Ultrashort Laser Pulses II

Relative importance of spectrum and spectral phase

Taylor expansion of the spectral phase

2nd and higher-order spectral phase distortions

The linearly chirped Gaussian pulse

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The frequency-domain electric field

The frequency-domain equivalents of the **intensity** and **temporal phase** are the **spectrum** and **spectral phase**.

Fourier-transforming the pulse electric field:

$$\mathcal{E}(t) = \tfrac{1}{2}\sqrt{I(t)}\,\exp\{i[\omega_0 t - \phi(t)]\} + c.c.$$

yields:

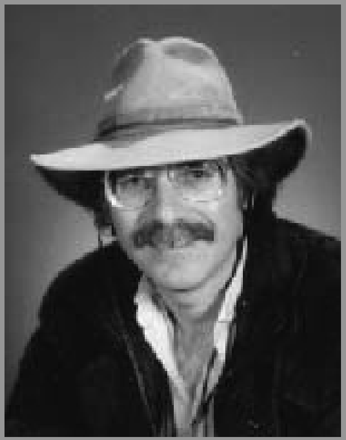
Note that ϕ and φ are different!

$$\tilde{\mathcal{E}}(\omega) = \tfrac{1}{2}\sqrt{S(\omega-\omega_0)}\,\exp\{-i[\varphi(\omega-\omega_0)]\} + \tfrac{1}{2}\sqrt{S(-\omega-\omega_0)}\,\exp\{+i[\varphi(-\omega-\omega_0)]\}$$

The **spectral phase** is the key quantity in determining the shape and duration of a short pulse.

The relative importance of intensity and phase

Photographs of Rick and Linda:



Composite photograph made using the spectral intensity of Linda's photo and the spectral phase of Rick's (and inverse-Fourier-transforming)

Composite photograph made using the spectral intensity of Rick's photo and the spectral phase of Linda's (and inverse-Fourier-transforming)

The spectral phase is more important for determining the result!

Frequency-domain phase expansion

Recall the Taylor series for $\varphi(\omega)$:

$$\boxed{\varphi(\omega) = \varphi_0 + \varphi_1 \frac{\omega - \omega_0}{1!} + \varphi_2 \frac{(\omega - \omega_0)^2}{2!} + \ldots}$$

where φ_0 is the **absolute phase**

$$\varphi_1 = \left.\frac{d\varphi}{d\omega}\right|_{\omega=\omega_0}$$ is the **group delay**

$$\varphi_2 = \left.\frac{d^2\varphi}{d\omega^2}\right|_{\omega=\omega_0}$$ is the **group-delay dispersion**

Only the first few terms are typically required to describe well-behaved pulses. Of course, one can also consider badly behaved pulses, for which the higher-order terms in $\varphi(\omega)$ must be retained.

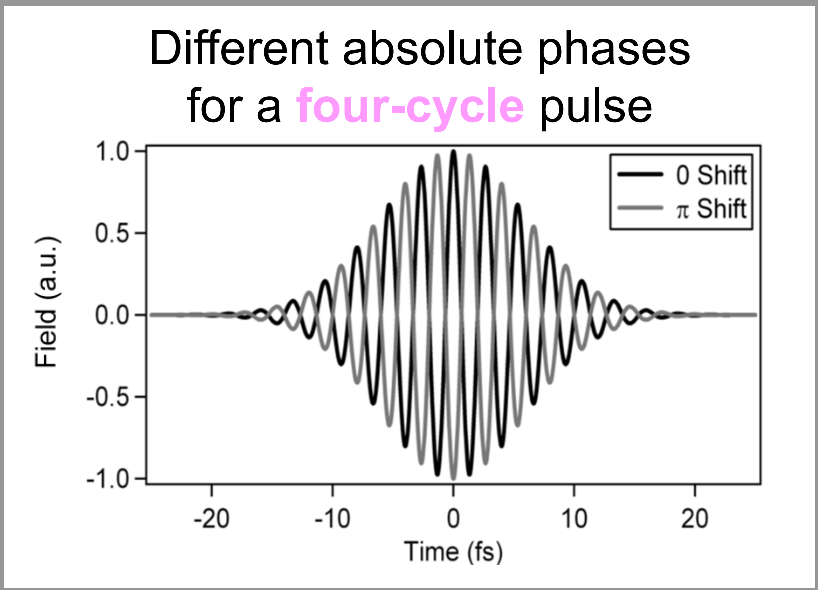
Zeroth-order phase: the absolute phase

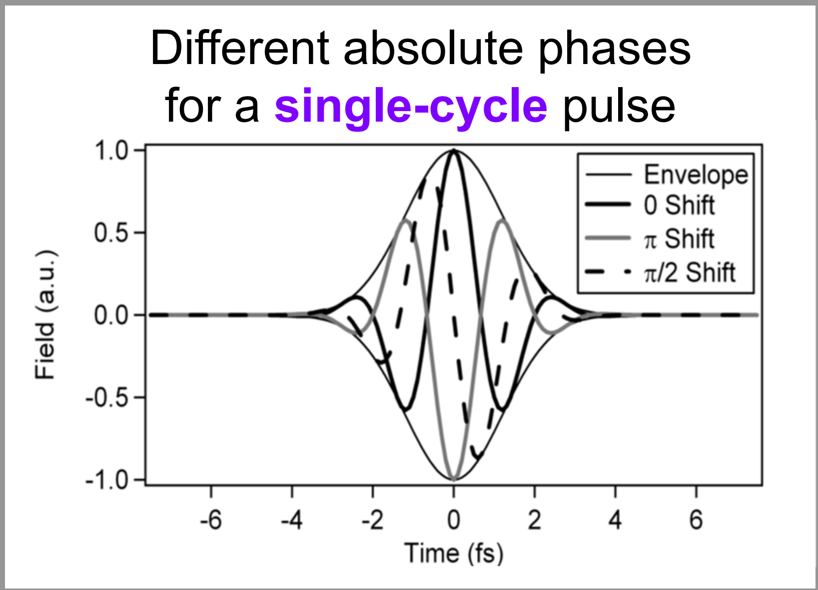
The absolute phase is the same in both the time and frequency domains.

$$f(t)\exp(i\phi_0) \supset F(\omega)\exp(i\phi_0)$$

An absolute phase of $\pi/2$ will turn a cosine carrier wave into a sine.

It's usually irrelevant,* unless the pulse is only a cycle or so long.





Notice that the two **four-cycle** pulses look similar, but the three **single-cycle** pulses are all quite different.

*there are some interesting and important exceptions to this!

Group delay

Just as the time derivative of the temporal phase tells us about the variation of the (local) center frequency as the pulse evolves in time:

$$\frac{d\phi}{dt} = \omega_0 - \omega_{inst}(t)$$

So, the frequency derivative of the spectral phase tells us about the variation of the center of the pulse envelope at different frequencies. We call this the ***group delay vs. frequency***, $t_{gr}(\omega)$, given by:

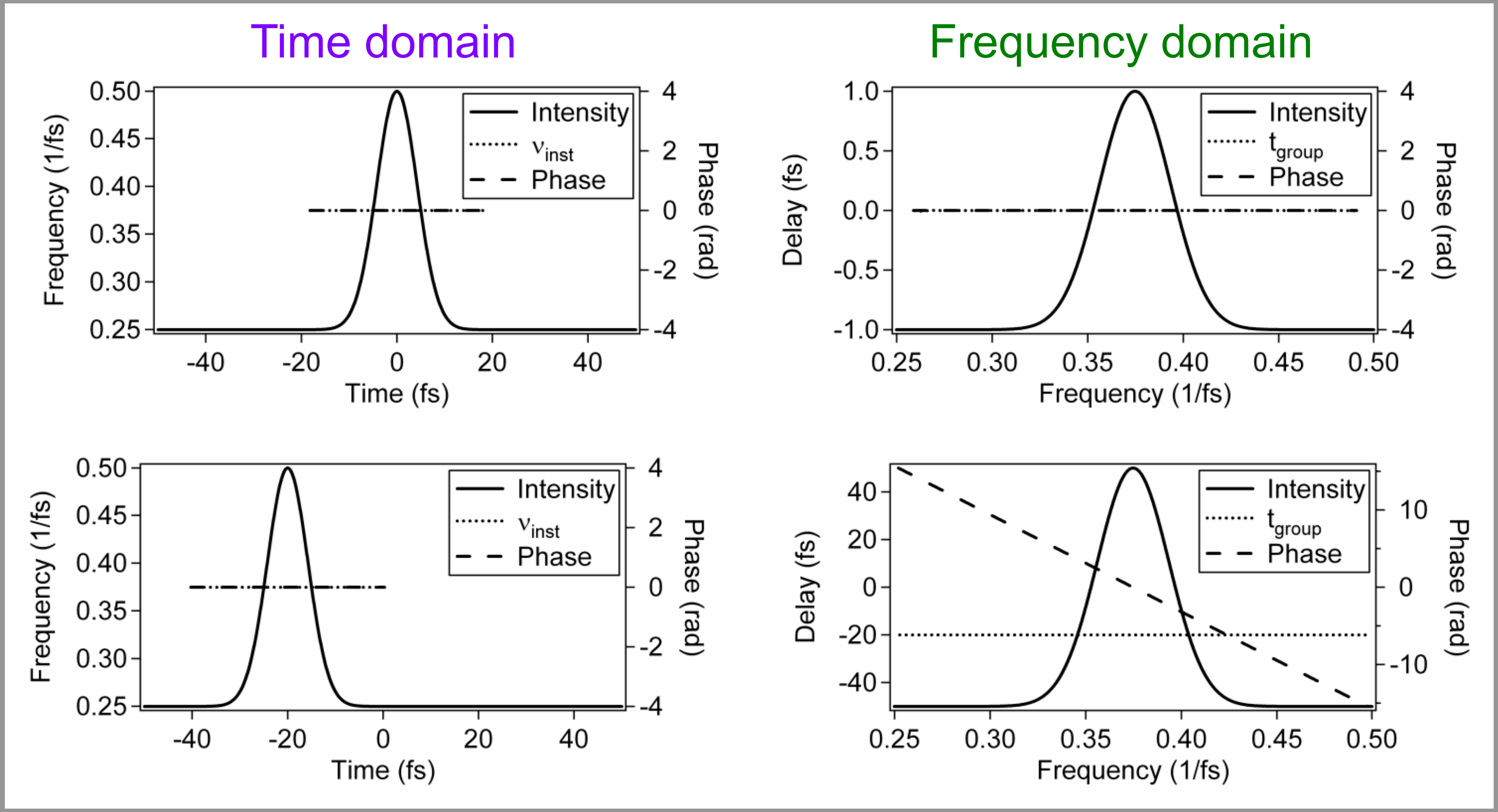
$$\boxed{t_{gr}(\omega) = d\varphi / d\omega}$$

First-order phase in frequency: a shift in time

By the Fourier-transform Shift Theorem, $f(t-\varphi_1) \supset F(\omega)\exp(i\omega\varphi_1)$

$\varphi_1 = 0$

$\varphi_1 = -20$ fs



Note that φ_1 does not affect the instantaneous frequency, but does affect the delay of the pulse envelope, i.e., the group delay = φ_1.

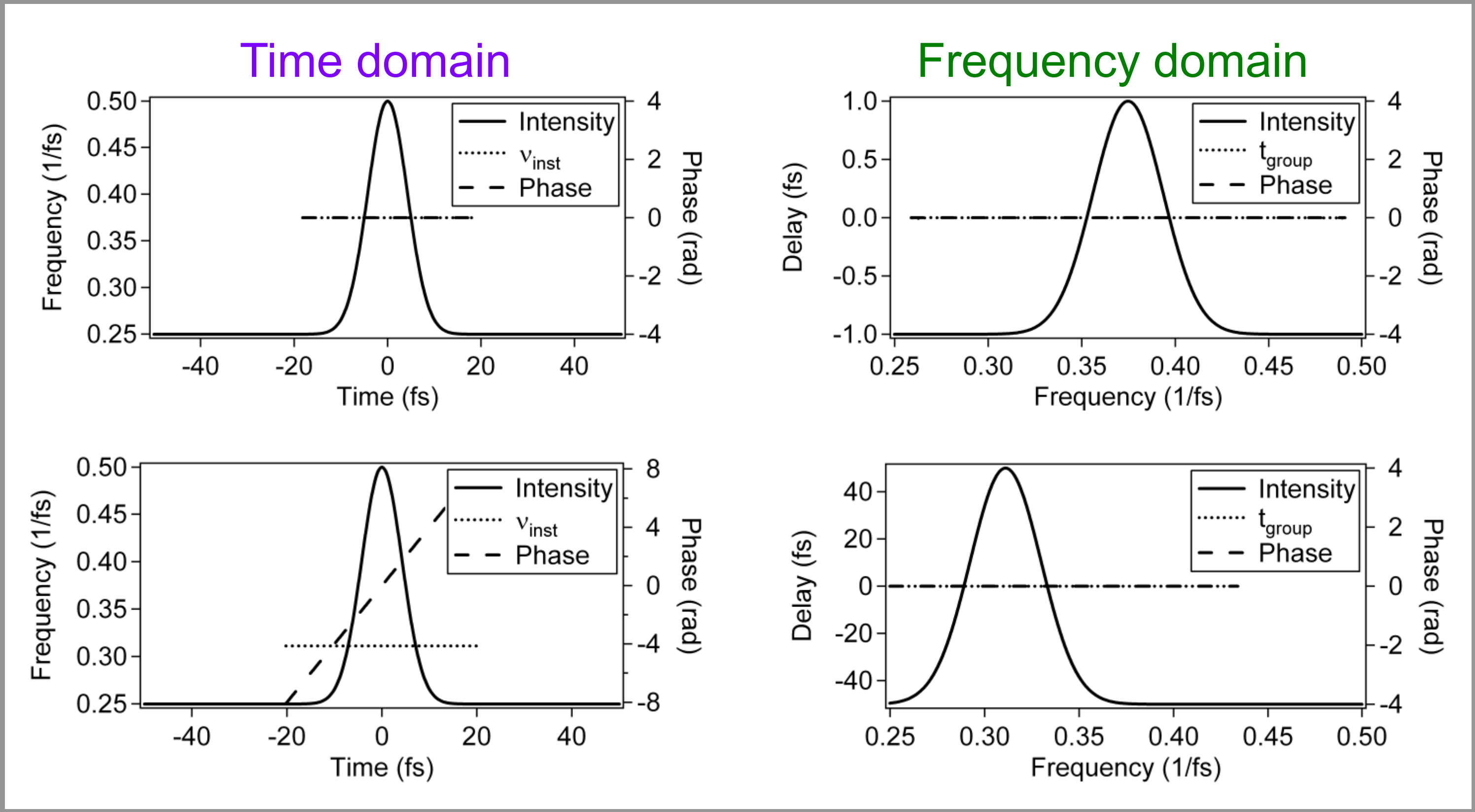
First-order phase in time: a frequency shift

By the Inverse-Fourier-transform Shift Theorem,

$$F(\omega - \phi_1) \subset f(t)\exp(-i\,\phi_1\,t)$$

$\phi_1 = 0\,/\,$fs

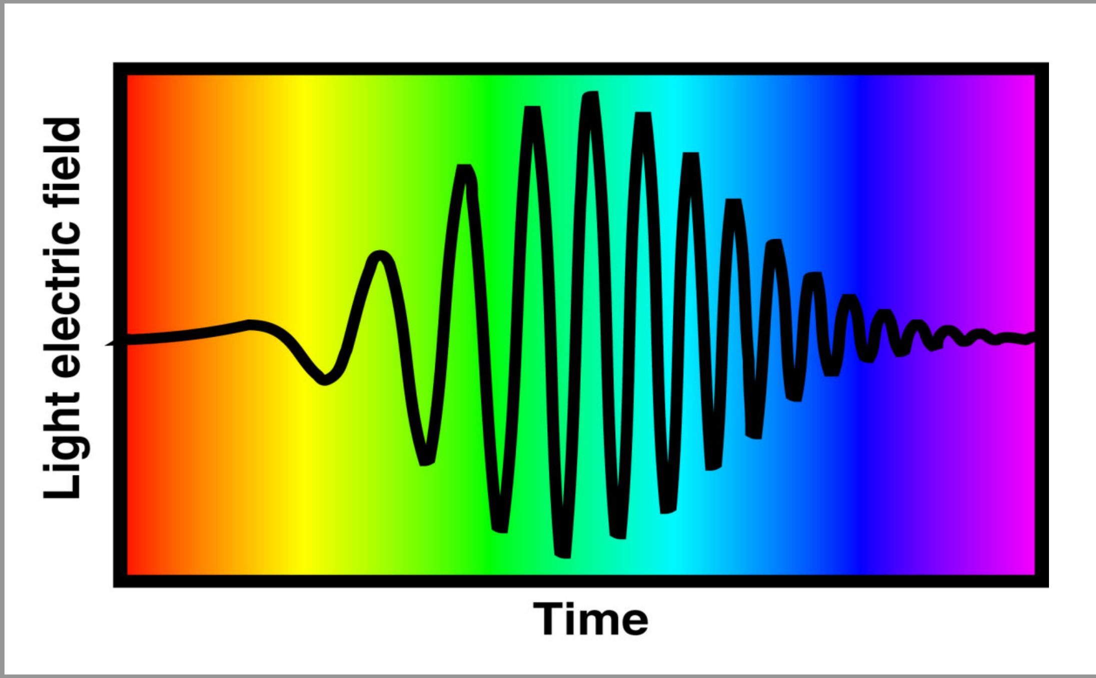
$\phi_1 = -.07\,/\,$fs



Note that ϕ_1 does not affect the group delay, but it does affect the shift of the spectrum, i.e., the change in instantaneous frequency = – ϕ_1.

Second-order phase: the linearly chirped pulse

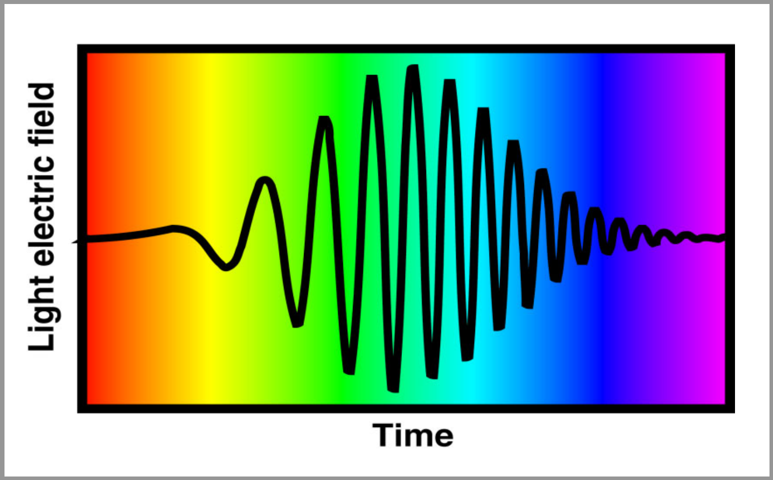
A pulse can have a frequency that varies in time.



This pulse increases its frequency linearly in time (from red to blue).

In analogy to bird sounds, this pulse is called a **chirped** pulse.

The Linearly Chirped Gaussian Pulse



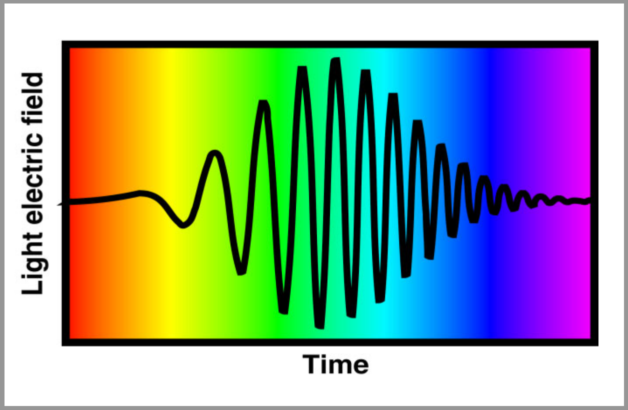
We can write a linearly chirped Gaussian pulse mathematically as:

$$\underbrace{\mathcal{E}(t) = E_0 \exp\left[-(t/\tau_G)^2\right]}_{\text{Gaussian amplitude}} \exp\left[i\left(\underbrace{\omega_0 t}_{\text{Carrier wave}} + \underbrace{\beta t^2}_{\text{Chirp}}\right)\right]$$

This expression puts t = 0 at the center (peak) of the pulse.

Note that for $\beta > 0$, when $t < 0$, the two terms partially cancel, so the phase changes slowly with time (so the frequency is low). And when $t > 0$, the terms add, and the phase changes more rapidly (so the frequency is larger).

The instantaneous frequency vs. time for a chirped pulse



The chirped pulse from the last slide has:

$$\mathscr{E}(t) \propto \exp\left[i\left(\omega_0 t - \phi(t)\right)\right]$$

where:

$$\phi(t) = -\beta t^2$$

Thus the instantaneous frequency is: $\omega_{inst}(t) \equiv \omega_0 - d\phi / dt$

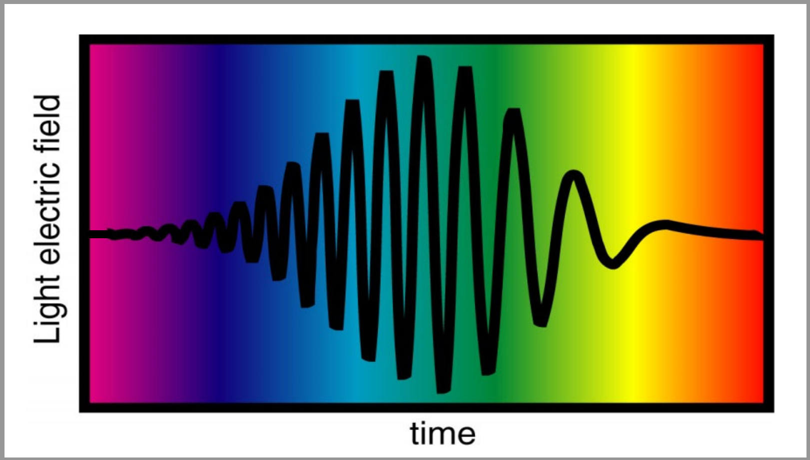
which is: $\omega_{inst}(t) = \omega_0 + 2\beta t$

So the frequency increases linearly with time.

The Negatively Chirped Pulse

We have been considering a pulse whose frequency **increases** linearly with time: a **positively** chirped pulse ("up chirp").

One can also have a **negatively** chirped (Gaussian) pulse, whose instantaneous frequency **decreases** with time ("down chirp").



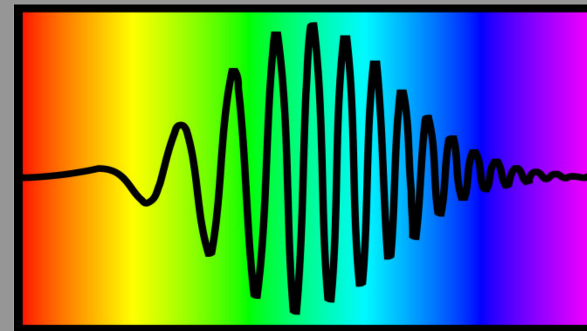
We simply allow β to be **negative** in the expression for the pulse:

$$\mathscr{E}(t) = E_0 \exp\left[-\left(t/\tau_G\right)^2\right]\exp\left[i\left(\omega_0 t + \beta t^2\right)\right]$$

And the instantaneous frequency will decrease with time:

$$\omega_{inst}(t) = \omega_0 + 2\beta t = \omega_0 - 2\left|\beta\right| t$$

The Fourier transform of a chirped pulse



Writing a linearly chirped Gaussian pulse as:

$$\mathcal{E}(t) \propto E_0 \exp\left[-\alpha t^2\right] \exp\left[i\left(\omega_0 t + \beta t^2\right)\right] + c.c \qquad \text{where } \alpha \propto \left(1/\tau_G\right)^2$$

or:

$$\mathcal{E}(t) \propto E_0 \exp\left[-\left(\alpha - i\beta\right)t^2\right] \exp\left[i\left(\omega_0 t\right)\right] + c.c.$$

Looks like a Gaussian with a complex width!

Fourier-Transforming yields:

neglecting the negative-frequency term due to the c.c.

$$\tilde{\mathcal{E}}(\omega) \propto E_0 \exp\left[-\frac{1/4}{\alpha - i\beta}\left(\omega - \omega_0\right)^2\right]$$

A chirped Gaussian pulse Fourier transforms to another Gaussian.

Rationalizing the denominator and separating the real and imaginary parts:

$$\tilde{\mathcal{E}}(\omega) \propto \underbrace{\boxed{E_0 \exp\left[-\frac{\alpha/4}{\alpha^2 + \beta^2}\left(\omega - \omega_0\right)^2\right]}}_{\sqrt{S(\omega)}} \exp\left[-i\overbrace{\boxed{\frac{\beta/4}{\alpha^2 + \beta^2}\left(\omega - \omega_0\right)^2}}^{\varphi(\omega)}\right]$$

The pulse duration and spectral width

In the time domain, we have:

$$\mathscr{E}(t) \propto E_0 \exp\left[-\alpha t^2\right] \exp\left[i\left(\omega_0 t + \beta t^2\right)\right] + c.c$$

So the intensity $I(t)$ is given by: $I(t) \propto E_0^{\,2} \exp\left[-2\alpha t^2\right]$

And the duration (full-width at half-max) is: $\tau_{FWHM} = \sqrt{2\ln 2/\alpha}$

independent of the chirp parameter β

Meanwhile, in the frequency domain, we have:

$$\tilde{\mathscr{E}}(\omega) \propto E_0 \exp\left[-\frac{\alpha/4}{\alpha^2+\beta^2}(\omega-\omega_0)^2\right] \exp\left[-i\frac{\beta/4}{\alpha^2+\beta^2}(\omega-\omega_0)^2\right]$$

So the spectrum is:

$$S(\omega) \propto E_0^{\,2} \exp\left[-\frac{\alpha/2}{\alpha^2+\beta^2}(\omega-\omega_0)^2\right]$$

depends on β!

And the width of the spectrum (FWHM) is: $\Delta\omega_{FWHM} = \sqrt{\frac{8\ln 2}{\alpha}\left(\alpha^2+\beta^2\right)}$

The pulse duration and spectral width (cont.)

Pulse duration (FWHM) is: $\tau_{FWHM} = \sqrt{2\ln 2/\alpha}$

Spectral width (FWHM) is: $\Delta\omega_{FWHM} = \sqrt{\frac{8\ln 2}{\alpha}\left(\alpha^2+\beta^2\right)}$

As the chirp increases (β gets bigger), we see that the pulse width doesn't change, but the spectrum gets broader.

We know that a broader spectrum can produce a shorter pulse. The shortest possible pulse duration is inversely proportional to the spectral width:

$$\tau_{FWHM\text{, shortest possible}} \propto \frac{1}{\Delta\omega_{FWHM}}$$

Therefore:

A chirped pulse is longer than it could be (given its spectral width)!

Chirp not only means that different frequencies arrive at different times. It also means that the pulse is longer than the theoretical minimum duration.

The group delay vs. ω for a chirped pulse

The group delay of a wave is the derivative of the spectral phase:

$$t_{gr}(\omega) \equiv d\varphi / d\omega$$

For a linearly chirped Gaussian pulse, the spectral phase is:

$$\varphi(\omega) = \frac{\beta / 4}{\alpha^2 + \beta^2}\left(\omega - \omega_0\right)^2$$

So:

$$t_{gr} = \frac{\beta / 2}{\alpha^2 + \beta^2}\left(\omega - \omega_0\right)$$

The group delay is a linear function of frequency.

This is not the inverse of the instantaneous frequency, which is:

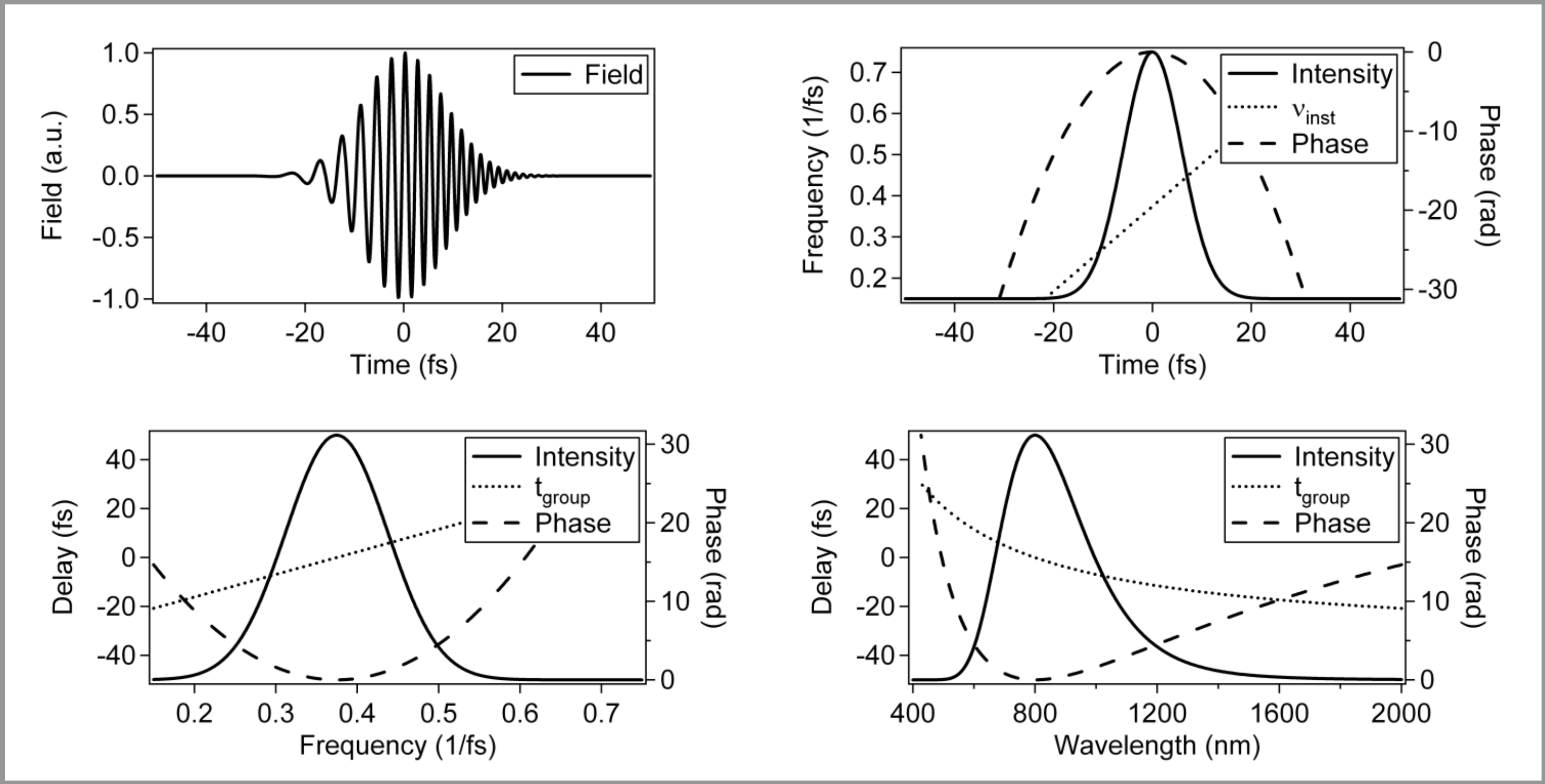
$$\omega_{inst}(t) = \omega_0 + 2\beta t$$

But when the pulse is long ($\alpha \to 0$): $t_{gr} = \dfrac{1}{2\beta}\left(\omega - \omega_0\right)$

which is the same result as $1/\omega_{inst}(t)$.

2^{nd}-order phase: positive linear chirp

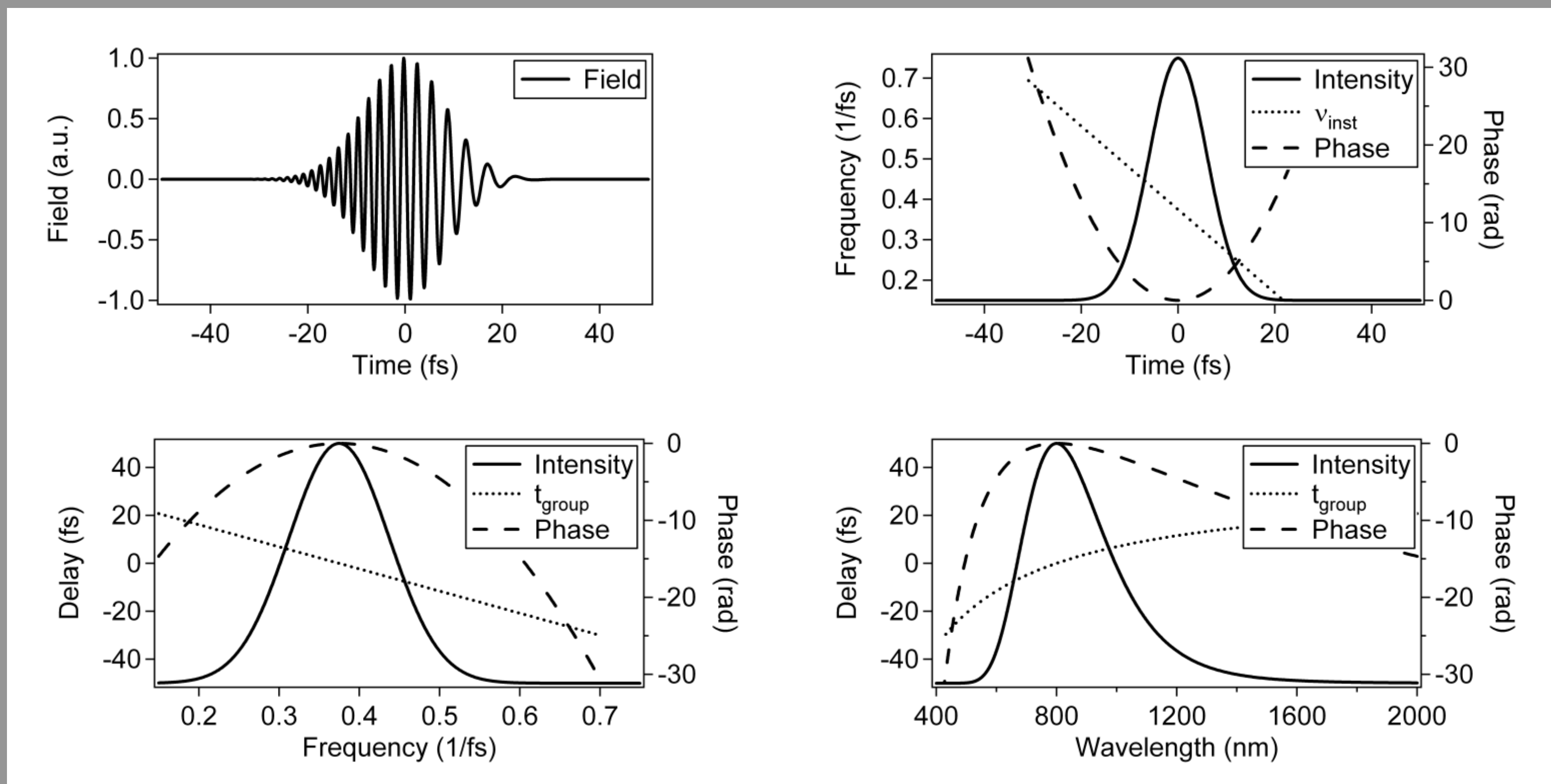
Numerical example: Gaussian-intensity pulse w/ positive linear chirp, φ_2 = 14.5 rad fs^2.



Here the quadratic phase has stretched what would have been a 3-fs pulse (given the spectrum) to a 13.9-fs one.

2^{nd}-order phase: negative linear chirp

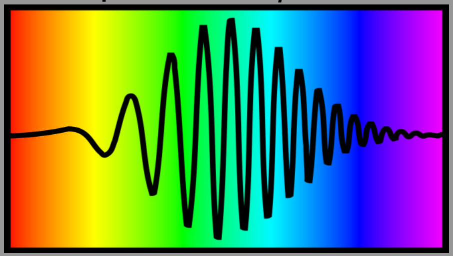
Numerical example: Gaussian-intensity pulse w/ negative linear chirp, $\varphi_2 = -14.5$ rad fs^2.



As with positive chirp, the quadratic phase has stretched what would have been a 3-fs pulse (given the spectrum) to a 13.9-fs one.

The TBP of a chirped Gaussian pulse

A linearly chirped pulse with $\beta > 0$



Pulse duration (FWHM) is: $\tau_{FWHM} = \sqrt{2\ln 2/\alpha}$

Spectral width (FWHM) is: $\Delta\omega_{FWHM} = \sqrt{\frac{8\ln 2}{\alpha}\left(\alpha^2 + \beta^2\right)}$

Using this definition of width, the TBP of this pulse is:

$$\Delta\omega_{FWHM} \cdot \Delta\tau_{FWHM} = \sqrt{\frac{2\ln 2}{\alpha} \cdot \frac{8\ln 2}{\alpha}\left(\alpha^2 + \beta^2\right)} = \left(4\ln 2\right)\sqrt{1 + \frac{\beta^2}{\alpha^2}} \approx 2.77\sqrt{1 + \frac{\beta^2}{\alpha^2}}$$

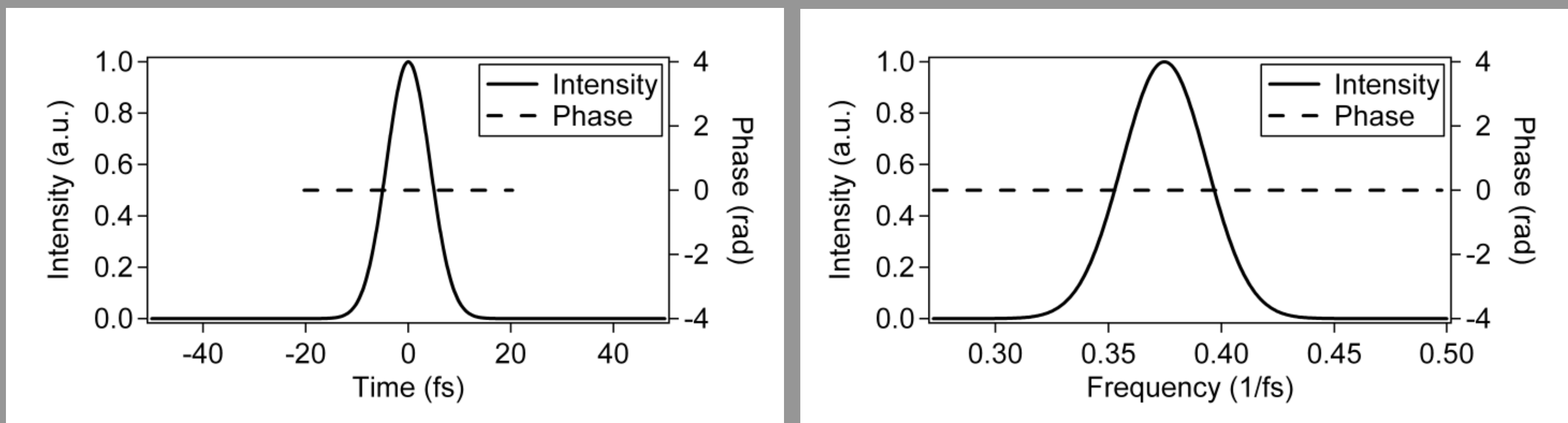
The TBP has a minimum value when $\beta = 0$.
It increases for both positive and negative chirp.

Thus:
For Gaussian pulse intensity, and using FWHM as the definition of width for the intensity and spectrum, the uncertainty principle says:

$\Delta\omega \cdot \Delta\tau \geq 2.77$ or $\Delta\nu \cdot \Delta\tau \geq 0.441$

Time-Bandwidth Product

Numerical example: A transform-limited pulse: A Gaussian-intensity pulse with constant phase and minimal TBP.



For the *angular* frequency and different definitions of the widths:

$\text{TBP}_{\text{rms}} = 0.5$ $\quad$ $\text{TBP}_{\text{eff}} = 3.14$

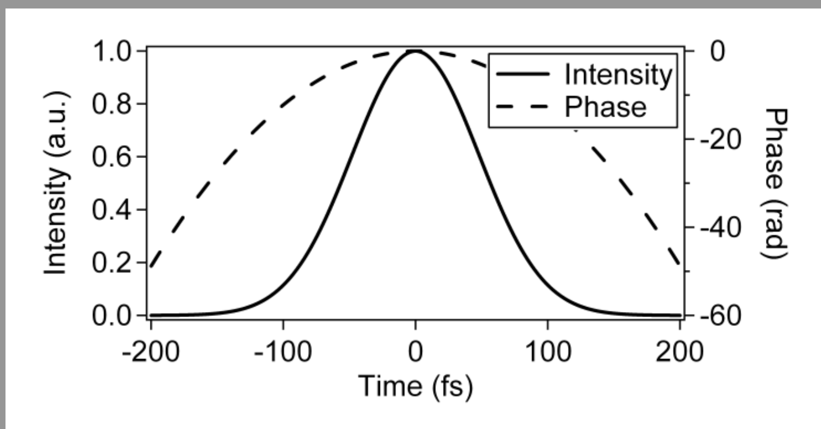
$\text{TBP}_{\text{HW1/e}} = 1$ $\quad$ $\text{TBP}_{\text{FWHM}} = 2.77$

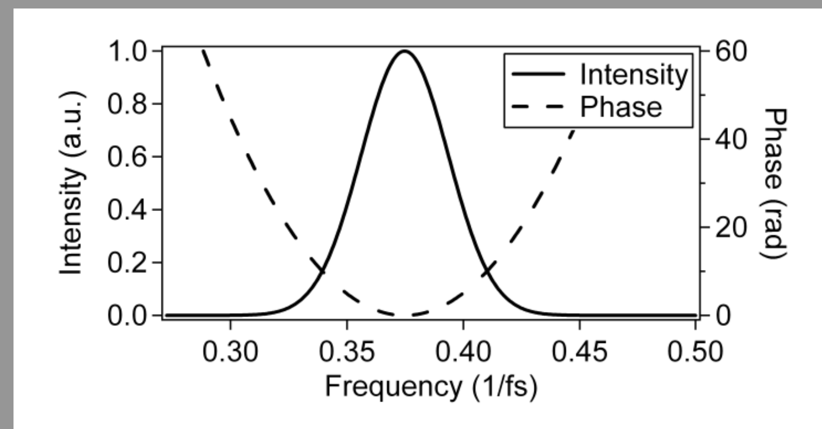
Notice that this definition yields an uncertainty product of π, not 2π; this is because we've used the intensity and spectrum here, not the fields.

Divide by 2π for the *cyclical* frequency $\Delta t_{rms}\,\Delta\nu_{rms}$, etc.

A linearly chirped pulse with no structure can also have a large time-bandwidth product.

Numerical example: A highly chirped, relatively long Gaussian-intensity pulse with a large TBP.





For the angular frequency and different definitions of the widths:

TBP_{rms} = 5.65

$TBP_{HW1/e}$ = 11.3

TBP_{eff} = 35.5

TBP_{FWHM} = 31.3

This pulse is about 11.3 times longer than it could be, given the spectrum.

Again: divide by 2π for $\Delta t_{rms}\, \Delta\nu_{rms}$, etc.

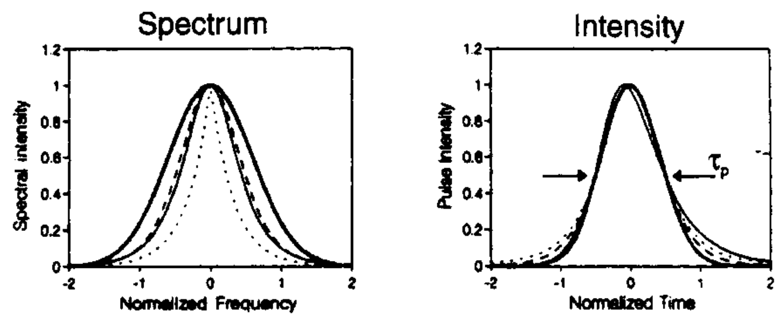
Temporal and spectral shapes and TBPs of some ultrashort pulses

Diels and Rudolph, Femtosecond Phenomena

Common pulse envelopes (with τ_p = Intensity FWHM):

Gaussian pulse	$\mathcal{E}(t) \propto \exp[-1.385(t/\tau_p)^2]$
sech - pulse	$\mathcal{E}(t) \propto \mathrm{sech}[1.763(t/\tau_p)]$
Lorentzian pulse	$\mathcal{E}(t) \propto [1 + 1.656(t/\tau_p)^2]^{-1}$
asymm. sech pulse	$\mathcal{E}(t) \propto [\exp(t/\tau_p) + \exp(-3t/\tau_p)]^{-1}$

Spectra for pulses with the same pulse width



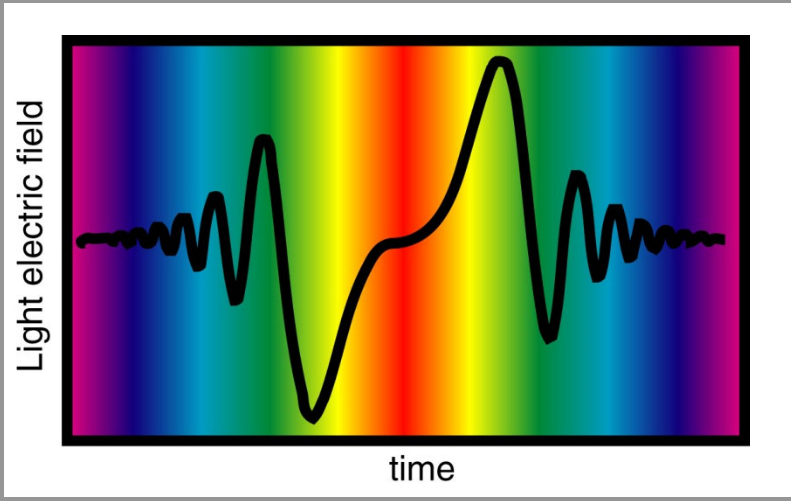
Field envelope	Intensity profile	τ_p (FWHM)	Spectral profile	$\Delta\omega_p$ (FWHM)	TBP
Gauss	$e^{-2(t/\tau_G)^2}$	$1.177\tau_G$	$e^{-(\omega\tau_G)^2/2}$	$2.355/\tau_G$	0.441
sech	$\mathrm{sech}^2(t/\tau_s)$	$1.763\tau_s$	$\mathrm{sech}^2(\pi\omega\tau_s/2)$	$1.122/\tau_s$	0.315
Lorentz	$[1+(t/\tau_L)^2]^{-2}$	$1.287\tau_L$	$e^{-2\lvert\omega\rvert\tau_L}$	$0.693/\tau_L$	0.142
asymm. sech	$\left[e^{t/\tau_a}+e^{-3t/\tau_a}\right]^{-2}$	$1.043\tau_a$	$\mathrm{sech}(\pi\omega\tau_a/2)$	$1.677/\tau_a$	0.278
rectang.	1 for $\lvert t/\tau_r\rvert \le \frac{1}{2}$, 0 else	τ_r	$\mathrm{sinc}^2(\omega\tau_r)$	$2.78/\tau_r$	0.443

Nonlinearly chirped pulses

The frequency of a light wave can also vary nonlinearly with time.

This is the electric field of a Gaussian pulse whose frequency varies quadratically with time:

$$\omega_{inst}(t) = \omega_0 + 3\gamma t^2$$



This light wave has the expression:

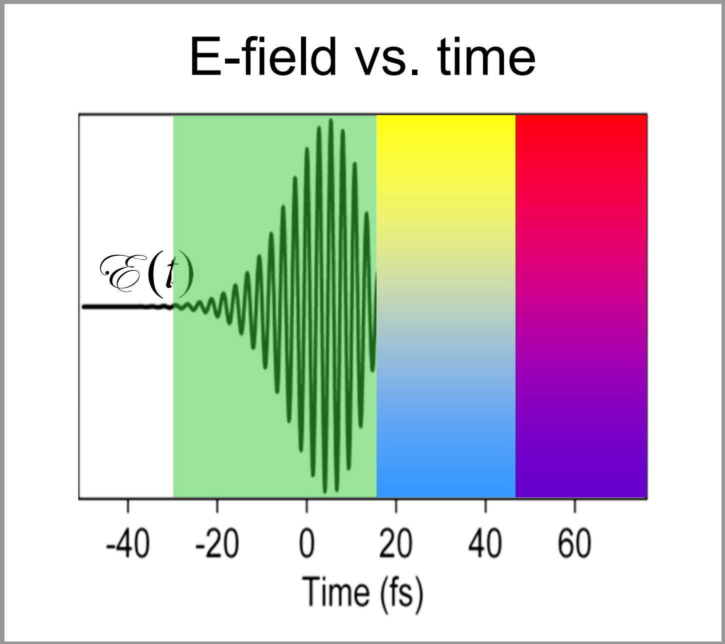
$$\mathscr{E}(t) = \operatorname{Re} E_0 \exp\left[-\left(t/\tau_G\right)^2\right]\exp\left[i\left(\omega_0 t + \gamma t^3\right)\right]$$

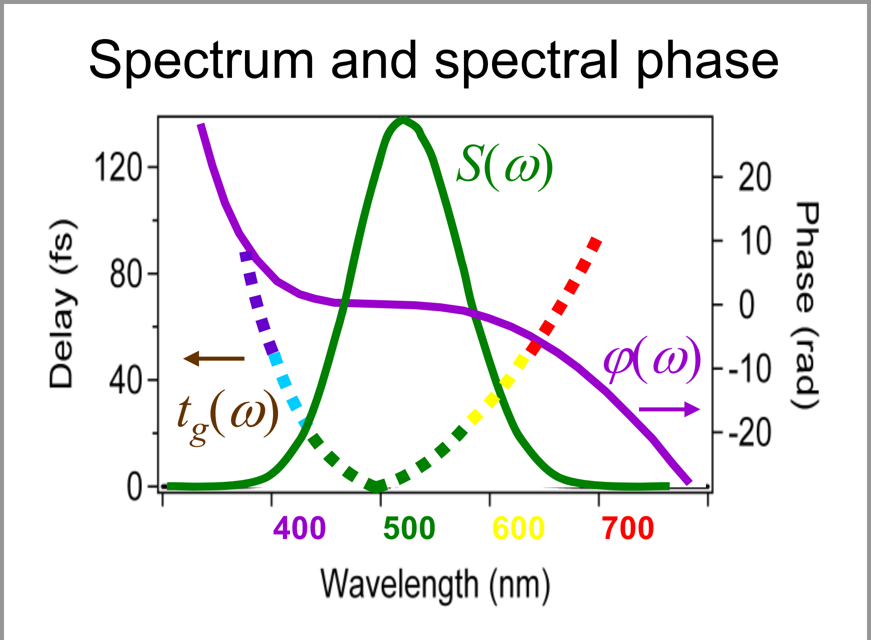
Arbitrarily complex frequency-vs.-time behavior is possible.

But, as before, we usually describe phase distortions in the frequency domain, not the time domain.

3^{rd}-order spectral phase: quadratic chirp

Longer and shorter wavelengths coincide in time and interfere (beat).



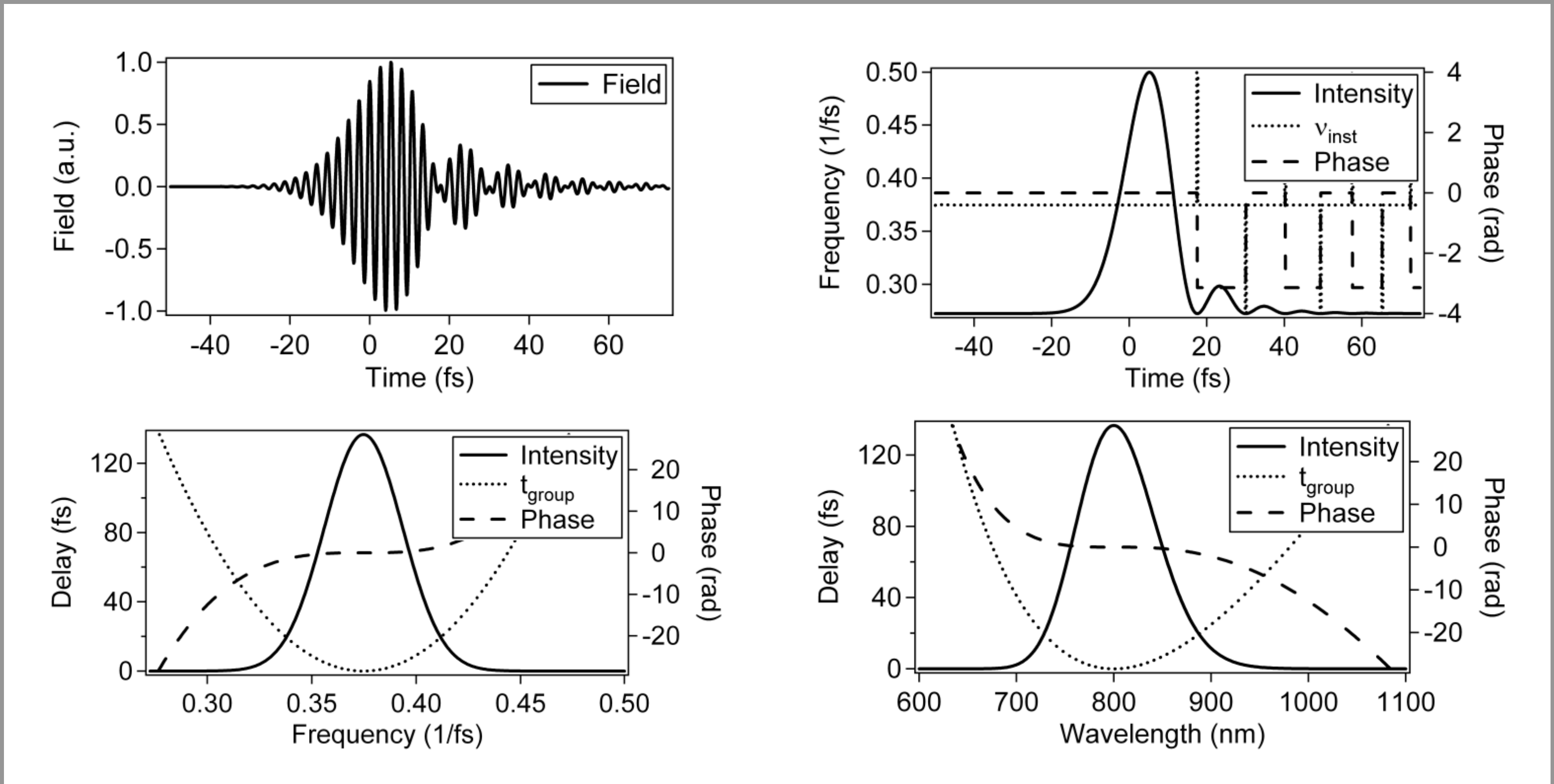


Because we're plotting vs. wavelength (not frequency), there's a minus sign in the group delay, so the plot is correct.

Trailing satellite pulses in time indicate positive spectral cubic phase, and leading ones indicate negative spectral cubic phase.

3^{rd}-order spectral phase: quadratic chirp

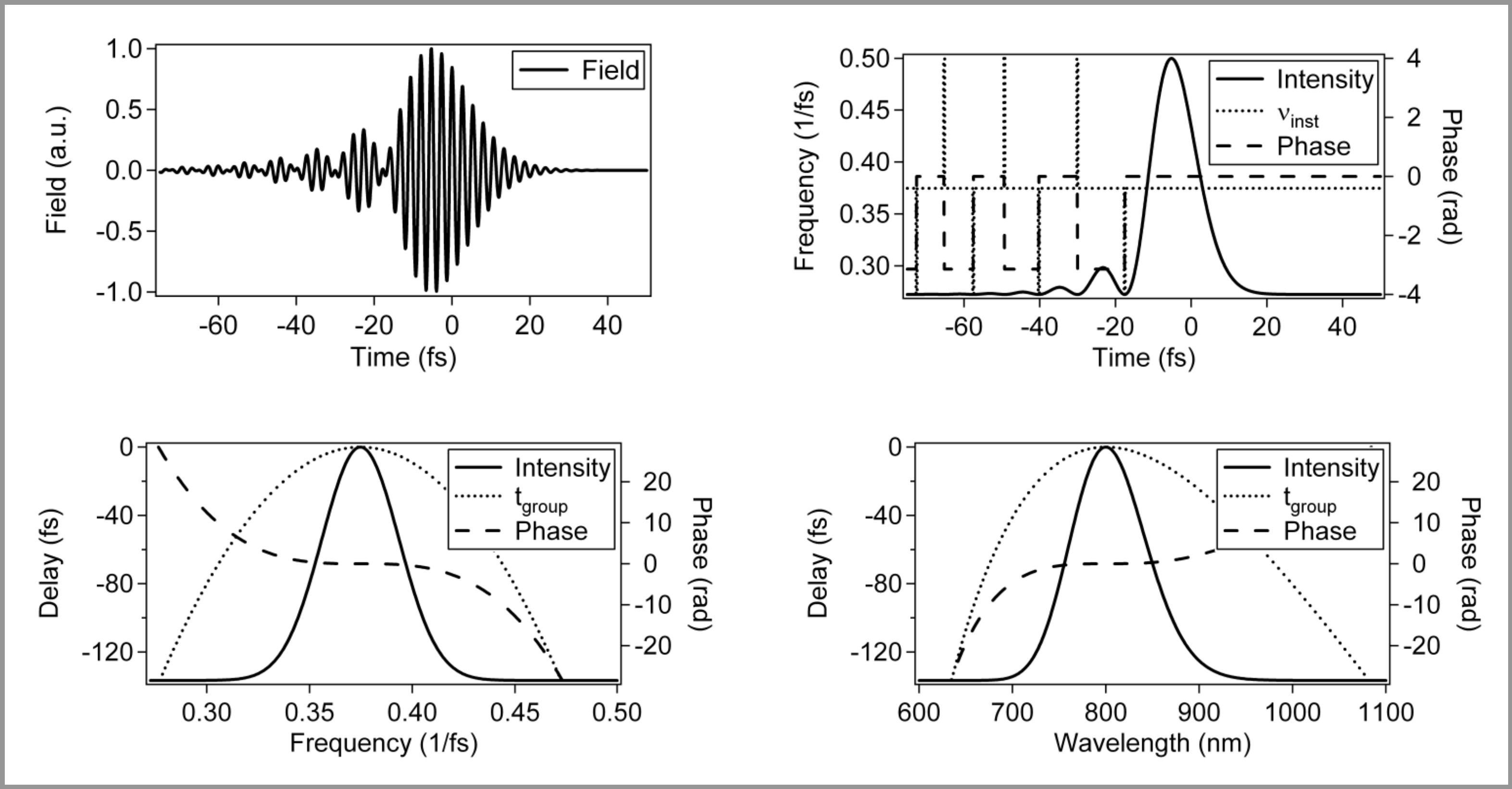
Numerical example: Gaussian spectrum and positive cubic spectral phase, with φ_3 = 750 rad fs^3



Trailing satellite pulses in time indicate positive spectral cubic phase.

Negative 3rd-order spectral phase

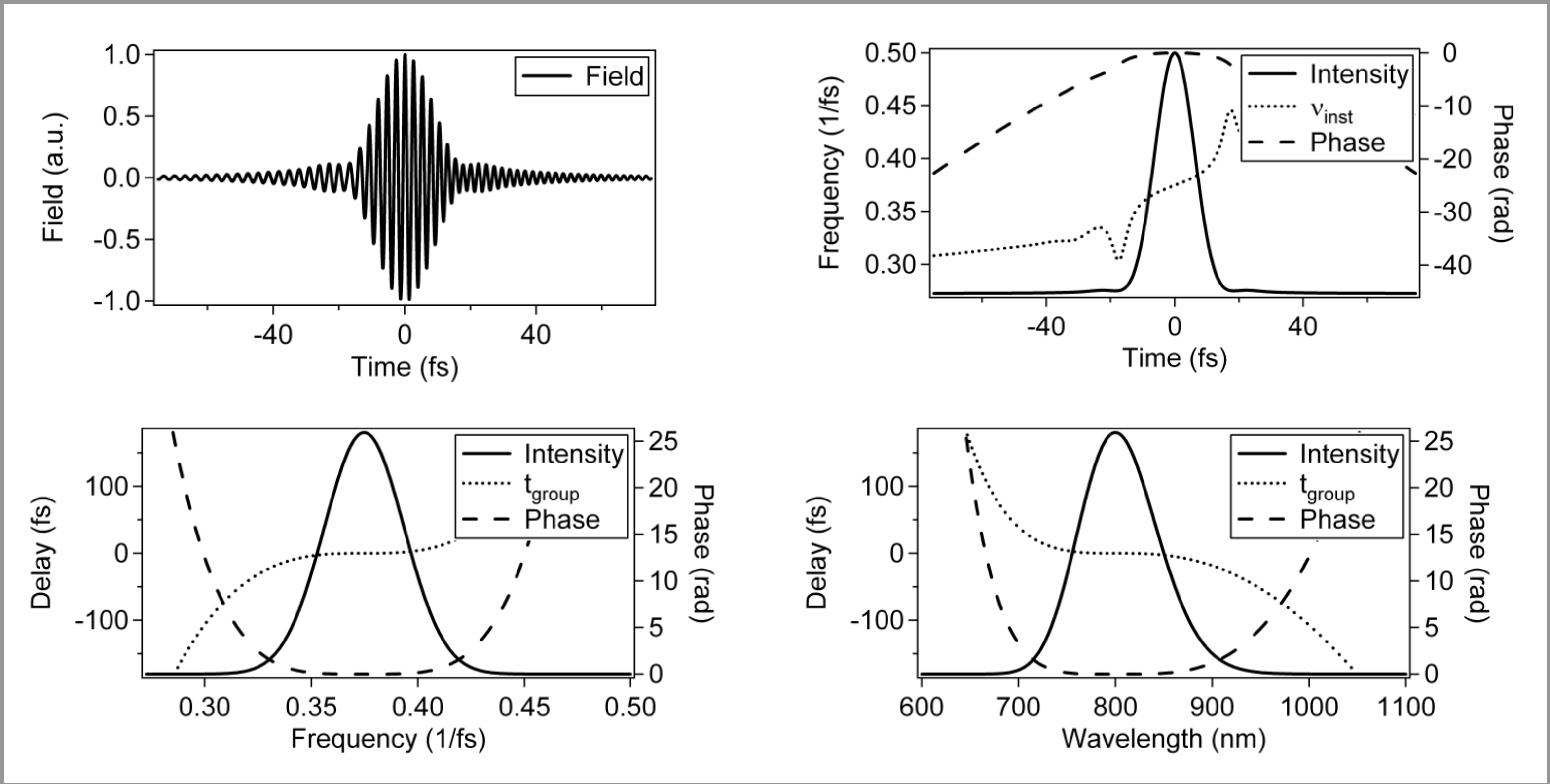
Another numerical example: Gaussian spectrum and negative cubic spectral phase, with $\varphi_3 = -750$ rad fs^3



Leading satellite pulses in time indicate negative spectral cubic phase.

4th-order spectral phase

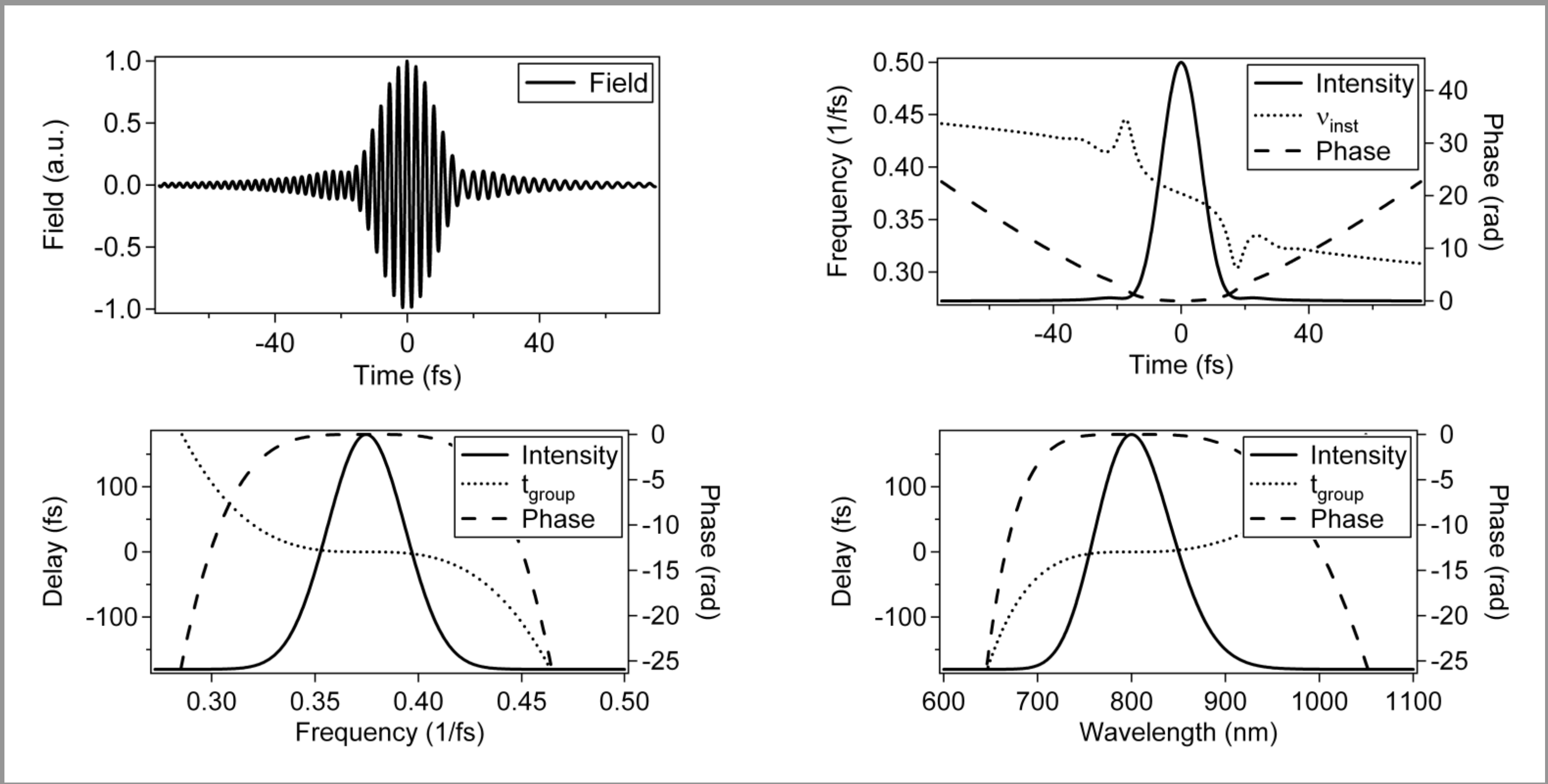
Numerical example: Gaussian spectrum and positive quartic spectral phase, φ_4 = 5000 rad fs^4.



Leading and trailing wings in time indicate quartic phase. Higher-frequencies in the trailing wing mean positive quartic phase.

Negative 4th-order spectral phase

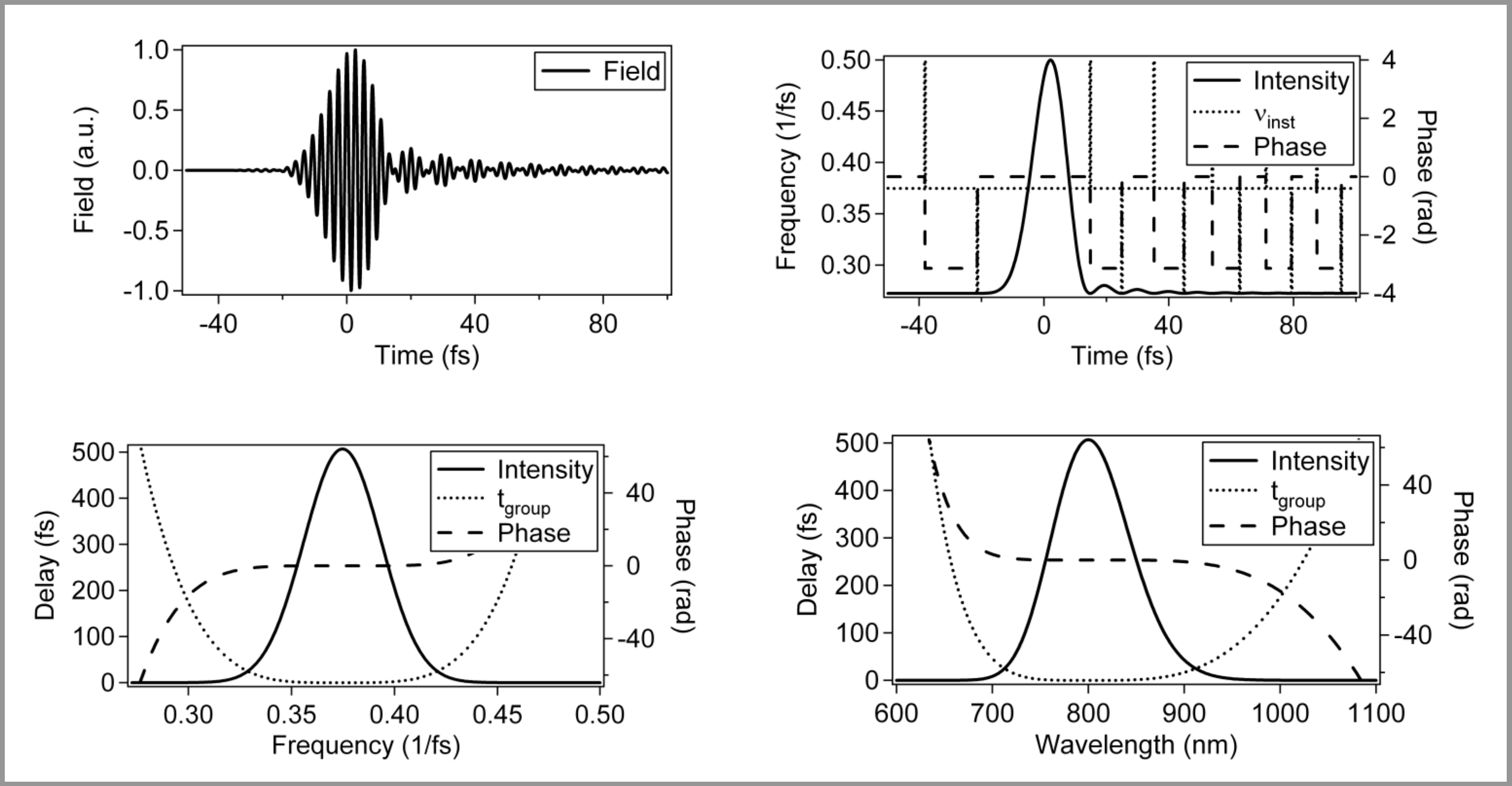
Numerical example: Gaussian spectrum and negative quartic spectral phase, φ_4 = –5000 rad fs^4.



Leading and trailing wings in time indicate quartic phase. Higher-frequencies in the leading wing mean negative quartic phase.

5th-order spectral phase

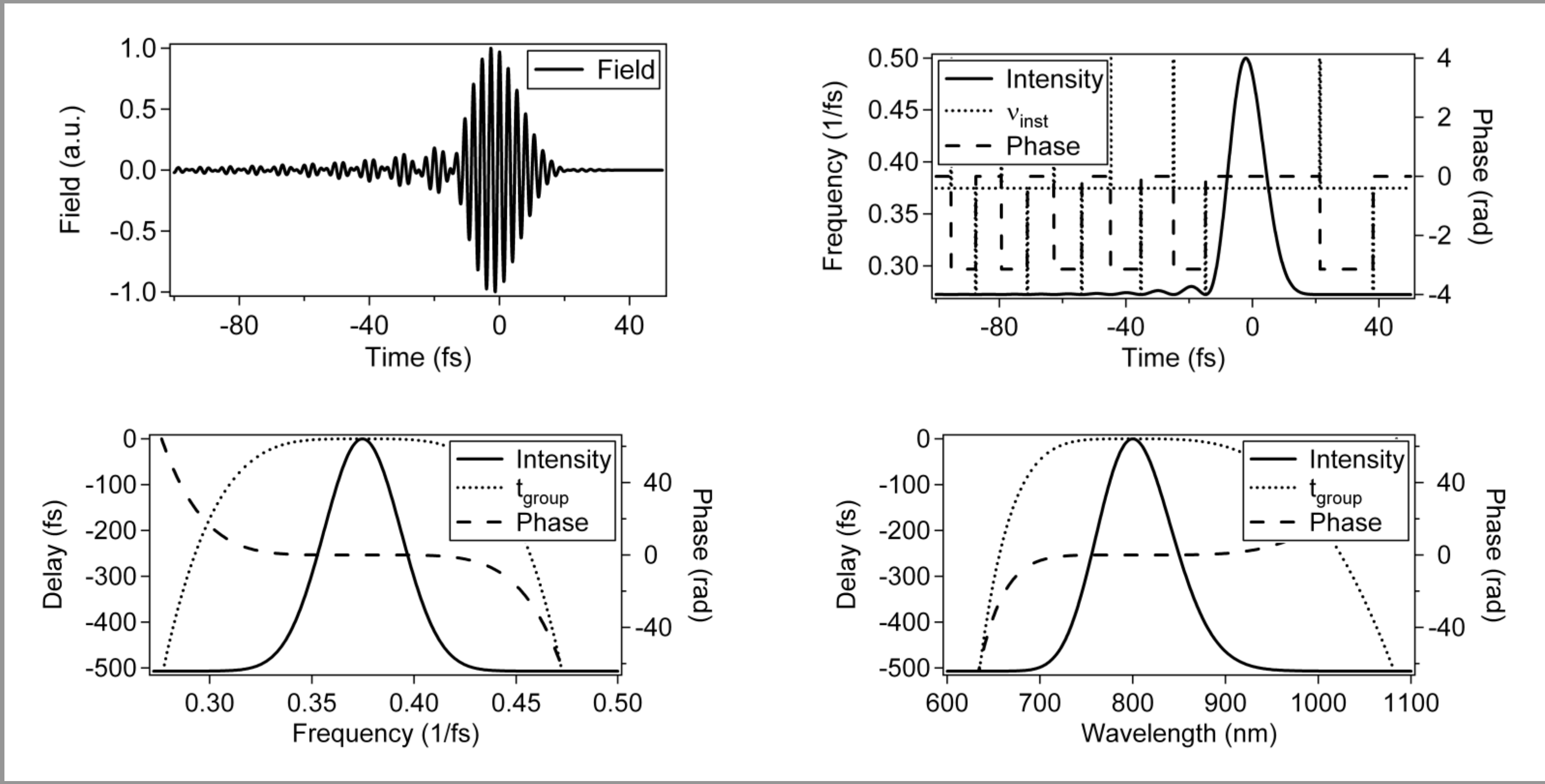
Numerical example: Gaussian spectrum and positive quintic spectral phase, $\varphi_5 = 4.4\times10^4$ rad fs^5.



An oscillatory trailing wing in time indicates positive quintic phase.

Negative 5th-order spectral phase

Numerical example: Gaussian spectrum and negative quintic spectral phase, $\varphi_5 = -4.4\times10^4$ rad fs^5.



An oscillatory leading wing in time indicates negative quintic phase.

The shortest pulse for a given spectrum has a constant spectral phase.

We can write the pulse width in a way that illustrates the relative contributions to it by the spectrum and spectral phase.

If $B(\omega) = \sqrt{S(\omega)}$, then the temporal width, Δt_{rms}, is given by:

$$\Delta t_{rms}^2 = \underbrace{\int_{-\infty}^{\infty} B'(\omega)^2 \, d\omega}_{\text{Contribution due to variations in the spectrum}} + \underbrace{\int_{-\infty}^{\infty} B(\omega)^2 \, \varphi'(\omega)^2 \, d\omega}_{\text{Contribution due to variations in the spectral phase}}$$

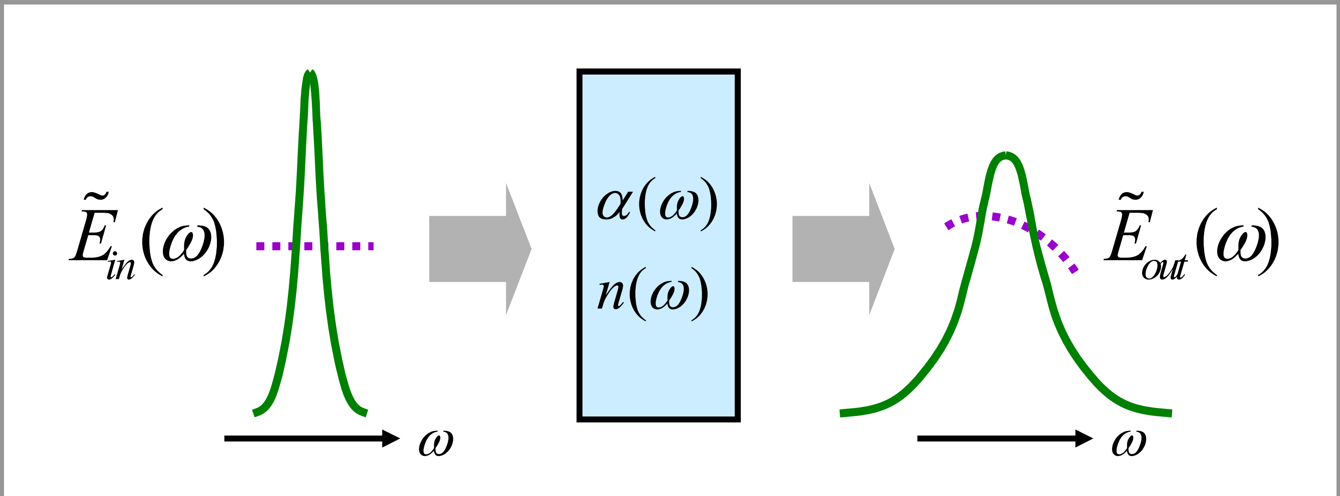
Note: this result assumes that the mean group delay has been subtracted from φ'. That is, the pulse is centered at $t_{gr} = 0$.

Notice that variations in the spectral phase can only increase the pulse width, never decrease it.
This *has to be true*, or else the uncertainty principle would be violated.

Pulse propagation in a medium

What happens to a pulse as it propagates through a medium?

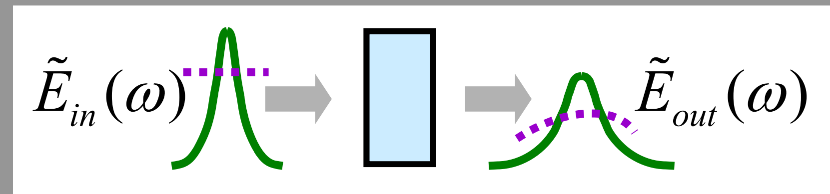
We always model (linear) propagation in the **frequency domain**. Of course, you must know the entire field (i.e., the intensity and phase) to do so.



$$\boxed{\tilde{E}_{out}(\omega) = \tilde{E}_{in}(\omega)\exp[-\alpha(\omega)L/2]\exp[-i\,n(\omega)\,k\,L]}$$

In the time domain, propagation is a convolution—much harder.

Pulse propagation (continued)



Rewriting this expression:

$$\tilde{E}_{out}(\omega) \;=\; \tilde{E}_{in}(\omega)\exp[-\alpha(\omega)L/2]\exp[-i\,n(\omega)\,k\,L]$$

using $k = \omega / c$:

$$=\; \tilde{E}_{in}(\omega)\exp[-\alpha(\omega)L/2]\exp[-i\,n(\omega)\,\frac{\omega}{c}\,L]$$

Separating out the spectrum and spectral phase:

$$\boxed{\begin{aligned} S_{out}(\omega) &= S_{in}(\omega)\exp[-\alpha(\omega)L] \\ \varphi_{out}(\omega) &= \varphi_{in}(\omega) + n(\omega)\,\frac{\omega}{c}\,L \end{aligned}}$$

Pulse propagation (continued)

$$\boxed{S_{out}(\omega) \;=\; S_{in}(\omega)\exp[-\alpha(\omega)L]}$$

$$\varphi_{out}(\omega) \;=\; \varphi_{in}(\omega) + n(\omega)\frac{\omega}{c}L$$

In transparent media, αL << 1, so the effect of absorption is often negligible.

Also, even when α is not negligible, it is often the case that $\alpha(\omega)$ does not vary too much within the spectrum of the pulse.

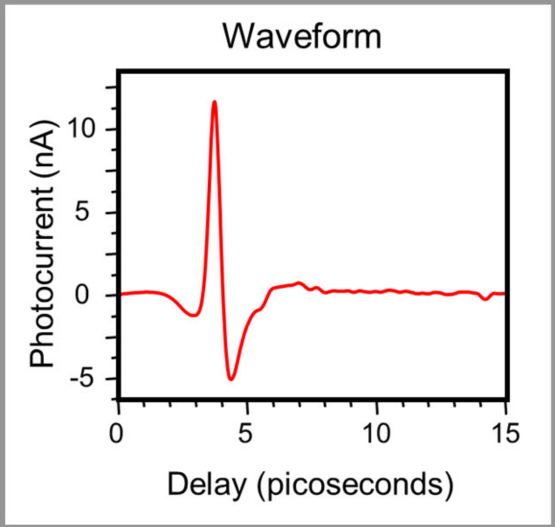
In which case, $S_{out}/S_{in} = \exp[-\alpha(\omega)L] \approx \text{constant}$

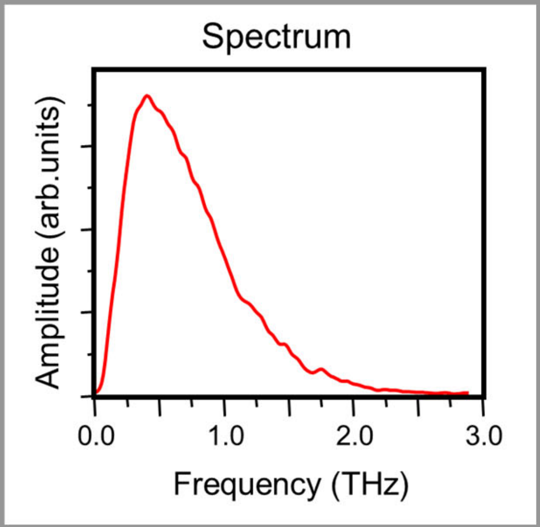
the effect of absorption is simply to decrease the spectrum uniformly by a constant value. This has no effect on the pulse duration.

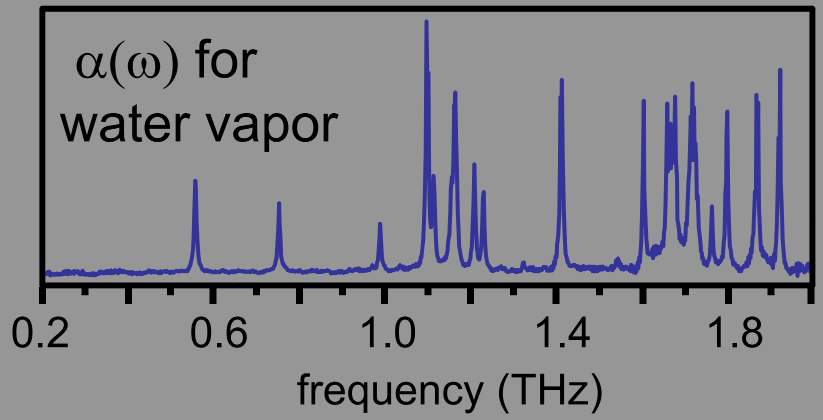
An interesting counter-example

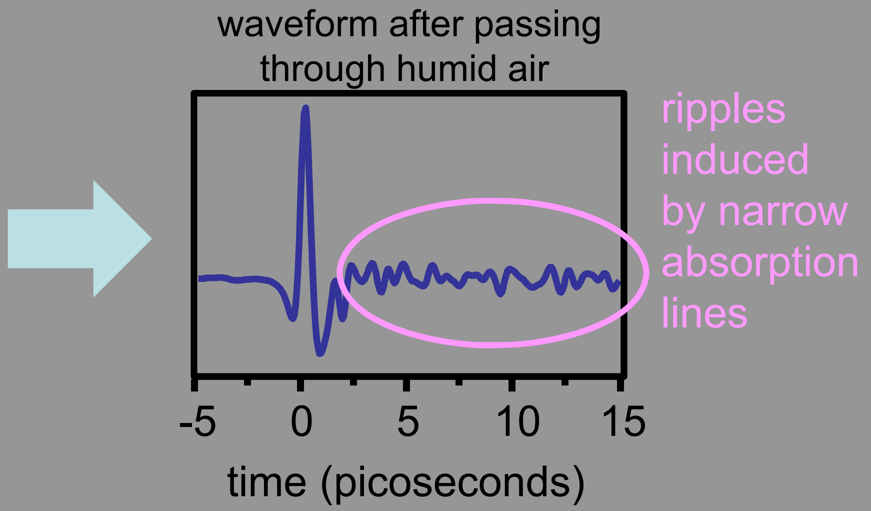
For ultra-broadband terahertz pulses, $\alpha(\omega)$ can vary a LOT within the bandwidth of the incoming pulse.

Absorption lines of gases are much narrower than the pulse spectrum.









Pulse propagation (continued)

$$S_{out}(\omega) = S_{in}(\omega)\exp[-\alpha(\omega)L]$$

$$\boxed{\varphi_{out}(\omega) = \varphi_{in}(\omega) + n(\omega)\frac{\omega}{c}L}$$

On the other hand, this term can have contributions that vary with any power of ω!

These terms add to the corresponding terms in the Taylor expansion for $\varphi(\omega)$, producing all manner of pulse distortions.

Next lecture: what does pulse propagation in a medium do to short pulses, and how do we describe it?