Observation of the fine structure of a hydrogen plasma in a microwave field by intracavity laser spectroscopy

R. A. Akhmedzhanov, I. N. Polushkin, Yu. V. Rostovstev, M. Yu. Ryabikin, Yu. M. Shagiev, and V. V. Yazenkov

Institute of Applied Physics, Academy of Sciences of the USSR
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The fine spectral structure of a hydrogen-deuterium plasma in an intense microwave field was studied experimentally by intracavity laser spectroscopy. Microwaves of frequency 38.5 GHz were generated by a 200 kW gyrotron and directed by a quasi-optical duct toward a sealed-off gas discharge tube. An organic dye laser pumped by a flashlamp generated radiation pulses of length 1.5 μm and spectral width 8 nm. The Balmer series of hydrogen and deuterium spectra are in quantitative agreement with the calculations. It is shown that these spectra can be used to determine the spectrum of the oscillating electric fields in a plasma.

1. INTRODUCTION

The rapid advances in research on the nonlinear interaction of plasmas with intense electromagnetic waves have stimulated interest in studies of how rapidly changing electric fields influence the energy spectrum of bound atomic electrons. The Stark shift and splitting of the energy levels alter the atomic absorption and emission spectra in the plasma and give rise to satellites due to multiphoton radiation, absorption, and Raman scattering. Additional optical transitions forbidden by the selection rules of linear optics become allowed.

According to the concepts developed in Refs. 1 and 2, an electric field of frequency Ω splits each level into a system of quasi-energy states (QES) separated by $p\hbar\Omega$ ($p = \mp 1, \mp 2,...$). The transitions among these states give rise to satellites of frequency $\omega_0 + q\Omega$ near the unperturbed line at frequency ω_0. The intensity of each satellite is the sum of the intensities of transitions for which the difference $p - p' = q$ of the QES indices is the same.

Attempts to observe this effect experimentally and to measure the satellite intensities by conventional plasma spectroscopic techniques have at best led only to qualitative results.[3] This apparently explains why the theoretical work has been confined to two-level systems or to transitions from the ground state of a hydrogen atom, with the fine spectral structure neglected.[4,5]

Intracavity laser (ICL) spectroscopy[6] is a highly sensitive technique which can record and accurately measure the intensity of the spectral components in the presence of rapidly varying fields, and it opens up new possibilities in the spectral diagnostics of plasma electromagnetic waves.

Experiments of this type are also of independent interest. As was remarked in Ref. 7, quasi-energy states are known to exist in systems having finitely many energy levels, and in particular, in two-level atoms. However, atoms have infinitely many levels, and in this case the existence of QESs is far from trivial. For example, it was shown in Ref. 8 that there exist infinite systems for which a periodic perturbation does not give rise to an equidistant spectrum.

2. EXPERIMENTAL APPARATUS AND MEASUREMENT PROCEDURE

The experiments were carried out in a pulsed discharge plasma in a *D-H* mixture at a total pressure of 0.5–10 Torr. The plasma was generated in sealed-off U-shaped glass tubes of diameter 8 mm by a current pulse generator (pulse length 0.1–5 ms, amplitude 10^{-2}–10 A). Figure 1 shows a block diagram of the experimental setup. The intense microwaves were generated by a cyclotron resonance maser (gyrotron) 1 with an output power of 200 kW per pulse and an operating frequency of 38.5 GHz (the pulse was 200 μs long).

A special converter transformed the microwaves from the gyrotron into a linearly polarized wave with a Gaussian transverse electric field distribution. The quasi-optical duct 2 and Teflon lens 3 generated an electromagnetic field of up to 6 kV/cm at the center of the gas discharge tube 4. We used a calibrated attenuator 5 in the measurement waveguide channel to monitor the microwave power, while attenuator 6 in the quasi-optical duct was used to vary the power continuously. The gas discharge tube 4 was equipped with optical windows at each end and was placed inside the laser cavity, which was bounded by the mirrors 7. The flashlamps 8 pumped a cell 9 filled with a solution of oxazine-17 dye (for spectral measurements near the H_α line) or coumarin-47 (near the H_β line) in ethanol; this enabled us to generate

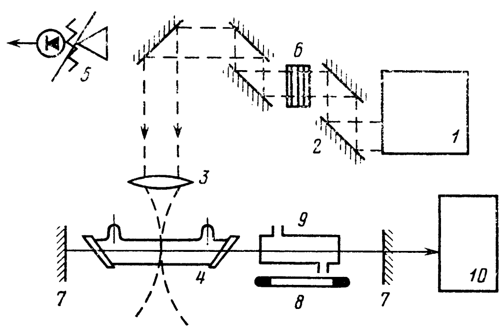

FIG. 1.

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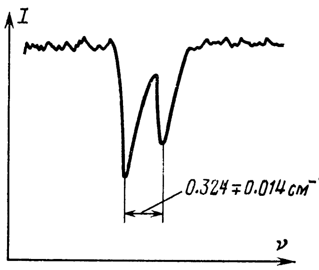

FIG. 2.

pulses of length $\approx 1.5\ \mu s$ and spectral width ≈ 8 nm. The laser spectrum was analyzed by a specially designed spectrograph 10 with a focal length of 1200 mm, a diffraction grating of period 1/75 mm and blaze angle 63°, and double dispersion; the resolution was equal to $4 \cdot 10^5$. We used a television system with a special synchronizing unit to record the spectrum on an oscilloscope screen.

In order to study how a rapidly varying electric field influences the plasma spectrum, one must simultaneously record spectral components which have nearly equal frequencies but whose amplitudes differ substantially by more than an order of magnitude. This imposes severe demands on the performance of the measuring and recording equipment, particularly on the spectral resolution and dynamic range. It was shown in Ref. 6 that the experimentally measurable ratio $I(\nu)/I_0$ of the intensities inside and outside the absorption line averaged over τ is determined by $\varkappa(\nu)l$, which depends on the selective absorber, and by L and τ:

$$I(\nu)/I_0=[1-e^{-\gamma(\nu)}]/\gamma(\nu), \qquad \gamma(\nu)=\varkappa(\nu)lc\tau/L. \tag{1}$$

Here $\varkappa$ is the absorption coefficient, l is the length of the cell, τ is the laser pulse length, and L is the length of the cavity. We monitored the instrument function of the recording system by recording the spectrum of a gas discharge tube placed in a cryostat cooled by liquid hydrogen. Figure 2 shows a typical spectrogram of the D_α Balmer line for deuterium under these conditions. Fine structure (splitting of the lower level) is plainly visible in all cases. The lines narrow appreciably and the contrast of the profile increases as the tube is cooled. We can use Eq. (1), the observed decay of the wings, and the measured halfwidths of the fine structure lines to deduce the lower bound $(2 \pm 0.2) \cdot 10^5$ for the resolution of the system from the data in Fig. 2.

Like any technique based on absorption, ICL spectroscopy has a limited dynamic range because the measurement errors increase for both small ($\gamma < 0.05$) and large ($\gamma > 3$) values of the effective optical thickness. One can optimize the conditions for measuring the weak spectral components by selecting the laser pulse τ and the concentration of the absorbing atoms in the discharge [this concentration determines the value of $\varkappa$ in (1)].

In most cases (except for very strong electric fields), near the center of the unperturbed line $\varkappa$ exceeds the gain of the dye, so that lasing stops at these frequencies. In the experiments we therefore generated a plasma in an *H-D* mixture with a partial pressure ratio $p_{H_2}/p_{D_2} = 5$–50. This ratio was chosen so that the intensity of the satellites of the H_α line in the microwave field was roughly equal to the intensity of the D_α line. By using an *H-D* mixture we were able to analyze the fine structure of the satellites under controlled conditions and determine the effects of the microwave field on the shape and width of the unperturbed line.

3. DISCUSSION OF THE EXPERIMENTAL RESULTS

The experiments revealed that the microwave field caused additional ionization of the plasma, with a corresponding increase in the concentration of absorbing atoms in the $2S_{1/2}$ and $P_{1/2,3/2}$ states. Two types of microwave breakdown were observed in the plasma, depending on the magnitude of the discharge current. At large discharge currents, the additional ionization increased the electron density n_e to a critical value, and in this case breakdown occurred on the leading surface of the plasma, from which the wave was reflected. The measurements showed that because of diffusion processes, the electron and excited atom densities increased everywhere inside the tube, even though the wave did not penetrate into the plasma.

For lower discharge currents, n_e also increased during microwave breakdown but remained below the critical value, and the plasma remained transparent to the microwave radiation. The gas pressure in the tube determined where the transition from one type of breakdown to the other occurred. All of our investigations were carried out for low discharge currents.

Under our experimental conditions we were able to record two additional pairs of absorption regions in the wideband laser spectrum; they were present in the microwave field and were frequency-shifted from the unperturbed lines by $-\Omega$, -2Ω (Stokes satellites) and Ω, 2Ω (anti-Stokes satellites). Because we employed an *H-D* mixture, we were able to determine the frequency scale of the recording system. Figure 3 shows some typical ICL spectrograms near the α-lines of the Balmer series for two concentrations of the absorbing atoms. We found that it was quite easy to make the absorption coefficients for the D_α line and $H_\alpha^{\pm 1}$ satellite comparable in order of magnitude (Fig. 3a). Since the partial pressure ratio p_{H_2}/p_{D_2} is known, in this case we can quite accurately determine the ratio of the absorption coefficients for the H_α and $H_\alpha^{\pm 1}$ spectral components. Similar measure-

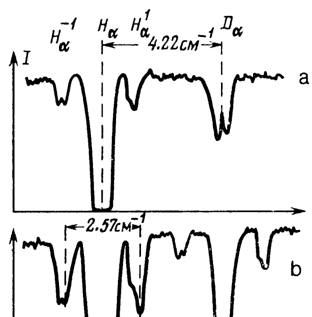

FIG. 3.

ments can be used to find the strengths of the oscillating electric fields in the plasma. According to (1), increasing the density of absorbing atoms (or the laser pulse length τ) will enhance the dip in the laser spectrum. Although the $D_\alpha^{\pm 1}$ satellites are also observed (Fig. 3b), the error in measuring the absorption coefficient for D_α is very large in this case.

It should be noted that the magnitude of the satellites depends on (among other things) the linear dimensions of the region in which the plasma interacts with the rapidly varying electromagnetic field. On the other hand, the effective absorption coefficient for the unperturbed lines depends on the entire length of the plasma column. In order to eliminate form-factor effects, the intensity of the microwave field must be measured in terms of the relative "intensity" of the first and second satellites. However, the spectral structure of the α-line at the microwave frequency (38.5 GHz) is such that the second anti-Stokes satellite H_α^2 nearly coincides in frequency with the Stokes satellite D_α^{-1}, which makes it much more difficult to interpret the experimental results. On the other hand, if one employs a recording system with a vidicon (600 lines on the screen with a contrast of 0.025), the H_α^{-2} and D_α^{+1} satellites cannot be measured simultaneously. Finally, only qualitative information can be gained from using photospectrograms recorded for large modern plasma devices. Things are considerably simpler if one operates near the β-lines of the Balmer series. The H_β and D_β lines are somewhat more widely spaced (5.5 cm^{-1} vs 4.2 cm^{-1}), so that the H_β^2 and D_β^{-1} satellites can be observed simultaneously, as was demonstrated in Ref. 9. However, for various reasons which will become apparent below, we were particularly interested in studying the spectrum near the α-lines.

The main experimental findings may be summarized as follows.

1. We analyzed in detail the profile of the D_α line (the halfwidth of the fine structure lines and the decay of the wings) in gas discharge tubes with a large ratio p_{H_2}/p_{D_2} ($\gtrsim 10$) in intense microwave fields. Under these conditions (Fig. 3a) the $H_\alpha^{\mp 1}$ lines were plainly visible, but the $D_\alpha^{\mp 1}$ deuterium satellites were too weak to appreciably distort the wings of the central D_α line. The experiments showed that the profile of the central line is independent of the electric field amplitude under our conditions (microwave frequency well above the frequency for transitions between the fine structure sublevels for both the upper and lower levels).

2. For laser and microwave fields polarized in the same direction, the satellites for the unperturbed lines were considerably more intense than for crossed laser and microwave fields. This fact can be exploited for polarization measurements, which are generally highly accurate and sensitive.

3. In all cases the satellites had a well-defined fine structure whose characteristic scale was roughly the same as for the fine structure of the level with principal quantum number $n = 2$. The Stokes and anti-Stokes satellites of the H_β line had nearly the same shapes and were located symmetrically about the unperturbed line (to within the instrument function of the recording system described above). However, the behavior of the spectrum near the H_α line was completely different—in the case the distances between the Stokes and anti-Stokes satellites were equal to 2Ω and 4Ω, and the entire spectrum was red-shifted by $(6 \pm 2) \cdot 10^{-2}$ cm^{-1} from the center of the one-photon absorption line. This shift is particularly apparent in the photospectrograms. The satellite profiles always differed considerably, and the anti-Stokes satellite was 30–40% more intense than the Stokes satellite.

In order to explain this behavior, we calculated the spectrum for absorption of the laser probe field $1/2[\mathbf{E}_l \times \exp(i\omega t) + \text{c.c.}]$ by a hydrogen atom in a microwave field $1/2[\mathbf{E}\exp(i\Omega t) + \text{c.c.}]$ with allowance for the fine structure of the levels $n = 2,3$ (H_α line) and $n = 4$ (H_β line). The intensities of the first Stokes and anti-Stokes satellites are determined by the probabilities $W_{\nu\mu}^{+}$, $W_{\nu\mu}^{-}$ for two-photon absorption and Raman scattering, respectively, because transitions between the quasi-energy states can be regarded as multiphoton transitions between the unperturbed stationary state [here ν is the number of sublevels of the lower ($n = 2$) level, and μ is the corresponding number for the upper levels ($n = 3,4$)]. Expressions for $W_{\nu\mu}^{\pm}$ can be derived by using perturbation theory to solve the time-dependent Schrödinger equation, or from the following general expression for the composite matrix element in the theory of two-photon transitions[10]:

$$W_{\nu\mu}^{\pm}=\frac{1}{8\hbar^4}\left|\sum_{\nu',\mu'}\frac{V_{\nu\nu'}V^{l}_{\nu'\mu}}{\omega_{\nu\nu'}\mp\Omega}+\frac{V^{l}_{\nu\mu'}V_{\mu'\mu}}{\omega_{\mu\mu'}\pm\Omega}\right|^2 q(\omega-\omega_{\mu\nu}\pm\Omega). \quad (2)$$

Here $V = -\mathbf{dE}$ is the Hamiltonian for the dipole interaction between an atom and the electromagnetic field; $q(\omega - \omega_{\mu\nu} \pm \Omega)$ is the lineshape function.

If we use (2) and results from the quantum theory of angular momentum to evaluate the matrix elements, we get the result

$$\sigma_{\nu\mu}^{\pm}=8\pi\hbar\omega_{\nu\mu}W_{\nu\mu}^{\pm}/c|E_l|^2 \quad (3)$$

for the two-photon transition cross section.

The vertical lines in Fig. 4 show the absorption cross sections calculated for the fine-structure transitions

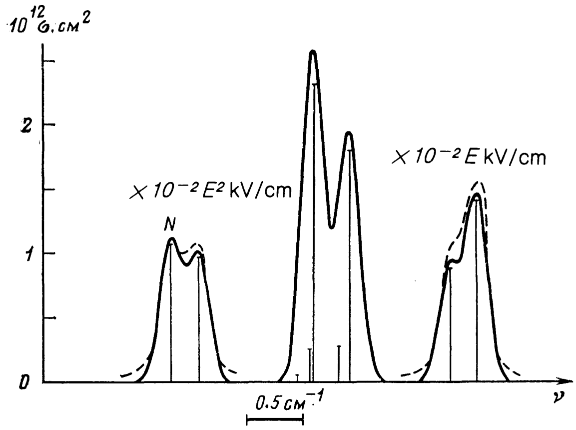

FIG. 4.

$n = 2 \rightarrow n = 3$. The solid curve shows calculated profiles $\sigma(\nu)$ found by assuming Doppler broadening at $T = 400$ K. The dashed curves give the results obtained by using Eq. (1) to analyze the experimental data in Fig. 3b.[1)] We see that the experimental and calculated results are in very satisfactory agreement, which improves further if the dashed curve is normalized at some other point. The deviation of the satellite wings from the calculated profile can be traced to the fact that the calculations neglected the influence of Stark broadening, and to the usual large error in measuring the absorption for small absorption coefficients.

As is the case for the one-photon absorption lines, the structure of the satellite falls into two spectral groups which correspond to transitions from two fine-structure sublevels of the $n = 2$ level which are separated by an energy gap of 0.364 cm^{-1}. In turn, each group consists of several lines whose intensities differ by a factor of ten or more, and the spectral structure is determined by the fine structure of the upper level ($n = 3$ for H_α and $n = 4$ for H_β). For both groups, the highest-frequency lines are also the most intense ones in the one-photon absorption spectrum. However, the satellites are associated with forbidden transitions (i.e., which require more than one photon). We therefore expect a satellite red-shift equal in order of magnitude to the Dirac splitting of the upper level (this splitting is proportional to n^{-3}).

Calculations show that the centroid of the first satellites in the H_α line is shifted by 0.072 cm^{-1}, as is also clearly shown by our experimental results. By contrast, the satellites of the H_β line are shifted by only 0.03 cm^{-1}.

4. CONCLUSIONS

Our experimental and theoretical investigations yielded the following conclusions.

1. Analysis of results like those shown in Figs. 2–4 for the α- and β-lines of the hydrogen and deuterium Balmer series shows that either the quasi-energy-state theory or the multiphoton transition theory can satisfactorily explain the experimentally observed fine structure of the satellites in a microwave field. There is no justification for the claim (see, e.g., Ref. 11) that the sensitivities of the shapes of the α- and β-lines to microwave fields are not the same.

2. Techniques based on intracavity laser spectroscopy can be used to measure the spectral components produced by the rf Stark effect in plasmas.

The procedure needed to analyze intense laser radiation can be automated easily by using ordinary photoelectric detectors. We predict that these techniques will prove very useful in measuring oscillating electric fields in plasmas.

[1)] We note that we made no attempt to measure the absolute values of the absorption coefficients. Results such as the ones shown in Fig. 3 therefore yield only relative values of $\gamma(\nu)$. In Fig. 4 the rescaled experimental values are normalized by the results calculated at the point N. The scale along the frequency axis was chosen on the assumption that the fine-structure components of the satellites are separated by 2Ω.

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