MULTIPASS MATRIX-ARRAY SYSTEMS — PROMISING LONG-PATH-LENGTH SYSTEMS FOR HIGH-RESOLUTION SPECTROSCOPY

S.M. Chernin and E.G. Barskaya

N.N. Semenov Institute of Chemical Physics of the Academy of Sciences of the USSR, Moscow
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High-transmission three- and four-objective multipass matrix-array systems, which provide an extremely long optical path in absorption, have been developed for application in high-resolution spectroscopy and are currently used in practice. In these systems images are formed on field mirrors in the form of compact rectangular matrix arrays and the number of whose rows and columns is adjustable. The number of passes for mirrors with highly reflecting coatings reaches 500. Distinguished by their structural simplicity, matrix-array systems have high optical quality and performance characteristics. They can also be used as delay lines for optical signals in laser technology. Based on their possibilities, matrix-array systems represent a new generation of multipass systems.

In the last few years interest in multiple-reflection mirror systems has increased significantly. This has happened for a number of reasons, one of the most important being the acute problem of preserving the environment. High-precision instruments for gas analysis require multipass systems, which are the basic part of universal and sensitive optical gas analyzers. To determine microconcentrations of pollutants in the atmosphere it is best to use multipass systems on long paths combined with tunable lasers.

Multiple-reflection optical systems are of special interest for high- and ultrahigh-resolution IR spectroscopy. The long optical absorption path obtained in multipass cells makes it possible to study gases at low pressures. Under these conditions gases satisfy well the Bouguer-Lambert law. It is the ideal case when the recorded shape of spectral lines is not distorted by collisional processes and intermolecular interactions. In addition it is possible to achieve extremely high spectral resolution and to interpret the fine structure of the molecules. In the opposite situation the high resolution of the instrument is of no use, since it does not correspond to the conditions of the experiment.

Yet, in spite of the fact that spectral instruments are constantly being improved fro» year to year, long-path-length cells, which are still based on White's multipass system (1942),[1] remain practically unchanged. Many research spectroscopists have attempted to adapt this well-known mirror system for solving modern problems. This has not always been successful.

The rapid development of laser technology over the last few years has led to the development of a new spectroscopy — high-resolution laser spectroscopy. For the new spectroscopy it has been found that long-path-length cells based on White's classical system[1] are obsolete owing to the ^possibility of realizing a very large number of passes as well as owing to a number of optical and operational drawbacks. It is now necessary to reexamine and improve the most important optical parameters of the classical multipass system,[1] employed in spectroscopy as a universal system, and to develop a series of' mirror systems for different problems arising in modern science.

We have developed a series of fundamentally new multiple-reflection optical systems, of which the multipass matrix-array systems (MMSs) have been found to be most promising for high-resolution spectroscopy.[2,3] Though these systems have relatively simple constructions they lake it possible to realize in a small volume an extremely large number of passes of light owing to the fact that intermediate images are created by a compact rectangular matrix array. They are widely used in our country and have been recognized abroad.[4–8] The possibility of using MMSs together with large-angular-aperture sources makes them irreplaceable when combined with high-resolution laser diode spectrometers.

The well-known multipass mirror systems with very long path length, such as White's system (1976)[9] and Hanst's system,[10] have many operational drawbacks and they are too cumbersome. Thus, for example, in order to increase the number of rows in the image additional objectives and collecting elements must be inserted each time. To realize 220 passes this system would have to contain, on the whole, 20 mirrors. Doubling the number of rows, which Horn and Pimental[11] did by adding a flat corner reflector to White's system,[1] was found to be inadequate for obtaining a very large number of reflections, and additional losses of light occur. The variants of Herriot's multipass system,[12] which are widely used abroad in laser spectroscopy as optical delay lines, are Intended only for sources with a small angular aperture and they have a very complicated image pattern, practically precluding readjustment during an experiment. The system of Schulz-DuBois,[13] also intended for use as an optical delay line, does not have an exit point because it is closed in the form of an optical cavity, though it does contain the optimal number of mirrors. Moreover, for vertical separation of images a refracting corner reflector having a complicated configuration and a focusing lens is inserted into the mirror system.

We were charged to solve the problem of developing a highly stable multipass system, in which the number of reflections is very large and the images are arranged compactly in a rectangular matrix array and in which the number of mirror elements is limited. We first had to solve a more specific problem — to transfer, using new reflecting elements, images to different vertical positions on a collecting element in order to construct successive rows of a matrix array.

We borrowed from Chernin's earlier multiple-reflection optical system,[14] in which a narrow collecting element was placed next to a large concave mirror, the idea of using a concave mirror for successive transfer of images. Similarly we placed a narrow auxiliary collecting element next to the large main collecting element. This auxiliary element will direct each time rays from the last image, formed in a double row, onto an additional objective, which has not yet participated in the construction of these images. In addition, in contrast to a flat corner reflector the concave mirror, collecting element will not cause losses of light due to vignetting when light sources with a large angular aperture are used. Next, we had to find a fundamentally new combination of objectives, whose arrangement and interaction and the manner in which they are anchored would make it possible to construct successive rows of images on field mirrors and would ensure that these images are stable. In designing the system of objectives we used the same efficient connection for mirror pairs as that used in Barskaya's system;[15] this ensured that the images would be stable.

Based on these two conditions, adopted beforehand, and all of the best features of White's classical system[1] we started to develop variants of the matrix-array system. This is how the three-objective and, somewhat later, the four-objective multipass matrix-array systems, each of which is of value in itself, arose. The multipass systems developed are essentially a combination of several mismatched but intercoupled confocal spherical cavities in which the emergent radiation is placed in a particular position.

THREE-OBJECTIVE MATRIX-ARRAY SYSTEM

Figure 1 shows a schematic diagram of the spatial arrangement of the mirrors in the system and the manner in which they move and are anchored. The broken circles depict the arrangement of the images on the surface of the collecting element. The numbers on the images show the order in which they are formed when the system is adjusted for 30 passes.

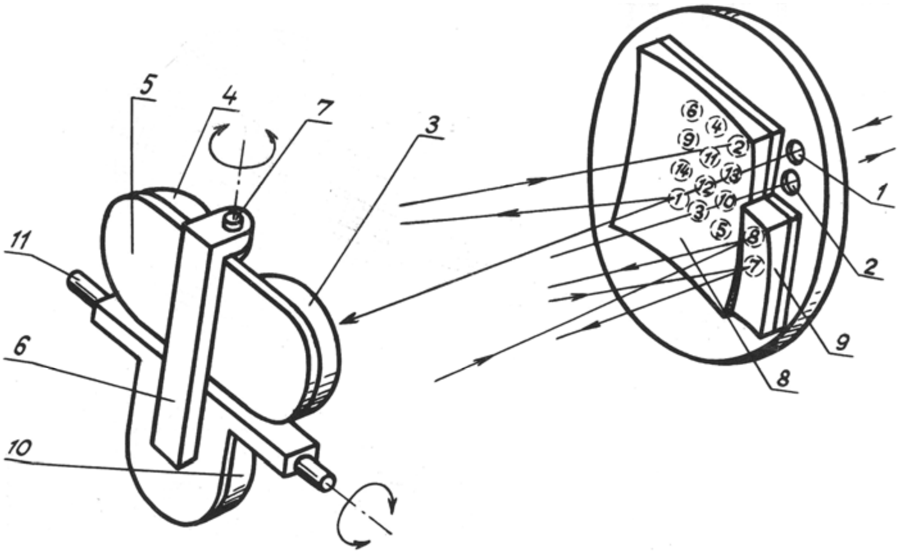

Fig. 1. Three-objective multipass matrix-array system

The connected objectives *3* and *4*, anchored securely to the plate *5* of the sectional mobile holder *6*, were placed opposite the entry and exit openings *1* and *2* at a distance equal to the radius of curvature of the mirrors. The reflecting objectives *3* and *4* are connected with one another at a fixed angle, whose value is chosen starting from the fact that the distance between, the centers of curvature of these objectives corresponds to one-half the horizontal step of the images. The plate *5*, together with the objectives *3* and *4*, can rotate around the vertical axis 7. A collecting mirror *8* is placed opposite the pair of objectives *3* and *4* next to the entry and exit openings *1* and *2*. The center of curvature of the collective mirror *8* lies at the center of symmetry of the objectives *3–4*, so that it reflects the connected objectives *3* and *4* onto one another. An auxiliary collecting mirror *9*, in the form of a narrow strip, adjoins the collecting mirror *8* below the entry and exit openings *1* and *2*. It is oriented so as to reflect the objective *3* on an additional objective *10*, placed on the holder *6* near the connected objectives *3* and *4*. The mobile holder can rotate around the horizontal axis *11* together with the reflecting objectives *3*, *4*, and *10*. The axes *7* and *11* are mutually perpendicular.

The system operates as follows. Radiation from the wide-aperture or laser source passes through the entry opening 1 and strikes the reflecting objective *3*, which creates an intermediate image of the entry opening on the surface of the collecting mirror *8*. The distance from the entry opening to its first image along the horizontal axis depends on the rotation of the plate *5* with the objectives *3* and *4*. After the first image is formed the radiation is reflected from the collecting mirror 8 onto the objective *4*, which forms a second image next to the entry opening 1 on the very edge of the collecting mirror *8*. The connected reflecting objectives *3* and *4* form successive images, shifted by precise distance, of the entry opening on the collecting mirror until two rows of images are formed. The last image falls on the auxiliary collecting mirror *9*; the radiation reflected from it passes to the additional reflecting objective 10, which rereflects it with a vertical shift to the collecting mirror *9*. The radiation is then directed onto the objective *3* as if from a new entry opening. The next pair of rows of images is formed and the process is repeated until the last image reaches the exit opening *2*.

The number of rows in the matrix array of images is controlled by the inclination of the entire holder 6 with three objectives *3*, *4*, and *10* around the horizontal axis *11*. The number of rows is directly proportional to the angle of inclination of the holder *6*. The number of images in each row, i.e., the number of columns in the matrix array, is changed by rotating the plate *5* with the objectives *3* and 4 — anchored securely relative to one another — around the vertical axis *7*. The number of columns is directly proportional to the angle of rotation of these mirrors in the direction away from the openings *1* and *2*.

When the connected objectives *3* and *4* rotate together their centers of cruvature slide along the surface of the collecting mirror *8*, the distance between them remaining constant. When the holder 6 is tilted they are displaced vertically, but the vertical distance between the line of the centers of curvature of the connected objectives *3–4* and the center of the additional objective 10 remains constant. The last image falls onto the exit opening *2* of the system not for any arbitrary rotation of the plate *5* and the holder *6* but only in positions corresponding to fixed displacements of the first image equal to multiples of the indicated intercenter distances.

The adjustable number of passes in the system forms a series of numbers, corresponding to definite ratios depending on the number of rows and columns of the matrix array of images. The number of passes N can be determined from the relation

$$N = 2(mn - 1), \tag{1}$$

where m = 2, 4, 6, 8, ... is an even positive integer equal to the number of rows and n = 1, 2, 3, ... is a positive integer equal to the number of columns.

The three-objective matrix-array system makes it possible to increase significantly the optical path length by increasing the number of rows without increasing the width of the collecting mirror. The system produces stable images. The compact arrangement of images in the matrix array reduces the aberrations of the field of view. The ability to control rotation in mutually perpendicular planes with a constant angular step makes possible more accurate and reliable adjustment of the number of passes. The system is especially suitable for use together with high-transmission, high-resolution, laser IR spectrometers when mounted in a tubular cell.

FOUR-OBJECTIVE MATRIX-ARRAY SYSTEM

Figures 2*a* and *b* show the arrangement of the reflecting elements in a multipass matrix-array system. They also show how the objectives are mounted and how they move. The collecting mirrors and the objectives are shown from the working side of the mirrors. In Fig. 2*a* *10* is the main collecting mirror and *11* is the auxiliary collecting mirror. The numbers in the broken circles depict the order in which the images are formed when the system is adjusted for 90 passes. Figure 2*b* shows a variant of the arrangement of the reflecting objectives *3*, *4*, *5*, and *6* on the circular holder *7*; this variant is used when the multipass system is mounted in a tubular cell. The primed numbers (Figs. 2*a* and *b*) indicate the centers of curvature of the corresponding mirrors. The connected objectives *3–4* and *5–6*, anchored securely on the mobile holder *7* in a single unit, are positioned opposite the entry and exit openings *1* and *2* at a distance equal to the radius of curvature of the mirrors. The holder 7, together with the objectives *3*, *4*, *5*, and *6*, can rotate around the vertical axis *8* and the horizontal axis *9*. The collecting mirror *10* placed opposite the objectives is located directly adjacent to the entry and exit openings *1* and *2*. The center of curvature of the collecting mirror *10* (the dot *10′*) lies at the center of symmetry of the pairs of objectives *3–4* and *5–6*. For this reason the collecting mirror *10* forms an image of both the objectives *3–4* and *5–6* on one another. An auxiliary collecting mirror *11* in the form of a narrow strip adjoins the collective *10* below the entry and exit openings *1* and *2*. It is oriented so as to image the objectives *3* and *5*, belonging to different pairs, one one another. Its center of curvature (the point *11′*) lies at the center of symmetry of the objectives *3–5*.

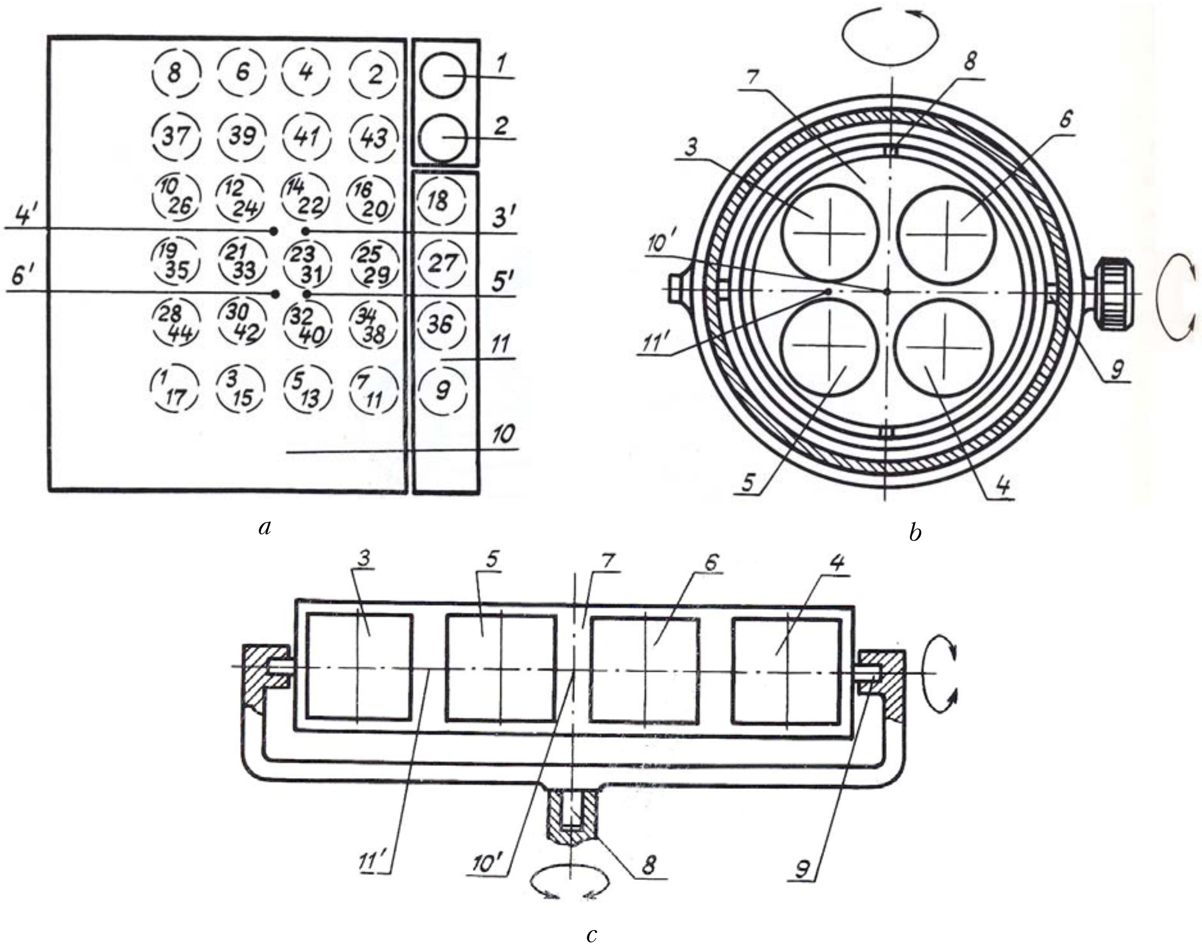

FIG. 2. The multipass four-objective matrix-array system: a) collecting mirrors; b) collecting mirrors on a circular holder; c) row variant of the arrangement of the objectives.

The system operates as follows. Radiation from the wide-aperture or laser source passes through the entry opening *1* and strikes the reflecting objective *3*, which forms an Intermediate image of the entry opening on the surface of the collecting mirror *10*. The distance from the entry opening to its first image along the horizontal axis depends on the rotation of the holder *7* — the set of objectives — and the magnitude of this vertically downward displacement depends on the inclination of the holder. After the first image is formed the radiation is reflected from the collecting mirror *10* onto the objective *4*, which forms a second image next to the entry opening 1 on the very edge of the collecting mirror *10*. The connected reflecting objectives *3* and *4* from successive images, displaced by a precise distance, of the entry opening until two rows of images are constructed and the last image reaches the collecting mirror *11*. The radiation reflected from it is directed to the objective *5* by the next pair. Next the reflecting objectives *5* and *6* successively focus images, forming the next two rows; but the last image does not again fall on the collecting mirror *11*. The connected objectives *3* and *4* once again start to operate. This continues until the last image falls on the exit opening *2* of the system.

When the holder *7* is rotated the centers of curvature of the securely connected objectives *3*–*4* and *5*–*6* (the points *3*′, *4*′ and *5*′, *6*′) slide along the surface of the collecting mirror *10*, the distance between them remaining constant. When the holder *7* is tilted these centers are also shifted vertically, their relative position remaining constant. The number of rows in the matrix is controlled by tilting the entire holder *7* around the horizontal axis *9*. The number of columns in the matrix array is changed by rotating the holder *7* around the vertical axis *8*. The number of columns is directly proportional to the angle of rotation of the holder in the direction away from the openings *1* and *2*. The last image falls onto the exit opening *2* not for arbitrary angles of rotation and inclination of the holder *7*, but only in positions corresponding to fixed displacements of the first image equal to multiples of the indicated intercenter distances.

Since the centers of curvature of the objectives in different pairs are separated vertically by a definite distance a double superposition of images is created in the matrix arrays in a manner so as to preserve the principle of superposition.

The number of passes N in the multipass matrix-array system can be calculated from the relation

$$N = (m - 1)(4n - 2), \quad (2)$$

where m = 2, 4, 6, 8, ... is an even positive integer equal to the number of rows and n = 1, 2, 3, ... is a positive integer equal to the number of columns.

Double superposition of the images occurs in the matrix array for $m \geq 4$ and $n \geq 2$.

In addition to having the best qualities of the three-objective system the four-objective system also has the following advantages. The number of passes can be adjusted more accurately and more reliably because the adjustment mechanism has been simplified. In spite of the extremely large number of passes the images formed in the four-objective system are exceptionally stable. When the system is perturbed (jolts, shocks, vibrations) the positions of the even images are completely self-compensated, so that errors in their position do not accumulate. In addition, all even and odd images in the matrix array repeat, with corresponding accuracy, the position of the entry opening and it first image with any number of passes. The optical path length is almost two times longer with collecting mirrors of the same size. In the four-objective matrix-array system the energy load on all objectives is uniform; this is important for many practical problems. The compact arrangement of the images in the matrix array and their double superposition create an additional effect of the system — aberrations of the field of view are significantly reduced. For laser sources with small angular divergence the system can be regarded as being virtually free of aberrations.

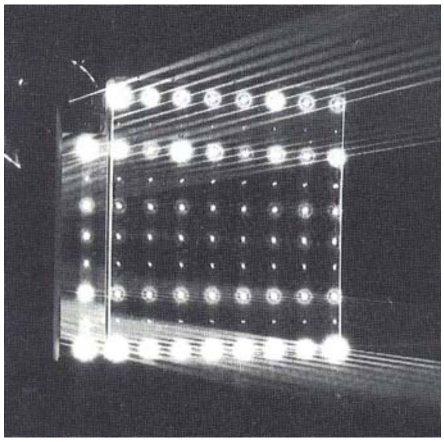

FIG. 3. Photograph of the matrix array of images on collecting mirrors after 306 passes of He-Ne laser radiation.

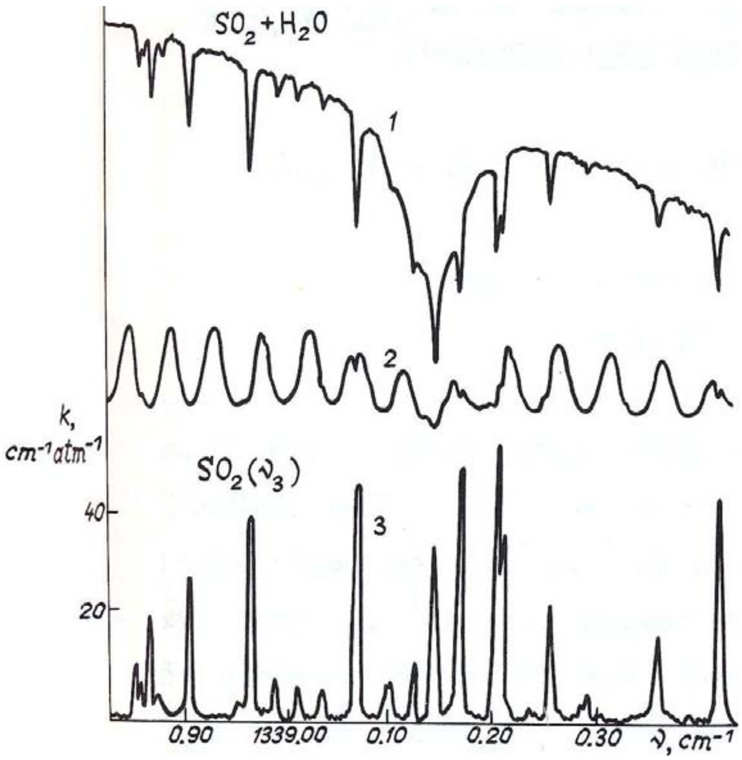

FIG. 4. Spectra obtained with a diode laser spectrometer equipped with a multipass matrix-array cell: 1) the narrow absorption peaks correspond to the vibrational rotational line of the ν_3 band of SO_2; the wide line corresponds to absorption by water in the atmosphere outside the cell; the narrow absorption line of water vapor is marked near the top of the wide line; 2) the reference spectrum of the Fabry-Perot etalon (the free dispersion field is 0.0397 cm^{-1}); 3) the spectrum of the absorption coefficient of SO_2 (ν_3).

Figure 3 shows a photograph of a matrix array of images on the collecting mirrors. This photograph clearly shows that there are no distortions after 306 passes of He-Ne laser radiation. The splitting of the points that is observed is associated with shifting of the superposed images. The last images in the form of ordinary points of the matrix array (second row from the top) have the same size and shape of the initial outlines of the cross section of the laser beam at the point of entry into the system. Only the exit point, which falls on the canted face of the auxiliary collecting mirror, destroys somewhat the sharply defined geometry of the matrix array.

Multipass cells based on a three-objective matrix-array system, enclosed in a cylindrical case have a larger relative opening than analogous cells with a four-objective matrix-array system with the same dimensions. Since the actual dimensions of the pupil of the system affect the aberrations of a wide beam most, for the three-objective system we performed a theoretical analysis of the aberrations.[16] In Ref. 16 it was found, in addition, that the conditions for focusing of an image on the exit slit and compensation of astigmatism were the same. The investigations performed in Ref. 16 made it possible to make a number of practical recommendations for constructing and operating MMSs.

Multipass three- and four-objective matrix-array systems make it possible to mount the objectives in one row. This is especially convenient in cases when the system is not mounted in an airtight tubular cell, for example, in systems placed on open paths. Figure 2*c* shows a uniform arrangement of objectives in a four-objective matrix-array system.

Multipass matrix-array systems have been included in the plan of the Member Countries of the Council of Mutual Economic Assistance "Instruments and methods for optical-spectroscopic measurements". Figure 4 shows spectra obtained at the Institute of General Physics of the Academy of Sciences of the USSR using a diode laser spectrometer equipped with a multipass matrix-array cell (Institute of Chemical Physics of the Academy of Sciences of the USSR). The air pressure was equal to 30 torr. The concentration of sulfuric anhydride was equal to 30 ppm. The optical path length was equal to 80 m (90 passes).

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