Effects of field-induced splitting of levels having high angular momenta

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We investigate manifestations of field-induced splitting in the spectrum of a test field in the presence of a strong field resonant to an adjacent transition in a three-level system with states degenerate in the orientations of the angular momentum. We show that since the splitting effect is different for different $M$-transitions the spectrum of the test-field operation consists of a set of individual spectral components that can overlap at $|J| > 1$ and form a broad common spectrum. We note that the envelope of the integral spectrum includes narrow spectral structures that preserve information on the natural line width. We analyze the dependence of the shapes of the spectra on the types of transitions in a three-level system and on the polarization states of the test and strong fields. For linear and circular polarization of the strong field, the problem is solved by a rigorous quantum-mechanical approach. A classical description of the orientation of the angular momentum is used in the case of elliptic polarization of the strong field.

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1. INTRODUCTION

Level splitting in the field of intense resonant radiation (sometimes called the dynamic Stark effect) is one of the fundamental effects of nonlinear spectroscopy.\(^1\)\(^2\) It has been thoroughly studied within the framework of the model of nondegenerate states. Its strongest manifestation is the splitting of the amplification (absorption) lines of test radiation.

It is known at the same time, however, that real states of atoms and molecules are degenerate in the $M$-projections of the angular momentum. Since the matrix elements of the operator of the interaction with radiation are different for different $M$-transitions, analysis of field splitting of levels by using the model of nondegenerate states is inapplicable to real objects. In the particular cases of linear or circular polarization of the strong radiation, it has been qualitatively understood that each $M$-transition produces its own splitting effect; in the upshot, the spectrum of the amplification (amplification) of test radiation should contain a corresponding number of components that correspond to the individual $M$-transitions. However, no concrete analysis of the relative positions of these components on the frequency scale and of their relative intensities has, to our knowledge, been carried out.

Of special interest are transitions between levels with large angular momenta $J$. In this case the number of spectral components can be so large that they begin to overlap. This raises the question of the envelope of the spectrum, i.e., of the actual shape of the spectral line that should be observed in experiment.

Finally, if the strong radiation has an arbitrary (elliptic) polarization, even a qualitative picture of the field splitting can be obtained on the basis of the previously developed premises. Moreover, a rigorous quantum-mechanical solution of the corresponding problem is fraught with considerable mathematical difficulties. These difficulties can, however, be overcome in the case of large $|J|$ by using a method\(^1\)\(^4\) based on the classical description of the orientation of the angular momentum.

We investigate here the effect of field-induced splitting of levels predominantly under conditions when the lines of individual $M$-transitions overlap. The problem is treated in one of the traditional formulations: the absorption (amplification) spectrum of test radiation is analyzed in the presence of a strong field resonant with an adjacent transition. We consider the cases of circular and linear polarization of the strong field, when a rigorous quantum-mechanical solution can be obtained, as well as the case of elliptic polarization with a classical description of the orientation of $J$.

We show that in a sufficiently intense field the spectrum has a doublet structure and the forms of the doublet components depend significantly on the polarizations of both the strong and test fields, as well as on the ratio of the angular momenta of the combining levels. One of the salient features of the spectrum is the possible existence of narrow structures and abrupt boundaries, that preserve the information on the natural (in the absence of the strong field) line width.

The bulk of the present data on nonlinear spectroscopy (in particular, with respect to field splitting), was obtained for atomic objects. Nonlinear spectroscopy of molecules, which typically have large values of $J$, is now undergoing intensive development. We hope that the results of the present paper can contribute to a correct interpretation of the experimental data of nonlinear-spectroscopy experiments on molecular objects.

2. COMPONENTS OF THE SPECTRUM OF TEST RADIATION UNDER CONDITIONS OF STRONG FIELD SPLITTING

We recall first the main feature of the field splitting effect in the nondegenerate-state model. Let the strong monochromatic radiation be at resonance with the $m-n$ transition, and the test radiation with the $m-l$ transition (Fig. 1). In the case of homogeneous broadening, the work $P_\omega$ of the test field is described by the expression\(^3\)\(^5\)

$$P_\omega = 2\hbar\alpha_{mn} G_{\omega} |\Omega| \{ [\Gamma_{\omega m} - (\Omega_{\omega} - \Omega) \{ (\Omega_{\omega m} - \Omega_{\omega n}) - (\Omega_{\omega n} - \Omega_{\omega}) \} + (\Omega_{\omega m} - \Omega_{\omega n}) + (\Omega_{\omega n} - \Omega_{\omega}) \} \} \{ [\Gamma_{\omega n} - (\Omega_{\omega} - \Omega) \{ (\Omega_{\omega n} - \Omega_{\omega m}) + (\Omega_{\omega m} - \Omega_{\omega}) \} \} \} \}$$

$$G = E_\omega \omega_{mn}/2\hbar, \quad G_\omega = E_\omega \omega_{ml}/2\hbar, \quad \Omega = \omega - \omega_{mn}, \quad \Omega_{\omega} = \omega - \omega_{ml}.$$
Here $E$ and $\omega$ are the amplitude of the electric field and the frequency of the strong radiation. The subscript $m$ marks the corresponding characteristics of the test radiation; $\Gamma_m$ and $\Gamma_m'$ are relaxation constants that characterize the broadening of the spectral line in the $i-j$ transitions; $N_i$ is the population of the level $i$ in the absence of radiation.

Expression (2.1) contains the population $P_{ni}$ of the level $n$ and the off-diagonal element $V_{nm}$ of the density matrix, due to the interaction with the strong field. They satisfy the equations:

$$\rho_{nm} = N_n \frac{G_{nm}}{\Gamma_m} + \frac{N_m}{\Gamma_m'} \rho_{mn},$$

$$\rho_{mn} = -G_{nm} \frac{N_n}{\Gamma_m} + \frac{N_m}{\Gamma_m'} \rho_{nm},$$

where $\rho_{nm}$ and $\rho_{mn}$ are the relaxation constants of the levels $m$ and $n$. The quantity $\kappa$ is the so-called saturation parameter.

The dependence of $P_{ni}$ on the test-radiation frequency $(\omega, \Gamma_i, \Gamma_i')$ is separated in explicit form in (2.1). The field-splitting effect is due entirely to the presence of the quantity $G_{12}$ in the denominator of (2.1). It manifests itself most strongly under the conditions $|G_1|, |G_2| \gg \Gamma_i', \Gamma_i$, i.e., when the strong field is intense enough. In this case it is convenient to rewrite (2.1) in the form

$$P_{ni} = \frac{2\omega_0 |G_{12}|^2}{\Gamma_i + \Gamma_i'} \left[ \frac{A_{ni}N_m - B_{ni}N_m}{\Gamma + (\Omega_{ni} - \Omega)} - \frac{A_{ni}N_m - B_{ni}N_m}{\Gamma + (\Omega_{ni} + \Omega)} \right].$$

It is distinctly seen here that the spectral line is a doublet whose components have a Lorentz shape with identical half-width and their spacing depends on the strong-field intensity. The ratio of the intensities of the components depends on $\Omega$. In the important particular case $\Omega = 0$ (or $|G_{12}|^2 \ll \Gamma_i$, which means an even higher radiation intensity) we have

$$P_{ni} = \frac{2\omega_0 |G_{12}|^2}{\Gamma + (\Omega_{ni} - \Omega)} \left[ \frac{A_{ni}N_m - B_{ni}N_m}{\Gamma + (\Omega_{ni} - \Omega)} + \frac{A_{ni}N_m - B_{ni}N_m}{\Gamma + (\Omega_{ni} + \Omega)} \right].$$

We consider now a situation wherein the levels $m, n, \ldots$ are degenerate in the projections $J_m, J_n, \ldots$ of the angular momenta. We assume for the sake of argument that the strong and test fields have the same linear polarization. Figure 2 shows schematically the optical transitions due to these fields. It is clearly seen from this figure that the overall system of levels and transitions breaks up into a set of three-level subsystems of the type shown in Fig. 1. Each of them can be described by the equations given above. The resultant work of the test field is given by the relation

$$P_{ni} = \sum P_{ni}(M),$$

where $P_{ni}(M)$ pertains to an individual $M$-subsystem ($M$ will hereafter be taken to mean the projection of the angular momentum of the level $m$). The results (2.1)-(2.4) remain in force for $P_{ni}(M)$, provided the following substitutions are made:

$$G_{12} \rightarrow |G_1|^2 \rightarrow |G(M)|^2 \rightarrow |G|^2 f(M),$$

$$G_{12} \rightarrow |G(M)| \rightarrow |G|^2 f(M),$$

where $f$ is the vector-addition coefficient.

We shall analyze hereafter transitions with large level angular momenta $(J_m, J_n, J_i, \ldots)$. In accord with the asymptotic values of the vector-addition coefficients, the expressions for $f(M)$ take on the form shown in Table I. If the test radiation is circularly polarized or has the same linear polarization as the strong radiation, the data in the table are valid for $f_0(M)$ (with allowance for the natural substitution $J_0 = J_{ni}$). If, however, the test field has an orthogonal linear polarization...
polarization $f_{n}(M)$ takes the following form:

$$f_{n}(M) = \begin{cases} f_{n}(1 - |M|^{2}/P), & J_{n} = J_{n} \\ f_{n}(1 + |M|^{2}/P), & J_{n} = J_{n} + 1 \end{cases}$$

(2.8)

We proceed now to analyze the absorption (amplification) spectrum of the test radiation. We assume that most values of $M$ satisfy the conditions of large field splitting, i.e., $|G| > \{f, f', f''\}$, and that Eqs. (2.3) and (2.4) hold for the individual three-level $M$-subsystems. We consider first the simplest case $\Omega = 0$, to which Eq. (2.4) with the substitution (2.6) corresponds. Thus, the resultant spectrum is a superposition of individual line pairs with different intensity and position in the frequency scale. According to (2.4), the distribution of the intensity among the lines is governed entirely by the function $f_{n}(M)$, i.e., it depends on the polarization of the test radiation and on the ratio of the angular momenta of the levels $m$ and $l$. The positions of the lines on the frequency scale relative to the point $\Omega_{2} = 0$ are set by the value of $|G| \sqrt{f(M)}$, i.e., they are connected with the polarization and intensity of the strong field and with the ratio of $J_{n}$ and $J_{n}$. The lines are distributed in the interval between the values $\Omega_{2} = 0$ and $\{|G| \sqrt{f(M)} \}$, depending on the polarization of the strong field and on the type of the $m-n$ transition. For linear polarization and for $\Delta_{n} = J_{n} - J_{n} = 0$, as well as for circular polarization and $\Delta_{n} = \pm 1$, the quantity $\{G \sqrt{f(M)} \}$ depends linearly on $M$ (see Table 1), the lines are therefore equidistant.

In those cases when $f(M) = \{1 - |M|^{2}/P\}$, the lines become strongly condensed on the edges of the spectral interval $\{|G| \sqrt{f(M)} \}$. This case is shown in Fig. 3. The relative intensity of the spectral components is unevenly distributed in all cases. In accordance with the different $f_{n}(M)$ for the different polarization states of the test field and different types of the $m-n$ transition, a greater weight is possessed either by components located closer to the line center (see Fig. 3a) or by those farthest from it (Fig. 3b). The positions of the components on the frequency scale are governed as before only by the characteristics of the strong field and of the $m-n$ transition. The qualitative results for $\Omega_{1} = 0$ remain in force also for $\Omega \neq 0$. Their main consequence is that both the distribution of the components over the spectrum and their relative intensities have an exceedingly strong dependence on the field polarizations and on the relations between $J_{n}$, $J_{n}$, and $J_{n}$. As a consequence, the form of the resultant spectrum undergoes appreciable modifications.

3. SHAPE OF THE PRINCIPAL PART OF THE SPECTRUM ENVELOPE

We assume that the natural width of the radiation line ensures overlap of the individual $M$-components of the spectrum. Then, in analogy with the known inhomogeneous-broadening situation, the resultant spectrum is described by a relatively smooth envelope. If the number of components is large enough $|J_{n}, J_{n}, J_{n}|$, the summation over $M$ in (25) can be replaced by integration. In the case of strong field splitting an integrand of the form (2.3) contains a "peaked" function of $M$ with smoothly varying parameters. Taking this circumstance into account and putting formally $\Gamma_{n} \rightarrow 0$ we can obtain the integration results in the form

$$P_{n} = \sum_{m} \Gamma_{n}(y_{0})^{1/2} \delta(y_{0}) C_{n}(y_{0}) \{1, 0, I, \}$$

(2.9)

$$D = \alpha_{n}$$

(3.1)

Here $N_{n}$ and $N_{nm}$ are the total population differences of the corresponding levels in the absence of radiation. The summation in (3.1) is over those values of $y_{0}$ for which the inequality

$$|G(y_{0})|^{2} = \Omega_{0} \Omega_{n} - \Omega_{nm} = \Omega_{0} \Omega_{n} - \Omega_{nm}$$

(3.2)

is satisfied. This relation specifies, in particular, the spectral-region boundaries in which $P_{n}$ differs substantially from zero. There are two such regions:

$$\Omega_{0} \Omega_{n} = \Omega_{nm}$$

(3.3)

Thus the envelope of the spectrum is also a doublet, but its components are to a certain degree "inhomogeneously" broadened.
We present for $P_\sigma$ a summary of the formulas, which corresponds to expression (3.1) under different polarization conditions and for different types of transitions.

For $A_{\sigma_1} = 0$ and $A_{\sigma_2} = 0$ we have

$$\Phi = \frac{1}{a} \left[ (t-u) |G(p)|^2 - 2u |G(p)|^2 (1 - 2u |G(p)|^2) \right].$$

(3.4)

For $A_{\sigma_1} = 0$ and $A_{\sigma_2} = \pm 1$

$$\Phi = \frac{1}{a} \left[ (t-u) |G(p)|^2 - 2u |G(p)|^2 (1 - 2u |G(p)|^2) \right].$$

(3.5)

For $A_{\sigma_1} = \pm 1$ and $A_{\sigma_2} = 0$

$$\Phi = \frac{1}{a} \left[ (t-u) |G(p)|^2 - 2u |G(p)|^2 (1 - 2u |G(p)|^2) \right].$$

(3.6)

And for $A_{\sigma_1} = 1$ and $A_{\sigma_2} = \pm 1$

$$\Phi = \frac{1}{a} \left[ (t-u) |G(p)|^2 - 2u |G(p)|^2 (1 - 2u |G(p)|^2) \right].$$

(3.7)

We have used here the symbol $u = \Omega_G (\Omega_2 - \Omega_1)$. The + sign corresponds to circular polarization of the strong field.

In all cases, with exception of the transitions $|A_{\sigma_1}| = 1$ and $|A_{\sigma_2}| = 1$, the value of $P_\sigma$ does not depend on the polarization of the test field.

Typical forms of the spectrum at $\Omega = 0$ are shown in Fig. 4. The spectrum is symmetrical about the point $\Omega_G = 0$ and has in many cases a doublet structure. The latter is absent in four cases, when the function $\Phi$ in (3.4)-(3.7) is proportional to $u^{-1/2}$ (see also Figs. 4c and 4h). The spectrum boundaries correspond to the maximum splitting effect for individual $M$-components and is located at $|\Omega_G| = |\Omega_2|/\sqrt{2}$. Figures 4c and 4b show that the field splitting effect can be quite pronounced even in the envelope of the spectrum, provided definite polarization conditions are satisfied. In this case $P_\sigma$ is proportional to $(1 - 2u |G(p)|^2)^{-1/2}$, which indicates a rapid growth of $P_\sigma$ when $|\Omega_G|$ approaches the edges of the contour, or even a divergence. The increase of the intensity on the edges of the contour is due to condensation of the $M$-components of the spectrum (see Fig. 3 and its discussion).

Divergence, on the other hand, sets in when $\Omega$ is neglected. We note that in all cases the area of the contours on Fig. 4 is the same. The reason is that the area of the contour of each individual $M$ component (the integral of expression (2.4) for $P_\sigma (M)$ with respect to $\Omega_G$) is determined only by the value of $|G(p)|^2$. Therefore the area of the integral contour depends in turn, by virtue of the properties of the Clebsch-Gordan coefficients, only on $|G(p)|^2$, i.e., on the intensity but not on the polarization state of the test field.

We consider now the more general case $\Omega \neq 0$. We note that at $\Omega \neq 0$ the terms proportional to $N_{\sigma_1}$ and $N_{\sigma_2}$ have substantially different frequency dependences and they must be considered separately. We agree to assign a plus sign to the first term $|N_{\sigma_1}| > 0$ and a minus sign to the second $|N_{\sigma_2}| > 0$. The corresponding spectra calculated from Eqs. (2.4) for $P_\sigma (M)$ with respect to $\Omega_G$ is determined only by the value of $|G(p)|^2$. Therefore the area of the integral contour depends in turn, by virtue of the properties of the Clebsch-Gordan coefficients, only on $|G(p)|^2$, i.e., on the intensity but not on the polarization state of the test field.

FIG. 5. Spectra at $\Omega \neq 0$ in the scale $|\Phi G(p)|^2/|G(p)|^2 |N_{\sigma_1} - N_{\sigma_2} G(p) + \Omega_1 + \Omega_2|$. a) $A_{\sigma_1} = 0, A_{\sigma_2} = 1, \pm 1$; b) $A_{\sigma_1} = A_{\sigma_2} = 1$; c) $A_{\sigma_1} = 0, A_{\sigma_2} = 1, \pm 1$; d) $A_{\sigma_1} = 1, A_{\sigma_2} = 0, \pm 1$; e) $A_{\sigma_1} = 1, A_{\sigma_2} = 0, \pm 1$; f) $A_{\sigma_1} = 1, A_{\sigma_2} = 0, \pm 1$; the solid and dashed lines show the contours for coinciding and orthogonal polarizations of the test field.
The asymptotic values of $g(x)$ and $h(x)$ at $x \to -\infty$ are described by rather unwieldy expressions which will not be given here. We note only that if the spectral intervals receive comparable contributions from all the $M$-components of the corresponding doublet component. For this reason the line wing also decreases quite slowly (slower than $|\Delta \Omega_n|^3$).

If $\Omega = 0$ and there is no doublet splitting of the line (Figs. 4b, 4c, 4b), the region of $\Omega_n = 0$ calls for a special analysis. Indeed, this spectral region is formed by $M$-components for which the field splitting is small $|G(M)|$ smaller than or comparable with the relaxation constants. Consequently the approximate formula (2.3) is not applicable here. It is easy to verify on the basis of this result that following a shift within the spectral line $|x| > 0$ the asymptotic behavior $|x| < 0$ is described by rather unwieldy expressions which will not be given here. We note only that if the spectral intervals receive comparable contributions from all the $M$-components of the corresponding doublet component. For this reason the line wing also decreases quite slowly (slower than $|\Delta \Omega_n|^3$).

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5. ELLIPTIC POLARIZATION OF STRONG FIELD

The rigorous quantum-mechanical solution of the problem becomes exceedingly more complicated if the strong field is elliptically polarized. At large value of the quantum numbers $J_{\ell m}$ and $J_{\ell'} m'$, however, a classical description of the orientation of the angular momenta is found to be effective. In other words, one introduces the elements of the density matrix $\rho_{ij}(s)$, where $s$ is a unit vector in the angular-momentum direction. As shown in Ref. 4, $\rho_{ij}(s)$ satisfies exactly the same equations as the elements $\rho_{ij}$ in the model of nondegenerate states. The only formal difference lies in the additional (parametric) dependence of $\rho_{ij}$ on $s$. This dependence is due to the $G(s)$ and $G_{ij}(s)$ dependences, for which the following equations are valid

$$G(s) = \sum G_{ij} D_{ij}^m(\theta, \phi, 0), \quad G = E_d d_m / 2(2J + 1)^{3/2},$$

$$G_{ij}(s) = \sum G_{ij} D_{ij}^m(s), \quad G^* = E_d d_m / 2(2J + 1)^{3/2}. \quad (5.1)$$

Here $E_d$ and $E^*_d$ are the circular components of the electric vectors of the fields; $\theta$ and $\phi$ are the azimuthal and polar angles of the vector $d$, and $D_{ij}^m$ is the Wigner rotation matrix.

The work of the test field is given by the natural expression

$$P_l = J \int_{\cos \theta_1}^{\cos \theta_2} \rho_{ij}(s) ds,$$ (5.2)

where Eq. (2.1) remains valid for $P_l(s)$, recognizing that now $G$ and $G_{ij}$ depend on $s$ in accordance with (5.1).

The summation over the discrete variable in (2.5) is replaced in (5.2) by integration. This means that the line contour described by (5.2) is directly a smooth envelope. It is clear therefore that from the viewpoint of the problem considered the classical description of the angular-momentum orientation contains automatically the assumption that neighboring elements of the spectrum components overlap. It is assumed that $P_l(s)$ does not depend on the angle $\theta$ and it is recognized that $\cos \theta = M / J_i$, then Eq. (5.2) is a transition from summation in (2.5) to integration.

We shall assume that the condition $|G(s)| \Gamma_{\ell m} \Gamma_{\ell'} m'$ is satisfied in the entire range of variation of $\theta$ and $\phi$. It is then possible to use the approximation (2.3) and, in addition, integrate explicitly in (5.2) with respect to one of the variables (say, $\theta$). We then obtain (cf. (3.1))

$$P_l = \frac{D}{2 \cos \theta_1} \sum_{j} \left| G_y(\phi, \psi) \right|^2 \left| \frac{d}{d\psi} G_y(\phi, \psi) \right|^2 d\psi,$$

$$y = \cos \theta, \quad G_y(\psi, \phi) = G_y(s), \quad G^* = G(s), \quad (5.3)$$

where the summation is over values of $\psi$ satisfying the equation $|G_y(\phi, \psi)|^2 = 0$ on $G(s) = 0$.

The factor $D$ is described by Eq. (3.1) as before.

Subject to the conditions under which the problem was solved in the preceding sections, we can choose a coordinate frame in which the functions $G_0(\phi, \psi)$ and $G(\phi, \psi)$ do not depend on $\psi$. In this case, as should be the case, Eq. (5.3) coincides with (3.1).

It will be more convenient in the analysis that follows to integrate in (5.2) with respect to the angle $\theta$. In the coordinate system where the propagation directions of the strong and test waves coincide with the $x$ axis we obtain

$$P_l = \frac{D}{\sin \theta} \int \rho_{ij}(\theta, \phi, 0) \left| \frac{d}{d\phi} G_y(\phi, \psi) \right|^2 d\phi,$$

$$x = \cos \theta, \quad G_y(\theta, \phi) = G_y(s), \quad G^* = G(s), \quad (5.4)$$

where $\rho_{ij}(\theta, \phi, 0) \Gamma_{\ell m} \Gamma_{\ell'} m'$ is the circular component of the electric vector of the field $\rho_{ij}(\theta, \phi, 0)$. It is then possible to use the approximation (5.2) and, in addition, integrate explicitly in (5.2) with respect to one of the variables (say, $\theta$). We then obtain (cf. (3.1))

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$$y = \cos \theta, \quad G_y(\phi, \psi) = G_y(s), \quad G^* = G(s), \quad (5.5)$$

where the summation is over values of $\psi$ satisfying the equation $|G_y(\phi, \psi)|^2 = 0$ on $G(s) = 0$.

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The factor $D$ is described by Eq. (3.1) as before.

Subject to the conditions under which the problem was solved in the preceding sections, we can choose a coordinate
The levels manifests itself quite unusually. First, the components of the known doublet are subject to a strong-field polarization-ellipse major axis. In contrast to the cases of linear and circular polarizations of the strong field (see Figs. 5b and 5f), an abrupt increase of the intensity is observed here not at the ends of the lines, but in the internal regions. Consequently, when the strong-field polarization ellipse is smoothly varied from linear to circular, a smooth displacement of the sharp peak should be observed from one edge of the doublet component to the other.

6. CONCLUSION

The analysis in the present paper shows that in systems with large rotational quantum numbers the field splitting of the levels manifests itself quite unusually. First, the components of the known doublet are subject to a strong "inhomogeneous" broadening. Sometimes the doublet structure disappears completely. In the cases when it is preserved, each of the components is represented by an envelope that occupies a spectral region that exceeds considerably the natural line width.

The shape of the doublet components depends very strongly on the polarization conditions and on the type of transition. In some cases the intensity increases sharply on the edges or in the internal parts of the lines. The characteristic scale of the corresponding peaks is governed by the value of Γ.

Despite the large broadening of the components, the spectrum retains information on the natural width. Besides the already noted peak width, this information is contained in the abrupt edges of the lines. In addition, in some cases (at J = 0) in the vicinity of the spectral line (D_m = 0) additional peaks or dips can arise having a natural width and amplitude that depends on the ratio of the relaxation constants.

To be able to observe the considered effects in experiment, for example in vibrational-rotational transitions of molecules, the following conditions must be satisfied. First, the absorption line must be sufficiently isolated, i.e., the distance to the other lines should exceed substantially the natural (in most cases, Doppler) width. Second, the resonant-radiation intensity must be high enough to cause field splitting on individual M-components; the splitting must exceed the Doppler width, but not be high enough to excite higher vibrational states. These conditions can be satisfied for a large class of relatively simple molecules, at least for almost all diatomic molecules. To be specific, we present the following estimates for HF, a typical working gas in chemical lasers. The rotational constant for HF is ~6 × 10^{-11} Hz, and the line shift due to anharmonicity is ~3 × 10^{11} Hz (Ref. 7). Recognizing that the Doppler width for the vibrational transitions of HF is ~10^{-6} Hz, we verify that the absorption spectrum of HF is very sparse. Using the data of Ref. 8 on the value of the Einstein coefficient A for an individual rotational transition of HF (J = 2 × 10^6 sec^{-1}), we find that the field splitting exceeds by 10 times the Doppler width at a radiation intensity ~10^7 W/cm². This by far not the record value of the HF laser radiation intensity.

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