Nonlinear Optics and Squeezed Light with Four-Wave Mixing

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outline

• a “black box” approach to nonlinear media
• dispersion relations for nonlinear optics
• 4-wave mixing (4WM) in atomic vapors
• a slow light example; complications to the “black box”
• non-classical light from 4WM
outline (continued)

• phase insensitive and phase sensitive amplifiers
• squeezing in multiple spatial modes
• an SU(1,1) interferometer
• 4-wave mixing in atoms
• an atomic SU(1,1) interferometer
an aside – or why do I want a nonlinear optics “black box”

What do I mean by a “black box?”
I want to put a light beam through this box and have it “transformed” in some way (intensity, phase, velocity, direction,...) ...

...but then I also want to describe it just in terms of what comes out, and not have to worry about just how the light beam was transformed.

in particular, can I look at light coming through an object, see that it is “slowed down” or delayed, and then simply describe the object by an index of refraction?
the black box

I will want to end up describing a black box for slow light, using EIT in the box, and then simply be able to look at the slow light out, and deduce the dispersion relation from the gain/loss in the system as a function of frequency, without worrying that I had to bring in another beam of light to make it happen...

Can I ignore the extra light beam(s) and treat this as a “block of glass”? 
The “Abraham force” problem:

What is the momentum of a photon in a medium?

Or, what is the correct choice of the stress-energy tensor for the electromagnetic field?

And can we do an experiment to tell the difference?

First let's consider a thought experiment...
Consider a block of glass on a frictionless surface

and a pulse of light passing through... (no reflections)

...what happens to the block of glass?
what is the momentum of a photon in a medium of index $n$?

naive approach #1:

note that the frequency of oscillation, $\nu$, is a constant

\[
\begin{align*}
\text{vacuum:} & \\ 
\lambda &= \frac{c}{\nu} \\
p &= \hbar k = \frac{h}{\lambda} = \frac{h\nu}{c}
\end{align*}
\]

\[
\begin{align*}
\text{medium:} & \\ 
\lambda &= \frac{c}{n\nu} \\
p &= \hbar k = \frac{nh}{\lambda} = \frac{nh\nu}{c}
\end{align*}
\]
what is the momentum of a photon in a medium of index $n$?

naive approach #2:

vacuum:

$p = m v$

$m = \frac{E}{c^2}$

$p = \frac{E}{c^2} c = \frac{h \nu}{c}$

medium:

$p = m v$

$v = \frac{c}{n}$

$p = \frac{E}{c^2 n} = \frac{h \nu}{nc}$
So, which is it?

\[ p = \frac{\hbar k}{\lambda} = \frac{nh}{c} \quad \text{or} \quad p = mv = \frac{E}{c^2} \frac{c}{n} = \frac{h\nu}{nc} \]

Minkowski \quad \text{or} \quad \text{Abraham}?

In terms of forces, the difference between the Minkowski and Abraham expressions is represented by the “Abraham force”
the stress-energy tensors

Abraham form:

\[
\begin{pmatrix}
S^A_{ik} & S^A_{4k} \\
S^A_{k4} & S^A_{44}
\end{pmatrix} = \begin{pmatrix}
-\frac{1}{2}(E_i D_k + E_k D_i) - \frac{1}{2}(H_i B_k + H_k B_i) + \frac{1}{2} \delta_{ik} (\vec{E} \cdot \vec{D} + \vec{H} \cdot \vec{B}) & (\vec{E} \times \vec{H})_k \\
\frac{i}{c} (E \times H)_k & \frac{1}{2}(\vec{E} \cdot \vec{D} + \vec{H} \cdot \vec{B})
\end{pmatrix}
\]

Minkowski form:

\[
\begin{pmatrix}
S^M_{ik} & S^M_{4k} \\
S^M_{k4} & S^M_{44}
\end{pmatrix} = \begin{pmatrix}
-E_i D_k - H_i B_k + \frac{1}{2} \delta_{ik} (\vec{E} \cdot \vec{D} + \vec{H} \cdot \vec{B}) & (\vec{E} \times \vec{H})_k \\
ic(\vec{D} \times \vec{B})_k & \frac{1}{2}(\vec{E} \cdot \vec{D} + \vec{H} \cdot \vec{B})
\end{pmatrix}
\]
The choice of stress-energy tensor
(or, here, the resultant force on a dielectric medium)

Minkowski (1908)

\[ f^M = -\frac{1}{2} E^2 \nabla \varepsilon - \frac{1}{2} H^2 \nabla \mu \]

Abraham (1909)

\[ f^A = f^M + \frac{\varepsilon \mu - 1}{c^2} \frac{\partial}{\partial t} (\vec{E} \times \vec{H}) \]

plus other choices given by a bunch of other authors, including Einstein & Laub, Peierls, Nelson,...
the “Abraham force”

\[ f^A = f^M + \frac{\epsilon \mu - 1}{c^2} \frac{\partial}{\partial t} (\vec{E} \times \vec{H}) \]

This term is known as the “Abraham force.”

It is generally small and has only been observed under “quasi-static” field conditions.

Note that: \( \vec{S} = \vec{E} \times \vec{H} \) is the Poynting vector.
a gedanken experiment

Balazs, 1953
Consider the center of mass motion

\[ mct = m(x + \xi + L) + M\xi \]

\[ m(x + n(L + \xi)) = m(x + \xi + L) + M\xi \]

\[ m(n - 1)(L + \xi) = M\xi \]

\[ m(n - 1)L = \xi[M - m(n - 1)] \]
Consider conservation of momentum.

If the block is going to move "forward," and momentum is going to be conserved while the pulse is in the medium,...

Then the photons must have less momentum in the medium than in vacuum.

\[ p = \frac{hv}{c} \]

\[ p' = \frac{hv}{nc} \] (Abraham’s answer)
m(n-1) acts as the “effective mass” of the light pulse

this mass can become comparable to the mass $M$, of the “slab” if the slab is as light as possible; for instance, a collection of about $10^6$ atoms in a BEC

if $M = m(n-1)$ then the displacement becomes infinite! (this ignores effects of the velocity of the medium on the light propagation)

if $n < 0$ you get “faster than light” propagation (or if $M < m(n-1)$) and the displacement changes direction!
The choice of stress-energy tensor
(or, here, the resultant force on a dielectric medium)

Minkowski (1908)

\[ f^M = -\frac{1}{2} E^2 \nabla \varepsilon - \frac{1}{2} H^2 \nabla \mu \]

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\[ f^A = f^M + \frac{\varepsilon \mu - 1}{c^2} \frac{\partial}{\partial t} (\vec{E} \times \vec{H}) \]

plus other choices given by a bunch of other authors, including Einstein & Laub, Peierls, Nelson,...
Abraham vs. Minkowski on the displacement of the block

Look at the time-averaged forces due to an optical field.

Abraham gives the expression from above:

\[ \xi^A = \frac{m(n - 1)L}{M} \]

approximated for \( M >> m(n-1) \)

Minkowski gives an expression with the opposite sign!

\[ \xi^M = \frac{m(1 - n)nL}{M} = -n\xi^A \]

note, that interpretation is straightforward only if \( n = n_{\text{phase}} \)

(explicitly not what I want!)
A real experiment that will not answer this question.  
(… but might answer a related one!)

You need a very light block of glass, with a very high index of refraction, on a completely frictionless surface… not so easy…

A BEC can, under some circumstances, behave like a block of glass (sort of), can be given a huge index of refraction (sort of), and can sit on a frictionless surface (no, really - that part is easy!).
The proposed experiment

Pulse:
3 x 10^6 photons/1 \mu s
= 1 \mu W at 589 nm
1 ms duration
m = E/c^2 = 10^{-26} kg

"Block of glass" = BEC
3 x 10^6 sodium atoms
M = 1.2 \times 10^{-19} kg
L = 100 \mu m

index n
n_g = d\omega/dk
EIT / "slow light"
n = 10^6  \ c/n = 300 m/s
transit time* = 100\mu m/300m/s = 0.3\mu s

\xi = \frac{m(n-1)L}{M - m(n-1)}
\xi \sim 0.1 L = 10 \mu m

m(n-1) = 10^{-20} kg
“EIT” and “slow light”

To get the large values of the group velocity index that are necessary, we need to employ "slow light" techniques, or "electromagnetically-induced transparency".

An extra beam "dresses" the atoms in such a way that the pulse of light sees an apparently large dispersion:

\[ \frac{\omega_p}{\omega_c} \]  

60 MHz  

1.8 GHz

\[ F=2, \ m_F=-2 \]  

\[ F=1, \ m_F=-1 \]

\[ F=3, \ m_F=-2 \]

\[ F=2, \ m_F=-2 \]

17 m/s or \( n_g \sim 2 \times 10^7 \)

unfortunately, transmission isn't amazing

The "real" momentum that is associated with an atom (polariton?) depends on the angle between the two Raman beams required for producing the EIT effect.

If the beams are orthogonal then the analysis of momentum conservation along the direction of the pulse is the same as for a "passive" medium like glass.
3x10^6 atoms and 1 \( \mu \text{W} \) (3x10^6 photons/\( \mu \text{s} \)) with a transit time of 0.3\( \mu \text{s} \) gives us about 10^6 polaritons in steady state (1/3 of the atoms).

For orthogonal beams each polariton carries 1 \( \hbar k \) of momentum along the direction of the beam (and 1 \( \hbar k \) orthogonal as well).

For sodium the recoil velocity is 3 cm/s and on average 1/3 of the atoms carry this recoil.

A 1 cm/s average velocity times a 1 ms duration pulse just as expected
Dispersive form of Abraham’s force

if we calculate the term with dispersion included we get (see Landau, Lifshitz, and Pitaevskii, Electrodynamics of continuous media):

\[
f^A = \frac{\varepsilon \mu - 1}{c^2} \frac{\partial}{\partial t} (\vec{E} \times \vec{H}) \\
+ \frac{\partial \omega \varepsilon \mu}{2 \partial \omega} \frac{\partial}{\partial t} (\vec{E} \times \vec{H})
\]
Abraham vs. Minkowski with a dispersive medium

Abraham gives:

\[ \xi^A = \frac{m(n_g - 1)L}{M} \]

approximated for \( M >> m(n-1) \)

Minkowski still gives an expression with the opposite sign:

\[ \xi^M = -\frac{m(n - 1)n_g L}{M} \]

note: the Abraham version is independent of the phase index

in the example discussed above,
for \( n = 1.001, n_g = 10^6; \xi^A = 10 \mu m; \xi^M = -0.01 \mu m \)
a BEC as a block of glass

If you drive a Raman transition with an $\hbar k$ of recoil, an individual atom will be kicked out of the condensate - the BEC needs to act as a “solid.”

If the recoil is about 0.1 $\hbar k$ or less, then the recoil is less than the speed of sound in the medium and the condensate should recoil as a whole, generating phonons, but not ejecting atoms.

The Raman beam that “dresses” the atoms for the EIT effect must then be at a “small” angle and the momentum exchange along this direction must be accounted for; this complicates the interpretation, and makes the transparency requirement more stringent.
spontaneous light scattering

residual absorption in “EIT” process is a severe problem
-Hau paper had only 65% transmission

assume that:
- we have our control beam at 90 degrees
- $3 \times 10^6$ atoms in the “block”
- one spontaneous scattering event removes the atom
- we can afford to lose $\sim 30\%$ of the atoms and still see the effect

$$1\mu W = 3 \times 10^6 \text{ photons}/\mu s$$
$$\times 1 \text{ ms pulse} = 3 \times 10^9 \text{ photons}$$

in this scenario we can afford only 0.1% absorption
(the need for a small angle for the control beam makes this worse)
can it be done?

If the EIT effect can really be made transparent...

- This can be a “big” effect and can be done as a qualitative, rather than quantitative experiment and still distinguish between the Abraham and Minkowski choices.

- The experiment also gives a graphic demonstration of “faster-than-light” effects and interesting things that happen with negative index of refraction materials.
So, what went wrong?

\[ p = \frac{nh}{\lambda} = \frac{nh\nu}{c} \quad \text{vs.} \quad p = mv = \frac{E}{c^2} \frac{c}{n} = \frac{h\nu}{nc} \]

answer: You cannot assume that \( p = \hbar k \) in the medium. This assumes that the photon carries all the momentum and that the medium carries none (as does Minkowski).
an “effective medium”
(a “black box” medium)

I would like to treat the medium (BEC with EIT control beam) as a block of glass with an index of refraction and ignore the fact that I had to “dress” the medium with the control beam to give it the properties that I want.

Can I describe the medium with a summary of the desired dispersion relations? Can I measure the dispersion relations and ignore the physics of what goes on inside the medium?

causality and dispersion relations

(a) a pulse appears after a given time

(b) a perfect absorber removes one frequency

(c) “suddenly” light had to have appeared at all times

this violation of causality can only be resolved by having the absorber changing the phase on all the other waves present
linear optics dispersion relations

\[ n(\omega) - 1 = \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\alpha(\Omega) \, d\Omega}{\Omega^2 - \omega^2} \]

relates the index of refraction to an integral over all frequencies of the linear absorption coefficient generally known as Hilbert transforms – in electrical networks, particle scattering, etc.

This sort of relation is well-established in linear optics but often not considered in nonlinear optics.

For nonlinear optical effects a nondegenerate form of the dispersion relation is appropriate; describing an index change at \( \omega \) due to the presence of a strong perturbing field at \( \Omega \) is related to an integral over \( \omega' \) of the nonlinear absorption due to the perturbing field at \( \Omega \).
linear optics dispersion relations

\[ n(\omega) - 1 = \frac{c}{\pi} \mathcal{P} \int_{0}^{\infty} \frac{\alpha(\Omega) \, d\Omega}{\Omega^2 - \omega^2} \]

relates the index of refraction to an integral over all frequencies of the linear absorption coefficient

Cauchy principle value integral over analytic continuation into the upper half-plane
Linear dispersion relations

$\phi$ is a function of a complex variable $\omega = \omega_r = i \omega_i$ which is defined in the upper half of the complex plane and is analytic there. (the real axis does not have to belong to range of analyticity.

the function $\phi$ is square-integrable

A function that is square integrable is zero for all negative values of its argument if and only if its Fourier transform is a causal transform.

So, you can have a pole (divergence) on the real axis, but the simple relation doesn’t work with a pole in the upper half-plane.
causality

\[ P(t) = \int_{-\infty}^{\infty} R(\tau) E(t - \tau) \, d\tau \]

polarization is related to the electric field by a response function ("Green's function")

\[ P(\omega) = \chi(\omega) E(\omega) \]

\( \chi \) is the susceptibility and is related to the response function:

\[ \chi(\omega) = \int_{-\infty}^{\infty} R(\tau) e^{i\omega \tau} \, d\tau \]

essentially a Fourier transform relation (to within constants)
Causality (cont.)

causality: effect cannot precede cause, thus $R(T)$ (response to a impulse at $T$) has to be 0 for $T < 0$

thus the integral only has to cover $t > 0$

$$R(T) = R(T)\theta(T)$$

$\theta(T)$ is the step function at $T$

$$\chi(\omega) = \chi(\omega)^* \left( \frac{\delta(\omega)}{2} + \frac{i}{2\pi\omega} \right)$$

$$= \frac{\chi(\omega)}{2} + \frac{i}{2\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi(\Omega)}{\omega - \Omega} \, d\Omega$$

$$= \frac{1}{i\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi(\Omega)}{\Omega - \omega} \, d\Omega$$

Fourier transform

product becomes a convolution in frequency

$\theta$ gives the 2 terms

combined to give the usual form
Thus, Kramers-Kronig relation is in some sense a restatement of the causality condition.

In optics we see this as a relation between the real and imaginary part of the complex index, that is, relating the real (index) and the imaginary (absorption) parts. In other contexts we look for a relationship between the amplitude and the phase, but these are not equivalent, so that when this sort of relationship does not exist we should not be bothered; it simply means that the phase or amplitude cannot be expressed simply in terms of one or the other of the real or imaginary parts.
complex phase shift: \( \eta(\omega) = \eta_r(\omega) + i \eta_i(\omega) \)

scattering amplitude:
\( H(\omega) = \exp(i\eta(\omega)) = \exp(-\eta_i(\omega)) \exp(i\eta_r(\omega)) \)

\( \eta_i(\omega) = -\ln|H(\omega)| \) and \( \eta_r(\omega) = \arg(H(\omega)) \)

\[ \arg \tilde{H}(\omega) = d_0 \omega - \frac{2\omega}{\pi} \int_0^\infty \frac{\ln|\tilde{H}(\Omega)|}{\Omega^2 - \omega^2} d\Omega, \]

in this case, for instance, the imaginary part of the phase response does not determine the real part of the phase response uniquely – \( d_0 \) is undetermined. If \( H(\omega) \) has zeroes in the upper half-plane it is even more complicated.
the imaginary part of the phase shift determines
the magnitude of the scattering amplitude, but
knowledge of the imaginary part of the phase shift
does not determine the real part of the phase shift

it is the real and imaginary parts of the scattering
amplitude that are related by the dispersion
relation, not the phase and amplitude
some objects that violate the simple dispersion relations

reflective Fabry-Perot filter (Gires – Tournois interferometer)

100% reflector

shows resonance behavior in the phase shift on reflection but the amplitude is constant

Also a birefringent filter will apparently give a phase shift without altering the transmission… what is the problem?

what went wrong?

Again, one is asking the wrong question of the dispersion relations – they do not relate amplitude and phase, but rather real and imaginary parts...

We can recover a useful dispersion function by looking at the coupling of modes; a vector dispersion relation.

These authors measured the transmission through a medium that they constructed for microwaves. (We might now refer to this as a meta-material.) It showed the same transmission, but different dispersion for two different polarizations. (oriented 40 and 50 degrees to their “photonic crystal” fast axis.

![Graph](image)

solid = measured
dotted = calculated from measured phases
vector K-K relations

while we have discussed the electromagnetic field as though it is a scaler field, it is actually a vector. If the components of the vector are coupled in the medium, then a vector form of the dispersion relation needs to be used.

\[ b_i(t) = \int_{-\infty}^{\infty} g_{ij}(t - \tau)a_j(\tau) d\tau \]
\[ = \int_{-\infty}^{\infty} g_{ij}(t')a_j(t - t') dt', \]

summing over repeated indicies

for input and output vectors \( \mathbf{a} \) and \( \mathbf{b} \), and a Green’s function response matrix \( g \).

Fourier transforming

\[ \tilde{B}_i(\omega) = \tilde{G}_{ij}(\omega)\tilde{A}_j(\omega), \]
vector dispersion relations

The photonic crystal in their case (or a birefringent tuner, for example) couples the two orthogonal polarizations, so that the scalar Kramers-Kronig relation is not applicable.

A vector relation can be written as:

\[
\text{Re} \tilde{G}_{ij}(\omega) = \frac{2}{\pi} P \int_{0}^{\infty} \frac{\Omega \text{Im} \tilde{G}_{ij}(\Omega)}{\Omega^2 - \omega^2} d\Omega,
\]

\[
\text{Im} \tilde{G}_{ij}(\omega) = -\frac{2\omega}{\pi} P \int_{0}^{\infty} \frac{\text{Re} \tilde{G}_{ij}(\Omega)}{\Omega^2 - \omega^2} d\Omega,
\]
one can obtain a relation between the real and imaginary parts of each component:

\[
\text{Re} \tilde{G}_{ij}(\omega) = \frac{2}{\pi} P \int_0^\infty \frac{\text{Im} \tilde{G}_{ij}(\Omega)}{\Omega^2 - \omega^2} d\Omega,
\]

\[
\text{Im} \tilde{G}_{ij}(\omega) = -\frac{2\omega}{\pi} P \int_0^\infty \frac{\text{Re} \tilde{G}_{ij}(\Omega)}{\Omega^2 - \omega^2} d\Omega,
\]

Basically, any pair of coupled modes can be described in this way... with energy flowing between the modes. The implications for negative dispersion media (generating superluminal or “fast” light), is that causality, in fact, requires it!

Amplitude-phase K-K relations

\[ \arg H(\omega) = d_0 \omega - \frac{2\omega}{\pi} \int_0^\infty \frac{\ln|H(\Omega)|}{\Omega^2 - \omega^2} d\Omega, \]

Calculated using amplitude-phase relation; works for the case where the zeroes are in the lower half-plane, not for the case where they are in the upper half-plane.

Sometimes it works, sometimes it doesn’t…
nonlinear optics

Now that we have some idea of what can go wrong, we can look at a “nonlinear black box” or a “black box” with something optically nonlinear inside.

We can write an expression for a perturbation, $\zeta$, to the index; turn on, say, a light field that alters the properties of a medium – what can you say about the change in the index due to this perturbation?

$$\Delta n(\omega; \zeta) = \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\Delta \alpha(\omega'; \zeta)}{\omega'^2 - \omega^2} \, d\omega'$$

a nonlinear dispersion relation

the perturbation must be held constant
Nonlinear susceptibility $\chi^{(3)}$ relates the polarization to the driving fields

$$P^{(3)}(t) = \chi^{(3)} E_1(t) E_2(t) E_3(t)$$

$$\chi^{(n)}(\omega_1, \omega_2, \ldots, \omega_n) =$$

$$\int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \cdots \int_0^\infty d\tau_n R^{(n)}(\tau_1, \tau_2, \ldots, \tau_n) e^{i(\omega_1 \tau_1 + \omega_2 \tau_2 + \cdots + \omega_n \tau_n)}$$

Now we have a response function for $n$ fields:

$$R^{(n)}(\tau_1, \tau_2, \ldots, \tau_n) = R^{(n)}(\tau_1, \tau_2, \ldots, \tau_n) \theta(\tau_i)$$
\[ \chi^{(n)}(\omega_1, \omega_2, \ldots, \omega_n) = \frac{1}{i\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi^{(n)}(\omega_1, \omega_2, \ldots, \Omega, \ldots, \omega_n)}{\Omega - \omega_i} d\Omega \]

integral is over one frequency – all others are held constant

\[ \Delta n(\omega_a; \omega_b) = \frac{c}{\pi} \mathcal{P} \int_{0}^{\infty} \frac{\Delta \alpha(\omega; \omega_b) d\omega}{\omega^2 - \omega_a^2} \]

change in index due to a perturbation at another frequency

A Kramers-Kronig type relation can thus be given for an “effective” material consisting of the original material plus an electromagnetic field of fixed frequency and intensity.

note: this does not work if the perturbation to the system is due to the field that you need to integrate over in this relation; thus self-action effects are not well handled!
for instance, for a saturated 2-level atom, often noted to violate K-K relations...

\[ \chi(\omega) = \frac{\mu^2 \Delta N_0 (\omega_0 - \omega) + i/T_2}{\varepsilon_0 \hbar (\omega - \omega_0)^2 + (1 + I/I_s) / T_2^2} \]

poles occur at

\[ \omega = \omega_0 \pm (i/T_2)(1 + I/I_s)^{1/2} \]

since there is a pole in the upper half plane it is no longer analytic in this regime.

The perturbation is the excited state population and this is altered by the driving field. If one changes the problem to consider probing the 2-level atom with a weak field while keeping the saturating driving field (perturbation) constant, you can again write a K-K-type relation:
\[
\chi(\omega; \Omega) = \left( \frac{\mu^2 \Delta N_0}{\varepsilon_0 \hbar} \right) \left( \frac{(\omega_0 - \omega) + i/T_2}{(\omega_0 - \omega)^2 + 1/T_2^2} \right) \left( \frac{(\omega_0 - \Omega)^2 + 1/T_2^2}{(\omega_0 - \Omega)^2 + (1 + I/I_s)/T_2^2} \right)
\]

\[
= \chi_0(\omega) \frac{(\omega_0 - \Omega)^2 + 1/T_2^2}{(\omega_0 - \Omega)^2 + (1 + I/I_s)/T_2^2}
\]

written for a pump with intensity \( I \) and frequency \( \Omega \).

\( \chi_0 \) is the low-power limit of the susceptibility from the first two factors.

This satisfies the K-K relations for a saturated 2-level atom, provided that we make the perturbation (saturation) be due to a separate field than the one we want to consider as a probe.
Thus, we have removed the problem for a 2-level atom, provided that we make the perturbation (saturation) be due to a separate field than the one we want to consider.

Imaginary part of the susceptibility for the degenerate 2-level atom (left) and the nondegenerate case (right) where the saturation is provided by another field; this now obeys a K-K relation with the real part.

Thus, we have removed the problem for a 2-level atom, provided that we make the perturbation (saturation) be due to a separate field than the one we want to consider.
Similarly, such relations can be given for static field perturbations to the system ($\Omega = 0$). For instance, photorefractive crystals can be described in this way.

Generally, there will be a relationship between the gain/loss or amplitude response and the dispersion of the medium, but it will sometimes not be simple, especially if “self action” effects are important.
gain media

Causality alone is sufficient for establishing the dispersion relations in passive dielectric media. This is no longer true in active dielectric or magnetic media because instabilities can exist there.

Media for which we have to worry are things like lasers, with gain saturation (nonlinearity) or instability.

amplifiers – gain is “linear” and you can describe it with linear susceptibilities and a complex refractive index... unless there is saturation or some instability...

instabilities

absolute, convective, or global instabilities

absolute: the field blows up at a particular point in space
convective: the field blows up, but the point moves; self-focusing, for instance
global: the boundary conditions lead to the field blowing up; a finite gain medium in a cavity

Absolute instabilities arise when $n(\omega)$ cannot be identified with an analytic function in the upper complex half-plane ($\text{Im}(\omega) > 0$); if $\varepsilon(\omega)\mu(\omega)$ contains poles or odd-order zeroes.
Even though media that satisfy the usual K-K relations are causal, not all causal media satisfy K-K relations.

- For instance, a response function may increase exponentially; this implies a pole in the upper half-plane.

Singularities in the upper half-plane do not indicate non-causality, but rather instability or diverging fields.
The magic of dispersion relations

Note that it is not that there is no relation between the real and imaginary parts of the susceptibility when we cannot write a K-K type relation; one can write a relation similar to the K-K relation, however to write the relation you need to evaluate the residues at the poles in the upper half-plane. Unfortunately to do this one needs to know the functional form of the susceptibility, thereby making it less useful. (That is, you cannot use either just the real or just the imaginary part of $\chi$ to determine the other.

Causality is still obeyed.
basics of 4WM

a “parametric” process; it leaves the (atomic) medium in the state that it started in...

it does not have to involve a “real” transition and can be off-resonant
basics of 4WM

Not all of the fields have to be the same energy, but energy should be conserved!
phase matching conditions

energy conservation: \( \omega_1 + \omega_2 = \omega_3 + \omega_4 \)

momentum conservation: \( \mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{k}_4 \)

note that the momentum is the momentum in the medium!
phase matching for our conditions

energy conservation: \( \omega_0 + \omega_0 = \omega_p + \omega_c \)

momentum conservation: \( \mathbf{k}_0 + \mathbf{k}_0 = \mathbf{k}_p + \mathbf{k}_c \)

The index of refraction of the atomic vapor, and k-vectors of probe and conjugate, are changed by the intensity of the pump beam; phase matching is only achieved at a small angle from the pump.
4WM in hot Rb vapor

$^{85}$Rb in a double-$\Lambda$ scheme

- $\sim$120°C cell temp.
- $\sim$1 GHz detuned
- $\sim$400 mW pump
- $\sim$100 $\mu$W probe
  - narrowband
  - no cavity
4WM in materials

- photons do not interact on their own, so we need a material
- $\chi^{(2)}$ interaction (3WM) in optics (parametric downconversion) requires asymmetric materials or surfaces
- $\chi^{(3)}$ interaction in optics is provided by a nonlinear material – this can happen in symmetric materials like vapors
- nonlinear atomic interactions (4WM in matter-waves) – this does not require another “medium”!
4-wave mixing

• in general, we have 4 fields coupled by a nonlinear interaction ($\chi^{(3)}$)

• this can be used to mix different frequencies or directions to generate new frequencies or phase conjugate beams

"forward" geometry

\[ \omega_1, k_1 \quad \omega_3, k_3 \quad \omega_2, k_2 \quad \omega_4, k_4 \]

"backward" geometry

\[ \omega_1, k_1 \quad \omega_2, k_2 \quad \omega_3, k_3 \quad \omega_4, k_4 \]
what do we expect from 4WM in a double-lambda system?

intensity correlations (parametric process)

leads to intensity difference squeezing

entanglement

EPR violations?
4WM - with a difference

typically:
a near-resonant, 2-level system

alternatively:
an off-resonant, 4-level system

4WM based on ground-excited coherences and ensuing absorption and spontaneous emission noise

Slusher, et al., 1985

4WM based on ground-ground coherences reduces these noise sources

Harris, Lukin, Vuletic...
entangled twin beams

- intensity difference squeezing tells you about correlations
- entanglement requires two variables simultaneously squeezed: intensity-difference and phase-sum correlations
- inseparability of the wavefunction
- continuous-variable EPR violations (guarantee for mixed state) require a level of squeezing beyond 50% (3 dB) in each variable
FWM and Atomic Coherence

- Use double-$\Lambda$ system.

\[
\begin{align*}
P_p &= \epsilon_0 \chi_p^{(1)}(\omega_p) E_p + \epsilon_0 \chi_p^{(NL)}(\omega_p = \omega_1 + \omega_2 - \omega_c) e^{i\Delta k z} E_c^* \\
P_c &= \epsilon_0 \chi_c^{(1)}(\omega_c) E_c + \epsilon_0 \chi_c^{(NL)}(\omega_c = \omega_1 + \omega_2 - \omega_p) e^{i\Delta k z} E_p^*
\end{align*}
\]

- For generation of squeezing need FWM term to dominate.
Response of Double-Λ System

Ideal Case

No Atomic Coherence
Slow Light

EIT (less loss) spectrum with associated dispersion

gain features imply dispersion and slow light as well
slow light

an example of other things to go wrong

in 4WM we have coupled propagation equations

for the fields; clearly there is a “self-action” of

the probe due to the generation of the

conjugate, acting back on the probe...
coupled propagation

$\chi(3)$ expression for a 4-level atom given by:

This will tell you the small-signal dispersion; i.e., the group velocity for a weak pulse. The presence of gain in the system, however, means that not all small signals will stay small!
4WM gain spectrum

experiment

Calculated gain line (without Doppler broadening)

can calculate $v_g$ as well...

$v_g$-conjugate = 2 $v_g$-probe!
pulse delay

\[ \delta = 10 \text{ MHz} \]
300 mW pump
10% broadening

\[ \delta = 22 \text{ MHz} \]
250 mW pump
5% broadening

pulse breakup when detuned closer to Raman absorption dip

70 ns FWHM
Gain \( \sim 13 \)

![Graph showing pulse delay and its effects with different pump powers and detunings.](image)
delay-locking of matched pulses

probe initially travels at $v_g$, then locks to the conjugate with velocity $2v_g$ when delay reaches lock point determined by gain

Lesson learned: we can’t always pay attention to just the dispersion relation!

You have to be careful when you consider a “black box” and you can’t just ignore the physics inside the box, and small signal response is not always the best indicator.

Here the 4WM process gives us coupled propagation of the two fields, so that one is not keeping “the perturbation” fixed...