Laser and Particle Beams (2000), *18*, 529–534. Printed in the USA.

On the optical analysis of the ray path-lengths in the diffraction of femtosecond XUV and soft X-ray pulses

P. VILLORESI
Laboratorio di Elettronica Quantistica - DEI, Università degli Studi di Padova, via Gradenigo 6a, 35131 Padova, Italia and INFM-Istituto Nazionale per la Fisica della Materia - Padova

(Received 21 July 1999; Accepted 31 October 1999)

Abstract

During the diffraction of a femtosecond pulse with a grating, apart spectral filtering, there occurs a time stretching due to the difference in the path length among the rays diffracted, even of equal wavelength. The reason resides in the action of the grating: to depart from the behavior of the mirror, that is, $\alpha = \beta$, and to direct the rays in directions whose paths are groove-by-groove different of $m\lambda$. The compensation of this broadening is studied in the following by using a pair of grating in a particular mounting. The configuration is studied for the application in the selection of one single laser harmonics in the framework of high order harmonic generation processes.

1. INTRODUCTION

The evolution in the knowledge of the physics of the generation of laser pulses have pushed the lower limit of the pulse duration to the femtosecond time-scale. Recently, the generation of pulses of a few optical cycles was reported (Nisoli *et al.*, 1997; Nisoli *et al.*, 1998). The use of a femtosecond pulse combined with the chirped phase amplification (Strickland & Mourou, 1985) have opened the ways to a vast field of nonlinear processes in which the matter is exposed to radiation whose intensity may reach values from 10^{14} to 10^{20} W/cm^2 (Mourou *et al.*, 1998). As in other areas, the understanding of these phenomena requires the observation of the radiation emitted. In these domains, this is no longer bound to the visible or near infrared spectral region, where the laser wavelength usually is, but extend to the vacuum ultraviolet, the soft X-rays and sometimes also to hard X-rays (McPherson *et al.*, 1994; McPherson *et al.*, 1996). The time duration of this short wavelength emission is of the order of that of the laser or even shorter.

In some cases, as in the generation of high order laser harmonics (Corkum, 1993; Salières *et al.*, 1998), the radiation emitted in the spectral region of 50–5 nm have shown a high intensity (Protopapas *et al.*, 1997). This fact may lead to study schemes of experiments in which this radiation is used, at the same way of conventional sources such as a synchrotron or a laser-produced plasma. In this spectral region, the optical schemes for these experiments must use only reflecting optics, due to the lack of transparent materials (Samson, 1967). On the other hand, the selection of one single harmonic with a conventional soft X-ray monochromator introduces a time broadening that strongly reduces the pulse intensity. The reason of this effect is the grating diffraction, in which the rays that are aimed toward a direction different from the specular one, a diffraction order, reach the focal plane of the monochromator following paths that have different lengths. At the diffraction order m, the ray of wavelength λ diffracted by a given groove of the grating has a path difference of $m\lambda$ with respect to that diffracted by the nearby groove. The total time difference is obtained by the multiplication of this quantity times the total number of illuminated grooves N and by the division by the velocity of light c:

$$\Delta\tau = \frac{Nm\lambda}{c} = \frac{\Delta_{OP}}{c} \tag{1}$$

where Δ_{OP} is the total path length difference. This effect is irrelevant if the duration of the pulse is longer of the maximum time difference between the shortest and longest path, that is usually lower than one picosecond.

Address correspondence and reprint requests to: Paolo Villoresi, Laboratorio di Elettronica Quantistica - DEI, Università digli Studi di Padova, via Gradenigo 6a, 35131 Padova, Italy. E-mail: paolo.villoresi@dei.unipd.it

https://doi.org/10.1017/S0263034600183259 Published online by Cambridge University Press

By using the grating equation for the angle of incidence α and diffraction β,

$$m\lambda = d(\sin\alpha - \sin\beta) \tag{2}$$

where d is the grating period, the quantity $\Delta\tau$ is derived as (Born & Wolf, 1980)

$$\Delta\tau = \frac{Nd}{c}(\sin\alpha - \sin\beta) = \frac{Nm\lambda}{c}. \tag{3}$$

This effect produces a rotation of the front of the diffracted pulse that leaves the grating. For a pulse shorter than $\Delta\tau$, there are two consequences: (a) the increase of the pulse duration as seen from an observer on the exit slit of the monochromator and (b) the reduction of the section of the pulse normal to the propagation direction at a given time. This latter process can cause an increase of the diffraction spot-size in the focalplane, for large value of $\Delta\tau$ and will be discussed separately in the following.

2. NEEDS OF A COMPENSATED MONOCHROMATOR

The theory of the ultrashort pulse diffraction has been extensively studied for many processes, most of them nonlinear (Akhmanov *et al.*, 1992; Rullière, 1998). In particular, the laser pulse compressor (Treacy, 1969; Martinez, 1986) and stretcher (Martinez, 1987), that is at the base of chirped phase amplification, make use of the tilted front produced by the diffraction combined with the cancellation of the spectral dispersion by a pair of grating. Moreover, in the propagation of a femtosecond beam, an increase in the duration of the pulse due to the decoupling of the spatial frequency components for their different diffraction have been found (Christov, 1985).

On the other hand, the optics for the selection of an ultrashort pulse usually are designed for the visible or near infrared regions. The extension to the vacuum ultraviolet of these techniques is the purpose of this work. The first application for this is to select one single harmonic from the spectrum generated in the interaction with a gas jet or with condensed matter. In the case of the interaction of a few femtoseconds pulse and laser intensity between 10^{14} and 10^{15} Wcm^{-2}, the generation of the odd harmonics of orders above two hundred have been observed, and of time duration that is expected to be shorted than the laser pump pulse (Schafer *et al.*, 1996; Christov *et al.*, 1998).

In order to reduce the reduction of the intensity of the ultraviolet beam due to the time broadening caused from the grating, here we propose the design of a compensated monochromator where all the paths within the aperture have the same optical path. At the same time, the instrument has to select a suitable spectral portion, using the grating dispersion. Due to the spectral region of interest of the high order harmonics, the extreme ultraviolet (XUV, 100–30 nm) and the part of the soft X-ray (30–4 nm), the optical setup must use only reflecting components (Samson, 1967). Moreover, in this latter region, an acceptable reflectivity of optical surfaces is limited to the grazing incidence. This requirement is a further limitation to the design of the monochromator.

The scale length, relevant to the differences of path lengths, is that of pulse. This is $l = c\tau$, and, for a pulse of 5 fs, this is of 1.5 μm. Due to diffraction for $\lambda = 60$ nm and using a grating with 900 grooves per mm illuminated for 10 mm, from Eq. (3) $\Delta\tau = 18$ ps at first order. This value is more than three orders of magnitude larger than the duration of the laser pulse and of its harmonics. On the other hand, also the optical aberrations that affect the monochromator can cause further time broadening. Also in this case, the reason is the difference in the path length of the rays gathered by the instrument at different angles (Mahajan, 1991). By using a grating in off-axis mountings, the most serious aberration is astigmatism (Beutler, 1945; Welford, 1991). For a grazing incidence, other terms may also cause path length differences comparable to the pulse l value. Therefore, for a compensated optical design, both the diffraction of the grating and the dominant aberrations have to be corrected.

The first aspect is solved by using two identical gratings in opposite dispersion, so that $\alpha_1 = \beta_2$ and $\alpha_2 = \beta_1$. If $m = +1$, the rays diffracted by the first grating have a path length that increase from the side close to the entrance slit going to the opposite. By inverting the order, the result is also reversed. The second grating is then mounted in opposite order with respect to the first. The compensation of the length of different rays can also be obtained by keeping the same order of diffraction and by exchanging the relative arrival order on the surface: the ray that arrives at the position closest to the slit in the first grating, then arrive farther to the source in the second grating. This second scheme exchanges the arrival orders and is shown in Figure 1b while the first exchanges the diffraction orders and is shown in Figure 1a.

The difference between the two mountings is the combination of their relative dispersion: while in the first case the total dispersion is zero, in the second the total dispersion is twice that of a single grating. The compensated monochromator must adopt the first scheme, because the nonzero dispersion at the final focalplane is equivalent to the decoupling of the spectral components mentioned above.

In order to correct the astigmatism of the grating, an aspheric surface has to be chosen for each grating. The classical solution for a finite source and image is given by the torus grating (Haber, 1950; Welford, 1965). In this case, the stigmatic conjugation is obtained by a proper choice of the tangential radius R, and the sagittal one ρ, as in

$$\frac{\rho}{R} = \cos\alpha\cos\beta. \tag{4}$$

This condition is valid for a given pair of α and β, that is, for a given λ and m. For small rotations of the grating, the de-

https://doi.org/10.1017/S0263034600183259 Published online by Cambridge University Press

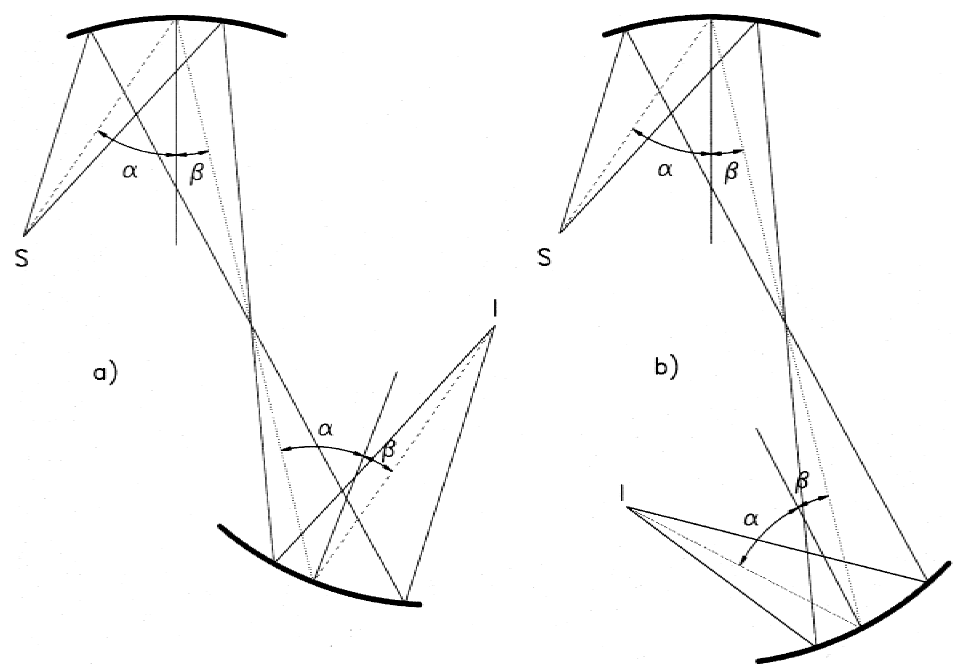

Fig. 1. Optical schemes that compensate the differences in the path length: (a) by exchanging the diffraction order; (b) by exchanging the arrival order on the grating.

parture from the stigmatic condition is also small, as will be verified later.

The scheme of the compensated monochromator that results from this discussion is the following: the beam containing the fundamental harmonic, used as a pump, and its harmonics is the source. Its size and divergence are very small, so the instrument can operate without an entrance slit (Salières *et al.*, 1996). The beam illuminates the first grating and is then diffracted. Either the internal or external orders can be used. Let's use the internal order for the first grating. In the focalplane of the grating is placed a slit, in order to select a given harmonic order. After the slit, a second grating is placed, in order to compensate the path length dispersion. This will have opposite diffraction order with respect to the first grating, and so the external one. The orientation of the grating is that of Figure 1a, in which the ray that first encounters the surface of the first grating does the same with the second one. The second grating is illuminated by a single harmonic, and there is no need for an exit slit. Thus, at its focal plane there is the nondispersed and focused image of the source at the wavelength selected. The rotation of the two gratings make the selection of a different order possible in a scheme of constant deviation angle $\Phi = \alpha + \beta$. The slit width is chosen by considering the particular profile of the harmonic spectrum under study, the diffraction spot-size, and the effect of the front rotation mentioned above.

3. RAY TRACING OF A TOROIDAL GRATING

The direction cosines of the rays incident on the grating have to be transformed into those diffracted by expressing in vector form, the grating diffraction formula Eq. (2). This latter derives from the Fermat principle of the least action for the optical path *OP* relative to a ray emerging from the source $S = (\xi, \eta, \zeta)$, reflected from the grating in the point $P = (x, y, z)$ and directed to the image $I = (\xi', \eta', \zeta')$ (Born & Wolf, 1980; Haber, 1950). We are using a coordinate system in which the grating vertex is at the origin. Thus, this quantity is expressed as:

$$OP = SP + PI + \frac{m\lambda}{d}x \tag{5}$$

with the convention that the grooves are on equispaced planes normal to $\hat{\mathbf{g}}$, that is parallel to axis $\hat{\mathbf{x}}$. The y-axis is parallel to the grooves and the z-axis normal to the grating surface, pointing toward its center. The Fermat principle here takes the form of the equality to zero of the derivatives of *OP* with respect to x and y.

The grating surface is expressed in this coordinate as

$$z = R - \sqrt{(R - \rho + \sqrt{\rho^2 - y^2})^2 - x^2}, \tag{6}$$

https://doi.org/10.1017/S0263034600183259 Published online by Cambridge University Press

and the incident and diffracted direction cosines as:

$$\hat{\mathbf{r}} = (L, M, N) = \left(\frac{\xi - x}{AP}, \frac{\eta - y}{AP}, \frac{\zeta - z}{AP} \right) \tag{7}$$

and

$$\hat{\mathbf{d}} = (L', M', N') = \left(\frac{\xi' - x}{AP'}, \frac{\eta' - y}{AP'}, \frac{\zeta' - z}{AP'} \right). \tag{8}$$

In these terms, the Fermat principles can be expressed as:

$$L + L' + (N + N') \frac{\partial z}{\partial x} - \frac{m\lambda}{d} = 0 \tag{9}$$

and

$$M + M' + (N + N') \frac{\partial z}{\partial y} = 0 \tag{10}$$

where the quantities to be found are the components of $\hat{\mathbf{d}}$. To this system of equations, the normalization of $\hat{\mathbf{d}}$

$$L'^2 + M'^2 + N'^2 = 1. \tag{11}$$

has to be added.

It is straightforward to derive the N component as:

$$N' = -\frac{A_2}{A_1} + \frac{1}{A_1}\sqrt{A_2^2 - A_1 A_3} \tag{12}$$

where

$$A_1 = \left(\frac{\partial z}{\partial x}\right)^2 + \left(\frac{\partial z}{\partial y}\right)^2 + 1, \tag{13}$$

$$A_2 = B_1 \frac{\partial z}{\partial x} + B_2 \frac{\partial z}{\partial y}, \tag{14}$$

$$A_3 = B_1^2 + B_2^2 - 1, \tag{15}$$

$$B_1 = \frac{-m\lambda}{d} - L - N \frac{\partial z}{\partial x}, \tag{16}$$

and

$$B_2 = -M - N \frac{\partial z}{\partial y}. \tag{17}$$

The other two terms are then:

$$L' = B_1 - N' \frac{\partial z}{\partial x} \tag{18}$$

and

$$M' = B_2 - N' \frac{\partial z}{\partial y}. \tag{19}$$

That completes the definition of the diffracted director cosine.

Table 1. *Parameters of the pair of grating for the XUV and soft X-ray regions.*

	XUV	soft X-ray
R (m)	0.35	2
ρ (m)	0.23	$9.7 \cdot 10^{-3}$
grooves/mm	200	100
Φ	70.5°	172°
f/#	50	100

4. DESIGN OF THE MONOCHROMATOR

The ideas exposed above are here applied in two cases in order to demonstrate the effectiveness of the compensation. The XUV and the soft X-ray regions are considered separately, because in the latter the incidence must be grazing, while this is not required in the former. The parameters of the pair of grating to be used in the two cases are summarized in Table 1. They consider the need to keep the setup as compact as possible and the effect that the optical aberrations are a growing function of the R value. Moreover, the dispersion was kept small in order to reduce the total path difference. For the XUV case, the plate factor has results of about 14 nm/mm, for a slit normal to the chief ray. By considering the difference with the nearby harmonics, the separation on the focal plane between the two gratings is about 500 μm. This figure is greater than the diffraction resulting from the front rotation. In this case, the instantaneous f/# can be derived from the general methods of Akhmanov *et al.* (1992) as:

$$f/\#_{inst} = \frac{Rm\lambda}{c\tau d} \tag{20}$$

Table 2. *Predictions for the XUV spectral region.*

Harmonic	λ (nm)	Δ_{OP} (μm)	$\Delta_{OPsingle}$ (μm)
11	72.7	0.7	102
15	53.3	0.5	75
21	38.1	0.4	53

https://doi.org/10.1017/S0263034600183259 Published online by Cambridge University Press

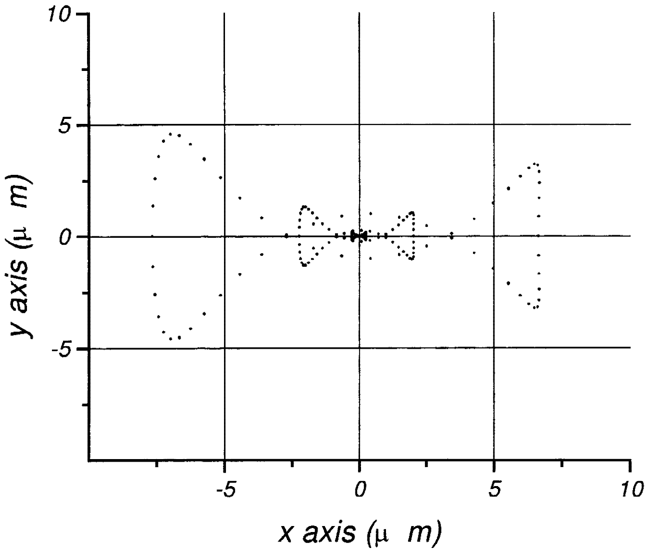

Fig. 2. Ray plot at the final focal plane for H141 at 5.7 nm. Here the *x*–*y* axis are intended to be normal to the chief ray and with the *y* axis parallel to the dispersion plane.

and give in the case of $\tau = 5$ fs the value for the diffraction spot of about 200 μm. This latter value can be taken as slit width, that corresponds to the selection of a single harmonics.

Table 3. *Predictions for the soft X-rays spectral region.*

Harmonic	λ (nm)	Δ_{OP} (μm)	$\Delta_{OPsingle}$ (μm)
111	7.2	0.7	14
141	5.7	0.7	12
171	4.7	0.6	10

The case of soft X-rays has a plate factor of 5 nm/mm. In this case, the diffraction has a negligible effect in the intermediate focal plane, of less than 10 μm.

The ray tracing of the setup has been carried out in order to estimate the geometrical aberrations. The XUV case reported in Table 2 is very well corrected, while the soft X-rays reported in Table 3 have a larger spot size, even if in this case it is also smaller than the diffraction spot. The examples of the ray tracing of H141 is shown in Figure 2, which is enclosed in an area of 10 μm $\times$ 15 μm.

The view of the ray paths for the same harmonic is shown in Figure 3 in order to depict the orientation of the setup. The observer looks from the source toward the focal plane to the path of the chief and of marginal rays of a square pupil.

The dependence of the total path *OP* on λ has been calculated. In all these cases, it gives a small contribution to the Δ_{OP} using a realistic linewidth of the harmonics. As an ex-

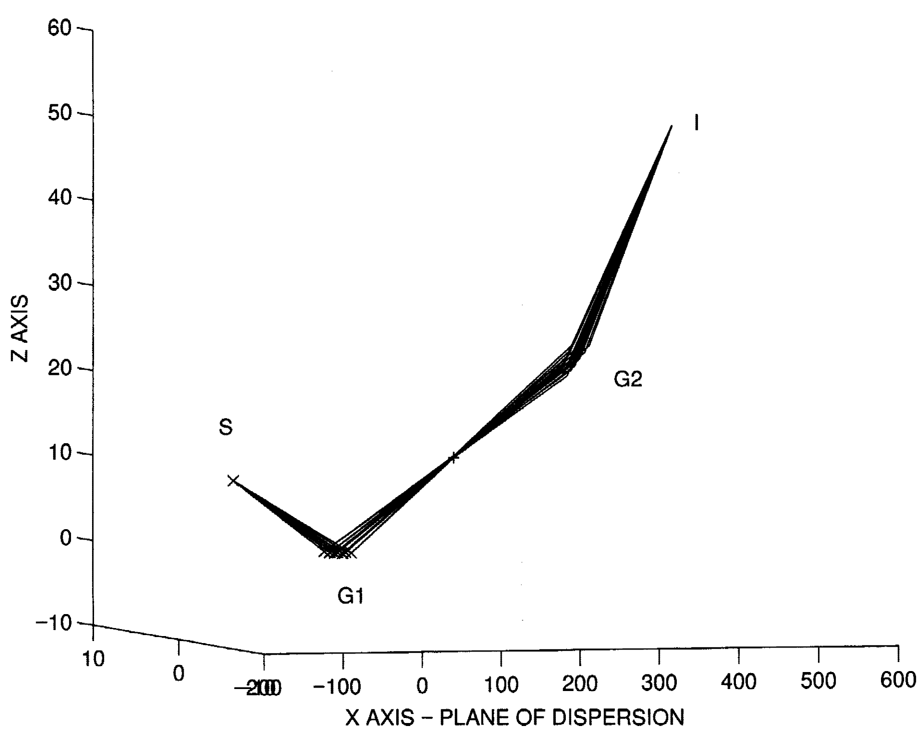

Fig. 3. Sketch of the rays for the H141. The source *S* illuminates the first grating *G*1, then the second grating *G*2 and finally is imaged at point *I*.

https://doi.org/10.1017/S0263034600183259 Published online by Cambridge University Press

ample, the value of $dOP/d\lambda$ for H15 is of -1.8 μm/nm and for H141 is of -3.1 μm/nm.

5. CONCLUSIONS

The design of a compensated monochromator has been discussed and two examples corresponding to the setup for the XUV and the soft X-ray were presented. The major steps were done to correct the dominant optical aberrations, to compensate the difference in path length of a single grating by using a second grating, identical to the first, and to adopt a mounting that cancels the total dispersion of the two grating. This method is intended to achieve the selection of a single harmonic order, or of a portion of a broad spectrum pulse, without a blur in its temporal structure, and in particular to preserve the duration even in the case of few femtoseconds.

ACKNOWLEDGMENTS

The present work was partially supported by the Italian INFM-Progetto di Ricerca Avanzata *FEXRAYS*. The author wishes to thank Professor Giuseppe Tondello and Drs. Maria-Guglielmina Pelizzo, Luca Poletto, and Paolo Ceccherini for helpful discussions and suggestions.

REFERENCES

Akhmanov, S.A., Vysloukh, V.A. & Chirkin, A.S. (1992). *Optics of femtosecond laser pulses*, Am. Inst. Physics, New York.
Beutler, H.G. (1945). *J. Opt. Soc. Am.* **35**, 311.
Born, M. & Wolf, E. (1980). *Principles of Optics*, Oxford: Pergamon.
Christov, I.P. (1985). *Opt. Commun.* **53**, 364.
Christov, I.P. *et al.* (1998). *Phys. Rev. A* **57**, R2285.
Corkum, P.B. (1993). *Phys. Rev. Lett.* **71**, 1994.
Haber, H. (1950). *J. Opt. Soc. Am.* **40**, 153.
Mahajan, V.N. (1991). *Aberration theory made simple*, Vol. **TT6**, SPIE Optical Washington: Engineering Press.
Martinez, O.E. (1986). *J. Opt. Soc. Am. B* **3**, 929.
Martinez, O.E. (1987). *IEEE J. Quantum Electron.* **QE-23**, 1385.
McPherson, A. *et al.* (1994). *Nature* **370**, 631.
McPherson, A. *et al.* (1996). *J. Phys.* **B29**, L291.
Mourou, G.A. *et al.* (1998). *Phys. Today* p. Jan/22.
Nisoli, M. *et al.* (1997). *Opt. Lett.* **22**, 522.
Nisoli M. *et al.* (1998). *IEEE J. Selected Topics in Quantum Electronics* **4**, 414.
Protopapas, M. *et al.* (1997). *Rep. Prog. Phys.* **60**, 389.
Rullière, C. (1998). *Femtosecond laser pulses*, Springer, Berlin.
Salières, P. *et al.* (1996). *J. Phys. B* **29**, 4771.
Salières, P. *et al.* (1998). In B. Bederson, H. Walther (ed.), *Advances in Atomic, Molecular and Optical Physics*, IOP.
Samson, J.A.R. (1967). *Techniques of Vacuum Ultraviolet Spectroscopy*, New York: John Wiley.
Schafer, K.J. *et al.* (1996). In *Generation, Amplification and Measurement of ultrashort laser pulses III*, Vol. 2701, p. 248, Bellingham: SPIE.
Strickland, D. & Mourou, G.A. (1985). *Opt. Communic.* **56**, 219.
Treacy, E.B. (1969). *IEEE J. Quantum Electron.* **QE-5**, 454.
Welford, W.T. (1965). In *Progress of Optics*, Vol. 4, p. 243, North Holland, Amsterdam.
Welford, W.T. (1991). *Aberrations of optical systems*, Bristol: Adam Hilger.