Doppler-Free Intermodulated Optogalvanic Spectroscopy

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Intermodulated optogalvanic spectroscopy, a new method of Doppler-free saturation spectroscopy, is described. Preliminary values of 725±4 MHz for the ^{3}He $3^3D_2(F=\frac{3}{2})$-$3^3D_1(F=\frac{1}{2})$ hyperfine splitting, and of 5830±6 MHz for the $3^3D_1(F=\frac{1}{2})$-$3^3D_1(F=\frac{3}{2})$ hyperfine splitting obtained using this method are reported. Intermodulated optogalvanic spectroscopy is limited in sensitivity only by shot noise in the direct current sustaining the discharge, and compares favorably in sensitivity with other Doppler-free methods.

In this Letter we describe a Doppler-free version of optogalvanic spectroscopy which promises to be a powerful tool for studying optical transitions in gaseous discharges. Optogalvanic detection is an especially convenient spectroscopic technique because many visible and infrared transitions between excited states are readily accessible in a discharge. Furthermore, the spectra of nonvolatile materials are conveniently observed using optogalvanic detection.[1] Although the Lamb dip has been observed optogalvanically,[2] most previous studies using this detection technique have been Doppler limited. Doppler-free intermodulated optogalvanic spectroscopy (IMOGS) as described herein, is closely related to intermodulated fluorescence spectroscopy,[3] but uses the detection scheme of optogalvanic spectroscopy. The sensitivity of IMOGS is fundamentally and experimentally limited only by shot noise in the direct current sustaining the discharge. Doppler-free IMOGS complements other Doppler-free methods,[4-6] and may prove to be most useful in regions of the spectrum where other methods are limited in sensitivity by laser noise, detector noise, or lack of high-quality polarizers. We report preliminary measurements of hyperfine splittings in the 3^3D level of ^{3}He obtained using IMOGS on the ^{3}He 2^3P-3^3D transition.

Optogalvanic spectroscopy is based on the detection of an impedance change in a gaseous discharge, produced by irradiation with a laser tuned to an atomic transition occurring within the discharge. This change in discharge impedance is detected as a change in voltage across a ballast resistor by a lockin amplifier tuned to the chopping frequency of the laser. A spectrum with ordinary Doppler broadening is obtained by tuning the laser across an optically thin transition.

Doppler-free IMOGS has a similar detection scheme to Doppler-limited optogalvanic spectroscopy. In IMOGS, however, the laser beam is split into two components of roughly equal intensity. One beam is chopped at a frequency f_1 and sent through the discharge. The second beam is chopped at a different frequency f_2 and sent through the discharge in the opposite direction. Because the two beams propagate in opposite directions, they interact, in general, with different velocity groups of atoms under the Doppler profile of a given transition. When the laser is tuned within one homogeneous width of line center, the two beams interact with the same group of atoms. Nonlinearities caused by the two beams acting to saturate the same atoms then give rise to Doppler-free optogalvanic signals at sum (f_1+f_2) and difference (f_1-f_2) modulation frequencies; the sum frequency is conveniently detected with a lockin amplifier.

We have studied the 2^3P-3^3D transition in ^{3}He as a demonstration of IMOGS. This transition is of particular interest because, to our knowledge, no Doppler-free study of it has been reported, and only one limited radio-frequency study of the 3^3D level has been reported in which the $3^3D_3(F=\frac{5}{2})$-$3^3D_3(F=\frac{7}{2})$ splitting was measured.[7] Furthermore, the structure of this ^{3}He level is amenable to precise theoretical calculation.

A schematic diagram of the apparatus is shown in Fig. 1. The dc-discharge cell is a commercial

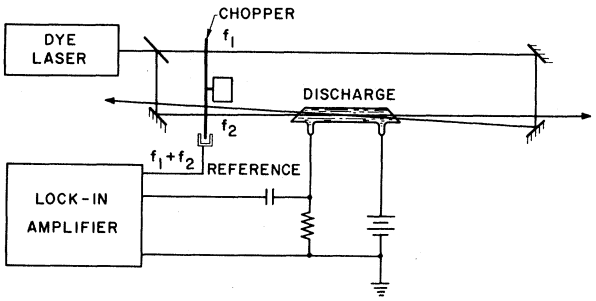

FIG. 1. Experimental apparatus for intermodulated optogalvanic spectroscopy.

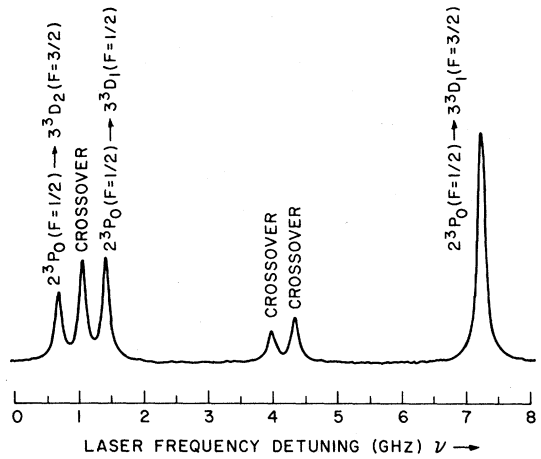

FIG. 2. Intermodulated optogalvanic spectrum of part of the ^{3}He 2^3P-3^3D transition at 587.5 nm.

He-Ne laser tube (Jodon Engineering Associates CE 16.7-2CS) with a ^{3}He partial pressure of 1.62 Torr and a Ne partial pressure of 0.23 Torr. It is operated with a discharge current of 5.5 mA and a tube voltage of 700 V. Tunable laser radiation is provided by an actively stabilized Rhodamine 6-G dye laser (Coherent Inc. 599-21). The dye-laser beam is split into two beams of roughly equal intensity. One beam is chopped at a frequency of 600 Hz and the other at 840 Hz by the same mechanical chopper, which also provides a reference signal at the sum frequency of 1440 Hz. Changes in discharge-tube current are determined by measuring the voltage across a 100-kΩ ballast resistor with a lockin amplifier tuned to 1440 Hz (PARC HR-8). The dc voltage across the ballast resistor is blocked by a coupling capacitor. The noise current in a 1-Hz bandwidth at 1440 Hz is on the order of 10^{-11} A, consistent with the shot-noise limit for the given conditions.

A typical partial scan of the 587.5-nm 2^3P-3^3D multiplet of ^{3}He is shown in Fig. 2. Only the part of the multiplet connected to the 2^3P_0 level is shown although a large number of components and crossovers associated with the 2^3P_1 and 2^3P_2 levels can also be observed. The laser power is attenuated to 0.3 W/cm^2 to avoid power broadening. The observed linewidth of 130 MHz is due primarily to pressure broadening at the ^{3}He partial pressure of 1.62 Torr, and is consistent with previous measurements.[8]

TABLE I. Hyperfine splitting of the ^{3}He 3^3D level.

	Experimental (MHz)	Theoretical (MHz)
$3^3D_2(F=3/2)$-$3^2D_1(F=1/1)$	725 ± 4[a]	750[b]
$3^3D_1(F=1/2)$-$3^3D_1(F=3/2)$	5830 ± 6[a]	5760[b]
$3^3D_3(F=5/2)$-$3^3D_3(F=7/2)$	55.8 ± 0.8[c]	0[b]

[a] This work. [c] Ref. 7. [c] Ref. 7.

Preliminary values for hyperfine splittings of the 3^3D level are given in Table I, with quoted uncertainties of 1 standard deviation. Calculated values are shown for comparison.[9] We are currently investigating possible systematic errors due to pressure shifts, and attempting to resolve the rest of the multiplet.

It is interesting to compare the sensitivity of IMOGS to that of Doppler-free saturated absorption spectroscopy. In IMOGS, the signal is detected as a change in the discharge current, rather than a change in probe-beam intensity. Both signal and noise are very different in form from those in saturated absorption spectroscopy. The use of a two-level model in the Doppler-broadened, weakly saturated limit greatly facilitates the comparison. Although the ^{3}He 2^3P-3^3D multiplet is not a simple two-level system, we use parameters from the experiment to quantify the comparison. Let n_u and n_l be the density of atoms in the upper and lower levels, respectively, of the transition under steady-state discharge conditions. The change in these densities at line center modulated at the sum frequency of the laser modulation frequencies is

$$\Delta n_u - \Delta n_l \equiv 2\,\Delta n = (n_u - n_l)(\Delta\nu_H/\Delta\nu_I) I_1 I_2/(8 I_{\rm sat}{}^2), \quad (1)$$

where $\Delta\nu_H$ and $\Delta\nu_I$ are the homogeneous and inhomogeneous widths, respectively, I_1 and I_2 are counter-propagating beam intensities, and $I_{\rm sat}$ is the saturation intensity. For simplicity we have ignored the difference in level degeneracy and assumed that the discharge is optically thin. Equation (1) follows from a standard derivation of the change in absorption or gain[3,10] due to saturation by including a factor, $\Delta\nu_H/\Delta\nu_I$, representing the fraction of atoms in the velocity distribution which interact with the laser beams.

Recombination in the positive column of a low-pressure glow discharge is negligible, although elec-

trons are lost by ambipolar diffusion to the wall.[11] We assume as an approximation that all excess electrons are collected with no gain. The signal current is

$$i_s = eVR\Delta n, \tag{2}$$

where e is the electron charge, V is the volume of the positive column perturbed by the laser beams, and R is the difference in ionization rates of the upper and lower levels. The limiting noise in IMOGS is due to fluctuations in discharge current, rather than fluctuations in laser intensity. We observe shot-noise–limited current fluctuations at audio modulation frequencies in a carefully designed discharge tube. The rms shot-noise current in a bandwidth B due to a direct current i_{dc} is[12]

$$i_N = (2ei_{dc}B)^{1/2}. \tag{3}$$

The signal (S) to noise (N) ratio in IMOGS is given by

$$(S/N)_{IMOGS} = VR(n_u - n_l)(\Delta\nu_H)I_1I_2/[16(\Delta\nu_I)I_{sat}^2(2Bi_{dc}/e)^{1/2}]. \tag{4}$$

In the widely used method of Doppler-free saturated absorption spectroscopy, an intensity modulation ΔI_1 on a laser beam of intensity I_1 is produced by the presence of a chopped saturating beam of intensity I_2. The signal at line center is

$$\Delta I_1/I_1 = \alpha l I_2/4 I_{sat}, \tag{5}$$

where α is the absorption coefficient of the sample of length l, and I_{sat} is the saturation intensity of the transition. The signal-to-noise ratio in saturated absorption spectroscopy is

$$(S/N)_{SAS} = \alpha l I_2/4\delta I_{sat}, \tag{6}$$

where δ is the fractional intensity fluctuation within the detection bandwidth.

The signal-to-noise ratios of IMOGS and of saturated absorption spectroscopy are then related by

$$(S/N)_{IMOGS}/(S/N)_{SAS} = (V/l)(R/A)(I_1/h\nu)\,\delta/[4(2B\,i_{dc}/e)^{1/2}], \tag{7}$$

where A is the Einstein coefficient for the transition, h_ν is the photon energy, and I_{sat} has been approximated as

$$I_{sat} = A\,h\nu\Delta\nu_H(n_u - n_l)/\alpha\Delta\nu_I. \tag{8}$$

In the approximation for I_{sat} it is assumed that radiative decay is the primary mechanism restoring steady-state–discharge populations. In the case of a transition with a much smaller A coefficient than that of the ^{3}He transition, the A coefficient in Eqs. (7) and (8) should be replaced by $1/2\tau$, where τ is the inversion lifetime.[10] The inversion lifetime is determined by collisional and radiative processes. For the ^{3}He 2^3P-3^3D transition, A is 7×10^7 sec^{-1},[13] and R is approximated as 2.7×10^6 sec^{-1}, the rate of associative ionization of the 3^3D level at a He partial pressure of 1.62 Torr.[14] The rate of associative ionization of the 2^3P level is zero because it is well below threshold. The presence of Ne in the commercial discharge tube is neglected. He 3^3D atoms can chemionize ground-state Ne atoms whereas He 2^3P atoms cannot; so reactions involving Ne atoms may enhance the optogalvanic effect. The difference in electron-impact ionization rates should in general be included in R. In the experiment, V/l is 0.03 cm^{-2}, I_1 is 0.3 W/cm^2, i_{dc} is 5.5 mA, and B is 1 Hz. For typical cw dye lasers, the fractional intensity fluctuation δ is far from the shot-noise limit. At the least-noisy modulation frequencies, we observe a δ of the order of 10^{-4} with a detector bandwidth of 1 Hz. Using these parameters, we estimate that $(S/N)_{IMOGS}/(S/N)_{SAS} = 100$.

It is apparent that with favorable reaction rates IMOGS can be more sensitive than saturated absorption spectroscopy. Other spectroscopic techniques have been devised to improve the sensitivity of saturated absorption spectroscopy. The fractional intensity fluctuations δ may be reduced by intensity stabilizing the laser, or their effect may be reduced by a differential-detection scheme.[4] Saturated interference spectroscopy[5] and Doppler-free laser polarization spectroscopy[6] have been demonstrated. Each of these techniques has its own set of performance requirements for optics and detectors which must be met if the technique is to provide a substantial improvement in sensitivity over saturated absorption spectroscopy. IMOGS may be most useful in regions of the spectrum where low-noise detectors, interferometric-quality optics, or high-quality polarizers are unavailable. IMOGS is also an ex-

tremely simple technique to use, requiring a bare minimum of optical components.

The possibility of performing Doppler-free intermodulated spectroscopy in an electrodeless radio-frequency discharge deserves careful investigation. Superregenerative radio-frequency detection may provide a useful form of optogalvanic detection.

In conclusion, intermodulated optogalvanic spectroscopy has been demonstrated to be a powerful new method of performing Doppler-free saturation spectroscopy in a discharge. We report preliminary hyperfine splittings of the 3^3D level of ^{3}He obtained using this method. Finally, our calculations indicate that IMOGS compares favorably in sensitivity to other Doppler-free techniques.

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Extended Two-Level Theory of the Exponential Index of Multiphoton Processes

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A nonperturbative formula is given for the r-photon-resonant, $(r+p)$-photon ionization rate of an atom and from it is derived an expression for the exponential index.

The exponential order, or index, of an atomic multiphoton process is defined by

$$K = \partial \log(\text{rate})/\partial \log(\text{intensity}). \quad (1)$$

It provides a convenient way to parametrize experimental data[1] because it does not require an absolute measurement of either the observed rate or the inducing laser's intensity. Clearly if the rate is simply proportional to the intensity, $R = \text{const} \times I^n$, then one has $K = n$, for all I. However, it is found experimentally that not only R but K also depends on I. This occurs when the laser frequency is such that an integer number of laser photons and an allowed atomic transition are near to resonance.

In spite of the phenomenological utility of the index K, there is no satisfactory theory for it. A satisfactory theory should be able to (a) apply both to ionization and to purely bound-bound multiphoton processes; (b) express K analytically and in terms of parameters significantly simpler than K itself; (c) treat the region very near to resonance; (d) apply at laser intensities both above and below the saturation level; and (e) accommodate standard line-broadening mechanisms (spontaneous emission, collisions, laser bandwidth, Doppler effect). The purpose of this note is to present a theory for multiphoton rates that

