Local microwave field measurements in plasmas by resonant laser-induced fluorescence of hydrogen atoms

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(Submitted 4 April 1985)
Zh. Eksp. Teor. Fiz. **89**, 1648–1655 (November 1985)

Experimental and theoretical results are reported on the simultaneous interaction of atoms in a hydrogen plasma with an intense microwave field and with a laser beam of frequency equal to the resonance frequency for the H_α transition. The 38.5 GHz microwaves were generated by a gyrotron of power ~ 100 kW and directed by a quasioptical duct to a sealed gas discharge tube filled with hydrogen. A coherently pumped tunable organic dye laser produced 20-ns pulses of power 20 kW and linewidth 0.008 nm. The measured dependences of the fluorescence signal on the laser and microwave intensity agree quantitatively with calculations. It is shown that one can measure microwave fields in plasmas with good time and space resolution (10 ns, 10^{-2} cm) by recording the dependence of the fluorescence signal on the laser intensity.

1. INTRODUCTION

The radiation that accompanies the interaction between plasmas and intense electromagnetic waves is of great interest to researchers, primarily because of its practical applications, which include microwave propagation in the atmosphere, the design of efficient plasmachemical reactors, the development of UV and IR lasers, applications to fusion experiments, etc. The need to maximize the available information concerning the properties of the plasma and microwaves and how they change in space and time during a pulse has stimulated the development of new diagnostic techniques which combine adequate sensitivity and dynamic range with high space-time resolution and do not require inserting probes into the plasma. Saturation laser resonance spectroscopy, and in particular resonant laser fluorescence (RLF), is an example of such a technique. In RLF, information is extracted from the fluorescence signal which is generated when the upper level of a spectral transition resonant with the laser radiation is spontaneously deexcited. Saturation laser resonance spectroscopy can be used to probe low-density plasmas with absorbing particle densities $N \gtrsim 10^7$ cm^{-3}. The time resolution is determined by the laser pulse length or by the recording system, and resolutions of 1–10 ns are readily achievable. Spatial resolutions of $\sim 10^{-2}$ cm[1)] are possible using the RLF technique, in which the receiving system picks up signals that originate where the laser and probing beams cross.

In plasmas of low or moderate density ($N_e \lesssim 10^{15}$ cm^{-3}), the allowed atomic transitions are readily saturated by quasimonochromatic radiation from ordinary tunable lasers of power $\sim 10^4$ W (by "quasimonochromatic" we mean that the laser line is much narrower than the absorption line).[1] However, our measurements in a cool gas-discharge plasma revealed that stepwise and associative ionization severely limit the increase $\Delta N_e/N_e$ in the electron density to less than 1–2%. Techniques based on saturation laser spectroscopy may therefore be regarded as remote diagnostic methods.

RLF diagnostics has been used to measure concentrations of excited atoms and molecules,[1,2] electron densities,[3] and spectral linewidths in plasmas.[4] In this paper we consider using RLF from hydrogen atoms to measure oscillating electric fields in plasmas.

2. FORMULATION OF THE PROBLEM

A technique was developed and experimentally tested in Ref. 4 for measuring the saturation parameter G_0 for a spectral transition in a two-level atom excited resonantly by laser radiation. If no microwave field is present, the saturation is given by

$$G_0 = 4\pi T_1 T_2 d_{ab}^2 I_l / c\hbar^2, \quad (1)$$

where I_l is the power flux density of the monochromatic laser radiation, d_{ab} is the dipole matrix element, and T_1 and T_2 are the longitudinal and transverse relaxation times.

If the homogeneous width of the absorption line is much greater than the inhomogeneous width,[2)] the population of the upper level is given by

$$\Delta N_b = \frac{G_0}{2} \frac{(N_a^0/g_a - N_b^0/g_b)}{(\omega_{ba}-\omega)^2 T_2^2 + 1 + G_0} \frac{2 g_a g_b}{g_a + g_b}. \quad (2)$$

Here g_a and g_b are the statistical weights of the (degenerate) levels a and b; N_a^0 and N_b^0 are the level populations in the absence of laser radiation.

One can deduce G_0 experimentally from the slope of the dependence $1/B_f$ $(1/I_l)$; the reciprocal of the fluorescence signal B_f is proportional to ΔN_b, and I_l is the laser intensity.

The laser-induced transition probabilities change when the microwave field is turned on, because multiphoton processes involving a laser photon $\hbar\omega$ and an arbitrary number of microwave field quanta $q\hbar\Omega$ can occur. As before, the population of the upper level is characterized by the saturation parameter (1), but now both the line width and the effective dipole moment of the transition in general depend on the amplitude E of the microwave field. If we express the saturation parameter as $G(E) = G_0 f(E)$, we can find $f(E)$ by measuring the dependence B_f (I_l) with and without the microwave field:

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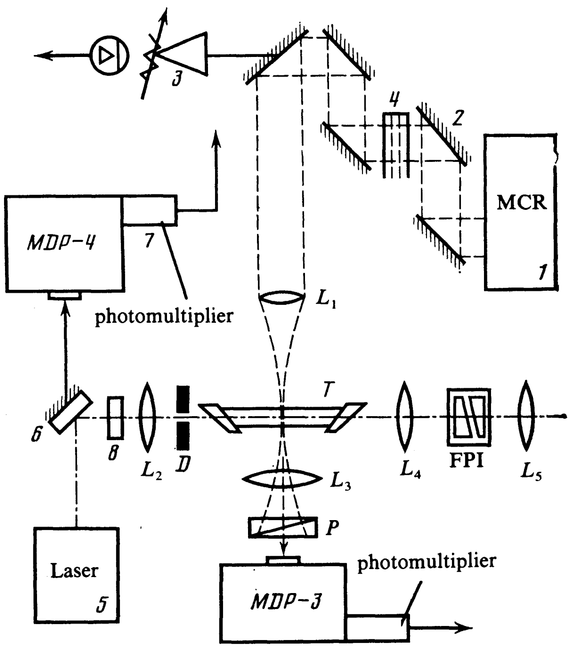

FIG. 1. Schematic of experimental system: 1) gyrotron; 2) quasi-optic forming line; 3,4) attenuators; 5) tunable laser; 6) beam-splitting window; 7) laser intensity monitor; 8) neutral filter; L_1, Teflon lens; $L_2 - L_5$, glass lenses; T, gas-discharge tube; P, polarizer; FPI, Fabry-Perot interferometer.

$$f(E)=\Delta\left(\frac{1}{B_f^{\,0}}\right)\Delta\left(\frac{1}{I_l^{\,E}}\right)\left[\Delta\left(\frac{1}{I_l^{\,0}}\right)\Delta\left(\frac{1}{B_f^{\,E}}\right)\right]^{-1}. \qquad (3)$$

3. EXPERIMENT

Figure 1 shows a schematic of the experimental setup. A gyrotron 1 (cyclotron resonance maser) served as the intense microwave source; the output power reached 200 kW per pulse and the pulse length was $\approx 200\,\mu$s at the operating frequency 38.5 GHz. We used a special converter to linearly polarize the gyrotron radiation and form a near-Gaussian transverse electric field. After this conversion, the microwaves reached a quasioptical forming line 2 and were then focused to a spot of diameter 12 mm (half-intensity) by a Teflon lens of focal length ~ 20 cm so that the field strength at the waist was

$$E_0\,[\mathrm{V/cm}]\approx 22P^{1/2}\,[\mathrm{W}]. \qquad (4)$$

Because the conversion factor of the entire microwave-transforming system was $\approx$40–45%, the maximum field was less than 6 kV/cm. The microwave power was monitored by a calibration attenuator 3 in the measuring waveguide channel; attenuator 4 in the quasioptical channel was used to continuously vary the microwave power. A constant-current discharge was ignited in a hydrogen-filled discharge tube T, which was located at the focus of lens L_1 and contained optical windows of diameter 8 mm at either end. The discharge current and hydrogen pressure were chosen to ensure that the microwave field did not cause any additional ionization of the plasma in the positive column; this was verified by analyzing oscilloscope traces of the spectral line intensities and discharge current during and after the microwave pulse.

The light source was a tunable dye laser 5 (cresyl violet in ethanol) which was longitudinally pumped by second-harmonic YAG laser light ($\lambda = 530$ nm) as described in detail in Refs. 1 and 4. The laser was tuned to the wavelength (656.3 nm) of the H_α hydrogen line; the laser power was 20 kW, the pulse length was ~ 20 ns, and the linewidth was ~ 0.008 nm. The laser beam was directed onto a rotating mirror 6 which split the beam, sending a small part of it to the power monitor 7 and the remaining $\sim 90\%$ to the lens L_2, which focused the light through a replaceable neutral filter 8 into the center of the discharge tube. The radiation then passed into a Fabry-Perot interferometer, which was used to monitor the laser linewidth. Lens L_3 formed an image of the intersection of the laser and microwave beams onto the entrance slit of the receiving system consisting of an NDR-3 monochromator with resolution $\sim 3\cdot 10^4$, a photomultiplier, and an oscilloscope. The time resolution was measured using a semiconductor laser and square-wave pulse generator and was better than 10 ns. The field vectors $\mathbf{E}_l$ and $\mathbf{E}$ of the polarized laser and microwave radiation were parallel, while their wave vectors $\mathbf{k}_l$ and $\mathbf{k}$ were mutually orthogonal. We mounted a polarizer P with its transmission axis perpendicular to $\mathbf{E}_l$ ahead of the monochromator entrance slit in order to block the laser light scattered by the walls of the plasma tube. Another Fabry-Perot interferometer, crossed with the monochromator or with an interference filter, was used to measure the spectral linewidths of the plasma radiation. The laser was tuned roughly to the H_α absorption line by using the monochromator to simultaneously observe the laser spectrum and the luminescence spectrum of the auxiliary glow discharge in the hydrogen. Fine tuning was achieved by recording the amplitude of the fluorescence signal from the gas discharge tube in the absence of the microwaves, or by exploiting the optogalvanic effect. The experimental equipment operated at a frequency of ~ 1 Hz; the various components (the microwave oscillator, tunable laser, and recording system) were synchronized by a six-channel GZI-6 pulse generator with adjustable delays in all channels.

Most of the experimental data were obtained for a discharge current $i \approx 0.5$ A and hydrogen pressue 5 Torr. Under these conditions the electron density and temperature (measured using a double probe) were equal to $6\cdot 10^{11}\,\mathrm{cm}^{-3}$ and 2 eV, respectively. The gas temperature in the discharge was found by analyzing the profile of the helium and neon spectral lines (trace amounts of these inert gases were present in the tube); we found the value 400 ± 40 K.

We carefully studied how the microwaves changed the shape of the hydrogen absorption lines; the measurements were carried out by intracavity laser spectroscopy.[5] The plasma was placed inside the cavity of a wide-band laser which was pumped by a lamp and produced pulses of length 1 μs. We used a specially designed diffraction spectrograph with double dispersion and a focal length of 1200 mm. The spectrum was recorded on an oscilloscope equipped with a television tube. The resolution of the spectrograph and the recording system was better than one part in $1.5\cdot 10^5$. These experiments were carried out at low current densities, so that

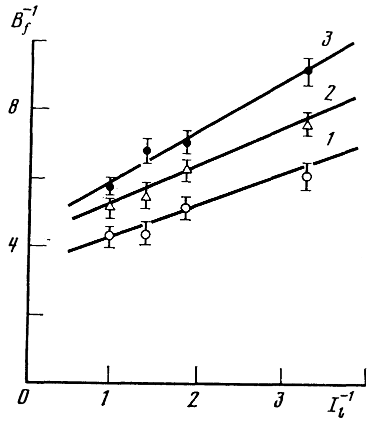

FIG. 2. Reciprocal B_f^{-1} of the fluorescence signal intensity as a function of I_l^{-1}, the reciprocal of the laser intensity, for microwave power $W = 0$ (1), 45 (2), and 60 kW (3).

the Stark broadening was negligible compared to the dominant Doppler broadening mechanism. We observed a doubly peaked profile for all microwave fields E, provided no additional ionization occurred; the profile corresponded to a temperature of ≈ 400 K and was determined by the fine structure of the H_α line. Calculations show that even a 20% increase in the Doppler width suffices to eliminate the fine structure completely. The wings of the line decay more gradually for the remaining broadening mechanisms, which should cause the fine structure to disappear still more rapidly. The persistence of the two peaks in the profile indicates that turning on the microwave field does not broaden the line but merely changes the effective dipole matrix element, and the quantity $f(E)$ in (3) may be expressed in the form

$$f(E)=d_{ab}^2(E)/d_{ab}^2(0). \quad (5)$$

Figure 2 shows the experimental dependence $B_f^{-1}(I_l^{-1})$, where B_f is the intensity of the fluorescence signal integrated over the width of the H_α line, and I_l is the laser radiation intensity; curves 1–3 correspond to microwave powers 0, 45, and 60 kW, respectively.

As in our previous experiments, we observed that B_f varied widely (by 50–150%) from pulse to pulse because of fluctuations in the plasma density with characteristic times 10^{-7} s and fluctuations in the laser energy and spectrum. Each point in Fig. 2 was therefore obtained by averaging the data found from 70–90 measurements. The experimental curves $B_f^{-1}(I_l^{-1})$ are nearly linear; according to (3) and (5), the dependence $d_{ab}^2(E)$ on the microwave field can be deduced from the ratio of the slopes corresponding to different E.

4. DISCUSSION

One can measure the field E if $f(E)$ is known theoretically. However, general information regarding $f(E)$ can also be found by qualitative arguments. Indeed, a static electric field will produce a Stark splitting of atomic energy levels with nonzero dipole moments $d_{\mu\mu}$, namely, $\Delta\epsilon_\mu = -d_{\mu\mu}E$. If the electric field oscillates at frequency Ω then according to Ref. 6 the atomic levels will oscillate harmonically, resulting in a modulation of the transition frequency for the two levels a and b with a deviation $\Delta\omega = (d_{\alpha\alpha} - d_{\beta\beta})E/\hbar$. Such an oscillation spectrum contains harmonics of frequency $\omega_{ba} + q\Omega(q = 0, \pm 1, \pm 2, \dots)$ and amplitude $J_q^2(\Delta\omega/\Omega)$, where J_q is the Bessel function of order q. For narrow-band modulation with $\Delta\omega/\Omega \ll 1$ the first harmonics are the important ones, because $J_q^2(x)$ decreases rapidly for small arguments as q increases. For a wide-band modulation, the individual harmonics have roughly equal amplitudes for $q < q^* \approx \Delta\omega/\Omega$ but decay rapidly to zero outside the modulation band $(q > q^*)$. This implies that turning on a microwave field will alter the effective transition probabilities at the frequency ω_{ab} and hence also the saturation parameter. This simple interpretation is supported by detailed quantum mechanical calculations which were carried out in Refs. 7 and 8 by different methods under somewhat different assumptions.

Among other things, we derived an expression for the population of the upper level in a two-level atomic system in a two-frequency electric field

$$\mathbf{E}_l \cos(\omega t+\varphi)+\mathbf{E}\cos\Omega t \, (\omega\approx\omega_{ba}\gg\Omega).$$

by solving the equations of motion for the density matrix and assuming a basis of H-atom wave functions in a low-frequency field[9]:

$$\Psi_\mu(\mathbf{r},t)=\varphi_\mu(\mathbf{r})\exp(-i\varepsilon_m t/\hbar - i d_{\mu\mu}E\sin\Omega t/\Omega t). \quad (6)$$

Here $\mu\in\alpha, \beta$; $m\in a, b$; β and α are the sets of parabolic quantum numbers of the states belonging to the top (b) and bottom (a) levels, respectively. One can show that

$$\Delta N_b = \frac{G(E)}{2} \cdot \frac{(N_a^0/g_a - N_b^0/g_b)}{(\omega_{ba}-\omega+q\Omega)^2 T_2^2+1+G(E)} \cdot \frac{2g_a g_b}{g_a+g_b}, \quad (7)$$

where

$$G(E)=4\pi T_1 T_2 d_{ab}^2(E) I_l/c\hbar^2$$

is the saturation parameter and

$$d_{ab}^2(E)=\sum_{\alpha,\beta} d_{\alpha\beta}^2 J_q^2(\Delta\omega_{\alpha\beta}/\Omega)$$

is the square of the effective dipole moment for the $a\rightarrow b$ transition. The result (7) is valid for high frequencies Ω:

$$\Omega\gg\max\{1/\tau, 1/T^*\}, \quad (8)$$

where $\tau = g_a g_b T_1/2(G_a + g_b)$ is the effective lifetime of atoms in the upper level and $T^* = 4T_1/G_0$ is the characteristic time of the laser-induced population changes.

Equation (7) for ΔN_b in an oscillating field is the same as Eq. (2), apart from the resonance condition and the effective dipole moment of the transition.[3)] The microwave field E alters the probability of laser-induced transitions—absorption and scattering at the frequency ω_{ab} becomes less probable and satellites form near the unperturbed line which correspond to transitions at the combination frequencies $\omega_{ba} + q\Omega(q\neq 0)$.

The values plotted in Fig. 2 and the calculations of $f(E)$ given in Ref. 8 lead to the following results for the microwave field strength [the values in parentheses were calculated using Eq. (4)]:

P, kW	45	60
E, kW/cm	4.3 ± 0.5	5.5 ± 0.6
	(4.7 ± 0.7)	(5.4 ± 0.8)

The close agreement between the field strengths measured by the RLF method and the values calculated from calorimetric measurements in a "cold" channel shows that plasma fields can be determined accurately from laser fluorescence intensity measurements as described above. In addition, the agreement shows that under our experimental conditions the transverse relaxation time T_2 is independent of the microwave amplitude E.

According to (8), the spectral width of the qth satellite is equal to $\Delta\omega_q = 2[1 + G(E)]^{1/2}/T_2$, and the condition $\Omega > \Delta\omega_q$ might appear to limit the usefulness of the RLF technique for fields of low frequency. (In our experiments we had $\Omega/2\pi c = 1.3$ cm^{-1} and $\Delta\omega_0/2\pi c = 0.5$ cm^{-1}.) However, one must bear in mind that for large modulation factors $\Delta\omega/\Omega \gg 1$, the spectral intensity of the frequency-modulated oscillations is greatest at the edge of the modulation band, i.e., for $q^* \sim \Delta\omega/\Omega$. In this case one must tune the laser to the frequency $\omega_{ab} + q^*\Omega$, and the RLF method is applicable if

$$q^*\Omega > \Delta\omega_q. \tag{9}$$

The limitations of the proposed method for probing plasmas with hot heavy components are also of interest; in this case the Doppler width greatly exceeds the collisional width. For $\omega u/c \gtrsim \Omega$, where $u = (2T_a/m_a)^{1/2}$ is the thermal velocity of the atoms, the ratio $G(E)/G_0$ becomes independent of the atomic temperature T_a because the broadening of the Maxwell velocity distribution increases the fraction of atoms with velocities $\omega V_x/c = q\Omega$ ($q = \pm 1, \pm 2$) and hence also the number of atoms absorbing resonantly at the satellite frequencies when the laser frequency ω is tuned to line center. In the extreme case when $\omega u/c \gg q^*\Omega$, i.e.,

$$T_a \gg T_0 = \left(\frac{\Omega}{\omega}\right)^2 \frac{mc^2}{2} q^{*2}, \tag{10}$$

the fluorescence signal becomes independent of the microwave field amplitude. In particular, the RLF technique proposed above becomes inapplicable for temperatures $T_a \gg 200$ eV if we assume weak fields ($q^* = 1$) and $\lambda_l = 6.56 \cdot 10^{-5}$ cm and $\lambda_{\text{microwave}} = 0.1$ cm. This restriction is clearly less stringent for stronger fields.

One can also measure E by using a single laser pulse. This requires a tunable narrowband dye laser which is pumped by a lamp and generates a bell-shaped pulse at least several microseconds long. A monochromator with two exit slits (or else two monochromators) is needed to analyze the spectrum of the fluorescence signal. The commercially available MDR-4 monochromator is very convenient here (its rotating output mirror must be replaced by a semitransparent mirror). As usual, the width of one of the exit slits is set equal to the width of the entrance slit, while the second slit is opened wide enough so that the photomultiplier receives the total signal from all the harmonics (this signal is easily shown to be equal to the fluorescence signal in the absence of the microwave field). One can study the time behavior of the field during the laser pulse by monitoring the time dependence of the laser intensity I_l while measuring $B_f^E(t)$ and $B_f^0(t)$.

We are indebted to M. A. Miller and E. A. Oks for helpful discussions regarding the experimental results and their interpretation.

[1)]The spatial resolution is usually limited by the threshold sensitivity of the recording system and by the need to achieve an adequate signal/noise ratio.

[2)]The opposite case was considered in Ref. 4.

[3)]We originally planned to derive and discuss Eq. (7) in more detail; however, a recent paper[8] has made this unnecessary.

[1]R. A. Akhmedzhanov, I. N. Polushkin, Ya. I. Khanin, and V. V. Yazenkov, Fiz. Plasma **8**, 333 (1982) [Sov. J. Plasma Phys. (1982)].

[2]G. T. Razdobarin and I. P. Folomkin, Zh. Tekh. Fiz. **49**, 1353 (1979) [Zh. Tekh. Fiz. **49**, 753 (1979)]; V. S. Burakov, P. Ya. Misakov, P. A. Naumenkov, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **26**, 547 (1977) [JETP Lett. **26**, 403 (1977)]; Pis'ma Zh. Tekh. Fiz. **4**, 1362 (1978) [Sov. Tech. Phys. Lett. **4**, 549 (1978) [Zh. Prikl. Spektrosk. **29**, 413 (1978)].

[3]K. Tsuchida, S. Miyake, K. Kadota, and J. Fujita, Plasma Phys. **25**, 99 (1983).

[4]R. A. Akhmedzhanov, I. N. Polushkin, Ya. I. Khanin, and V. V. Yazenkov, Fiz. Plasma **10**, 865 (1984) [Sov. J. Plasma Phys. **10**, (1984)].

[5]V. M. Baev, T. P. Belikova, É. A. Sviridenkov, and A. F. Suchkov, Zh. Eksp. Teor. Fiz. **74**, 43 (1978) [Sov. Phys. JETP **47**, 21 (1978)].

[6]D. I. Blochinzev, Phys. Zs. Sow. Union **4**, 501 (1933).

[7]V. A. Kovarskiĭ, Zh. Eksp. Teor. Fiz. **57**, 1217 (1969) [Sov. Phys. JETP **30**, 663 (1969)]; in: Opticheskie i Kineticheskie Éffekty v Sil'nom Élektromagnitnom Pole (Optical and Kinetic Effects in Strong Electromagnetic Fields), Shtinitsa, Kishinev (1974) p. 3.

[8]V. P. Gavrilenko and E. A. Oks, Pis'ma Zh. Tekh. Fiz. **10**, 1443 (1984) [Sov. Tech. Phys. Lett. **10**, 609 (1984)].

[9]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1964)].

Translated by A. Mason