평면거울법에 의한 수은—아르곤 방전중의 6^3P 수은 원자의 농도 결정

Determination of the Concentration of 6^3P Mercury Atoms in a Hg-Ar Discharge by the Plane-Mirror Method

지 철 근* · 여 인 선**
(Chee, Chol-Kon · Yeo, In-Seon)

요 약

본 연구의 목적은 스펙트럼의 재흡수를 이용하여, 수은-아르곤 방전중의 6^3P 여기원자 농도를 결정하는 데 있다. 먼저, 평면거울을 사용한 경우의 재흡수 이론을 전개한 다음, 이를 수은-아르곤 방전에 적용시켜, 복잡한 미세구조를 갖는 수은의 visible-triplet lines 에 있어서의 스펙트럼의 흡수계수와 측정된 흡수율사이의 수치적 관계를 구하였다. 이와함께, 흡수계수와 흡수원자농도사이의 관계를 이용하여, 방전관의 관벽온도를 변화시킨 경우와 방전전류를 변화시킨 경우에 있어서, 측정 흡수율에 상응하는 수은의 6^3P 여기원자 농도를 구하였고 다음과 같은 결과를 얻었다.

i) 측정범위내에서 준안정원자의 농도는 6^3P_1 상태의 농도보다 높다.

ii) 방전전류를 증가시킴에 따라 435.8[nm]의 재흡수가 급격히 증가하고, 그결과 6^3P_1 여기상태의 농도는 준안정원자에 비해 상대적으로 높은 증가속도를 보인다.

iii) 방전전류 증가시 준안정원자와 전자사이의 충돌이 빈번해져서 준안정원자의 증가속도가 둔화하는 대신 축적여기 또는 전리의 확률이 높아진다.

1. Introduction

The positive column of low-pressure mercury-argon discharges has been the subject of many publications because of its wide application, especially in lighting fields, and a great deal of researches are being made to improve the discharge characteristics. Since the radiation characteristics depend mainly on the rate of producing the excited states of mercury atoms, it is necessary to know the variation in the concentration of the excited-state atoms. If the concentration can be explicitly determined, various physical processes involving the production and annihilation of excited mercury atoms during the interaction with other particles may be well described.

There are a number of techniques for determining the properties of the positive-column plasma. The probe-technique gives informations about the properties at a well-defined spot inside the discharge, but has a drawback of possible disturbances. On the contrary, the radiation emitted by it gives information without disturbing the plasma. For example, if transition probabilities are known, the radiation provides the concentration of atoms excited to the initial level of the lines. And reabsorption measurements make it possible to calculate the concentration of the lower level of the line concerned. 1)2)3)4)

The purpose of this paper lies in determining the concentration of excited states by using the reabsorption method, and in establishing its simple but effective performance. In this paper, the reabsorption of the visible-

---

*서울대학교 전기공학과 교수
**전남대학교 전기공학과 전임강사

triplet lines of mercury is studied to determine the concentration of 6^3P excited atoms in a low-pressure mercury-argon discharge. This study proceeds in the following sequences:

(1) The theory of reabsorption of a parallel beam is developed for the case of plane-mirror method, thus yielding a relationship between the absorption coefficient and the measured absorption.

(2) Then, the theory is applied to a low-pressure mercury-argon discharge, the absorption being measured for the visibletriplet lines of mercury.

(3) The concentration of 6^3P atoms is determined from the relationship obtained in (1) for relevant measured absorption.

2. Theoretical considerations

2. 1 The absorption coefficient

A gas discharge emitting a certain spectral line also absorbs this line in a measure depending strongly upon the concentration of the atomic state corresponding to the lower energy level of the line. It is well known that concentration of atoms at the level is related to the absorption coefficient:[2][3][4]

$$\int_{S.L.} k(\nu)\,d\nu = \frac{\lambda_o^2}{8\pi}\,\frac{g_2}{g_1}\,A_{21}\,N_1 \tag{1}$$

where $k(\nu)$ is the absorption coefficient at the frequency ν of the line, λ_o, corresponding to the transition from state 2 to state 1whose probability is denoted by A_{21}, and g_1, g_2 represent the statistical weights of the lower and upper levels, respectively. And the integration should be done over the spectral line considered.

If the absorption line has only a single component and has a Doppler contour, then

$$\int_0^{\infty} k(\nu)\,d\nu = \frac{1}{2}\sqrt{\frac{\pi}{\ln 2}}\,k_o\,\triangle\nu_D \tag{2}$$

where k_0 is the absorption coefficient at the center of the line and $\triangle\nu_D$ is the Doppler width. If the line consists of several components, all with Doppler contours, the frequency dependence of the absorption line is then of the form:[2]

$$k(\nu) = \sum_{\iota} k_{o,\iota}\, e^{-\left(2\ln 2\,\frac{\nu-\nu_{o,\iota}}{\Delta\nu_D}\right)^2} \tag{3}$$

Here $\nu_{o,l}$ are the frequencies of the individual hyperfine structure components, and $k_{o,l}$ the height at their centers. Or, the component may be specified by its relative intensity $k_{o,l}$ and its shift $\triangle\nu_l$ from the central component of the line:

$$k(\nu) = k_o \sum_{\iota} k_{o,\iota}\, e^{-\left\{\frac{2\ln 2}{\Delta\nu_D}(\nu-\nu_o-\Delta\nu_\iota)\right\}^2} \tag{4}$$

where k_0 is a factor which determines the absolute magnitude of the absorption coefficient. This expression, along with Eq.(1), establishes the relationship between k_0 and N_1.

2. 2 Reabsorption using the plane-mirror method[2]

The absolute value of k_0 can be determined by relating the measured absorption of a given line with a theoretical estimation. When the reabsorption is measured using the plane-mirror method as illustrated in Fig.1, the ratio between the line intensities with the mirror and those without it are considered. Here the reabsorption means the absorption of the light that has been emitted after experiencing self-absorption. The reflected light at the mirror surface can be thought of as if itemerged from imaginary tube.

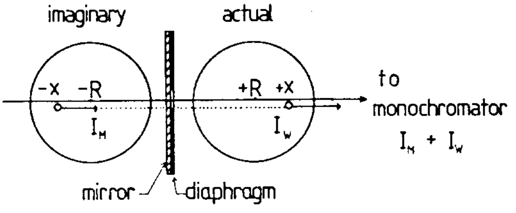


그림 1. 평면거울을 이용한 재흡수 측정.
Fig. 1. Reabsorption measurement using a plane-mirror.

The line intensities for frequencies between ν and $\nu + d\nu$ emerging from the actual discharge tube, $Iw(\nu)d\nu$, and those from the imaginary one, $Im(\nu)d\nu$, are expressed as follows:

$$I_W(\nu)\,d\nu = k(\nu)\,d\nu \int_0^{2R} \phi_2(x)\, e^{-\int_x^{2R} k(\nu)\phi_1(\xi)\,d\xi}\,dx \tag{5}$$

$$I_M(\nu)\,d\nu)\,d\nu = k(\nu)\,d\nu \int_{-2R}^{0} \phi_2(x)\, e^{-\int_x^{2R} k(\nu)\phi_1(\xi)\,d\xi}\,dx \tag{6}$$

where $\phi_1(x)$ and $\phi_2(x)$ are the distributions of absorbing (of state 1) and emitting atoms (of state 2) over the cross section of the discharge tube, respectively. Since the quantity $IM(\nu)d\nu$ represents the reduced portion of the original bean $IW(\nu)d\nu$ as the result of reabsorption the amount of absorbed light is equal to $IW(\nu)d\nu-Im(\nu)d\nu$. Thus, the absorption A_L for a spectral line is given by:

$$A_L = 1 - \frac{\int_0^\infty I_M(\nu)\,d\nu}{\int_0^\infty I_W(\nu)\,d\nu} \qquad (7)$$

This is related to the measured ratio M^* between the intensities with the mirror and those without it which may be expressed as

$$M^* = 1 + r\frac{\int_0^\infty I_M(\nu)\,d\nu}{\int_0^\infty I_W(\nu)\,d\nu} \qquad (8)$$

where r is the effective reflection coefficient of the mirror.

Now, the absorption A_L can be obtained from the eperimental value M^* and can be expressed as a function of k_0 through numerical integration(Eq.(9)), one can find out the corresponding value of k_0.

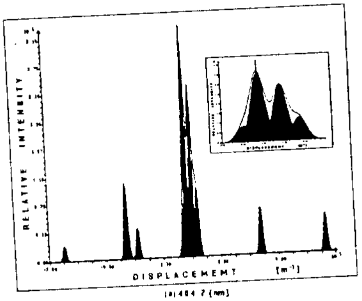
$$A_L = 1 - (M^* - 1)/r$$

$$= 1 - \frac{\int_0^\infty d\nu \int_0^{2R} k(\nu)\,\phi_2(x)\,e^{-k(\nu)\int_0^{2R}\phi_1(\xi)d\xi}\,e^{-k(\nu)\int_0^{x}\phi_1(\xi)d\xi}dx}{\int_0^\infty d\nu \int_0^{2R} k(\nu)\,\phi_2(x)\,e^{-k(\nu)\int_0^{2R}\phi_1(\xi)d\xi}\,e^{+k(\nu)\int_0^{x}\phi_1(\xi)d\xi}\,dx} \qquad (9)$$

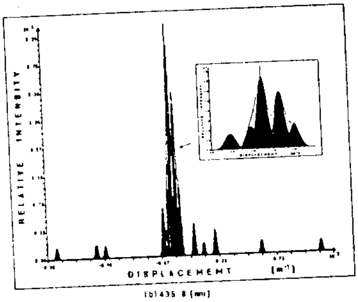
2. 3. Application to the visible-triplet lines of mercury

Since mercury spectra exhibit complex hyperfine structures and consequently consist of i components, the calculation of the concentration N_1 must be done according to Eq.(4). The relative intensities and the positions of the components are taken from Ref. 4, and are shown in Fig. 2. The transition probability A_{21} can be calculated from the lifetime of 7^3S_1 given by Holt and Pipkin[5] and the branching ratio given by Koedam and Kruithof.[4] Then, Xaccording to E.(1), the concentration N_1 can be expressed in terms of k_0 and T, the gas temperature.

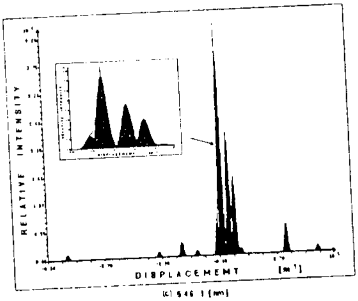
The results of calculation for A21 and N1 are given



(a) 404.7 (nm)



(b) 435.8 (nm)



(c) 546.1 (nm)

그림 2. 수은의 가시 3중선 스펙트럼의 초미세구조

Fig. 2. Hyperfine structures of the visible-triplet lines of mercury:
(a) 404.7[nm] (b) 435.8[nm] (c) 546.1[nm]

표 1. 수은 원자의 6^3p 여기 상태의 농도 N_1.

Table.1. The concentration N_1 of 6^3p states of mercury atoms.

$N_1 = \frac{8\pi g_1}{g_2 A_{21}\lambda_0^2}\int_{S.L.} k(\nu)\,d\nu$ Where $k(\nu)$ exhibits hyperfine structures.

Line[nm]	Series notation	g_2/g_1	$A_{21}[sec^{-1}]$	$N_1[m^{-3}]$
404.7	$7^3S_1-6^3P_0$	3	1.71E7	1.19E14$\sqrt{T}*k_0$
435.8	$7^3S_1-6^3P_1$	1	4.71E7	1.04E14$\sqrt{T}*k_0$
546.1	$7^3S_1-6^3P_2$	3/5	5.78E7	7.19E13$\sqrt{T}*k_0$

in Table 1.

And the integration (Eg.(9)) is performed numerically to obtain the relationship between k_0 and A_L, on the assumption that the distribution of absorbing atoms is of a parabolic shape:

$$\phi_1(x) = 1-\left(\frac{x-R}{R}\right)^2 \tag{10}$$

where R is the radius of the discharge tube. Futhermore, since, for most low-pressure discharges, the electron distribution also takes the same form as the above one, the rate of excitation of 7^3S_1 state becomes proportional to

$$\left\{1-\left(\frac{x-R}{R}\right)^2\right\}^2 \tag{11}$$

With these expressions, Eq.(10)-(11), Eq.(9) yields the k_0-A_L relationship for the visible-triplet lines of mercury, and are plotted in Fig. 3 for A_L against k_0R.

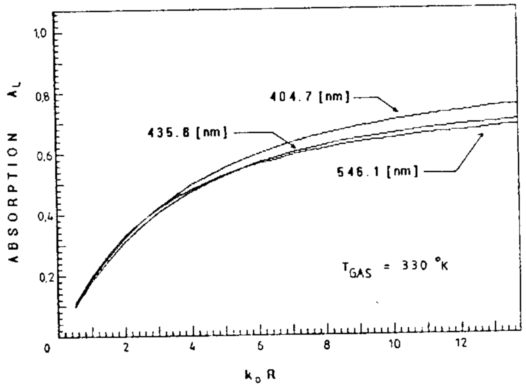


그림 3. A_L과 k_0R 사이의 관계.

Fig. 3. Relationship between A_L and k_0R.

3. Experiment

Fig. 4 shows the diagram of the apparatus used for the experiment. The central ray of the beam is collected by a monochrometer after being collimated. A photomultiplier tube is used as a radiation detector and the resulting singal is sent to a recorder. Reabsorption measurements are made by operating a shutter which is located between the discharge tube and the mirror, thus giving the value M*.

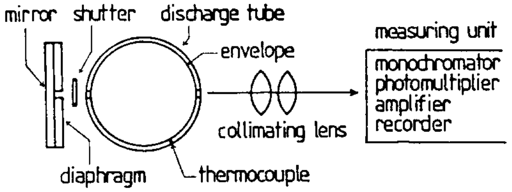


그림 4. 재흡수측정을 위한 실험 장치.

FIG. 4. Diagram of the apparatus for the reabsorption measurement.

At the same time, the temperature of the tube wall is measured with a thermocouple which is attached to the coolest point on the tube wall. Since the gas temperature within the discharge tube is, due to the internal heating, slightly higher than that on the tube wall, a value of 20[°C] will be taken for the compensation.[6]

The intensity ratio M* is measured in the two cases:

i) with the tube-wall temperature varied at a constant discharge current of 370 [mA], and

ii) with the discharge current varied at a constant ambient temperature of 20 [°C].

Variation of the tube-wall temperature is made by heating the entire room in still air in order to lessen the inhomogenity in the temperature, while the discharge current being controlled by adjusting the resistance of the ballast. In both cases, a discharge tube of radius 1.5[cm], filled with mercury plus argon of 3 [torr], is employed as a sourcs. And an average value of 0.9 is used for the effective reflection coefficient of the mirror.

4. Results and discussion

Results of the experiment are plotted in Fig. 5. As the tube-wall temperature rises, the absorption for all 6^3P states in. crease due to the rise in mercury vapor pressure (Fig. 5(a)). On the other hand, when the discharge current is varied, the absorption by 6^3P_1 state shows rapid increase, while the others vary only slightly (Fig. 5(b)), which will lead to the rapid rise in the concentration of 6^3P_1 state. But, within the range considered, the line, 546.1 [nm], is most absorbed, and thus a high value is expected for k_0.

The corresponding value of k_0 is obtained from the k_0-A_L plot, and then is used for the calculation of N_1.

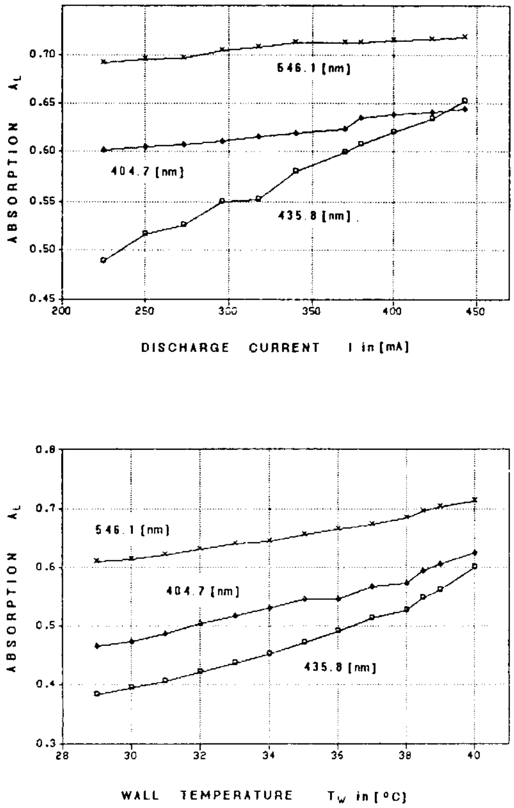


그림 5. 재흡수 측정의 결과

FIG. 5. The measured reabsorption A_L

(a) with the tube–wall temperature T_W varied at a constant discharge current of 370 [mA]

(b) with the discharge current varied at a constant ambient temperature of 20 [°C].

The final results are plotted in Fig. 6: the concentration N_1 (a) as a function of the tube–wall temperature and (b) as a function of the discharge current. A particular attention is paid to the sharp rise in the concentration of 6^3P_1 state in the case of varying the discharge current. As the current increases, the internal heating raises the vapor pressure of mercury, that is, the population in all the states. But since 6^3P_0 and 6^3P_2 states are metastable, their collisions with electrons become more frequent as the current increases, leading to the relatively sharp increase in the concentration of 6^3P_1 state.

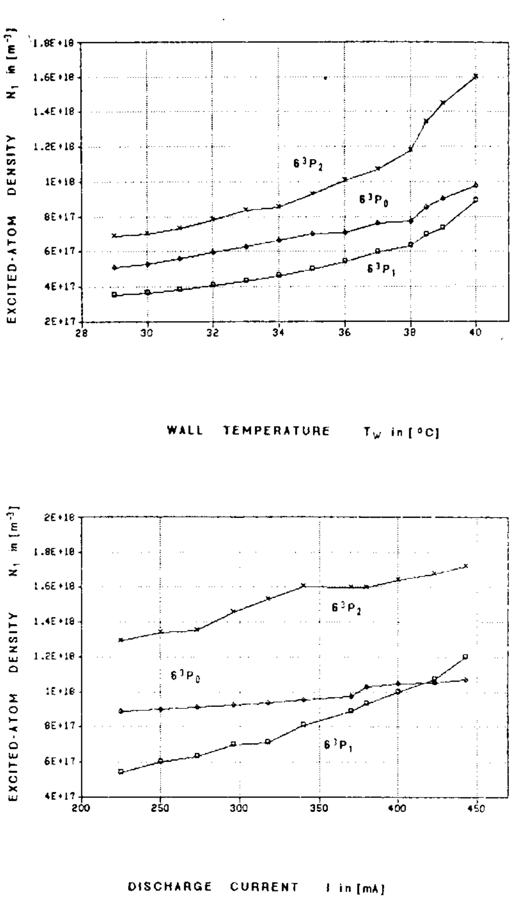


그림 6. 수은 원자의 6³P 여기상태의 농도변화

Fig. 6. The concentration of 6^3P states of mercury atoms

(a) as a function of the tube–wall temperature

(b) as a function of the discharge current.

5. Conclusion

The theory of reabsorption is developed for the plane–mirror method, and thus it is possible to relate the measured absorption to the concentration of atoms which absorb a particular spectral line exhibiting hyperfine structures. Application of the theory to a Hg–Ar discharge is made to determine the concentration of 6^3P states of mercury atoms, along with the measurement of reabsorption of the visible–triplet lines. These results shows:

(1) Within the range considered in the experiment, the concentrations of the metastable states exceed that of 6^3P_1 state.

(2) A relatively rapid increase is noticed in the absorption of 435.8[nm] as the discharge current is raised, which leads to a sharp rise in the concentration of 6_3P_1 state of mercury atoms.

(3) As the current increases, the collisions between electrons and metastable atoms become more frequent, and thus higher probality is expected for the cumulative excitation or ionization.

From the consideration given so far, it is concluded that the theory of reabsorption provides a simple but effective method in determining the concentration of excited atoms, and succeeds in the application to the visible–triplet lines of mercury.

References

1) W. Elenbaas, "Light sources", Macmillan, 1972, pp. 91.

2) Y.M. Kagan, et al., "The Use of the Reabsorption Method for a Line with Complex Structures", Opt. Spectrosc., Vol. 22, pp. 486–488. 1967.

3) A.C.G. Mitchell, M.W. Zemansky, "Resonance Radiation and Excited Atoms", Cambridge, 1971, pp. 92–175.

4) M. Koedam, A.A. Kruithof, "Transmission of the Visible Mercury Triplet by the Low Pressure Mercury– Argon Discharge", Physica, Vol. 28, pp. 80–100, 1962.

5) R.A. Holt, F.M. Pipkin, "Precision Measurement of the Lifetime of the 7^3S_1 State of Atomic Mercury", Phys. Rev., Vol. 9(2), pp. 581–584, 1974.

6) C. Kenty, et al., "Gas Temperatures and Elastic Losses in Low Pressure Mercury–Argon Discharges", J. Appl. Phys., Vol. 22(8), pp. 1006–1011, 1951.