Structure of quasi-energy spectrum of hydrogen in a microwave field; theory and experiment

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Experimental and theoretical investigations are reported of the absorption spectra of a hydrogen plasma near the D_α line in microwave fields of frequency 2.94, 9.1, and 40.5 GHz. The use of an intracavity laser spectrometer with high spectral and temporal resolution has made it possible to track the change of the form of the absorption profile of the D_α line as a function of the intensity and frequency of the field in two cases; the field frequency exceeds appreciably or is comparable with the fine splitting of the levels of the D_α transition. In particular, an energy shift of the atomic levels was recorded in an alternating electric field. Comparison of the experimental results with calculations in the framework of the quasi-energy method has shown that for an adequate interpretation of the experiments it is necessary to take into account the fine structure of the hydrogen energy levels.

1. INTRODUCTION

Research into the influence of oscillating electric field on the spectral characteristics of atomic systems is attracting at present great interest, primarily as applied to diagnostics of electric fields in a plasma. More accurate and subtle experimental methods are being developed, chracterized by high sensitivity, high temporal, spatial and spectral resolutions, and a large volume of information that calls for higher automation of the gathering and reduction of the experimental data. Great promise in this direction is offered by the method of intracavity laser spectroscopy,[1,2] which permits high-accuracy investigation of the spectra of laser-radiation absorption by hydrogen and deuteron atoms placed in a microwave electric field.[3,4] These atoms are almost always present in the plasma as impurities or as technological additives. The attained experimental accuracy and the use of modern data reduction methods uncovers extensive possibilities for measuring the parameters of electric-field oscillations in a plasma.

At the same time, if the accuracy of the experimental data is to be increased, more stringent requirements must be met by the employed theoretical premises. Simulation of an atom by a two-level system and neglect of the fine structure of the hydrogen terms[5–7] can no longer provide an adequate interpretation of the observation results.

We report here an experimental and theoretical investigation of the absorption spectra near the α lines of the hydrogen and deuterium Balmer series in microwave fields of various frequencies. We justify the need for taking into account the fine structure of the hydrogen levels, and propose a method of calculating the spectral characteristics in intense fields, when a substantial role is played by multiphoton processes. We discuss the experimental results of investigating the distortion of plasma spectra at various ratios of the field frequency Ω to the line width. Comparison of the calculation results with experiment reveals a number of heretofore unknown singularities in the dependences of the spectral characteristics on the amplitude and frequency of the field. In particular, a decrease of the distance between the maxima of the fundamental line, due to the shift of the fine-structure levels by the microwave field, has been observed for the first time. We show also that in the case of sufficiently strong fields the intensity ratio of the first satellites becomes dependent on the amplitude of the field strength.

In Sec. 2 we describe a method of calculating the quasi-energies and the quasi-energy states of a one-electron atom, with allowance for the fine structure of the levels. Section 3 contains the results of experiments on the α line of the Balmer series in microwave fields of various frequencies. These frequencies can be both higher than or comparable with the fine splitting of the levels. In Sec. 4 we demonstrate the good agreement between the results of calculations by the procedure of Sec. 2 and the experiments. Section 5 is devoted to a discussion of the possibility of using the regularities observed by us for diagnostics of electric fields in a plasma. The main deductions of the work are formulated in the Conclusion.

We use in equations a system of units in which $\hbar = m = c = 1$ (m is the electron mass).

2. NEED FOR ALLOWANCE FOR THE LEVEL FINE STRUCTURE IN CALCULATIONS OF THE HYDROGEN QUASI-ENERGY SPECTRUM

The spectrum of hydrogen in an oscillating electric field having a cyclic frequency Ω and a peak intensity E_Ω is known to be defined as the solution of the nonstationary Schrödinger equation with a Hamiltonian that depends periodically on the time

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r},t)=[\hat{H}_0(\mathbf{r})+V_0\cos\Omega t]\Psi(\mathbf{r},t). \qquad (1)$$

Here $\hat{H}_0$ is the zeroth-approximation Hamiltonian (in the absence of an oscillating field); for the case considered below, of a linearly polarized electric field directed along the z axis, $V_0 = ezE_\Omega$.

It is convenient to seek a solution of (1) by the method of quasi-energies (QE) and quasi-energy states (QES) developed in Refs. 8 and 9. In investigations of the behavior of hydrogenlike systems in external electric fields, the customarily employed zeroth approximation was a solution of the nonrelativistic Schrödinger equation for an electron in a Coulomb field (see, e.g., Refs. 7 and 10) (the energy levels

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are then $2n^2$-fold degenerate, where n is the principal quantum number). This seemed quite natural, since the level fine splitting due to the spi-orbit interaction is small for hydrogen and plays as a rule a noticeable role only in the case of multiply charged hydrogenlike ions, where the relativistic effects are altogether quite substantial (see, e.g., Ref. 11). Thus, in the case of a constant electric field, the fine splitting $\Delta E_{fs}(n)$ of the level n of a one-electron ion with nuclear charge z can be neglected if it is much smaller than the level splitting due to the electric field, i.e., for strong enough fields of intensity F (in a.u.) that satisfies the condition $F \gg \alpha^2 z^5/n^5$. A linear Stark effect is observed in such fields. Corresponding to lower field strengths is the quadratic Stark effect, which goes over gradually into linear one with increase of intensity.[11,12] As shown in Ref. 13, the line profile calculated with allowance for the fine splitting of the levels differs in this case substantially from the results of the nonrelativistic analysis.

In the case of alternating external field, allowance for the fine splitting of the levels is necessitated not only by the value of the intensity amlitude E_Ω, but also by the frequency Ω of the field. If this frequency is much lower than the spacing between the levels (and is, of course, not resonant), while the field is weak enough, each level of a multiply charged ion can be regarded as isolated, and admixtured to it from all the remaining levels can be accounted for by perturbation theory.[14] When Ω is of the order of $\Delta F_{fs}(n)$, the influence of the field becomes more complicated. For the states of the first excited configuration of multiply charged ions with different nuclear charges, this question was considered in Ref. 15. It was shown in particular, with ions having $z = 20$ as the example, that the arrangement of the QE levels has a complicated dependence on the frequency and intensity amplitude of the field. The coefficients of the corresponding QES also depend on the frequency and intensity amplitude in a more complicated manner than in the case of degenerate hydrogen-atom levels.[6] This alters in turn the dependence of the character of the dependence of the transition probabilities for the satellite lines.[16] Thus, in a study of the spectral characteristics of multiply charged one-electron ions and intense external fields, there is undisputed need for taking into account relativistic effects, primarily the splitting of the fine structure of the levels.

We return now to the hydrogen atom. Note that the influence of fields of frequency $\sim 10^6$ GHZ on the fine structure levels of an ion with $Z = 20$ was considered in Ref. 15. The fine-structure splitting, as is well known, varies in proportion to Z^4, so that if we study the effect of microwave fields of frequency 1–100 GHz on a hydrogen atom, the external-field frequency and the fine-structure splitting of the hydrogen levels are related approximately in the same manner as in the case of multiply charged ions.[15] Thus, it is natural to expect neglect of the fine structure of the hydrogen levels in a microwave field to lead to an incorrect description of the observed phenomena. Indeed, in Ref. 3 was observed a distinct asymmetry of the intensities of the "red" (Stokes) and "blue" (anti-Stokes) first satellites of the Balmer-series α line in a field $\Omega/2\pi = 49.5$ GHz, whereas theoretical calculations in the degenerate-levels approximation yield the same intensity for both satellites. Note, incidentally, that details of the fine structure of the satellites were also observed in Ref. 3.

Thus, the foregoing theoretical arguments, and the experiments already performed, point quite convincingly to the need for taking into account relativistic effects in the study of the influence of microwave fields on the hydrogen atom. Consequently, $\hat{H}_0(\mathbf{r})$ in Eq. (1) must be chosen to be the Dirac Hamiltonian for an electron in the Coulomb field of the nucleus. The solution of (1), as usual in the quasi-energy method, is sought in the form of the series

$$\Psi_\varepsilon(\mathbf{r}, t) = e^{-i\varepsilon t} \sum_{p=-\infty}^{\infty} \varphi_p(\mathbf{r}) \exp(-ip\Omega t), \tag{2}$$

where ε is the quasi-energy. The radial functions $\varphi_p(\mathbf{r})$ will be sought, in turn, in the form of expansions in the basis of the Dirac wave functions ψ_{njlm}:

$$\varphi_p(\mathbf{r}) = \sum_{njlm} a_{njlm,p} \psi_{njlm}(\mathbf{r}). \tag{3}$$

The problem reduces then to finding the expansion coefficients $a_{njlm,p}$ which are the roots of an infinite system of homogeneous linear equations

$$(\varepsilon - \varepsilon_{nj}{}^0 + p\Omega) a_{njlm,p} = \sum_{n'j'l'm'} V_{njlm}^{n'j'l'm'} (a_{n'j'l'm',p-1} + a_{n'j'l'm',p+1}), \tag{4}$$

where $V_{njlm}^{n'j'l'm'}$ is a matrix element of the operator V_0 from (1). If the external field is linearly polarized (this is just the case considered hereafter), $V_{njlm}^{n'j'l'm'}$ reduces simply to a matrix element of the coordiate z:

$$V_{njlm}^{n'j'l'm'} = {}^1/_2 e E_\Omega z_{njlm}^{n'j'l'm'}. \tag{5}$$

General expressions for the matrix element $Z_{njlm}^{n'j'l'm'}$ between the Dirac functions are given, e.g., in Ref. 11. It must be noted, however, that each matrix element is a sum of two terms, the first is the matrix element between the upper components of the Dirac bispinor, and the second between the lower ones. Since the second term is of order $(\alpha Z)^2$ relative to the first, it can be discarded in the case of the hydrogen atom, in contrast to multiply charged ions.[15] We can therefore use the expression from Ref. 17 for the matrix element $Z_{njlm}^{n'j'l'm'}$ in the case of hydrogen.

The general equation (4) is valid at arbitrary frequency or field amplitude. If, however, the field amplitude E_Ω is small compared with the atomic field $E_{at} = 5.14 \cdot 10^9$ V/cm, and the frequency is much lower, as in the case of microwave fields, than the optical frequencies $\omega_{nn'}$ corresponding to transitions with change of the principal quantum number n, we can confine ourselves in the derivation of the function $\varphi_\beta(\mathbf{r})$ [Eq. (3)], in the lowest order of perturbation theory in the parameter E_Ω/E_{at}, to summation over all the eigenfunctions with specified value of the principal quantum number. Mixing of states with different n and n' is significant only in the next higher orders of perturbation theory (this question was investigated for the case of multiply charged one-electron ions in Ref. 18, where it was shown that the role of the higher orders of perturbation theory increases with increase of the nuclear charge Z).

The spectrum of a real atom in an alternating electric field is a superposition of all transitions between the QES of the entire multiplet. In investigations of specific spectral

lines of a hydrogen plasma it is therefore necessary to take into account all the singularities that result from the fine splitting of both the lower level and the upper one. For arbitrary values of the frequency Ω and of the field intensity amplitude E_Ω, the system (4) can be solved only numerically, and to ensure the required calculation accuracy it is necessary to take into account in (2) a sufficiently large number of harmonics. The results of calculations carried for the α line of the Balmer series, and their comparison with experiment, are discussed in Sec. 4. below.

3. EXPERIMENTAL INVESTIGATION OF THE SHAPE OF THE α LINE OF THE BALMER SERIES IN MICROWAVE FIELDS OF VARIOUS FREQUENCIES

The investigations were carried out in a pulse discharge plasma in a mixture of deuterium and hydrogen with a total pressure 0.5 to 2 Torr. The experimental setup is described in Ref. 3. The charge density and the gas pressure were in all cases such that the lines were broadened mainly by the Doppler mechanism, the width being 0.3–0.4 cm^{-1} and 0.2–0.3 cm^{-1} for the hydrogen and deuterium atoms, respectively. The plasma was produced in sealed glass tubes inserted in a waveguide and placed inside the cavity of a pulsed tunable multimode laser based on solutions of organic dyes in ethanol and pumped by a lamp. The lasing spectrum (width $\sim$10nm, pulse duration 1–2 μs) was analyzed by an double dispersion autocollimation spectrograph (DAS) having a focal length 1300 and a resolution $\lambda/\delta\lambda \sim 3\cdot 10^5$.

The absorption-line shape was investigated at three different field microwave frequencies: $\nu_1 = 2.94$ GHz, $\nu_2 = 9.1$ GHz, and $\nu_3 = 40.5$ GHz. The first is of the order of the splitting of the fine structure of the upper level ($n = 3$), the second of the order of the fine structure of the lower level ($n = 2$), and the third exceeds the level multiplet splitting by several times. The experiments at frequencies ν_1 and ν_2 were performed with magnetron oscillators. The power of the low-frequency 2.94-GHz magnetron was 600 kW, ensuring a field strength up to 6–7 kV/cm inside the waveguide (in the absence of plasma). In the case of ν_2, however, the field intensity in the cold waveguide did not exceed 3 kV/cm. In the investigations at 40.5 GHz we used a gyrotron and the electromagnetic wave was focused through a quasi-optical line and a teflon lens into the plasma volume, so that a value of E_Ω up to 6 kV/cm could be obtained. In all cases, the microwave polarization direction coincided with that of the laser radiation.

The investigations revealed a fundamental difference between the absorption spectra in the three considered cases. Thus, at the frequency $\nu_1 (1\lambda_1 = 0.098\ cm^{-1})$ close to the splitting of the upper level $\Delta E_{1/2\ 3/2}(n = 3) = 0.108\ cm^{-1}$, not only is the absorption line noticeably broadened, but an additional structure is produced.[19] A typical spectrogram obtained by intra-cavity laser spectroscopy (ICLS) is shown in Fig. 1a. The fine details of the structure are distinctly observed, but the spectral interval between the absorption maxima is not equal to (nor is it a multiple of) 0.1 cm^{-1}, but always exceeds the multiplet splitting of the lower level. The overall line broadening is appreciable (by more than a factor of two).

An unexpected change occurs in the character of the spectrum in a field of frequency ν_2 $(1/\lambda_2 = 0.304\ cm^{-1})$. A typical ICLS plot is shown in Fig. 1b. In particularly, the

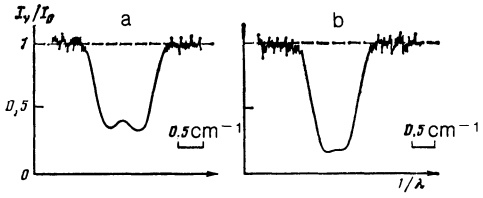

FIG. 1. ICLS plots of plasma in a microwave field: a—frequency 2.94 GHz, b—9.1 GHz.

multiplet structure, which is clearly observed both in the absence of a microwave field and at high frequencies of the field,[3] practically vanishes. The line broadens, but not as much as in the first case.

Finally, at the frequency ν_3, which exceeds the multiplet splitting of the levels, it is possible to record reliably satellites at distances $\pm\Omega$ from the H_α line.[2,3] An increase of the sensitivity of the intracavity laser spectroscope (or of the density of the absorbing atoms) permits also observation of satellites at distances $\pm 2\Omega$. A detailed study of the corresponding experimental results may reveal new features (compared with those noted in Ref. 3).

First, it was observed that turning on a 40.5 GHz microwave field shifts the atomic terms—the energy gap between the fine-structure levels $2s_{1/2}$, $2p_{1/2}$ and $2p_{3/2}$ decreases (see Sec. 4 below). This follows from the decrease of the measured value of the spectral distance Δ between the maxima of the absorption coefficient of the D_α line in the presence of a microwave field (averaged over 20 realizations of the spectrograms). A comparison was made with the analogous value in the absence of a field (9 realizations). It turned out that both sets of experimental data have near-normal distributions. Their reduction in accordance with the general rules of statistical analysis[20] has shown that $\Delta_1/2\pi c = 0.319 \pm 0.010\ cm^{-1}$, $n_1 = 9$, $s_1 = 0.023\ cm^{-1}$ in the case $E_\Omega = 0$ and $\Delta_2/2\pi c = 0.295 \pm 0.005\ cm^{-1}$, $n_2 = 20$, $s_2 = 0.016\ cm^{-1}$ in the case $E_\Omega \neq 0$ (here n is the size of the selection, and s is the selection standard). Comparison of s_1^2/s_2^2 with the Fisher quartiles shows that the difference between the selection standards is statistically insignificant with a significance level 0.05. It is therefore possible to compare the obtained average values of Δ_1 and Δ_2 and conclude that Δ is decreased in the presence of microwaves.

Second, we measured the ratio of the integral absorption coefficients of the anti-Stokes ($\varkappa^+$) and Stokes ($\varkappa^-$) satellites. To this end, the experimental values of I_ν/I_0 (I_ν is the time-integrated lasing intensity at the frequency ν in the presence of selective losses, and I_0 is the intensity outside the absorption lines) were used to calculate the effective absorption coefficient

$$\gamma_{\text{eff}} = \varkappa_\nu lc\tau/L. \qquad (6)$$

Here l and L are the lengths of the absorbing layer and of the cavity, and τ is the laser-pulse duration. The integral $\int \gamma_\nu^{\text{eff}} d\nu$ was next calculated. The details of the reduction of the ICLS plots and the possible measurement errors are discussed in detail in Ref. 21. The electromagnetic field causes an increase of atoms absorbing in the state $n = 2$. The experiments were performed at different values of the discharge current. The values of γ_{eff} varied therefore in different realizations. The quantity $\varkappa^+/\varkappa^-$ was determined as the coeffi-

cient of linear regression of $\varkappa^+$ on $\varkappa^-$ over 14 realizations of $\gamma_{\text{eff}}^{\pm}$ and found to be 1.17 ∓ 0.08.

Clarification of the gist of the physical processes that lead to the observed effects calls for a thorough theoretical analysis. In the next section we use for this analysis the calculated QE and QES of hydrogen and deuterium with allowance for the fine splitting of levels, using the procedure of Sec. 2.

4. CALCULATION RESULTS AND COMPARISON WITH EXPERIMENT

Computer solution of the system (4) yielded the QE values and the QES wave functions:

$$\Psi_{n,m}^{(k)}(\mathbf{r},t)=\exp\{-i\varepsilon_{n,m}^{(k)}t\}\sum_{j,l}\sum_{p=-N}^{N}a_{njlm,p}^{(k)}\psi_{njlm}(\mathbf{r})\exp\{-ip\Omega t\} \tag{7}$$

for the lower ($n=2$) and higher ($n=3$) states of the α lines of the Balmer series in hydrogen and deuterium. Note that since the microwave field is linearly polarized, the projection of the total angular momentum is conserved, and to find the QES wave functions (3) it is necessary to take into account the mixing of the Dirac functions with identical values of m, a circumstance taken into account in (7). The state with maximum value of m for a given n ($|m|=\frac{5}{2}$ for $n=3$ and $|m|=\frac{3}{2}$ for $n=2$) is not changed by the microwave field. Thus, to find the functions (7) it is necessary to solve in the case $n=2$ the two-level problem ($|m|=\frac{1}{2}$) and in the case $n=3$ the three-level ($|m|=\frac{3}{2}$) and five-level ($|m|=\frac{1}{2}$) problems, with account taken of a sufficiently large number of harmonics (to ensure a correct calculation of the coefficients $a_{njlm,p}^{(k)}$ in (6)). In actual calculations, N is chosen such that its further increase produces practically no change of those coefficients that must be taken into account in the calculation of the absorption cross section. Obviously, N depends on the field parameters, increasing with increase of E_Ω and with decrease of Ω.

The square of the dipole moment of the transition between the states $\psi_{2,m}^{(i)}$ and $\psi_{3,m'}^{(k)}$ is of the form

$$|D_i^k(m,m')|^2=\sum_q|D_i^k(m,m';q)|^2 g(\varepsilon_{3,m'}^{(k)}-\varepsilon_{2,m}^{(i)}+q\Omega-\omega). \tag{8}$$

Here $|D_i^k(m,m';q)|$ determines the absorption and the satellite frequencies shifted by $q\Omega$ from the ground line, and $g(\nu)$ is the spectral function. Since the polarizations of the laser and microwave fields had the same direction in the experiments described above, contributions to the observed absorption are made only by transitions with $\Delta m=0$. Consequently, the calculations using Eq. (8) must be carried out in two cases: $m=m'=\pm\frac{3}{2}$, $m=m'=\pm\frac{1}{2}$.

If $i=1$ is assumed to number the state $\psi_{2\,\frac{3}{2}\,\frac{3}{2}}$, we can write in the case $|m|=\frac{3}{2}$ (with allowance for summation over the momentum components $\pm m$)

$$|D_i^k(q)|^2=2\left|\sum a_{3j2\frac{3}{2},q}^{(k)}z_{2\frac{3}{2}i\frac{3}{2}}^{3j2\frac{3}{2}}\right|^2. \tag{9}$$

For transitions between states with $|m|=\frac{1}{2}$, the dipole matrix element takes the form

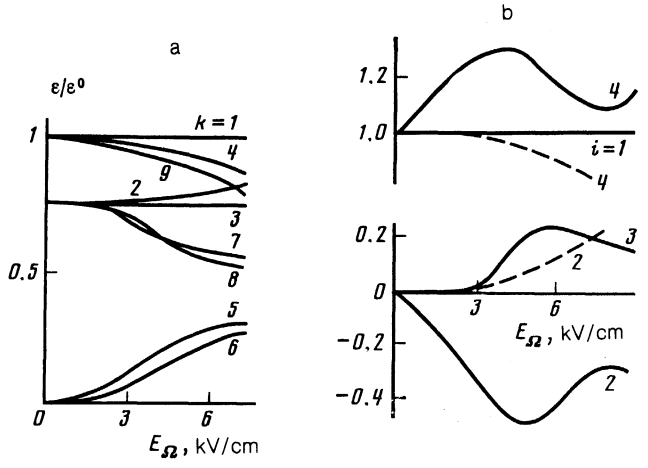

FIG. 2. Quasi-energy states: $a-n=3$ ($1-d^{5/2\,5/2}$, $2-p_{3/2\,3/2}$, $3-d_{3/2\,3/2}$, $4-{}_{5/2\,3/2}$, $5-s_{1/2\,1/2}$, $6-p_{1/2\,1/2}$, $7-p_{3/2\,1/2}$, $8-d_{3/2\,1/2}$, $9-{}_{5/2\,1/2}$); $\varepsilon^0=\varepsilon_{3,5/2}-\varepsilon_{3,1/2}$; $b-n=2$ ($1-p_{3/2\,3/2}$, $2-s_{1/2\,1/2}$, $3-p_{1/2\,1/2}$, $4-p_{3/2\,1/2}$) in a field of frequency 4.5 GHz (solid curves) and 9, 1 GHz) (dashed); $\varepsilon^0=\varepsilon_{2,3/2}^0-\varepsilon_{2,1/2}^0$

$$|D_i^k(q)|^2=2\left|\sum_p\sum_{jl}\sum_{j'l'}a_{2jl\frac{1}{2},p}^{(i)}a_{3j'l'\frac{1}{2},p+q}^{(k)}z_{2jl\frac{1}{2}}^{3j'l'\frac{1}{2}}\right|^2. \tag{10}$$

The contour to be observed is determined by a convolution of the spectral components with a Gaussian contour.

We proceed now to describe the actual calculation. We consider first the case $\nu_3(1/\lambda_3=1.35\ \text{cm}^{-1}$. Figure 2a shows the QE values of states $n=3$ as functions of the field amplitude E_Ω, while Fig. 2b (solid lines) shows the values for the states $n=2$. in both cases the energy is reckoned from the Dirac level $\varepsilon_{n,1/2}^0$ and the ordinate is $\varepsilon/(\varepsilon_{nj=n-1/2}^0-\varepsilon_{n,1/2}^0)$. As seen from Fig. 2a, all the solutions of the five-level problem ($k=5$ to 9) are complicated functions of the field strength, and it is quite obvious that this problem cannot be replaced by a simpler one. At the same time, the locations of the levels $k=2$, 3, 4 on Fig. 2a and of $i=2$, 3, 4 (solid line) in Fig. 2b might offer hope that the two-level approximation suffices for a description of the states $3d_{5/2\,3/2}$, $3p_{3/2\,3/2}$ and $2p_{3/2\,1/2}$, $2s_{1/2\,1/2}$ in the considered range of E. However, even though the levels $3d_{3/2\,3/2}$ and $2p_{1/2\,1/2}$ are shifted by the field quite insignificantly, the corresponding wave functions (7) differ noticeably from the results of calculations in the two-level approximation, and this is directly reflected in the values of the dipole matrix elements (9) and (10).

As seen from the arrangement of the curves in Figs. 2a and 2b, the relative shift of the QE decreases with increase of E_Ω, and it is this which decreass the spectral interval, described in Sec. 3, between the fine-structure lines. Comparison of the calculation and experiment yields an estimate of the electric field intensity amplitude, 3.2–3.9 kV/cm. This result agrees with measurements, performed under similar conditions, of the local values of E_Ω by the resonant laser fluorescence method.[22]

Figure 3 shows the calculated ratio $\sigma^{(\pm1)}/\sigma^0E_\Omega^2$ (E_Ω is in kV//cm) vs E_Ω ($1-\sigma^{(-1)}$, $2-\sigma^{(+1)}$). The dashed lines correspond to the weak-field-approximation calculation used to interpret the results in Ref. 4 and described in detail in Ref. 23. The weak-field region, as seen from the figures, is determined by the condition $E_\Omega\leqslant1$ kV/cm. In stronger

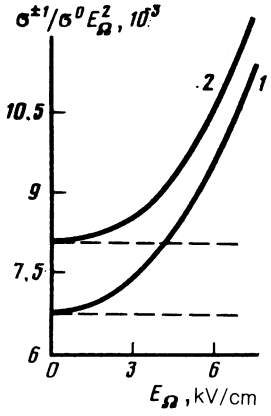

FIG. 3. Ratio of the cross sections one one- and two-photon transitions in a 40.5 GHz field: $1-\sigma^{(-1)}$, $2-\sigma^{(+1)}$.

fields the ratio $\sigma^{(\pm 1)}/\sigma^0$ increases more rapidly than E_Ω^2. In addition, in fields that are strong enough the relative intensity of the first satellites also depends on E_Ω. Indeed, in weak fields the intensities of the two first satellites are proportional to E_Ω^2 (Refs. 14, 16, 23) and their ratio, which differs from unity, is naturally independent of the field strength. When the field strength becomes so large that the Stark splitting of the levels exceeds considerably their fine structure, the latter can be neglected and the results obtained for degenerate levels[6] can be used. The Stokes and anti-Stokes satellites are in this case symmetric and their intensity ratio, equal to unity, is likewise independent of the field strength. Between the two indicated limiting cases there exists an appreciable region of fields in which the ratio of the intensities of the first satellites depends on E_Ω in a complicated manner.

The procedure proposed in Sec. 2 permits a calculation, in a unified manner, of the intensities of not only the first but also of the succeeding satellites. Figure 4 shows the calculated ratio of the absorption cross sections of the second and first satellites (likewise divided by E_Ω^2, where E_Ω is in kV/cm). In the next section we shall discuss the possibilities of using these results for more precise plasma diagnostics.

Similar calculations were performed for the cases $\nu_1(1\lambda_1 = 0.098\ \text{cm}^{-1})$ and $\nu_2(1\lambda_2 = 0.304\ \text{cm}^{-1})$. By way of example, Fig. 2b (dashed lines) shows the QE of the lower state of ν_2. The calculations have shown that as Ω decreases the number of harmonics that must be taken into account

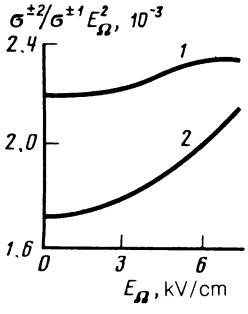

FIG. 4. Ratio of cross sections of three and two-photon transitions in a 40.5 GHz field: $1-\sigma^{(-1)}$, $2-\sigma^{(+1)}$.

increases, so that the observed contour is determined by a convolution of many dozens of spectral components. The set of absorption contours calculated for these fields at different E_Ω is given in Ref. 23. We did not succeed in discerning in the spectral line shape any universal parameters that depend on Ω or E_Ω and whose simultaneous measurement would permit a determination of this quantity from the experimental data without other *a priori* information. The dependence of the line shape on Ω and E_Ω is quite complicated. Thus, in a 2.94-GHz field the spectral distance between the maxima increases with increase of E_Ω, and additional extrema appear at sufficiently large E_Ω. In the case of 9.1-GHz field, this parameter is nonmonotonic: in weak fields (up to $E_\Omega \sim 2$ kV/cm) the spectral interval decreases while the line width is simultaneously increased, after which the interval begins to increase. Thus, the only possibility of checking on the calculations is to compare the experimental $\gamma^{\text{exp}}(\nu)$ for a known line-profile frequency with analogous theoretical $\gamma^{\text{theor}}(E_\Omega, \nu)$.

In such a comparison there exists, generally speaking, a leeway in the choice of three parameters: the field amplitude E_Ω, the frequency shift $\Delta\nu_0$ relative to some arbitrarily chosen frequency ν_0, and the measured experimental absorption cross section (or effective coefficient). One must not forget that the spectral function $g(\nu)$ in (8) is, in the language of probability theory, none other than the probability distribution density of the absorption at the frequency ν. It is meaningful therefore to compare spectral contours normalized by the condition

$$\int_{-\infty}^{\infty} g(\nu)\, d\nu = 1.$$

This obviates the need for choosing a third parameter.

Experimental results similar to those shown in Fig. 1 were used to calculate the quantities[21]

$$\gamma^{\text{exp}} = \frac{I^\nu}{I^0} = \frac{1-\exp(-\gamma_{\text{eff}})}{\gamma_{\text{eff}}}.$$

The effective absorption coefficient γ_{eff} is defined in Eq. (6). Numerical values of $\gamma_i^{\text{exp}} = \gamma^{\text{exp}}(\nu_0 + ih)$, $i = 1...n$ ($n = 161$ in the case considered) were fed to the computer. This series was compared with the calculated $\gamma^{\text{theor}}(E_\Omega, \nu_0 + \Delta\nu_0 + ih)$, obtained for different E_Ω with interval $\Delta E_\Omega = 0.1$ kV/cm. We take as an estimate of the maximum likelihood a contour $\gamma^{\text{theor}}(E_\Omega, \nu)$ that minimizes the quantity $M(\Delta\nu_0, E_\Omega)$:

$$M = \sum_{i=1}^{n} \frac{1}{s_i^2} [\, \gamma^{\text{exp}}(\nu_0 + ih) - \gamma^{\text{theor}}(E_\Omega, \nu_0 + \Delta\nu_0 + ih)\,]^2, \quad (11)$$

where s_i^2 is the empirical-distribution variance of the ith measurement.

It is known[11] that the quantity (11) is an effective maximum-likelihood estimate obeying a Pearson distribution, if the quantities γ_i^{exp} have in various realizations a normal distribution. We did not check on this fundamental hypothesis in the described experiments, since the value of E_Ω varied uncontrollably in different realizations. Special investigations,[21] however, have shown that the basic hypothesis is confirmed under conditions when a substance with selective losses independent of time were introduced into the cavity of

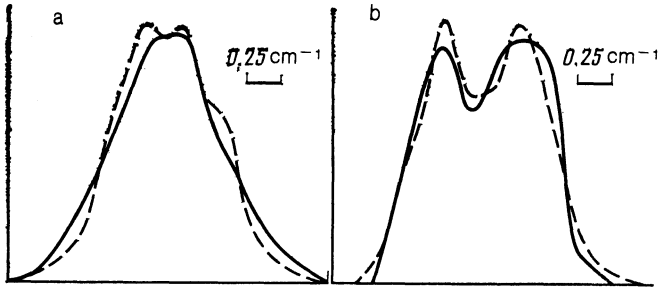

FIG. 5. Comparison and experimental (solid curve) absorption contours: a—9.1 GHz, b—2.94 GHz.

the ICL spectrometer. It was shown in addition that in this case the difference between the empirical-distribution variances is statistically insignificant, so that one can assume that $s_i^2 \approx s^2$.

The random component of the error in the estimate of the field intensity E_Ω was determined from its variance

$$DE_\Omega = \alpha_{EE}^{-1} M_{min}/(n-2), \qquad (12)$$

where α_{EE}^{-1} is a diagonal element of the error matrix that is the inverse of the matrix

$$\alpha_{a_j a_k} = \sum_{i=1}^{n} \frac{1}{s^2} \frac{\partial \gamma^{\text{theor}}}{\partial a_j} \frac{\partial \gamma^{\text{theor}}}{\partial a_k},$$
$$a_j,\ a_k = E_\Omega,\ \Delta\nu_0.$$

The total error of the estimate is determined by the random and "instrumental" component. The latter is in fact the "scale factor," or the interval ΔE_Ω in the calculations of γ^{theor} (in our calculations $\Delta E_\Omega = 0.1$ kV/cm). Figures 5a and 5b illustrate the comparison of the experimental (solid curve) and theoretical (dashed) data.

The procedure employed to formalize the maximum-likelihood estimates yields the parameters that determine the calculated contour, as well as their variance for arbitrary sets of contours obtained within the framework of any one of the theoretical models. Thus, the calculations of Ref. 24, in which no account was taken of the multiplet splitting, call for the appearance in a 9.1-GHz field, of a line-profile fine structure which does not accord with experiment; no such effect was observed, although the spectral resolution of the apparatus was quite sufficient. As shown above, no such fine structure of the line profile appears in our calculations.

The foregoing analysis of the results is convincing evidence of the need for taking the fine structure of the hydrogen levels into account in the interpretation of experimental data, in all cases when the electric-field frequency is comparable with the multiplet splitting.

5. NEW MEANS FOR DIAGNOSTICS OF ELECTRIC FIELDS IN A PLASMA

Spectroscopy of a plasma in oscillating microwave fields is for us not an end in itself. This subject is of interest primarily because of the feasibility of the inverse diagnostic task, that of determining the parameters (field intensity and frequency) of electric fields in a plasma from its optical spectra near Balmer-series lines. Let us examine the possibilities uncovered here in light of our experimental and theoretical investigations.

Interpretation of the experimental data is subject to no doubt under conditions when the field frequency exceeds the linewidth due either to its fine splitting or to the action of various broadening mechanisms, including the instrumental one. Optical spectra of a plasma contain observable satellites of unperturbed lines, with magnitude and form that can be adequately described by calculation procedure described in Sec. 2. For the amplitude one uses at present mainly the ratio of the itensity of the first satellites to the intensity of the main transition (see, e.g., Refs. 4 and 25). It must be recognized then that, as shown by our calculation, this quantity increases with increasing field amplitude E_Ω more rapidly than E_Ω^2 (see Fig. 3). The use of calculation results (similar to those shown in Fig. 3 for a 40.5 GHz field) permits a more accurate measurement of E_Ω.

In addition, to measure fields in a plasma it may be convenient to use the observed (see Sec. 4) dependence of the relative intensity of the first satellites on E_Ω. In this case it is necessary to measure the intensities of two lines of approximately equal magnitude, and there is no need to worry about the absence of laser shutoff in the region near the unperturbed line.[4] The form factor plays no noticeable role here, and the influence of the instrumental contour on the experimental result can be easily and adequately taken into account. Unfortunately, under the conditions of our experiments the quantity $\sigma^{(+)}/\sigma^{(-)}$ depends on the field quite weakly, decreasing from 1.125 at $E_\Omega \lesssim 1$ kV/cm to ~ 1 at $E_\Omega \approx 10$ kV/cm. Such a weak dependence does not permit a substantial increase of the field measurement accuracy. The accuracy can be improved, however, for other frequencies ($\nu \geqslant 15$ GHz), since the dependence of on E_Ω becomes stronger with decrease of frequency.

Finally, the field-intensity amplitude can be measured by using the calculated ratio of the cross sections of the first and second satellites (see Fig. 4). Measurements of this quantity permit, as indicated in Ref. 4, measurements of the electric field intensity in a plasma produced in a hydrogen-deuteron mixture with known ratio of the partial pressures of the two components. In this case, most importantly, the plasma regions in which there is no electric field have no longer any influence.

The situation is less obvious under condition when the field frequency is comparable with the line field. In this case spectroscopic diagnostics cannot, in our opinion, be regarded as an autonomous method. The interpretation of the experimental results calls for additional information on the possible field frequencies. This information can be obtained from theoretical models or by alternate diagnostics methods. Calculation of the absorption line contour by the procedure described in Sec. 2, and comparison of the experimental and theoretical contours, similar to that carried out in Sec. 4, will permit an estimate of the validity of the assumptions made concerning the field frequency and concerning the possible value of the field strength.

6. CONCLUSION

The foregoing theoretical and experimental investigations of hydrogen spectra in a microwave discharge lead to the following conclusions concerning the possibility of field diagnostics in a plasma.

1. Unambiguous intepretation of the spectra is possible only when the frequency of the oscillating field exceeds the

measured line width. (This requirement not only limits the range of the experimental conditions, but also determines the characteristics of the spectral apparatus and of the recording system.) Under these conditions, a spectral structure is observed with Stokes and anti-Stokes satellites, the distances of which from the central component of the line make it possible to determine the field frequency Ω. Analysis of the form of the satellites, of the ratio of their intensities to the central component and to one another, and of the spectral spacing of the maxima yields (if Ω is already known) the amplitude of the harmonic electric field intensity.

2. When the frequency of the plasma fields does not exceed the linewidth, spectroscopic diagnostics cannot be regarded as an autonomous method. Spectroscopic measurements can be used only as tests, as criteria for the agreement of the experimental data with some hypothesis concerning the frequencies of the plasma fields.

3. In all cases, an adequate interpretation of the experimental results calls for taking into account the fine structure of the hydrogen level. This can be done by numerically solving the nonstationary equation (1) with a Dirac Hamiltonian for an electron in the field of a nucleus, as the zeroth approximation with allowance for a sufficiently large number of harmonics in the wave function of the quasi-energy state.

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