=\left|1-\frac{\mathrm{j}\theta}{4}\left[\begin{array}{l}-\left(1+\frac{P_{\mathrm{E}}^2}{2}\frac{\left(1+\mathrm{j}V+2m\right)^2}{2m\left(1+\mathrm{j}V\right)}\right)\left(\begin{array}{l}2\mathrm{j}\tan^{-1}\left\{\frac{2mV}{\left[\left(1+2m\right)^2+V^2\right]\left(t_{\mathrm{c}}/2t\right)+1+2m+V^2}\right\}\\+\ln\left\{\frac{\left[1+2m/\left(1+2t/t_{\mathrm{c}}\right)\right]^2+V^2}{\left(1+2m\right)^2+V^2}\right\}\end{array}\right)\\-P_{\mathrm{E}}^2\left[\frac{2m}{\left(1+\mathrm{j}V\right)}\ln\left(1+2t/t_{\mathrm{c}}\right)+\left(\frac{2t/t_{\mathrm{c}}}{1+2t/t_{\mathrm{c}}}\right)\right]\end{array}\right]\right|^2 \tag{59}$$

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