SPATIAL AND TEMPORAL OPTO-GALVANIC EFFECTS IN RARE GASES
S. Brunker, S. Haydon

To cite this version:
S. Brunker, S. Haydon. SPATIAL AND TEMPORAL OPTO-GALVANIC EFFECTS IN RARE GASES. Journal de Physique Colloques, 1983, 44 (C7), pp.C7-55-C7-63. <10.1051/jphyscol:1983705>. <jpa-00223262>

HAL Id: jpa-00223262
https://hal.archives-ouvertes.fr/jpa-00223262
Submitted on 1 Jan 1983

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
SPATIAL AND TEMPORAL OPTO-GALVANIC EFFECTS IN RARE GASES

S.A. Brunker and S.C. Haydon

Physics Department, University of New England, Armidale, N.S.W. 2351, Australia

Abstract - A pulsed tunable dye laser is used, in conjunction with spatial and temporal techniques developed for the study of pre-breakdown ionization phenomena, to provide information about opto-galvanic phenomena in neon. A steady-state weakly ionized gas is examined thus ensuring that localised space charge effects have negligible influence. Signal averaging facilities are used to reveal fine structure in the opto-galvanic signals. This structure is identified with resonance photon, \( \text{Ne}^+ \) and \( \text{Ne}_2^+ \) influences. Application of the techniques to \( \text{N}_2 \) is considered briefly.

I - INTRODUCTION

A large variety of electrical switching and control devices rely for their successful operation on the complex behaviour of an ionized gas. Unfortunately the consequences of collision interactions between the neutral and charged particles themselves and of each with the boundary surfaces of the device are often extremely sensitive to gas purity and surface conditions. It then becomes extremely difficult to maintain sufficient control of the gas behaviour to ensure a reproducible response when precise threshold conditions are prescribed. In these laboratories, for example, observations of the critical breakdown voltage of low pressure nitrogen gas subjected to uniform electric fields have shown that this critical voltage can vary from \( \sim 1200 \) to \( \sim 450 \) volts as the quality of the gas sample is progressively improved. These dramatic effects have appeared during extensive investigations of both the spatial and temporal growth of the ionization in this \( /1-4/ \) and other gases \( /5/ \). Sufficient control of the breakdown mechanism is now available to enable us to investigate the possibility of achieving very sensitive perturbations of the threshold breakdown conditions by optical means using external light sources. This opto-galvanic effect was first demonstrated by Penning \( /6/ \) who was able to change the voltage breakdown threshold in a neon cell containing a small quantity of argon by allowing light from another neon cell to fall on it. The change in the gas behaviour was a consequence of the optical quenching of metastable atoms of neon. Similarly in nitrogen it is the contribution of the metastable particles to ionization processes that is responsible for the remarkable behaviour of the ionized gas.

In considering whether such opto-galvanic effects can be exploited for practical
purposes two major problems of a fundamental nature arise. Firstly the basic gas discharge phenomena should be well understood, reproducible and amenable to quantitative analysis. Secondly the opto-galvanic effect induced should be the consequence of a perturbation of a single level of a selected particle. In this context it was attractive in the first instance to carry out extensive investigations in $N_2$ gas because the dominant $N_2(A^3Σ^+)$ metastable particle has only sufficient energy to contribute to the ionization growth through surface interactions with the boundary electrodes. By contrast the metastable particles in neon gas are energetically capable of both surface and volume ionization processes and also lead, when optically perturbed, to further complexity because of the subsequent generation of high energy resonance radiation. However, appropriate tunable dye lasers are readily available with which to selectively perturb the metastable populations in neon gas. The situation is not so favourable for the $N_2$ system.

In these circumstances we have applied the opto-galvanic effect itself to unravel the complexities of the laser-induced perturbation spectroscopy of rare gas systems such as neon and continued at the same time to examine the basic processes controlling the reproducibility of both spatial and temporal ionization growth in the pre-breakdown regime in $N_2$. For the opto-galvanic studies in neon we have concentrated exclusively on the pre-breakdown phenomena where the added complications of space charge distortions and related effects are not present. This has required more demanding experimentation in order to observe and record the nanoampere opto-galvanic signals obtained under these conditions. This paper describes the opto-galvanic techniques used for investigating weakly ionized gases and discusses briefly the potential applications of the procedures to molecular gases such as nitrogen.

II - EXPERIMENTAL PROCEDURES FOR PRE-BREAKDOWN INVESTIGATIONS

These have been described in detail elsewhere [2]. The following brief outline provides the background necessary to appreciate the opto-galvanic phenomena reported in the present paper. Observations of the spatial growth of ionization are made using plane-parallel electrode geometry at constant $E/N$ (electric field/gas density). Analysis of the variation of ionization current with electrode separation, $d$, then yields the appropriate primary and secondary ionization coefficients. The first coefficient ($\alpha/N$) is substantially independent of the spatial coordinate and is constant for a fixed value of $E/N$. The secondary coefficient $\omega(d)$ is, on the other hand, strongly spatially dependent when significant contributions to the ionization growth are made by metastable particles. Both observation and prediction are

![Figure 1. Apparatus for spatial and temporal ionization growth studies.](image-url)
consistent provided that a quenching parameter, $\mu/\alpha$, is appropriately adjusted. The quantity, $\mu/\alpha$, is given by $\alpha^{-1} \sqrt{G/D}$, where $G$ is the volume destruction and $D_m$ is the diffusion coefficient for the metastable particles. Both $G$ and $D_m$ can be evaluated from the corresponding temporal investigations of the ionization growth. Here the external light source (Fig. 1) producing the initial electrons is chopped periodically giving access to the time constant for the growth or decay of ionization due to the metastable particles. Typical temporal signals for neon and nitrogen are shown in (Fig. 2) and from the shape of the in $I$ vs $t$ curve derived from this signal, it has been possible, for some special conditions in the case of $N_2$, to evaluate both $D_m$ and $G$ using an analysis first proposed by Molnar. The special conditions are determined by the need to control the level of impurity in the gas sample so that only one dominant metastable particle exists. This is achieved by appropriate ultra-high vacuum procedures (Fig. 1), the demands in this respect having been generally more severe for our investigations in neon than in nitrogen.

The strong dependence of the observations on the quality of the gas purity reflects the sensitivity of the metastable particle population to the quenching action of the common trace impurities. From the fundamental point of view, therefore, the possibility of replacing the influence on the metastable particle population of complex particle-collision quenching with a controlled optical quenching, using a tunable dye laser, is very attractive. The opto-galvanic signal that results from such a laser-induced perturbation then provides direct access to the precise role of the metastable states in the ionization growth process. The experimental procedure now used to observe and record such opto-galvanic signals is shown schematically in Figure 3.

The experiment first establishes a steady state ionization current between two plane-parallel electrodes immersed in a suitable gas at a pressure $p$ (gas number density $N$) in a uniform electric field. This steady-state pre-breakdown condition requires a constant electron current $I_0$ generated at the negative electrode by external U.V. irradiation. The magnitude of this steady-state ionization current can, in principle, be calculated, given the form of the electron energy distribution function, information about the collision cross-sections of the various excitation and ionization processes, the corresponding drift velocities of the charged particles, diffusion coefficients of the neutral metastable particles and efficiencies of the neutral metastable particle and efficiencies of surface ionization processes taking place at the boundary walls. Similar procedures will also identify the consequences of selectively perturbing the populations of any single metastable particle energy level and should predict the fine structure of the anticipated opto-galvanic signal.

A pulsed dye-laser has been used in these first pre-breakdown experiments to create the population changes. The 337 nm output from the nitrogen laser, Figure 3,
operated in conventional transverse electric field configuration was line-focused by means of a cylindrical lens on to the rhodamine B dye cell incorporated into the cavity of a Hansch-type system. The tuned radiation (linewidth 0.02 nm) could be arranged either to illuminate the whole volume or, by appropriate aperturing, to interact with a selected small volume of gas at any position between the electrodes. The change in ionization current brought about by the laser-induced perturbation produces a corresponding voltage drop across $R$ which can in turn be suitably amplified by a Tetronix AM 502 differential amplifier and recorded. Signal averaging is essential and the experiment is initiated by a trigger signal from the LSI 11. This activates the controller, the analogue-to-digital converter and its associated memory. It is also used to trigger either the high tension supply to the chopped ultra-violet light source which generates the initial electrons or the nitrogen laser which emits a 10 ns (FWHM) pulse of 337 nm radiation to initiate the dye-laser emission. The experiment could be monitored during the data accumulation period by means of a Tetronix Model 7904 CRO and the final opto-galvanic signal could either be displayed on the Hewlett Packard model 7004B X-Y recorder or sent to a PDP11/40 and/or DEC 20 for storage and further analysis.

The system was examined for any signal distortion produced by the pre-amplifier by observing a rectangular test signal having a rise-time $< 100$ ns. No distortion was observed for times greater than 700 ns so that only information in the first 7 channels of signals averaged with 100 ns/channel need be rejected. Tests were also applied to ensure that no distortion of the opto-galvanic signals occurred as a result of the load resistor $R$.

III - EXPERIMENTAL RESULTS

This simplified energy level diagram relevant to the laser-induced perturbation is shown in both Paschen and Russel-Saunders notation in Figure 4(a). When tuned to $\lambda = 614.3$ nm the 10 ns dye laser pulse raises $1s_\alpha$ metastable particles to the $2p_\alpha$ state from which they decay to the $1s_\beta$, $1s_\delta$, and $1s_\gamma$ levels. The $1s_\beta$ and $1s_\delta$ states are resonance states and subsequently decay to the $1s_\gamma$ ground state emitting resonance radiation at $\lambda = 73.6$ nm and 74.3 nm respectively. This pulse of radiation provides an internal pulsed source of electrons which is subsequently amplified and gives rise to an opto-galvanic signal. The 10 ns perturbation disturbs the steady-state ionization for some tens of milliseconds and the particular form of the opto-galvanic signal will depend on the value of $E/N$ applied across the ionization region. It consists of a fast and slow component (Fig. 4(b)), the former associated with the relatively fast resonance photon and ionic drift processes, the latter with the slower, diffusion dominated re-population of the depleted metastable levels.
(a) Phenomena associated with the fast component

With improved data acquisition facilities extra information about the fine structure of the fast component of the opto-galvanic signals has appeared and this is summarised in Figure 5 for a variety of E/N values throughout the range from about 80 to 283 Td (E/p ~ 30 to 100 V cm$^{-1}$ torr$^{-1}$). At even lower values of E/N (e.g. E/N = 42.45 Td curve 1, fig. 6) a sharp peak is observed at the earliest times followed by a flat plateau. With increasing E/N subsidiary peaks appear in the plateau region whilst the first sharp peak becomes progressively less pronounced. The origin of these various peaks is associated with the complex processes contributing to secondary ionization in neon. These secondary processes include resonance and non-resonance photons, positive ions and diffusing metastable particles all interacting with the boundary surfaces. The ionization coefficients describing these processes can be functions of both the gas density N, and the value of E/N, and this adds complications to the interpretation. This is demonstrated most clearly in Figure 6 which shows a series of opto-galvanic signals taken at constant E/N = 42.45 Td but at different pressures. The appearance of two peaks in the plateau region as p increases is a consequence of a three-body collision to produce a molecular state from the atomic ion

$$\text{Ne}^+ + 2\text{Ne} \rightarrow \text{Ne}_2^+ + \text{Ne}.$$
Figure 6. Variation of fine structure of fast components with pressure at constant $E/p = 15 \text{ V cm}^{-1} \text{torr}^{-1}$. (Ne$^+$ and Ne$_2^+$ peaks indicated for curve 5)

At the higher values of $E/N$ in Figure 5, where the experiments were undertaken at sufficiently low pressures for the above reaction not to occur, the subsidiary peaks are associated with atomic ions only. Convincing evidence for this interpretation is provided by analysis of the time intervals between the successive peaks in the fine structure. Given the electrode separation, $d$, these can be converted into values of the drift velocity of the charged particles responsible for the peaks. For the larger values of $E/N$ corresponding to low pressure the values obtained are in excellent agreement with more precise, independent determinations of the drift velocity for Ne$^+$ ions /10/. A similar analysis applied to the fine structure of the curves of Figure 6 for low $E/N$ shows that the drift velocities associated with the two subsidiary peaks are in the ratio 1:2. The ratio of the drift velocities of Ne$_2^+$ and Ne$^+$ at this $E/N$ measured by Beatty and Patterson /11/ is 0.53 and supports the contention that the origin of the second of the three peaks is the Ne$_2^+$ ion.

The first peak at the earliest times is, in all cases, associated with resonance photons. This peak becomes less pronounced with increase in $E/N$ and is not discernible at the largest values with the time resolution at present available to us. As a consequence the magnitude of the first ionic Ne$^+$ peak relative to the resonance photon contribution increases with $E/N$ and the time interval to the first ionic peak decreases because of the increase in ionic drift velocity. The changes in relative magnitude of the resonance photon and ion contribution is to be expected on the basis of the results of an earlier investigation of the unperturbed temporal growth of ionization by Willis and Gray Morgan /12/. They recorded the time-dependent growth of ionization as a function of $\%$ overvoltage for $8 < E/p < 90 \text{ V cm}^{-1} \text{torr}^{-1}$ and analysed their data by the procedures given by Thomas /13/ and by Davies and Evans /14/ each of which is based on the earlier work of Phelps /15/ and Davidson /16/. In addition to the resonance photon processes they also considered a source of secondary ionization from non-resonance photons liberated in the dissociation of molecular metastable neon formed in the three-body collision.

$$\text{Ne}^* + 2\text{Ne} \rightarrow \text{Ne}_2^* \rightarrow \text{Ne} + 3\text{Ne} + hv.$$  

Such a process would be more significant at high gas densities and low values of $E/N$ and in the present investigations, which do not extend to gas number densities higher than $\sim 7 \times 10^{17} \text{ cm}^{-3}$ or $E/N$ values $< 40 \text{ Td}$, the contribution from this source is relatively small. Their complex analysis requires knowledge of the magnitudes of the generalised secondary coefficient $\omega/\alpha$ as well as of $\alpha/N$ and the former was obtained by application of the Townsend criterion for the onset of breakdown to their measured ionization currents. Whilst some doubt surrounds the validity of
such a procedure /2/ nevertheless the derived values of the secondary coefficients for the various processes which they subsequently derive are of interest to the present observations. The ratios of the derived coefficients for positive ions ($\gamma$) and resonance photons ($\delta'/\alpha$) which they quote are shown in Figure 7. The corresponding information appropriate to the present experiments is convoluted into the structure of the opto-galvanic signal and appropriate computer-aided procedures are at present being developed to retrieve the values of the coefficients appropriate to conditions where one or more of them may be spatially dependant. For the present purposes we simply recognise that the magnitudes of the individual peaks reflect the size of the contributions from the secondary processes and for comparison we also show the ratios of the actual peaks themselves in Figure 7. The same trend with E/N is observed and indicates that the structure of the observed opto-galvanic signals is consistent with the earlier observations.

It seems clear, therefore, from these observations with the new laser-induced perturbation approach that an opportunity now exists for a more direct experimental attack on the problem of unravelling the relative contributions of a variety of complex processes to the ionization growth. With appropriate improvements in the resolution of the data acquisition facility combined with corresponding changes in the geometry of the ionization chamber itself it should be possible to establish directly the various contributions to the ionization growth.

(b) Spatial variation of the opto-galvanic signals

That part of the complex secondary ionization mechanism associated with the diffusion of metastable particles to the boundary surface is a spatially dependent process /2/. Consequently a laser-induced perturbation technique should be able to examine these spatial characteristics by confining the perturbation to well-defined regions of the ionization volume. Preliminary studies using a focused dye laser beam confirmed that the shape of the opto-galvanic signals varies with position and a more controlled study was made by aperturing the laser beam cross-section as indicated in Figure 8. Also shown in this figure are typical records for a specific value of E/N. The marked changes in shape are not a consequence of any intensity variations across the beam profile of the perturbing laser. The effect of varying the output intensity of this laser using neutral density filters to vary the intensity from 10 to 90% of the direct beam intensity showed no changes in the shape of the signals. The explanation of the phenomena is associated with the spatial variations of the number densities of metastable particles across the ionization region and detailed examination of this situation is at present in progress.

The time constants associated with the slow recovery of the ionization current due to diffusion of metastable particles to the boundaries were evaluated by plotting the
Figure 8. Spatially dependant OG signals showing fast and slow components for constant $E/p = 100 \text{ V cm}^{-1} \text{ torr}^{-1}$, $p = 1 \text{ torr}$.

$I$ vs $t$ variations for those portions of the opto-galvanic signals beyond the minimum value. In each case the time constant, as judged by the slope of the $\ln I$ vs $t$ plot for each perturbed position, is identical. No evidence appears indicating any contributions to the recovery from more than one metastable particle. This is consistent with the view that the number density of the other $1s_4$ metastable atoms is some two orders of magnitude less than that of the $1s_4$ metastable atom. It might also be expected that the diffusion-controlled recovery characteristics of the slow component of the opto-galvanic signals should also be similar to the growth characteristics observed when the equilibrium steady-state current is established repetitively by using a chopped u.v. light source to generate the initial electrons (Fig. 2(a)). Figure 9 compares the corresponding $\ln I$ vs $t$ data thus confirming expectations. An explanation for the changes in the observed time measured from the time of generation of the perturbing laser pulse to the time corresponding to the minimum value of the opto-galvanic signals, as the position of the perturbation is varied, requires detailed consideration of the spatial dependence of number density of the metastable particles. Appropriate computer-based programmes are being developed to examine this matter more thoroughly.

IV - APPLICATIONS TO OTHER GAS SYSTEMS

Reference was made in §I and II above to fundamental studies of the basic processes controlling the reproducibility of both the spatial and temporal ionization growth in $N_2$. These have been reported elsewhere //4/ and highlight the dramatic

Figure 9. Semi-log plot of difference $I_T - I(t)$ as a function of time for constant $E/p = 100 \text{ V cm}^{-1} \text{ torr}^{-1}$. ($-$) data of figure 2(a); (x,o,$\bullet$) data of figures 8(a),(b),(c).
consequences of improving the purity of the gas samples. With progressive removal of the common gaseous impurities high energy metastable particles remain unquenched and their concentrations build up significantly. This leads to situations where, very close to the threshold for breakdown, extremely sensitive control of the ionization current can be exercised through very small changes to the electrical parameters. Such a situation opens up the prospect of achieving similar control by optical means through the use of opto-galvanic effects as described in the present paper. When two or more metastable particles are involved in the ionization growth, and this is common in many gas laser mixtures as well as in the specific case of \( \text{N}_2 \) that has arisen, then the ability provided by the technique described here to display directly the contributions to the ionization growth by the individual processes should be particularly helpful in unravelling the complex phenomena.

ACKNOWLEDGEMENTS

This particular work is supported by the Australian Research Grants Scheme and has also benefited from help provided by the Australian Institute of Nuclear Science and Engineering. One of us (S.A.B.) also wishes to thank the Electrical Research Board of Australia for its financial support towards a post-graduate scholarship. The authors are also indebted to Mr G. Cooper for his assistance in developing the data acquisition facilities used in the investigations.

REFERENCES

6. PENNING, F.M., Physica 8 (1928) 137.