Optogalvanic Observation of Ionization Waves in Hollow-Cathode Discharges

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(Received 14 April 1986)

A damped oscillation at about 5 kHz was observed on the optogalvanic signals of a Ne hollow-cathode discharge on illumination by a pulsed dye laser at the resonant frequency of the Ne $2p_8$-$1s_5$ transition. The signal fits the formula $\Delta V(t) = V_0 e^{-t/\tau} \sin(\omega t + \theta)$, and the values of the parameters were determined for several positions of illumination and discharge currents. The oscillation originates from the ionization wave in the positive-column region, and a qualitative interpretation is presented based on the theory of striation.

PACS numbers: 51.50.+v, 52.80.Hc

In the past, moving striations and ionization waves have been investigated by the application of voltage or current pulses to probes, the discharge electrodes, or external ring electrodes,[1] and the optical observation by Ausschnitt and Bjorklund[2] of laser-induced striations in a hydrogen discharge plasma, in which the direct photoionization of hydrogen atoms caused the transient excitations of striations, suggested that striations can be excited by the optogalvanic (OG) method. Photon-induced ionization changes in a neon-filled hollow-cathode discharge have been reported by Smyth, Keller, and Crim,[3] and it has been shown that substantial changes in the Ne^+ concentration result from the excitation of neutral-neon transitions by irradiation with a cw tunable dye laser.

We report here on the OG signals of a hollow-cathode discharge of Ne which has been found to undergo a damped oscillation when a pulsed dye laser was used. The current and the position-of-illumination dependent features of the damped oscillation have led us to conclude that the ionization wave which is excited by the laser pulse inside the positive-column region of the discharge is causing the oscillation.

The experiment was carried out on a see-through hollow-cathode lamp[4] operated at a condition where no self-sustained striations took place, and by tuning of a pulsed dye laser (Princeton Applied Research model 2100), which has a duration time of 1 nsec with typical output power of 20 μJ/pulse, at 633.4 nm, the transition wavelength of the Ne $2p_8$-$1s_5$ line. Transient OG signals as obtained by directing the laser beam at various positions within the discharge as shown in Fig. 1 were detected and fed into a programmable digitizer (Sony Techtronics model 390AD) which had an input impedance of 1 MΩ. The sample rate was 1 MHz and 30 pulses were accumulated. Typical examples of the observed signals are shown by the solid curves in Fig. 2. Similar signals appeared on tuning of the laser to the $2p_4$-$1s_5$ transition at 594.0 nm, but the irradiation at nonresonant wavelengths failed to give detectable signals. The oscillating component of the signals was simulated by fitting with the formula,

$$\Delta V(t) = V_0 e^{-t/\tau} \sin(\omega t + \theta), \tag{1}$$

where $\Delta V(t)$ in millivolts is the OG signal at time t after the firing of the laser pulse, and V_0, τ, ω, and θ are parameters. Table I lists the derived values of the parameters for each configuration of the experiment (Fig. 1), and the calculated curves are shown by dashed lines and the residue signals of the observed minus the calculated signals by dotted curves in Fig. 2. The short-time signals for $t < 250$ μsec as seen in Fig.

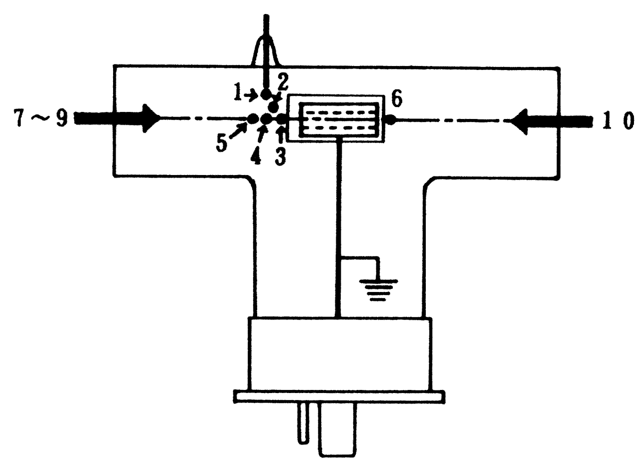

FIG. 1. Directions and positions of the laser illumination. Circles and arrows show, respectively, that the laser beam was directed perpendicular and parallel to the axis of the cathode cylinder, and the attached numbers refer to the first column of Table I.

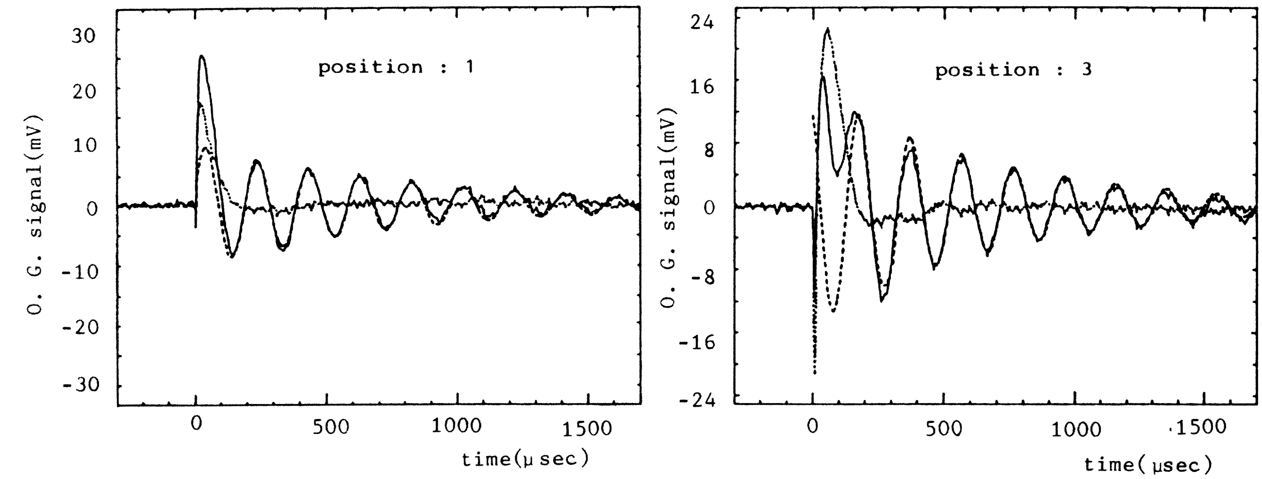

FIG. 2. Transient OG signals of Ne $2p_8$-$1s_5$ line at 633.4 nm (see legend of Fig. 1 for the attached numbers). Solid curves are the observed signals, the dashed lines are the simulated curves, and the dotted curves corresond to the residual, i.e., the observed minus the simulated signals.

2 are left out of the present analysis. The variation of temporal behavior with the spatial location as observed for the OG signal of molecular iodine[5] did not show up in the present experiment.

Of three possible sources of the damped oscillation, oscillation due to electrical circuitry and the excitation of acoustic waves are eliminated leaving the ionization wave as the sole candidate. It is inconceivable that the illumination of the plasma at the transition frequency of neutral Ne can give rise to a transient inductance with constant value of several henries. Acoustic waves are exempted on consideration that the sound velocity in Ne is 450 m/sec at 19 °C. The observed frequency of about 5 kHz gives the wavelength of 90 mm requiring a boundary length of 45 mm for an axial mode and a diameter of 53 mm for a γ_{10} radial mode, both of which do not coincide with any part of the discharge tube. The current-dependent feature of the oscillation is another reason to disregard the acoustic wave. On the other hand, the operation of the discharge tube above 5 mA gives rise to self-sustained oscillation of about 5 kHz and this undoubtedly is the ionization wave. We note the near coincidence of this frequency with those of the OG signals. The initial amplitude V_0 is much larger when the position of illumination is between the electrodes, i.e., in the region where the ionization wave takes place. Actually, the variation of ω, τ, and V_0 with the discharge current can be inter-

TABLE I. Derived values of parameters for various optical configurations of illumination and discharge current.

Optical config.[a]	I[b] (mA)	V_0 (mV)	τ (μsec)	ω[c] (msec^{-1})	θ[d] (rad)	θ/ω (μsec)
1	3.5	9.74	810	32.1	0.2	6.2
2	3.5	48.1	670	32.1	0.8	25
3	3.5	14.7	700	32.1	2.2	69
4	3.5	3.55	680	32.1	1.5	47
5	3.5	0	...	...	...	...
6	3.5	2.03	680	32.1	5.6	175
7	3.5	22.7	650	32.1	3.8	118
8	3.0	18.0	550	30.1	4.0	133
9	2.5	13.8	350	28.0	3.6	129
10	3.5	16.2	700	32.1	3.7	115

[a]The directions and positions of illumination as shown in Fig. 1.
[b]Discharge current.
[c]Angular frequency of the damped oscillation.
[d]Phase factor θ as derived on the assumption that V_0 is positive and that $\Delta V(t=0)=0$. In the case that $V_0<0$, $\pi/2=1.57$ has to be subtracted from θ, and if $\Delta V(t=0)$ takes the maximum value then another factor of $\pi/4$ has to be added.

preted in terms of an ionization wave as described below.

The illumination of the discharge gas by pulsed laser light whose frequency is in resonance with the Ne atom leads to a spatially and temporally localized increase of the population density of Ne in its $2p$ levels at the expense of the population of the metastable $1s$ levels. It will be safe to assume that the collisional mixing among the levels in the $2p$ manifold and among those in the $1s$ manifold is fast compared with the time scale of the present experiment. The change in the degree of ionization is due to the resulting change in the rate of ionizing collisions that directly or indirectly involve the Ne($2p$) states. The change in the number of fast electrons due to the change in the frequency of superelastic collisions of excited Ne with the electrons is believed to be one of the important mechanisms of the OG effect in glow discharges,[6] and contributions from multiphoton ionization might also affect the degree of the ionization of plasma. The localized and abrupt change of ionization will trigger the excitation of the ionization wave, and the change in the degree of ionization, either transient or oscillating, results in a change of the impedance of the discharge plasma as a whole that causes the transient OG signal. According to the theory of ionization described by Lee, Bletzinger, and Garscadden,[7] which was based on the ideas of Pekarek,[8,9] a pulse-excited ionization at $t=0$ causes the ion density $n_+(z,t)$ to vary according to the expression

$$n_+(z,t)=n^0(z)+\Delta n e^{-t/\tau}\sin[k_i(-\beta t+z+\epsilon)], \tag{2}$$

where z is the coordinate taken along the direction of the ionization wave, $n^0(z)$ is the ion density under stationary state, Δn is the initial amplitude of the ionization wave, and k_i is the magnitude of the wave vector whose value is defined by the boundary condition $k_i=2\pi/\lambda_i$, where λ_i is the wavelength of the ionization wave. Quantities τ and β determine the dependence of the decay time and frequency on discharge conditions. The phase term ϵ is introduced to take care of such factors as the dependences on the location and intensity of the pulsed illumination or on the discharge current (see Ref. 7 for further details and assumptions involved in the above quantities and equations).

The conductivity $\sigma(z,t)$ of a plasma at point z and time t can be expressed by

$$\sigma(z,t)=\alpha_e n_e+\alpha_+ n_+(z,t), \tag{3}$$

where n_e and n_+ are the densities of electrons and ions, respectively, and α_e and α_+ are proportionality constants. A uniform distribution of electrons is assumed since the electrons move sufficiently fast that a local buildup of the electron density is unlikely.[8,9] The impedance of the positive-column region is then given by

$$R=R_0+\Delta R(t)=q\int[1/\sigma(z,t)]dz, \tag{4}$$

where R_0 and $\Delta R(t)$ are the equilibrium and transient values of the impedance, respectively, q is a constant, and the integration is taken over the region where the ionization wave extends. From Eqs. (2) and (3) we obtain $\sigma(z,t)=\sigma^0(z)+\Delta\sigma_+(z,t)$, where $\sigma^0(z)=\alpha_e n_e+\alpha_+ n_+^0(z)$ and $\Delta\sigma_+$ is the laser-induced change in conductivity, and

$$\frac{1}{\sigma(z,t)}=\frac{1}{\sigma^0(z)[1+\Delta\sigma_+(z,t)/\sigma^0(z)]}\approx\frac{1}{\sigma^0(z)}\left[1-\frac{\Delta\sigma_+(z,t)}{\sigma^0(z)}\right]. \tag{5}$$

Substitution of Eq. (5) into Eq. (4) gives

$$R_0=q\int[1/\sigma^0(z)]dz=q\int\{1/[\alpha_e n_e+\alpha_+ n_+^0(z)]\}dz, \tag{6}$$

$$\Delta R(t)=-q\int\{[\alpha_+\Delta n_+ e^{-t/\tau}\sin[k_i(-\beta t+z+\epsilon)]/[\alpha_e n_e+\alpha_+ n_+^0(z)]^2\}dz. \tag{7}$$

The ionization wave as expressed by Eq. (2) is thus converted into the oscillation of plasma impedance as given by Eq. (7). The ionization wave inside the positive column might also induce an oscillation in the number of ions that hit the cathode surface which adds to the OG effect through ion current and the secondary emission of ions and electrons.

The frequency ν_i and wavelength λ_i of the oscillation are given as[7]

$$\nu_i=k_i\beta/2=\beta/\lambda_i \tag{8}$$

and

$$\lambda_i=2\pi/k_i=2L, \tag{9}$$

where L is the linear dimension of the ionization wave's extent. The phase velocity of the wave is equal to β, and Eqs. (8) and (9) give the value of β as 89, 96, and 105 m/sec for steady-state discharge currents of 2.5, 3.0, and 3.5 mA, respectively, with use of the interelectrode distance of 10 mm. It is seen from Table I that the decay time τ increases with the discharge current.

In conclusion, a damped oscillation was observed on the optogalvanic signals when a Ne hollow-cathode discharge was illuminated by a resonant laser pulse and this has been explained as due to an ionization wave stimulated by the pulsed excitation of Ne from the $1s_5$

level to the $2p_8$ level. A quantitative analysis was not possible because the geometry of the discharge tube was not designed for that purpose and the exact pressure of the fill gas was not known.

The ease of temporal, spatial, and intensity control of laser light and simple setup of the OG experiment provides a technique which is suited for extensive investigations of various kinds of gas discharges.

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