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# A New Approach to Impedance Atomic Spectroscopy*

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**Abstract.** Resonant laser-induced changes of the dielectric constant of atomic or molecular gases are detected as deviations in the oscillation frequency of a circuit including the sample itself as a component. Heterodyne detection allows high sensitivity. The new non-intrusive technique is demonstrated in the spectroscopic investigation of neon and sodium atoms.

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Impedance spectroscopy is based on the detection of small changes in the electrical properties of a gaseous sample in the presence of resonant radiation. When the sample is a dc discharge, changes in the flowing current are directly measured and the term "optogalvanic" (OG) is used. Although many other detection schemes have been applied, in most of the cases the discharge current is still the physical parameter which is probed by the detection apparatus. In radio frequency excited discharges, for example, the optogalvanic signal has been detected as a change in the current feeding the oscillator [1], by external coils inductively coupled to the plasma [2], or by internal passive electrodes [3].

We present here a new scheme for impedance spectroscopy. In this technique the physical parameter which is probed is not a current, but the dielectric constant of the medium constituting the sample. Although this approach is related to the previous ones, as the complex dielectric constant depends on the conductivity, it appears to be of much wider applicability. In fact, the atomic or molecular sample (not necessarily a discharge) is introduced as a component in a resonant circuit by capacitive or inductive coupling; any change in the dielectric constant of the sample will affect the circuit itself, changing its $Q$ or its oscillation frequency. The possibility of using a frequency heterodyne scheme for the measurement of these changes with respect to a stable unperturbed oscillator makes this technique extremely sensitive.

We have applied this technique as a demonstration to dc and rf discharges in Ne, and to a sample of sodium vapour. In the case of discharges we observed an improvement in the sensitivity by more than one order of magnitude compared with conventional OG detection; both Doppler-limited and sub-Doppler spectroscopy could be performed. Measurements on sodium, on the other hand, had the main purpose of demonstrating the wide applicability of this detection scheme, in particular when it is important that the probe does not intrude.

## 1. Experimental Setup

A schematic diagram of the experimental apparatus is shown in Fig. 1. The sample cell is placed between two parallel copper plates; they form a capacitor ($C \approx 3$ pF) which is connected as a component of a resonant circuit. A separate internal resonant circuit acts as a very stable, high-$Q$ reference. The resonant frequencies of both the circuits are probed by a radio-frequency signal ($\nu_p \approx 10$ MHz) generated by an internal oscillator which is periodically tuned ($\nu_t = 1$ kHz). The resonance of the internal, stable, reference circuit gives a start signal, while the resonance of the external circuit containing the sample gives a stop signal. The temporal separation of these two signals is converted into a voltage signal and gives a measure of the difference $\Delta f$ between the two frequencies of resonance. For a change of 0.1 pF in the value of the capacitance an

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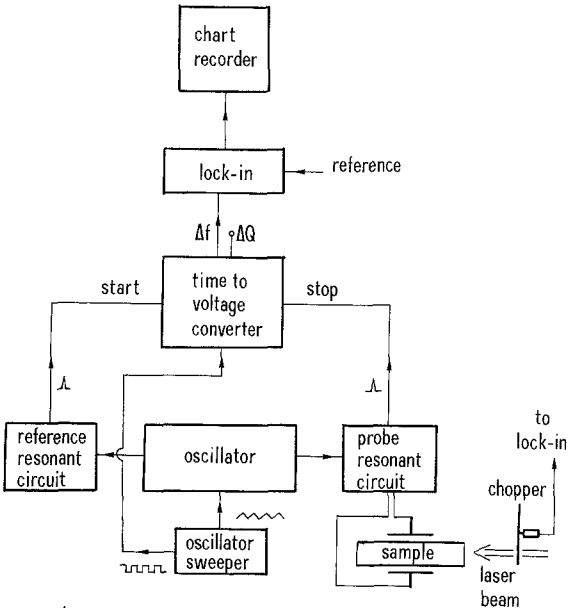


**Fig. 1.** Schematic illustration of the new detection method. The sample is part of the capacitance of the probe resonant circuit. Heterodyne with the reference resonant circuit allows high sensitivity measurement of laser-induced variations in the oscillation frequency

output signal of about 100 mV results; this gives a measure of the obtainable sensitivity. In a similar way the changes in the merit factor $Q$ of the external circuit are measured. To improve the sensitivity, lock-in detection has been used; the laser beam was mechanically chopped and sent through the sample cell. The $\Delta f$ and $\Delta Q$ signals were measured by a lock-in amplifier with an integration time of 0.3 s.

The neon transitions investigated in this experiment start from high-energy excited levels. The use of an electrical discharge created an adequated population of these levels and, at the same time, allowed the detection of conventional OG signals for comparison. The schemes for the excitation of the discharge and the detection of the OG signal have been described elsewhere [4].

In the case of sodium the sample was a sealed pyrex bulb containing or not neon as a buffer gas. Here resonant fluorescence light was detected for comparison instead of the OG signal.

## 2. Results and Discussion

Figure 2 shows a typical spectrum of neon transitions between excited levels, in the 580–630 nm wavelength range, obtained by the detection technique described above. A cw dye laser was operated at a bandwidth of 20 GHz over the tuning range of rhodamine 6G dye; the laser beam was mechanically chopped at a frequency of 80 Hz and directed into a rf excited discharge operated at a pressure of 1 Torr. The probe capacitor was formed by two parallel copper plates ($2 \times 4$ cm$^2$) placed on either side of the discharge tube. Both $\Delta f$ and $\Delta Q$ signals could be recorded with a good signal-to-noise ratio. The $\Delta f$ signal is reported in the figure. Similar results were obtained with dc positive column discharges.

The spectra we obtained are the same as the simultaneously recorded OG signals (not reported in the figure) from the point of view of relative intensities and sign of the lines. As we shall discuss later, the

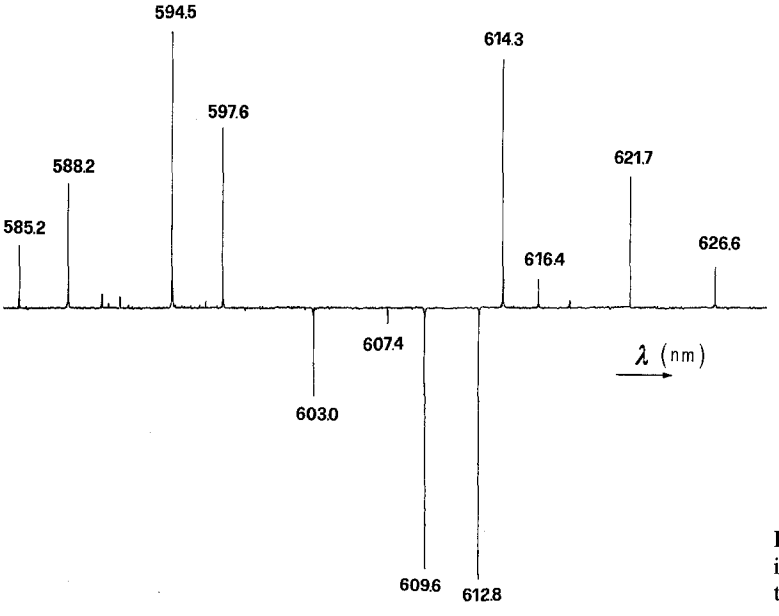


**Fig. 2.** Broadband recording of the neon transitions in the rhodamine 6G wavelength range using the technique described in Fig. 1

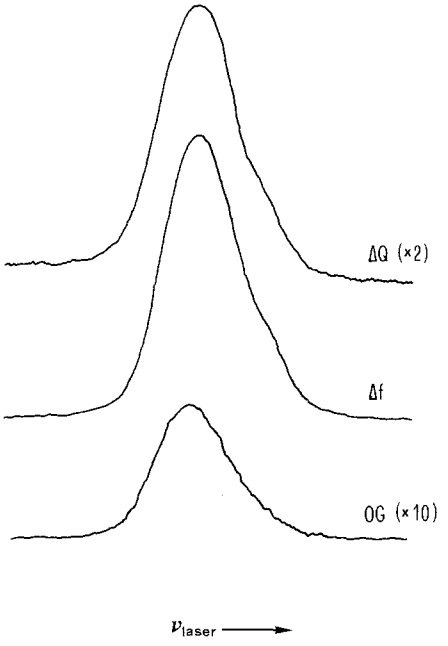


**Fig. 3.** Simultaneous recordings of the Doppler broadened 607.4 nm Ne transition obtained with the new $\Delta f$ and $\Delta Q$ techniques and with conventional optogalvanic detection. Excited levels were populated in a He–Ne (4.8/0.2 Torr) discharge. Note the change in the sensitivity scale

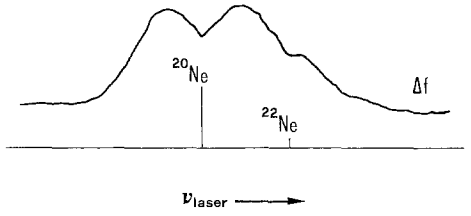


**Fig. 4.** Saturated spectroscopy of Ne with the $\Delta f$ detection method

sample dielectric constant depends on the density of free electrons $n_e$. Positive or negative signals in Fig. 2 hence reflect the sign of the laser induced variation of $n_e$. To characterize the new detection method better we studied the weak 607.4 nm ($1s_4$–$2p_3$) Ne transition using a narrow band ring dye laser (bandwidth $<5$ MHz). Both Doppler-limited and saturation sub-Doppler signals could be recorded with our technique; their intensities generally exceeded the ones obtained with OG detection by one order of magnitude. The improvement in sensitivity is evident in Fig. 3 where Doppler-limited recordings are reported, obtained with $\Delta f$ and $\Delta Q$ detection and with OG detection. The scale used for the lock-in amplifier was respectively 10 times and 2 times more sensitive for OG and $\Delta Q$ signals than for $\Delta f$ signals. For the measurement in Fig. 4 two counterpropagating beams were sent into the discharge cell; the resulting Lamb dips can be observed in the Doppler profile, as recorded with the new $\Delta f$ method. This is not, of course, the best scheme to observe sub-Doppler signals in a discharge, because of the presence of the large Doppler background. For instance, intermodulated [5] or polinex [6] spectroscopy would be much more suitable; but, because of the limitation on the laser beam chopping frequency, determined by the relatively low tuning frequency $\nu_t$ of the probe oscillator, we could not observe very good intermodulated signals with either $\Delta f$ or $\Delta Q$ detection or with OG detection. It should be possible to improve the present apparatus for sub-Doppler investigations by increasing the tuning frequency.

In all the cases we considered, the signals we obtained by the new detection method seemed to correspond to the OG signals. This observation suggests that the main mechanism involved in the generation of the $\Delta f$ and $\Delta Q$ signals is, as in the case of OG signals, the change in the number of electrons in the sample caused by the interaction of the atoms with laser radiation. To test this hypothesis we applied our detection scheme to a sample of sodium, exciting the strong 589.0 nm resonance transition by laser radiation. Strong $\Delta f$ and $\Delta Q$ signals could be observed also in this case but only if the cell contained several Torrs of neon buffer gas and the cell itself was heated to about 300° C. It is well known [7] that in these conditions associative ionization is very effective in ionizing sodium atoms, producing free charges in an initially neutral sample. This confirms our hypothesis that a change in the number of free charges under laser irradiation is the origin of the signals we observe by our technique. In fact, the variation of the complex dielectric constant $\Delta\varepsilon$, resulting from a change $\Delta n_e$ in the free electron density, can be expressed as:

$$\Delta\varepsilon = -m\omega/(\omega - i\beta)\Delta n_e(4\pi e^2), \tag{1}$$

where use has been made of the well known expression for the Lorentz conductivity of a medium [8]. In the present case $\omega \equiv 2\pi\nu_p$ and $\beta$ is the electron collision frequency.

In our system the capacitor containing the sample can be considered as an ideal capacitor in parallel with a resistor; the resistor takes account of the leaks between the metallic plates and the absorption of the probing 10 MHz radio frequency by the plasma in the cell. Any variation in the density of charges produces changes in the real and imaginary parts of the complex dielectric constant, that is changes in the values of the equivalent capacitance and resistance. Changes in the resonance frequency and in the $Q$ of the circuit including the capacitor result. The technique we have described consists, in fact, in the accurate measurement of this variation.

## 3. Conclusions

We have demonstrated the possibility of performing optical impedance spectroscopy on samples of different type by a very simple new technique. The sensitivity obtainable can be very high and seems to be limited only by shot noise in the sample itself.

Because of its wide applicability and non intrusiveness, this technique lends itself to the study of processes involving the formation of unstable molecules or reactive atoms. A possible application is, for example, the diagnosis of flames where detection of photoelectrons has already proved to be a useful tool [9]. The new technique should be more sensitive and offers the advantage of not perturbing the charged particles kinetics by external static fields.

A possible improvement could be achieved by increasing the probe oscillator frequency. For example, going from the rf range used here to the microwave region, by replacing the capacitor with a suitable cavity containing the sample would increase the $Q$ of the system considerably and then the detection sensitivity.

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