Ultrafast thermal refractive nonlinearities in bistable interference filters

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A novel, angularly resolved excite-probe technique is used to discriminate ultrafast refractive and absorptive nonlinearities in interference filters and to determine the sign of the nonlinearity. The results explain observations of nanosecond switch-on and switch-off of optical bistability in ZnSe-based filters.

INTRODUCTION

Optical bistability in ZnSe-based interference filters has been studied by a number of groups.[1-16] The devices themselves continue to be used as a basis for prototype all-optical computing circuitry, taking advantage of the cascadability and logic-level restoration achievable, and of the 2-D uniformity.[17-27] It is well established that the origin of the bistability is the ZnSe refractive-index change induced thermally as a consequence of band-tail absorption at the operational wavelength; typically the 514-nm argon laser line and the 633-nm He–Ne laser line have been used.[5,8] Under circuit conditions, reported switching times are in the 10-μsec to 1-msec range, and corresponding power levels are in the range 100–1 mW.[28] Submilliwatt operation has been predicted for optimized cavity configurations.[29]

In the present work the ultimate limit to the optothermal switching time is investigated, namely, the time for carrier energy relaxation and subsequent lattice heating. The carrier lifetime also controls the magnitude of the optoelectronic nonlinear refraction. The results, obtained here for the thin-film material pertaining to conventionally grown interference filters, imply that it will not be possible to achieve optoelectronic switching in devices made of this nonepitaxial, poorly crystalline material.

Two experimental techniques are used. First, by monitoring the transmission of a cw beam, before and after a single 25-psec-duration pulse, thermally induced refractive effects of subnanosecond response times are resolved. Second, the probe transmission delay dependence, for excite-probe experiments on interference filters, shows that a refractive-index change of thermal origin is induced over a period of less than 20 psec.

NANOSECOND SWITCHING

The experimental arrangement for the present switching studies is shown in Fig. 1. Radiation at 633 nm, from a cw He–Ne laser, is incident upon a nonlinear interference filter. Picosecond pulses are provided from a passively mode-locked Nd:YAG system with Pockels cell single-pulse switch-out and amplification. Harmonic generation in KDP produces 15–30-psec pulses at 532-nm wavelength. The incident, transmitted, and/or reflected 633-nm radiation was monitored before and after each 532-nm pulse. The incident, transmitted, and reflected 532-nm pulse energies were also monitored.

Three samples were investigated. The first was a Fabry–Perot filter consisting of a ZnSe spacer of approximately 500-nm thickness, surrounded by reflective stacks, each containing four high–low index-layer pairs. This sample was fabricated by Optical Coatings Limited, Scotland, using conventional thermal-deposition techniques. A second sample had a 6.8-μm-thick spacer grown under ultrahigh-vacuum conditions, between conventionally deposited stacks. The spacer was deposited as a molecular beam nonepitaxial layer by K. Lewis, at the Royal Signals and Radar Establishment, Malvern.[8] Both samples have previously been studied for cw optical bistability at 633 nm.[6] The second sample was designed as a cavity of optimized absorption; it operates at lower switching powers and is considerably more stable to long-term drift than sample 1. A hysteresis loop stable over 5 h has been observed for sample 2.[30] The third sample was prepared by depositing a 15-nm-thick aluminum layer at the dielectric–air interface of a sample of the first structure. Use of this metal coating was originally designed in order to decrease the required power levels. Marginal reduction was obtained at 633 nm, but once again stability was improved. This structure is now commonly used at various wavelengths and is termed a beat structure (bistable étalon using absorbed transmission).[31]

Sample 1 above has a narrow bandpass in the 630-nm region and, oriented at 12° to the red He–Ne beam, gives the cw transmission response plotted in Fig. 2(a). There is an adjacent Fabry–Perot resonance peaked at 552 nm for normal incidence. By using an angle of incidence of 45° for the 532-nm pulses, we obtain the maximum fluence into the filter. A red-beam spot size ($1/e^2$ radius) of $\sim$10 μm was used. The green-beam spot size (r_0) used was 100 μm, so as to ensure good overlap and a uniform effect across the red spot. Initially the He–Ne power was set at $\sim$30 mW so as to give an output in the lower branch of the bistability region [position A in Fig. 2(a)]. Figure 2(b) shows the effect of a single green pulse introduced at time t =0. Within 1 nsec the red transmission increases and settles at level B, the upper bistable level. [The small oscillations shown in Fig. 2(b) are due to the presence of several axial modes in the He–Ne laser output.] The nanosecond result is noted to be the

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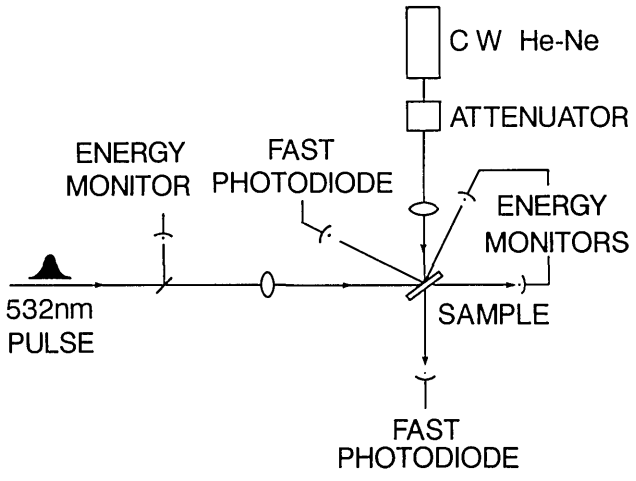

Fig. 1. Experimental configuration for fast optical switching.

limit of the detector response for the cw laser beam. The energy absorbed from the 532-nm pulse, in the spatial volume of the probing 633-nm beam, was of the order of 10 nJ.[32]

Figures 3(a) and 3(b) show the corresponding responses obtained if the transmission is set initially in the upper branch of the bistable loop (position B). This is achieved by raising the cw input beyond the switch-up point (to 36 mW) and then reducing it back to 30 mW; the output is then stable at B while only the cw beam is present. Nanosecond, detector-limited rapid reduction of the red-beam transmission is followed by recovery, on a microsecond time scale, to the initial level of the upper branch. The transmission spectrum of sample 2 contains many Fabry–Perot peaks in the visible; this fact and the slightly wedged nature of the spacer (due to the deposition configuration) allowed for normal incidence operation at 633 nm and near-normal incidence for the switching pulse at 532 nm. Once again nanosecond switch-down was observed from an initial setting in

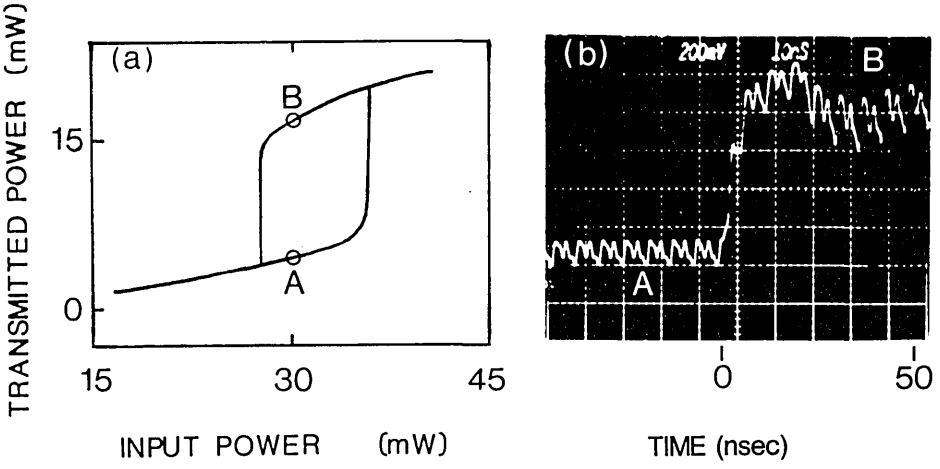

Fig. 2. (a) Nonlinear cw response at 633 nm for filter sample 1; (b) nanosecond switch-on from position A to position B, induced by a single 532-nm pulse.

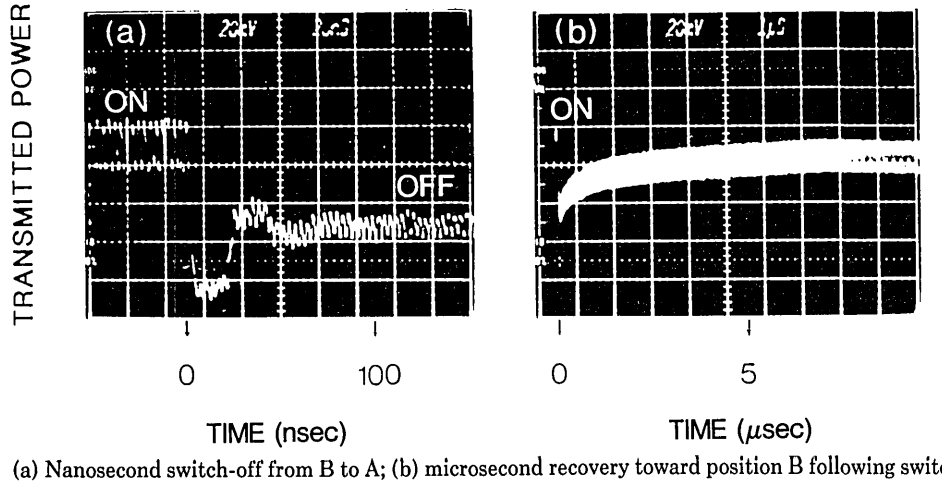

Fig. 3. (a) Nanosecond switch-off from B to A; (b) microsecond recovery toward position B following switch-off.

the upper bistable branch.[33] The improved stability of this sample also allowed us to confirm that the switching was in no way associated with response drift.

In the above filters forced switching relies on the availability of transmission resonances at both the holding (cw) and switching (picosecond) wavelengths. For the metallic-coated sample the restriction on the switch pulse is relaxed. Illumination was from the substrate side with the red, so that the cavity was effective, but directly onto the absorbing metal layer for the picosecond pulse. This device, best operated in the reflection mode, was switched by using picosecond pulses at arbitrary angles and at either 532-nm or 1.06-μm wavelength. The switching times were of the order of 500 nsec in all cases.

These switching experiments, on the uncoated samples, are similar to those undertaken by Bigot *et al.*[13–16] However, the latter authors used 308-nm, 15-nsec and 355-nm, 30-psec switching pulses, which are strongly absorbed in the filter-front-stack layers. Switch-on times of 2 and 150 nsec were observed. It is possible that the slower time scales, similar to those obtained with the metallic-coated sample, are associated with the need to conduct heat into the spacer region. In the present work the 532-nm radiation is absorbed primarily directly in the ZnSe spacer. The switching energies used here are at least an order of magnitude less than those used in Refs. 13–16 despite the similar switch-pulse spot sizes.

PICOSECOND LATTICE HEATING

Improved time resolution for the refractive nonlinearities was achieved by the following excite-probe technique. From a single 532-nm doubled-YAG pulse a small fraction (<3%) was split off and time-delayed by using a stepper-motor-controlled translation stage. The excitation beam was incident onto an interference filter at an angle $\theta_{ex} = \theta_m$ for which the transmission (and the internal absorption) was maximized. For excitation pulses of a specified energy the material excitation is thus always the same. This excitation will lead to absorptive and refractive effects on the probe beam. A refractive-index change at a given time after the excitation pulse produces a change in the spacer path length and therefore a displacement of the angle at which the filter transmission is a maximum. The presence of such a displacement may be detected by probing at incident angles to either side of θ_m. Suppose that an increase in transmission is observed for an angle $\theta_{p>} > \theta_m$, while for the same time delay the transmission at $\theta_{p<} < \theta_m$ decreases, then the angle at which the filter transmission is a maximum must have increased. Such a result identifies both the presence of a refractive effect and the sign of the index change. Nonlinear absorption increases (or decreases) would produce a decrease (increase) in the transmission at both $\theta_{p>}$ and $\theta_{p<}$.

Figure 4 shows the delay dependencies of the probe transmission for three incident probe angles (1) $\theta_{pm} = \theta_m$ (but in a different direction to the transmitted excitation in order to discriminate between the transmitted excitation and probe beams), (2) $\theta_{p<}$ chosen such that the linear transmission was 40% of that at θ_m, and (3) $\theta_{p>}$ also chosen for this transmission value but on the other side of θ_m.

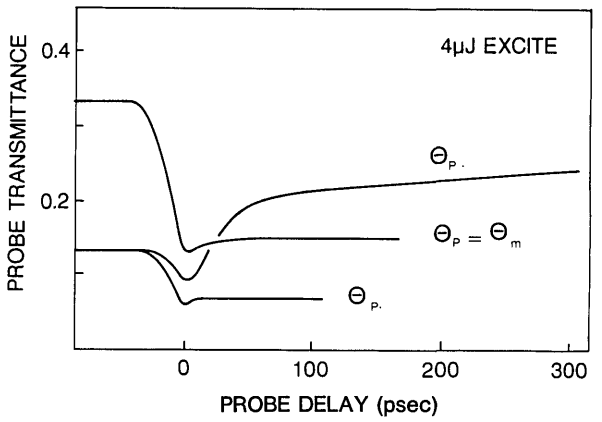

Fig. 4. Probe pulse transmission versus delay, following an excitation pulse of ~4-μJ energy. The initial probe transmission is set to be either on the Fabry–Perot peak ($\theta_p = \theta_m$) or near the half-maximum position $\theta_{p>}$, $\theta_{p<}$ (see text).

A range of excitation-pulse energies was found for which the transmission changes at $\theta_{p>}$ and $\theta_{p<}$ were indeed of opposite signs for both samples 1 and 2. Figure 4 refers to sample 1 experiments. For the probe at angle θ_{pm} then the transmission is reduced for short delays, when the excite and probe pulses overlap in time, and then increases gradually, but remains below the initial level even after 400 psec. A similar response is obtained for $\theta_{p<}$. In complete contrast, for $\theta_{p>}$ the initial reduction in transmission is followed by a rapid (20-psec) increase to a level well above that before the excitation and a continuing increase during the 400-psec observation time.

ANALYSIS

In the above experiments there is strong linear absorption, generating carriers into the ZnSe band tail ($\alpha \sim 10^3$ cm^{-1} at 532 nm). In addition, for pulse energies above 0.2 μJ we have observed nonlinear reflection and transmission; the sample absorptance increases by a factor of 2 for 4-μJ pulses. This nonlinear absorption is a combination of two-photon interband excitation and free-carrier absorption by the photogenerated carriers. The excited carriers lose energy by recovery to the band extrema and then by interband recombination. Both effects lead to an elevated temperature and an associated positive refractive-index change. The index change associated with the excited carriers themselves is negative. In other materials, e.g., InSb and GaAs, these opposing nonlinearities can lead to regenerative oscillations in the response of Fabry–Perot samples. That is, it is possible for carrier excitation to cause optoelectronic switching from low to high transmission followed by sample heating that causes reverse switching back to a low transmission.[34] In ZnSe the thermal effect is known to dominate cw switching. Nevertheless, the fast switch-down of Fig. 3(a) could possibly be interpreted to be a consequence of rapid generation of carriers, their recombination leading to slower heating and therefore explaining the gradual recovery to the high-transmission state.

The excite-probe technique provides the definitive experiment to show that induced carrier switching is not being

observed. All the above experiments, for time scales exceeding 20 psec, can be explained in terms of thermal-refractive-index effects alone.

The results shown in Fig. 4 are explained by the following: (1) A reduction of the cavity finesse of the interference filter during the excitation pulse. This is consistent with an increase in the absorption coefficient of the probe pulse. (2) For time delays above 20 psec the angular dependence of the probe response indicates a movement of the transmission peak to larger angles. The phase thickness of the spacer, to the probe beam, is $nD\cos\theta_p$; this is equal to a half-integer multiple of the vacuum wavelength at the transmission peak. Hence an increase in the θ_p value at the peak implies an increase in the refractive index, n. In turn this indicates an optothermal effect. (3) The longer-term (400-psec) gradual changes in the probe transmissions are consistent with a gradual cooling, based on the assumption that the 4-μJ pulse has driven the transmission peak slightly beyond $\theta_{p>}$. This was confirmed by the use of different excite-pulse energies. For 2 μJ the probe transmission rises, but not so high as at 4 μJ; for greater than 4 μJ the transmission rise is lower and changes to a decrease beyond 10 μJ. This latter we attribute to driving the low-angle half-maximum point beyond $\theta_{p>}$. We estimate that a refractive-index change of 3×10^{-2} is required in order to drive the transmission peak through $\theta_{p>}(2n\Delta n = \sin^2\theta_m\Delta\theta)$. Given that the thermo-optic coefficient of the ZnSe is of the order of 1.5×10^{-4} K^{-1} (Ref. 8) the required temperature rise is 200 K. An energy relaxation of a few microjoules in the irradiated volume will produce such a rise.

A simplification of the analysis of Bigot *et al.*[14] and Grun[16] provides a qualitative explanation of the present switching observations. We ignore spatial gradients, the laser radiation is taken to be plane-wave, and the thermal diffusion is characterized by an effective relaxation time (τ). The response to the cw monitoring beam is then expressible as

$$\frac{d\Delta T}{dt} = \frac{aI_0}{1 + F\sin^2(\phi_0 + b\Delta T)} - \frac{\Delta T}{\tau}. \tag{1}$$

This equation describes the rate of temperature change at any given instantaneous value of the temperature. The coefficient a contains reflectivity and absorption factors but is only weakly temperature dependent, as is the finesse factor F. b is the temperature coefficient of the cavity phase, proportional to the thermo-optic coefficient, and ϕ_0 is the phase at the ambient temperature. Figure 5 shows the radiative driving force and the thermal heat sink terms on the right-hand side of Eq. (1), plotted as functions of ΔT. The points of intersection of the Airy function and the load line give the steady-state values of ΔT. The corresponding steady-state filter transmissions are directly proportional to the vertical coordinate at the intersections. The unstable nature of the intermediate steady-state (point C) is illustrated directly from consideration of a small increase of ΔT with respect to the steady-state value. Under such a perturbation the temperature driving force exceeds the sink and ΔT will move toward the upper stable point, B. This and the solution at A are the upper and lower bistable branches for the particular I_0 value, corresponding to A and B in Fig. 2(a).

Given the conclusions from the excite-probe measurements, then for the switching experiments the sample experiences an increase in temperature, ΔT_1, that is instantaneous on the detector-resolution time scale:

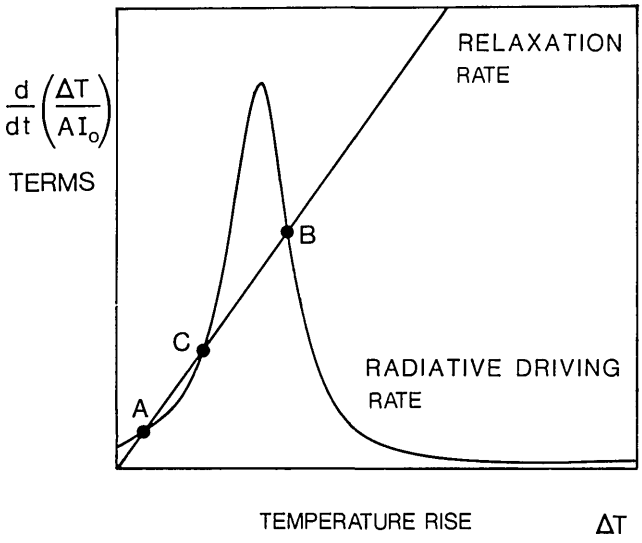

Fig. 5. Schematic of terms in the filter dynamics [see Eq. (1) and associated text].

$$\Delta T_1 \sim \Delta E/(\rho C_p \pi r_0^2 D). \tag{2}$$

Here ΔE is the energy absorbed from the switching pulse, D is the thickness of the absorbing region of density ρ and specific heat C_p, r_0 is the $1/e^2$ spot radius of the pulsed radiation. Estimates of a 50–200-K temperature rise in the experiments are consistent with requirements for switching.[6] Hence, from position A the transmission will increase suddenly, provided that $\Delta T_1 \simeq \Delta T_B$, i.e., for a range of pulse energies. For large pulse energies the transmission may decrease initially, followed by a gradual increase as the temperature recovers to the stable point ΔT_B, where it is maintained by the absorption of the cw beam. This explains the observed rapid switch-on, achieved for pulse energies in the range 10–20 μJ (for sample 1).

Similarly, starting from position B, the 532-nm pulse will produce an instantaneous reduction in transmission associated with the increase in ΔT, followed by recovery to the initial level on the cooling time scale (Fig. 3). Immediately following the switching pulse the high-temperature region of the filter is essentially confined within the spacer. The cooling time therefore has an initial lower limit value of the order of $\tau = \rho C_p D^2 \kappa_s^{-1}/16$, where κ_s is the substrate conductivity. The calculated value of τ is 0.5 μsec in sample 1 and 80 μsec in sample 2. These values explain the observed experimental recovery times following switch-off. As one approaches the steady-state solutions, however, the effective time constant approaches the far slower value of $\rho C_p r_1^2 \kappa_s^{-1}/4$, where r_1 is the cw beam-spot radius. Finally we note that because the cavity response is influenced primarily by spacer heating, then for the aluminum-coated sample thermal conductivity from the metal through the reflective stack is necessary. This explains the 500-nsec observed switching times, as opposed to instantaneous (i.e., subnanosecond) switching.

DISCUSSION

Using a variation of the excite-probe technique we determine the carrier energy relaxation time of the thin-film ZnSe in interference filters to be less than 20 psec. This result is attributed to an intraband relaxation of carriers excited high in the bands and to rapid trapping or interband recombination.[34] The latter we associate with the microcrystalline nature of the material in the filters and the resultant rapid recombination at the grain surfaces. It is noted that no photoluminescence signals are detectable for the ultrahigh-vacuum-grown material[35]; this would suggest the dominance of trapping. By contrast recent time-resolved four-wave mixing experiments in polycrystalline bulk ZnSe indicate nanosecond-time-scale recombination.[36] The grain size is considerably larger in the bulk material, and the band edge is dramatically sharper.[19] As a consequence of the rapid energy relaxation, 20-psec optothermal switching is in principle possible (at high powers) in the filters. Optoelectronic nonlinearities, associated with the photoexcited carriers themselves, are commensurately unlikely to be observed. At the carrier concentrations necessary for optoelectronic switching there is strong nonlinear absorption and associated rapid energy relaxation and lattice heating. In further experiments, no energy dependence of the excitation pulse transmission peak angle (θ_m) was observed, presumably owing to the competition between carrier and thermal effects during the pulse itself.

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