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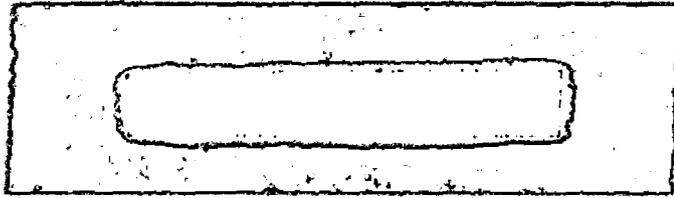
AN INTERFEROMETRIC STUDY OF ATOMIC
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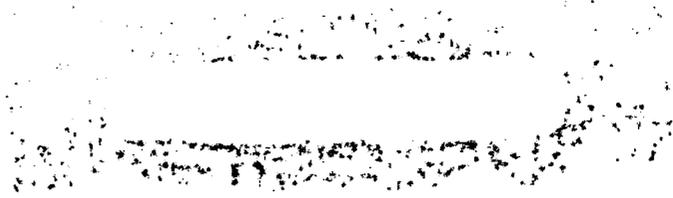
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Robert Berg

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University of California, Berkeley, California

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ABSTRACT

Optical conditions required for accurate measurement of atomic line shapes and relative line intensities with a Fabry-Perot interferometer are discussed. Measurements have been made on the 5461 Å line of three commercial mercury lamps.

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INTRODUCTION

Recent developments in optical spectroscopy and related fields have highlighted the importance of characterizing light sources as to line shapes and intensities. Narrow atomic spectral lines of high intensity are required as wavelength standards¹, for high resolution Raman spectroscopy², for certain absorption studies³, and in the recently developed field of optical pumping.¹⁵

Because of their special requirements many researchers in these fields are even making their own lamps. The causes of line broadening and distortion being well known⁴, the typical lamp now being constructed is filled at low pressure, to avoid pressure broadening and self reversal; operated at low temperature, to limit Doppler broadening; and excited by microwave power, to minimize electromagnetic field broadening.

In this laboratory atomic spectral lamps are used to excite molecular fluorescences⁵. Because each molecular electronic-vibrational absorption occurs over a range of rotational energy levels, broad exciting lines are not only permissible but desirable, so long as they are sufficiently intense. However, no data are available on the line profiles of commercially available lamps and there are very few references in the recent literature to measurement of both line width and intensity of laboratory made sources. (See Table I).

Because of the dearth of such data a study of lamps available in our laboratory was begun by Dr. Earl Worden⁸. Line profiles were observed on 3 meter and 21 foot grating spectrographs and absolute intensities were determined by comparing lamp intensity to the intensity of a tungsten filament lamp of known blackbody temperature. The results of that study suggested work at higher resolution. Accordingly, this project was undertaken to obtain more accurate line profiles by using a Fabry-Perot interferometer.

Table I
Data on recently reported line sources

Line	Half Width	Absolute Intensity watts-cm ⁻² -steradian ⁻¹	Intensity* in °K	Reference
Hg 5461 Å	.03 Å	.010	6300	6
	.006	.0001	3700	6
	< .20	.025	> 5100	7

It was not possible to use this method for comparing intensities with a continuous blackbody source as on a spectrograph because of the narrow free spectral range of the interferometer; no filter was available to select a line from the blackbody continuum as narrow as the atomic lines under investigation.

Mercury high pressure lamps producing rather broad lines at 5461 Å were studied because of their use in current research. In addition, a narrow 5461 Å line was observed for comparison, as well as to survey the more stringent optical conditions for obtaining high resolution and accurate narrow line profiles.

* Absolute intensity is expressed as a blackbody temperature, T °K. This is the temperature a blackbody must have to produce the observed intensity, according to Planck's equation

$$I_{\lambda} = \frac{2hc^2}{\lambda^5 (e^{hc/\lambda kT} - 1)} \quad \text{ergs-sec}^{-1} \cdot (\text{unit solid angle})^{-1} \cdot \text{cm}^{-2} \cdot (\text{unit wavelength in cm})^{-1}$$

The wavelength interval taken as a measure of line width is the half-width, or width at half intensity.

OPTICS

Apparatus

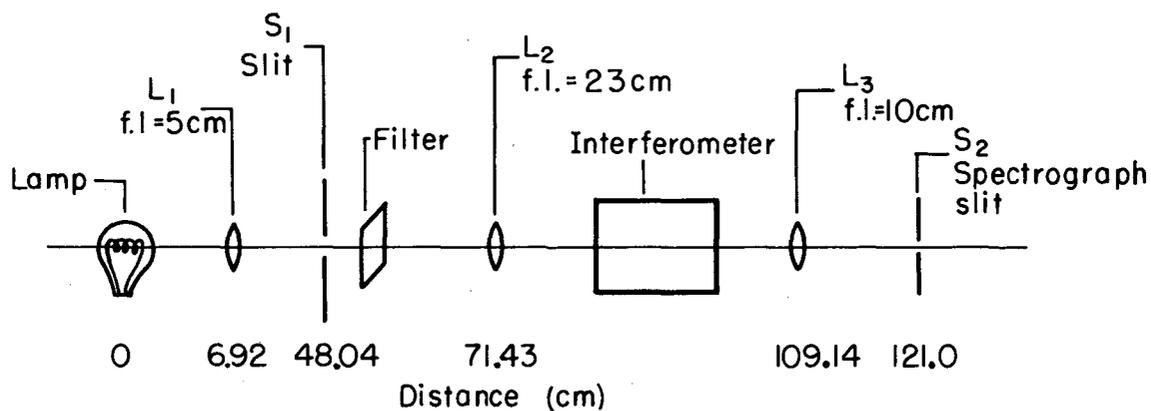
The optical arrangement used is shown in Fig. 1. A greatly enlarged image of the lamp is focused on slit S_1 . In this way that portion of the lamp which is being observed can be defined. In most of the lamps there was an obvious temperature gradient, but in all cases the geometric center of the arc or discharge was viewed. Lens L_2 collimates the light to the interferometer and L_3 focuses the interference pattern on the slit of the spectrograph. The Fabry-Perot etalon was designed by Professor Jenkins of the Physics Department. An interferometer plate area of maximum diameter 3.5 cm was exposed. The spectrograph was a Hilger (medium) glass prism instrument; it served the double purpose of separating the different atomic lines and providing a means of photographing the interference patterns. Kodak type III photographic plates were used as a compromise among several requirements; for relative intensity comparison between several similar lamps, high contrast; for correct line shape, low granularity. The filter (s) was used not only to isolate particular lines of interest but also to decrease the light intensity by a known amount so that exposures of several lamps of different intensities might be taken for the same fixed time (1 minute).

Alignment

Two of the extreme light rays are traced through the optics in Fig. 2. It is clear that because the rays pass within all the indicated optical stops, the limiting aperture is the collimating lens of the spectrograph (not shown). Thus, to properly fill the optics with light it was necessary only to observe from the spectrograph plate holder that the spectrograph optics were filled.

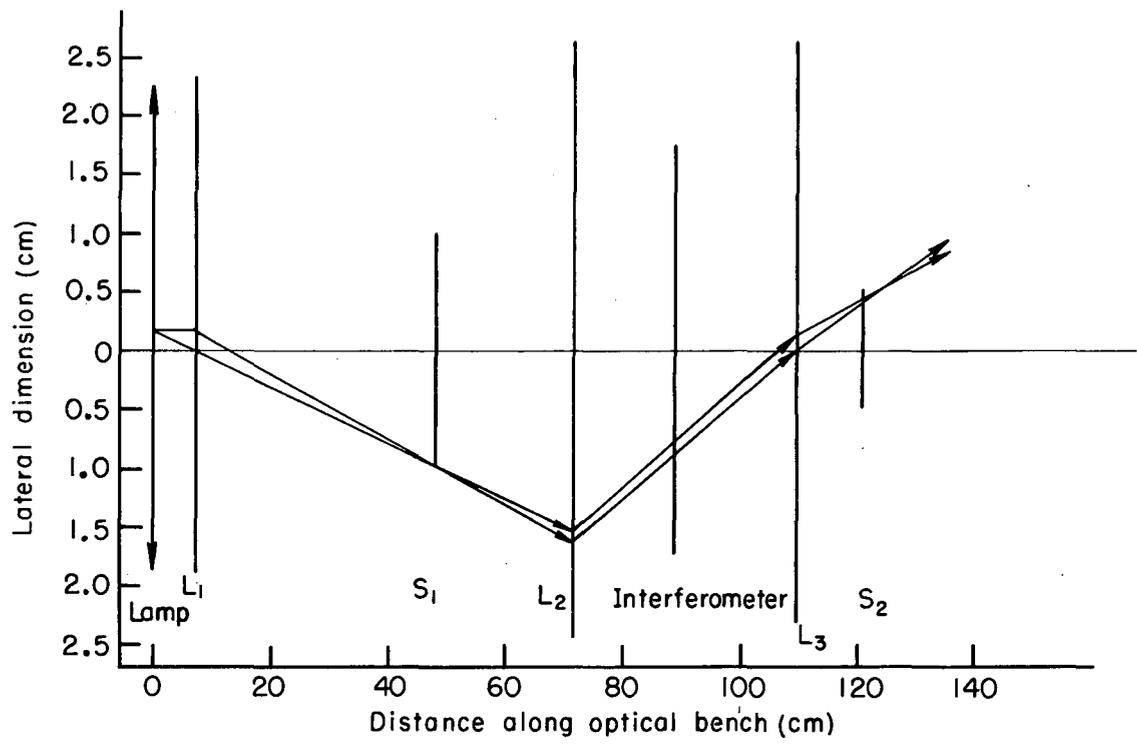
To align the interferometer plates to parallelism a less complicated optical system will do. Fig. 3 shows the schematic arrangement. The first lens collimates the light from the lamp. The final lens is slightly concave, giving a virtual image of the interference pattern.

Detailed observations of the interference fringes was made possible by replacing the final lens in Fig. 3 by a lens of large focal length. The magnified image could be observed in its focal plane with a viewing lens of short depth of focus. (These optics will be referred to as Fig. 3 modified.) In this way, with high magnification, all of the spectral hyperfine structure could be observed. Unfortunately, the optics of Fig. 1 did not offer high magnification and in some cases hyperfine lines which could be resolved by Fig. 3 modified optics overlapped



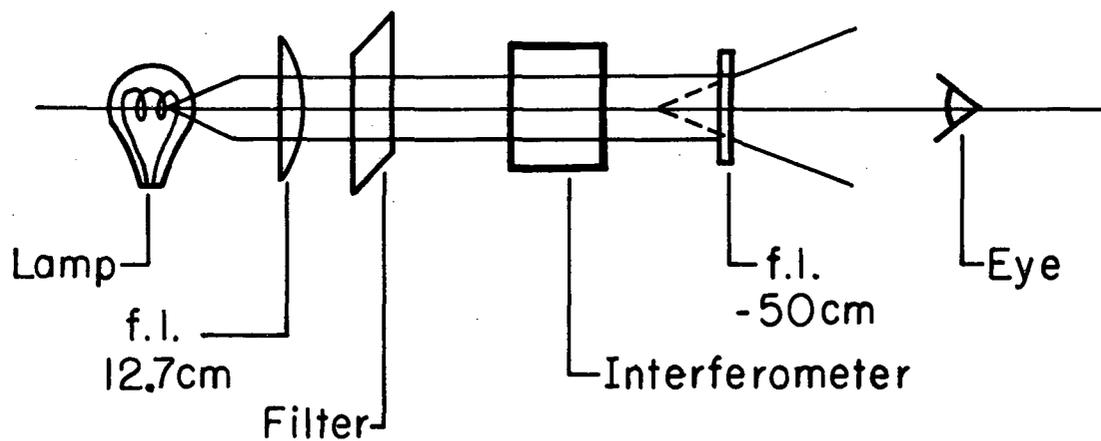
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Fig. 1. Optical arrangement for photographing interference fringes.



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Fig. 2. Extreme rays traced through optics of Fig. 1.



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Fig. 3. Arrangement for visual observation of interference fringes.

in the image plane of the spectrograph. This put a definite limitation on the resolving power of the system.

In the case of the complex hyperfine structure of the 5461 Å line only the most intense component was of interest and exposures were taken only long enough to record that component. However, it would have been difficult to photograph, for example, the complete structure of the Hg 5461 Å line because the long exposure required for the weak components would have so overexposed the strong one that it would have been broadened. Also, the resolution was so poor that several of the components would have overlapped.

Non-Ideal Interference

As the work progressed a rather interesting unexpected phenomenon became apparent. The intensity of the fringes of the narrow 5461 Å line of the mercury germicidal lamp fell off rapidly from the central fringe (Fig. 4a). This effect was studied in some detail and the significant evidence is summarized below.

- (a) On removing the Fabry-Perot etalon, the line spectrum could be photographed and showed no intensity variation.
- (b) Regardless of the position of the interferometer ring system with respect to the optical axis the central ring was always brightest.
- (c) The broad line of the mercury high pressure lamps did not show this intensity variation.
- (d) In the previous experiment multilayer dielectric-coated interferometer plates of very high reflectivity (probably > 99%), therefore of high theoretical resolution, were used. When these were replaced by silver coated plates of lower reflectivity, the intensity gradient decreased (Fig. 4b).

In an attempt to eliminate the intensity gradient the optical system of Fig. 5 was tried. Under these conditions the narrow line appeared as in Fig. 4c. The intensity gradient was completely eliminated. (However, the central fringe in this exposure has unusually low intensity. This result is presumably due to the fact that the spectrograph optics could not be filled for this geometry; the central interference ring was the bottom ring in the exposure and apparently was not getting enough light at that extreme point of the spectrograph plate holder.) Note that Fig. 5 is almost identical to Fig. 1 - the difference is that L_1 collects light over a smaller solid angle in Fig. 5 and the extra diaphragm selects an even narrower cone of light to pass through the interferometer.

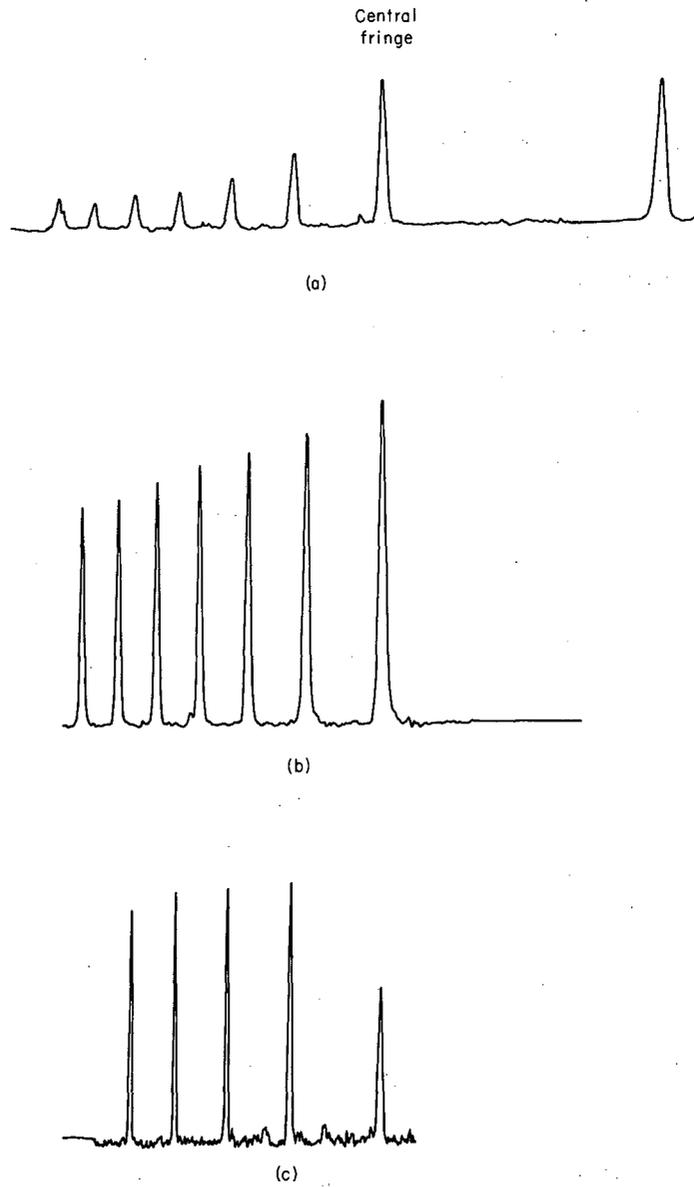
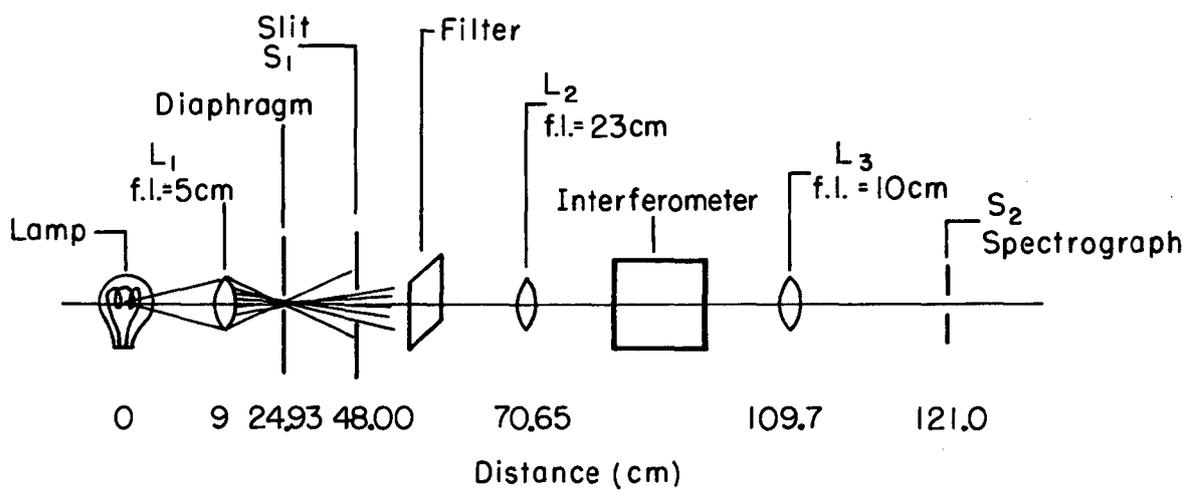


Fig. 4. Variation of appearance of interference fringes with different optics.
(a) Germicidal lamp line with optics of Fig. 1 and high reflectivity interferometer plates.
(b) Same, with plates of lower reflectivity.
(c) Same, with optics of Fig. 5.



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Fig. 5. Optical arrangement for eliminating fringe intensity gradient.

The following explanation is offered for this series of observations¹⁰. Consider an ideal high resolution interferometer profile (shown exaggerated in Fig. 6a). Suppose parts of the reflecting surfaces are slightly out of the reflecting plane (i.e. suppose plate imperfections or varying thickness of the metal film). This will produce subsidiary fringes somewhat displaced from the mean position (Fig. 6b) because they correspond to a different effective spacing between the plates. The central fringe is broadest and the ideal width of the fringes decreases regularly. A displacement is not significant at the central fringe, but components of other fringes may be shifted sufficiently to decrease the resultant peak intensity.

The same result will be obtained if flat plates are not aligned sufficiently parallel. (This adjustment was limited by the nature of the etalon and the dependence on the optics of Fig. 3 to check the adjustment by observing the interference rings.) Both effects will be observed most noticeably

- (a) in intrinsically narrow lines as opposed to broad lines,
- (b) where the reflectivity is high, allowing for high resolution.

The optics of Fig. 5 apparently reduced the usable area of the interferometer plates, minimizing the variation of effective spacer thickness and making the adjustment to parallelism less difficult. We conclude that the same result could have been obtained by stopping down the interferometer. The changed angular spread of the light cone to the interferometer should have made no difference since reflectivity is insensitive to small changes of incident angle near 90 degrees⁹.

Similar observations on the effect of surface and alignment imperfections have been made in some detail by Dufour and Pica¹⁴.

From the above discussion it is expected that as the observed fringe intensity decreases the calculated half-width should increase. Calculation of the interference pattern of Fig. 7 top shows that the half-width increases and peak intensity decreases in such a way that their product is a constant (Table III). Thus with suitable corrections all the calculated intensities can be equalized. However, the very existence of the gradient means that the measured peak intensity is a minimum and the measured half-width is a maximum.

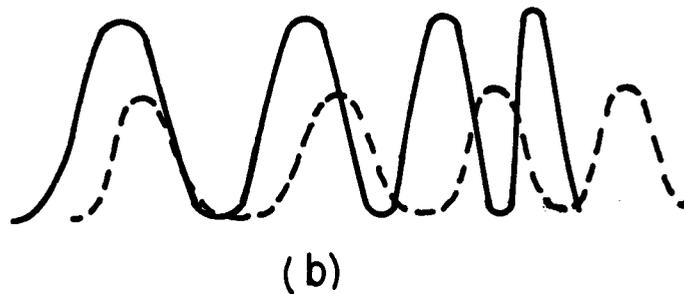
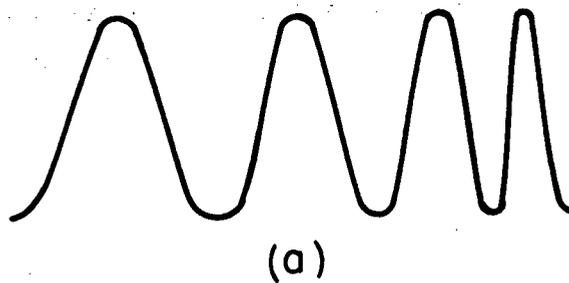
MERCURY LIGHT SOURCES

Line

Central component of 5461 Å ($7^3S_1 \longrightarrow 6^3P_2$)

Operating Conditions

- (a) General Electric germicidal (G18T6). Powered by G.E. 4 watt ballast transformer, Catalog #586848.



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Fig. 6. (a) Ideal fringe intensity distribution.
(b) Intensity distribution due to defects.

(b) G.E. H100A4 high pressure lamp, 0.85 amp D.C.

(c) Osram mercury lamp, 1.1 amp A.C. (rated current).

More details on lamp construction and operation are given by Worden⁸.

Complete Line Structure

Because of the large number of relatively abundant isotopes there are 14 hyperfine components in this line. The central group of 5 lines has a width of $.117 \text{ cm}^{-1}$ or $.035 \text{ \AA}$.

Interference Pattern

(a) Germicidal lamp

Inspection with optics of Fig. 3 modified showed 7 distinct lines in the hyperfine structure corresponding to the 7 groups in which the 14 hyperfine components fall. Only the most intense line (corresponding to the central group of 5 hyperfine components) was photographed.

(b) H100A4 and Osram

On warming up both lamps showed hyperfine structure which was gradually smeared out into a continuum. After several minutes both lamps showed strong self-reversal.

Conditions of Exposure and Development

The transmission of the interferometer plates is given in Table II. Reflectance was assumed to be related to transmission according to the results of Kuhn and Wilson¹³.

Plates	Line	Transmission (%) ¹²		Spacer	Average reflectance of 2 plates (%)
		#1	#2		
Set A (old surface)	Hg 5461 Å	1.00	2.35	1.77 mm	.96

To calibrate the photographic plate response to light intensity in order to find the half-intensity points for half-width determinations, and to determine relative intensities of different sources exposed on the same plate, a step-weakener exposure was taken on each plate before development.

Plates were developed in Kodak D-19 for 3 minutes under continuous brushing with a cotton wad. The tracings from a Leeds and Northrup Recording Microphotometer appear in Fig. 7.

Analysis of Data

The germicidal lamp shows the intensity gradient previously discussed. The large background intensity in the interference traces of the high pressure lamps is due to the strong continuum in the mercury spectrum. However, the intensity of successive peaks with respect to the continuum level is essentially constant. The AH⁴ lamp shows a completely self-reversed line; for the particular spacer used, successive side bands overlap.

The important parameters were obtained from measurements according to the methods of Meissner¹³. A least squares analysis of data for 5 fringes was carried out for the constant difference in the square of the fringe radii ΔR^2 and the fractional order of interference at the center, ϵ . Half-widths calculated for successive fringes of the broad, high pressure lines were identical. As previously mentioned, the product of half-width and peak intensity for the successive peaks of the germicidal lamp line was constant. This is shown in Table III. Because of the overlap of the AH⁴ side peaks, their true half-width is in doubt.

Table III

Variation of Intensity and Half-Width in the Interference Fringes of a Narrow Line (5461 Å)

Fringe	Measured Half-width in Å (X)	Relative Intensity (Y)	Product XY
0 (central fringe)	.052	23.0	1.20
1	.080	14.5	1.16
2	.103	11.5	1.18
3	.111	10.0	1.11

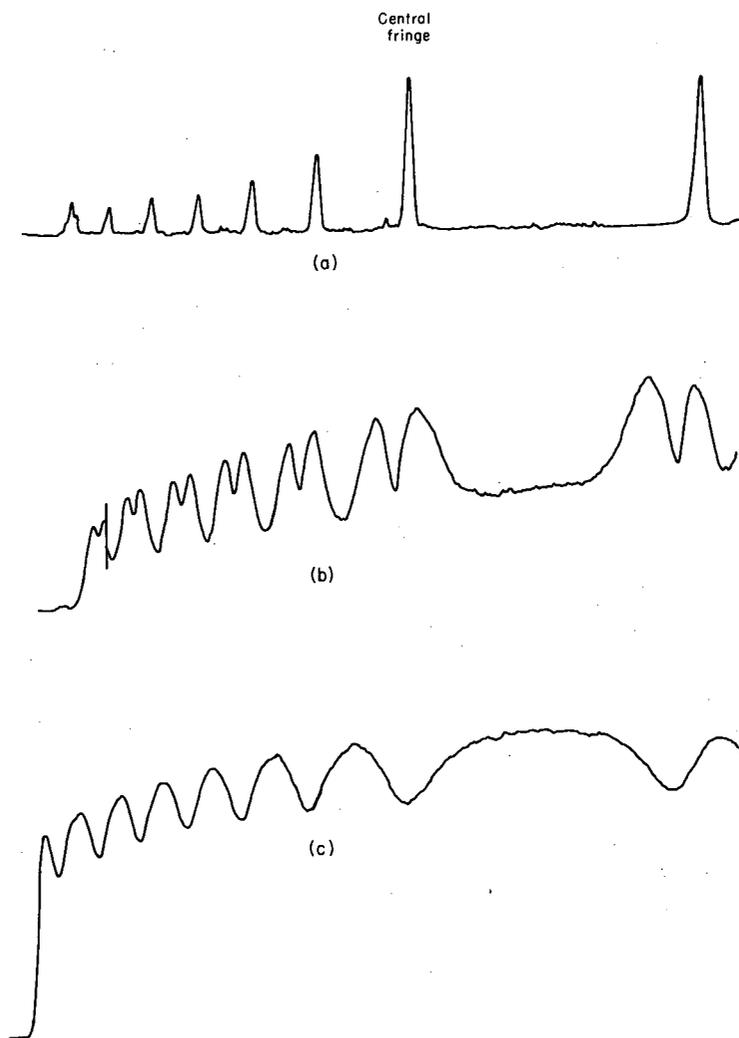


Fig. 7. Traces of 5461 Å line.
(a) G. E. germicidal lamp
(b) Osram lamp
(c) G. E. H100A4 lamp.

The measured half-width was corrected for instrumental broadening of the interferometer¹³, given by

$$A_{\lambda} = \frac{\lambda^2}{2\pi t} \frac{1-r}{\sqrt{r}}$$

The spacer thickness, t , and the reflectance, r , are given in Table II. It is also of some value to know the position of each spectral line relative to some fixed wavelength. The center of the Osram lamp self-reversal was taken to be the true position of the 5461 Å line. Displacements could be calculated¹³ from

$$\nu_a - \nu_b = \frac{\epsilon_a - \epsilon_b}{2t}$$

$$\lambda_a - \lambda_b = (\nu_b - \nu_a) \frac{\lambda}{\nu}$$

where ϵ is the fractional order of interference at the center of the interference ring system for a line of frequency ν . The data on the 5461 Å line shapes are given in Table IV. In taking an average value of half-width from measurements of several interference rings, most weight was given to the central ring because of higher resolving power there.

Table IV
Line Shapes and Intensities Hg 5461 Å

Lamp	Measured Half-width (Å)	Interferometer Instrumental Half-width Å	Corrected Half-width (Max. value Å)	Relative Position on Wavelength Scale (Å)	Relative Intensity at Peak
Osram					
center				0.00	
1st peak	.22	.011	.21	+ .16	59
2nd peak	.22	.011	.21	- .13	59
H100A4 (G.E.)					
reversed center				0.00	
peak appx.	.46	.011	appx. .45	+ .32	100
Germicidal (G.E.)	.050	.011	.039	- .009	1

Relative Intensities

Relative lamp intensities were determined in the following way. Exposures of the several lamps were for a constant time, 1 minute. To produce approximately uniform intensity of exposure, suitable Kodak Wratten Light Filters were used; filter transmission at the wavelength of interest was determined on a Cary Model 11 Recording Spectrophotometer. A small correction was made for the different final exposure intensities which showed up in the microphotometer tracing; for the high pressure lines, the continuum on the tracing was taken as the zero of intensity.

It is estimated that 86% of the center of the osram line was self-reversed while the G.E. AH4 was completely self-reversed. Because of side peak overlap in the latter case, the intensity for each peak was taken as half of the observed intensity.

CONCLUSION

Because of the strict optical requirements attending the proper use of the Fabry-Perot interferometer the results reported here, as already mentioned, suffer from a significant systematic error. This points to the importance of making more accurate measurement on commercial lamps not only for their own sake, but so that line shape and intensity "standards" may be readily available to the many research workers who will be making interferometric measurements in the near future.

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The value of high resolution spectroscopy in connection with our fluorescence studies was realized by Professor Leo Brewer who suggested this work and supported it with his comments and encouragement.

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