

INTRODUCTION

We report the first observation of an optovoltaic effect generated in saturated neutral optically thick cesium vapor irradiated with a monochromatic diode laser. The design of the Cs vapor cell device and the experimental setup are presented, and the preliminary spectral optovoltaic signal features are qualitatively attempted.

The spectral and photonic properties of optically thick alkali metal vapors are much less known, more problematic, and raise new spectroscopic challenges compared to the case of optically thin vapors. There are several reasons for this lack of experimental spectral data; one of them is the fact that conventional absorption, emission, and fluorescence spectroscopic methods are not appropriate for the analysis of an optically thick vapor. In order to obtain spectral information essential for the in-depth understanding of optically thick metallic vapors in the vicinity of a surface, systematic studies of the spectral characterization of saturated Cs vapors in closed electrodeless cells, in both the linear^{1,2} and nonlinear regimes, have been conducted in our laboratory. In order to complement the spectroscopic investigation of optically thick Cs vapor, we are now exploring a spectroscopic method based on the detection and measurements of voltage signals associated with an irradiated volume of the vapor. To this end, we used a new design of the Cs cell (produced in our laboratory), with two internal probe electrodes. It appeared that the resonantly irradiated Cs vapor cell exhibited spectral electric activity without the assistance of an external electric voltage, which we call the *optovoltaic effect*. This effect is different from the conventional optogalvanic effect in which glow discharge in hollow cathode cells³ are used. This new effect is also different from the one used for the detection and investigation of laser-desorbed silver in a low-pressure argon atmosphere⁴ and the one studied in optogalvanic signals due to laser-heated electrodes in hollow cathode plasmas.⁵ The preliminary results are promising and stimulate further research in this original axis of experimental atomic spectroscopy, based on the observation of electric effects induced on probe electrodes without an external bias voltage in a volume of a metallic vapor.

The aim of this note is to present the preliminary observations of this resonant spectral optovoltaic phenomenon and propose a simple qualitative explanation of the features of the effect.

DESCRIPTION OF THE CESIUM VAPOR CELL DEVICE AND THE EXPERIMENTAL SETUP

The design of the Cs vapor cell (with no buffer gas) used in our experiment is shown in Fig. 1. The glass cell, produced in our laboratory, consists of a cylindrical body, a reservoir of Cs connected at right angles to the body, and two parallel-plate internal electrodes of different sizes. The glass cell is first heated in a 10^{-6} torr vacuum environment in order to eliminate contaminants and impurities and then filled with a pure Cs under a controlled environment and by the usual procedures. One electrode is ~ 4.8 cm long, ~ 0.9 cm in width, and ~ 1 mm thick,

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New Spectral Optovoltaic Effect in Optically Thick Neutral Cesium Vapor

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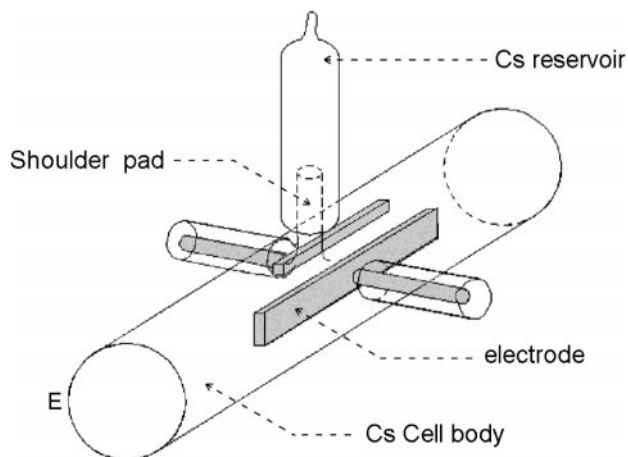


FIG. 1. Design of the cesium cell. E designates the cell entrance window. (For detailed description, see text.)

and the other (starting at half the length of the cylindrical body of the cell) is ~ 2.4 cm long, 1 mm in width, and 1 mm thick. The electrodes are separated by a gap of about 4 mm. The front end of the largest electrode is situated at 0.6 cm from the internal surface of the cell entrance window. Electrodes with different sizes have been chosen in order to take advantage of the spatial asymmetry regarding the electrical effects in the cell, particularly increasing the electric detection efficiency due to the electrode edge effects. The inner diameter and inner length of the cell body are ~ 2.4 cm and ~ 5.8 cm, respectively. The internal structure of the reservoir prevents the Cs liquid from dropping into the cylindrical body. The cell body was enclosed in an aluminum block, leaving only the windows uncovered. A stack of aluminum rings was placed around the Cs reservoir, vertically oriented. The device was placed on a horizontal hotplate (MIRAK model HP 72935), under a Pyrex glass cover, and heated to a constant programmed temperature between 100 and 150 °C. A saturated Cs neutral vapor was thus obtained. The temperature of the cell and the vapor were measured using two thermocouples, one in contact with the cylindrical body of the cell and the other in contact with the top of the Cs reservoir (which is the coolest point of the cell). In this configuration, the body of the cell is kept about 10 °C hotter than the coolest point of the cell in order to avoid the condensation of the Cs on the windows, the inner walls of the body, and the electrodes.

The block-diagram of our experimental setup is schematically illustrated in Fig. 2. The Cs vapor is excited by a monochromatic diode laser (Environmental Optical Sensors, Inc., Model LCU 2001 M), with a bandwidth lower than 10 MHz. We chose the atomic hyperfine structure transition lines [$6^2S_{1/2}$ ($F_g = 4$) \rightarrow $6^2P_{3/2}$ ($F_e = 3,4,5$)] and [$6^2S_{1/2}$ ($F_g = 3$) \rightarrow $6^2P_{3/2}$ ($F_e = 2,3,4$)] at wavelength 852.2 nm, where F_g and F_e are the quantum numbers of the hyperfine levels of the ground and excited states, respectively. The spectral stepping of the diode laser sweep around the 852.2 nm resonance line was carried out by using a piezoelectric device having a resolution of a few MHz. The diode laser beam was split in two parts. The weaker part of the split beam was directed into a Fabry–Pérot interferometer, which permitted the calibration of

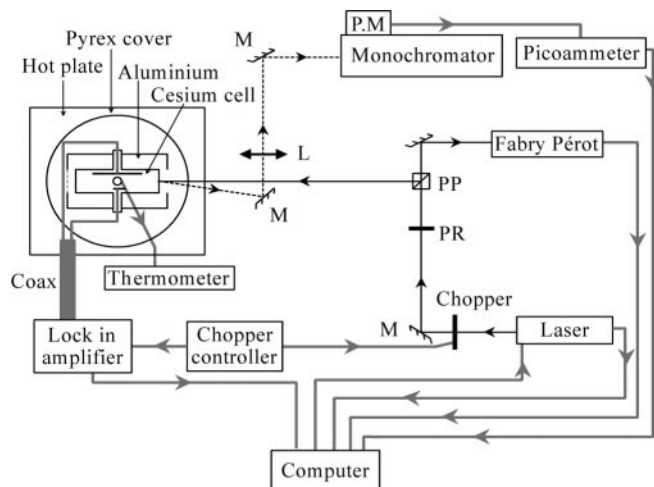


FIG. 2. Diagram of the experimental setup (L: Lens, M: Mirror, PM: Photomultiplier Tube, PP: Polarizing Prism (Beam Splitter), PR: Polarization Rotator, and Coax: coaxial cable).

the explored spectral band. The stronger beam was directed at an angle of incidence of $\sim 2^\circ$ with respect to the normal of the cell entrance window surface. The arrangement of the experimental setup permitted us to perform, simultaneously, measurements of laser-induced retrofluorescence spectra and electrical signals at the electrodes of the cell. For comparison, the retrofluorescent signal, observed at an angle of $\sim 16^\circ$ with respect to the normal of the entrance window surface, was captured by a 0.5 m Jarrell-Ash Czerny–Turner spectrometer. The retrofluorescence signals are used to scale the frequency axis of the optovoltic spectra. The electrodes of the Cs cell were directly connected, through a shielded cable (with a BNC connector at one end), to a lock-in amplifier (Stanford Research Systems, Model SRS 530), used in a voltmeter mode having a 100 M Ω voltage input impedance and 25 pF capacitance. A shielding of the recording cables was necessary as the signal was very sensitive to the ambient magnetic fields. The use of a lock-in amplifier has the benefit of permitting the extraction of the signal associated with the modulated laser beam from the total noisy signal. No external bias voltage was applied to the electrodes of the Cs cell. The reference signal for the synchronous detection was generated by the controller of a chopper, which mechanically modulated the amplitude of the laser beam at an appropriate frequency of about 644 Hz.

RESULTS

When the diode laser is tuned far away from the 852.2 nm ($6^2S_{1/2} \rightarrow 6^2P_{3/2}$) resonance line, the voltage measured at the electrodes of the Cs cell is negligible compared to the maximum optovoltic signal in the resonance spectral band. When the laser is off, the signal is zero. However, when the laser is scanned through the atomic resonant 852.2 nm ($6^2S_{1/2} \rightarrow 6^2P_{3/2}$) transition line, a voltage (optovoltic signal) was detected whose magnitude depended on the intensity, the frequency of the laser beam, and the temperature of the vapor. We observed that by approaching the laser beam close to an electrode or by shifting it from the cell axis the optovoltic signal was affected. For

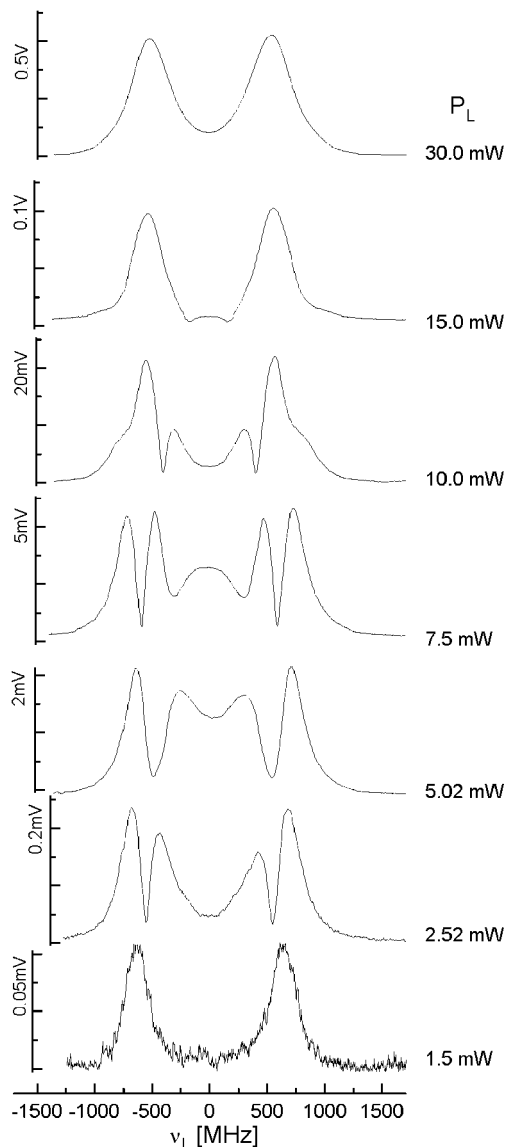


FIG. 3. Experimental optovoltic spectra: Dependence on laser power at fixed temperature $T_v = 136\text{ }^\circ\text{C}$ for the transition $6^2S_{1/2} (F_g = 3) \rightarrow 6^2P_{3/2} (F_e = 4,3,2)$.

recording the spectral signals given in the following, the laser beam was in coincidence with the cell axis. The amplitudes of the optovoltic signals were in the range between $50\text{ }\mu\text{V}$ and 500 mV , as indicated by the full-range sensitivity of the lock-in amplifier, corresponding to laser powers in the interval from 1 mW to 30 mW (laser beam diameter of $\sim 0.014\text{ cm}^2$) and atomic number densities in the range of 10^{13} – $10^{14}\text{ atoms cm}^{-3}$. At these atomic densities, the Cs vapor was optically thick at 852.2 nm resonance in the linear regime ($k_v l \gg 1$, where k_v is the spectral absorption coefficient of the medium), over a vapor thickness of the order of a few wavelengths ($l \sim 10\lambda_L$). The laser powers used correspond to an optical saturation regime (nonlinear). The signal strength evolves by four orders of magnitude while the laser power evolves by a factor of 30. Below 1 mW , we did not observe optovoltic signal.

Figure 3 shows the spectral optovoltic signals corresponding to the $[6^2S_{1/2} (F_g = 3) \rightarrow 6^2P_{3/2} (F_e = 2,3,4)]$

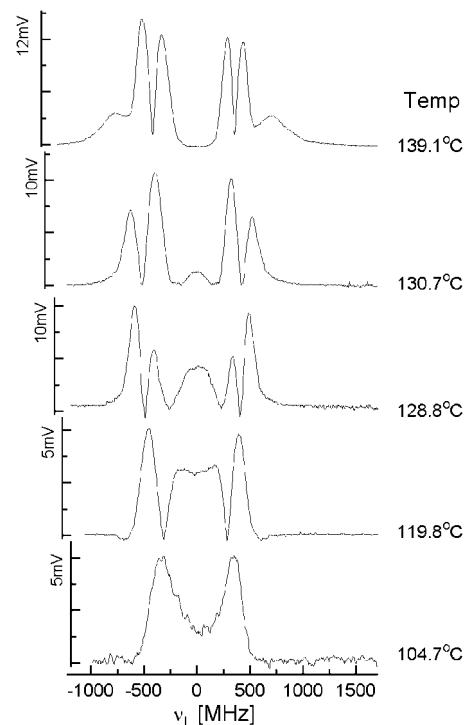


FIG. 4. Experimental optovoltic spectra: Dependence on the vapor temperature at fixed laser power $P_L = 10\text{ mW}$ for the transition $6^2S_{1/2} (F_g = 4) \rightarrow 6^2P_{3/2} (F_e = 5,4,3)$.

atomic excitation channel for different laser powers (1.5 , 2.52 , 5.02 , 7.5 , 10 , 15 , and 30.0 mW) and recorded at a constant vapor temperature, $T_v = 136\text{ }^\circ\text{C}$. In order to display signals of different amplitudes simultaneously on the same graph, the amplitudes of the experimental results are presented in arbitrary units for different scale ranges of the lock-in amplifier used. One may note the particular evolution of the structures of the spectra. At a laser power of 1.5 mW , the spectral profile has two peaks, symmetrically located at $\Delta\nu_L \sim 658\text{ MHz}$ on both sides of the zero detuning of the $[6^2S_{1/2} (F_g = 3) - 6^2P_{3/2} (F_e = 2,3,4)]$ line center. The spectral half width at half-maximum of the individual peaks is about 130 MHz . By gradually increasing the laser power, a new symmetrical structure (made up of satellite peaks) appears at 2.5 mW on both sides of the zero detuning, and then at 7.5 mW a peak centered at zero detuning emerges. At 10 mW , the central peak disappears. Moreover, one may notice a relative decrease in the intensities of the satellite peaks. At 30 mW , a two-peak profile, similar to Fig. 3 (1.5 mW), is observed. However, the individual peaks are broader (half width at half-maximum $\sim 210\text{ MHz}$) and are located symmetrically at approximately $\pm 500\text{ MHz}$, on both sides of the zero detuning.

Figure 4 represents the spectral optovoltic signals corresponding to the $[6^2S_{1/2} (F_g = 4) \rightarrow 6^2P_{3/2} (F_e = 3,4,5)]$ atomic excitation transition channel at various vapor temperatures (104.7 , 119.8 , 128 , 130.7 , and $139.1\text{ }^\circ\text{C}$) for a fixed laser power of 10 mW . The number density of the Cs vapor is $1.9 \times 10^{13}\text{ atoms cm}^{-3}$ at $104.7\text{ }^\circ\text{C}$ and $1.2 \times 10^{14}\text{ atoms cm}^{-3}$ at $139.1\text{ }^\circ\text{C}$, as determined by the Langmuir–Taylor vapor pressure equation.⁶ As in the preceding figure, the different scale ranges of the lock-in amplifier used are indicated on the y-axis of the graphs.

At 104.7 °C the spectral signal exhibits two peaks at about ± 350 MHz, on both sides of the zero detuning, whose half width at half-maximum (of each peak) is ~ 125 MHz. As the temperature increases from 104.7 to 139 °C, the spectral signal exhibits satellite peaks. For example, at 119.8 °C symmetric satellite peaks appear on both sides of the zero detuning from the $[6^2S_{1/2} (F_g = 4) - 6^2P_{3/2} (F_e = 3,4,5)]$ line center and, at 130.7 °C, a peak centered at zero detuning is observed. We note that the relative intensities of the satellite peaks undergo significant changes for weak variations of the vapor temperature between 128 °C and 130 °C. At 139.1 °C, the peak centered at zero detuning disappears and distortions on the wings of the spectrum are observed.

DISCUSSION

The configuration of our experimental setup and the atomic spectral properties of the observed signals are distinct from the setups used and results obtained by the conventional optogalvanic spectroscopy in glow discharges.^{7,8} Our experimental device behaves like a current (or voltage) generator, featuring original spectral properties. The spectral optovoltic signal is sensitive to the temperature of the vapor and the frequency and intensity of the exciting monochromatic diode laser. Our experiment is different from that of Brandenberger⁹ in that (1) it was implemented in a pure alkaline vapor (Cs) while that of Brandenberger was performed in inert gas (Kr), (2) there was no plasma discharge in our cell while in that of Brandenberger, a pair of external ring-shaped electrodes was used to produce a radio frequency (RF) discharge plasma in the cell. However, in both cases, no external bias voltage was applied (or needed) to collect the optovoltic signal from the cell. By using a laser beam at the wavelength 852.2 nm, we exclude the non-resonant process of direct one-photon photoionization of the Cs atom from ground level. The energy of a laser photon ($h\nu_L = 1.45$ eV) is not high enough to produce the direct ionization of the atoms of the vapor in ground state ($E_i = 3.89$ eV is the ionization potential of the Cs atom), nor to produce a photoelectric effect on the glass and electrode surfaces ($\phi_{Cs} = 2.14$ eV is the workfunction of a Cs thin solid film). Thermoelectronic emission at the temperatures used and electron multiplication due to electron-atom collisions are also excluded. The relaxation processes due to elastic collisions in an optically thick vapor (10^{14} atoms/cm³) are important and the evolution of the atomic optical coherencies are strongly perturbed. In our experimental conditions, the extra phase information contained in the Bloch equation is lost or irrelevant. In this case, the ac Stark effect (or Rabi splitting effect) is negligible. Note, also, that the spectral Doppler width (~ 450 MHz) is very important compared to the laser width (10 MHz).

We attribute the origin of the optovoltic signal to the existence of four concurrent and necessary phenomena in the volume of the cell irradiated by the laser: the presence of an internal long-range electrical field without an external bias voltage, the polarization of the Cs vapor, the generation of electron-ion pairs by the effect of the laser radiation, and the surface ionization of the excited Cs

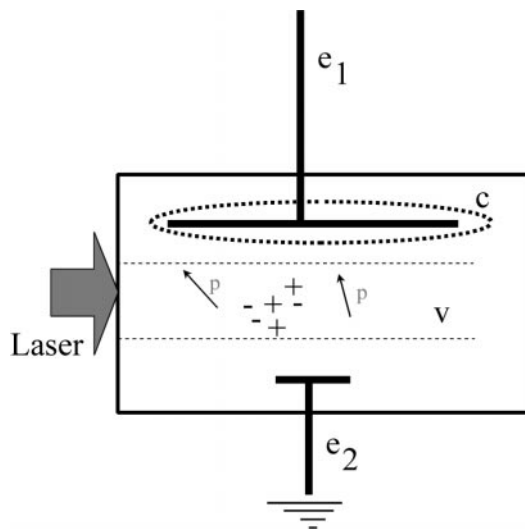


FIG. 5. Schematic view of electrical effects in a Cs vapor cell irradiated by a diode laser. e_1 and e_2 are electrodes (e_2 is ground-connected), C indicates space charge surrounding the surface of electrode e_1 in contact with the vapor, V is the illuminated volume of vapor, and p designates polarized atom.

producing space charges surrounding the surface of the electrodes.

In order to make our preliminary qualitative assumption clear, let us consider Fig. 5, which shows a schematic representation of a cylindrical dielectric cell containing a Cs vapor optically thick at resonance. Two metallic plane probe electrodes are introduced in the vapor; the ground-connected one is used as the reference. The cell is irradiated by a laser beam along the axis of the cell. The diameter of the laser beam is smaller than the gap between the two electrodes. This schematization of the cell is a simple model reasonably close to our experimental conditions. The floating electrode can be an important source of electric charges by surface ionization processes forming a cloud of charges surrounding the electrode surface due to its contact with the Cs vapor.^{10,11} This sheath of electrical charges, therefore, constitute a space charge generating an internal long-range electrical field inside the cell, without the assistance of an external voltage. An important optical pumping of the excited $6^2P_{3/2}$ level results from the laser-atom interaction in the Cs vapor irradiated by the diode laser tuned at the resonant 852.2 nm ($6^2S_{1/2} - 6^2P_{3/2}$) atomic transition, thus generating a macroscopic polarization of the illuminated volume of the Cs vapor. Generation of electron-ion Cs_2^+ pairs also occurs in the vapor due to the laser-photosensitized ionization by binary collisions of 6^2P cesium atoms ($2Cs(6^2P_{3/2}) \rightarrow Cs_2^+ + e^-$).¹² The polarization of the vapor can disturb the space charge surrounding the probe electrode. The electrical charges, particularly the electrons, produced by the photo-sensitized ionization phenomenon are driven by the action of the long-range internal electrical field. Each heavy positive ion in the Cs vapor simultaneously attracts several electrons in its closeness^{13,14} before neutralization by the recombination process, thus inducing variations in the space charge distribution. The whole movement of the free charges (electrons and positive ions) produces an electrical current, which can supply an electrical signal to an external cir-

cuit. Two running modes of the external circuit are possible: the photo-current and the photo-voltage modes. The origin of the optovoltaic effect is associated with this electrical current and is observed, in our case, in the photo-voltage mode.

According to the preceding qualitative viewpoint, we can write the optovoltaic signal S_{OV} as a complicated non-trivial function F :

$$S_{OV} = F[k(I_L, \nu_L, T_V), i_c, i_p] \quad (1)$$

where $k(I_L, \nu_L, T_V)$ is the nonlinear absorption coefficient (corresponding to the resonant $[6^2S_{1/2}(F_g) \rightarrow 6^2P_{3/2}(F_e)]$ atomic transition), which depends on the laser intensity I_L , the laser frequency ν_L , and the Cs vapor temperature T_V ; i_c is the electrical current associated with the generation of electron-ion Cs_2^+ pairs; and i_p is the current due to the laser-induced polarization of the Cs vapor excited at the $6^2P_{3/2}$ level. Since there are two types of charge carriers (electrons and positive ions), we can write

$$i_c = i_+ + i_- \quad (2)$$

where i_+ is associated with the positive ions and i_- with the electrons.

The value of S_{OV} measured without an external voltage supply cannot be directly used to determine the atomic spectral properties of the optically thick vapor. The magnitude of the current intensities i_c and i_p are governed, in a complicated way, by the properties of the atomic absorption coefficient, the nature and the geometric shape of the probe electrodes in contact with the vapor, and the spatial positioning of the laser beam relative to the electrodes. The nonlinear optovoltaic phenomenon observed in an optically thick vapor is non-trivial to interpret. The interpretation of the spectral optovoltaic signal brings about several problems that stimulate further development of the potential application of this effect as a diagnostic tool for characterizing an optically thick metallic vapor. Before attempting any quantitative theoretical investigation, additional experiments are necessary to confirm our assumptions regarding the origin of the spectral optovoltaic signal in a neutral Cs vapor irradiated by a monochromatic diode laser tuned over the resonance line. No experimental work has yet been conducted with an applied external low voltage (~ 2 V) on the electrodes. This is an interesting question; works have been initiated but at this stage no appropriate answer can be proposed.

These optovoltaic signals, obtained as a function of the laser frequency, the power of the exciting laser beam, and the temperature of the vapor, exhibit unusual spectral characteristics different from those obtained by using conventional optogalvanic, retrofluorescence, and selective reflection spectroscopic methods. This new optovoltaic phenomenon that we observed appears as a basic effect for a potential new laser spectroscopic method for the analysis of optically thick alkali metal vapors.

We believe that this note on the observation of a new optovoltaic effect in neutral metallic vapor will arouse an interest among the spectroscopists' community and stimulate work to push ahead this new area of laser spectroscopy.

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