

Kinetics of holographic refractive-index gratings in rare-earth-sensitized glasses

George S. Dixon, Abdulatif Y. Hamad, and James P. Wicksted

Department of Physics and Center for Laser and Photonics Research, Oklahoma State University, Stillwater, Oklahoma 74078

(Received 23 February 1998)

Diffusion of small modifier cations over distances large compared to the interatomic spacing is used to account quantitatively for the kinetics of photoinduced refractive-index gratings that can be holographically written in rare-earth-doped glasses. A model is developed that includes diffusion activated by nonradiative relaxation of the rare-earth sensitizers, drift under the space-charge field or externally applied fields, and trapping of the mobile modifiers. In this model the refractive-index contrast of the grating arises from the modulation of the concentration of small modifiers. The model accounts well for experiments on Eu^{3+} -doped silicate glasses reported here. [S0163-1829(98)04525-1]

I. INTRODUCTION

Rare-earth-sensitized silicate, phosphate, and germanate glasses can support refractive-index gratings created by exciting into states with strong nonradiative decay channels.¹⁻¹¹ These holographic gratings commonly exhibit a transient component and a persistent component. The extreme stability of the persistent gratings (essentially permanent when read with 632 nm or longer wavelengths at room temperature) in these glasses makes them excellent candidates for optical demultiplexers, filters, and read-only optical memories. The fast-transient gratings, also found in these systems, have been proposed for signal modulators⁷ and have promise as wavelength-selective switches. There is great potential for forming these glass-based devices as integrated fiber-optic systems. Grating strengths in these glasses have been steadily improved by optimization of the composition and processing parameters.

The transient component has been identified as an excited-state population grating in Eu^{3+} -doped glasses⁴ by its decay rate, which is identical with that of the long-lived 5D_0 excited state. A similar identification has been made with the 1D_2 state in Pr^{3+} -doped glasses.⁶ The source of the persistent gratings has proven more elusive. Behrens and co-workers⁴⁻⁶ attributed it to hot-phonon-mediated tunneling of network modifiers at the sites of the excited rare earths that undergo nonradiative decay. The change in the local refractive index of the glass in their model was the result of alteration in the immediate neighbors of the rare-earth sensitizers. Their mechanism has been challenged by Broer, Bruce, and Grokiewicz¹⁰ who did not observe persistent gratings in binary $\text{Na}_2\text{O-SiO}_2$ glasses doped with Er and with Pr; they did, however, produce both persistent and transient gratings with Eu doping. They raised the possibility of a multi-step electronic process as the source of the persistent gratings.

In this paper the defect physics of the persistent gratings in these glasses is investigated. The persistent gratings in these materials are attributed to a source with well-established roots in glass chemistry. Kinetic equations for the formation and optical erasure of these holographic gratings are developed and their predictions are compared with experiment.

II. MODEL

We consider the persistent refractive-index modulation to be due to an underlying modulation in the concentration of small network modifiers. This differs from the proposal of Behrens and co-workers⁴⁻⁶ in which the environment, but not the local concentration, of the modifiers is altered when the grating is written. The dependence of the refractive index of glasses on the concentrations of network modifiers is familiar to glass chemists. Appen¹² has shown that the refractive index of oxide glasses can be expressed as

$$n_d = \sum_i n_{d,i} c_i / 100, \quad (1)$$

where the c_i are the concentrations of the component oxides in mol % and the $n_{d,i}$ factors are their contributions to the refractive index.

In this picture it suffices to calculate the density $M(x,t)$ of mobile modifiers to obtain the strength of the *persistent* grating. To be mobile a modifier must be small enough in diameter to pass along the interstices of the network. This consideration indicates that light alkali or alkaline-earth modifiers, e.g., Na, Li, and Mg, will be the most important chemical species for formation of permanent gratings. This is consistent with previous experimental observations.^{4,8} These modifiers must also be bound in wells that are shallow enough that they can be excited out of them by the hot phonons available locally. Because of this, the density of *mobile* modifiers of a given chemical species may be substantially smaller than the total density of that species.

The modulation of the concentration of modifiers arises by hot-phonon-driven diffusion from the native composition. As in the proposal of Behrens and co-workers,⁴⁻⁶ the hot phonons that drive the production of the persistent grating are produced by nonradiative relaxation of the rare-earth excited state. Figure 1 shows a simplified energy-level diagram for the excitation-relaxation scheme of the rare-earth sensitizer. It is initially excited to the upper excited state $\langle 1^* \rangle$ from which it can relax radiatively to the ground-state manifold or nonradiatively to the lower excited state $\langle 2^* \rangle$. The lower excited state subsequently relaxes radiatively to the ground-state manifold. R_0 , R_1^* , and R_2^* represent the densi-

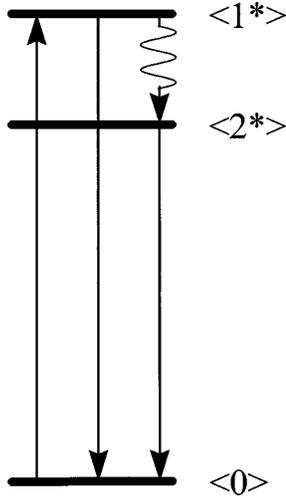


FIG. 1. Simplified energy-level diagram for rare-earth sensitizers. The $\langle 1^* \rangle \rightarrow \langle 2^* \rangle$ transition is nonradiative.

ties of rare earths in the respective states. If the lower excited state is long-lived compared to the upper state, the strength of the transient will be proportional to R_2^* and, thus, to the density of hot phonons.

This leads to a total refractive-index modulation for the grating given by

$$\Delta n(x,t) = gR_2^*(x,t) + \sum_i n_{di} \frac{c_i}{100} \frac{\Delta M_i(x,t)}{\bar{M}_i}, \quad (2)$$

where the n_{di} and c_i are the factors from Eq. (1) above for the mobile modifiers, $\Delta M_i(x,t)$ is their local excess density, \bar{M}_i is the mean total density of the i th chemical species, and g is a proportionality factor for the strength of the transient grating.

Transport equation. For simplicity, suppose that only one species of modifier is mobile. The mobile modifiers are distributed over a uniform density of sites S . The mean density of *mobile* modifiers is M_0 . In the native state of the glass these mobile modifiers are also distributed uniformly. The effect of a gradient in the light intensity in the sample is to produce a corresponding gradient in the probability that a modifier can hop among neighboring sites. One-dimensional diffusion resulting from the gradient in the hopping probability and drift under the action of the space-charge field and an external electric field are considered. All fields and the net redistribution of modifiers are parallel to the gradient.

First consider only the diffusion. Suppose that each site can accommodate only one modifier at a time. The density of empty sites to which the modifiers may diffuse is $O(x,t) = S - M(x,t)$. Let a be the mean separation of sites. Then the kinetics of the modifier distribution are given by

$$\begin{aligned} \frac{\partial M(x,t)}{\partial t} = & -\gamma(x,t)M(x,t)[O(x+a,t) + O(x-a,t)] \\ & + [\gamma(x+a,t)M(x+a,t) \\ & + \gamma(x-a,t)M(x-a,t)]O(x). \end{aligned} \quad (3)$$

$\gamma(x,t)$ is the rate parameter for hopping between adjacent sites. It is optically stimulated by the high-energy phonons

produced in nonradiative relaxation of the excited rare-earth modifiers. We take $\gamma(x,t)$ to be proportional to the local hot-phonon density $\phi(x,t)$ so that $\gamma(x,t) = \gamma_0 \phi(x,t)$.

Since $a \ll \Lambda$, the period of the grating, it is convenient to pass to the continuum limit by letting, for any position-dependent property $f(x,t)$,

$$f(x \pm a, t) \approx f(x, t) \pm \frac{\partial f(x, t)}{\partial x} a + \frac{1}{2} \frac{\partial^2 f(x, t)}{\partial x^2} a^2. \quad (4)$$

In this limit

$$\begin{aligned} \frac{\partial M(x, t)}{\partial t} = & -\gamma_0 a^2 \left[\phi(x, t) M(x, t) \frac{\partial^2 O(x, t)}{\partial x^2} \right. \\ & \left. - O(x, t) \frac{\partial^2 \phi(x, t) M(x, t)}{\partial x^2} \right]. \end{aligned} \quad (5)$$

Recalling that $O(x, t) = S - M(x, t)$, this becomes

$$\begin{aligned} \frac{\partial M(x, t)}{\partial t} = & +\gamma_0 a^2 \left[\phi(x, t) M(x, t) \frac{\partial^2 M(x, t)}{\partial x^2} \right. \\ & \left. + [S - M(x, t)] \frac{\partial^2 \phi(x, t) M(x, t)}{\partial x^2} \right]. \end{aligned} \quad (6)$$

Under conditions of uniform illumination $\phi(x, t) = \phi_0$, so that Eq. (6) reduces to

$$\frac{\partial M(x, t)}{\partial t} = +\gamma_0 \phi_0 S a^2 \left[\frac{\partial^2 M(x, t)}{\partial x^2} \right]. \quad (7)$$

To include the effects of the space charge and applied fields, express the transport using the equation of continuity

$$\frac{\partial M(x, t)}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (8)$$

and divide \mathbf{J} into diffusion and drift contributions

$$\mathbf{J} = -\eta \nabla M + \mu M \mathbf{E}. \quad (9)$$

Letting q be the charge on the mobile modifiers, the diffusion constant η and the mobility μ are related by the Einstein relation

$$\mu = \frac{q}{k_B T} \eta. \quad (10)$$

The electric field \mathbf{E} is the sum of the applied field \mathbf{E}_A and the space-charge field \mathbf{E}_S given by application of Gauss's law to the modifier distribution. The diffusion term in the equation of continuity is identical with Eq. (6). Under uniform illumination

$$\mu = \frac{q}{k_B T} \gamma_0 S a^2 \phi_0. \quad (11)$$

When the illumination is nonuniform, a local mobility can be defined by replacing ϕ_0 by $\phi(x)$ in Eq. (11) above. This gives

$$\nabla \cdot \mathbf{J}_{\text{drift}} = \frac{q}{k_B T} \gamma_0 S a^2 \frac{\partial}{\partial x} [\phi(x, t) M(x, t) E(x, t)]. \quad (12)$$

It can also be expected that the glass contains sites that bind the mobile modifiers too tightly for them to escape. In lightly doped material these might simply be sites too far from the rare-earth activators for the hot phonons to be effective in mobilizing modifiers from them. Their effect on the transport is included by following the buildup of trapped modifiers.

$$\frac{\partial N(x,t)}{\partial t} = \gamma_T \phi(x,t) M(x,t) [S_T - N(x,t)]. \quad (13)$$

Here $N(x,t)$ is the density of trapped modifiers, S_T is the density of deep traps, and γ_T is the rate constant for the trapping.

The full-transport equation, including diffusion, deep traps, and drift, can be expressed as

$$\begin{aligned} \frac{\partial M(x,t)}{\partial t} = & + \gamma_0 a^2 \left[\phi(x,t) M(x,t) \frac{\partial^2 M(x,t)}{\partial x^2} \right. \\ & \left. + [S - M(x,t)] \frac{\partial^2 \phi(x,t) M(x,t)}{\partial x^2} \right] \\ & + \frac{q}{k_B T} \gamma_0 S a^2 \frac{\partial \phi(x,t) M(x,t) E(x,t)}{\partial x} - \frac{\partial N(x,t)}{\partial t}. \end{aligned} \quad (14)$$

As noted above, the space-charge field can be expressed as a function of the modifier densities using Gauss's law:

$$\nabla \cdot \mathbf{E}_S = \frac{q}{\epsilon} [M(x,t) + N(x,t) - M_0]. \quad (15)$$

Since each term in the transport equation (14) is proportional to the hot-phonon density $\phi(x,t)$, which results from the excitation and relaxation of the sensitizers, the gratings formed by this process will be permanent in the dark.

It is often the case experimentally that there are far more sites for mobile modifiers than there are mobile modifiers, i.e., $S \gg M_0$. In this case Eq. (14) simplifies to

$$\begin{aligned} \frac{\partial [M(x,t) + N(x,t)]}{\partial t} = & \gamma_0 S a^2 \left[\frac{\partial^2}{\partial x^2} [\phi(x,t) M(x,t)] \right. \\ & \left. - \frac{q}{k_B T} \frac{\partial}{\partial x} [\phi(x,t) M(x,t) E(x,t)] \right]. \end{aligned} \quad (16)$$

This expression rather than the full-transport equation (14) will be used in the following paragraph. The full-transport equation is needed only when the density of mobile modifiers approaches the density of available sites so that it becomes difficult to write gratings. Glasses of this latter type may be of little technical interest.

Sinusoidal gratings. Refractive-index gratings in these materials are produced by the interference of two overlapping laser beams ("write beams") on resonance with a rare-earth optical transition and detected by Bragg diffraction of a longer-wavelength probe beam off-resonance (the "read beam"). To allow for the possibility that the intensities I_1 and I_2 of the two write beams are not equal, it is

convenient to define a "fringe contrast factor" $m = 2(I_1 I_2)^{1/2} / (I_1 + I_2)$.¹³ Then, with $x=0$ chosen to lie on an intensity maximum,

$$I(x) = (I_1 + I_2) [1 + m \cos(Kx)], \quad (17)$$

where $K = 2\pi/\Lambda$ is the wave number of the grating. Taking the hot-phonon density to be proportional to the local light intensity gives

$$\phi(x) = \frac{\phi_0}{2} (2 + m e^{iKx} + m e^{-iKx}). \quad (18)$$

This allows the gratings to be expressed as Fourier series

$$M(x,t) = \sum_{p=-\infty}^{\infty} M_p(t) e^{ipKx} \quad (19)$$

and

$$N(x,t) = \sum_{p=-\infty}^{\infty} N_p(t) e^{ipKx} \quad (20)$$

for the mobile and trapped modifiers, respectively. The electric field will have the form

$$E(x,t) = E_A - \frac{iq}{\epsilon K} \sum_{p \neq 0} \frac{M_p(t) + N_p(t)}{p} e^{ipKx}. \quad (21)$$

Since the left-hand sides of these relations are real, it follows that $M_{-p} = M_p^*$, etc. If the applied field $E_A = 0$, all the Fourier coefficients for M and N are real so that $M_{-p} = M_p$, etc. The orthogonality of the complex exponentials allows the transport and trapping equations (16) and (13) to be reduced to a set of coupled-rate equations.

$$\begin{aligned} \dot{M}_n + \dot{N}_n = & -\beta_n (2M_n + mM_{n+1} + mM_{n-1}) \left(1 + \frac{iqE_A}{k_b TK} \right) \\ & + \frac{q^2 \beta_n}{\epsilon k_B TK^2} \sum_{p \neq 0} \frac{n}{p} (2M_{n-p} + mM_{n-p+1} \\ & + mM_{n-p-1}) (M_p + N_p) \end{aligned} \quad (22)$$

and

$$\begin{aligned} \dot{N}_n = & \gamma_T \phi_0 S_T (2M_n + mM_{n+1} + mM_{n-1}) \\ & - \gamma_T \phi_0 \sum_p N_p (2M_{n-p} + mM_{n-p+1} + mM_{n-p-1}), \end{aligned} \quad (23)$$

where $\beta_n = \gamma_0 \phi_0 S K^2 a^2 n^2 / 2 = n^2 \beta_1$. The first term in Eq. (22) arises from diffusion and from drift under the applied field; the second arises from drift under the space-charge field. Note that these rate equations describe both writing conditions $m \approx 1$ and optical erasure (bleaching) by uniform illumination $m = 0$. Optical erasure is often done by blocking one of the write beams; in this case $\phi_0^{\text{erase}} = \phi_0^{\text{write}}/4$, assuming proportionality between light intensity and hot-phonon production.

It is important at this juncture to examine the physical effect of the trapping given by Eq. (23). It should be emphasized at the outset that the trapping is *not* necessary for the

production of gratings that are fixed in the dark. This is primarily the result of the diffusion of modifiers to a modulated distribution that is fixed when illumination at the write-beam wavelength ceases, but can be erased by subsequent uniform illumination. The deep traps have two effects on the gratings. (1) They cause the gratings to decay slowly with extended writing times because some of the modifiers are back-diffusing from shallow sites. These may be trapped at sites that are more nearly uniformly distributed than would be the case if there were no trapping. This effect is especially important when the number of deep traps is comparable to the number of mobile modifiers. (2) They lead to a grating that is resistant to bleaching even by light of the same wavelength as the write beams. This is of considerable practical interest since many of the applications that have been proposed for photorefractive media require that holograms be written and read at the same wavelength. This can minimize the erasure problem that many photorefractive materials exhibit when read on resonance.

The first-order Bragg diffraction from the persistent grating is due solely to the $M_1 + N_1$ Fourier amplitude. The family of coupled-rate equations represented by Eqs. (22) and (23) can be solved numerically so that their predictions can be compared with experiment. The transient gratings decay with the lifetime, < 3 ms, of the terminal excited-state density R_2^* . On the time scale of the growth and decay of the persistent gratings, > 10 s, this is instantaneous. The transient grating is included by a step function that turns on and off with the write beams. The amplitude of the refractive-index modulation in Eq. (2) is then

$$\Delta n = gR_2^* + n_d \frac{c}{100} \frac{M_1 + N_1}{\bar{M}}. \quad (24)$$

We shall refer to this Δn amplitude as the refractive-index contrast.

This model can also be adapted to describe materials in which the refractive-index contrast arises from the electro-optic effect, as is the case in many photorefractive materials. In that limit the present model is similar to the hopping model of Feinberg *et al.*¹⁴ and is related in its results to the band-transport model of Kukhtarev *et al.*¹⁵

There are four parameters in Eqs. (22)–(24) that can be adjusted to fit experiments. β_1 principally sets the rate of growth and decay of the persistent gratings. M_0 determines the number of mobile modifiers and, thus, the strength of the persistent gratings. The product $\gamma_T \phi_0 S_T$ sets the rate at which mobile modifiers become trapped (S_T is a separate parameter if almost all the traps become filled), and thus, the decay of the grating under extended writing conditions. Finally, the transient grating is set by gR_2^* .

III. EXPERIMENT

The experiments reported here were performed using the conventional nondegenerate four-wave-mixing configuration. Gratings were written with the 465.8-nm line of an Ar^+ laser operating in the TEM₀₀ mode. This line pumps the ${}^7F_0 \rightarrow {}^5D_2$ transition of the Eu^{3+} sensitizer. The Gaussian profile of the beam was confirmed with a laser beam profiler. Gratings were detected with the first-order Bragg diffraction

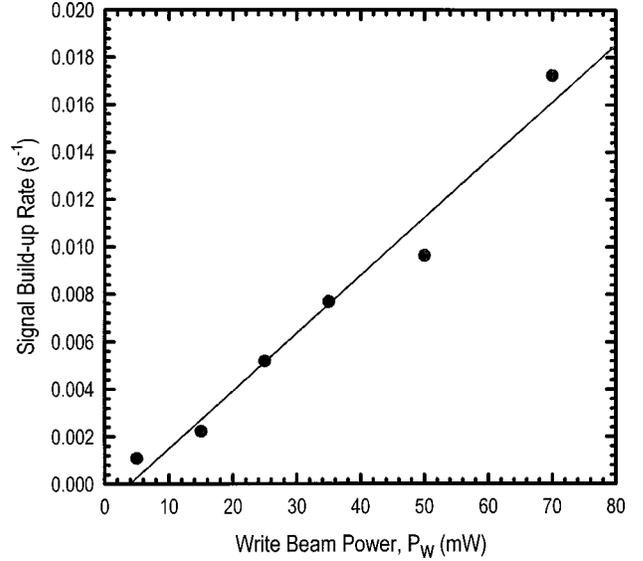


FIG. 2. Power dependence of the growth rate of the gratings.

of 632.8-nm light from a He-Ne laser. Each of the laser beams was focused to a 200- μm -diam spot. In all of the experiments reported here a write-beam crossing angle $2\theta = 4.25^\circ$ was used. The phototube used as the detector was carefully calibrated so that the absolute scattering efficiencies of the gratings could be measured. The refractive-index contrast was calculated for each grating using the method of Hamad and Wicksted.¹⁶ This has allowed quantitative comparison of gratings in different samples.

The glass samples had a base composition $70\text{SiO}_2 \cdot 15\text{Na}_2\text{O} \cdot 12\text{MgO} \cdot 3\text{Al}_2\text{O}_3$. Sample B9 was doped with 2.5-mol % Eu_2O_3 , and sample B10 was doped with 5-mol % Eu_2O_3 . This ratio of Eu concentration between the two samples was confirmed by optical-absorption measurements.

IV. COMPARISON WITH EXPERIMENT

The transport and trapping equations (22) and (23) allow the kinetics of growth and decay of the gratings to be modeled and compared with experiments. A basic assumption of the model is that the transport is driven by nonradiative relaxation of the rare-earth excited state. In our samples this nonradiative relaxation is between the 5D_2 and 5D_0 states of Eu^{3+} . Since, as Eqs. (22) and (23) show, the transport and trapping coefficients are taken to be linear in the amplitude ϕ_0 of the hot-phonon density, the rate of growth of the persistent gratings are predicted to be linear in the write-beam power P_W due to the proportionality between ϕ_0 and P_W . Figure 2 shows the growth rate of gratings, as measured by the reciprocal of the time to reach half maximum, in sample B10. The linear dependence exhibited by these data confirms the validity of the assumption of a linear driving mechanism for production of the persistent gratings. This also seems to us to rule out the multistep electronic processes suggested by Broer, Bruce, and Grokiewicz¹⁰ as a source for the persistent gratings.

Figure 3 displays results for two write-block-erase cycles in sample B10 with a total laser power of 50 mW. During the initial 30 s both write beams are blocked to establish the

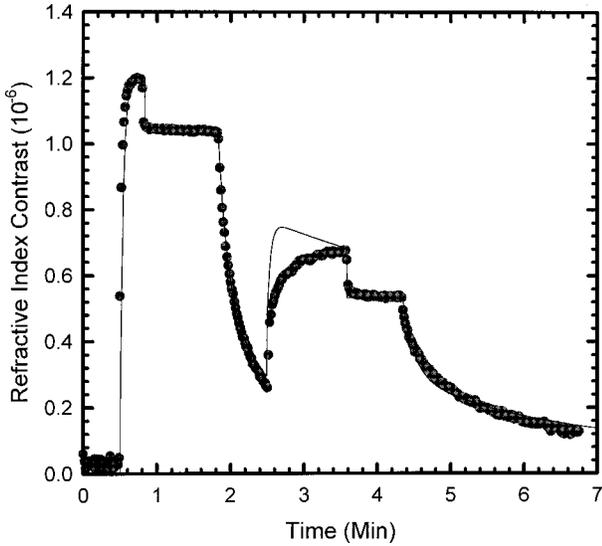


FIG. 3. Grating kinetics of sample B10 at 50 mW of write-beam power for two write-block-erase cycles. The curve is from the present model as described in the text.

background level. Next the write beams are unblocked and the grating is written through maximum. The write beams are then blocked for 60 s to allow the persistent grating to be observed. Finally, one of the write beams is unblocked to erase the persistent grating. Then the cycle begins again by rewriting the grating. The curve in the figure is a fit to the data using the model presented here. β_1 , M_0 , and gR_2^* were adjusted to fit the leading edge, the maximum, and the transient, respectively, in the first write cycle. $\gamma_T\phi_0S_T$ was adjusted to fit the decay of the persistent grating between the first and second blocking cycles. All fits of the model to data were ‘‘eyeball’’ only and were done by successive approximations of the fitting parameters. The fitting parameters are displayed in Table I. The mobile modifier concentration M_0 , assumed to be Na, is of the order of 1 ppm of the total Na concentration in the glass.

The model provides an excellent fit to all the data except during the middle stages of the second write cycle. This discrepancy is readily understood by the differences between the simple model developed here and real glasses. Since all the mobile modifiers are taken to have the same γ_0 rate parameter, the model implicitly assumes the same depth for all the shallow wells in which they reside. Ionic conductivity experiments show that real glasses possess a wide spectrum of well depths for small modifiers.¹⁷ It is reasonable to expect that the modifiers will be distributed into wells that are deeper on the average (compared to available hot phonons) after optical erasure than was the case for the native distribution of modifiers. This native distribution approximates a thermal distribution at the fictive temperature of the glass.

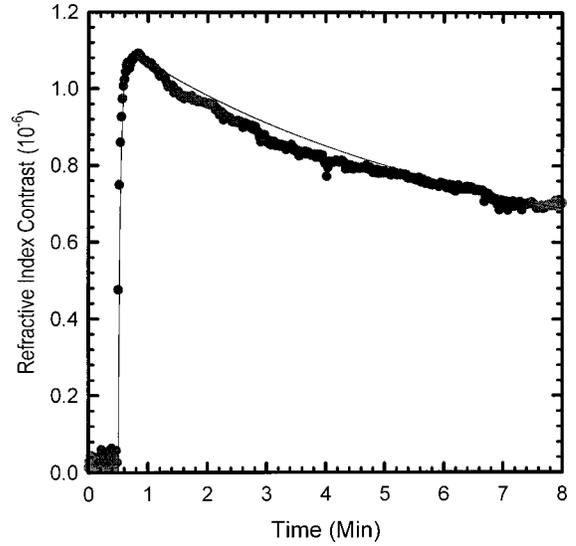


FIG. 4. An extended write cycle for sample B10 under the same conditions as in Fig. 3.

Thus, the model predicts a faster regrowth of the grating than is possible in the physical glass.

To test the applicability of the parameters used to fit the data in Fig. 3, an extended write cycle was performed at a neighboring fresh location in sample B10. These results are displayed in Fig. 4. The curve is the model calculation for continuous writing using the same parameters as were used to fit Fig. 3 with the exception that M_0 has been reduced slightly to agree with the smaller maximum produced at this new location. Again, there is excellent agreement between the model and the experiment.

When the transient grating is due to a long-lived terminal state in the nonradiative relaxation process, as is the 5D_0 state of Eu^{3+} , the strength of the transient grating is not only proportional to the density of excited sensitizers R_2^* , it is also proportional to the density ϕ_0 of hot phonons in the glass. Thus, β_1 , M_0 , and $\gamma_T\phi_0S_T$ should all scale from sample to sample in proportion to R_2^* as measured by the strength of the transient grating. Note that it may be important to use this excited sensitizer density rather than the total sensitizer density to scale among samples of different sensitizer concentration. Branching ratios for radiative and nonradiative relaxation of the initial excited state may differ for sites of different local symmetry, and it cannot be guaranteed that different concentrations of sensitizer will have the same distribution over the various possible sites.

Figure 5 shows data for sample B9 collected under the same excitation conditions as were used in sample B10. The curve is calculated from the model using the ratio of the measured transients to scale the remaining three parameters from their values found for sample B10. It should be empha-

TABLE I. Fitting parameters.

Sample no.	Eu_2O_3 conc. (mol %)	Laser power (mW)	β_1 (s^{-1})	M_0 (m^{-3})	$\gamma_T\phi_0S_T$ (s^{-1})	q (C)	$\Delta n_{\text{transient}}$
B10	5	50	0.158	2.44×10^{22}	0.0415	0	3.00×10^{-8}
B9	2.5	50	0.0395	6.10×10^{21}	0.0138	0	7.50×10^{-9}

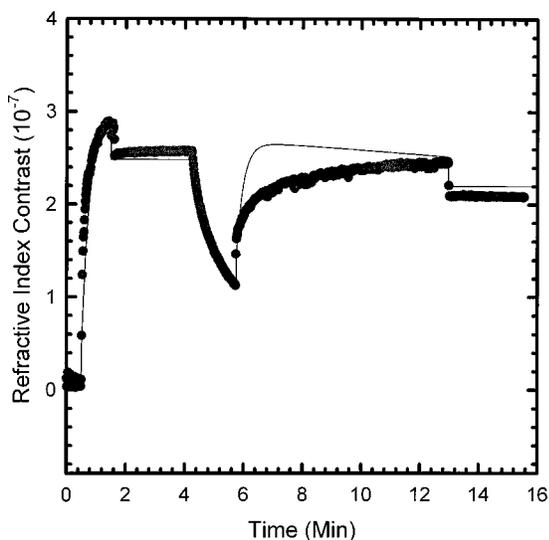


FIG. 5. Two write-block-erase cycles for sample B9 under the same conditions as in Fig. 3 for sample B10.

sized that the curve shown in Fig. 5 has only *one* free parameter—the strength of the transient (proportional to R_2^*). The agreement between model and experiment speaks for itself. Fitting parameters for the data in Figs. 4 and 5 are displayed in Table I.

Although the model has been formulated to include space-charge effects, an interesting feature of the fitting parameters is that the charge for the mobile modifiers q , here taken to be zero, must be much smaller than the elementary charge. This is because it is necessary to neutralize the space-charge field in order to obtain the observed grating strength and kinetic behavior. The choice of $q=0$ does not necessarily imply that the mobile modifiers are uncharged in their native state. There may be electrons in deep-donor states that can be excited into the conduction band by 465-nm light. A possible

candidate for such a deep donor would be a nonbridging oxygen that does not have a compensating cation among its immediate neighbors. Because the space-charge field tends to limit diffusion, it may be more difficult to write gratings at longer wavelengths where electrons cannot be excited into the conduction band of the glass. This could account for the observation of Broer, Bruce, and Grokiewicz¹⁰ that gratings could not be written in Er-sensitized glasses using resonant absorption at 980 nm.

The results of this model are in qualitative agreement with previously published observations of the grating kinetics.^{1–11} A quantitative comparison with these earlier data is not possible since they were not reported as refractive-index changes.

V. SUMMARY

It has been shown that the kinetics of the persistent gratings in rare-earth-sensitized silicate glasses are well accounted for by long-range diffusion of small modifiers mediated by the hot-phonon field resulting from nonradiative decay of the rare-earth sensitizers. The refractive-index contrast is due to the resulting modulation of the modifier concentration. This model is suggestive of ways in which the photorefractive efficiencies of these glasses might be improved through the increase of weakly bound modifiers or by the introduction of larger modifiers that tend to stretch the glass network.

ACKNOWLEDGMENTS

It is a pleasure to thank L. Pierre de Rochemont for supplying the samples used in this study. This material is based on work supported in part by the U.S. Army Research Office under Grant No. DAAH04-96-1-0322 and the National Science Foundation under Grant No. DMR-9705284.

¹E. G. Behrens, F. M. Durville, and R. C. Powell, *Opt. Lett.* **11**, 653 (1986).

²F. M. Durville, E. G. Behrens, and R. C. Powell, *Phys. Rev. B* **34**, 4213 (1986).

³F. M. Durville, E. G. Behrens, and R. C. Powell, *Phys. Rev. B* **35**, 4109 (1987).

⁴E. G. Behrens, F. M. Durville, and R. C. Powell, *Phys. Rev. B* **39**, 6076 (1989).

⁵R. C. Powell, E. G. Behrens, and F. M. Durville, *Mater. Sci. Forum* **50**, 115 (1989).

⁶E. G. Behrens, R. C. Powell, and D. H. Blackburn, *J. Opt. Soc. Am. B* **7**, 1437 (1990).

⁷E. G. Behrens, R. C. Powell, and D. H. Blackburn, *Appl. Opt.* **29**, 1619 (1990).

⁸V. A. French, R. C. Powell, D. H. Blackburn, and D. C. Cranmer, *J. Appl. Phys.* **69**, 913 (1991).

⁹M. M. Broer, A. J. Bruce, and W. H. Grokiewicz, *J. Lumin.* **53**, 15 (1992).

¹⁰M. M. Broer, A. J. Bruce, and W. H. Grokiewicz, *Phys. Rev. B* **45**, 7077 (1992).

¹¹A. Munoz, R. J. Reeves, B. Taheri, and R. C. Powell, *J. Chem. Phys.* **98**, 6083 (1993).

¹²A. A. Appen, *J. Anal. Chem. USSR* **24**, 1141 (1951).

¹³See, for example, T. J. Hall, R. Jaura, L. M. Connors, and P. D. Foote, *Prog. Quantum Electron.* **10**, 77 (1995).

¹⁴J. Feinberg, D. Heiman, A. R. Tanguay, Jr., and R. W. Hellwarth, *J. Appl. Phys.* **51**, 1297 (1981).

¹⁵N. V. Kukhtarev, V. B. Markov, S. G. Odulov, and V. L. Vinetskii, *Ferroelectrics* **22**, 949 (1979).

¹⁶A. Y. Hamad and J. P. Wicksted, *Opt. Commun.* **138**, 354 (1997).

¹⁷See, for example, S. H. Martin, and C. A. Angell, *J. Non-Cryst. Solids* **83**, 185 (1986).