# Single-step switching at a nonlinear interface

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Theoretical predictions of the behavior of a light beam crossing a plane interface between a linear and nonlinear dielectric medium indicate staircased multiple threshold switching in nondiffusive media when an incident Gaussian beam instead of a plane wave is assumed. We report experimental findings showing unexpected high-contrast single-threshold switching, which may have important implications for optical signal processing. No evidence was found for the existence of bistable behavior indicated by previous experiments.

The nonlinear interface (NI) is commonly known in the literature as a plane interface between an ordinary dielectric and a nonlinear dielectric material having an intensity-dependent refractive index. The first theoretical discussions of such an interface were carried out in the late 1970s investigating the intensity dependence of the reflectivity of plane waves incident on the NI (see Ref. 1 and references therein). These studies predicted that under appropriate conditions the reflectivity of an NI would not only be intensity dependent, but would exhibit optical hysteresis resulting in bistability. The highly nonlinear behavior suggested possible application[1] of the NI for (ultrafast) optical switches, scanners, logical elements,[2] etc.

In spite of the potential importance of the NI for compact all-optical digital processors,[3] essentially only three experimental investigations of the NI have been published to date: a pulsed experiment[4,5] using a $CS_2$-glass interface, and two cw experiments using a LiF-liquid[6] (small quartz particles suspended in water) or glass-liquid[7] (polystyrene spheres in water) interface. The first two experiments investigated intensity dependence of NI reflectivity, while the third studied the reflected beam profile.

It is our belief that the reported low (100–80% or 70%) switching contrast[4–6] (maximum reflectivity change between low and high intensity) and multiple thresholds (indicating beam breakup) discouraged any further experimental investigation of the NI. In addition, the experimental results[6] seemed to agree qualitatively with the predictions of a two-dimensional diffusionless NI model[1] for an incident Gaussian beam, implying that realistic NI applications would, at best, be severely limited.

The purpose of this paper is to show that the NI can have, for certain types of materials, fundamentally different behavior than previously thought. The first step in this direction was taken by Cuykendall and Andersen[2,8] introducing a diffusion mechanism in the two-dimensional NI model which delocalized the induced index change. This resulted in a more plane-wave-like behavior preventing the transmitted beam from breakup into multiple channels, thereby significantly extending the potential range of NI application. Based on that indicated behavior, an optical integration architecture was developed[3,9] (with an ideal NI as the single computing element) in order to assess the possible worth of such a device for optical computing.

Stimulated by these results, we repeated the Smith-Tomlinson experiment[6] under more controlled and slightly different conditions. Our experimental setup is shown in Fig. 1. A $p$-polarized output of a cw laser (514.5 nm) was focused to a theoretical $1/e$ amplitude radius $w_0 \approx 5$ $\mu$m at the interface with a far-field diffraction angle $\Psi_D \approx 1.3°$. The beam was attenuated by a $\lambda/2$-plate polarizer combination with a maximum rotation speed of 45°/111 sec. Roughly 5% of the incident beam was reflected to a power meter ($I$) while an identical power meter ($R$) measured the intensity of the beam after reflection at the interface. Both power meters were triggered simultaneously and read by computer each second. The linear medium was an optically polished $6\times1\times1$-$cm^3$ single crystal of LiF with a refractive index $n_0=1.391$. It was mounted horizontally in a 360° rotator allowing a relative (absolute) angle determination of 0.002° ( <0.3°). The nonlinear medium consisted of an aqueous suspension of 0.277-$\mu$m polystyrene spheres[10] in a volume concentration of 7.7% with a calculated[11] effective nonlinear Kerr coefficient $n_2 \approx 0.44$ $cm^2$/MW and low-intensity refractive index $n_0-\Delta \approx 1.353$. Thus the critical glancing angle for total internal reflection is $\Psi_{crit} \approx 13.4°$.

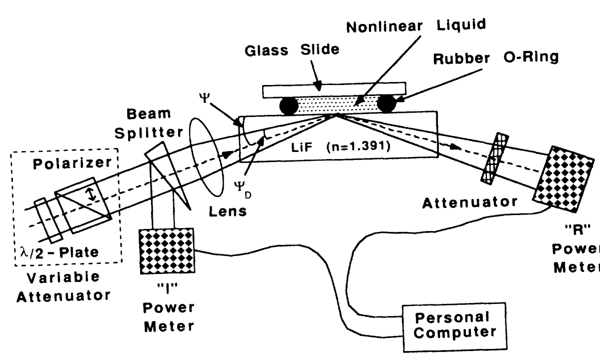


FIG. 1. Experimental setup.

 

In addition to polystyrene and water, the nonlinear liquid contained controlled amounts of surfactant and was saturated with LiF. The saturation avoided etching of the LiF crystal surface with water, so that the reflected intensity no longer decreased with time due to increasing surface roughness. Exposure of the LiF crystal to the nonlinear liquid for as long as 5 h did not cause any visible damage to the crystal surfaces. However, the switching contrast still decayed somewhat with time. Furthermore, we measured an absolute reflectivity and slope decrease with time (due to laser-induced particle aggregation and/or deposition at the interface). Thus the LiF crystal was repolished every time it was exposed to the nonlinear liquid to achieve identical starting points.

Unfortunately, when very small particles come too close together, they tend to agglomerate, irreversibly changing such nonlinear particle suspensions. This can happen spontaneously through Brownian diffusion, or be induced by a light intensity gradient, and is more likely to happen near a solid surface. In order to influence this phenomenon, we added different amounts of the nonionic surfactant (Triton X-100) to the pure polystyrene suspension. Without surfactant the latex globally flocculated into macroscopic aggregates due to the presence of ions from dissolved LiF. Adding surfactant prevented flocculation through stabilization by hydrophilic coating. It was found that the presence of surfactant strongly influenced the intensity dependence of the reflectivity. This can be seen clearly in Fig. 2, which shows measured intensity dependencies of the NI reflectivity for an incident glancing angle of $\Psi=7.3°$. Each curve is normalized against the low-intensity value for easier comparison. Calculated values for $w_0$, $n_2$, and $\Delta$ were used to convert from incident power to normalized intensity $n_2 I_{max}/\Delta$. Figures 2(a)–2(c) show typical curves obtained without surfactant. The measured intensity dependence of the NI reflectivity was also affected by the local condition of the flocculated nonlinear medium. Small translations of the laser focus along the interface result in significantly different behavior [see Figs. 2(a)–2(c)], and are therefore nearly impossible to reproduce. But in all cases we observed a small switching contrast. However, high contrast (roughly 3 to 1) was obtained with a concentration of $\approx 0.5\%$ surfactant and is shown in Fig. 2(d). The initial experiments showed some hysteresis, but after improving the experimental setup and NI preparation, no hysteresis could be found in the explored parameter region. [See, for example, the curves in Fig. 2 which contain several intensity cycles (low-high-low) for each case.]

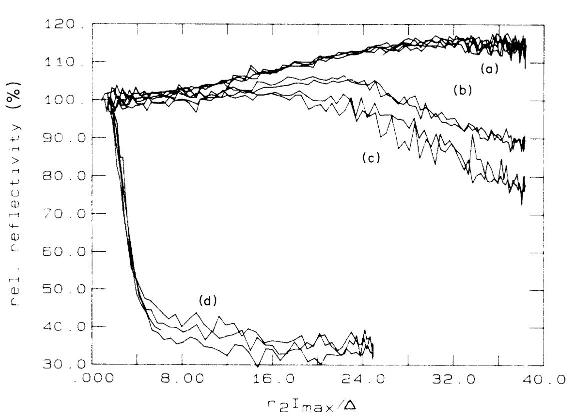


FIG. 2. Measured intensity dependence of the relative reflectivity of the NI for an incident glancing angle $\Psi=7.3°$: (a)–(c) typical curves obtained without surfactant showing sensitivity to local agglomerate variation in the nonlinear medium; (d) observed switching result with roughly 0.5% surfactant.

Our results differ significantly from those found earlier for a similar experiment,[6] where roughly four-times smaller quartz particles in a three-times higher volume concentration were used. The authors reported multiple thresholds with low switching contrast (100–70%) and observed transient bistable behavior. Using that same suspension we were not able to reproduce the multiple threshold results of Ref. 6, and attained a more than 10% reflectivity decrease only by using a one-year-old (partially diffusion agglomerated) quartz sphere suspension. Moreover, we observed a strong, nearly irreproducible time dependence of the reflectivity, especially at high intensities, suggesting that laser-induced particle aggregation or deposition influenced the data, making them hard to interpret. Since much better results were obtained with the polystyrene spheres once we learned how to stablize the interface and influence the switching behavior, we did not further pursue the quartz sphere experiments.

For comparison, we consider briefly the plane wave and standard (diffusionless) two-dimensional Gaussian model predictions. The nonlinear refractive index is assumed to have the usual phenomenological Kerr dependence

$$n=n_0-\Delta+n_2 I , \tag{1}$$

where $n_0$ is the index of refraction in the linear medium, $\Delta$ is a small positive refractive-index offset, $n_2(>0)$ is the optical Kerr constant, and $I$ is the local intensity of the light. Since these models and their associated predictions have already been extensively discussed,[1,2,12] we present here only those results relevant for the parameter choices in this paper. Figure 3 shows the plane-wave prediction for an incident glancing angle $\Psi=0.55\Psi_{crit}$, where $\Psi_{crit}$ is the critical glancing angle for low intensity. Note the predicted bistable behavior when one changes from low to high and back to low intensity $(1\rightarrow2\rightarrow3)$. Figures 4 and 5(a) show the CRAY X-MP/48 computer results obtained from the standard two-dimensional model with $n_0=1.391$ for an incident Gaussian beam having an intensity $I_{max}$ and a minimum $1/e$ amplitude radius of $w_0=10\lambda=10\ \mu m$ for the focus at the interface (neglecting reflections at the interface). The respective index offsets are $\Delta=0.038$ (Fig. 4) and $\Delta=0.01$ [Fig. 5(a)]. The ratio $\Psi/\Psi_{crit}$ is identical (0.55) in Figs. 2–5. The computer program required excessive run time (resolution) for the relatively large offset $\Delta=0.038$ case at intensities higher than shown in Fig. 4. Nevertheless, the general trend can be seen in Fig. 5, since at constant $\Psi/\Psi_{crit}$ the "average" reflectivity versus normalized intensity $n_2 I/\Delta$ is, to first approximation, independent of $\Delta$ for the same

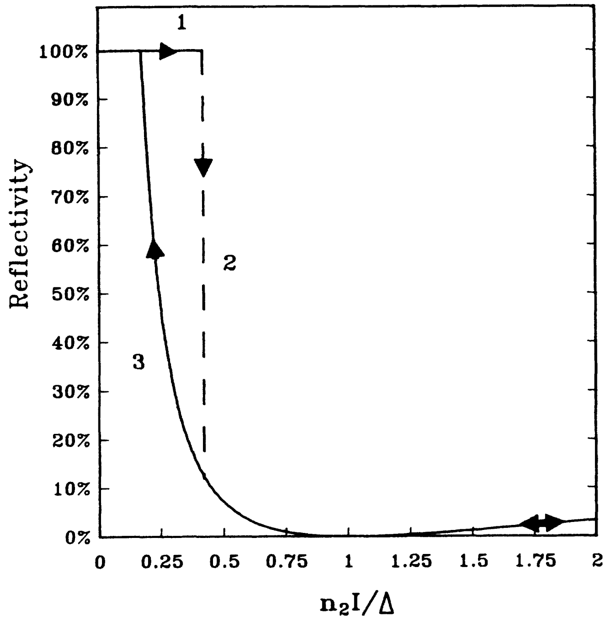


FIG. 3. Intensity dependence of NI reflectivity in the case of an incident plane wave for $\Psi/\Psi_{crit}=0.55$.

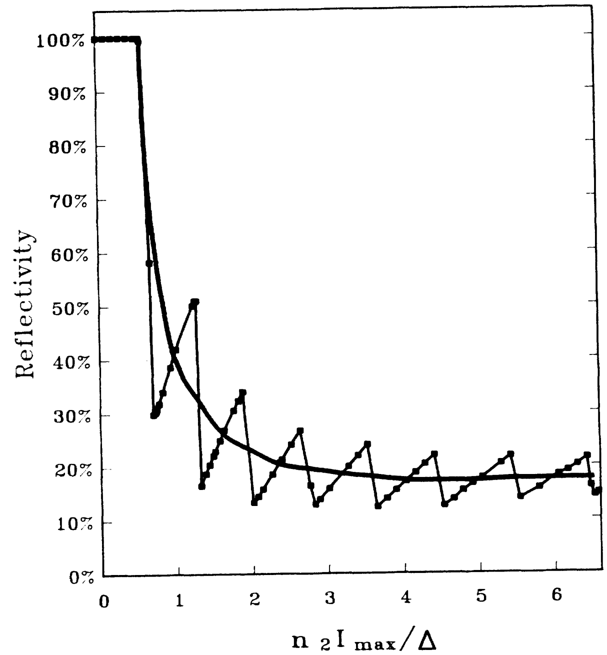


FIG. 5. (a) Diffusionless two-dimensional NI model prediction for $\Psi=3.8°$, $n_0=1.391$ $\Delta=0.01$, and $w_0=10$, $\lambda=10$ $\mu$m; and (b) smoothed reflectivity dependence (thick line).

focusing conditions. Although the local "steps" are more pronounced for a lower offset $\Delta$, the average path through the steps [see Fig. 5(b)] in both Figs. 4 and 5(a) are alike.

Comparing Figs. 3, 4, 5, and 2(d) one can classify the plane-wave behavior, the standard (diffusionless) two-dimensional Gaussian model prediction, and the experimental data. The plane-wave theory entails a single threshold, very high switching contrast ($100\rightarrow0$ %), and optical bistability. The standard Gaussian model predicts multiple thresholds related to the formation of additional self-focused transmitted channels, slightly increased threshold intensity, and much lower contrast ratio for the first step. No statement can be made regarding bistability, since without refractive-index memory the program effectively models only increasing intensity (see Fig. 3, $1\rightarrow2$). The experimental reflectivity, on the other hand, stabilizes after the first threshold, showing no sign of additional steps, has a significantly higher threshold intensity than the Gaussian model predicts, and shows no hysteresis for intensity changes which are much slower than the recovery time of the nonlinear medium ($\approx 10$ s). Approximate agreement between the experiment and Gaussian model can be achieved by smoothing out the local reflectivity jumps [see Fig. 5(b)] and rescaling the normalized intensity by a factor of roughly 4.

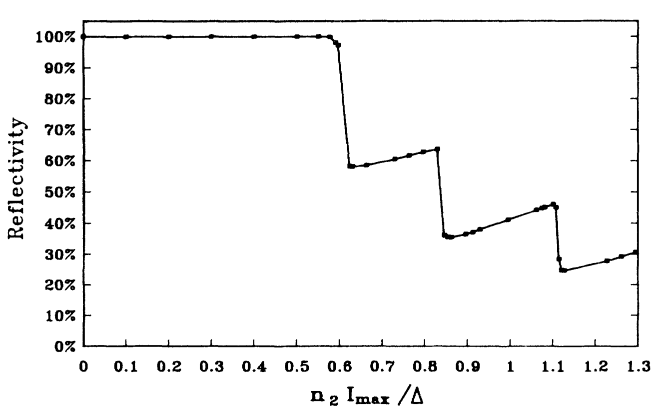


FIG. 4. Diffusionless two-dimensional NI model prediction for an incident Gaussian beam at glancing angle $\Psi=7.3°$, $n_0=1.391$, $\Delta=0.038$, and $w_0=10$, $\lambda=10$ $\mu$m.

In order to understand what is causing the experimental results to deviate from the Gaussian model and approach plane-wave-like behavior, we briefly analyze the particular nonlinear medium. A very simple model describing the nonlinear behavior of these suspensions of dielectric spheres in an electromagnetic field gradient has been given.[6,11] In short, the spheres experience a force which attracts them in the high-intensity region, changing therefore the particle density locally, resulting in a refractive-index change. These light forces are opposed by thermal diffusion (Brownian motion) of the particles. As the light-induced force on the spheres is proportional to the gradient of a field, it can be expressed in terms of a potential $\phi$. The quasistationary equilibrium sphere-density distribution caused by this potential can be found from a standard diffusion model in which the sphere density is proportional to $\exp(-\phi/kT)$. Assuming the validity of this model we find that for a not-too-intense light beam ($\phi_{max} \lesssim kT$) the induced density distribution is only negligibly sharper than the intensity profile. Under such conditions the thermal diffusion counterbalances the optical pressure, resulting in a density distribution which follows the intensity variation. The effect of Brownian diffusion in this particular system is thus to simulate a diffusionless Kerr-like nonlinearity, where the local induced index change varies directly as the local intensity, and Eq. (1) remains valid.

This simple model neglects, among other effects, the actual sphere size ($\approx\lambda/2$), the agglomeration and the Coulomb repulsion (attraction) due to surface charges (induced dipole moment), and dissolved ions. Developing a model which includes these effects is beyond the scope of this paper. However, it is clear that the spheres cannot be packed more densely than hexagonal or cubic close packing permits. This gives an upper limit of $3.7\Delta$ for the induced index change in the polystyrene water suspension. Long before that limit is reached the increased viscosity and Coulomb repulsion will slow down the response by saturating the nonlinear index, thus deviating from Eq. (1). We believe that such a saturation process is primarily responsible for washing out the multiple thresholds by preventing an overshooting of the desired index change $\Delta$.

More than one factor is probably responsible for the higher experimental threshold in Fig. 2(d) (not observed in Ref. 6): (i) the spatial profile of our laser was not perfectly Gaussian, precluding exact calculation of the spot size at the interface; (ii) the polystyrene spheres have a surface charge of roughly 1.82 $\mu C/cm^2$, causing a smaller effective nonlinear Kerr coefficient and saturation; and (iii) the effect of the surfactant on the Kerr coefficient is not yet fully understood, since it is still unclear if it influences only the laser-induced local particle aggregation or deposition, or if it also changes the refractive-index response. Although the effective offset $\Delta$ is probably slightly smaller than calculated due to a local sphere-density gradient near the solid LiF surface, this would only partially compensate for the above effects.

We have demonstrated that under certain conditions a nonlinear interface can have much higher switching contrast and more plane-wave-like behavior (single threshold) than previous experiment and theory have indicated. Furthermore, we have seen that the present two-dimensional Gaussian model needs corrections to describe the new experimental results. A three-dimensional model might be necessary to correctly predict the behavior of the NI. However, a more likely explanation for the observed differences is that the nonlinear medium is not behaving strictly as the simple description[6,11] predicts. Saturation and other effects (such as diffusion, deposition, convection, etc.) may completely change the switching behavior by deviating from the linear Kerr dependence at higher intensities. If it can be confirmed[13] that saturation of the nonlinear refractive index is the main source for the observed plane-wave-like behavior of the NI, it would have significant impact on the potential for NI application in ultrafast optical signal processing, since most saturation effects are virtually instantaneous. There is a good possibility that better understanding of the various influences may also lead to a novel use of the NI to measure ultrafast phenomena. This seems to be one of the rare cases where reality shows better results than predictions made from simple models, leaving hope that more is to come.

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[1] W. J. Tomlinson, J. P. Gordon, P. W. Smith, and A. E. Kaplan, Appl. Opt. **21**, 2041 (1982).

[2] R. Cuykendall, Appl. Opt. **27**, 1772 (1988).

[3] R. Cuykendall and K. H. Strobl, J. Opt. Soc. Am. B **6**, 877 (1989).

[4] P. W. Smith, J.-P. Hermann, W. J. Tomlinson, and P. J. Maloney, Appl. Phys. Lett. **35**, 846 (1979).

[5] P. W. Smith, W. J. Tomlinson, P. J. Maloney, and J.-P. Hermann, IEEE J. Quantum Electron **QE-17**, 340 (1981).

[6] P. W. Smith and W. J. Tomlinson, IEEE J. Quantum Electron. **QE-20**, 30 (1984).

[7] G. Delfino and P. Mormile, Opt. Lett. **10**, 618 (1985).

[8] R. Cuykendall and D. Andersen, Opt. Lett. **12**, 542 (1987).

[9] K. H. Strobl and R. Cuykendall, Appl. Opt. (to be published).

[10] Surfactant-free product of Interfacial Dynamics Corp.

[11] P. W. Smith, P. J. Maloney, and A. Ashkin, Opt. Lett. **7**, 347 (1982).

[12] A. E. Kaplan, Pis'ma Zh. Eksp. Teor. Fiz. **24**, 132 (1976) [JETP Lett. **24**, 114 (1976)]; Zh. Eksp. Teor. Fiz. **72**, 1710 (1977) [Sov. Phys.—JETP **45**, 896 (1977)].

[13] We are presently conducting experimental and theoretical investigations in order to determine the influence of saturation on nonlinear interface switching behavior. (*Note added in proof.* Numerical confirmation has recently been achieved as well as further experimental evidence.)