

### Absorption spectra of strongly driven two-level atoms

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An analysis of the well-known absorption spectrum of a strongly driven two-level atom, in terms of the response of the individual *mode* of the atomic system, is given. It is shown how the response of a given mode leads immediately to the dispersionlike behavior of the absorption spectrum for frequencies in the neighborhood of the Rabi frequency.

The absorption spectrum of a strongly driven two-level atom is known to have several anomalous features.<sup>1-4</sup> For example (see Fig. 1), one finds that there are regions of frequency in which absorption is negative, i.e., instead of absorbing energy from the probe field, the atom emits into it. This can also be viewed as a parametric process,<sup>3</sup> in which the incident signal is amplified. Another important feature of the spectrum is that for frequencies close to the Rabi frequency, the spectrum behaves like the real part of the dielectric constant in the usual dispersion theory<sup>5</sup> (Fig. 2). This feature is quite anomalous as normally one would think that the absorption spectrum

should behave more like the imaginary part of the dielectric constant (Fig. 2), which is indeed the case for low driving fields (Fig. 1).

In this paper, we examine the response of the individual "mode" of the atomic system and compare this response with the response of an oscillator to a driving field. A comparison of the two response functions shows why the absorption spectrum in the vicinity of the Rabi frequency shows the dispersionlike behavior.

The absorption spectrum of a strongly driven two-level atom with energy separation  $\omega_0$  is given by (cf. Refs. 2, 3, and 6)

$$S_A(\omega) \propto \text{Re } f(z) \Big|_{z=i(\omega-\omega_L)}, \tag{1}$$

$$f(z) = [(z + T_2^{-1} - i\Delta)(z + T_2^{-1} + i\Delta)(z + T_1^{-1}) + 4\alpha^2(z + T_2^{-1})]^{-1} \\ \times [2\alpha^2 + (z + T_2^{-1} + i\Delta)(z + T_1^{-1}) - 2\alpha^2(i\Delta + T_2^{-1})^{-1}(z + T_2^{-1} + i\Delta)], \quad \Delta = \omega_0 - \omega_L \tag{2}$$

where  $2\alpha$  is the Rabi frequency and other symbols have standard meanings. Figure 1 shows the behavior of  $S_A(\omega)$  for a weakly driven ( $\alpha = \frac{1}{10}\gamma$ ) and a strongly driven ( $\alpha = 10\gamma$ ) two-level atom for the radiative relaxation of the atom ( $T_1^{-1} = 2\gamma = 2/T_2$ ,  $W^{(0)} = -\frac{1}{2}$ ), and for the case when the driving field is at resonance ( $\Delta = 0$ ). In order to understand the behavior of  $S_A$  in the vicinity of  $2\alpha$ , we examine the response equations for an oscillator for applied frequencies close to the resonance frequency, and response at  $\omega$ :

$$\ddot{x} + 2\gamma\dot{x} + \omega_0^2 x = \eta \epsilon(t), \quad \epsilon(t) = \epsilon_0 e^{-i\omega t}, \tag{3}$$

and hence

$$x(\omega) = \eta \mathcal{E}_0 (\omega_0^2 - \omega^2 - 2i\omega\gamma)^{-1} \\ \approx \eta \mathcal{E}_0 (-2i\omega_0)^{-1} [\gamma + i(\omega_0 - \omega)]^{-1} \\ = \eta \mathcal{E}_0 (2\omega_0)^{-1} \chi(\omega_0 - \omega) \text{ for } \omega \sim \omega_0. \tag{4}$$

Therefore for  $\omega \sim \omega_0$ , Eq. (3) can be approximately reduced to

$$\dot{x} + \gamma x + i\omega_0 x \approx \eta \mathcal{E}(t) (-2i\omega_0)^{-1}. \tag{5}$$

The well-known behavior of the real and imaginary parts of  $\chi(\omega)$  is shown in Fig. 2. It is clear that

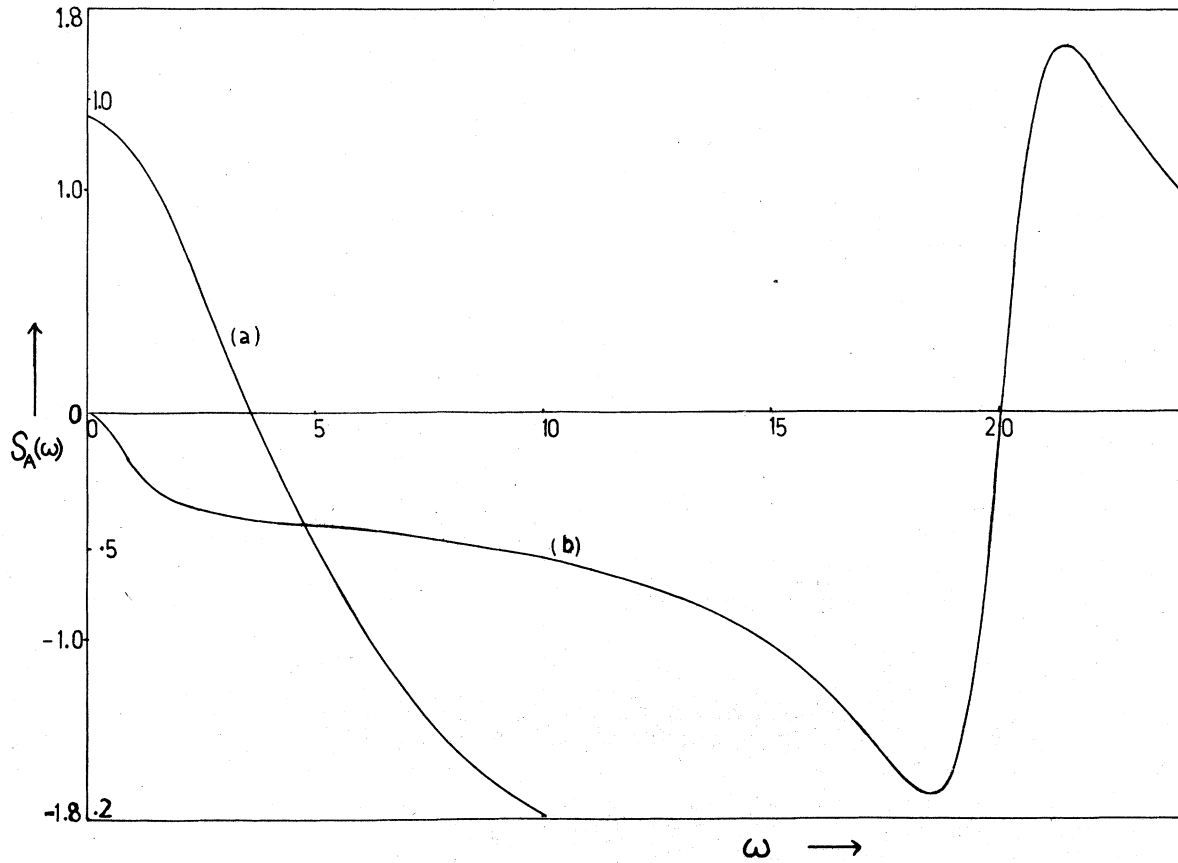


FIG. 1. Absorption spectra  $S_A(\omega)$  as a function of frequency, in units of  $\gamma$ , for the radiative relaxation of the atom and for (a)  $\alpha = \frac{1}{10}\gamma$ , (b)  $\alpha = 10\gamma$ .

the real part of  $\kappa(\omega)$  [solution of Eq. (5)] has the dispersionlike behavior, whereas the imaginary part has absorptionlike behavior. The form of the coefficient multiplying  $\mathcal{E}$  is crucial for the analysis of the response equations for the strongly driven two-level atom. The equations giving the linear response of a two-level atom are

$$\dot{\psi} = (A_0 + A_1)\psi + D, \quad A_0 = -\frac{1}{T_2} \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \frac{T_2}{T_1} \end{bmatrix}, \quad (6)$$

$$A_1 = i\alpha \begin{bmatrix} 0 & 0 & 2 \\ 0 & 0 & -2 \\ 1 & -1 & 0 \end{bmatrix},$$

$$D = 2i\alpha_1 \begin{bmatrix} \psi_3^{(0)} e^{i\Omega t} \\ -\psi_3^{(0)} e^{-i\Omega t} \\ \frac{1}{2}(\psi_1^{(0)} e^{-i\Omega t} - \text{c.c.}) \end{bmatrix},$$

where  $\psi$  represents the linear response of the variables  $\langle S^+ \rangle$ ,  $\langle S^- \rangle$ :

$$\psi_1 = \langle S^+ \rangle - \psi_1^{(0)}, \quad \psi_2 = \langle S^- \rangle - \psi_2^{(0)}, \quad \psi_3 = \langle S^e \rangle - \psi_3^{(0)}, \quad (7)$$

with  $\psi_i^{(0)}$  denoting the equilibrium values of the variables  $\langle S^+ \rangle$ ,  $\langle S^- \rangle$ , and  $\langle S^e \rangle$  and  $\alpha_1$  denoting the coupling with the weak probe field. When  $\alpha = 0$ , then the driving term in the equation for  $\psi$ , is purely imaginary, since  $\psi_3^{(0)}$  is real, and hence the usual situation prevails, i.e., the expected absorption spectrum is obtained. When  $\alpha$  is small but not zero, a straightforward second-order perturbation theory with respect to  $\alpha$  shows that

$$\dot{\psi}_1 = -T_2^{-1}\psi_1 + i2\alpha_1\psi_3^{(0)}e^{i\Omega t} + 2i\alpha\psi_3', \quad (8)$$

where  $\psi_3'(t)$  is the linear response to first order in  $\alpha$ , which is now known and is real by definition. The above equation shows that the driving term is purely imaginary, and hence the expected behavior of the absorption spectrum is obtained. For large values of  $\alpha$ , we have to use a nonper-

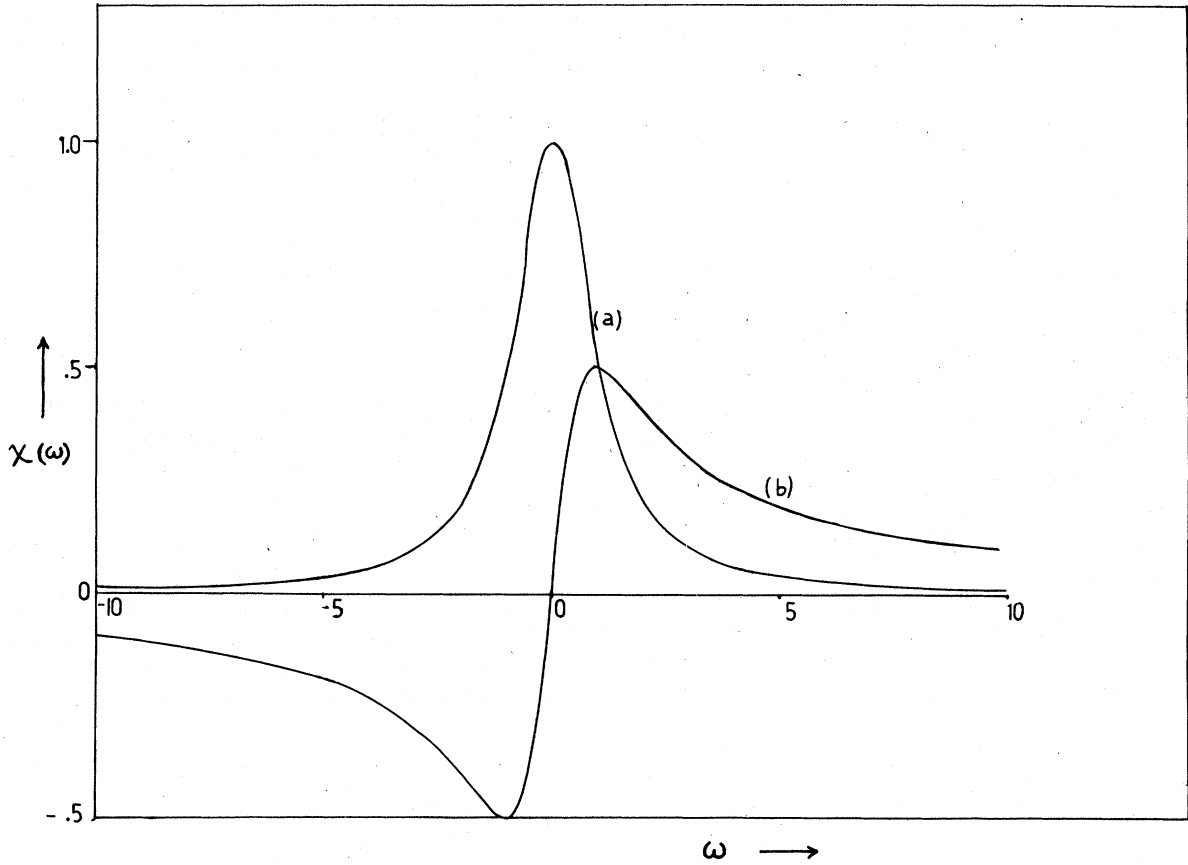


FIG. 2. (a) Imaginary and (b) real parts of the susceptibility  $\chi(\omega)$  [Eq. (5)] as a function of the frequency.

turbative method. For this purpose, we work in a representation in which  $A_1$  is diagonal. Let  $S$  be the matrix that diagonalizes  $A_1$ , i.e.,

$$S^{-1}A_1S = \Lambda, \quad \Lambda = 2i\alpha \begin{bmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{bmatrix}, \tag{9}$$

$$S = \frac{1}{\sqrt{3}} \begin{bmatrix} \sqrt{\frac{3}{2}} & 1 & -1 \\ \sqrt{\frac{3}{2}} & -1 & 1 \\ 0 & 1 & 1 \end{bmatrix}, \quad S^{-1} = \frac{\sqrt{3}}{4} \begin{bmatrix} 2\sqrt{\frac{2}{3}} & 2\sqrt{\frac{2}{3}} & 0 \\ 1 & -1 & 2 \\ -1 & 1 & 2 \end{bmatrix}.$$

Then the matrix  $S^{-1}\psi \equiv \Phi$  satisfies the equation

$$\dot{\Phi} = (B_0 + B_1 + \Lambda)\Phi + \mathcal{D}, \tag{10}$$

where

$$B_0 = -\frac{1}{2} \left( \frac{1}{T_1} + \frac{1}{T_2} \right) \begin{bmatrix} \frac{2}{T_2} \left( \frac{1}{T_1} + \frac{1}{T_2} \right)^{-1} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix},$$

$$B_1 = \frac{1}{2} \left( \frac{1}{T_1} - \frac{1}{T_2} \right) \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix},$$

$$\mathcal{D}_1 = -4\sqrt{2} \alpha_1 \psi_3^{(0)} \sin \Omega t,$$

$$\mathcal{D}_2 = \mathcal{D}_3^* = 2\sqrt{3} i \alpha_1 (\psi_3^{(0)} \cos \Omega t + \frac{1}{2} \psi_1^{(0)} e^{-i\Omega t} - \frac{1}{2} \psi_2^{(0)} e^{i\Omega t}). \tag{11}$$

The column matrix  $\mathcal{D}$ , on using the form of  $\psi_i^{(0)}$ , reduces to

$$\mathcal{D} = \begin{bmatrix} -4\sqrt{2} \sin \Omega t \alpha_1 \psi_3^{(0)} \\ \sqrt{3} \alpha_1 \cos \Omega t \left[ 2i \psi_3^{(0)} \left( 1 - \frac{i}{2T_1 \alpha} \right) - \frac{W^{(0)}}{T_1 \alpha} \right] \\ \sqrt{3} \alpha_1 \cos \Omega t \left[ -2i \psi_3^{(0)} \left( 1 + \frac{i}{2T_1 \alpha} \right) - \frac{W^{(0)}}{T_1 \alpha} \right] \end{bmatrix}. \quad (12)$$

Equation (10) gives the response of each "mode" with frequencies 0,  $-2\alpha$ ,  $2\alpha$ . In this equation the terms involving  $B_1$  are counter-rotating terms as  $\alpha$  is large, and therefore, for strong driving fields the coupling of  $\Phi_1$  to  $\Phi_2$  etc. can be ignored, as one is justified in making the rotating wave approximation.<sup>7</sup> Moreover, for strong fields  $\psi_3^{(0)} \rightarrow 0$ , and therefore,

$$\mathcal{D} \approx \frac{-\sqrt{3} \alpha_1 W^{(0)}}{T_1 \alpha} \cos \Omega t \begin{bmatrix} 0 \\ 1 \\ 1 \end{bmatrix}. \quad (13)$$

The response of  $\langle S^+ \rangle$  near  $\omega - \omega_L \approx 2\alpha$  will be  $\langle S^+ \rangle$

$\approx -3^{-1/2} \Phi_3$  as  $\alpha$  is large and hence other components  $\Phi_1$ ,  $\Phi_2$  will contribute negligibly small for  $\omega - \omega_L \approx 2\alpha$ . Since the absorption spectrum is proportional to the imaginary part of the response function and hence near  $\omega - \omega_L \approx 2\alpha$ , the absorption spectrum will essentially be given by the imaginary part of the response of  $\Phi_3$ . The equation of motion for  $\Phi_3$  from Eqs. (10)–(13) is

$$\begin{aligned} \dot{\Phi}_3 = & - \left[ 2i\alpha + \frac{1}{2} \left( \frac{1}{T_1} + \frac{1}{T_2} \right) \right] \Phi_3 \\ & - \frac{\sqrt{3} \alpha_1}{\alpha T_1} W^{(0)} \cos \Omega t. \end{aligned} \quad (14)$$

It is important to note that the driving term in (14) is real, i.e., *the effective field driving the mode is now out of phase by a factor of  $\frac{1}{2}\pi$  as compared to the field in Eq. (5)*, and this is the crucial point. We have seen earlier that the driving term in Eq. (5) was purely imaginary. It is therefore clear that the imaginary part of  $\Phi_3$  will behave like the real part of  $x$ . This explains why the absorption spectrum of a strongly driven two-level atom shows the dispersionlike behavior for frequencies close to the Rabi frequency.

<sup>1</sup>V. M. Fain and Ya-I. Khanin, *Quantum Electronics* (MIT, Cambridge, Mass., 1969), Sec. 23.

<sup>2</sup>B. R. Mollow, *Phys. Rev. A* **5**, 2217 (1972).

<sup>3</sup>S. Haroche and F. Hartmann, *Phys. Rev. A* **6**, 1280 (1972); S. L. McCall, *ibid.* **9**, 1515 (1974); B. R. Mollow, *ibid.* **7**, 1319 (1973).

<sup>4</sup>F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, *Phys. Rev. Lett.* **38**, 1077 (1977).

<sup>5</sup>Cf. M. Born and E. Wolf, *Principles of Optics* (Per-

gamon, London, 1970), p. 93.

<sup>6</sup>G. S. Agarwal, *Phys. Rev. A* **18**, 1490 (1978).

<sup>7</sup>The author is grateful to Professor Cohen-Tannoudji, who has used this in the calculation of the characteristics of the resonance fluorescence spectrum of a multilevel atom [C. Cohen-Tannoudji and S. Reynaud, *J. Phys. B* **10**, 345 (1977)], for pointing out this approximation.