predict a linear dependence of the signal intensity on each of the three pulses of Fig. 1. Particularly when studying dilute guests in a matrix of other species, it is important to make use of intense pulses and enhancement by multiple resonances. This optimizes signal-to-noise ratio and discriminates against interfering signals. It is precisely for these conditions that perturbative calculations are inapplicable.

We have observed nonlinearities in the response to both preparation and probing pulses under multiple resonance conditions where \( \omega_3 \) is near both the \( 0-0 \) transition and a \( v-v' \) transition of the guest pentacene molecule. A doubly rotating frame formalism has been developed for treating the time evolution of the system to all orders in the applied fields. Various limiting cases for the dependence of the signal on the pulse parameters have been treated analytically.

Comparison with experiment allows an estimate of the relevant Rabi frequencies and the optimum pulse parameters. In order to account for the observed intensity and spectral distribution around \( \omega_3 \), contributions to the signal from vibronic free induction decay after the probe pulse are included, along with the transient aspects of the scattering during the probe pulse.

The investigations were supported by the Netherlands Foundation for Chemical Research (S.O.N.) with financial aid from the Netherlands Organization for the Advancement of Pure Research (Z.W.O.). D.P.W. gratefully acknowledges a Z.W.O. postdoctoral fellowship.

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Coherent Transient Phenomena

Quantum Treatment of the Cooperative AC-Stark Effect

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PACS: 33

A recent paper by Boyd et al. [1] discuss the possibility of amplification of Rabi sidebands of the resonance fluorescence spectrum from a N two-level medium driven by a strong electric field. Using a classical four-wave parametric argument they show that a probe field with frequency near the Rabi-sidebands may withdraw energy from the pump field and may be amplified by propagation through the medium. This treatment is similar to the classical treatment of superfluorescence where a small coherent field is injected in the medium to simulate the quantum noise. In the quantum treatment presented here, only the strong driving field illuminates the medium. Then, the field operator at a given depth \( z \) is the sum of the incident field and of the atomic response resulting from the Maxwell equations with sources. In the direction of the incident field, the slowly varying amplitude operator for the Rabi frequency may be written [2] as

\[
\Omega(z, t) = \Omega_0 + \frac{i}{N\tau_R} \sum_{s} \bar{R}_s + \frac{i}{N\tau_R} \sum_{s} \delta R_s(z, \tau),
\]

where \( \tau \) is the retarded time, and \( \tau_R \) is the usual superradiant time \( (\tau_R = 4\pi N \mu^2 \omega_0 / c) \). In (1) we decompose the slowly varying part of the lowering atomic operator \( \bar{R}_s = \bar{R}_s + \delta \bar{R}_s \), into its equilibrium and fluctuating parts. We neglect the diffraction and the backward field emitted by the atoms.

The spectral properties of the field are studied by means of the correlation function of the fluctuating part \( \Delta \Omega(z, \tau) \) of the atomic response [last term in (1)]

\[
\Gamma_{\Delta \Omega}(z, \tau) = \langle \Delta \Omega(z, \tau) \Delta \Omega^*(z, 0) \rangle
\]

\[
= \frac{1}{N\tau_R^2} \sum_{s \neq t} \langle \delta \bar{R}_s(\tau) \delta \bar{R}_t(0) \rangle + \frac{1}{N\tau_R} \sum_{s} \langle \delta \bar{R}_s(\tau) \delta \bar{R}_s(0) \rangle.
\]

Fig. 1. Spectral density of the fluctuating atomic response at the cell exit, central peak \( (\omega - \omega_0) \) and one sideband \( (\omega - \omega_0 - \Omega) \). Upperstate lifetime \( T_1 = 10^2 \Omega_0^{-1} \), dipole relaxation time \( T_2 = 2 \times 10^2 \Omega_0^{-1} \), \( \Omega_0 \tau_R = 0.25 \). --- \( \gamma(0), \gamma_0(0), \gamma_{\text{inc}}(\omega), \gamma_{\text{scat}}(\omega), \gamma_{\text{oop}}(\omega) \)
The temporal evolution of operator $R_j(t)$ is derived from the Bloch equations. $\Gamma_{ad}(\xi, \tau)$ is calculated with the following assumptions:

(H1) $\xi l \ll I_0/I_S$ where $\alpha^{-1}$ is the Beer's length, $l$ the length of the cell, $I_0$ and $I_S$ the incident and saturated intensities respectively. This hypothesis means that the mean value of the atomic response is much smaller than $\Omega_0$, i.e. the medium is weakly absorbing. Then, $R_j$ in (1) is independent of $j$.

(H2) Third and higher order correlation functions like $\langle \delta R_j(t) \delta R_k(\tau) \delta R_\Delta(0) \rangle$, etc... are neglected in order to get closed coupled equations for second order cross correlation functions. A necessary condition for the validity of this approximation is shown to be $\Omega_0 \tau_R > 0.1$.

The spectrum $\gamma(\omega)$ of the atomic response $\Delta \Omega$ at the cell exit ($\xi = l$) is the Fourier transform (F.T.) of $\Gamma_{ad}(l, \tau)$. It is the sum of two terms $\gamma_{\text{coop}}(\omega)$ and $\gamma_{\text{inc}}(\omega)$. The "cooperative" spectrum $\gamma_{\text{coop}}(\omega) = \sum_{j \neq l} I_{jk}(\omega)$ is the F.T. of the first term in (2), and concerns the correlation between two different atoms, while the "incoherent" spectrum $\gamma_{\text{inc}}(\omega) = \sum I_{jk}(\omega)$ is the F.T. of the second term in (2) and concerns auto-correlation of the same atom. In this treatment each atom is simultaneously illuminated by the incident field and the forward response of other atoms. We show that $I_{jk}(\omega)$ is independent of j.

Let us precise that hypothesis (H1) may be avoided without any trouble, although (H2) is necessary to our treatment. However we recall that third and higher order correlation functions are required for high atomic densities ($\Omega_0 \tau_R \ll 1$).

We show that they have a noticeable contribution especially for the incoherent part of the spectrum at high atomic density ($\Omega_0 \tau_R \ll 1$). This result is shown on Fig. 1 where the usual one-atom ac-Stark effect $[3] \gamma(\omega)(I_R = 0)$ is compared to the expression of $\gamma_{\text{inc}}(\omega)$ when the response field of other atoms is taken into account by the way of $I_R \neq 0$. This could be experimentally checked by detecting the fluorescence spectrum perpendicularly to the axis both at the entrance and the exit of the cell. Any spectral function $I_{jk}(\omega)$ varies exponentially with the distance $\Delta \xi$-z, $I_{jk}(\omega) \propto e^{i(\omega t_1 - z)} e^{i(\omega t_2 - z)}$ for $z_1 - z_2 \approx 10 \mu s e^{i(\omega t_2 - z)}$ for $z_1 - Z_2$. The gain $g(\omega)$ is plotted in Fig. 2 as a function of $\omega$. Its behaviour illustrates quite well the variation $\gamma_{\text{inc}}(\omega)$ with respect to $\omega$ in Fig. 1. Finally we show that there is a coherent amplification near the sidebands $\omega = \omega_0 \pm \Omega$.

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We report here a time-resolved experiment demonstrating the role played by superelastic collisions (SEC) in efficient ionization of strontium vapor under pulsed resonant excitation. During the last few years there has been considerable growth in the experiments concerned with substantial ionization of an atomic vapor laser-excited to the first resonance level. In order to interpret these results it has been necessary to invoke the effects of electron heating through SEC [1]: the energy of any free electrons is increased by SEC with excited atoms. Consequently an ionization time is necessary to reach the burnout ionization. The purpose of this paper is to reveal the time evolution of the free electrons heated through SEC with excited atoms. The first time-resolved experiment, free of multiphoton processes, shows how the medium can be spontaneously burned out after the end of the laser excitation.

The beam provided by a pulsed tunable dye laser is focused ($I_0 \sim 30$ $\text{MW/cm}^2$) into an absorbing cell filled with strontium vapor ($10^{14}$ to $10^{16}$ $\text{at/cm}^2$) and krypton ($\sim 100$ Torr) has a buffer gas. The backward fluorescence is observed through a monochromator. We have tuned the laser line to populate the $5s^2\text{P}_1$ resonance level ($\lambda$ = 460.7 nm). The laser induced fluorescence comes not only from high lying levels of neutral strontium but also from the fluorescence levels of ionized strontium. The significant emission that we observed from the resonance levels of the ion requires a large ionization of strontium followed by excitation to these levels. This may be achieved by electrons having gained energy. We have studied the time evolution of the two ion resonance lines $\lambda_R$ = 407.8 nm and $\lambda_R$ = 421.5 nm for various atomic densities $n_0$. For a density larger than $2 \times 10^{15} \text{at/cm}^3$, the signal exhibits a maximum delayed with respect to the laser pulse. The corresponding delay time $\tau$ decreases from 14 $\mu s$ to 50 $\mu s$ as the atomic density increases. The time behavior of the fluorescence can be qualitatively understood as follows.

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During the laser pulse (15 ns) large population is created in the metastable $5s^5\text{p}_2\text{p}$ states, via optical pumping. Multiphoton ionization, laser induced Penning ionization, associative ionization give rise to a rapid seed of free electrons with low energy $e$. When the laser is turned off, the population of metastable atoms remains stored. The seed electrons gain energy through SEC which quench the metastable level and give rise to an exponential growth in the

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**Time Resolved Study of Superelastic Collisions in Laser Excited Strontium Vapor**

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PACS: 34

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