A simple far-infrared laser interferometer for measuring electron densities in reactive low-temperature plasmas

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(Received 31 May 2005; accepted 13 October 2005; published online 23 November 2005)

A sensitive far-infrared (fir) interferometer for electron density measurements in reactive low-temperature plasmas is described. The instrument is based on an optically pumped fir laser (wavelength range 50–600 μm depending on the working gas) and makes use of the nonlinear relation between output power and cavity loss. The fir beam, which leaves the resonator through a coupling hole in the end mirror, is reflected back into the cavity, such that the coupling hole behaves like a variable “leak” with a loss rate depending on the phase of the reentering wave relative to the standing wave within the resonator. As a result of the feedback, the output intensity undergoes strong nonlinear variations if the optical distance of the external mirror is changed by small amounts, Δz. The power variation is monitored through a small opening in the external mirror. Test experiments using a wavelength of 432.6 μm and a Schottky-diode detector have yielded a minimum detectable pathlength variation of Δz=0.4 μm, corresponding to a change of the line-integrated electron density n_e×L of about 5×10^15 m^-2. A first application to argon plasmas in inductively coupled rf discharges has been made, and the results have been compared to concomitant Langmuir probe measurements. © 2005 American Institute of Physics.

[DOI: 10.1063/1.2134355]

I. INTRODUCTION

Reactive low-temperature plasmas of reduced pressure (typically a few hundred Pa or less) have found numerous technological applications, in particular, for the modification of surfaces (e.g., the production of integrated circuits or the treatment of textiles, mechanical tools, and medical implants). In these plasmas the gas temperature is close to ambient, and the chemistry is strongly dependent on the energy delivered by hot electrons. Hence the number density of free electrons, n_e, is a key parameter for characterizing the plasma properties and predicting the gas composition. In practice, however, precise measurements of n_e turn out to be rather difficult in a reactive environment: Langmuir probes^1 are susceptible to uncontrolled coatings of the probe tip that deteriorate the reliability, optical emission spectroscopy^1 has to deal with complex molecular spectra that are hard to analyze, and incoherent light scattering^2 suffers from a very small number of scattered photons in the presence of ample background radiation.

An alternative diagnostic technique is interferometry, which makes use of the fact that the index of refraction μ of a nearly collisionless, unmagnetized plasma depends on the electron density according to^3

\[ \mu = \sqrt{1 - \omega_p^2/\omega^2} \approx 1 - (r_0/2\pi)\lambda^2 n_e \]  

[ω and λ denote the probing wave frequency and wavelength, respectively, ω_p is the plasma frequency, and r_0 = e^2/(4πε_0ε_n) = 2.818×10^{-15} m is the classical electron radius; the approximate expression holds for ω_p^2/ω^2 = (r_0/π)^2 n_e ≪ 1]. Owing to the change of μ, an electromagnetic wave traveling through a plasma along a path L experiences a phase shift Δφ relative to free-space propagation, which amounts to

\[ \Delta \phi = r_0 \lambda \int n_e \, dz. \] 

Since Δφ is proportional to the product of the probing wavelength and the line-integrated electron density, typical values of n_e~10^{17} m^-3 and L~0.1 m require wavelengths in the microwave range (>1 cm) in order to obtain sizable phase shifts of π/10 or more. On the other hand, the scale length of density variations in these plasmas is of the order of a centimeter, which can hardly be resolved by means of rather wide microwave beams. Despite this deficiency, several microwave interferometers operating at λ~0.4 cm have been constructed and successfully applied to rf-heated low-temperature plasmas.4,5

In order to improve the spatial resolution, Schulz-von der Gathen and co-workers^6,7 developed a quasi-optical 1 mm interferometer with a Gaussian probing beam that is focused in the plasma center to a waist of about 5 mm in diameter. The setup resembles the phase-modulated Mach-Zehnder configuration invented by Véron,^8 where the reference beam is diffracted off a rotating cylindrical grating in order to introduce a small frequency shift. In this way the plasma-induced phase shift is transmitted to a low-frequency beat signal and can be measured with high accuracy. By mounting the optical components on a very rigid, vibration-isolated baseplate and employing sensitive Schottky barrier diodes, the authors achieved a phase resolution of better than 10^{-4} of 2π (corresponding to a line-integrated electron density of 2×10^{14} m^-2).
In this contribution we propose a sensitive and fairly plain scheme that is reminiscent of a coupled-cavity laser interferometer\textsuperscript{b–11} and makes use of the nonlinear relation between output power and cavity loss. The device consists of a homemade far-infrared (fir) laser, an external feedback mirror, and a Schottky-diode detector. Besides technical simplicity and ease of operation, the instrument offers a further improvement of the spatial resolution due to the relatively short wavelength of the fir radiation (between about 50 and 600 \(\mu\)m depending on the laser gas). In the following section we give a description of the principle of operation. Then the practical realization is outlined, and the results of the first test experiments are presented. Finally, some options for future developments are discussed.

II. PRINCIPLE OF OPERATION

A schematic drawing of the optical system is shown in Fig. 1. The main component is a cw fir waveguide laser consisting of a cylindrical dielectric tube and two flat end mirrors (details concerning the pumping of the active medium have been omitted for the sake of clarity and will be considered in the next section). A small fraction of the circulating power leaves the cavity through a coupling hole in one of the end mirrors (\(M_2\)) and is reflected back by an external feedback mirror (\(M_3\)). The latter is slightly concave and positioned such that its radius of curvature matches the spherical phase front of the (nearly) Gaussian laser beam. Consequently, \(M_3\) transforms the phase front back to its original size and shape at the coupling hole, which appears to be “closed” if the reentering wave is in phase with the standing wave inside the laser resonator. Shifting the external mirror by a quarter of a wavelength along the beam direction brings the reflected wave out of phase and reduces the circulating power in the cavity. In other words, the coupling hole behaves like a variable “leak” with a loss rate depending on the optical distance \(z\) between \(M_2\) and \(M_3\). The resulting power variation can be monitored by means of a small opening in the external mirror and a fir detector placed directly behind.

In order to estimate the dependence of the laser output power \(I_{\text{out}}\) on the distance \(z\), we follow the general treatment by Siegman\textsuperscript{12} and express \(I_{\text{out}}\) as a fraction of the circulating power \(I_{\text{circ}}\):

\[
I_{\text{out}} = \delta_M \cdot I_{\text{circ}}
\]  

(\(\delta_M < 1\) represents the total external coupling, which is normally given by the transmission of the end mirrors). The circulating power \(I_{\text{circ}}\) depends on the unsaturated gain and loss coefficients of the laser medium (\(\gamma_0\) and \(\alpha\), respectively), on the external coupling \(\delta_M\), and on the cavity length \(L_c\) according to

\[
I_{\text{circ}} = \left( \frac{2L_c \gamma_0}{2L_c \sqrt{2} \alpha + \delta_M} - 1 \right) I_{\text{sat}},
\]

where \(I_{\text{sat}}\) is the saturation intensity (i.e., the intensity for which the stimulated emission rate equals the spontaneous emission rate of the laser transition). For \(\gamma_0 \gg \alpha + \delta_M/2L_c\) (which is usually valid in optically pumped fir lasers) we can neglect the second term in the large parentheses and write the output power as

\[
I_{\text{out}} = \frac{\delta_M \gamma_0 I_{\text{sat}}}{\alpha + \delta_M/2L_c}.
\]

Recalling the above-mentioned concept of a variable “leak,” we may assume that the loss coefficient \(\alpha\) is a periodic function of the distance \(z\) of the external mirror \(M_3\):

\[
\alpha(z) = \alpha_0 - \alpha_1 \cos(2kz) = (\alpha_0 - \alpha_1) + 2\alpha_1 \sin^2(kz)
\]

\((k=2\pi/\lambda\) is the wave number of the laser radiation, and the factor of 2 in the argument of the cosine function accounts for the double path to and from \(M_3\)). If we substitute this expression for \(\alpha\) in Eq. (5), the output power as a function of \(z\) takes on the same form as the transmission function of a scanning Fabry–Perot interferometer:

\[
I_{\text{out}}(z) = \frac{\delta_M \gamma_0 I_{\text{sat}}}{(\alpha_0 - \alpha_1 + \delta_M/2L_c) + 2\alpha_1 \sin^2(kz)}
\]

\[= \frac{I_{\text{max}}}{1 + K \sin^2(kz)}.\]

Although the derivation is somewhat heuristic, this expression describes the actual performance of the laser interferometer quite well, if we expand it by an additional term \(B\) to allow for a negative offset:

\[
I_{\text{det}}(z) = \frac{A}{1 + K \sin^2(kz)} - B.
\]

As an example Fig. 2 shows the detected power as a function of the mirror separation \(z=z_0+\Delta z\) for a laser wavelength of \(\lambda=184.3\ \mu\text{m}\).
184.3 \mu m [the working gas was CH$_2$F$_2$ pumped by the 9R32 line of a CO$_2$ laser; the intensity was registered by means of a pyroelectric detector (Mullard 802-CPY) in combination with a mechanical chopper]. The full line is a fit according to Eq. (8) where the constants $A$, $B$, and $K$ have been adjusted to match the narrow peaks.

Since the parameter $kz$ represents the phase of the laser beam at $M_3$, it can be altered, not only by moving the external mirror but also by placing a refractive medium in the beam path that is sufficiently homogeneous across the beam diameter to leave the wave fronts undistorted. In case of a weakly ionized low-temperature plasma, the density gradients are small enough to satisfy this condition, and the phase change $\Delta(kz) = \Delta \varphi$ as given by Eq. (2) can be measured fairly directly by displacing $M_3$ to compensate for the plasma-induced variation of the output power. Denoting the required distance by $\Delta z$ and replacing the wavelength $\lambda$ in Eq. (2) by the wave number $k$, we obtain the line-integrated electron density

$$\int L n_e dz = \frac{k}{2 \pi r_0} \Delta \varphi = \frac{k^2}{2 \pi r_0} \Delta z.$$  

(9)

Obviously, the sensitivity of the laser interferometer depends on the “working point” and is highest in the wings of the power peaks where the slope $\Delta I_{le}/\Delta z$ is maximal (see Fig. 2).

### III. EXPERIMENTAL SETUP

The centerpiece of the instrument is an optically pumped far-infrared interferometer$^{13}$ as shown in Fig. 3. In order to keep the design as simple as possible, we use a dielectric waveguide (Pyrex tube, length 1.5 m, inner diameter 3.2 cm), which is terminated at both ends by flat mirrors made of polished aluminium disks. Both mirrors are attached to a support structure by means of adjusting screws, and one of them is mounted on a translation stage for tuning the cavity length $L_c$. Each mirror contains a coupling hole, which serves to admit a CO$_2$ laser beam for optical pumping at one end of the cavity and to extract the far infrared radiation at the other end. The input hole has a diameter of 2 mm and is eccentric such that the pump beam can be aligned at a slight angle relative to the axis in order to avoid feedback into the CO$_2$ laser. The extraction hole for the far infrared beam is centered on axis. Its diameter ranges from 2 to 10 mm depending on the laser gas and on the selected wavelength (the optimum size is found by maximizing the slope of the power peak in the final layout of the interferometer). The input and output couplers are sealed off by a ZnSe Brewster window and a crystal quartz plate, respectively. The quartz window is transparent for far infrared but opaque in the CO$_2$ wavelength range and serves also as a blocking filter for the pump radiation.

The cavity can be filled with a variety of molecular gases that lase in the far infrared regime when pumped by suitable infrared lasers. To date we have used methylene fluoride (CH$_2$F$_2$), methyl alcohol (CH$_3$OH), and formic acid (HCOOH) at filling pressures of $\sim$10 Pa (the latter two substances are liquids with a vapor pressure well above the filling pressure, which makes their application particularly easy). The pump source is a standard, grating-tunable CO$_2$ laser that can be operated at many discrete wavelengths around 10 \mu m. For fine tuning of the pump frequency, the front mirror of the CO$_2$ laser is mounted on a piezoelectrical translator so that the resonator length can be adjusted by a dc voltage. Typical pump powers are 10–40 W in a cw mode, yielding far infrared laser radiation at a power level of a few mW (note that the output coupling hole is optimized for interferometer performance and not for maximum far infrared power). The gases, wavelengths, and operating conditions that we have used so far are summarized in Table I.

The interferometer is set up by placing an external mirror at some distance $z_0$ from the output coupler and reflecting the far infrared beam back through the coupling hole. The exact position depends on the radius of curvature $R$ of the mirror, on the wavelength $k=2\pi/\lambda$ of the laser radiation, and on the waist $w_0$ of the Gaussian beam according to

$$z_0 = (R + \sqrt{R^2 - k^2w_0^2})/2.$$  

(10)

Since $w_0$ corresponds roughly to the radius of the coupling hole, the term $k^2w_0^2$ is normally much smaller than $R^2$, and Eq. (10) is well approximated by $z_0 \approx R$.

In our first test setup we added two additional mirrors guide and focus the far infrared beam through a plasma chamber (Fig. 4). The external mirrors have a curvature of $R=0.4$ m ($M_3$) and $R=0.9$ m ($M_4$), resulting in a beam diameter of 5 mm inside the chamber. The far infrared power ($\lambda=432.6$ \mu m) is monitored through a small hole ($\sim$3 mm diameter) in the external mirror by means of a Schottky-diode detector (Radiometer Physics GmbH, optimized for 432.6 \mu m).

### IV. FIRST TEST EXPERIMENTS

By mounting the external mirror $M_3$ on a translation stage and varying the distance $z=z_0+\Delta z$ to the output cou-
pler, strong changes of the fir laser intensity are observed. Figure 5 shows the amplitude of the detector signal as a function of $z$, and the full line is a fitted curve according to Eq. 8. At the steepest slope of the power peak, a shift of the mirror by $\Delta z = 1 \mu m$ causes a change of the signal amplitude by $5 mV$ [Fig. 5(b)]. Since the noise level of the detector signal amounts to about $2 mV$, the minimum detectable variation of the optical pathlength becomes $\Delta z_{\text{min}} = 0.4 \mu m$, and Eq. 9 yields a minimum detectable change of the line-integrated electron density of

$$n_e L_{\text{min}} = \frac{2 \pi}{r_0 \lambda} \Delta z_{\text{min}} = 5 \times 10^{15} m^{-2}. \quad (11)$$

First measurements of the electron density in argon plasmas were performed in an inductively coupled GEC reference cell, which is a standard source for the production of low-temperature plasmas at reduced pressure (details of the construction and operation may be found elsewhere). The cell is equipped with crystal quartz windows that are tilted by $28^\circ$ to prevent reflections into the laser cavity and to provide maximum transmission for the fir wavelength used.

The line-integrated electron density was determined from the change of the optical pathlength when the plasma was switched off. In order to minimize the influence of mechanical vibrations and small fluctuations of the laser power, the following procedure was applied: the external mirror $M_3$ was put on a translation stage and moved by a stepper motor at a constant velocity of about $100 \mu m/s$ to produce a sequence of ten signal peaks. After five peaks the plasma was turned off, causing a slight increase of the optical path and hence a little jump toward the next maximum. By fitting the signal trains before and after the switching separately, the constant peak separation in stationary conditions corresponding to a mirror displacement by $\lambda/2$ and the reduced distance between the fifth and sixth peaks could be determined fairly accurately, yielding a resolution of the line-integrated electron density of $3 \times 10^{15} m^{-2}$. The improvement in comparison with Eq. (11) is at the expense of time resolution.

In a series of measurements the filling pressure and flow rate of the argon gas were kept constant at 1.33 Pa and 10 sccm, respectively, while the applied rf power $P_{\text{gen}}$ (13.56 MHz) was varied between 100 and 300 W. The plasma power $P_{\text{in}}$ (i.e., the power absorbed by the plasma) was somewhat lower than the applied power $P_{\text{gen}}$. The difference

FIG. 4. Experimental setup for first measurements in a GEC reference cell (two additional mirrors serve to focus the probing beam inside the plasma chamber to a diameter of 5 mm).

FIG. 5. Voltage of a Schottky-diode detector in dependence on $\Delta z$ for a laser wavelength of $\lambda_{\text{FIR}} = 432.6 \mu m$: (a) squares: signals measured in setup of Fig. 4, solid line: fit function according to Eq. (8); (b) first derivative of the fit function.

FIG. 6. Signal detected for mirror displacement at constant speed (for details see the text).
the interferometer measurements yield approximately twice as high as the probe measurements. A corresponding discrepancy has been reported by Miller et al.,\textsuperscript{16} who did a comparison between a microwave interferometer ($\lambda=3.8$ mm) and a Langmuir probe in an inductively coupled GEC reference cell under very similar conditions. In this work the interferometer data were found to be higher than the probe data by a factor of $\sim 1.5$. The difference to our results is most likely due to the rather wide microwave beam that averages over a larger plasma volume (including regions of less density).

A detailed discussion of possible reasons for the observed deviations between interferometer and probe measurements can be found in a paper by Overzet and Hopkins,\textsuperscript{15} who applied both techniques to a capacitively coupled GEC reference cell. Generally, the Langmuir probe appears to be prone to measurement errors when the ion mean-free path becomes smaller than the probe radius. However, owing to its high spatial resolution, the probe can provide a fair estimate of local variations of the electron density, whereas the interferometer yields the mean density along the beam path with high accuracy.

FIG. 7. Line-integrated electron densities versus plasma power $P_p$ as determined by the FIR interferometer and a scanning Langmuir probe.

was calculated as described by Miller et al.\textsuperscript{16} The results are shown in Fig. 7, where the line-integrated electron densities for different plasma powers $P_p$ are compared with concomitant measurements by means of a commercial Langmuir probe.\textsuperscript{18} The probe tip (8 mm in length and 50 $\mu$m in diameter) was moved across the plasma to cover the laser beam path and to provide sufficient data for calculating the line-integrated density $\int n_e dl$. The data analysis was based on the Orbital Motion Limited (OML) theory\textsuperscript{19} and made use of the probe current at the plasma potential. As can be seen in Fig. 7, the interferometer measurements yield approximately twice as high values as the probe measurements. A corresponding discrepancy has been reported by Miller et al.,\textsuperscript{16} who did a comparison between a microwave interferometer ($\lambda = 3.8$ mm) and a Langmuir probe in an inductively coupled GEC reference cell under very similar conditions. In this work the interferometer data were found to be higher than the probe data by a factor of $\sim 1.5$. The difference to our results is most likely due to the rather wide microwave beam that averages over a larger plasma volume (including regions of less density).

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V. FUTURE DEVELOPMENTS

The sensitivity of the interferometer can be further improved by the application of modulation techniques without changing the laser parameters or the detector system. For example, the optical path of the FIR beam can be altered periodically with the help of a polymer foil attached to a mechanical chopper. The thickness of the foil and the distance of the mirror $M_3$ are selected in such a way, that in the absence of plasma both working points (with and without foil) are located on different sides of the intensity peak at the same height (see Fig. 8 for an illustration). Under these conditions any change of the optical pathlength produces a change of the (differential) intensity, which is twice as high as it would have been without modulation, because the (absolute) intensities on both sides of the peak change in opposite directions by approximately equal amounts. In addition, the modulation reduces the error due to low-frequency fluctuations of the laser power, because both working points are affected in the same direction, leaving the differential measurement nearly unfluenced. Another quite obvious advantage of applying the modulation is the possibility to utilize lock-in amplifier techniques for a further improvement of the signal-to-noise ratio.

In order to overcome the inherent limitations of a mechanical chopper and to raise the modulation frequency to the MHz range, we are planning to replace the device by a semiconductor of high resistivity that is transparent in the FIR region. If such a semiconductor is irradiated by a diode-laser, emitting photons of an energy higher than the band gap, electrons will be lifted into the conduction band.\textsuperscript{20} Because these free electrons will change the refractive index of the semiconductor, it should be possible to manipulate the optical pathlength by varying the intensity of the diode laser, which is readily done by modulating the diode current even at a fairly high frequency. A first test of this scheme is in preparation.

FIG. 8. An illustration of the modulation technique (for details see the text).

ACKNOWLEDGMENTS

The authors would like to thank Professor J. Uhlenbusch, University of Duesseldorf, Germany for providing them with a grating-tunable CO$_2$ pump laser and various mechanical components that have been used for the construction of the FIR cavity. Support of this work by the Sonderforschungsbereich 591 of the Deutsche Forschungsgemeinschaft is gratefully acknowledged.


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