Laser multipass system with interior cell configuration

Jacek Borysow,[1,*] Alexander Kostinski,[1] and Manfred Fink[2]

[1]Department of Physics, Michigan Technological University, Houghton, Michigan 49931, USA

[2]Department of Physics, The University of Texas at Austin, Austin, Texas 78712, USA

*Corresponding author: jborysow@mtu.edu

We ask whether it is possible to restore a multipass system alignment after a gas cell is inserted in the central region. Indeed, it is possible, and we report on a remarkably simple rearrangement of a laser multipass system, composed of two spherical mirrors and a gas cell with flat windows in the middle. For example, for a window of thickness d and refractive index of n, adjusting the mirror separation by $\approx 2d(1 - \frac{1}{n})$ is sufficient to preserve the laser beam alignment and tracing. This expression is in agreement with ray-tracing computations and our laboratory experiment. Insofar as our solution corrects for spherical aberrations, it may also find applications in microscopy.   © 2011 Optical Society of America

OCIS codes: 300.6450, 170.5660, 120.0120, 170.0170, 300.0300.

1. Introduction

Multipass cells are used widely in spectroscopy as well as in industrial and medical settings [1] and are available commercially. Multipass cells are particularly important when native signals are weak, as in Raman spectrospcopy, in dealing with absorption measurements of low concentration or weakly absorbing species. The signal-to-noise ratio in absorption experiments increases with the path length. The most frequently used configurations are the White cell [2,3] and the Herriot cell [4]. While the former typically uses spherical mirrors, the latter (also known as an off-axis resonator) has numerous variants [4–7] with either spherical mirrors or astigmatic mirrors. New multipass cell designs for absorption spectroscopy are still being reported in the literature, e.g., based on cylindrical mirrors, with a path length of more than 10 m, despite a small volume [8].

In addition to the difficulty of very small cross sections, in Raman spectroscopy, the experimenter must collect the scattered light and analyze its spectral components. The process involves imaging of a small scattering volume onto the entrance slit of a grating spectrograph of a hundred micrometers wide and a few millimeters long. Therefore, the essential requirement for multipass configuration is that the excitation light source (laser) beam traverses the same small scattering region repeatedly. This is why the relatively large multipass cells developed for absorption measurements cannot be used in Raman spectroscopy without major modification.

An early multipass Raman tube was reported by Weber and co-workers [9] where laser light traveled multiple times between flat mirrors and the Raman signal was collected by concave mirrors. The first design of a multipass cell, capable of producing a very high flux of laser light at a small focal region, was demonstrated by Hartley and Hill [10]. Their light trapping system used an ellipsoidal mirror and a flat mirror positioned in such an arrangement that the laser light bouncing between the two mirrors eventually collapsed on the major axis of the ellipsoidal mirror. However, this system required custom manufacturing of the ellipsoidal mirror and was very difficult to align. The Raman multipass cell under consideration in this paper was first described by Kiefer *et al.* [11] and later extensively used by the authors of [12–15]. It is relatively simple to align, can be built from off-the-shelf components, and offers very large signal gains (up to 50) in comparison to a single-pass cell configuration.

2. Multipass Cell Alignment: Interior Cell Configuration

In the course of our work on Raman spectroscopy of diluted gases (number densities below 10^{14} cm^{-3}), we have used a particular multipass cell configuration [14,15]. The major components of the Raman multipass cell are (i) a pair of 50.2 mm diameter concave mirrors made out of BK7 glass with a 100.0 mm radius of curvature, separated by a distance of about 200 mm. The nominal reflectivity of the mirrors at normal incidence is better that 99.99% and (ii) a small cylindrical vessel (5 cm long and 3.5 cm in diameter) containing gas samples placed between the mirrors. The gas vessel has 3 mm thick windows made out of BK7 glass mounted on each side. The windows have antireflection coating to minimize losses. The alignment of the multipass cell without the sample vessel inside is straightforward and suitable to a laboratory environment as the entire system is in the open air and an experimenter can readily trace the laser beam's path.

However, once a sample cell is inserted in the central region, the alignment is ruined. The disturbing influence of the glass vessel, with two windows of width d, is illustrated by the comparison of Figs. 1(a) and 1(b). Figure 1(a) shows the laser beam traveling between two spherical mirrors crossing each time at one of the two very tight spots in the middle. Figure 1(b) shows trajectories of the laser beam traveling between the mirrors in the presence of the interior gas cell with 3 mm thick windows. The computations were done using the optical design program ZEMAX [16]. The windows displace the original trajectories so that the reflected laser beams no longer cross the same spot inside the gas vessel, defeating the purpose of the multipass cell. The scattering volume created by these laser beams in the multipass system is greatly increased by inserting the glass cell, as is apparent in Fig. 1(b). This effect is akin to spherical aberrations, which are so ubiquitous in microscopy.

Is it possible to correct for the spherical aberrations so that the alignment of the multipass system is restored? Specifically, in the presence of the cell inside, one still needs to maintain the condition that every beam crosses the same point upon consecutive reflections from one of the spherical mirrors. Hence the question arises: is there a simple or at least a practical way to compensate for the distortion, caused by the windows, thereby rendering Raman spectroscopy feasible? The question is quite general and is likely to arise in many situations, e.g., microscopy. At first sight, the answer is negative as the optical path differences vary from ray to ray. Furthermore, defocusing has enlarged the laser beam.

3. Simple Solution

It is important to note that the geometric effect of the incident ray passage through the window is a phase delay, yielding a vertical shift (normal to the propagation direction of the laser). From a purely
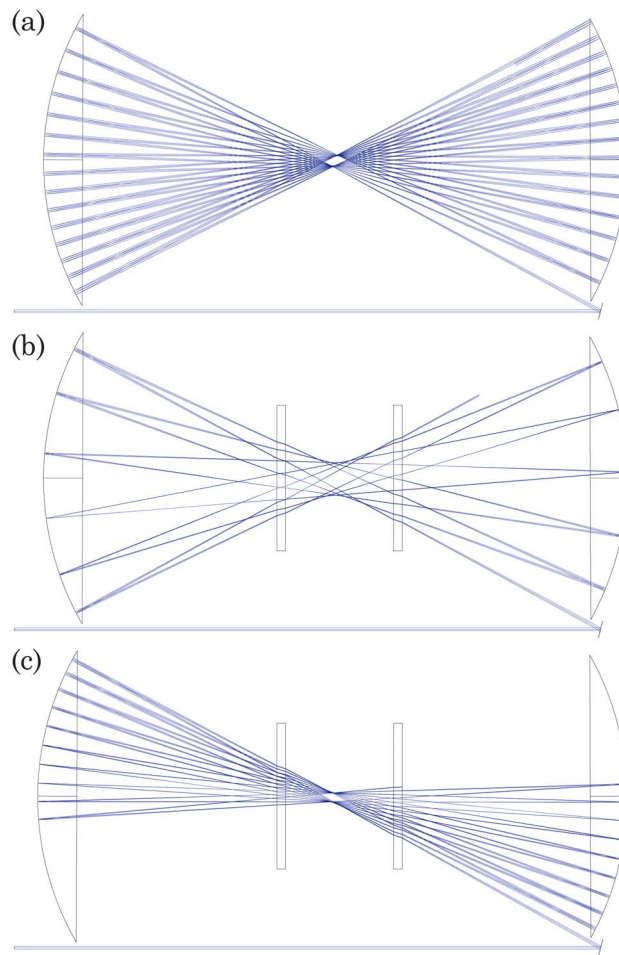

Fig. 1. (Color online) Multipass system built from a pair of 50.2 mm diameter concave mirrors with a 100.0 mm radius of curvature, separated by a distance of about 200 mm. (a) Consecutive laser beams inside a multipass system without interior disturbance. (b) The same, but with a gas vessel placed between the spherical mirrors. The windows in the gas sample are made out of BK7 glass of index of refraction equal to 1.5 and thickness 3 mm. (c) Laser beam trajectories for multipass cell with a 3 mm thick glass window cell inside: realigned as given by Eq. (3).

geometric point of view, this is akin to the Goos–Hänchen shift, e.g., see Jackson [17], Fig. 7.7, p. 308.

Figure 2(a) shows the laser beam displacement caused by the gas sample window of thickness d. We assume here for simplicity that the initial laser beam traveled along the radius of the left spherical mirror in the multipass cell. The displaced beam arrives at the mirror below the normal and is reflected at the angle equal to the angle of incidence, not crossing the incoming beam. The original beam traveling along the mirror radius would have been reflected onto itself without any deviation. The beam is displaced by

$$\Delta y = \frac{d}{\cos(\beta)} \sin(\alpha - \beta), \qquad (1)$$

where d is a thickness of the window, α is an angle of incidence of the laser beam, β is an angle of the
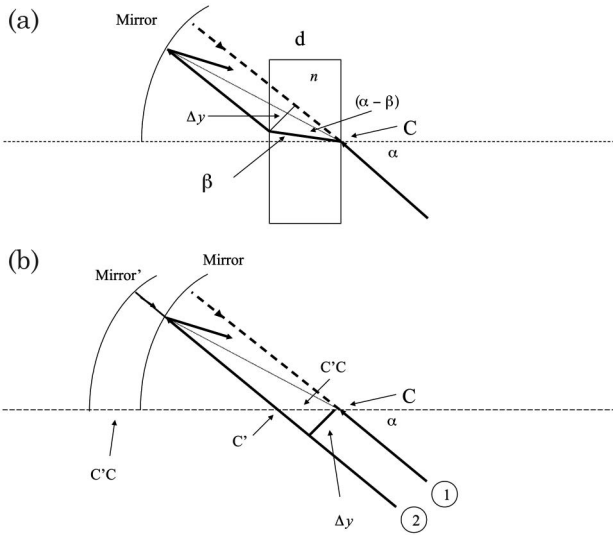
Fig. 2. (a) Displacement of the laser beam passing through a glass windows of thickness d. The net effect of the double refraction at the window walls is a vertical shift $\Delta y = \frac{2d}{\cos(\beta)} \sin(\alpha - \beta)$. (b) Restoring alignment of the multipass system with the glass window inside: (1) original laser beam and (2) displaced laser beam. Mirror, original position of the spherical mirror; Mirror', corrected position of the spherical mirror. Mirror moved horizontally by $CC' \approx 2d(1 - \frac{1}{n})$.

refracted beam computed from Snell's law, and n is the index of refraction of the BK7 window at 780 nm.

Can a horizontal shift of the mirror restore the alignment? Indeed, this is the case as shown in Fig. 1(c). The ray-tracing drawing in Fig. 1(c) is the result of ZEMAX [16] calculations and was obtained by increasing the separation between the spherical mirrors until the laser beams traveling inside the cell crossed the same spots again.

Upon closer examination of the laser ray in Fig. 2, we realized that the left mirror of the multipass system must be moved along the optical axis until the laser beam, displaced by the cell window, aligns along the radius of the left mirror. Therefore, the mirror must move to the left by the distance CC':

$$CC' = \frac{\Delta y}{\sin(\alpha)}. \tag{2}$$

Substituting for Δy from Eq. (1) in Eq. (2) yields

$$CC' = 2\frac{d}{\sin(\alpha)\cos(\beta)} \sin(\alpha - \beta) = 2d\left(1 - \frac{\tan(\beta)}{\tan(\alpha)}\right), \tag{3}$$

where the factor of 2 accounts for both windows in the path of the laser beam. After this correction, the laser beam arrives at the mirror at normal incidence and is reflected on itself (Fig. 2(b)) and later displaced by the windows in the gas cell just so that it arrives undisturbed at the right mirror of the multipass cell. For small angles α, Eq. (3) can be written as
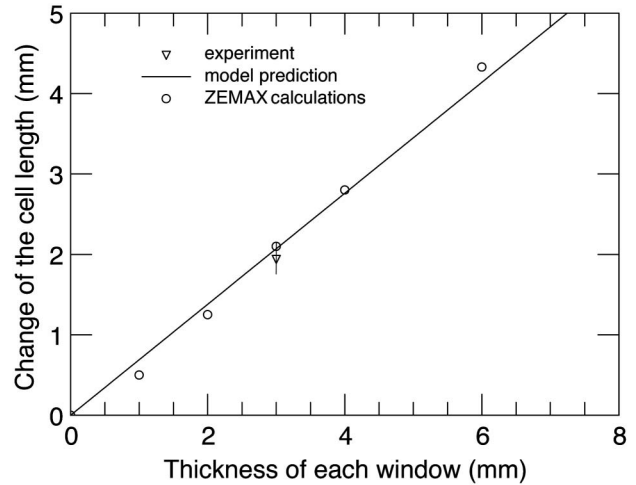

Fig. 3. Horizontal shift of the spherical mirror, required for alignment of the multipass system as a function of the thickness of the windows of the gas cell: (1) simple approximate analytic expression (solid line), (2) ZEMAX simulations (circles), and (3) laboratory experiment (triangle). The vertical line is an estimate of our accuracy to judge optimal alignment.

$$CC' = 2d\left(1 - \frac{\tan(\beta)}{\tan(\alpha)}\right) \approx 2d\left(1 - \frac{\sin(\beta)}{\sin(\alpha)}\right)$$
$$\approx 2d\left(1 - \frac{1}{n}\right), \tag{4}$$

where n is the index of refraction of the gas sample window.

We have tested the proposed alignment procedure for inside cell windows as thick as 6 mm, by calculating the required shift versus the window thickness d, using the optical design code ZEMAX [16]. Additionally, in the laboratory we measured the required shift of the multipass configuration by placing two 3 mm thick windows inside it. An excellent agreement between experiment, simulations, and model predication computed from Eq. (4) can be seen in Fig. 3. The solid line in Fig. 3 is the result of the model calculations. The circles are the results of the simulation done using ZEMAX [16] software, and the triangle is the outcome of the laboratory experiment.

4. Concluding Remarks

We posed a question of realignment of the multipass system in the interior cell configuration. Here we presented a surprisingly simple solution to this seemingly complex problem. We verified our simple expression for the required correction for the interior of the glass vessel with flat windows by numerical simulations with ZEMAX and by a laboratory measurement. Our solution essentially corrects for spherical aberrations and therefore can have applications in microscopy as well.

References

1. W. Demtröder, *Laser Spectroscopy: Basic Concepts and Instrumentation*, 3rd ed. (Springer-Verlag, 2003).
2. J. U. White, "Long optical paths of large aperture," J. Opt. Soc. Am. **32**, 285 (1942).
3. J. U. White, "Very long optical paths in air," J. Opt. Soc. Am. **66**, 411–416 (1976).
4. D. Herriott, H. Kogelnik, and R. Kompfner, "Off-axis paths in spherical mirror interferometers," Appl. Opt. **3**, 523–526 (1964).
5. J. P. McManus, P. L. Kebabian, and M. S. Zahniser, "Astigmatic mirror multipass absorption cells for long-path-length spectroscopy," Appl. Opt. **34**, 3336–3348 (1995).
6. Ch. Dyroff, A. Zahn, W. Freude, B. Janker, and P. Werle, "Multipass cell design for Stark-modulation spectroscopy," Appl. Opt. **46**, 4000–4007 (2007).
7. G. Durry, Th. Danguy, and I. Pouchet, "Open multipass absorption cell for *in situ* monitoring of stratospheric trace gas with telecommunication laser diode," Appl. Opt. **41**, 424–433 (2002).
8. D. Das and A. C. Wilson, "Very long optical path-length from a compact multi-pass cell," Appl. Phys. B **103**, 749–754 (2010).
9. A. Weber, S. P. S. Porto, L. E. Cheesman, and J. J. Barrett, "High-resolution Raman spectroscopy of gases with cw-laser excitation," J. Opt. Soc. Am. **57**, 19–28 (1967).
10. D. L. Hartley and R. A. Hill, "A highly efficient light-trapping cell for Raman-scattering measurements," J. Appl. Phys. **43**, 4134–4136 (1972).
11. W. Kiefer, H. J. Bernstein, H. Wieser, and M. Danyluk, "The vapor-phase Raman spectra and the ring-puckering vibration of some deuterated analogs of trimethylene oxide," J. Molec. Spectrosc. **43**, 393–400 (1972).
12. J. Sabbaghzadeh, W. Buell, J. Holder, and M. Fink, "A very narrow high throughput Rayleigh filter for Raman spectroscopy," Appl. Phys. B **60**, S261–S265, (1995).
13. R. Claps, J. Sabbaghzadeh, and M. Fink, "Raman spectroscopy with a single-frequency, high-power, broad-area laser diode," Appl. Spectrosc. **53**, 491–496 (1999).
14. J. Borysow and M. Fink, "Doppler width limited near-infrared Raman spectrometer," Appl. Spectrosc. **60**, 54–56 (2006).
15. J. Borysow and M. Fink, "NIR Raman spectrometer for monitoring protonation reactions in gaseous hydrogen," J. Nucl. Mater. **341**, 224–230 (2005).
16. Focus Software, Inc., ZEMAX optical design program, version 8 (1999).
17. J. Jackson, *Classical Electrodynamics*, 3rd ed. (Wiley, 1997).