Stark broadening of spectral lines of atoms in the field of multimode laser radiation

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A theoretical study is reported of the Stark broadening of atomic spectral lines by nonmonochromatic laser radiation. An allowance is made for the finite number of longitudinal modes emitted by the laser. An analytic expression obtained for the absorption line profile demonstrates a strong dependence of this profile on the number of modes. Relationships are obtained between the frequencies of intermode beats, natural width of an atomic transition, and average (over a time much longer than the reciprocal of the frequency of intermode beats) Stark shift of atomic levels. Conditions are found under which the average Stark shift in a multimode field can be determined directly from the shift of an atomic transition, by analogy with a monochromatic field. The cases of small and asymptotically large numbers of modes are considered separately. In the latter limiting case the line profile is described by an expression obtained earlier using a series of stochastic models for laser radiation.

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One of the most important manifestations of the effects of high-power radiation on atoms and molecules in modern laser spectroscopy is the Stark shift of the energy levels.[1-3] In the simplest case of a nondegenerate level and an idealized monochromatic field $F=F_0\cos\omega t$ far from resonances of frequency ω with natural oscillation frequencies of an atom the Stark shift of the position of a level n is a quadratic function of F_0:

$$\eta_n=\alpha_n(\omega)F_0^2, \tag{1}$$

where $\alpha_n(\omega)$ is the dynamic polarizability of an atom in the nth state at a frequency ω. Radiation from real lasers is nonmonochromatic and the most typical reason for the finite width of the laser spectrum is the emission of a considerable number of longitudinal modes. The intensity of the field of such radiation can be represented in the form

$$F=\sum_{k=1}^{N}F_k\cos(\omega_k t+\varphi_k), \tag{2}$$

where F_k, ω_k, and φ_k are the amplitude, frequency, and phase of the kth mode; N is the number of longitudinal modes which are emitted. We shall assume that the frequencies ω_k form an equidistant spectrum ($|\omega_{k+1}-\omega_k|=\pi c/L$, where L is the length of the laser resonator and c is the velocity of light) and are grouped within a spectral interval $\Delta\omega=\pi cN/L$ near the central frequency

$$\bar{\omega}=\frac{1}{N}\sum_{k=1}^{N}\omega_k$$

($\Delta\omega$ is the spectral width of laser radiation). It is convenient to rewrite Eq. (2) in the form

$$F=\mathrm{Re}\,F_0\exp(i\bar{\omega}t)f(t), \tag{3}$$

$$f(t)=\frac{1}{F_0}\sum_{k=1}^{N}F_k\exp(i\Delta_k t+i\varphi_k),\qquad \Delta_k=\omega_k-\bar{\omega}. \tag{4}$$

The function $f(t)$ describes modulation of the radiation whose carrier frequency is $\bar{\omega}$ and whose average intensity is $F_0^2=\sum F_k^2$; this modulation results from intermode beats and its characteristic frequency is $\pi c/L$. The amplitudes and phases of the individual modes, F_k and φ_k also depend on (fluctuate with) time. However, if the mode structure of laser radiation is highly pronounced, these fluctuations (which contribute to the spectral width of a single mode) have a characteristic time $\tau_k \gg L/\pi c$. This means that in time intervals $\sim L/\pi c$, in which modulation of the envelope $f(t)$ appears, the amplitudes and phases of the individual modes remain constant.

The formal definition of the Stark shift of a level in the field of Eqs. (2)–(4) averaged over a time interval $T\gg\Delta\omega^{-1}$ is[4]

$$\eta_{nN}=\sum_{k=1}^{N}\alpha_n(\omega_k)F_k^2, \tag{5}$$

which is identical with Eq. (1) if the dependence $\alpha_n(\omega_k)$ is sufficiently smooth: $[\alpha_n(\omega_k)\approx\alpha_n(\bar{\omega})]$. We are now faced with the central problem: how is the Stark shift of Eq. (5) manifested in the observed spectral characteristics of atoms and molecules? The Stark broadening of spectral lines in nonmonochromatic fields has already been investigated (see, for example, Refs. 5–10). However, these investigations have ignored totally the actual mode structure of laser radiation. It has been assumed that nonmonochromatic laser radiation can be described by an expression for the field intensity of the type given by Eq. (3), where $f(t)$ is a steady-state random process of the discontinuous Markov type[5] or a complex Gaussian process (with a Rayleigh amplitude distribution).[6-10] These stochastic models of radiation are justified by the fact that, for an asymptotically large number N modes which are out of phase, the function $f(t)$ has—according to Eq. (4)—a considerable number of fairly random peaks. If we ignore the fact that the maximum amplitude of a peak is finite for any finite value of N, and if we consider the limit $N\to\infty$, we can model $f(t)$ by a steady-state (for example, Gaussian) random process. However, in some practical applications of laser spectroscopy the number of modes N is small (see, for example, Refs. 11–13). Therefore, it is essential to know what are the manifestations of the Stark shift of the levels under these conditions. Moreover, if we use the description of the radiation field given by Eqs. (2) and (3) and the number of modes is small ($N \sim 10$), there are no *a priori* grounds for assuming the envelope $f(t)$ to be a random process, for example one of the Gaussian type, and this is true at least for time intervals shorter than the fluctuation times of the amplitudes and phases of the individual modes but longer than the intermode beat time. Moreover, it is interesting to determine whether the Stark broadening of spectral lines can be found, including the case $N \gg 1$, directly from the initial "dynamic" description of laser radiation given by Eqs. (2)–(4) without additional assumptions reducing to ordering of $f(t)$ by a random process of special type. The present paper is concerned with the last two points.

We shall consider probe radiation of frequency Ω and the profile of an absorption line of an atom undergoing a transition from the ground state 1 to an excited state 2. We shall assume that an atom is in the field of multimode laser radiation of intensity F [see Eqs. (3) and (4)] and that $\hbar\bar{\omega} \ll \varepsilon_2 - \varepsilon_1$ (ε_i is the atomic energy in the ith state) so that by itself this laser radiation does not excite the atom. We shall simplify calculation of the Stark shift in the field described by Eqs. (3) and (4) by assuming that $|\varepsilon_3 - \varepsilon_2 - \hbar\bar{\omega}| \ll \hbar\bar{\omega}$ and that a similar equality applies to other natural frequencies of the atom, so that we can find the field-perturbed atomic spectrum by considering only the dipole-coupled states 2 and 3 whose mixing by the field under these conditions is maximal. We shall assume that the characteristic time $\Delta\omega^{-1}$, which governs the changes in the envelope $f(t)$, satisfies the condition

$$\hbar\Delta\omega \ll |\varepsilon_3-\varepsilon_2-\hbar\bar{\omega}|. \tag{6}$$

We shall also assume that the following inequality is obeyed:

$$|d_{23}|^2F_0^2|f(t)|^2 \ll (\varepsilon_3-\varepsilon_2-\hbar\bar{\omega})^2. \tag{7}$$

Then, in the second order of perturbation theory for the energy we can easily find the time-dependent shift of the level 2:

$$\eta(t)=\eta+\frac{2\eta}{F_0^2}\sum_{i<j=1}^{N}F_iF_j\cos(\Delta_{ij}t+\varphi_{ij}), \tag{8}$$

$$\eta={}^1/_4|d_{23}|^2F_0^2(\varepsilon_3-\varepsilon_2-\hbar\bar{\omega})^{-1},\quad \Delta_{ij}=\Delta_i-\Delta_j,\quad \varphi_{ij}=\varphi_i-\varphi_j. \tag{9}$$

Here, d_{ij} is the dipole matrix element and η is the average [in the sense of the definition given by Eq. (5)] Stark shift of the level 2. The validity of the approximation (8) and the errors resulting from its use are discussed more rigorously in the Appendix. Equation (8) allows us to obtain the following expression for the nondiagonal matrix element ρ_{21} of the density matrix, which governs the absorption by an atom of weak probe radiation of frequency Ω (and of field intensity F_Ω):

$$i\hbar\dot{\rho}_{21}=[\varepsilon_2-\varepsilon_1-\eta(t)-i\gamma]\rho_{21}-{}^1/_2d_{21}F_\Omega e^{-i\Omega t}. \tag{10}$$

Here, $\gamma=\hbar/T_2$ and T_2 is the transverse relaxation time. We shall ignore the population of the excited states in the absorbing system of atoms ($\rho_{11}=1$). We can easily find the steady-state, after a time $t \gg \hbar/\gamma$, solution of Eq. (10). This solution can then be employed in the usual way to find the imaginary part of the complex polarizability which governs the power of the probe radiation absorbed by the system:

$$\chi''(\Omega)=\frac{1}{\hbar}|d_{12}|^2\operatorname{Re}\frac{1}{T}\int_0^T dt\int_{-\infty}^0 d\tau\exp\left\{\frac{i}{\hbar}(q-\eta-i\gamma)\tau+ig(t,\tau)\right\}, \tag{11}$$

$$g(t,\tau)=\frac{4\eta}{\hbar F_0^2}\sum_{i<j=1}^{N}\frac{F_iF_j}{\Delta_{ij}}\sin\left(\frac{1}{2}\Delta_{ij}\tau\right)\cos\left[\frac{1}{2}\Delta_{ij}(\tau+2t)+\varphi_{ij}\right], \tag{12}$$

$$q=\varepsilon_2-\varepsilon_1-\hbar\Omega. \tag{13}$$

Equation (11) is averaged over the measurement time T. Clearly, T should be related to the width of the probe radiation spectrum $\delta\Omega$ by the inequality $T \gg \delta\Omega^{-1}$. We shall now consider a number of special cases which follow from Eqs. (11) and (12).

LASER RADIATION WITH FEW LONGITUDINAL MODES

The simplest case is the one with two modes (bichromatic fields). The situation with $N=2$ has been investigated earlier in connection with some nonlinear effects.[13,19,20] We shall assume that

$$T \gg \max(\Delta_{12}^{-1}, \delta\Omega^{-1}). \tag{14}$$

It now follows from Eqs. (11) and (12) that

$$\chi''(\Omega)=\frac{1}{\hbar}|d_{12}|^2\operatorname{Re}\int_{-\infty}^0 J_0\left(\frac{4\eta F_1F_2}{\hbar F_0^2\Delta_{12}}\sin\left(\frac{1}{2}\Delta_{12}\tau\right)\right)\times\exp\left(\frac{i}{\hbar}(q-\eta-i\gamma)\tau\right)d\tau. \tag{15}$$

Here, $J_n(x)$ is a Bessel function. We shall use[21]

$$J_0(2x\sin y)=\sum_{n=-\infty}^{+\infty}J_n^2(x)e^{2iny}. \tag{16}$$

We then find

$$\chi''(\Omega)=|d_{12}|^2\sum_{n=-\infty}^{+\infty}J_n^2\left(\frac{2\eta F_1F_2}{\hbar F_0^2\Delta_{12}}\right)\frac{\gamma}{(\varepsilon_2-\varepsilon_1-\eta-\hbar\Omega-n\hbar\Delta_{12})^2+\gamma^2}. \tag{17}$$

To be specific, we shall assume that $\eta > 0$. If $\gamma < \eta \ll \hbar\Delta_{12}$, then $\xi = 2\eta F_1F_2/\hbar F_0^2\Delta_{12} \ll 1$ and the absorption spectrum reduces to one line corresponding to $n=0$:

$$\chi''(\Omega)=\frac{\gamma|d_{12}|^2}{(\varepsilon_2-\varepsilon_1-\hbar\Omega-\eta)^2+\gamma^2}. \tag{18}$$

Thus, in the presence of laser radiation the absorption line shifts without a change in the profile by an amount equal to the average Stark shift, in the same way as in the case of a monochromatic field. If $\gamma \ll \hbar\Delta_{12} \lesssim \eta$ (i.e., if $\xi \gtrsim 1$ for $F_1 \approx F_2$), the absorption spectrum consists of a set of equidistant lines of width γ (which are satellites of the main $n=0$ line) and the intensities of these lines are governed by $J_n^2(\xi)$, where the frequency $\Omega = (\varepsilon_2 - \varepsilon_1 - \eta)/\hbar$ in the absorption spectrum is not distinguishable so that in the average Stark shift of the energy levels cannot in this case be observed directly. The appearance of a line structure in the absorption spectrum (i.e., the appearance of satellites of the n

$=0$ line) is due to activation of channels of the electron Raman scattering of the laser radiation. For $\eta/\hbar\Delta_{12} < 1$ these satellites are described by odd orders of perturbation theory. For example, the satellite with $n=1$ corresponds to the Raman scattering accompanied by the absorption of a photon $\hbar\Omega$ and a photon $\hbar\omega_1$ and by the stimulated emission of a photon $\hbar\omega_2$. This process can be described in the third order of perturbation theory and its probability is proportional to F_Ω^2 and F_0^4. The same result follows from Eq. (17) for $\eta/\hbar\Delta_{12} < 1$. In the case described by $\eta/\hbar\Delta_{12} > 1$, the first nonvanishing order of perturbation theory fails to describe this process. It is then necessary to sum all possible re-emissions of laser photons, described by higher orders of perturbation theory. This is essentially ensured by the calculation scheme used in the present study and based on the adiabatic approximation (see the Appendix).

We shall now consider the profile of an absorption band in the case of practical importance when $\hbar\Delta_{12} \ll \gamma < \eta$. It is convenient to find it directly from Eq. (15) because in the range of importance in the process of integration with respect to τ we then have $\Delta_{12}\tau \ll 1$. Assuming that $\sin\frac{1}{2}\Delta_{12}\tau \approx \frac{1}{2}\Delta_{12}\tau$, we obtain

$$\chi''(\Omega) = \frac{|d_{12}|^2}{(2R)^{1/2}} \{R^{1/2} + \gamma^2 - [q - \eta(1-\mu)][q - \eta(1+\mu)]\}^{1/2},$$
$$R = \{[q-\eta(1-\mu)]^2 + \gamma^2\}\{[q-\eta(1+\mu)]^2+\gamma^2\}, \tag{19}$$
$$q = \varepsilon_2 - \varepsilon_1 - \hbar\Omega, \quad \mu = 2F_1F_2/F_0^2.$$

According to Eq. (19), the absorption line profile is a strongly broadened curve with two maxima at $\hbar\Omega = \varepsilon_2 - \varepsilon_1 - \eta(1 \pm \mu)$ (Fig. 1). For comparison, Fig. 1 shows (by a dashed curve) a Lorentzian absorption profile in the presence of a two-mode field of the same average intensity but satisfying the condition $\gamma < \eta \ll \hbar\Delta_{12}$. A profile of the type shown in Fig. 1 is derived also in Ref. 16 but under very different assumptions relative to the experimental arrangement. It is assumed that the measurement time T is much shorter than the reciprocal of the intermode beat frequency ($\hbar/\gamma \ll T \ll 1/\Delta_{12}$), so that not even a single complete beat of the field amplitude occurs during the measurement time. Averaging is carried out over a set of successive measurements, each of which being characterized by a random

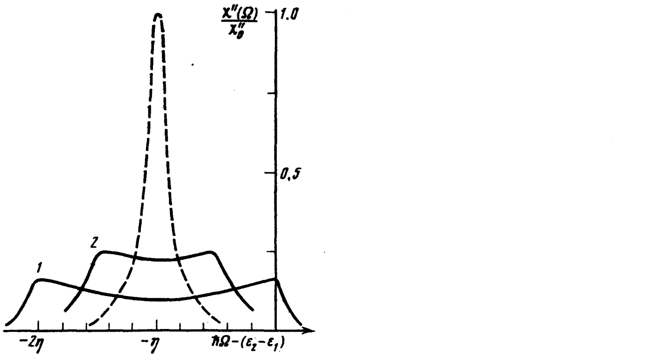

FIG. 1. Frequency dependence $\chi''(\Omega)$ in the $N=2$ case: $\chi_0'' = |d_{12}|^2/\gamma$, $\gamma = 0.1\eta$; curve 1 corresponds to $\hbar\Delta_{12} \ll \gamma$, $F_1 = F_2$ ($\mu = 1$); curve 2 corresponds to $\hbar\Delta_{12} \ll \gamma$, $F_1 = 1/4F_2$ ($\mu \approx 0.47$); the dashed curve represents the case when $\hbar\Delta_{12} \gg \gamma$.

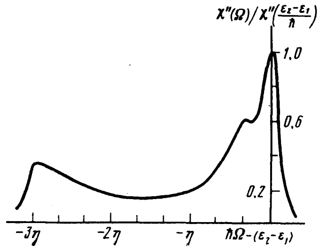

FIG. 2. Frequency dependence $\chi''(\Omega)$ in the $N=3$ case: $F_1 = F_2 = F_3 = F_0/\sqrt{3}$.

value of the phase shift φ_{12}.

We shall consider briefly the case $N=3$. We shall confine ourselves to the conditions described by $\max|\Delta_{ij}| \ll \gamma/\hbar$ and we shall assume that $F_1 = F_2 = F_3 = F_0/3^{1/2}$ and that the modes are locked. Under these assumptions we obtain the following expression for $\chi''(\Omega)$:

$$\chi''(\Omega) = \frac{|d_{12}|^2}{2\pi} \int_{-\pi}^{+\pi} \frac{\gamma\, d\varphi}{(q - \eta - {}^4/_3\eta\cos\varphi - {}^2/_3\eta\cos 2\varphi)^2 + \gamma^2}. \tag{20}$$

If $\eta \gg \gamma$, the Lorentzian in Eq. (20) can be replaced with the δ function which gives

$$\chi''(\Omega) = \frac{3|d_{12}|^2}{8(3q)^{1/2}}\theta(q)\left\{\frac{\theta[3(\eta-q)+2(3\eta q)^{1/2}]}{[3(\eta-q)+2(3\eta q)^{1/2}]^{1/2}} + \frac{\theta[3(\eta-q)-2(3\eta q)^{1/2}]}{[3(\eta-q)-2(3\eta q)^{1/2}]^{1/2}}\right\}$$
$$\theta(x) = 1, \quad x > 0; \quad \theta(x) = 0, \quad x < 0. \tag{21}$$

It is clear from Eq. (21) that $\chi''(\Omega)$ has three maxima at $q=0$, $q=\eta/3$, and $q=3\eta$. The square-root divergences at these points are removed by allowing for the fact that the value of γ is finite. The resultant cumbersome expression will not be given here. A typical line profile described by Eqs. (20) and (21) is shown in Fig. 2. In the complex band shown in Fig. 2, as in the case demonstrated in Fig. 1 (curve 1), the average Stark shift η appears only in the average form.

MULTIMODE LASER RADIATION

We shall now consider the case when $N \gg 1$. As in the few-mode case, the condition for direct observation of the average Stark shift (5) is the condition under which Eq. (11) can be simplified by dropping $g(t, \tau)$. This can be done when the average Stark shift is considerably less than the width of the laser radiation spectrum, i.e., when

$$\eta \ll \hbar\Delta\omega, \tag{22}$$

which is in agreement with the qualitative ideas put forward in Ref. 4. In the alternative case of $\eta > \hbar\Delta\omega$, we have a complex absorption band where η does not appear directly. A simple analytic expression for this band can be obtained if $\hbar\Delta\omega \ll \gamma$. Then, in the range of importance in integration with respect to τ we can obtain $\sin\frac{1}{2}\Delta_{ij}\tau \approx \frac{1}{2}\Delta_{ij}\tau$ and it is then easy to carry out integration with respect to t assuming that the mode amplitudes are equal ($F_i = F_0/N^{1/2}$ is the rectangular form of the spectrum) and the mode phases are not matched. We then find

$$\chi''(\Omega)=\frac{1}{\hbar}|d_{12}|^2\,\mathrm{Re}\int_{-\infty}^{0}e^{i(q-i\gamma)\tau/\hbar}\sum_{n=0}^{\infty}\left(-\frac{i\eta\tau}{\hbar N}\right)^n\frac{1}{n!}d\tau$$
$$\times\sum_{k_1k_2\ldots k_N=0}^{n}(n!)^2(k_1!\,k_2!\ldots k_N!)^{-2} \qquad (23)$$

$(k_1+k_2+\cdots+k_N=n)$. If $N\gg1$, then

$$\sum_{k_1k_2\ldots k_N}\ldots\to n!\,N^n$$

(see Refs. 14 and 15), which gives

$$\chi''(\Omega)=|d_{12}|\int_0^{\infty}e^{-X}\frac{\gamma\,dX}{(q-\eta X)^2+\gamma^2}. \qquad (24)$$

Equation (24) is derived using the familiar representation

$$n!=\int_0^{\infty}e^{-x}x^n\,dx.$$

Equation (24) can be represented also in the form

$$\chi''(\Omega)=-\frac{|d_{12}|^2}{\eta}\,\mathrm{Im}\left\{e^{-(q-i\gamma)/\eta}\,\mathrm{Ei}\left(\frac{q-i\gamma}{\eta}\right)\right\}. \qquad (25)$$

Here, $Ei(x)$ is the exponential integral. The simplest expression for $\chi''(\Omega)$ is obtained for $\eta\gg\gamma$:

$$\chi''(\Omega)=\frac{\pi|d_{12}|^2}{\eta}\exp\left\{-\frac{\varepsilon_2-\varepsilon_1-\hbar\Omega}{\eta}\right\}\theta(\varepsilon_2-\varepsilon_1-\hbar\Omega). \qquad (26)$$

Equation (26) has been obtained earlier[5,7,8] on the assumption that the intensity of laser radiation can be regarded as a random process with an exponential distribution (Rayleigh amplitude distribution). Here the same expression is obtained directly by multimode description of the radiation field (2) without any additional assumptions.

We shall now introduce moments σ_m of an absorption band

$$\sigma_m=\int_{-\infty}^{+\infty}\left(\Omega-\frac{\varepsilon_2-\varepsilon_1}{\hbar}\right)^m F(\Omega)\,d\Omega. \qquad (27)$$

Here, $F(\Omega)$ is the normalized function of the band profile

$$F(\Omega)=\chi''(\Omega)\left[\int_{-\infty}^{+\infty}\chi''(\Omega)\,d\Omega\right]^{-1}. \qquad (28)$$

Using Eq. (11), we easily obtain for $\eta\gg\gamma$

$$\sigma_m=(-1)^m\hbar^{-m}\eta^m\frac{1}{T}\int_0^T|f(t)|^{2m}\,dt$$
$$=(-1)^m\hbar^{-m}\eta^m\frac{1}{N^m}\sum_{k_1k_2\ldots k_N=0}^{m}\frac{(m!)^2}{(k_1!\,k_2!\ldots k_N!)^2} \qquad (29)$$

$(k_1+k_2+\cdots+k_N=m)$. According to Eq. (29), the center of gravity of an absorption band is independent of N ($\sigma_1=-\eta/\hbar$). Higher moments depend on the number of modes N, for example, $\sigma_2=2\eta^2\hbar^{-2}(1-1/2N)$. [The values of the sum in Eq. (29) are given in Ref. 14 for $m=1$–6.] The moments σ_m practically cease to depend on N only for $N\gg m^2$. The corresponding asymptotic value is $\sigma_m=(-1)^m\hbar^{-m}\eta^m m!$, which naturally corresponds to the asymptotic expressions for the line profile (24)–(26). It is clear from Eq. (29) that the mth moment of the absorption band σ_m is proportional to the mth correlation moment of the intensity of N-mode radiation. Consequently, an analysis of an absorption band can be used to reconstruct the intensity distribution function of N-mode radiation, i.e., it can give important information on the statistical properties of such radiation.

In addition to the experimental arrangement discussed above, we can also conceive a different configuration when the source of the Stark broadening of an absorption line is a pulsed laser emitting radiation with amplitudes and phases of the individual modes varying from pulse to pulse in a random manner. Each measurement is carried out during one pulse at fixed values of F_k and φ_k. The line profile is found by averaging over a large number of separate pulses (measurements). It is assumed that the number of measurements is so large that it represents a complete ensemble of random quantities F_k and φ_k. In describing the results of such experiments the quantity $\chi''(\Omega)$ [see Eq. (11)] should be additionally averaged over the ensemble of the separate realizations F_k and φ_k subject to the additional condition

$$\sum_{k=1}^{N}F_k^2=F_0^2=\mathrm{const},$$

which expresses the fact that the average intensity of laser radiation is reproduced from pulse to pulse. Thus, the observed absorption line profile is

$$\bar{\chi}''(\Omega)=C_N\int\prod_{k=1}^{N}F_k\,dF_k\,d\varphi_k\,\delta\left(F_0^2-\sum_{k=1}^{N}F_k^2\right)$$
$$\times\exp\left\{-\frac{1}{\sigma}\sum_{k=1}^{N}F_k^2\right\}\chi''(\Omega,\{F_k,\varphi_k\}). \qquad (30)$$

Here, C_N is the normalization constant; $\chi''(\Omega,\{F_k,\varphi_k\})$ is governed by Eq. (11); complex random quantities $F_k\exp(i\varphi k)$ are assumed to have a Gaussian distribution with the same variance σ (the radiation spectrum is assumed to be rectangular). In the case of a narrow spectrum ($\Delta\omega\ll\gamma/\hbar\lesssim\eta/\hbar$), Eq. (30) can be reduced to

$$\bar{\chi}''(\Omega)=|d_{12}|^2\int_0^{\infty}\mathcal{P}(X)\frac{\gamma\,dX}{(q-\eta X)^2+\gamma^2}, \qquad (31)$$

$$\mathcal{P}(X)=C_N\int\prod_{k=1}^{N}F_k\,dF_k\,d\varphi_k\,\delta\left(F_0^2-\sum_{k=1}^{N}F_k^2\right)$$
$$\times\delta\left(X-\frac{1}{F_0^2}\left|\sum_{k=1}^{N}F_ke^{i\varphi_k}\right|^2\right)\exp\left(-\frac{1}{\sigma}\sum_{k=1}^{N}F_k^2\right),$$
$$\int_0^{\infty}\mathcal{P}(X)\,dX=1. \qquad (32)$$

The quantity $\mathcal{P}(X)$ is the distribution function of the intensity (in units of F_0^2) of multimode radiation. This function has been calculated earlier[13,17,22] and it is given by

$$\mathcal{P}(X)=\frac{N-1}{N}\left(1-\frac{X}{N}\right)^{N-2}\theta\left(1-\frac{X}{N}\right). \qquad (33)$$

If $\eta\gg\gamma$, it follows from Eqs. (31) and (33) that

$$\bar{\chi}''(\Omega)=\frac{\pi|d_{12}|^2}{\eta}\frac{N-1}{N}\left(1-\frac{\varepsilon_2-\varepsilon_1-\hbar\Omega}{N\eta}\right)^{N-2}$$
$$\times\theta\left(1-\frac{\varepsilon_2-\varepsilon_1-\hbar\Omega}{N\eta}\right)\theta(\varepsilon_2-\varepsilon_1-\hbar\Omega), \qquad (34)$$

i. e., the absorption band profile repeats the distribution function of the intensity of multimode laser radiation. In the limit $N\to\infty$, Eq. (34) reduces to Eq. (26).

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APPENDIX

We shall now consider the precision of the approximation (8) for $\eta(t)$ obtained in the second order of perturbation theory. The expression for $\eta(t)$ includes the average Stark shift η given by Eq. (9) and its validity is governed[1] by the inequality (7), which can be satisfied by relatively moderate fields F_0. On the other hand, the validity of the second term in Eq. (8), which governs the intensity of the satellites ($n\neq 0$) of the main absorption line ($n=0$), is not ensured by the inequality (7), as we shall show below. Moreover, there is literally no parameter containing F_0 which would ensure the validity of the time-dependent part of $\eta(t)$ for any value of n. In the case of sufficiently high values of $|n|$ the approximation (8) gives an incorrect result for the intensity of the nth satellite when F_0 is low. We shall show this by finding the wave function of an excited atomic state bearing in mind that $|\varepsilon_3-\varepsilon_2-\hbar\bar{\omega}|\ll\hbar\bar{\omega}$ and we shall express this wave function in the form

$$\Psi(t)=[a_2(t)|2\rangle e^{i\bar{\omega}t/2}+a_3(t)|3\rangle e^{-i\bar{\omega}t/2}]\exp\left\{-\frac{it}{2\hbar}(\varepsilon_2+\varepsilon_3)\right\}, \qquad (A1)$$

where $a_2(t)$ and $a_3(t)$ satisfy the usual system of equations in the two-level approximation (see p. 176 in Ref. 23), which follows from the secular Schrödinger equation. The condition (6) justifies the adiabatic approximation for the solution of the system,[23,24] i. e., it allows us to assume

$$a_{2,3}=b_{2,3}\exp\left\{-\frac{i}{\hbar}\int\varepsilon(t)\,dt\right\},$$

and we then find that the slow time dependence of $b_{2,3}$ can be ignored in the spirit of the adiabatic approximation. We thus obtain the system of equations

$$[\varepsilon+{}^1/_2(\varepsilon_3-\varepsilon_2-\hbar\bar{\omega})]b_2+{}^1/_2d_{23}F_0f(t)\,b_3=0,$$
$$[\varepsilon-{}^1/_2(\varepsilon_3-\varepsilon_2-\hbar\bar{\omega})]b_3+{}^1/_2d_{32}F_0f^*(t)\,b_2=0, \qquad (A2)$$

whose nontrivial solution is possible only for

$$\varepsilon(t)=\pm\lambda(t),\quad \lambda(t)={}^1/_2[(\varepsilon_3-\varepsilon_2-\hbar\bar{\omega})^2+|d_{23}F_0f(t)|^2]^{1/2}. \qquad (A3)$$

The approximation (8) follows from Eq. (A3) if the expansion of the square root occurring in the definition of $\lambda(t)$ is confined to the first nonvanishing term with F_0^2. In the simplest case of two modes we can obtain the result for $\chi''(\Omega)$ without invoking the approximation (8). In particular, we can obtain an expression of the (17) type where $J_n^2(\eta/\hbar\Delta_{12})(F_1=F_2=F_0/2^{1/2})$ is replaced by the quantity $|U_n|^2$ defined by

$$U_n=\frac{\Delta_{12}}{2\pi}\int\limits_0^{2\pi/\Delta_{12}}\cos\frac{\beta(t)}{2}\exp\left\{-\frac{i}{\hbar}\int\limits_0^t\lambda(t')\,dt'+i(S+n\Delta_{12})t\right\}, \qquad (A4)$$

$$\beta(t)=\operatorname{arctg}\frac{d_{23}F_0f(t)}{\varepsilon_3-\varepsilon_2-\hbar\bar{\omega}},\quad S=\frac{\Delta_{12}}{2\pi\hbar}\int\limits_0^{2\pi/\Delta_{12}}\lambda(t)\,dt. \qquad (A5)$$

When the inequality (7) is obeyed, we can expand as a series the root occurring in the definition of S. We then obtain

$$\hbar S={}^1/_2(\varepsilon_3-\varepsilon_2-\hbar\bar{\omega})+\eta, \qquad (A6)$$

which implies the validity of Eq. (9) for the average Stark shift. In general, it is not possible to expand the root $\lambda(t)$ of Eq. (A3) occurring in the integrand of Eq. (A4). If $\eta/\hbar\Delta_{12}\ll 1$ and $|n|>1$, the integral in Eq. (A4) relating to the rapidly oscillating function inside the integrand is exponentially small and, therefore, extremely sensitive to the approximations in the quantity $\lambda(t)$. The terms dropped in the approximation (8) of the expansion $\lambda(t)$ make a contribution to $|U_n|^2$ of the same order as those which are retained. However, if $\eta/\hbar\Delta_{12}\gtrsim|n|$, the integral in Eq. (A4) is not exponentially small and the sensitivity to the approximation in $\lambda(t)$ disappears. When the condition for the above adiabatic approximation $|\varepsilon_3-\varepsilon_2-\hbar\omega|/\hbar\Delta_{12}\equiv\vartheta\gg 1$ [see Eq. (6)] is obeyed, the integral (A4) can be estimated by the steepest-descent method. (Details of ways of estimating integrals of this type can be found in an earlier paper by the present author.[25]) We find that when $p<\vartheta$ [$p=2(S/\Delta_{12}-n)\approx\vartheta+2\eta/\Delta_{12}\hbar-2n$], then

$$|U_n|^2\sim\exp\left\{2p\operatorname{Arsh}\frac{r}{\varkappa\vartheta}-\frac{2\vartheta}{(1+\varkappa^2)^{1/2}}D\left(\delta_1,\frac{1}{(1+\varkappa^2)^{1/2}}\right)-\frac{2pr}{(\vartheta^2(1+\varkappa^2)-p^2)^{1/2}}\right\},$$
$$r=(|p^2-\vartheta^2|)^{1/2},\quad \varkappa^2=\frac{2|d_{23}|^2F_0^2}{(\varepsilon_3-\varepsilon_2-\hbar\bar{\omega})^2}, \qquad (A7)$$
$$\delta_1=\arcsin\left(\frac{1+\varkappa^2}{1+\varkappa^2\vartheta^2/r^2}\right)^{1/2},$$

$$k^2D(\delta,k)=F(\delta,k)-E(\delta,k),$$

$F(\delta,k)$ and $E(\delta,k)$ are elliptic integrals of the first and second kind, respectively. It follows from Eq. (A7) that if $\eta/\hbar\Delta_{12}\ll 1$, then

$$|U_n|^2\sim\left(\frac{\vartheta+p}{2\vartheta}\right)^{2p}\left(\frac{e^2\varkappa^2}{16}\frac{\vartheta+p}{\vartheta-p}\right)^{\vartheta-p},\quad e=2{,}72\ldots.$$

Under these conditions, we have

$$J_n^2(\eta/\hbar\Delta_{12})\sim(e\eta/2\hbar\Delta_{12}n)^{2n},$$

which gives

$$\rho_n=\frac{|U_n|^2}{J_n^2(\eta/\hbar\Delta_{12})}\sim\exp\left\{2n+2(\vartheta-n)\ln\left(1-\frac{n}{\vartheta}\right)\right\}. \qquad (A8)$$

If $n/\vartheta<1$, then $\rho_n\propto\exp(n^2/\vartheta)$, i. e., the intensity of the nth satellite of the main absorption line estimated from Eq. (A7) is ρ_n times higher than the value obtained using the approximation (8). If $\vartheta=10$, then $\rho_n=2.7$ even for $n=3$. An estimate of $|U_n|^2$ similar to that given by Eq. (A7) is also obtained if $p>\vartheta$ and $(p^2-\vartheta^2)^{1/2}/\varkappa\vartheta>1$ (these inequalities are satisfied for $n=0$ and $|n|>2\eta/\hbar\Delta_{12}$). We then find

$$|U_n|^2\sim\exp\left\{\frac{2\vartheta}{(1+\varkappa^2)^{1/2}}D\left(\delta_2,\frac{1}{(1+\varkappa^2)^{1/2}}\right)-2p\operatorname{Arsh}\frac{r}{\varkappa v}+2p\frac{(p^2-\vartheta^2(1+\varkappa^2))^{1/2}}{r}\right\}, \qquad (A9)$$
$$\delta_2=\arccos\frac{\varkappa\vartheta}{r}.$$

It also follows from Eq. (A9) that $\rho_n\propto\exp(n^2/\vartheta)$ for $\eta/\hbar\Delta_{12}\ll 1$. Thus, the approximation (8) is quantitatively unsuitable for finding the intensities of the Raman satellites located on both sides of the main absorption line

($n=0$) when $\eta/\hbar\Delta_{12}<1$, i.e., when the absorption spectrum has a line structure and the main absorption line is shifted relative to the absorption line of a free atom by an amount equal to the average quandratic Stark shift η. The error due to the approximation (8) rises exponentially on increase in the satellite number. The correct expression for $\chi''(\Omega)$ is obtained by replacing in Eq. (17) the quantity $J_n^2(\eta/\hbar\Delta_{12})$ with $|U_n|^2$ which is estimated by Eqs. (A7) and (A9). If $\eta/\hbar\Delta_{12}\gg 1$, the ratio ρ_n differs little from unity and the approximation (8) is satisfactory at least for $\eta/\hbar\Delta_{12}\gtrsim|n|$. It is these values of $|n|$ that are important in the formation of a Stark-broadened absorption band when the laser spectrum is narrow ($\hbar\Delta_{12}\ll\gamma\lesssim\eta$). Thus, the absorption bands corresponding to Eq. (19) are described well by the approximation (8). It is easily shown that this applies to the case $N>2$, i.e., to Eqs. (20), (21), and (24)–(26).

[1]N. B. Delone and V. P. Kraĭnov, Atom v sil'nom svetovom pole (Atom in a Strong Optical Field), Atomizdat, M., 1978, p. 159.

[2]L. P. Rapoport, B. A. Zon, and N. L. Manakov, Teoriya mnogofotonnykh protsessov v atomakh (Theory of Multiphoton Processes in Atoms), Atomizdat, M., 1978, p. 42.

[3]H. Walther, in: Laser Spectroscopy of Atoms and Molecules (Topics in Applied Physics, Vol. 2), Springer Verlag, Berlin, 1976, p. 1 (Russ. Transl., Mir, M., 1979, p. 68).

[4]N. B. Delone and V. P. Kraĭnov, Preprint No. 83 (in Russian), Lebedev Institute, Academy of Sciences of the USSR, Moscow, 1979.

[5]L. D. Zusman and A. I. Burshteĭn, Zh. Eksp. Teor. Fiz. 61, 976 (1971) [Sov. Phys. JETP 34, 520 (1972)].

[6]S. G. Przhibel'skiĭ, Opt. Spektrosk. 35, 715 (1973) [Opt. Spectrosc. (USSR) 35, 415 (1973)].

[7]V. A. Kovarskiĭ and N. F. Perel'man, Zh. Eksp. Teor. Fiz. 68, 465 (1975) [Sov. Phys. JETP 41, 226 (1975)].

[8]A. M. Bonch-Bruevich, S. G. Przhibel'skiĭ, V. A. Khodovoĭ, and N. A. Chigir', Zh. Eksp. Teor. Fiz. 70, 445 (1976) [Sov. Phys. JETP 43, 230 (1976)].

[9]P. V. Elyutin, Opt. Spektrosk. 43, 542 (1977) [Opt. Spectrosc. (USSR) 43, 318 (1977)].

[10]A. T. Georges, P. Lambropoulos, and P. Zoller, Phys. Rev. Lett. 42, 1609 (1979).

[11]B. Decomps, M. Dumont, and M. Ducloy, in: Laser Spectroscopy of Atoms and Molecules (Topics in Applied Physics, Vol. 2), Springer Verlag, Berlin, 1976, p. 283 (Russ. Transl., Mir, M., 1979, p. 325).

[12]C. Lecompte, G. Mainfray, C. Manus, and F. Sanchez, Phys. Rev. A 11, 1009 (1975).

[13]F. Sanchez, Nuovo Cimento B 27, 305 (1975).

[14]I. V. Tomov and A. S. Chirkin, Kvantovaya Elektron. (Moscow) No. 1, 110 (1971) [Sov. J. Quantum Electron. 1, 79 (1971)].

[15]J. L. Debethune, Nuovo Cimento B 12, 101 (1972).

[16]V. A. Kovarskiĭ, N. F. Perel'man, and S. S. Todirashku, Kvantovaya Elektron. (Moscow) 1, 2417 (1974) [Sov. J. Quantum Electron. 4, 1344 (1975)].

[17]A. V. Masalov, Kvantovaya Elektron. (Moscow) 3, 1667 (1976) [Sov. J. Quantum Electron. 6, 902 (1976)].

[18]N. B. Delone, V. A. Kovarskiĭ, A. V. Masalov, and N. F. Perel'man, Preprint No. 120 (in Russian), Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow, 1978; Usp. Fiz. Nauk 131, 617 (1980) [Sov. Phys. Usp. 23, (1980)].

[19]P. A. Apanasevich, Osnovy Teorii vzaimodeĭstviya sveta s veshchestvom (Fundamentals of the Theory of Interaction of Light with Matter), Nauka i Tekhnika, Minsk, 1977.

[20]S. P. Goreslavskiĭ and V. P. Kraĭnov, Zh. Eksp. Teor. Fiz. 76, 26 (1979) [Sov. Phys. JETP 49, 13 (1979)].

[21]I. S. Gradshteĭn and I. M. Ryzhik, Tablitsy integralov, summ, ryadov i proizvedeniĭ, Fizmatgiz, M., 1963 (Tables of Integrals, Series, and Products, Academic Press, New York, 1965).

[22]J. Gersten and M. Mittleman, in: Electron and Photon Interaction with Atoms (Proc. Intern. Symposium at University of Stirling, Scotland, 1974, ed. by H. Kleinpoppen and M. R. C. MacDowell), Plenum Press, New York, 1976, p. 553.

[23]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika, Nauka, M., 1974, p. 230 (Quantum Mechanics, Pergamon Press, Oxford, 1974).

[24]L. I. Schiff, Quantum Mechanics, 2nd ed., McGraw-Hill, New York, 1955 (Russ. Transl., IIL, M., 1959, 246).

[25]N. F. Perel'man, Zh. Eksp. Teor. Fiz. 68, 1640 (1975) [Sov. Phys. JETP 41, 822 (1975)].

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