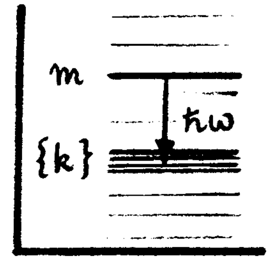
Discussion of Exponential Decay Law.



9) EXAMPLE Exponential Decay of the Initial State m.

Use FERMI'S GOLDEN RULE to find population of initial state m at time t into an emission process: $E_m^{(0)} \to E_k^{(0)} + \hbar\omega$.

1. Let $\mathcal{P}_m(t)$ be probability of finding system in state m at time t. The probability $\mathcal{P}_m(t+dt)$ of finding m at time $t+dt$ is fixed by 2 factors, viz. (A) m existed at time t, (B) m did not make a transition in t to $t+dt$.

So/ $$\mathcal{P}_m(t+dt) = \underbrace{\mathcal{P}_m(t)}_{(A)}\underbrace{[1-W\,dt]}_{(B)}$$

w/ $W = W(m\to\{k\})$ of Eq.(29). (30)

(A) = prob. of m occupied at time t,
(B) = prob. of no $m\to k$ transitions in next dt ($W\,dt$ = prob. of YES, so $(1-W\,dt) \leftrightarrow$ NO; W is time-independent.

2. Expand (30) to 1st-order ∞simals...

$$\mathcal{P}_m(t) + \left(\frac{d\mathcal{P}_m}{dt}\right)dt = \mathcal{P}_m(t)[\cancel{1}-W\,dt] \Rightarrow \boxed{\mathcal{P}_m(t) = \mathcal{P}_m(0)\,e^{-Wt}}. \quad (31)$$

We get the exponential decay law: population of state m declines exp'lly.

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3. QM puzzle: $m \rightleftarrows \{k\}$ should engage in a "quantum oscillation" (p. tD8), and thus m should be replenished just as often as it is depleted... so how does it decay? Answer: the $\{k\}$ have slightly different energies, so the replenishment $\{k\}\to m$ provides "feedback" amplitudes at different phases; these amplitudes tend to cancel, so m can suffer a net loss.

4. QM Objection: Eq.(30) tacitly assumes the classical idea that the act of fixing system in state m at time t [i.e. $\mathcal{P}_m(t)$] does not influence its future development [i.e. no transition in dt]. Instead of (30), we should write QM'ly:

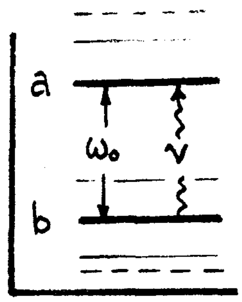
$$\to \mathcal{P}_m(t+dt) = |a_m(t+dt)|^2 = \mathcal{P}_m(t)\left|1+(\dot{a}_m/a_m)dt\right|^2, \quad \mathcal{P}_m = |a_m|^2. \quad (32)$$

Can (32) be reconciled with (30)? The answer is YES, and $m\to\{k\}$ gives exponential decay. We shall provide more details via Weisskopf Wigner Theory.

Two-level Problem: AM–FM dependence of transitions.

10) Starting from the lowest order transition amplitude $a_k^{(1)}(\infty) = -\frac{i}{\hbar}\int_{-\infty}^{\infty} V_{km}(\tau)\, e^{i\omega_{km}\tau}\, d\tau$ in Eq. (23), we have demonstrated quantum oscillations, Fermi's Golden Rule for transition rates, and the plausibility of exponential decay for excited states. Much more can be done with the $a_k^{(1)}(\infty)$'s, but we will mention only one more application, before moving on to higher order terms in this theory (i.e. the $a_k^{(\mu)}(t)$, $\mu > 1$), and also alternative ways of looking at time-dependent transitions.

$\boxed{\text{EXAMPLE}}$ AM–FM dependence of transitions.



1. Consider a "two-level" QM system†, where the levels are represented by amplitudes $a(t)$ & $b(t)$ and are (initially) separated in energy by $\hbar\omega_0$. An absorptive transition $b \to a$ is driven @ freq. $\nu \simeq \omega_0$ by an external field, represented by a potential $U(t, \nu)$. By Eq. (23) above, the final state amplitude is given (to lowest, or leading order) by*

$$\rightarrow i\,a(\omega_0, \nu) = \int_{-\infty}^{\infty} U(t,\nu)\, e^{i\omega_0 t}\, dt, \tag{33}$$

and it is a fcn of the frequencies ω_0 & ν. Other than being "weak" (i.e. $|\max\{U\}| \ll \omega_0$), there is no restriction on the form of the coupling $U(t,\nu)$.

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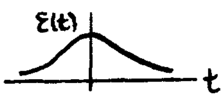
2. If U were monochromatic @ freq. ν, it could be written: $U = \mathcal{E}(t)\, e^{-i\nu t}$, where $\mathcal{E}(t)$ is an envelope fcn (usu. in form of a pulse: 

). To generalize this a bit, allowing U to have a frequency spectrum, we write:

$$\rightarrow U(t,\nu) = \mathcal{E}(t)\int_{-\infty}^{\infty} \delta(\omega - \nu)\, e^{-i\omega t}\, d\omega. \tag{34}$$

[next page]

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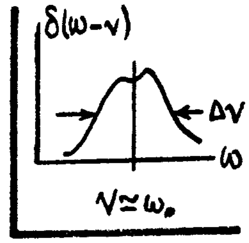
* Notation is a bit simplified... $\hbar$ has been incorporated in U, subscripts "ab" have been dropped on U, $\omega_0 = \omega_{ab}$, etc., and we've eliminated the $^{(1)}$ on the ampl. a.

† The system may actually have many more levels, but we can concentrate on just two, if the transition $b \to a$ is "tuned"... i.e. if the driving freq $\nu \simeq \omega_0$.

Transition Lineshape : AM-FM dependence.



The spectral fcn $\delta(\omega-\nu)$ is peaked around $\omega \sim \nu \sim \omega_0$ but is otherwise arbitrary. The coupling $U(t,\nu)$ in Eq. (34) can be adjusted in <u>3 ways</u>: the central frequency ν can be tuned, the envelope fcn $\mathcal{E}(t)$ can be changed around [this allows "AM" (i.e. <u>amplitude</u> modulation) adjustment], and the spectrum $\delta(\omega-\nu)$ can be chosen [allowing "FM" (i.e. <u>frequency</u> modulation) modifications].

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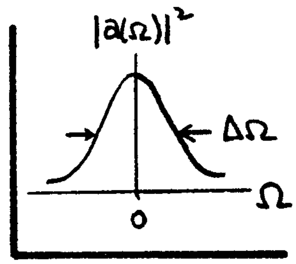
3. Now put (34) into (33)...

$$i a(\omega_0,\nu) = \int_{-\infty}^{\infty} dt\, e^{i\omega_0 t}\, \mathcal{E}(t) \int_{-\infty}^{\infty} d\omega\, e^{-i\omega t}\, \delta(\omega-\nu) \int \begin{matrix} \text{define}: \Omega = \omega_0 - \nu \\ \text{let}: k = \omega_0 - \omega \end{matrix}$$

$$= \int_{-\infty}^{\infty} dk\, \delta(\Omega-k) \underbrace{\int_{-\infty}^{\infty} dt\, \mathcal{E}(t)\, e^{ikt}}_{2\pi \tilde{\mathcal{E}}(k),\ \tilde{\mathcal{E}} = \text{F.T. of } \mathcal{E};\P}$$

so//

$$\boxed{i a(\Omega) = 2\pi \int_{-\infty}^{\infty} dk\, \underbrace{\delta}_{\text{FM}}(\underbrace{\Omega-k}_{\text{tuning}})\, \underbrace{\tilde{\mathcal{E}}(k)}_{\text{AM}}}, \quad \Omega = \omega_0 - \nu = \text{"}\underline{\text{detuning freq.}}\text{"} \tag{35}$$



A plot of $|a(\Omega)|^2$ vs Ω gives the "lineshape" for the $b \to a$ transition; generally we get a <u>resonance</u> @ $\Omega = 0$ (i.e. $\nu = \omega_0$), where the transition is most easily driven. Evidently we can "adjust" the $b \to a$ lineshape by adjusting the FM & AM factors flagged in Eq. (35).

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4. The AM-FM adjustments indicated in $a(\Omega)$ of Eq. (35) have practical applications... e.g. we may wish to <u>suppress</u> the absorption when broadcasting an EM pulse $U(t,\nu)$ through a dispersive medium, or to <u>enhance</u> absorption for NMR diagnostic studies. Pulses may be chosen which either broaden or narrow the absorption resonance... see R.T. Robiscoe, Phys. Rev. A **40**, 4781 (1989), and Prob. # O.

¶ We use the convention: $F(t) = \int_{-\infty}^{\infty} f(\omega)\, e^{-i\omega t}\, d\omega$, and inverse: $f(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} F(t)\, e^{i\omega t}\, dt$.

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Absorptive line broadening or narrowing by frequency-modulated pulses

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We consider an absorption process in a two-level atom for which the driving pulse is controlled in both its frequency modulation (FM) and its amplitude modulation (AM), or temporal shape. In the weak-signal limit, we find that a variety of FM-AM combinations can provide either broadening or narrowing of the absorption line shape, along with an enhancement or suppression of the atomic absorptivity. We develop a simple analytic criterion for the driving pulses which induce absorptive line broadening or narrowing, and give examples of each type. The absorptivity increments can be exploited in spectroscopic and signal broadcast applications.

INTRODUCTION

Recently, on the subject of driving absorptive transitions in a sample of two-level atoms, it was noted that by using special types of excitation pulses whose spectral content and temporal shape are carefully controlled, the absorption line profile can be significantly enhanced.[1,2] Furthermore, these pulses, with specially chosen frequency modulation (FM) and amplitude modulation (AM) features, propagate through the atomic medium without substantial distortion. Clearly, such control over the pulse absorption and propagation characteristics has important spectroscopic applications, as well as use for signal broadcast devices. In this Brief Report, we show, in the weak signal limit, that when the pulse AM-FM content can be controlled while driving an atomic transition, there are many pulse choices which can either enhance or suppress the atomic absorption. Moreover, we derive a simple analytic criterion for deciding which pulse shapes lead to enhanced or suppressed absorptivity.

We shall work within first-order perturbation theory for transitions in a two-level atom which are driven by an incident em (electromagnetic) pulse. For simplicity, we ignore the space dependence of the pulse and the induced atomic polarization, etc., and we also ignore any relaxation mechanisms for the atoms per se. Thus our calculation is restricted to weak pulses incident near resonance on atoms which show no collective effects. However, even in this simple system, we can display some novel effects on the absorption line which are connected with the pulse AM-FM content.

When an absorptive atomic transition $b \rightarrow a$ is driven near its resonant frequency ω_0 by a *monochromatic* pulse of slowly varying amplitude and long duration τ, the absorptive linewidth $\Delta\omega$ is limited only by the natural widths of the states b and a; if the states are long lived, then $\Delta\omega \sim 1/\tau \rightarrow 0$, and the absorption line is arbitrarily narrow. If, however, the driving pulse has an FM component with an intrinsic frequency spread $\Delta\nu$, then the $b \rightarrow a$ linewidth is generally broadened by just this amount. In what follows, we show it is possible to compensate for this FM broadening by proper choice of the temporal shape (i.e., AM content) of the driving pulse. In general, both the $b \rightarrow a$ linewidth and total absorption can be decreased or increased by appropriate choice of the pulse AM component.

PERTURBATION ANALYSIS

For an absorptive transition $b \rightarrow a$ at frequency ω_0 in a two-level atom, driven by a weak coupling pulse $U(t)$, first-order time-dependent perturbation theory gives the final-state amplitude as[3]

$$ia = \int_{-\infty}^{\infty} U(t)\exp(i\omega_0 t)dt \ . \tag{1}$$

In this approximation, state b is initially fully populated and is assumed to be negligibly depleted by transitions $b \rightarrow a$; state a is then populated according to the Fourier transform of $U(t)$, so the $b \rightarrow a$ absorption is sensitive to the spectral content of $U(t)$. Usually, $U(t)$ has frequency components $\nu \sim \omega_0$, near resonance, so that the $b \rightarrow a$ absorption is relatively "large." In this case, the nature of the absorption can change markedly with the frequency components carried by $U(t)$.

As a suitably general coupling pulse in Eq. (1), we consider an FM pulse of nominal frequency $\nu \sim \omega_0$ with an overall envelope $V(t)$,

$$U(t) = V(t) \int_{-\infty}^{\infty} \delta(\omega-\nu)\exp(-i\omega t)d\omega \ . \tag{2}$$

The envelope $V(t)$, which is the AM component of the pulse, has a nominal duration τ and it vanishes as $t \rightarrow \pm\infty$. The spectral function $\delta(\omega-\nu)$ specifies the frequency content of $U(t)$, beyond that contained in $V(t)$; we assume that $\delta(\omega-\nu)$ is peaked at $\omega \sim \nu \sim \omega_0$, and that it is normalized $[\int_{-\infty}^{\infty} \delta(\omega-\nu)d\omega = 1]$. If $\delta(\omega-\nu)$ were a δ function, then $U(t) = V(t)e^{-i\nu t}$ would be nominally monochromatic.

Upon substituting Eq. (2) into Eq. (1), it is easy to show that the absorptive transition amplitude is given by a convolution of Fourier transforms[4]

$$\boxed{ia(\Omega)/2\pi = \int_{-\infty}^{\infty} v(k)\delta(\Omega-k)dk, \quad \Omega = \omega_0 - \nu \ .} \tag{3}$$

$v(k)$ is the Fourier transform of the pulse envelope function $V(t)$, and Ω is the "detuning frequency." The absorption line shape may be plotted as $|a(\Omega)|^2$ versus Ω; normally, this plot shows a strong resonance at $\Omega = 0$.

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