Laser spectroscopy of calcium in hollow-cathode discharges

R. L. Cavasso-Filho

Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, C.P. 6165, Campinas, São Paulo, 13083-970, Brazil

A. Mirage

Instituto de Pesquisas Energéticas e Nucleares C.P. 11049, São Paulo, SP 05422-970, Brazil

A. Scalabrin, D. Pereira, and F. C. Cruz

Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, C.P. 6165, Campinas, São Paulo, 13083-970, Brazil

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We investigated the use of hollow-cathode discharges for high-resolution and high-sensitivity spectroscopy, using atomic calcium. Spectra with sub-Doppler resolution of CaI transitions at 423 (resonant), 610, 612, 616, 645, 657 (intercombination), and 672 nm were obtained by optogalvanic saturation spectroscopy in lamps filled with argon (0.6 and 2.5 Torr) and krypton (0.6 Torr). A Doppler background that is due to velocity-changing collisions, which may severely limit the resolution, can be greatly reduced by the choice of buffer gas. Sub-Doppler linewidths comparable with those achieved in atomic beams have been obtained, making a properly chosen hollow-cathode lamp a convenient tool for high-resolution spectroscopic experiments, providing wavelength references for laser frequency tuning. The sensitivity of optogalvanic detection and the excitation of most electronic levels by the discharge make these lamps attractive also for investigating weak and excited level transitions with the use of a simple experimental setup.

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1. INTRODUCTION

Atomic calcium ($Z = 20$) has been shown to be an attractive element for high-resolution and precision spectroscopy. Its strong $^1S_0$–$^1P_1$ resonant transition, at 423 nm, permits laser cooling and trapping,[1–7] which greatly reduce systematic shifts and spectral-broadening effects. Calcium also offers a narrow and weak $^1S_0$–$^3P_1$ intercombination transition, at 657 nm, which has been used in high-resolution spectroscopy,[8] atomic interferometry, [9,10] and precision measurements[11] and as a reference for high performance atomic frequency standards in the optical region.[12,13] Because the most abundant isotopes of alkali earth elements (for example, ^{40}Ca) have no hyperfine structure, they can be interesting also for studying cold collisions,[14] which in this case could have the advantage of a potentially simpler theoretical analysis. These studies, combined with the possibility of reaching ultracold temperatures as was recently demonstrated for strontium,[15] will be important for the eventual achievement of quantum degenerate regimes in alkali earth atoms.

In experiments that involve high-resolution or precision spectroscopy in atomic beams or cold atoms, lasers often may have to stay tuned for long times near the atomic transitions of interest. Interferometric wavemeters can be used to tune the laser frequency with an accuracy of 10^{-6} or better, but they are not convenient for frequency locking or when several lasers are required. When the transition of interest departs from the ground state, frequently an auxiliary vapor cell of the same element is used for locking the laser frequency, because it represents a cheap and easily fabricated sample of atomic vapor in which the resonances can be detected with a good signal-to-noise ratio. In fact, for strong transitions such as resonant transitions in alkali elements, fluorescence can easily be detected in an atomic beam or a magneto-optical trap (MOT). However, such is not the case for narrow and weak transitions, such as the calcium intercombination transition at 657 nm ($\Delta\nu \approx 400$ Hz; $A_{21} = 2600\ \mathrm{s}^{-1}$), which require either high-sensitivity techniques for detection in cells or some special care for detecting fluorescence in a beam or a MOT. For instance, a calcium reference cell would have to be heated at least to 400 °C. Tuning to or detecting a weak and narrow transition can be particularly cumbersome when the transition's wavelength is not precisely known. When the optical transition of interest involves excited levels, a vapor cell of the same element is not useful. Alternatives can include some sort of excitation of these levels, for example, by creation of a rf plasma, or the use of a cell with a different atom or molecule as the wavelength reference. A good example is iodine (I_2), which has thousands of closely spaced narrow lines. They are strong in the visible but weak in the near infrared, requiring heating and the use of long cells.

Here we investigate the use of hollow-cathode lamps

(HCLs) as an efficient way to produce a high-density vapor of calcium atoms for spectroscopy. Combined with the optogalvanic effect, they represent a simple and effective alternative for atomic detection with high sensitivity and detection of weak transitions. The lamp design can permit the use of laser techniques that employ counterpropagating beams, to eliminate broadening caused by the first-order Doppler effect. Because the electric discharge can excite most electronic levels of the neutral atom, beside producing ions these lamps offer the natural possibility of investigating transitions between excited levels of neutral atoms and ions. We obtained spectra of CaI transitions at 423 (resonant), 610, 612, 616, 645, 657 (intercombination), and 672 nm, with Doppler-limited and sub-Doppler resolution, using saturation spectroscopy combined with optogalvanic detection.[16] We used two lamps filled with argon, at pressures of 0.6 and 2.5 Torr, and one lamp filled with krypton, at a pressure of 0.6 Torr. For the lamps filled with argon, velocity-changing collisions (VCCs) are responsible for a large Doppler pedestal that broadens the line and severely limits the resolution. This pedestal is reduced in the case of the lamp filled with krypton, for which sub-Doppler resonances with widths as small as 25 MHz were observed. A calcium lamp may, in particular, be a convenient tool in other experiments with this element, such as in laser cooling and trapping, for which it can be used as a vapor cell, supplying wavelength references for laser frequency tuning and locking but not restricted to transitions from the ground state.

2. EXPERIMENTAL DETAILS

Two of the advantages of a hollow-cathode discharge compared with other kinds of glow discharge are the possibility of operation at large currents (up to a few amperes) and the high sputtering efficiency that can be obtained, making this type of discharge well suited for investigations of refractory elements.[16] The experimental setup is simple, with no need for an oven to produce samples at temperatures near 1000 °C or higher. We used three homemade hollow-cathode lamps that are similar to others that we reported previously.[17] A schematic diagram, along with electrical connections, is included in Fig. 1. The lamps are 16 cm long and were made with a Pyrex tube of of 3.5-cm diameter. They were filled with argon (at pressures of 2.5 and 0.6 Torr) or krypton (at a pressure of 0.6 Torr). Two ring titanium anodes were placed at 0.5 cm on each side of the cylindrical steel cathode, where a small calcium tube was inserted. The lamps could be operated over 100 mA, with typical voltages of 100 V, and therefore had a cathode that was water cooled. A ballast resistor of 3.0 kΩ and a 400-V, 150-mA power supply were used. We verified the efficient production of calcium vapor by sputtering through emission spectra that show most of the neutral calcium lines. As Fig. 1 shows, a spectrum analyzer was used for a study of the electrical noise in the discharge, which is important for optogalvanic detection.[18] For example, for the lamp filled with argon at 2.5 Torr, the spectral distribution was verified to be flat and at the shot level for currents above 30 mA and ballast resistances higher than 0.5 kΩ.

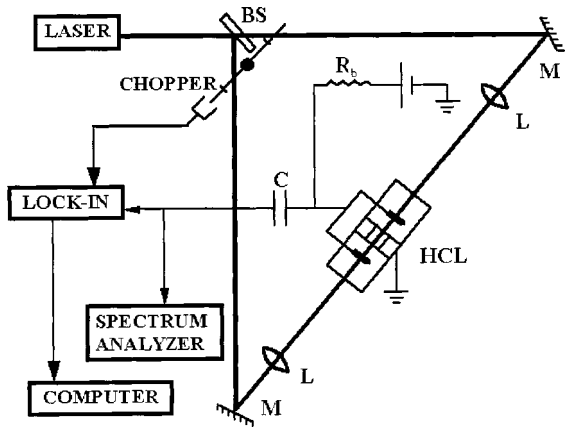

Fig. 1. Schematic diagram of intermodulated optogalvanic spectroscopy. The lamp construction diagram with electrical connections and a setup for noise measurement[18] are also included. BS, beam splitter; Ls, lenses; Ms, mirrors; R_b, ballast resistor; LOCK-IN, lock-in amplifier, C, capacitor.

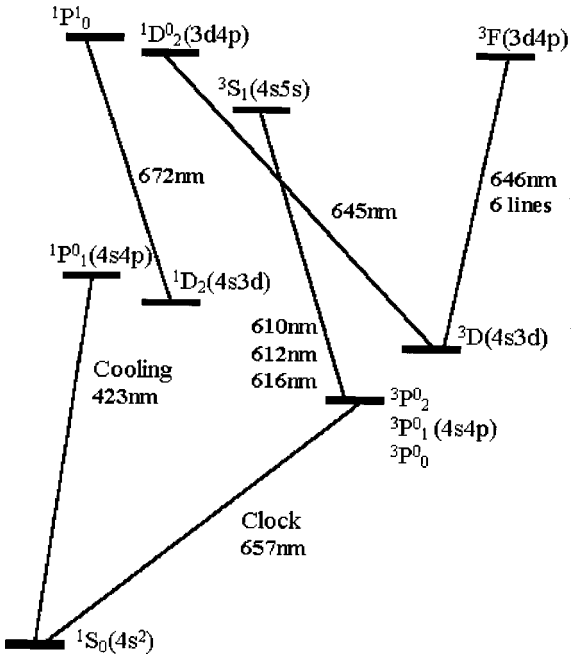

Fig. 2. Simplified level diagram of CaI.

We obtained optogalvanic spectra for the following transitions of CaI (a simplified diagram is shown in Fig. 2), listed here in decreasing order of transition rate (Einstein coefficient A_{21}) or absorption cross section: $^1S_0$–$^1P_1$ (423 nm, $A_{21} = 2.13 \times 10^8\ s^{-1}$, resonance transition; used for laser cooling and trapping), $^3P_{0,1,2}$–$^3S_1$ (610, 612, and 616 nm, $A_{21} = 7 \times 10^7\ s^{-1}$), $^1D_2$–$^1P^o_1$ (672 nm, $A_{21} = 1.5 \times 10^7\ s^{-1}$, used as repumping in a MOT[19] to depopulate the metastable $^1D_2$ level), ^3D–$^1D^o_2$ (645 nm, $A_{21} = 1.4 \times 10^6\ s^{-1}$), and $^1S_0$–$^3P_1$ (657 nm, $A_{21} = 2600\ s^{-1}$, intercombination; reference for optical frequency standard). Using the setup of Fig. 1, we obtained both Doppler-limited and sub-Doppler spectra for these transitions. Sub-Doppler resolution was achieved by saturation spectroscopy and either the configuration of intermodulated optogalvanic spectroscopy[16] or the setup in which a Lamb dip is observed[20] (also with optogalvanic

detection). In the first configuration, two counterpropagating laser beams with equal intensities are amplitude modulated at frequencies $f1$ and $f2$, and the optogalvanic signal is recorded at the sum frequency. In the second configuration, a weakly modulated probe beam is detected in the presence of a nonmodulated counterpropagating pump beam. To excite the red transitions we used commercial single-frequency dye lasers (linewidth, $\Delta\nu \leq 1$ MHz). For the resonant blue transition we used a homemade Ti:sapphire laser ($\Delta\nu \approx 100$ kHz), which was frequency doubled in a 1-cm-long potassium niobate crystal placed inside a power-enhancement cavity.[21] For 300 mW of power at 846 nm we could generate 80 mW at 423 nm. The transitions at 616 and 645 nm were at the edges of the gain curves of the dye lasers, which used R6G and DCM dyes, respectively, and because of the low laser power we obtained only spectra with poor signal-to-noise ratios (S/N) for these transitions.

3. RESULTS AND DISCUSSION

Although, in emission spectra taken for the 2.5-Torr Ca–Ar lamp, the relative intensities for the transitions that we investigated are proportional to their respective transitional probabilities (see Table 1), the magnitudes of the optogalvanic signals (OGSs) follow a different sequence, which we again list in decreasing order: 423, 610, 672, 612, and 657 nm. To give an idea of the numbers involved in the optogalvanic detection of these transitions (in the 2.5 Torr Ca–Ar lamp), we show in Table 2 extrapolated values for the laser power required for a S/N equal to 10, at line center and $I = 70$ mA, with amplitude modulation of the laser by a chopper (frequencies from 1 to 2 kHz) and phase-sensitive detection by a lock-in amplifier with an integration time constant of 300 ms. For the resonant transition the result is shown for $I = 30$ mA for reasons explained below. The magnitude of the OGS can be higher for transitions for which the lower level is metastable,[22] which is the case for the transitions at 610, 616, and 672 nm. This result explains the above sequence for the magnitude of the OGS.

In HCLs the atomic excitation depends on the electron temperature, which is higher than the gas temperature and is not linked to it. In all Doppler-limited spectra of the transitions that we investigated, the Doppler widths (FWHM) obtained from fittings to a Voigt profile varied from 1.3 to 2.2 GHz. These widths correspond to temperature ranging from 750 to 1200 K for CaI, within the current range in which we have mostly operated the lamps (up to 100 mA). Figure 3 shows a Doppler spectrum for the 423-nm line (FWHM of 2.2 GHz) in the 2.5-Torr Ca–Ar lamp, obtained simultaneously with a fluorescence spectrum in an atomic beam. This spectrum was obtained with the laser transverse to the atomic beam and thus has only a residual Doppler width of ~70 MHz, because of the divergence of the atomic beam. The difference between the positions of the maxima of the two curves or between the maximum of the beam spectrum and the center of gravity of the lamp spectrum was smaller than 34 MHz, the radiative linewidth of the 423-nm resonant transition. This indicates that a possible collisional shift will be relatively small for this transition.

An interesting point with respect to the magnitude of the OGS concerns its dependence on the lamp current and the geometry. The impedance change is caused mainly by atoms that are absorbing light between the electrodes, where the discharge is concentrated. The OGS is the corresponding voltage change measured across the electrodes (or across the ballast resistor). However, there

Table 1. Relative Intensities of Some CaI Lines Observed in Emission Spectra from the 2.5-Torr Ca–Ar Lamp Normalized to the Intensity of the Resonant Transition at 70 mA[a]

	Relative Intensity		
Calcium Line (nm)	30 mA	50 mA	70 mA
423	238	466	1000
445	20.2	50.6	170
610	0.25	0.63	2.03
612	0.66	1.75	5.57
616	1.06	2.78	8.61
657	0	0.08	0.15
672	0.10	0.18	0.38
715	0.28	0.48	0.86

[a] A double monochromator with a resolution of 0.1 Å was used.

Table 2. Laser Power Required for a S/N of 10 in Optogalvanic Detection of some CaI Transitions[a]

Calcium Line (nm)	Laser Power (mW) for an Optogalvanic S/N of 10
423	1.0
610	0.38
672	1.4
612	1.9
657	20

[a] Line center, $I = 70$ mA, except for the resonant line center, for which $I = 30$ mA; 2.5 Torr Ca–Ar lamp. The laser was amplitude modulated by a chopper at frequencies of 1–2 kHz.

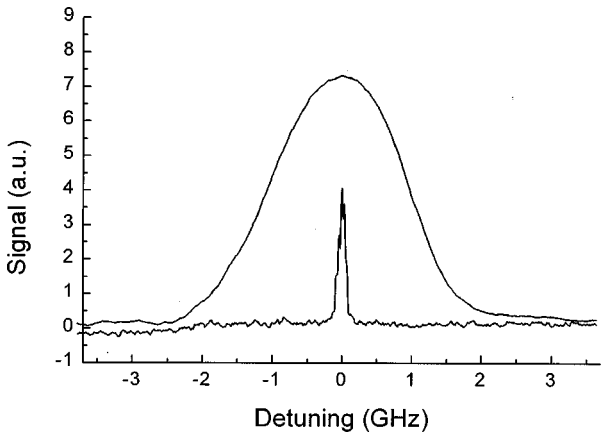

Fig. 3. Spectra of the CaI resonant transition at 423 nm, obtained simultaneously in the HCL (2.5 Torr Ca–Ar, $I = 30$ mA) with optogalvanic detection (upper curve) and in an atomic beam with detection of fluorescence (lower curve).

may be sputtered atoms all over the lamp, which absorb light but essentially do not contribute to the OGS. Usually the OGS increases with current, following the corresponding increase in density for the sputtered atoms. However, it is common to observe a decrease in the magnitude of the OGS above a certain current, which can be explained if we consider that this increasing atomic density is also creating more atoms outside the region between the electrodes. The increase in density of these atoms is of course followed by an increase of their absorption coefficient, with the result that they can absorb an appreciable fraction of the laser light before it reaches the region between the electrodes. This will cause a decrease in the magnitude of the OGS. Quantitative analysis of the magnitude of the OGS as a function of current, in lamps with designs similar to ours, should take this effect into account.[23] Figure 4 shows an example for the calcium resonant transition, where simultaneous Doppler-limited optogalvanic and optical absorption spectra have been recorded for the 2.5-Torr Ca–Ar lamp. The spectra in Fig. 4, obtained at 30 mA, show absorption by the lamp of 75% at the line center, which increases to 100% for 40 mA. At 50 mA the central portion of the atomic line, observed in optogalvanic detection, also starts to show attenuation, because of absorption from atoms located before the electrodes. At 80 mA this attenuation at the line center is also 100% for the OGS. For these current levels, the fluorescence for this strong transition in the lamp can easily be observed. Thus, just by changing the current in a HCL, for a given atomic line we can significantly change the absorption coefficient, producing a thin or a thick medium for spectroscopy, in a situation similar to that of a change in length for a vapor cell. This property is due only to the change in the atomic density of the sputtered atoms and can be useful for high-sensitivity spectroscopy of weak transitions. Considering the 75% absorption in Fig. 4, the transition rate for the resonant line ($A_{21} = 2.13 \times 10^8\ s^{-1}$), and a Doppler width (FWHM) of 2.2 GHz, we estimate a density of $10^{10}\ cm^{-3}$ for calcium atoms at a current of 30 mA. Table 1 indicates that this atomic density has an approximate quadratic dependence on current, because the intensities observed in the emission spectra are proportional to it.

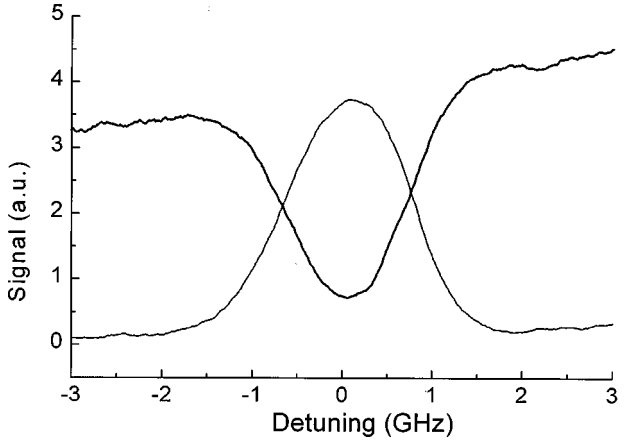

Fig. 4. Doppler-limited optogalvanic (lower curve) and optical absorption (upper curve) spectra of the calcium resonant transition at 423 nm, obtained at $I = 30$ mA.

Considering now the sub-Doppler spectra (Lamb dip and intermodulated), we observed that they are strongly affected by VCCs,[24] which can broaden the lines and severely limit the resolution. These collisions can be classified as weak or strong, depending on whether the corresponding frequency shifts are smaller or bigger than the homogeneous linewidth.[25] Because of their much lower concentration, the sputtered ^{40}Ca atoms collide chiefly with atoms from the buffer gas (^{40}Ar or ^{84}Kr in our lamps). The weak collision regime occurs only in extreme cases, in which there is a large mass ratio between the collision partners.[16] Thus the spectra from our lamps can be considered to be affected by strong collisions, which redistribute atoms within the Doppler profile. In particular, atoms with velocity $\mathbf{v}$, initially interacting with one of the laser beams, can be transformed into $-\mathbf{v}$ and then interact with the other counterpropagating beam. The effect of strong collisions in sub-Doppler spectra is therefore to give rise to a Doppler-broadened pedestal superimposed upon the homogeneous profile. The resultant line shapes consist of a Gaussian pedestal superimposed upon a sub-Doppler Lorentzian profile. The relative size of the Gaussian pedestal with respect to the homogeneous profile can be particularly high if the lower level of the transition is metastable, as it is in fact a function of radiative and collisional cross sections.[16,22] It is also a function of the mass ratio between the collision partners and can sometimes be reduced by the appropriate choice of buffer gas.[26] A quantitative analysis of the effect of VCCs on saturation spectroscopy was considered in the research reported in Refs. 24 and 25.

Figure 5 shows examples of Doppler and sub-Doppler spectra for the intercombination $^1S_0$–$^3P_1$ transition at 657 nm. The spectra in Fig. 5(a) were obtained with the lamp filled with argon at a pressure of 2.5 Torr, and Fig. 5(b) shows spectra obtained with the lamp filled with krypton at a pressure of 0.6 Torr. The influence of VCCs is evident. In Fig. 5(a) the sub-Doppler spectra (both Lamb dip and IMOGS) essentially show the broad Doppler pedestal, a few hundred megahertz wide, with the homogeneous signal hidden on it. Similar spectra were obtained for the other transitions. The intermodulated spectra, like those in Fig. 5(a), were fitted to an expression derived in Ref. 25 for the strong collision case, which describes the line shapes as a sum of a Gaussian (Doppler pedestal) and a Lorentzian (homogeneous signal). In these fittings, the Doppler widths obtained from the Doppler-limited spectra were fixed, and the adjusted parameters were the homogeneous linewidth and a parameter (C in Ref. 25) that describes the magnitude of the Doppler pedestal relative to the homogeneous profile. For the 2.5-Torr Ca–Ar lamp the linewidths (FWHM) vary from 400 MHz to 1 GHz. For instance, the smaller width was observed for the 612-nm transition, whose lower level ($^3P_1$) is relatively long lived (0.38 ms) but not as much as the metastable $^3P_0$ or $^3P_2$. These observed widths are much greater than radiative or typical collisional widths in HCLs at similar pressures.[27] In the spectra of Fig. 5(b), obtained with the krypton lamp, the Doppler pedestal is greatly reduced, and a narrower homogeneous peak, a few tens of megahertz wide, is observ-

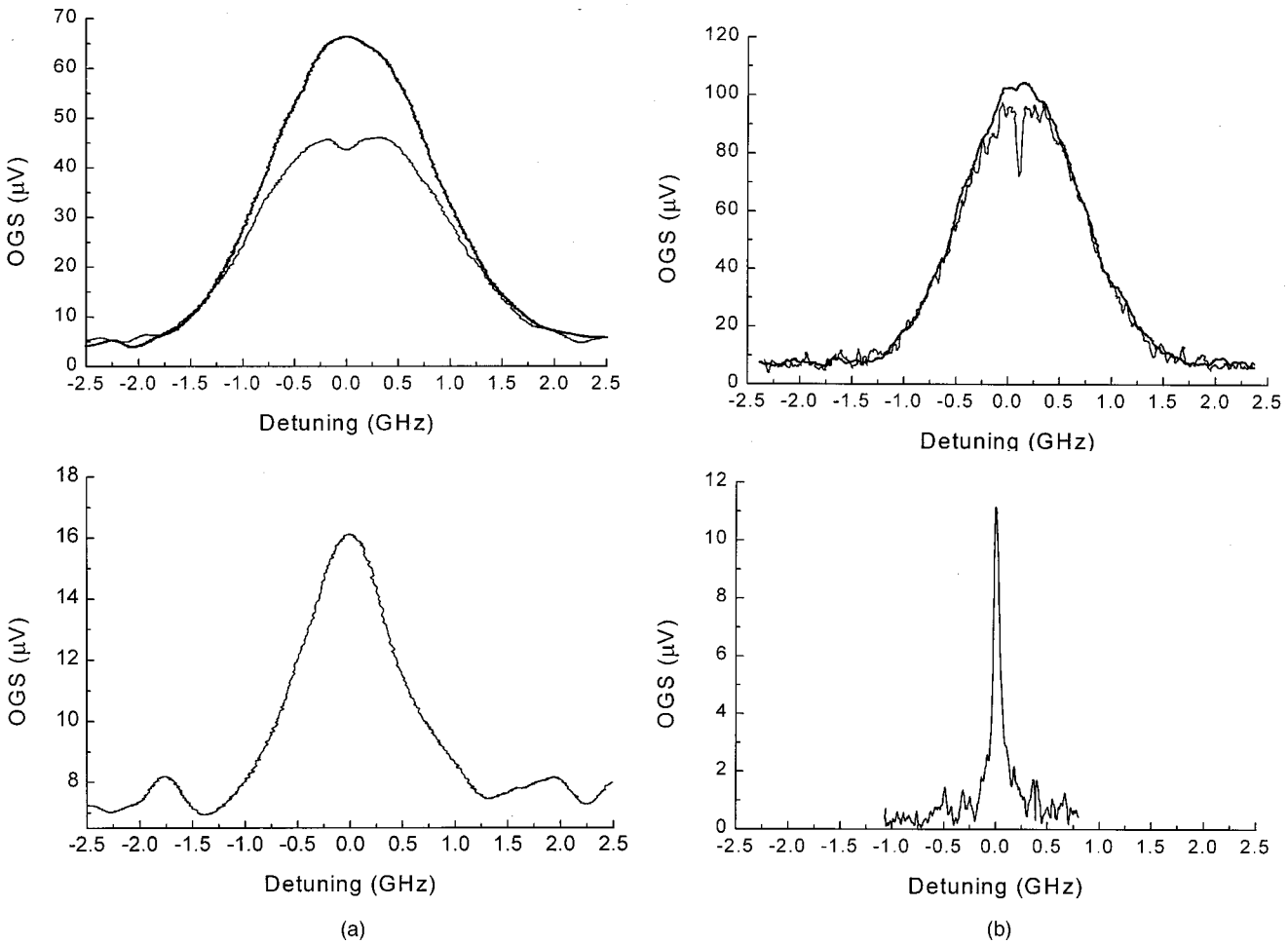

Fig. 5. Doppler and sub-Doppler optogalvanic spectra of the CaI intercombination transition at 657 nm. (a) Spectra obtained with a 2.5-Torr Ca–Ar lamp. A strong influence of VCCs is evident, giving rise to a broad Doppler pedestal. (b) Spectra obtained with a 0.6-Torr Ca–Kr lamp. The Doppler pedestal is greatly reduced. The lower curves are intermodulated optogalvanic spectra.[16] Lamb dips,[20] which can be used for laser frequency stabilization,[27] are also shown for both lamps.

able. A reduction of the Doppler pedestal was reported and analyzed in Ref. 26, and different line shapes, obtained for several buffer gases, were reported in Ref. 25. In all spectra we confirmed the dominant role of VCCs in broadening the sub-Doppler spectra and also the much less significant influence of power broadening and collisional broadening, because in this case similar results were obtained for the two lamps filled with argon at different pressure. A complete characterization of pressure broadening for high-resolution spectroscopy is usually difficult in HCLs because the discharge starts to be unstable at lower pressures. Such was the case for our lamp filled with argon at 0.6 Torr.

Homogeneous linewidths as low as 25 MHz were obtained, for example, for the 657-nm transition in the 0.6-Torr Ca–Kr lamp. This resolution is comparable with what is obtained in a slightly diverging atomic beam, owing to residual Doppler broadening. Considering the integration time constants that we used to achieve the signal-to-noise ratio in Fig. 5(b), we believe that a properly chosen HCL can be useful for tuning and locking lasers in high-resolution spectroscopic experiments involving alkali earth atoms in beams or magneto-optical traps. A HCL could possibly be used to give long-term stability, preventing drifts in a laser that has already been stabilized to a reference cavity. In the case of narrow and weak transitions between excited states, a HCL can be especially useful, working as a vapor cell whose use is not restricted to transitions departing from the ground state.

In conclusion, we have obtained spectra of CaI transitions (including the strong resonant and the weak intercombination transitions, at 423 and 657 nm) by using hollow-cathode discharges and optogalvanic detection. Two homemade Ca–Kr lamps, with pressures of 0.6 and 2.5 Torr, and one Ca–Kr lamp, with a pressure of 0.6 Torr, were used. Sub-Doppler resolution was obtained by optogalvanic saturation spectroscopy that employed the experimental configuration in which a Lamb dip is created or the configuration of intermodulated spectroscopy. The sub-Doppler spectra in the Ca–Ar lamps are strongly affected by VCCs, which give rise to a broad Doppler pedestal. It could be possible that such a strong effect of a VCC, observed for partners that have basically the same atomic mass (calcium and argon), could be similar in other lamps made with alkali earth atoms, such as Sr–Kr, Mg–Ne, or even Ba–Xe. The pedestal that was due to VCC was greatly reduced in the spectra taken with the Ca–Kr lamp, which proved to be better suited for high-

resolution spectroscopy. Homogeneous linewidths as small as 25 MHz (limited by pressure and power broadening) were obtained with this lamp for the 657-nm transition. Such resolution is comparable to what is obtained in a sligtly diverging atomic beam, owing to residual Doppler broadening. We believe that a properly chosen HCL can be particularly useful as an auxiliary tool for tuning or locking lasers in high-resolution experiments in atomic beams or MOTs. Their main advantages come from the simplicity of the experimental setup combined with the good S/N that can be achieved with the sensitive optogalvanic detection. This technique can be applied equally to transitions in different spectral regions, from the UV to the IR, dispensing, for example, different optical detectors. In addition, HCLs can be suitable for tuning to transitions between excited states, which are naturally populated by the electrical discharge. Some prospects for future research include the study of VCCs with other buffer gases (for example, Xe and Ne), the realization of precise measurements of collisional shifts, and tests of HCL-stabilized lasers in experiments with atomic beams and cold atoms.

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F. C. Cruz's e-mail address is flavio@ifi.unicamp.br.

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