Journal of the Korean Vacuum Science & Technology; JKVST
Vol.7, No.1, 2003, pp.18~22

Observation of Penning ionization using the optogalvanic effect

Kee Ju Jeong† and Jun-Hoi Lee*

Department of Physics Education, Kongju National University, Kongju 314-701
**Department of Electronic Materials Science, Deajeon University, Deajeon 300-716*

(Received January 16, 2003)

Abstract

The optogalvanic effect is proposed and demonstrated as a technique for Penning ionization in a discharge of mixtures of metal vapors and rare gases. The gadolinium and argon mixture is used as a prototype. The lowest metastable of argon, $3P_2$ ($1s_5$ in Paschen notation) at 93144 cm^{-1}, is within kT from the excited states of Gd ion. Thus Penning ionization occurs to an excited states of the ion. This process strongly alters the optogalvanic signal and has its own signatures.

Keywords : Penning ionization, optogalvanic signal, hollow cathode

1. Introduction

We report a technique of utilizing the optogalvanic (OG) effect for the study of Penning ionization [1]. This effect has been used successfully in other spectroscopic applications such as atomic and molecular spectroscopy [2,3], atomic line profiles [4], plasma diagnostics [5], etc. The optogalvanic effect is based on the response of the discharge to changes in the population distribution of the atomic levels. It has been shown in a detailed model of a discharge [6] that a larger population density in the higher excited states in a discharge results in an increase in electron density and ionization rates. Subsequently, the discharge conductivity and the voltage applied to the discharge may change. In the OG effect a change in the population distribution is induced by a laser in a cw mode whose photon energy matches one of the atomic transitions. This causes a voltage change which is detected on an oscilloscope. The voltage signal has a positive and negative signal depending on whether the lower level is metastable or not. A detailed theory for the time dependence of the OG signal by a diode laser has recently been published elsewhere [7]. Most of the early works on the OG effect dealt with one species of atoms in the discharge tube, mainly Ne. In this work we are concerned with atomic processes in discharge, specifically rare-earth metal vapor and Ar and propose and demonstrate the applicability of the OG effect to the Penning ionization.

A major factor contributing to the discharge conductivity is ionization of the rare gas atoms by electron impact and collisions among the excited atoms. In addition some positively charged ions collide with the cathode surface and liberate atoms. Some of the excited rare gas atoms may collide with the sputtered atoms and cause further ionization [1] as

$$Ar^* (1s_5) + M^0 \rightarrow Ar^0 + M^+ + e^- \qquad (1)$$

where M represents the sputtered atoms. Penning ionization as well as rare gas ionization take place at all current. Several other collisional ionization mechanisms are thought to be less than the electron impact and Penning process. Since the electron density is much

† E-mail : keeju@kongju.ac.kr

larger than that of sputtered atoms the effect of Eq. (1) is not noticed in general at high discharge currents. At high discharge current, electron impact ionization process described by

$$Ar^{*}(1s_5) + e^{-} \rightarrow Ar^{+} + 2e^{-} \qquad (2)$$

where $Ar^{*}(1s_5)$ is the Ar atom in its lowest metastable level.

An understanding of such phenomenon requires the study of temporal variation of OG signals. We have systematically recorded the temporal evolution of OG signals by atomic transitions of Gd/Ar, La/Ar HCD. We wish to deal mainly with metastable effect and its influence on the OG signal. Our aim is twofold, first to show a Penning ionization process exists in which the lowest argon metastable level ionizes rare-earth metal, and second to exhibit the use of the OG effect and its related signal in the study of Penning ionization.

2. Experiment

The experimental setup is shown schematically in Fig. 1. A commercial HCD tubes (Cathodeon Ltd., model 3QQAY/Gd, 3QQAY/La) were used in the experiment. It consists of a hollow cylindrical cathode inner diameter of 2 mm with two-ring anodes and contains argon gas at a pressure of about 5-8 mbar. The discharge was produced by a high voltage power supply (Bertan Associate, Inc., model series 105). Current limiting resistors of 10 kΩ were used for each anode and the OG signal was detected by blocking the dc voltage using a coupling capacitor of 0.01 μF. We used single-mode diode laser systems which have center wavelengths of 810 nm (New Focus Inc., 6200 controller with model 6225 laser head). An anarmorphic prism pair and an iris of 0.5 mm diameter were used to make an appropriate laser beam shape. The laser beam path was carefully djusted to pass through the center of the negative glow region inside the cathode. The laser beam power in front of the HCD tube was 1 mW and more than 90% of the input power was transmitted through the HCD tube with the discharge off. With the laser wavelength fixed at the center of a transition line for Ar, the dynamic OG signal was obtained with a digitizing oscilloscope (LeCroy Inc., model 9450). An acousto-optic modulator (NEOS Inc., model N23080-1SAS) was used as a beam chopper to switch the laser beam on and off.

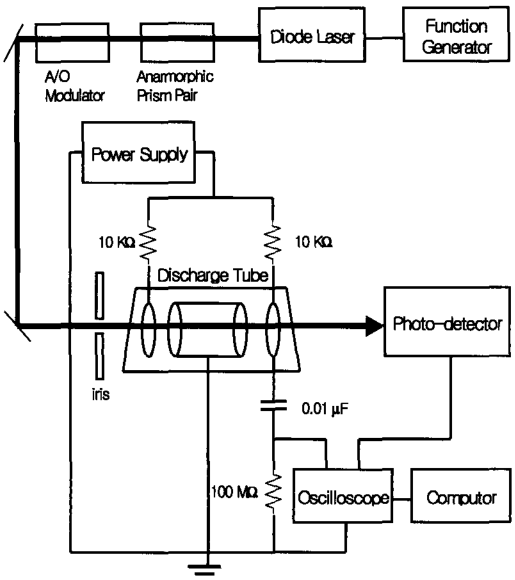

Fig. 1. Experimental setup.

3. Results and Discussion

Figure 2 shows a partial energy diagram of the relevant levels in the Penning ionization process and OG effect in Gd/Ar, La/Ar mixture discharges. Both Gd, La and Ar ground states appear at a common zero level, while Gd^{+}, La^{+} states appear above the Gd, La levels in order to illustrate the energy match Gd, La levels in order to illustrate the energy match occurring in Gd/Ar, La/Ar and yielding efficient Penning ionization process. The process is

$$Ar^{*}(1s_5) + Gd \rightarrow Ar^{0} + Gd^{+} + e^{-} + \triangle E(23\,cm^{-1})$$

$$Ar^{*}(1s_5) + La \rightarrow Ar^{0} + La^{+} + e^{-} + \triangle E(1570\,cm^{-1})$$

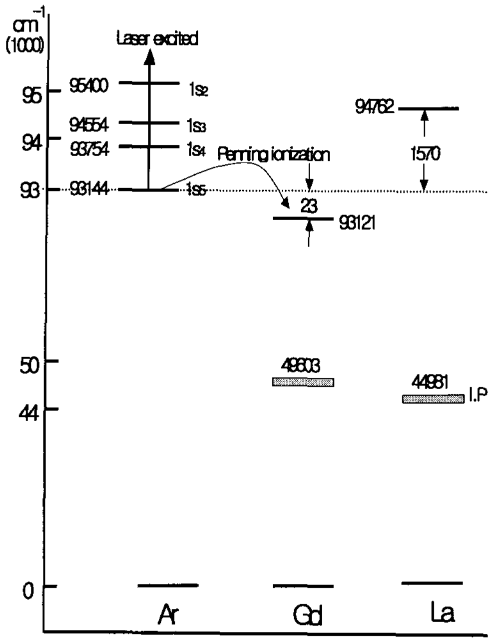

Fig. 2. Partial energy diagram for the relevant Gd, Gd^+ and La, La^+ and Ar atomic levels related to Penning ionization of Gd by argon.

where the difference in energy ΔE is the energy mismatch of the collision partners. The excited state of Gd^+ is at 93121 cm^{-1} [8] from the ground states of neutral gadolinium. This level is within the thermal energy kT which is ascribed to the translational energy of the colliding particles from the metastable level of argon $1s_5$ at 93144 cm^{-1} [9], where T is the translational temperature of the gas in the discharge. But the excited state of La^+ is at 94714 cm^{-1} [8] from the ground states of neutral lanthanum. This level is not within kT. The small energy mismatch is the main reason for Penning ionization via energy transfer collisional because energy transfer collisions between the lowest metastable atoms of the argon gas and the sputtered metal atoms are efficient whenever the mismatch in energy is less than 200 cm^{-1} [10]. Thus Penning ionization of gadolinium by argon metastable states can be readily achieved. The laser at a wavelength 801.479 nm depletes the argon metastable state. As a consequence, less Penning ionization of gadolinium occurs. These processes have a strong effect on the discharge characteristics and should influence and alter the OG signal.

The OG signals from in transition originating from the metastable levels are positive voltage signal at discharge current of 10 mA. The typical OG signals having a response time of about several tens μs to reach its steady state [7].

The OG signal is obtained by a diode laser at 801.479 nm to excited the argon metastable state $1s_5$ to $2p_8$ state in two HCD tubes. This transition is from the lowest metastable state, which is the most heavily populated state of argon. Fig. 3 shows the time-dependent OG signals obtained in the La/Ar HCD at discharge current of 6.5 and 10 mA. The laser excitation from the argon metastable level cause the negative peak of

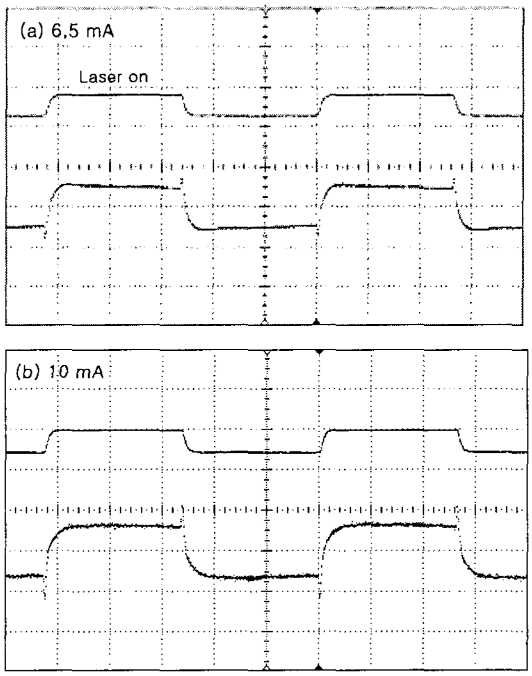

Fig. 3. Oscilloscope trace of the time-dependent OG signal of the $1s_5-2p_8$ transition in La/Ar HCD. The laser wavelength is 801.479 nm and discharge current is 6.5 and 10 mA. The upper trace indicate laser on. The down trace indicate the OG signal. The typical OG signal having a response time of about several tens μs to reach its steady state is appeared at all discharge currents.

signal. After fast relaxation, this is followed by a positive component as a consequence of the decrease in the metastable population. These two features are typical pure argon OG signal, occur in several tens μs in experiment. In other words the OG signal corresponds solely to transitions and enhanced ionization in the argon manifold, while lanthanum is a bystander as far as the OG effect is concerned.

Figure 4 shows the signal obtained in the Gd/Ar HCD at two currents. The OG signal at discharge current of 6.5 mA is Penning ionization dominated, while the higher current OG signal is not. This signal of Fig. 4(a) differs markedly from that of argon. This part of the

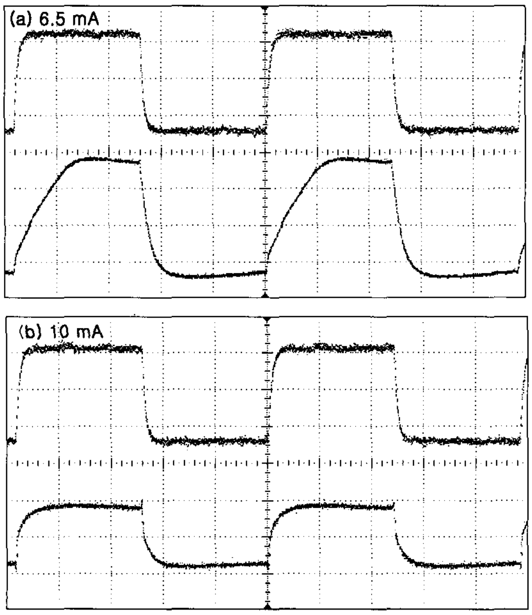

Fig. 4. Oscilloscope trace of the time-dependent OG signal for the $1s_5-2p_8$ transition at 801.479 nm in Gd/Ar HCD at the discharge current of 6.5 mA and 10 mA. The upper trace indicate laser on. The down trace indicate the OG signal. (a) The specific OG component which shows a response time of a few hundred μs. This effect is related to the Penning ionization. (b) The typical OG signal at discharge current of 10 mA. As the increasing discharge current, the Penning ionization effect on the OG signal is disappeared.

signal is dominant as is clearly exhibited by comparing Fig. 3(a) and 4(a). It has specific components having a response time of about a few hundreds μs to reach its steady state. The effect is related to the Penning ionization of gadolinium ground state to the excited level 93121 cm^{-1} of Gd^+ by metastable argon and is explained as follows. Resonant laser excitation from a argon metastable state reduces the Penning ionization of Gd due to the decrease of the metastable population. The resulting decrease production of Gd^+ and increase in Gd density cause a specific component OG signal. This phenomenon agrees with the earlier experimental observation of the Penning ionization effect in Ne HCD by direct measurement of both the buffer gas and the sputtered ions by Smyth et al [1].

With increasing discharge current after a special OG signal, the Penning ionization effect on the OG signal disappeared. The metal atom density decrease with increasing discharge current [1,11]. At higher discharge current, the increased electron in the hollow cathode cavity cause electron impact ionization to become the dominant ionization process. The Penning ionization plays a minor role under these conditions. Fig. 4(b) shows the dynamic OG signal at the discharge current of 10 mA. It has been understood that this typical OG signal feature is mainly due to the electron impact ionization process.

We have also checked the negative resistance region in voltage-current (V-I) curve of Cu/Ar and Cu/Ne HCDs [10]. The ion energy levels near the $1s_5$ level of Ar do not exist. Thus Typical OG signal is observed at all discharge current. The $1s_5$ levels of Ne is 134044 cm^{-1} and the nearest energy level of Cu ion is the 134237 cm^{-1} from the ground states of neutral copper. The energy mismatch is the 193 cm^{-1}. This level is within kT. Therefore the specific components having response time of about a few hundred μs in OG signal is observed. This also indicates the existence of Penning ionization process in Cu/Ne. Similar effects have been seen in Ca/Ne [12] and Fe/Ne [13] discharge tube by pulse laser. This makes the OG effect a powerful and attractive spectroscopic tools for investigating Penning ionization.

4. Conclusions

We have shown that the OG effect is an easy technique for the study of Penning ionization spectroscopy in hollow cathode discharge tube. The Penning ionization process is shown to play an important role in determining the OG effect and its time dependent OG signal. Our observation clearly suggests that Penning ionization is not characteristic to metastable levels alone and depends on the resonance in energy levels of the colliding particles. Penning ionization is efficient when the energy mismatch is within *kT*.

Acknowledgments

This work was supported by Kongju National University. Grant (2002-2221)

References

[1] K. C. Smyth, B. L. Bentz, G. Bruhn, and W. W. Harrison, J. Am. Chem. Soc. **101**, 797 (1979).

[2] K. C. Smyth, R. A. Keller, and F. F. Crim, Chem. Phys. Lett. **55**, 473 (1978).

[3] D. Feldmann, Optics. Comm. **29**, 67 (1979).

[4] T. F. Johnston, Jr., Laser Focus **14**, 58 (1978).

[5] C. P. Ausschnitt, G. C. Bjorklund, and R. R. Freeman, Appl. Phys. Lett. **33**, 851 (1978).

[6] R. Shuker, A. Gallahger, and A. V. Phelps, J. Appl. Phys. **51**, 1306 (1980).

[7] Jun-Hoi Lee and Kee Ju Jeong, J. Korean. Vacuum Soc. **11**, 119 (2002).

[8] W. C. Martin, R. Zalubas, and Lucy Hagan, NSRDS-NBS 60 (U.S. Department Commerce, 1978)

[9] C. E. Moore, *Atomic Energy Levels*, NSRDS-NBS 35 Vol. I (U.S. Department Commerce,1971)

[10] Jun-Hoi Lee, Hyuck Cho, E. C. Jung, and Jongmin Lee, J. Korean Phy. Soc. **38**, 99 (2001).

[11] R. Bleekrode and J. W. V. D. Laarse, J. Appl. Phys. **40**, 2401 (1969).

[12] R. Shuker. A. Ben-Amar, and G. Erez, J. Appl. Phys. **54**, 5685 (1983).

[13] B. R. Reddy and P. Venkateswarlu, Optics Comm. **85**, 491 (1991).