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Electrostriction mechanism of Bragg grating formation in germanosilicate fibres

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***Abstract*. An electrostriction model is proposed for the photorefractive effect observed during the writing of Bragg gratings in germanosilicate fibres. Electrostriction is caused by a spatial charge grating formed upon the exposure to UV radiation. According to our estimate, the contribution of electrostriction to the photorefractive effect under real writing conditions is comparable with the contribution from colour centres and exceeds the contribution from the electrooptical effect by more than an order of magnitude. The electrostriction model explains the production of the IIA type Bragg grating in fibres with a high content of germanium in the core, as well as a number of effects that could not be explained earlier.**

Keywords: *optical fibre, refractive index grating, charge transfer, electrostriction*

1. State of the problem

A self-organised stable periodic variation $\Delta n(z)$ of the refractive index along the axis of an optical fibre was first discovered in germanosilicate fibres upon a prolonged exposure to laser radiation [1]. After the development of the holographic technique for writing refractive index gratings (RIGs) by UV laser radiation [2], the in-fibre RIGs have found wide applications as narrowband mirrors and filters in modern fibreoptic systems and devices. An impressive result of the application of Bragg RIG is the creation of a family of efficient fibre lasers and Raman converters. Germanosilicate fibres are still preferable for fabricating Bragg gratings and for studying the photorefractive effect. However, the available experimental results are not sufficient for obtaining a clear idea about the mechanism of RIG formation, which restricts the possibilities of fabrication and application of Bragg gratings.

Bragg RIGs are divided into three types: type I (typical) RIGs, type II RIGs formed due to local melting, and type IIA RIGs having the N-shaped dependence of Δn on the exposure time (single oscillation). The Bragg grating production is initiated by the photoinduced conversion of germanium oxygen-deficient centres (Ge – ODC) caused by UV laser radiation. Several mechanisms of the photorefractive effect are considered:

(1) structural modifications of glass (densification, change of thermoelastic stresses) [3, 4] which make the maximum contribution (more than 10^{-3}) to Δn;

(2) an increase in the refractive index due to an increase in the induced absorption (colour centre mechanism) [2, 5]; $\Delta n = 10^{-4}$ at 1.5 μm is determined by the Kramers – Kronig relation;

(3) spatial separation of charges leading to an increase in polarisability due to the electrooptic effect [6].

The model of spatially periodic separation of charges [6] assumes the photoionisation of defects and the creation of an axially periodic electric field with a period ~ 0.5 μm in which the refractive index can be modulated with the amplitude ($\Delta n \sim 10^{-6}$) due to the Pockels effect. However, the strongest modulation ($\Delta n \approx 10^{-5}$) is expected in the case of the Kerr effect when charges are separated by a distance of ~ 1 nm and randomly distributed dipoles are separated by several nanometers [7].

The spatially periodic charge separation was not considered in subsequent works because of its smallness. The variation Δn caused by other effects correlates with the excitation intensity, so that the refractive-index maxima $n(z)$ in the Bragg grating should coincide with the maxima of the writing interference pattern. These mechanisms produce the *spatially coinciding (phased) subgratings* Δn. The only exception is the special case of the thermoelastic stress relaxation model (alternatively known as the stress relief model) [4], when a subgrating may be formed in opposite phase with other subgratings.

The competition between the mechanisms (the N-shaped dependence of the modulation amplitude Δn on the exposure time) was first discovered in fibres with a high concentration of GeO_2 [8]. Gratings produced under such conditions were grouped into a separate category (type IIA). Several phenomenological models of production of IIA type gratings were proposed [9], which were based on a successive formation of two types of defects (or the interaction between two processes) resulting in changes in n of opposite signs. At the beginning of the exposure, defects of type A are formed, which lead to an increase in the amplitude Δn and in averaged $\langle n \rangle$ (a RIG of type I is produced). During their accumulation, defects of type A are transformed into defects of type B, which cause a stronger photorefractive effect of the opposite sign (production of RIG of type IIA).

It is accepted that RIGs of type I and IIA are in opposite phases, although this conclusion is not fully in accord with

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the experimental results. The minimum in the N-shaped dependence of Δn does not reach zero, and sometimes represents a weak oscillation against a fairly monotonic background. This means that the oscillation of Δn results from a competition between the spatially dephased subgratings. This is possible if one of these subgratings is associated with a spatially periodic charge grating. However, this raises questions about the mechanism and amplitude of the effect.

To explain this phenomenon, we propose the electrostriction mechanism of the photorefractive effect caused by a spatially periodic charge grating [10]. In this paper, we provide a more detailed substantiation for this model and obtain some quantitative estimates for the effect.

2. Spatially periodic charge grating

To produce a Bragg grating at the resonance wavelength λ_B, a field of interfering laser beams with a period $\Lambda = \lambda_B/2n_{eff}$ is formed, where n_{eff} is the effective refractive index for the fibre mode. For the wavelength $\lambda = 1.5$ μm, the period is $\Lambda \approx 0.5$ μm. The charge separation occurs due to diffusion of photoelectrons from a strongly illuminated region to a weakly illuminated region at a distance shorter than $\Lambda/2 \approx 0.25$ μm to both sides of the interference fringe.

The donors of photoelectrons in germanosilicate glass are the Ge–ODC, some photoinduced defects, and the matrix itself in the case of multiphoton excitation. For a stable charge grating to be formed, stable electron traps are required. The role of such traps is played by the well-known paramagnetic centres Ge(I) produced due to trapping of electrons by four-coordinated germanium atoms. Under typical conditions of RIG production, the number of Ge(I) centres may reach 10^{18} cm^{-3}. Such centres, which have an absorption band at 4.4 eV with the half-width ~ 1 eV, may donate an electron upon excitation into this band, and may migrate to the weakly illuminated region upon repeated excitation. In addition, more numerous unidentified traps exist in germanosilicate glass [11, 12]. The charge separation will continue until the drift current in the electric field of charges compensates for the diffusion current, or until the donors or acceptors of photoelectrons are depleted.

Fig. 1 shows the schematic diagram for charge separation. According to the accepted model, the positive charges $(Ge–ODC)^+$ at photoionised electron donors are immobile. During the recombination process, a part of the photoelectrons does not return to the initial donors, as indicated by an increase in induced absorption with the exposure time. An excess positive charge is created after relaxation of excitation at the maximum of the interference fringe. The excess negative charge creates symmetric maxima at the periphery of each interference fringe. A repeated pulsed or a prolonged continuous excitation produces a steady-state distribution of the excess charges.

Saturation of Δn usually occurs after the exposure ~ 10 kJ cm^{-2}, while induced absorption is saturated after the exposure ~ 100 J cm^{-2} (e.g., to the radiation at 248 nm). Because a change in the induced absorption serves as an indicator of the charge transfer, the charge grating may be produced at the very beginning of the RIG fabrication. The charge grating is a periodic structure in which the diameter of the charged layer (equal to the core diameter 3–8 μm) is much larger than the separation between layers (~ 0.25 μm). This allows us to use the model of a flat capacitor to estimate the electric field strength and to neglect the mechanical reaction of the fibre cladding to the core deformation.

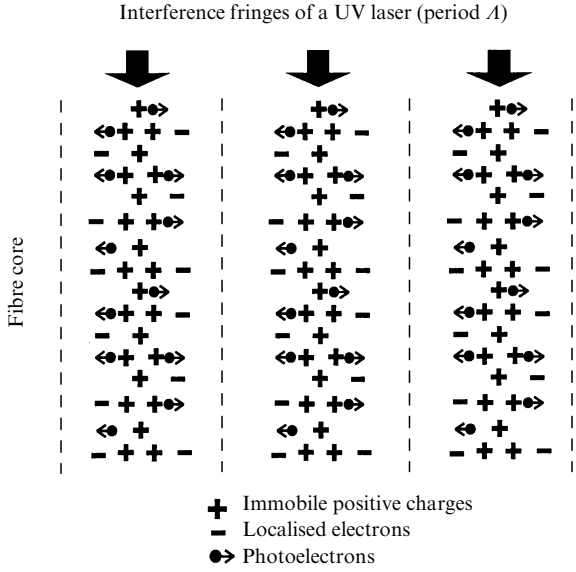

Figure 1. Charge separation in a fibre core.

3. Electrostriction

The direction of the electric field in a charge grating varies periodically along the fibre axis at the excess charge maxima where the field strength $E_d = 0$. As a result of electrostriction, the core will be most strongly compressed in the region of maximum E_d, while elongation will be observed in the region $E_d = 0$ according to the laws of elasticity. Thus, the charge grating produces an elastic periodic deformation $(V - V_0)/V = \Delta V/V = AE_d^2$ of the core volume, modulation of density and of the refractive index $n(z)$ ($\Delta V/V$ is the relative volume variation and A is the electrostriction coefficient).

To estimate the maximum field strength in the flat capacitor model, we assume that the effective excess charges are located at a certain effective distance from each other, while the field E_d (which is assumed homogeneous) corresponds to the maximum field in the gap between the charged layers. Really, it is natural to expect a smooth distribution of the excess charges, and, hence, of the field strength along the fibre axis, which will cause a smooth variation in the refractive index. Because electrostriction is a macroscopic effect, the proposed model will be applicable if the effective separation is not smaller than 20–30 nm.

According to Ref. [13], we can write for such a model

$$A = \left[\beta(\varepsilon - 1)\varepsilon^{-1} - \frac{d\varepsilon}{dp}\varepsilon^{-2}\right] \Big/ 8\pi, \qquad (1)$$

where ε is the dielectric constant; β is the isothermal compressibility of the medium; p is the pressure created by electrostriction. The second term in Eqn (1) corresponds to the contribution from photoelasticity. In reality, the photoelasticity and electrostriction make an additive contribution to the variation in n. Since we are interested in the variation of n caused by the variation in the sample volume under the action of the field, there is no need to separate these two

effects. Thus, the electrostriction coefficient can be written in the form

$$A_c \approx \frac{\beta(\varepsilon - 1)}{8\pi\varepsilon}. \quad (2)$$

We estimated the electrostriction coefficient from the fused silica constants, which do not differ significantly from those for the germanosilicate glass:

$$\beta = \frac{3(1-2\mu)}{E} = 2.9 \times 10^{-12} \text{ CGSE units}, \quad (3)$$

where E is Young modulus and μ is the Poisson ratio for fused silica. Using the value $\varepsilon = 3.77$, we obtain $A_c \approx 8.5 \times 10^{-14}$ CGSE units. Taking into consideration that

$$\frac{d\varepsilon}{dp} = 2n\frac{dn}{dp},\ n = 1.46 \text{ and } \frac{dn}{dp} = 1.0 \times 10^{-12} \text{ CGSE units [14]},$$

we can easily verify that the contribution of photoelasticity in formula (1) is only one tenth of the overall effect.

In real RIGs, the modulation amplitude is $\Delta n = 10^{-4}$. The variation of n is related to the volume variation by the expression

$$\frac{\Delta n}{n} \approx -\frac{1}{3}\frac{\Delta V}{V}. \quad (4)$$

To obtain the value $\Delta n = 10^{-4}$ due to electrostriction, the electric field E_d must be equal to 4.8×10^4 CGSE units $= 1.5 \times 10^7$ V cm^{-1}. This value is several times smaller than the breakdown threshold and is therefore attainable in principle. For thin charged layers in the interference-fringe geometry, we have

$$E_d = 4\pi\sigma, \quad \sigma = N_e \times 4.80 \times 10^{-10} \text{ CGSE units}, \quad (5)$$

where σ is the volume charge density and N_e is the electron concentration in a layer at the periphery of an interference fringe. To obtain $E_d = 1.5 \times 10^7$ V cm^{-1}, we must have $N_e = 7.6 \times 10^{12}$ cm^{-3}. This concentration is negligible compared to the concentration of electron traps.

The concentration of Ge–ODC is nearly proportional to the concentration of GeO_2 and is approximately equal to 10^{19} cm^{-3} for [GeO_2] = 10 %. During the writing of RIG, about half of the Ge–ODC are destroyed. The corresponding electron concentration $\sim 5 \times 10^{18}$ cm^{-3} is localised at a certain distance from the donors, i.e., participates somehow in the phototransfer. Thus, the charge transfer is not limited by the number of donors and traps, but the radiation intensity in the interference fringe must ensure a much higher concentration of photoelectrons than 7.6×10^{12} cm^{-3} in order to create conditions for the emergence of diffusion current.

To estimate the concentration of photoelectrons during RIG production, we used the results of Ref. [15] in which all the necessary parameters of the fibre and laser radiation are presented. The writing by a single laser pulse at 248 nm, with an energy of 20 mJ and a duration of 20 ns of Bragg gratings with $\Delta n \approx 10^{-5}$ whose thermal stability was much lower than the stability of induced absorption and structural variations was reported in Ref. [15]. We believe that such Δn gratings are associated with the spatially periodic charge grating with electron localisation at comparatively shallow traps [10]. Note that the Ge(I) centres can also be included in this category of traps because their thermal stability is much lower than the thermal stability of typical RIGs (200 °C as compared to $\sim 350-400$ °C under identical measuring conditions).

A single-mode fibre with a molecular concentration 15 % of GeO_2 in the core was used in Ref. [15]. The beam was focused by cylindrical optics and had an average energy density 450 mJ cm^{-2}. The concentration of photoelectrons can be estimated by assuming that the quantum yield of electron emission is equal to the initial efficiency of Ge–ODC decay, which is approximately equal to 0.1 [11, 12]. Taking the geometrical factors into account, the number of photons absorbed per pulse (averaged over the length of RIG) was 8.5×10^{13}, while the number of electrons emitted in the same period of time was 8.5×10^{12}, which corresponds to the average electron concentration 5×10^{15} cm^{-3} in the core and (at least) to a doubling of their concentration in the interference fringes $N_0 = 10^{16}$ cm^{-3}. A modulation of n with the amplitude $\Delta n = 10^{-5}$ by electrostriction requires the excess electron concentration 2.8×10^{12} cm^{-3}, i.e., a 0.03 % transfer of photoelectrons per pulse by a distance comparable to $\Lambda/2$. We are not aware of the actual path of photoelectrons during excitation and recombination.

The energy density used in Ref. [15] is about three times larger than the typical value for this quantity. Under typical writing conditions, the concentration of photoelectrons is expected to be $\sim 10^{15}$ cm^{-3}. Apparently, it is sufficient for creating an excess electron concentration of $10^{12}-10^{13}$ cm^{-3} during multiple pulse excitation.

Fig. 2 shows schematically the distributions of excess charges and field strength (intersection with the z axis corresponds to a change in the direction) relative to the distribution of the intensity of the writing radiation. These distributions illustrate the important features of the electrostriction subgrating of $\Delta n(z)$, namely, a spatial shift relative to the writing radiation and a clearly manifested spatial harmonic.

According to the known properties of germanosilicate glass [11], an increase in the GeO_2 concentration leads to an increase not only in the number of donors and electron traps, but also in the concentration of photoelectrons under chosen exposure conditions. The contribution of the electrostriction mechanism to Δn should increase superlinearly with the GeO_2 concentration. This explains the N-shaped dynamics of RIG production in fibres with a high GeO_2 concentration: initially, the RIG is formed due to the electrostriction mechanism, and then the contribution of other subgratings phased with the excitation intensity is manifested.

Let us turn to Ref. [15] again. The authors of this work observed a sharp increase in the modulation amplitude Δn with increasing the energy of single pulses above 30 mJ. For a pulse energy of 40 mJ, a relief grating with $\Delta n = 0.006$ was obtained and was found to be thermally stable up to 800 °C. According to the estimate obtained by the authors, the local temperature of grating writing could be as high as several thousand degrees. It was concluded that local melting was responsible for the production of the relief structure. In contrast to 'typical' gratings, such a grating was assigned to the type II by the authors of Ref. [15].

We believe that such an interpretation does not agree with the grating instability observed for a pulse energy of 20 mJ, when the local temperature also should exceed

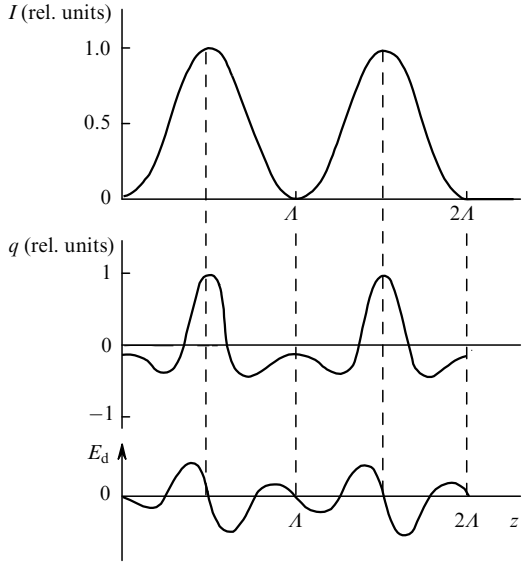

Figure 2. Distributions of excess charges q and the electric field strength E_d along the fibre axis relative to the distribution of the UV radiation intensity I.

1000 °C. Another explanation based on the electrostriction model can also be proposed. For energies of 20 and 40 mJ, the RIG is associated with the charge grating with the only difference that the electric breakdown that changes the structure of glass occurs for a higher pulse energy. This assumption is confirmed by the superlinear dependence of the photoionisation efficiency on the pulse intensity and the threshold character of the effect mentioned by the authors of Ref. [15].

4. Conclusions

The obtained estimates show that the electrostriction mechanism of Bragg RIG production with $\Delta n = 10^{-4}$ is possible in principle. Upon typical exposures, in this case, the macroscopic transfer of less than 0.1 % of all the photoelectrons available during exposure is required.

The photorefractive effect caused by electrostriction does not coincide spatially with other mechanisms. The electrostriction mechanism should dominate at the beginning of exposure and its contribution should increase with the concentration of GeO_2. The dynamics of change Δn of type IIA in fibres with a high concentration of GeO_2 is probably caused by a competition between the electrostriction mechanism and the structural rearrangement.

Apart from the fundamental period Λ, the charge grating should have a well-defined (preferably the first) spatial harmonic. The electrostriction mechanism in a specific fibre may or may not be manifested in the dynamics of Δn depending on the radiation intensity.

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References

1. Hill K O, Fujii Y, Johnson D C, Kawasaki B S *Appl. Phys. Lett.* **32** 647 (1978)
2. Meltz G, Morey W W *Proc. SPIE-Int. Soc. Opt. Eng.* 1516 185 (1991)
3. Poumellec B, Guénot P, Riant I, et al. *Opt. Mater.* **4** 441(1995)
4. Sceats M G, Atkins G R, Pool S B *Ann. Rev. Mat. Sci.* **23** 381 (1993)
5. Dong L, Archambault J-L, Russel P St J, Payne D N *Proc. Conf. on Opt. Comm. ECOC'94* (Firenze, Italy, 1994) Vol. 2, p. 997
6. Payne F P *Electron. Lett.* **25** 498 (1989)
7. Bernardin J P, Lawandy N M *Opt. Commun.* **79** 194 (1990)
8. Xie W X, Niay P, Bernage P, et al. *Opt. Commun.* **104** 185 (1993)
9. Dong P, Liu W F, Reekie L *Proc. SPIE-Int. Soc. Opt. Eng.* **2998** 49 (1997); *Opt. Lett.* 21 2032 (1996)
10. Dianov E M, Neustruev V B *Proc. SPIE-Int. Soc. Opt. Eng.* **4083** 132 (2000)
11. Neustruev V B *J. Phys.: Condens. Matter* **6** 6901 (1994)
12. Neustruev V B, Dianov E M, Kim V M, et al. *Fiber Integr. Opt.* **8** 142 (1989)
13. Landau L D, Lifshitz E M *Elektrodinamika sploshnykh sred* (Electrodynamics of Continuous Media) (Moscow: Nauka, 1982), p. 79
14. Mazurin O V, Strel'tsina M V, Shvaiko-Shvaikovskaya T P, in: *Svoistva stekol i stekloobrazuyushchikh rasplavov* (Properties of Glasses and Glass-Forming Melts) (Leningrad: Nauka, 1973), Vol. 1, p. 76
15. Archambault J-L, Reekie L, Russel P St *J Electron. Lett.* **29** 453 (1993)