

Line narrowing beyond natural linewidth in radiation matter interaction*

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Abstract. A review of the various linear and nonlinear methods used to obtain resolution beyond natural linewidth is given.

Keywords. Line narrowing; linewidth; radiation interaction; nonlinear spectroscopy; saturation phenomena.

1. Introduction

The observed width of an atomic/molecular transition is usually very different from the natural width because many sources of broadening contribute to such a width. These sources of broadening include the broadening due to (i) thermal motion of the atoms (ii) collisions among atoms and (iii) the temporal fluctuations of the exciting source used to prepare the system in excited states. Special techniques have been developed to eliminate the broadening effects due to these mechanisms. The resolution is still limited by the natural linewidth. The question arises how to resolve two lines lying within the natural linewidth of each other? In this article we review the various techniques which have been developed to achieve line narrowing beyond the natural linewidth. We will show that the resolution beyond natural linewidth is indeed possible.

Consider a quantum mechanical system with energy levels $|i\rangle$ having eigenvalues E_i . The system is assumed to be radiating in presence of external driving fields. The interaction Hamiltonian can be written in dipole approximation as

$$H = -\mathbf{d} \cdot \mathbf{E}_{\text{ext}}(t) - \mathbf{d} \cdot \mathbf{E}_{\text{vac}} \equiv H_{\text{ext}} + H_{\text{vac}}. \quad (1)$$

Here \mathbf{E}_{ext} is the external field which is used to prepare the system in the excited state. This field is in general time-dependent. The interaction with the vacuum of the radiation represented by the second term in (1) involves all the modes of the vacuum. The vacuum degrees of freedom can be eliminated in the standard fashion (Agarwal 1974). One finds that the dynamics of the atomic molecular system is given by the master equation for the density matrix

$$\begin{aligned} \frac{d}{dt} \rho_{ij} = & -i\omega_{ij} \rho_{ij} - i[H_{\text{ext}}, \rho]_{ij} - \Gamma_{ij} \rho_{ij} (1 - \delta_{ij}) \\ & - \sum_k \delta_{ij} (\gamma_{ki} \rho_{ii} - \gamma_{ik} \rho_{kk}), \end{aligned} \quad (2)$$

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where

$$\begin{aligned} \gamma_{ij} &= \frac{2}{3} \frac{\omega_{ji}^3}{c^3} |d_{ij}|^2, & E_j > E_i \\ &= 0, & E_j < E_i \end{aligned} \quad (3)$$

$$\Gamma_{ij} = \sum_k (\gamma_{ki} + \gamma_{kj}). \quad (4)$$

Thus $2\gamma_{ij}$ gives the transition rate for making a transition from the state $|j\rangle$ to $|i\rangle$ with the emission of a photon. Various Lamb shifts are already accounted for in the frequencies ω_{ij} . The atomic dynamics is completely specified. If H_{ext} is set zero, then (2) leads to the standard results on the line shapes and linewidths.

The question now arises as to how to design the experiments using a suitable initial preparation of the system or by making use of the nonlinearity of the radiation matter interaction so that the resulting line shapes are narrower than the natural linewidths. It is important to remember that the basic dynamical equation (2) contains the standard damping and shift parameters. Thus the basic interaction responsible for spontaneous emission is the usual one. It is only that the experimentalist does not measure the conventional shapes in properly designed experiments and thereby he improves his resolution.

2. Transient methods of line narrowing

In this section we discuss the delayed fluorescence methods (Dodd and Series 1978; Deech *et al* 1974; Figger and Walther 1974; Schenck *et al* 1973) used in the transient domain to obtain resolution beyond natural linewidth. In these methods one makes use of the appropriate bias function to collect and to analyse the data. Suppose the signal to be measured has the form

$$I(t) = (A + B \cos 2\delta t) \exp(-\Gamma t). \quad (5)$$

Such a signal is produced typically by a radiating system in a coherent superposition of two states separated by 2δ . The two excited states may be Zeeman states and thus 2δ can be changed by varying the magnetic field. The cosine Fourier transform of (5) has the usual form

$$\begin{aligned} I(\omega) &= \int_0^\infty dt \cos \omega t I(t) \\ &= \frac{A\Gamma}{\Gamma^2 + \omega^2} + \frac{B\Gamma}{2(\Gamma^2 + (\omega - 2\delta)^2)} + \frac{B\Gamma}{2(\Gamma^2 + (\omega + 2\delta)^2)} \end{aligned} \quad (6)$$

i.e. different components have half width Γ . Series and coworkers suggested that line narrowing can be obtained by introducing the bias function $f(t)$

$$S(\omega) = \int_0^\infty dt \cos \omega t f(t) I(t). \quad (7)$$

The simplest bias function $f(t)$ is $\theta(t - T)$ where T is the waiting time. In such a case $S(0)$ is

$$S(0) = \exp(-\Gamma T) \left\{ \frac{A}{\Gamma} + \frac{B}{\Gamma^2 + 4\delta^2} (\Gamma \cos 2\delta T - \omega_0 \sin 2\delta T) \right\}. \quad (8)$$

The signal $S(0)$ as a function of δ shows line narrowing which becomes narrower and narrower as T increases. The line narrowing occurs at the cost of considerable loss of the signal (due to the factor $\exp(-\Gamma T)$). Moreover the bias function also introduces the oscillations of the signal in the wings (Schenck *et al* 1973). These oscillations also become more pronounced as T increases. It has been suggested that some of these problems can be avoided by using a Gaussian bias function

$$f(t) = \exp[-(t-a)^2/b^2]. \quad (9)$$

whose width and position are related by

$$b^2 = 2a/\Gamma. \quad (10)$$

The signal now is

$$S(\omega) = \frac{1}{2} b(\pi)^{1/2} \exp\left(-\frac{\Gamma^2 b^2}{4}\right) \left\{ A \exp\left(-\frac{\omega^2 b^2}{4}\right) + \frac{1}{2} B \left[\exp\left(-\frac{b^2}{4}(\omega-2\delta)^2\right) + \exp\left(-\frac{b^2}{4}(\omega+2\delta)^2\right) \right] \right\}, \quad (11)$$

and hence the line narrowing will occur provided we choose

$$b > 2(\ln 2)^{1/2}/\Gamma. \quad (12)$$

The delayed fluorescence signals (8) have been investigated for the case of a number of atomic transitions in Na, Ba, etc. Walther and coworkers were able to see lines as narrow as one sixth of the natural linewidth (Figger and Walther 1974).

An interesting situation arises if both the levels $|1\rangle$ and $|2\rangle$, separated by ω_0 , involved in optical transition are decaying to some other states at the rates $2\gamma_1$ and $2\gamma_2$. Assume that the system is continuously pumped from the state $|2\rangle$ to the excited state $|1\rangle$ by an external monochromatic source of frequency ω_1 . The fluorescence signal to the lowest order in pump intensity will now be proportional to

$$S(0) = \left[\frac{\exp(-2\gamma_2 T)}{2\gamma_2} + \frac{\exp(-2\gamma_1 T)}{2\gamma_1} + \frac{2 \exp(-\gamma_{12} T)}{(\Delta^2 + \gamma_{12}^2)} \times (\Delta \sin \Delta T - \gamma_{12} \cos \Delta T) \right] \frac{1}{(\Delta^2 + (\gamma_1 - \gamma_2)^2)}; \quad (13)$$

$$\gamma_{12} = (\gamma_1 + \gamma_2); \Delta = \omega_0 - \omega_1,$$

In the limit $T \rightarrow 0$, one has

$$S(0) = (\Delta^2 + \gamma_{12}^2)^{-1}. \quad (14)$$

For large T , the width is $(\gamma_1 - \gamma_2)$ i.e. the line has a width which is the difference of the two rates of decay rather than the sum. Thus much narrower lines can be produced (at the cost of the loss of signal) if the two states decay at roughly equal rate (Meystre *et al* 1980; Lee *et al* 1981).

A further variation of the delayed fluorescence technique (Knight and Coleman 1980; Knight 1981) is to tailor the pulses so that the field envelop decays with time at the rate γ_L i.e. $\varepsilon(t) = \exp(-\gamma_L t) \varepsilon_0$. To lowest order in ε_0^2 the instantaneous fluorescence is

$$I(t) = \frac{1}{\Delta^2 + (\gamma_1 - \gamma_L)^2} \{ \exp(-2\gamma_L t) - 2 \cos \Delta t \exp[-(\gamma_1 + \gamma_L)t] + \exp(-2\gamma_1 t) \}. \quad (15)$$

Using (15) the delayed fluorescence signal is found to have a half width in the range $(\gamma_1 - \gamma_L) \leftrightarrow (\gamma_1 + \gamma_L)$. Thus for large values of the waiting time T , considerable line narrowing may result. Generalization of these methods to two-photon transitions is possible (Coleman *et al* 1981).

Another very interesting method has been suggested by Shimizu *et al* (1983). Their method switches the phase of the driving field by electronic means. Let us assume a driving field of the form

$$\begin{aligned} E(t) &= \varepsilon_0 \exp(-i\phi - i\omega t) + \text{c.c.} & t < 0 \\ &= \varepsilon_0 \exp(-i\omega t) + \text{c.c.} & t > 0. \end{aligned} \quad (16)$$

Defining the fluorescence signal in terms of the excited state population by

$$S_\phi(t) = \int_{-\infty}^t dt_0 \rho_{11}(t-t_0), \quad (17)$$

one can show that

$$\begin{aligned} F(t) &= S_\phi + S_{-\phi} - 2S_0 \\ &= \frac{|\varepsilon|^2 (\cos \phi - 1)}{(\Delta^2 + \gamma_{12}^2) [\Delta^2 + (2\gamma_1 - \gamma_{12})^2]} \{(\Delta^2 - \gamma_{12}^2 + 2\gamma_1\gamma_{12}) \\ &\quad \times [\exp(-\gamma_{12}t) \cos \Delta t - \exp(-2\gamma_1t)] - 2\Delta(\gamma_1 - \gamma_{12}) \\ &\quad \times \exp(-\gamma_{12}t) \sin \Delta t\}. \end{aligned} \quad (18)$$

The transient fluorescence $F(t)$ shows line narrowing if $2\gamma_1 > \gamma_{12}$. For Na D line $\gamma_1 = \gamma_{12}$ and hence using the present technique, one can expect narrower lines. The signal $F(t)$ again shows considerable oscillations which can be reduced by taking a weighted average. Using this method Shimizu *et al* (1983) have seen lines whose widths are about 40% of the natural linewidth in experiments on Na D lines. It should be noted that in this method the signal $F(t)$ is analysed rather than $S_\phi(t)$. This is because $F(t)$ yields symmetric line shapes.

3. CW methods based on nonlinear spectroscopy

We now discuss the methods based on the nonlinear interaction between the external radiation field and matter. The external fields are of cw type and the observed line shapes are related to the steady state nonlinear response of the medium. These cw methods were developed by Saxena and Agarwal (1982, 1985).

3.1 CW method based on the modulated fluorescence

Consider a quantum-mechanical system irradiated with an amplitude-modulated external field i.e. the field amplitude has the form

$$\varepsilon(t) = \varepsilon_0 \left(1 + M \sin \frac{\Omega}{2} t\right). \quad (19)$$

The fluorescence in the steady state is proportional to the excited state population $\rho_{11}(t)$. Let us assume that the experiment is performed on an atomic beam with orthogonal directions for the excitation and detection. In steady state $\rho_{11}(t)$ will have many modulated components. The modulated fluorescence to second order in external fields turns out to have the form ($|\alpha| = |\mathbf{d} \cdot \boldsymbol{\varepsilon}_0|$)

$$I^{(2)} = \frac{\alpha^2}{(\gamma^2 + \Delta^2)} \left(1 + \frac{M^2}{2}\right) - \frac{\alpha^2 M^2}{2(\gamma^2 + \Delta^2)} \cos \Omega t, \quad (20)$$

where the modulation frequency Ω has been assumed to be much smaller than Δ, γ ($\Omega \sim \text{kHz}, \gamma \sim \text{MHz}$). This signal has the usual width. The modulated signal to fourth order in external field turns out to have the structure (Saxena and Agarwal 1982)

$$I^{(4)} \approx -\frac{\alpha^4 M^4}{4} \cos 2\Omega t \frac{1}{(\gamma^2 + \Delta^2)^2}. \quad (21)$$

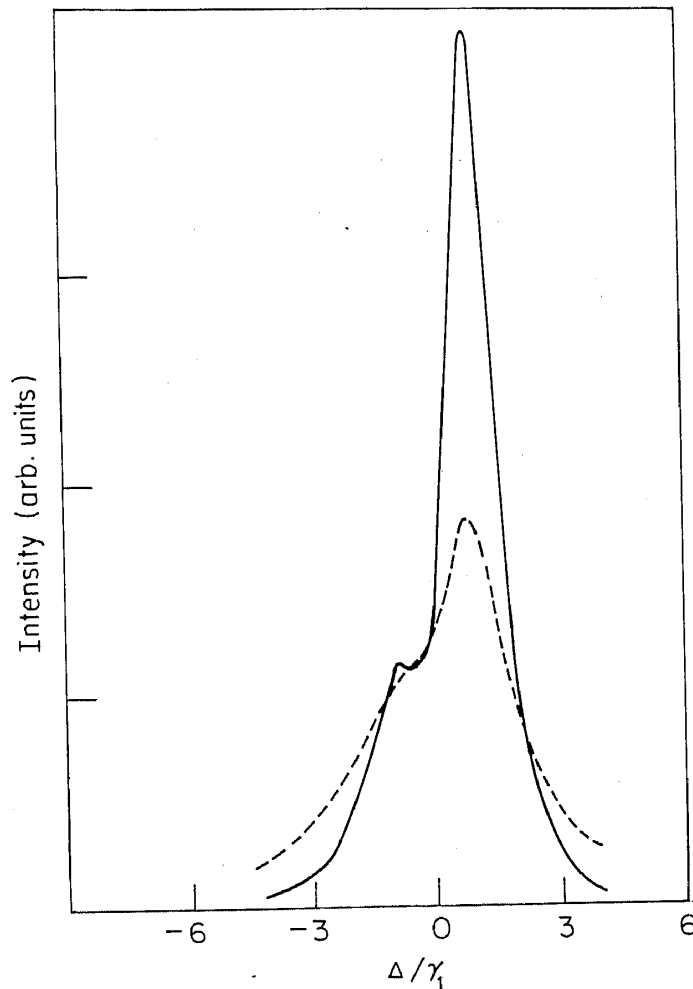


Figure 1. Resolution of two lines lying within the natural linewidth of each other. Modulated fluorescence, at 2Ω (Solid curve) and at Ω (dashed curve), is shown as a function of $\Delta/\gamma_1 = (\omega_0 - \omega_1)/\gamma_1$, and for other parameters equal to $\gamma_1 = 1, \gamma_2 = 1.5, 2\delta = 1.6$ (after Saxena and Agarwal 1982).

Thus the phase-sensitive detection of the signal at 2Ω will produce lines which are subradiative. The full width at half maximum is 1.3γ which is considerably less than the natural linewidth 2γ . Thus the nonlinear response yields line shapes with subnatural linewidths. This method is reminiscent of the method used by Sorem and Schalow (1972) to obtain Doppler-free signals.

It is clear from the above that this method may be used to resolve lines lying within the natural width of each other. For this purpose we examined the modulated fluorescence at 2Ω produced by two overlapping transitions $|1\rangle \rightarrow |3\rangle$, $|2\rangle \rightarrow |3\rangle$ with half widths γ_1 and γ_2 and with energy separation 2δ . The external field is detuned by an amount Δ from the centre of the two energy levels $|1\rangle$ and $|2\rangle$. The complete mathematical expression for the signal is given in the original paper (Saxena and Agarwal 1982). Figure 1 clearly shows that two lines lying within the width of each other can indeed be resolved by the present method.

3.2 Line narrowing in four-wave mixing

Nonlinear response can produce narrower signals as seen above in the context of modulated fluorescence. Here we consider the structure of four-wave mixing signals and demonstrate the possibility of subradiative structures under certain conditions (Saxena and Agarwal 1985). It is well known that the coherent four-wave mixing signal $I(2\omega - \omega_s)$, in the direction $2\mathbf{k} - \mathbf{k}_s$, produced by the action of two fields at ω and ω_s and with wave vectors, \mathbf{k} , \mathbf{k}_s is related to the third order susceptibility $\chi^{(3)}(\omega, \omega, -\omega_s)$ (Bloembergen *et al* 1978). The structure of $\chi^{(3)}$ depends on the energy levels involved in the optical transition and on the relaxation parameters of the system. Consider $j = 0$ to $j = 1$ transition in an atomic system in a magnetic field. Let 2δ be the energy separation between two Zeeman states $|1, 1\rangle$ and $|1, -1\rangle$ and let ω_0 be the transition frequency in the absence of the magnetic field. Let the system be irradiated with two circularly polarized fields of frequency ω travelling in opposite directions and oppositely polarized. The beam at ω_s is right hand circularly-polarized. The general form of $\chi^{(3)}$ and hence four-wave mixing signal can be calculated for all values of control parameters—magnetic field, frequencies ω and ω_s . The four-wave mixing signal can be scanned as a function of either of these parameters. In the special cases, one discovers lines with subnatural linewidth γ_s (full width at half maximum) as shown by the following examples:

$$(i) \text{ Pump-probe detuning scan, } \omega \neq \omega_s, \omega = \omega_0 \\ I(2\omega - \omega_s) \sim 1/[\gamma^2 + (\delta + \omega - \omega_s)^2]^2, \gamma_s \equiv 1.3\gamma, \quad (22)$$

$$(ii) \text{ Pump-atom detuning scan, } \omega \neq \omega_0, \omega = \omega_s, \delta = 0, \\ I(2\omega - \omega_s) \sim 1/[\gamma^2 + (\omega - \omega_0)^2]^3, \gamma_s = \gamma, \quad (23)$$

$$(iii) \text{ Magnetic field scan, } \omega = \omega_s = \omega_0, \delta \neq 0 \\ I(2\omega - \omega_s) \sim 1/[\gamma^2 + \delta^2]^4, \gamma_s \approx 0.87\gamma. \quad (24)$$

Note that in order to see such signals, experiments are to be performed on high density atomic beams, so that Doppler broadening is negligible. Note also that in the degenerate case $\omega = \omega_s$ [equations (23) and (24)], the susceptibility $\chi^{(3)}(\omega, \omega, -\omega)$ leads to narrower four-wave mixing signals. Thus narrower signals can be obtained whenever the result of the experiment is related to the degenerate susceptibility $\chi^{(3)}(\omega, \omega, -\omega)$. For example the narrowing resulting from (21) can be understood as

follows: In modulated fields, the system produces coherent radiation at $\omega + (3\Omega/2)$ through the susceptibility

$$\chi^{(3)}\left(\omega + \frac{\Omega}{2}, \omega + \frac{\Omega}{2}, -\omega + \frac{\Omega}{2}\right).$$

This coherent radiation can beat with the component at $\omega + \frac{1}{2}\Omega$ of the incident radiation to produce modulated fluorescence at 2Ω . Thus the modulated fluorescence is related to

$$\chi^{(3)}\left(\omega + \frac{\Omega}{2}, \omega + \frac{\Omega}{2}, -\omega + \frac{\Omega}{2}\right)$$

which in the limit of small Ω reduces to the third order susceptibility for the degenerate case.

4. Line narrowing using transient four-wave mixing methods

In the last section, we have seen how cw methods can lead to nonlinear response resulting in subradiative line shapes. Recently Zinth *et al* (1984) combined the delayed fluorescence technique with the nonlinear response of the system to produce much narrower Raman lines than the usual Raman lines with width $1/T_2$. More specifically they observed line narrowing using transient CARS. The input pump field ε_1 at ω_1 and the Stokes field ε_s at ω_s drive a coherent molecular vibration Q at $\omega_1 - \omega_s$ which is close to the molecular frequency ω_0

$$\ddot{Q} + \frac{2}{T_2} \dot{Q} + \omega_0^2 Q = \beta \varepsilon_1 \varepsilon_s^*, \quad t < t_0. \quad (25)$$

This coherent oscillation is probed by a third field ε_p at frequency ω_p . The probe field is delayed by T . The CARS signal at $\omega_0 + \omega_p \equiv \bar{\omega} \approx \omega_1 - \omega_s + \omega_p$ is analysed. The CARS signal I_c is related to $Q(t)$ by

$$I_c \sim |\varepsilon_p(t-T) Q(t) \exp(-i\omega_p t)|^2. \quad (26)$$

Assuming the probe field ε_p to be Gaussian

$$\varepsilon_p(t) \sim \exp\left\{-\frac{1}{2} \left(\frac{t-t_p}{t_p}\right)^2\right\}, \quad (27)$$

the CARS signal in the frequency domain turns out to have the form

$$I_c \sim \exp\left\{-\frac{(\omega - \bar{\omega})^2 t_p^2}{4 \ln 2}\right\} \exp\left\{\frac{(t_0^2/2 + t_p^2)}{4 \ln 2 T_2^2} - \frac{2T}{T_2}\right\}. \quad (28)$$

The width of the signal (28) is $2 \ln 2 / \pi t_p$ which interestingly enough depends on the width of the probe pulse. This results in the narrowing of the signal if $t_p > 1.4 T_2$ i.e. the width is less than $1/\pi T_2$. Zinth *et al* (1984) have resolved Raman lines in the CARS spectra of pyridine-methanol mixtures. Such lines were previously unresolved in spontaneous Raman spectra. This method is very closely related to the method of introducing a bias function as discussed in §2. For times $t > t_0$, $Q \approx Q(t_0) \exp(-i\omega_0 t - t/T_2)$.

The initial amplitude $Q(t_0)$ is determined from the solution of (25). If $t_0 \gg 1/T_2$, then $Q(t_0)$ will be the steady state response. The probe field ε_p acts like the bias function (9) with

$$a = T, b^2 = t_p^2/4 \ln 2, \Gamma = 2/T_2. \quad (29)$$

The condition $t_p > 1.4 T_2$ is just the narrowing condition (12).

5. Line narrowing and saturation phenomena

The last two sections discussed the nonlinear response and the physical results which were obtained using perturbative techniques. If the resonant fields become strong, then one has to account for the saturation effects. Recent calculations (Agarwal and Nayak 1984; Toptygina and Fradkin 1982; Agarwal *et al* 1984) have shown that narrowing can also result from the dynamical behaviour of a system in strong external fields. Such narrowing can have spectroscopic applications. Here we mention two examples.

Consider an optical transition which is simultaneously driven by two strong radiation fields at frequencies ω_1 and ω_2 . One can monitor the energy absorption from either of the two fields. Both the fields together dress the energy levels and these dressed levels will be reflected in the energy absorption measurements. Detailed calculations (Agarwal and Nayak 1984; Toptygina and Fradkin 1982) have shown the presence of a number of absorption peaks which are located at $\pm (g_1^2 + g_2^2)^{1/2}/n$ for the case $T_1 = T_2$ and $\omega_0 = \omega_2$. Here g_i 's denote the Rabi frequencies. The width of these absorption peaks is of the order $1/n T_2$. Thus in the simultaneous irradiation by two strong fields much narrower lines can result.

Another very interesting line narrowing arises in the context of laser-induced autoionization (Agarwal *et al* 1984 and references therein). Here one of the weakly bound states $|a\rangle$ lying in the continuum can ionize at the rate Γ due to configuration mixing. The state can also radiatively decay at the rate 2γ to one of the lower lying states say $|i\rangle$. We assume that the atom is driven by a strong laser field which transfers population to the autoionizing state. Thus the following processes are possible (i) laser-induced ionization from the initial state (ii) coherent oscillations induced by the laser field between the initial state $|i\rangle$ and the autoionizing state $|a\rangle$ (iii) autoionization of the state $|a\rangle$ (iv) radiative decay of $|a\rangle$ (v) autoionization of state $|a\rangle$ followed by the radiative recombination. The process (iv) and (v) can then be followed by laser excitation to the state $|a\rangle$. The mathematical formulation of this problem and many physical results are discussed in Agarwal *et al* (1984). The line shapes will have a width which will be determined by both radiative and autoionization widths. For example for weak incident fields, the ionization signals have a linewidth $\Gamma(\Gamma + \gamma)/[\Gamma + (\gamma/q^2)]$. Here q is called Fano's asymmetry parameter and depends on the configuration mixing. The situation changes drastically if the laser field driving the system is strong so that one has to calculate ionization signals to all orders in the external field strength. It turns out that the ionization signals show a doublet structure, the widths and locations of which are determined by the parameters like detuning $\alpha \equiv (2/\Gamma)(\omega_i - E_a)$, field strength

$$\Omega = \frac{2\pi}{\Gamma} |\langle E | H_{\text{ext}}^d | i \rangle|^2$$

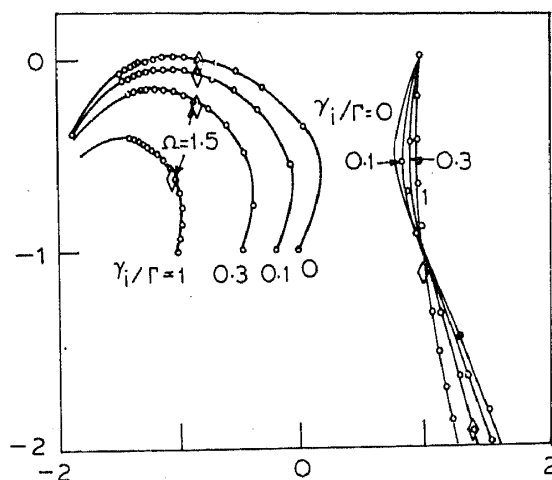


Figure 2. The complex zeros that determine the position and the width of the doublet in ionization signals as a function of laser strength Ω in the range 0 and 10. All parameters are in units of autoionization width. Other parameters are $\alpha = 1$, $q = 1$, $\gamma_1 = 0, 0.1, 0.3$ and 1.0 . The dots indicate the increments of 0.3 in Ω . Curves on the left show considerable narrowing as Ω is changed (after Agarwal *et al* 1984).

and Fano asymmetry parameter q . In figure 2 we show these peak positions and widths as the complex zeros with the real part giving the width and the imaginary part the peak position. An examination of these zeros shows that for $\Omega \simeq (1 + \alpha/q)$ one of the doublets will be extremely narrow i.e. its linewidth will be much smaller than $(\Gamma + \gamma)$. Thus this provides us with another example where considerable line narrowing results as a result of the saturation effects and the interference effects between the various channels. This certainly can lead to the resolution of two autoionizing lines lying within the width of each other.

Recently Devoe and Brewer (1983) have seen other manifestations of the line narrowing due to the interplay between the saturation effects and the finite correlation times associated with the relaxation mechanisms in certain systems. In such situations light scattering spectra have also been shown (Hanamura 1983; Agarwal 1985) to have extremely narrow structures.

We have thus shown how specially designed nonlinear spectroscopic methods can lead to much narrower lines and thus can yield resolution which is beyond the natural linewidth. Finally we mention that subnatural resolution has also been achieved by using the Ramsay fringe technique, the details of which can be found in Bergquist *et al* (1977) and Salour and Cohen-Tannoudji (1977).

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