Spatial dispersion of crystals as a critical problem for deep UV lithography

A. G. Serebryakov,* F. Bociort, and J. Braat

Delft Technical University, Delft, the Netherlands

The birefringence effect caused by spatial dispersion in crystals has been known for more than a hundred years, but until recently it was studied only from the viewpoint of theoretical physics. This article discusses the mathematical description of the effect as well as problems associated with the presence of the effect in modern lithographic optical systems and methods of solving these problems. © *2003 Optical Society of America*

BIREFRINGENCE CAUSED BY SPATIAL DISPERSION IN LITHOGRAPHY

The birefringence effect caused by spatial dispersion (SD) was discovered theoretically in 1877 by Lorentz.[1] In the 1960s, Agranovich and Ginzburg gave a theoretical description of the effect, based on Maxwell's equations.[2] Quantitative measurements for semiconductors were first made in 1972.[3]

The possibility that the SD effect would influence the image quality in lithography was a surprise for the lithography industry, since SD can be a critical problem for the creation of a new generation of lithography machines with a wavelength of 157 nm and lens elements made from crystalline materials.[4–6] The world's leading producers of microcircuits initiated practical measurements of birefringence for CaF_2, the most widely used crystal in lithography, carried out by the National Institute of Standards and Technology (USA).[7–9] The value obtained for 157 nm was $\Delta n = (11.8 \pm 0.4) \times 10^{-7}$ (where Δn is the difference between the refractive indices for the ordinary and extraordinary rays). The most critical consequences of SD in lithography are the presence of quasi-interference effects in the image plane of the optical system, caused by polarization of the rays, and doubling of the rays when they are refracted at each surface. Losses of image contrast in the optical system, caused by the presence of these effects, can be more than 50% of the initial value, depending on the optical layout. The producers of lithographic machines were already acquainted with birefringence caused by residual stresses in crystals after polishing. However, the SD value is several times as large as the previously known dispersion effect caused by residual stresses, and new optical layouts and solutions consequently need to be developed to compensate it.

SPATIAL DISPERSION IN CRYSTALS

The essence of the SD effect is that the permittivity ε depends not only on the wavelength but also on the direction in which the wave propagates in the medium. The effect must be taken into account in media with regular structure (crystals) if the wavelength of the radiation becomes comparable with the dimensions of the crystal lattice. The effect thus needs to allowed for in the far UV when using even those crystals that are considered isotropic under ordinary circumstances. In practice, the presence of this effect means that pairs of polarized rays can appear after refraction at each surface, with the refractive index for each of the rays depending on the wavelength and propagation direction of the incident ray.

In general, the displacement vector **D** and the electric field vector **E** are mutually dependent, following from Maxwell's equations, where the inverse dielectric tensor $\varepsilon_{ij}^{-1}(\omega,\mathbf{k})$ is determined by frequency ω and the direction of wave vector **k**:

$$E_i(\omega,\mathbf{k}) = \sum_j \varepsilon_{ij}^{-1}(\omega,\mathbf{k}) D_j(\omega,\mathbf{k}), \tag{1}$$

$$\mathbf{D} = -\frac{c^2}{\omega^2}[\mathbf{k}(\mathbf{k}\cdot\mathbf{E})] = -\frac{c^2}{\omega^2}[\mathbf{k}(\mathbf{k}\cdot(\varepsilon^{-1}\mathbf{D}))]. \tag{2}$$

Since the wave vector is $\mathbf{k} = k\mathbf{s} = (\omega\tilde{n}/c)(\omega,\mathbf{s})\mathbf{s}$, where c is the speed of light in vacuum and **s** is the unit vector along the propagation direction of the ray, we have

$$\mathbf{s}[\mathbf{s}\cdot(\varepsilon^{-1}\mathbf{D})] + \frac{1}{n^2}\mathbf{D} = 0, \tag{3}$$

where n is the refractive index.

Since vector **D** is orthogonal to the wave vector and $\mathbf{s}\cdot\mathbf{D} = 0$, it is possible to choose a coordinate system XYZ so that **D** lies in the XY plane and vector **s** is parallel to the Z axis:

$$\mathbf{s} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}. \tag{4}$$

Equation (3) for the XY plane can be simplified by introducing the transverse component $\varepsilon_\perp^{-1}$ of tensor ε^{-1}. Then the eigenvalue equation with eigenvectors **D**′ and **D**″ and eigenvalue $1/n^2$ can be written in general as

$$\varepsilon_\perp^{-1}\mathbf{D} = \frac{1}{n^2}\mathbf{D}. \tag{5}$$

For a single component of vector **D**,

$$\delta_{ij}D_j = \left(\varepsilon_{ij\perp}^{-1}(\omega,\mathbf{k}) - \frac{1}{n^2}\delta_{ij} \right) D_j, \tag{6}$$

where δ_{ij} is the Kronecker delta ($\delta_{ij}=1$ if $i=j$, and $\delta_{ij}=0$ if $i\neq j$). Since tensor $\varepsilon_{ij}^{-1}(\omega,\mathbf{k})$ is real and symmetrical, the eigenvalues are real and the eigenvectors are orthogonal. Consequently, in an anisotropic medium, the polarization states of $\mathbf{D}'$ and $\mathbf{D}''$ are determined by the two eigenvectors.

Thus, the physical properties of an anisotropic material lead to the existence of two mutually orthogonal plane-polarized waves that propagate through a medium characterized by tensor $\varepsilon_{ij}^{-1}(\omega,\mathbf{k})$. To determine the refractive index of the crystal, it is necessary to find the eigenvalues of the projection of tensor ε_{ij}^{-1} in a plane orthogonal to $\mathbf{k}$. The refractive indices for the two polarized rays are then determined from the eigenvalues; i.e., $n=\sqrt{1/\mathrm{eigval}(\varepsilon_{ij\perp}^{-1})}$. Since the relative value of the effect is small, a series expansion in powers of the wave-vector components can be used to determine the inverse dielectric tensor: $\varepsilon_{ij}^{-1}(\omega,\mathbf{k})=\varepsilon_{ij}^{-1}(\omega)$ $+i\delta_{ijl}(\omega)k_l+\beta_{ijlm}(\omega)k_lk_m$, where $\varepsilon_{ij}^{-1}(\omega)$ describes the ordinary frequency dispersion, tensor $\delta_{ijl}(\omega)$ characterizes the gyrotropy, and tensor $\beta_{ijlm}(\omega)$ shows how tensor $\varepsilon_{ij}^{-1}(\omega,\mathbf{k})$ depends on the SD.

In general, tensor $\beta_{ijlm}(\omega)$ has 81 components, but, because of the high degree of symmetry of the CaF_2 crystal (symmetry type O_h^5 or m3m), the number of independent components reduces to three: β_{1111}, β_{1122}, and β_{1212}. Moreover, in our case, $\delta_{ijl}(\omega)=0$. It can be shown that the amount of birefringence, i.e., the difference between the two refractive indices for the two polarization states, is determined from

$$\Delta n(\omega,\mathbf{k})=\frac{1}{2}n(\omega)^3(\beta_{1111}-\beta_{1122}-2\beta_{1212})$$
$$\times k^2(\mathrm{eigval}_2(\delta_{ij}l_i^2)_\perp-\mathrm{eigval}_1(\delta_{ij}l_i^2)_\perp),\quad (8)$$

where l_1, l_2, and l_3 are the direction cosines of the angles between vector $\mathbf{k}$ and the coordinate axes. The refractive-index variations can be determined by a single measurement of the birefringence and by computing the eigenvalues for each propagation direction.

SPATIAL DEPENDENCE OF THE BIREFRINGENCE

To describe the spatial dependence of the birefringence for an arbitrary propagation direction, it is necessary to choose a coordinate system for the crystal lattice. A Cartesian coordinate system is ordinarily used in optics in which
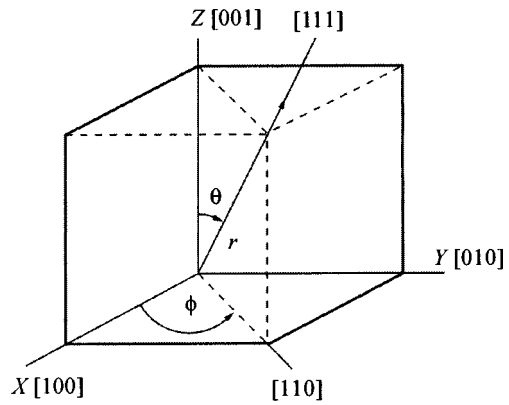

FIG. 1. Coordinate system of the crystal lattice.

the Z axis coincides with the optic axis, while the X and Y axes are orthogonal to it. In our case, there is also a coordinate system of the crystal lattice. This can be a Cartesian coordinate system coinciding with the edges of the CaF_2 cubic lattice or a system of spherical coordinates in which the ray intersects the lattice in a direction determined by radius vector $\mathbf{r}$, coinciding with unit wave vector $\mathbf{s}$ (Fig. 1). In this case, according to the rules adopted in crystallography, the directions of the coordinate axes are denoted as [100], [010], and [001], the face diagonals as [110], [101], and [011], and, finally, the cube diagonal as [111].

In general, since we have a tensor $\varepsilon_{ij}^{-1}(\omega,\mathbf{k})$ with known eigenvalues and eigenvectors and a measured birefringence for a certain definite direction like [100] or [111], we can rotate this definite tensor to an arbitrary propagation direction of the light wave. In a spherical coordinate system, we have an orthogonal set of vectors ($\boldsymbol{\theta}$, $\boldsymbol{\varphi}$, $\mathbf{r}$), and, to diagonalize a tensor in the plane of transverse $\mathbf{k}$, it is necessary to find the projection of the tensor in the two-dimensional space of $\boldsymbol{\theta}$ and $\boldsymbol{\varphi}$,

$$\mathrm{eigval}_{1,2}(\delta_{ij}l_i^2)_\perp=\mathrm{eigval}_{1,2}(R(\boldsymbol{\theta},\boldsymbol{\varphi})\delta_{ij}l_i^2R^{-1}(\boldsymbol{\theta},\boldsymbol{\varphi})),\quad (9)$$

where $R(\boldsymbol{\theta},\boldsymbol{\varphi})$ and $R^{-1}(\boldsymbol{\theta},\boldsymbol{\varphi})$ are rotation matrices. Then the eigenvalues of the tensor determine the refractive indices, and, to determine the polarization states, it is necessary to find the inverse projections of the eigenvectors in three-dimensional space. The value of the birefringence is determined from

$$\Delta n\sim\left(\mathrm{eigval}_2(\delta_{ij}l_i^2)_\perp-\mathrm{eigval}_1(\delta_{ij}l_i^2)_\perp=\sin^2\theta\sqrt{\frac{1}{16}\cos^4\theta(7+\cos 4\phi)^2+\frac{1}{4}\sin^4 2\phi+\frac{1}{4}\cos^2\theta\sin^2 2\phi(\cos 4\phi-5)}\right).$$
$$(10)$$

To describe the SD effect in a cubic crystal, coordinates θ and ϕ are varied from 0 to 2π. Then in a solid angle of 360° there are twelve maxima and fourteen minima of the birefringence (Fig. 2). The figure shows the refractive-index difference of various directions in a cubic crystal. Conse-

quently, a cubic crystal can be called heptaxial with respect to SD. In all, there are seven axes along which the effect equals zero: three axes coinciding with the cube edges {100}, and four cube diagonals {111}. However, these two sets of axes have different refractive indices. The maximum of the
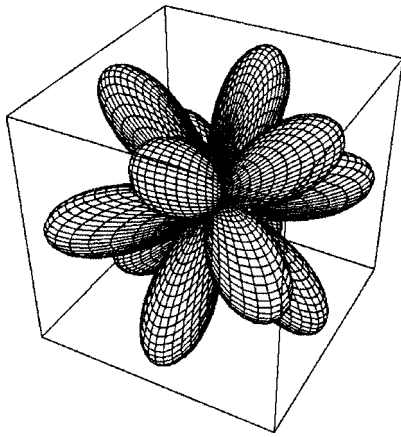
FIG. 2. Birefringence maxima and minima in a CaF$_2$ crystal.

FIG. 3. Angular dependence of the SD.

effect is observed in the {110} directions (the face diagonals). The most interesting behavior of the dependence of the birefringence is that in the plane of the cube diagonal, i.e., for $\phi = \pi/4$, $\theta = 0$ to $\pi/2$ (Fig. 3).
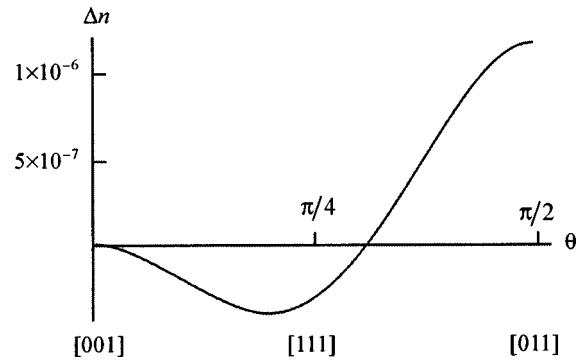
COMPENSATING THE SD EFFECT IN LITHOGRAPHY

The results of computing the birefringence for three directions are shown in Table I, where the character of the dependence of the SD over the pupil of the optical system is shown for an axial point of the image. For an off-axis point, some distortion of the radial pattern of the effect is observed.

TABLE I. Spatial dependence of birefringence.

Direction of optic axis	Character of variation of birefringence	Polarization state	
{100}			
{110}			
{111}			

To maintain the symmetry of the optical system, only three directions in the crystal—along {100}, {110}, and {111}—can be used for the optic axis, with the dependence of the effect on the angle of incidence being more symmetrical in the {100} and {110} directions than along {111}, where there is a change in the sign and the character of the variation of the effect. However, the {111} direction is preferable from the viewpoint of processing (polishing) the crystals, since the residual stresses are minimal for this direction. In principle, a random direction {abc} can be taken as the optic axis, but the computation of the effect and the technological problems can be unpredictable in this case.

The general strategy of compensating the effect is as follows: It can be seen from the dependence displayed in Fig. 3 that the effect can be mutually compensated, for example, by choosing the orientation of the components. Moreover, for the {100} and {111} directions, the effect is absent for rays close to the optic axis. The {100} direction has a symmetry of rotation of 45°, while {111} has a symmetry of 60°. Thus, it is possible to adjust the individual elements to each other, with the formation of a wavefront with minimum distortions. The minimum number of components for such a compensation method is four (0° for {100}, 45° for {100}, 0° for {111}, and 45° for {111}). The possibilities of compensation for an off-axis point are still unclear because of the complex character of the dependence of the effect.

Difficulties in designing such optical systems are also associated with the rate of calculating the ray path in the crystal, since, for each ray at each interface of the air–crystal medium, it is necessary to compute two refractive indices by the technique indicated above. Of the software available on the market, only one program (Code V, ©Optical Research Associates) includes a procedure for calculating the ray path taking SD into account. However, the doubling of the ray at each surface is ignored, whereas, even with twenty surfaces in the system, there can be $2^{20} \approx 10^{6}$ rays in the image plane for each object point.

A possible solution of the problem can be to create a "mixed" crystal. The SD effect is also present in other crystals with cubic symmetry, SrF_2 and BaF_2, but has the opposite sign. Thus, it is theoretically possible to create a crystal with the formula $Ca_{1-x}Sr_xF_2$ or $Ca_{1-x}Ba_xF_2$, in which SD for 157 nm is absent. However, in this case, the solution of the problem involves crystal synthesis.

CONCLUSION

This article has explained the main problems that arise in modern lithography in connection with the appearance of the spatial dispersion effect. A mathematical description of the effect and the possibilities of compensating it have been proposed.

*Email: a.serebriakov@tnw.tudelft.nl

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