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# Interferometric Measurement of Large Number Density of Metal Vapor in the Heat Pipe

By

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## Abstract

A method is proposed which permits one to determine a large number density of metal atoms along the absorption path in a heat-pipe cell. The proposed method requires a measurement of the anomalous dispersion near the absorption lines with known oscillator strengths by the use of a Mach-Zehnder interferometer and a stigmatic spectrograph. The method is tested on the resonance line of calcium and the first doublet of sodium. Its advantage over the hook method and the method by Frich *et al.* is demonstrated, and its precision and validity are discussed.

## 1. Introduction

A heat-pipe cell has been used in various experiments on metal vapors ever since it was devised by Vidal and Cooper in 1969.<sup>1)</sup> If we try to make quantitative photo-absorption measurements using this device as an absorption cell, we should know an integral  $\int_l N(x)dx$ , where  $N(x)$  is the number density of metal atoms in a pertinent state at a given position  $x$  along the light beam; and  $l$  is the total extent of the metal vapor in the cell. This paper is concerned with an interference method to determine the  $\int_l N(x)dx$  value from a measurement of the anomalous dispersion near the spectral lines with known oscillator strengths or  $f$  values.

One of the conventional methods to determine the  $\int_l N(x)f dx$  value from the dispersion measurement is the hook method. For this method we have some excellent reviews, *e.g.*, by Marlow<sup>2)</sup> and by Huber.<sup>3)</sup> In recent experiments of barium<sup>4)</sup> and rubidium,<sup>5)</sup> the hook method was actually applied to the resonance line or the second doublet with known  $f$  values to determine the  $\int_l N(x)dx$  values

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of these vapor atoms in the heat-pipe cell. However, when  $\int_l N(x)fdx$  becomes larger than  $10^{18} \text{ cm}^{-2}$ , it is difficult to apply the conventional hook method.

In the present paper, we propose a new interference method to determine the large  $\int_l N(x)fdx$  values from the interference fringe shift due to anomalous dispersion. Recently, Frich *et al.*<sup>6)</sup> proposed an interference method to determine the  $\int_l N(x)fdx$  value of  $10^{14} \text{ cm}^{-2}$ , using the fringe shift. In our proposed method we utilize the Mach-Zehnder interferometer, as in the case of the hook method and the method by Frich *et al.*

In the following section, the principle of the proposed method is shown. Section 3 describes the experiments in which the proposed method is applied to calcium and sodium vapor atoms in the heat-pipe cell. In §4, the proposed method is compared with the hook method and the method by Frich *et al.* In §5, a discussion is given about the precision and the validity of the proposed method.

## 2. Principle

We consider below atoms in the ground state. Also, we assume the uniform metal density  $N$  along the well defined absorption length  $l$ , so that  $\int_l N(x)dx = Nl$  for simplification. (These restrictions are not always necessary in the following discussion.)

When the Mach-Zehnder interferometer is used, the condition of the interference maximum is

$$(n-1)l - [(n'-1)l' + d] = k\lambda, \quad (1)$$

where  $n$  is the refractive index of a metal vapor layer and depends dramatically on  $\lambda$  near an absorption line;  $n'$  is that of a compensation medium and is weakly dependent on  $\lambda$ ;  $d$  is the path difference in the absence of a compensator and is independent of  $\lambda$ ; and  $k$  is an integer which gives the fringe order. Let us consider that  $k$  changes continuously as a function of  $\lambda$ . Then, a differential form of eq.(1) is

$$l \frac{d(n-1)}{d\lambda} - l' \frac{d(n'-1)}{d\lambda} = k(\lambda) + \frac{dk}{d\lambda} \lambda. \quad (2)$$

If we are interested in the spectral region outside the core of an isolated line at  $\lambda_0$  with an absorption oscillator strength  $f$ , the refractive index is given by

$$n-1 = \frac{e^2}{2\pi mc^2} \frac{\lambda_0^2 \lambda^2}{\lambda^2 - \lambda_0^2} Nf. \quad (3)$$

From eqs.(1), (2) and (3) we obtain

$$[A(\lambda) + B(\lambda)]Nl + K_m(\lambda) = K(\lambda), \quad (4)$$

where

$$A(\lambda) = \frac{e^2}{2\pi mc^2} \frac{\lambda_0^2 \lambda}{\lambda^2 - \lambda_0^2} f, \quad (4a)$$

$$B(\lambda) = \frac{e^2}{\pi mc^2} \frac{\lambda_0^4 \lambda}{(\lambda^2 - \lambda_0^2)^2} f, \quad (4b)$$

$$K_m(\lambda) = \frac{dk}{d\lambda} \lambda, \quad (4c)$$

and

$$K(\lambda) = \frac{1}{\lambda} [(n' - 1)l' + d] - l' \frac{d(n' - 1)}{d\lambda}. \quad (4d)$$

Note that  $K_m(\lambda)$  can be determined from the spectrum itself as

$$K_m(\lambda_i) = \frac{\lambda_i P_i}{\Delta \lambda_i} \quad (4c')$$

by counting the number  $P_i$  of the fringes within a wavelength interval  $\Delta \lambda_i$  at  $\lambda_i$ .

If we assume that  $d(n' - 1)/d\lambda$  is constant within the wavelength region concerned,  $K(\lambda)$  is given by

$$K(\lambda) - K(\lambda_c) = a \left( \frac{1}{\lambda} - \frac{1}{\lambda_c} \right), \quad (5)$$

where the two constants  $K(\lambda_c)$  and  $a$  are usually unknown. Equation (4) is therefore regarded as a set of equations with three unknown values:  $Nl$ ,  $K(\lambda_c)$  and  $a$ . Evidently, we can get many more equations than three by measurements at the various wavelengths  $\lambda_i$ . This allows us to determine the  $Nl$  value by the least squares method.

If we apply the present method to the case of closely spaced lines such as the  $D$  lines of alkali atoms, we have to replace  $A(\lambda)$  and  $B(\lambda)$  of eqs. (4a) and (4b) by

$$A(\lambda) = \frac{e^2}{2\pi mc^2} \sum_j \frac{\lambda_j^2 \lambda}{\lambda^2 - \lambda_j^2} f_j \quad (4a')$$

and

$$B(\lambda) = \frac{e^2}{\pi mc^2} \sum_j \frac{\lambda_j^4 \lambda}{(\lambda^2 - \lambda_j^2)^2} f_j, \quad (4b')$$

where  $\lambda_j$  is a wavelength of the  $j$ -th absorption line with an absorption oscillator strength  $f_j$ .

We now introduce another coefficient  $K_{\text{eff}}(\lambda)$  such as

$$K_{\text{eff}}(\lambda) = -k - l \frac{d(n'-1)}{d\lambda}. \quad (6)$$

From eqs.(1), (3), (4a), (4d) and (6), we obtain

$$K_{\text{eff}}(\lambda) = K(\lambda) - A(\lambda)Nl. \quad (7)$$

Two predicted features of  $K_{\text{eff}}(\lambda)$  and  $K_m(\lambda)$  are as follows: 1) Both  $K_{\text{eff}}(\lambda)$  and  $K_m(\lambda)$  approach  $K(\lambda)$  as  $\lambda$  is going far from the absorption lines. We call the region of  $K_{\text{eff}}(\lambda) \simeq K_m(\lambda) \simeq K(\lambda)$  the "line-free region". 2) At the hook positions  $\lambda_H$ ,  $K_m(\lambda)$  becomes 0 and  $K_{\text{eff}}(\lambda)$  becomes maximum or minimum since  $dk/d\lambda=0$  at  $\lambda_H$ .

From eqs.(4) and (7) we obtain

$$B(\lambda)Nl + K_m(\lambda) = K_{\text{eff}}(\lambda). \quad (8)$$

We can therefore expect that the plotting of  $K_{\text{eff}}(\lambda_i) = B(\lambda_i)Nl + K_m(\lambda_i)$  vs  $1/\lambda$  with a proper  $Nl$  value should show the maximum and minimum at the hook positions on both sides of an absorption line. In the present study, we will use this plotting to show a consistency with the  $Nl$  value determined by the least squares method.

### 3. Experimental

#### 3.1. Setup

Figure 1 shows a general experimental arrangement. Four mirrors (M1–M4) form the Mach-Zehnder interferometer. A heat-pipe cell with 5 mm thick quartz windows (C1) at both ends is placed in the test beam section. This heat-pipe cell is described in detail elsewhere.<sup>4)</sup> The interferometer is adjusted so that the lengths of both arms are almost equal to each other in the absence of the com-

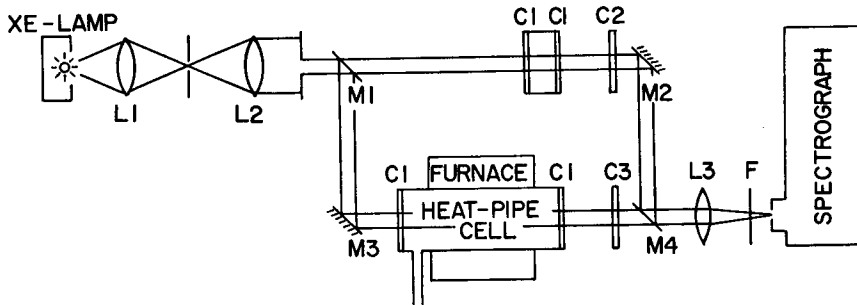


Fig. 1. General experimental arrangement: L1–L3, lenses; M1–M4, mirrors; F, a filter; C1–C3, quartz windows or plates.

pensation plates (C2 and C3), i.e.  $d \simeq 0$ . We utilize quartz plates of a different thickness, such as C2 and C3 to obtain fringes of a negative high order.

A xenon lamp was used as the continuum light source, and dispersion spectra were recorded on Kodak Tri-X film with a 170 cm Ebert-mounting spectrograph. A grating having 1200 grooves/mm was used in the first order, resulting in the linear dispersion of 4.7 Å/mm in the visible wavelength region. The slit width was 20–30  $\mu\text{m}$ . The exposure times were 1–4 sec. The Model 9825A Calculator and 9872A Graphics Plotter (HP) were used for the data reduction.

### 3.2. Dispersion measurement of calcium

Calcium vapor was contained in the heat-pipe cell which was heated to 1120–1242°K over the central 60 cm length. Krypton gas of 280 mb was used as buffer gas. Thus, our cell did not operate in the heat-pipe oven mode of Vidal and Cooper.<sup>1)</sup>

Dispersion spectra were observed in the wavelength region of  $\lambda$ 3825–4695 Å outside the core of the calcium resonance line  $\lambda$ 4227 Å with an  $f$  value of 1.75.<sup>7)</sup> Figure 2 shows an example of the densitometer traces of the dispersion spectra.

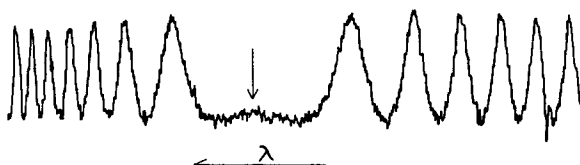


Fig. 2. The densitometer trace of the interferogram near the hook on the blue side of the resonance line of calcium  $\lambda$ 4227 Å. The arrow shows the hook position.

Emission lines from a helium discharge lamp were used as wavelength references, and the wavelengths of the fringes were measured by interpolation. In Fig. 3, the open circles show an example of the  $K_m(\lambda_i)$  values obtained by eq. (4c').

The proposed least squares method was applied to the blue wing of the resonance line of calcium  $\lambda$ 4227 Å. The unknown values  $Nl$ ,  $K(\lambda_0)$  and  $a$  in eqs.(4) and (5) were determined, where  $\lambda_c$  in eq.(5) was taken to be the wavelength of the resonance line  $\lambda_0=4227$  Å. A resonance line of impurity strontium  $\lambda$ 4607 Å prevented us from applying the proposed method to the red wing of the resonance line of calcium. In Fig. 3, the closed circles show  $[A(\lambda_i)+B(\lambda_i)]Nl+K_m(\lambda_i)$  of eq.(4) with the determined  $Nl$  value. The straight line of eq.(5) with the determined  $K(\lambda_0)$  and  $a$  values is shown with a solid line, which fits the closed circles very well. The open triangles show  $B(\lambda_i)Nl+K_m(\lambda_i)$  of eq.(8) with the determined  $Nl$  value. This plotting approaches  $K(\lambda)$  as  $\lambda$  is going far from

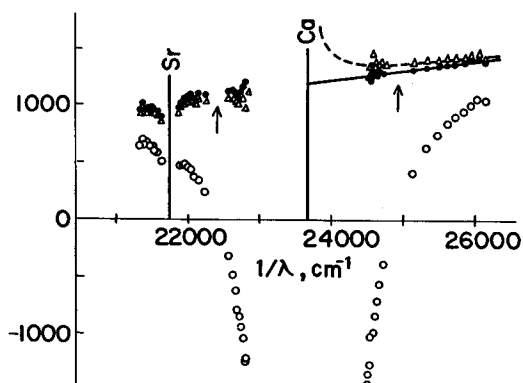


Fig. 3.  $K_m(\lambda)$ ,  $K(\lambda)$  and  $K_{\text{eff}}(\lambda)$  vs  $1/\lambda$  in the case of calcium:  $\circ$ ,  $K_m(\lambda_i)$ ;  $\bullet$ ,  $K(\lambda_i) = [A(\lambda_i) + B(\lambda_i)]Nl + K_m(\lambda_i)$ ;  $\triangle$ ,  $K_{\text{eff}}(\lambda_i) = B(\lambda_i)Nl + K_m(\lambda_i)$ ; a solid line, resultant  $K(\lambda)$ ; a dashed curve, resultant  $K_{\text{eff}}(\lambda)$ . Positions of the resonance line of calcium and impurity strontium are indicated by bars. The hook positions are indicated by arrows. The oven temperature was 1242°K. Determined  $Nl$ ,  $K(\lambda_0)$  and  $a$  values are  $1.99 \times 10^{18} \text{ cm}^{-1}$ , 1200 and 0.088 cm, respectively. See text for details.

$\lambda_0$ , and the maximum and minimum occur near the hook positions on both sides as predicted. A dashed curve fitting the open triangle shows the resultant  $K_{\text{eff}}(\lambda)$  of eq.(7).

### 3.3. Dispersion measurement of sodium

Sodium vapor was generated in the same cell as in the case of calcium. The cell was heated to 780–809°K. Krypton gas of 800 mb was added as buffer gas.

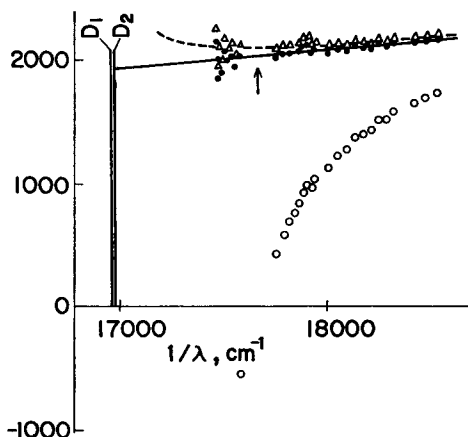


Fig. 4.  $K_m(\lambda)$ ,  $K(\lambda)$  and  $K_{\text{eff}}(\lambda)$  vs  $1/\lambda$  in the case of sodium with the same notation as in Fig. 3. The oven temperature was 809°K. Determined  $Nl$ ,  $K(\lambda_2)$  and  $a$  values are  $2.51 \times 10^{18} \text{ cm}^{-2}$ , 1930, and 0.15 cm, respectively.

Dispersion spectra were observed in the region of  $\lambda 5280\text{--}5730 \text{ \AA}$ , which corresponds to the red wing of the D lines  $\lambda 5896 \text{ \AA}$  and  $\lambda 5890 \text{ \AA}$ . Emission lines from an Hg-Ar discharge lamp were employed as wavelength references. Owing to a strong absorption by  $\text{Na}_2$  molecules we could not observe any interference fringes on the red wing of the D lines. The proposed method was applied to the above region, where  $\lambda_c$  in eq. (5) was taken to be the wavelength of the  $D_2$  line  $\lambda 5890 \text{ \AA}$ . The absorption oscillator strengths used here were 0.327 for  $D_1$  and 0.655 for  $D_2$ .<sup>8)</sup> An example of the results is shown in Fig. 4, with the same notations as in Fig. 3.

#### 4. Comparison with other methods

##### 4.1. Hook method

From eq. (8) with  $K_m(\lambda_H)=0$  we obtain

$$Nl = \frac{K_{\text{eff}}(\lambda_H)}{B(\lambda_H)}. \quad (9)$$

If  $K_{\text{eff}}(\lambda_H)$  is known, we can determine the  $Nl$  value by measuring the wavelength of the hook position. This is a basic idea of the hook method.

Let us consider for a while the case of a sufficiently small hook separation  $\Delta$ . We may approximate  $K_{\text{eff}}(\lambda_H) \simeq K(\lambda_0)$  for the isolated line at  $\lambda_0$ . Equation (9) then becomes

$$Nl = \left( \frac{e^2 \lambda_0^3}{\pi m c^2 \Delta^2} f \right)^{-1} K, \quad (10)$$

where  $K \equiv K(\lambda_0)$  is a so called hook constant. The hook constant can be determined by an interpolation of  $K_m(\lambda)$  in the line-free region. Thus, we can determine the  $Nl$  value by measuring the hook separation  $\Delta$ .

For a large hook separation, it is difficult to determine either  $K_{\text{eff}}(\lambda_H)$  or  $K(\lambda_0)$  from  $K_m(\lambda_i)$  by the interpolation as shown in Figs. 3 and 4. To test the validity of eq. (10) itself, we determined the  $Nl$  values from eq. (10), using the  $K(\lambda_0)$  values obtained by the least squares method. The results are in good agreement within a few percent with the  $Nl$  values determined by the least squares method. Thus, eq. (10) is considered to be a good approximation even for a large hook separation. This fact allows us to determine the  $Nl$  value approximately by measuring only the hook separation, once  $K(\lambda_0)$  is obtained. (Note that  $K(\lambda_0)$  is expected to be as well reproduced as far as  $d=0$  in each measurement.)

##### 4.2. Fringe shift method by Frich *et al.*

If we assume  $d(n'-1)/d\lambda=0$ , then eqs. (4d) and (7) become



$$K(\lambda) = \frac{1}{\lambda} d_{\text{eff}} \quad (11)$$

and

$$-k = \frac{1}{\lambda} d_{\text{eff}} - A(\lambda) Nl \quad (12)$$

respectively, where

$$d_{\text{eff}} = d + l'(n' - 1). \quad (13)$$

Equation (12) is regarded as a set of equations with three unknown values:  $k$ ,  $d_{\text{eff}}$  and  $Nl$ . Thus, we can determine the  $Nl$  value by the least squares method, as suggested by Frich *et al.*<sup>6)</sup>

However, in the situation where quartz plates are used to make the path difference, and dispersion spectra are observed in a large wavelength interval of several hundred angstroms, the above assumption  $d(n' - 1)/d\lambda = 0$  is not reasonable. Actually, the  $K(\lambda)$  of eq. (5) gives a straight line that does not pass through the origin in  $K(\lambda)$  vs  $1/\lambda$  plane as shown in Figs. 3 and 4, whereas the  $K(\lambda)$  of eq. (11) requires a straight line through the origin.

## 5. Discussion

To estimate the precision of the proposed method we use eq. (4). We have approximately

$$[A(\lambda) + B(\lambda)]\delta(Nl) \simeq -\delta P_i \frac{\lambda}{d\lambda}, \quad (14)$$

where  $\delta P_i$  denotes the measurement error in the number  $P_i$  of the fringes in eq. (4c'). If we assume  $\delta P_i \simeq 0.05$ , we find from eq. (14) a relative error of  $\delta(Nl)/Nl \leq 0.01$  under the present experimental conditions. In the present experiment, we actually determined both the  $Nl$  and  $K(\lambda_c)$  values with a high precision, better than 2 %, using the proposed least squares method. This shows a good agreement with the above prediction. However, the precision of the determined  $a$  value in eq. (5) was only slightly better than 10 %.

We have assumed that  $d(n' - 1)/d\lambda$  is constant. To test the validity of this assumption, we observed the interference fringes in the absence of metal vapor, and determined  $K_m(\lambda_i)$ , where  $K(\lambda) = K_m(\lambda)$ . The plot of  $K_m(\lambda)$  vs  $1/\lambda$  fits the straight line quite well within any interval of  $2000 \text{ cm}^{-1}$ . Thus the above assumption, and therefore eq. (5) are considered to be a sufficiently good approximation as long as the least squares fitting is restricted within  $2000 \text{ cm}^{-1}$ .

We now consider the effect of the rare gases admixed in the heat-pipe cell.

In the case when measurements are carried out under several rare-gas pressures keeping the cell temperature constant (ref. 9), we had better, if possible, readjust the Mach-Zehnder interferometer for every pressure so that  $d \approx 0$ . Then the resulting  $K(\lambda_c)$  and  $a$  values may be reproduced for each measurement.

There are possibilities that high-pressure rare gases affect the profile of the refractive index given by eq. (3). (As for the profile of the absorption coefficient, it is well known that high-pressure foreign gas induces an antisymmetric profile.) In the present experiments of calcium and sodium in the presence of krypton, the plotting of  $[A(\lambda_i) + B(\lambda_i)]Nl + K_m(\lambda_i)$  vs  $1/\lambda$  fits a straight line quite well, as shown in Figs. 3 and 4. In the case of calcium, the plotting of  $B(\lambda_i)Nl + K_m(\lambda_i)$  vs  $1/\lambda$  showed the maximum and minimum at the hook positions on both sides of the absorption line, as shown in Fig. 3. These facts suggest that the refractive index has approximately the same symmetric profile as described by eq. (3), with reasonable accuracy under the present experimental conditions.

We also tested the case of xenon. The results were quite similar to the case of krypton so far as dispersion spectra were observed on the blue wing. For calcium in xenon, however, the plotting of  $B(\lambda_i)Nl + K_m(\lambda_i)$  vs  $1/\lambda$  did not show the maximum at the hook position on the red side for the  $Nl$  value that gave the minimum at the hook position on the blue side. This asymmetric effect implies that the profile of the refractive index deviates from symmetric shape in the presence of xenon of  $\geq 200$  mb. We tentatively determined the  $Nl$  value in such a way that the plotting of  $B(\lambda_i) + K_m(\lambda_i)$  vs  $1/\lambda$  showed the maximum at the hook position on the red side. The results show a small disagreement—less than 10%—with the  $Nl$  values determined by applying the proposed least squares method to the blue wing.

In conclusion, we have proposed a method to determine large  $Nl$  values of the order of  $10^{18}$  cm<sup>-2</sup>, which are easily obtained in the heat-pipe cell operating under normal conditions of vapor pressure of several millibars. Compared with the difficulties of applying the hook method or the method by Frich *et al.*, the present method seems to be the most precise and reliable method for measuring such large  $Nl$  values.

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