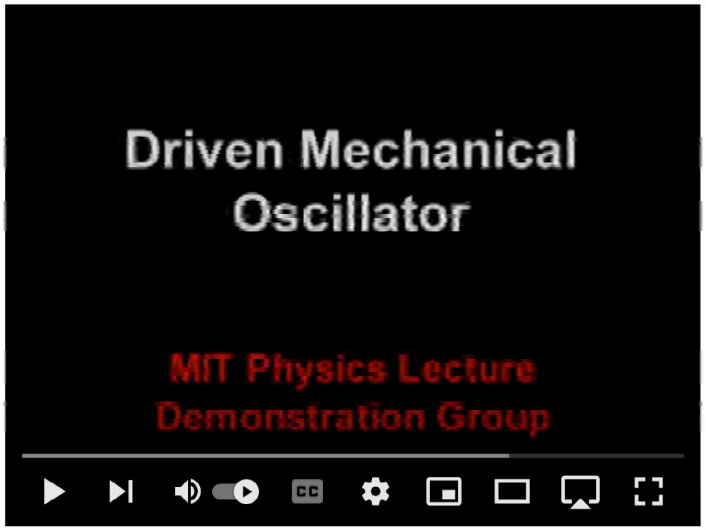
Light-Matter Interaction

Video of driven – damped harmonic oscillator

https://www.youtube.com/watch?v=aZNnwQ8HJHU



Complex polarizability:

$$\boxed{\begin{aligned} \vec{p} &= e\vec{x} = e\vec{a}e^{-i(\omega t - kz)} \equiv \alpha(\omega)\vec{\varepsilon}E_0 e^{-i(\omega t - kz)} \\ \alpha(\omega) &= \frac{e^2/m}{\omega_0^2 - \omega^2 - 2i\beta\omega} = \frac{e^2}{m}\frac{\omega_0^2 - \omega^2 + 2i\beta\omega}{(\omega_0^2 - \omega^2)^2 + 4\beta^2\omega^2} \end{aligned}}$$

Easy to show that if $\vec{E}(\vec{R},t) = \vec{\varepsilon}E_0 e^{-i(\omega t - kz)}$

and $\vec{P} = N\vec{p} = N\alpha(\omega)\vec{\varepsilon}E_0 e^{-i(\omega t - kz)}$

then the wave equation

$$\nabla^2\vec{E} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{E} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\vec{P}$$

takes the form

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{\varepsilon}E_0 e^{-i(\omega t - kz)} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\vec{\varepsilon}E_0 e^{-i(\omega t - kz)}$$

and thus $k^2 = \frac{\omega^2}{c^2}\left(1 + \frac{N\alpha(\omega)}{\varepsilon_0}\right)$

We define the complex index of refraction

$$\boxed{k^2 = \frac{\omega^2}{c^2}\left[1 + \frac{N\alpha(\omega)}{\varepsilon_0}\right] \equiv \frac{\omega^2}{c^2}n(\omega)^2}$$

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Complex Index of Refraction – Physical discussion

Let $\boxed{n(\omega) = n_R(\omega) + i n_i(\omega)}$

Plane wave propagation $k = n(\omega)\omega/c$

$$\vec{E}(z,t) = \vec{\varepsilon} E_0 e^{-i(\omega t - kz)}$$
$$= \vec{\varepsilon} E_0 e^{-i(\omega t - [n(\omega)\omega/c]z)}$$
$$= \vec{\varepsilon} E_0 e^{-n_i(\omega)\omega z/c} e^{-i\omega(t - n_R(\omega)z/c)}$$

We can now identify

$\frac{c}{\omega n_i(\omega)}$ ←	attenuation length
$\frac{c}{n_R(\omega)}$ ←	phase velocity

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Complex Index of Refraction – Physical discussion

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Plane wave propagation

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$\dfrac{c}{n_R(\omega)}$	←	phase velocity

Absorption

The intensity of a plane wave field E is

$$I_\omega(z) = \frac{1}{2} n_R(\omega) c \varepsilon_0 |E(c,z)|^2 = I_0(0) e^{-2n_i(\omega)\omega z/c}$$
$$\equiv I_0 e^{-a(\omega)z}$$

where the absorption coefficient is

$$a(\omega) \equiv 2 n_I(\omega)\,\omega/c = \frac{2\omega}{c} \mathrm{Im}\left[\left(1 + \frac{N\alpha(\omega)}{\varepsilon_0}\right)^{1/2}\right]$$

Possibility of gain ?

No – there is no energy source !

Light-Matter Interaction

Absorption and Dispersion in Gases

Approximations:

$|\omega_0-\omega| \ll \omega_0, \omega$ near resonance

$|n(\omega)| \sim 1$ weakly polarizable

Let $\omega_0^2-\omega^2 = (\omega_0+\omega)(\omega_0-\omega) \approx 2\omega(\omega_0-\omega)$

$$\alpha(\omega) = \frac{e^2/m}{\omega_0^2-\omega^2-2i\beta\omega} = \frac{e^2/2m\omega}{\omega_0-\omega-i\beta}$$

$$= \frac{e^2}{2m\omega}\,\frac{\omega_0-\omega+i\beta}{(\omega_0-\omega)^2+\beta^2}$$

Furthermore

$$n(\omega)^2 = 1+\frac{N\alpha(\omega)}{\varepsilon_0} = 1+\varepsilon,\ \varepsilon \ll 1$$

Expand to 1st order $(1+\varepsilon)^{1/2} \approx 1+\varepsilon/2$

Putting it together

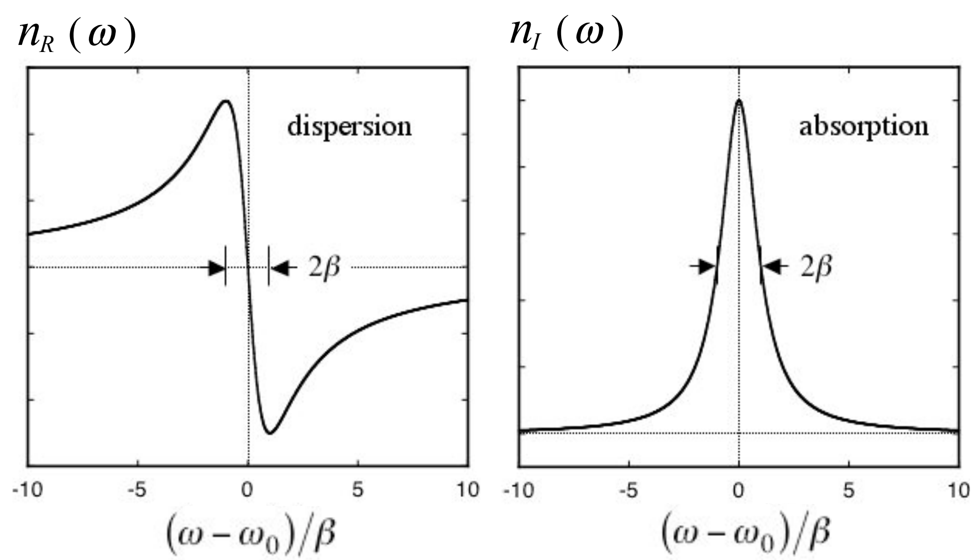
$$n_R(\omega) = 1+\frac{Ne^2}{4\varepsilon_0 m\omega}\,\frac{\omega_0-\omega}{(\omega_0-\omega)^2+\beta^2}$$

dispersive line shape

$$n_I(\omega) = \frac{Ne^2}{4\varepsilon_0 m\omega}\,\frac{\beta}{(\omega_0-\omega)^2+\beta^2}$$

Lorentzian line shape

General behavior:



Light-Matter Interaction

Putting it together

$$n_R(\omega) = 1 + \frac{Ne^2}{4\varepsilon_0 m\omega}\,\frac{\omega_0-\omega}{(\omega_0-\omega)^2+\beta^2}$$

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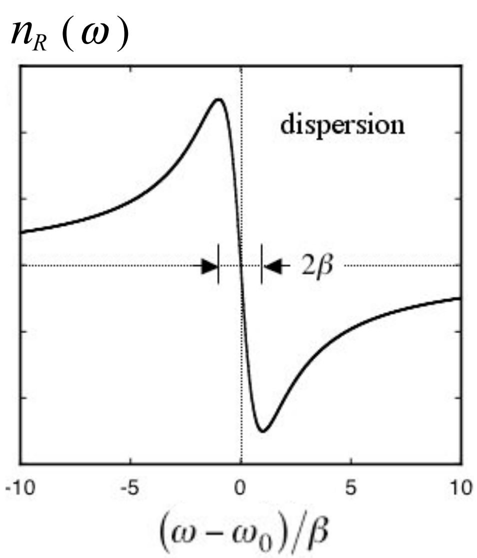
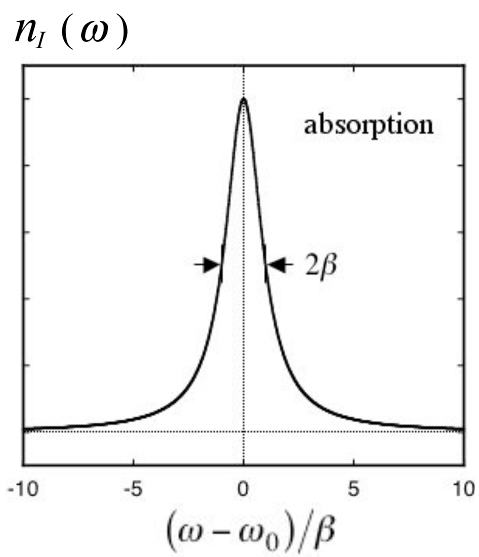




Note: for $|\omega_0-\omega| \gg \beta$

dispersion $\propto \dfrac{1}{(\omega_0-\omega)}$

absorption $\propto \dfrac{1}{(\omega_0-\omega)^2}$

➡ We can have loss-less dispersive media

Note: If we introduce the detuning $\Delta = (\omega_0-\omega)$ we can rewrite $n_R(\omega)$, $n_I(\omega)$ as

$$\boxed{\begin{aligned} n_R(\Delta) &= 1 + \frac{Ne^2}{4\varepsilon_0 m\omega}\,\frac{\Delta}{\Delta^2+\beta^2} \\ n_I(\Delta) &= \frac{Ne^2}{4\varepsilon_0 m\omega}\,\frac{\beta}{\Delta^2+\beta^2} \end{aligned}}$$

From the above we see that

$n_R(\omega) < 1$ for $\omega > \omega_0$ ➡ $\dfrac{c}{n_R(\omega)} \gg c$

Superluminal propagation ?

End 01-18-2024

Light-Matter Interaction

Begin 01-23-2024

Light-Matter Interaction

Free Electrons

Consider the limit $\omega \gg \omega_0$

effectively unbound electrons

This is a reasonable model of plasmas & metals

In this limit we have

$$\alpha(\omega) = \frac{e^2/m}{\omega_0^2 - \omega^2 - 2i\beta\omega} \approx -\frac{e^2}{m\omega}$$

$$n(\omega) = \sqrt{1 + \frac{N\alpha(\omega)}{\varepsilon_0}} \approx \sqrt{1 - \frac{Ne^2}{\varepsilon_0 m \omega^2}} \equiv \sqrt{1 - \frac{\omega_p^2}{\omega^2}}$$

We introduce the <u>Plasma Frequency</u>

$$\boxed{\omega_p = \sqrt{\frac{Ne^2}{\varepsilon_0 m}}}$$

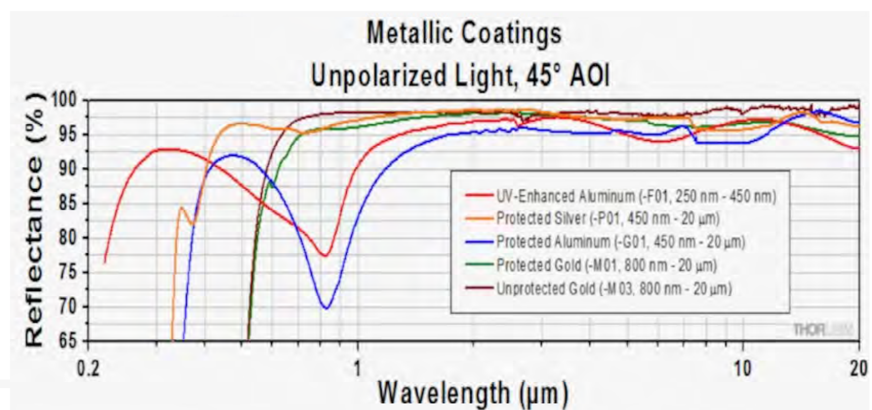
Let $\left.\begin{array}{l}\omega_0 \ll \omega \ll \omega_p \\ |\omega_0 - \omega| \gg \beta\end{array}\right\}$ $\Rightarrow$ $n(\omega)$ purely imaginary - but no loss!

We now have

$$\vec{E}(z,t) = \vec{\varepsilon} E_0 e^{-i\omega[t - n(\omega)z/c]}$$
$$= \vec{\varepsilon} E_0 e^{-i\omega t} e^{i(z/c)\sqrt{\omega^2 - \omega_p^2}}$$
$$= \vec{\varepsilon} E_0 e^{-i\omega t} e^{-b(\omega) z}$$

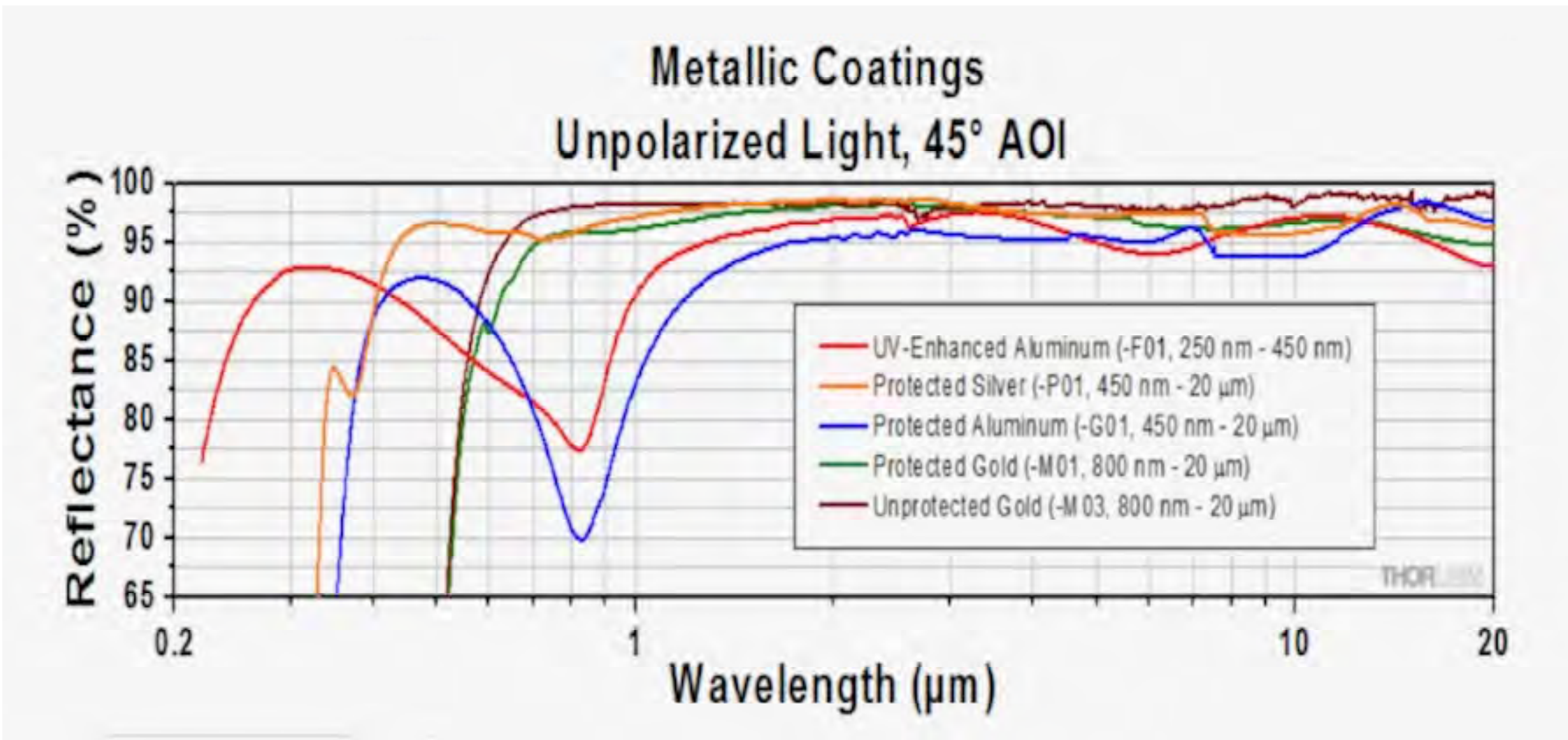
where $b(\omega) = -\frac{i}{c}\sqrt{\omega^2 - \omega_p^2}$

Reflection at surface penetration depth $\sim 1/b(\omega)$



Light-Matter Interaction, Free Electrons

Examples of this kind of medium includes plasmas, and metals such as aluminum, silver and gold which are known to be excellent mirrors for visible and IR radiation.



Quantum Theory of Light-Matter Interaction

Completed:

– Fully classical description of fields & Atoms

Next Step:

– Semiclassical description $\left\{ \begin{array}{l} \text{Classical field} \\ \text{Quantum atoms} \end{array} \right.$

Self-Consistent Description

Electromagnetic Field $\rightarrow$ Atom/Molecule/Solid

Needed: Quantum theory of atomic response analogous to classical $\vec{p} = \alpha(\omega)\vec{E}$

Note: In QM the dipole is an Observable

Observable = Hermitian operator $\hat{\vec{p}}$

Classical Field = C-valued vector $\vec{E}$

Cannot plug into Wave Eq. for classical field !

Wave Equation w/classical field & atoms

$$\boxed{\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\vec{P}, \qquad \vec{P} = N\vec{p}}$$

How do we deal with this mis-match?

Repeated measurements of $\vec{p}(t)$

Quantum fluctuations $\vec{p}(t) = \langle \vec{p}(t) \rangle + \Delta \vec{p}(t)$

where $\langle \vec{p}(t) \rangle = \langle \psi(t) | \hat{\vec{p}} | \psi(t) \rangle$ mean

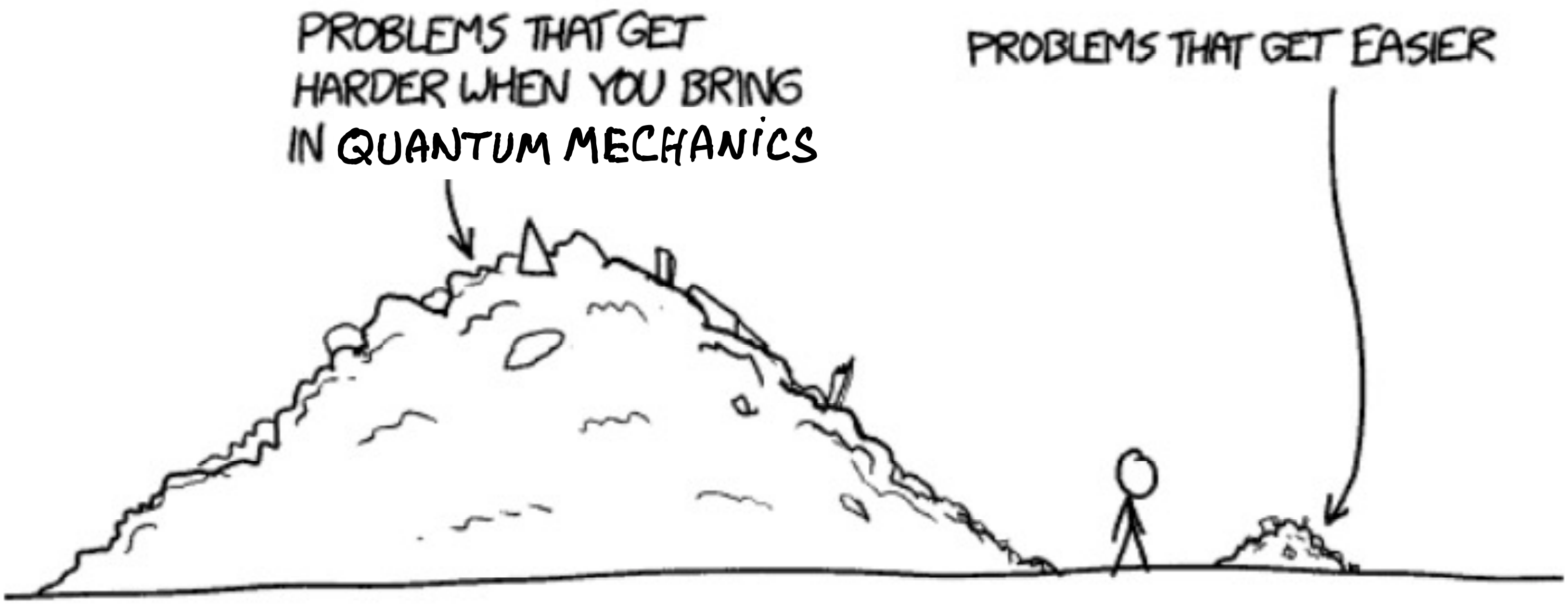
$\Delta\hat{\vec{p}}(t)$ fluctuations

Note: Given $|\psi(t=0)\rangle$ and $\vec{E}$ the mean $\langle \vec{p}(t) \rangle$ follows from the Schrödinger Eq., radiates <u>coherently</u> like classical $\vec{p}(t)$

$\langle \vec{p}(t) \rangle$ is a Real-valued vector (more later)

$\Rightarrow$ we can plug it into the Wave Eq.

Quantum Theory of Light-Matter Interaction



Source: xkcd.com

Quantum Theory of Light-Matter Interaction

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Electromagnetic Field $\rightarrow$ **Atom/Molecule/Solid** $\rightarrow$ **Electromagnetic Field**

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Quantum theory of atomic response

analogous to classical $\vec{p} = \alpha(\omega)\vec{E}$

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radiates <u>coherently</u> like classical $\vec{p}(t)$

$\langle \vec{p}(t) \rangle$ **is a Real-valued vector (more later)**

$\Rightarrow$ **we can plug it into the Wave Eq.**

Quantum Theory of Light-Matter Interaction

Wave Equation w/classical field & atoms

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Note:
- The Equations look very similar
- Polarizability, index of refraction, etc will be *very different* in some regimes
- Notably, the model is no longer linear in $\vec{E}$ and will lead to phenomena like saturation and wave mixing
- $\Delta\hat{\vec{p}}(t)$ represents quantum fluctuations driven by the empty modes of the EM field, a process also responsible for spontaneous decay.

Note: Do not identify $\langle \vec{p} \rangle$ and $\Delta\vec{p}$ with Stimulated and spontaneous emission. Those labels are not meaningful here.

Quantum Theory of Light-Matter Interaction

Wave Eq. w/classical field & quantum atoms

$$\boxed{\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\vec{P}, \quad \vec{P} = N\langle\vec{\mu}\rangle}$$

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Atom-field interaction

Hamiltonian: $\boxed{H = H_a + V_{ext}(\vec{R}, t)}$

H_a : time-independent atomic Hamiltonian

V_{ext} : time-dependent driving term, non necessarily a perturbation

Question: Time evolution of the atomic system? Is there a steady state?

Schrödinger Eq.: $\boxed{i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle = H|\psi(t)\rangle}$

Expand in basis $\{|\varphi_n\rangle\}$ of eigenstates of H_a

$$|\psi(t)\rangle = \sum_n a_n(t)|\varphi_n\rangle, \quad H_a|\varphi_n\rangle = E_n|\varphi_n\rangle$$

Quantum Theory of Light-Matter Interaction

Atom-field interaction

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Plug into S. E.

$$i\hbar \sum_n \dot{a}_n(t) |\varphi_n\rangle = \sum_n a_n(t) \left[E_n + V_{ext} \right] |\varphi_n\rangle$$

Take scalar product w/ $|\varphi_m\rangle$ on both sides

$$i\hbar \sum_n \dot{a}_n(t) \langle \varphi_m | \varphi_n \rangle = \sum_n a_n(t) \left[E_n \langle \varphi_m | \varphi_n \rangle \right] + \overbrace{\langle \varphi_m | V_{ext} | \varphi_n \rangle}^{V_{mn}}$$

On vector-matrix form this can be written

$\boxed{i\hbar \underline{\dot{a}} = \underline{\underline{H}}_a \underline{a} + \underline{\underline{V}} \underline{a}}$

S.E. in $\{|\varphi_n\rangle\}$ rep.

Still exact!

Quantum Theory of Light-Matter Interaction

Plug into S. E.

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Still exact!

Note: If $\underline{\underline{H}}_a$ and $\underline{\underline{V}}_{ext}$ are known we can do

- Perturbation Theory
  (OK for short times or "weak" driving fields)
- Numerical integration of the S. E.
- Few-level approximations to simplify and obtain analytical solutions outside the perturbative regime

General problem:
No analytical solution!

Atom-Light Interaction: 2-Level Approximation

General observation:

- Atoms and molecules often behave as if they have a single, dominant transition frequency
- We expect this when the freq. of the driving is resonant with one transition $|\varphi_n\rangle \rightarrow |\varphi_m\rangle$ and far off resonance with all others.

Interaction $\boxed{V_{ext} = -\vec{p}\cdot\vec{E}(t)}$

State space $\mathcal{D}im(\mathcal{E}) = 2\,,\ \{|1\rangle, |2\rangle\}$

$$|2\rangle \quad \updownarrow \hbar\omega_{21} \quad |1\rangle$$

State vector

$$|\psi(t)\rangle = a_1(t)\,|1\rangle + a_2(t)\,|2\rangle$$

Schröd. eq.

$$i\hbar\dot{a}_1 = E_1 a_1 + V_{11} a_1 + V_{12} a_2$$
$$i\hbar\dot{a}_2 = E_2 a_2 + V_{21} a_1 + V_{22} a_2$$

Interaction

$$V_{12}(t) = -\vec{p}_{12}\cdot\tfrac{1}{2}(\hat{\varepsilon}E_0 e^{-i\omega t} + c.c.)$$
$$V_{21}(t) = -\vec{p}_{21}\cdot\tfrac{1}{2}(\hat{\varepsilon}E_0 e^{-i\omega t} + c.c.)$$

Parity selection rule

Definition: $\vec{r} \rightarrow -\vec{r}$ is a reflection through the origin

Atomic Hamiltonian $H \propto \frac{1}{r} \Rightarrow H(\vec{r}) = H(-\vec{r})$

$\Rightarrow$ **Eigenstates** $\varphi(\vec{r}) = \pm\,\varphi(-\vec{r}) = \varphi(-[-\vec{r}])$

"+" for even parity
"-" for odd parity

two reflections equals the identity

The dipole $\vec{p}$ **is a vector operator** $\Rightarrow$ **transforms like a vector when** $\vec{r} \rightarrow -\vec{r}$

Thus $\vec{p}(\vec{r}) = e\vec{r} = -\vec{p}(-\vec{r})$ **and**

$\vec{p}_{nm} = \int d^3r\, \varphi_n^*(\vec{r})\,\vec{p}\,\varphi_m(\vec{r}) \neq 0$ **only when**

φ_n **and** φ_m **have** <u>**opposite parity**</u>

Parity rule: **No dipole moment in energy eigenstate !**

$$\boxed{\begin{array}{l} \vec{p}_{12} = \langle 1|\hat{\vec{p}}|2\rangle, \quad \vec{p}_{21} = \vec{p}_{12}^{\,*} \\ \vec{p}_{11} = \vec{p}_{22} = 0 \Rightarrow V_{11} = V_{22} = 0 \end{array}}$$