Stable Hysteretic Reflection of Light at a Nonlinear Interface

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Abstract. We have observed a hysteretic (bistable) reflection of a Gaussian laser beam at the interface between glass and a liquid solution of polystyrene microspheres as nonlinear medium with both branches of the hysteretic curve being stable, in contrast to the results of previous experiments.

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Nonlinear reflection of light at the interface between two dielectrics, one of which is linear and the other having an intensity-dependent index of refraction (the so-called nonlinear interface), has lately attracted a great deal of attention [1–8]. Since switching devices based on nonlinear interface effects do not include the Fabry-Perot resonator typical for most of the other optical bistable devices [9], they have potential [1] to become very fast logic elements and to use a light source with a broad spectrum. The first experimental results [2] using CS₂ as nonlinear material, clearly displayed the threshold behaviour predicted by analytic plane-wave theory [1] and demonstrated certain evidence of bistability also predicted in [1].

Since reflectivity experiments at nonlinear interfaces inevitably involve laser beams with limited cross-sections which therefore differ from the ideal plane-wave assumed in [1], various numerical simulations with one-dimensional limited Gaussian input beam have been reported with surprisingly contradictory results. Some of them ([3] and to some extent [4]) endorsed the existence of bistability for the Gaussian beam case while others [5] disputed it. The modeling study [5] also showed that the reflectivity exhibits not a single threshold, but a series of thresholds each of which is associated with the formation of a new self-trapping channel propagating in the nonlinear material away from the interface.

In order to resolve this controversy, a new experiment [6] was done using an "artificial" nonlinear medium [10] (an aqueous suspension of quartz microspheres) with a very large effective nonlinearity (but a slow response time). This made it possible to use a cw laser with a much better time resolution. The results [6] clearly displayed two thresholds as was expected from computer simulation [5]. However, when the intensity of the incident beam was swept downward through a threshold value, the hysteresis was observed; although the high-transmission branch of the hysteretic curve was found to be unstable and therefore the whole effect was reported as a non-bistable one. The high-transmission state persisted on the order of 300 times the response time of the nonlinearity. This quasi-bistability received no explanation. Results of a later experiment [7] were consistent with the hypothesis of bistability at the interface, although this experiment had a drawback in the shortness of the pulses used.

In this paper we report the results of an experiment essentially similar to that of [6], (and to some extent to [8]). In this new experiment, by varying the concentration and radii of the polystyrene microspheres (latex), and substituting ethanol for water (the liquid with which latex is normally supplied), we have found hysteresis with both branches of the hysteretic curve being stable over a long period of time (on the order of 2–3 min).
It is known that a particle with radius $r$ and refractive index $n_p$ immersed in a surrounding liquid with different refractive index $n_a$ moves, in the presence of an electromagnetic field, towards the high-field region (if $n_p > n_a$). This leads to a change in the optical density and (in a certain approximation) results in a Kerr-like nonlinearity, $\Delta \approx n_z I$, where $I$ is the intensity of the light in the medium, with $[10, 11]$:

$$n_z = 8 \pi^2 n_p^2 r^8 N (m - 1) (m^2 - 1) / 3 c k_0 (m^2 + 2),$$  

(1)

where $c$ is the velocity of light, $k$ is Boltzmann's constant, $\theta$ the absolute temperature, $N$ is the number of microspheres per unit volume, and $m = n_p / n_a$. In our case the nonlinearity coefficient $n_z$ is positive and very high ($\approx 10^3$ times that of $\text{CS}_2$).

Assume now that a light beam is incident from a linear medium with refractive index $n_0$ at an angle $\psi$ upon the boundary of nonlinear medium with slightly mismatched refractive index $n$, which depends on the beam intensity $[1]$:

$$n = n_0 - \Delta + n_z I,$$

(2)

where $\Delta$ is a small positive mismatch between the two indices at low intensity. If the incidence angle $\psi$ is less than the critical angle $\psi_c$, with $\psi_c = \arccos(n/n_0) \geq (2 \Delta / n_0)^{1/2}$, the beam will undergo total internal reflection (TIR) at the interface. In this case, we have no transmission, and in the nonlinear medium there is an evanescent field $[12]$. As the evanescent field increases, due to the positive Kerr constant the effective critical angle decreases which, in turn, gives rise to an increase in the evanescent field, closing the positive feedback loop. There is a threshold intensity at which a sudden jump from the TIR state to the transmission state occurs. Therefore, by sweeping the input intensity through the threshold, it is possible to drive the switching device from one state to another. When analyzing the behaviour of a Gaussian beam in TIR, one should also take into account the Goos-Hanchen effect $[13]$. This effect consists in a spatial shift in a light beam when it is totally reflected at the interface (Fig. 1) $[8]$, with the shift given by

$$\Delta x = \frac{\cot \psi |T|^2 \lambda}{2 \beta},$$

(3)

where $T$ is the transmission Fresnel coefficient, $\lambda$ is the wavelength of the incident light and $\beta = (n_p \cos \psi - 1)^{1/2}$. Since the optimal experimental conditions occur at the maximum Goos-Hanchen shift, we used this parameter as a good reference point in the experiment.

The experiment is divided into two steps. In the first, we maximize the Goos-Hanchen shift (the results of this experiment will be published later). Modifying the experimental setup we subsequently analyze the reflectivity of the system versus the incident intensity. In order to enhance the Goos-Hanchen shift detection, we made use of an optical lever; to obtain a scanning on the phototube, we employed a mirror rotating in front of it, which transformed the spatial separation into a temporal separation. In this way, the phototube receives first the totally reflected beam and then the shifted wave (Fig. 2). The experimental results, obtained in this first part, confirm, as expected, the existence of a shifted optical field in the TIR regime, and the dependence of the evanescent field on the angle of incidence. In order to perform studies on the reflectivity, we developed the second experimental step using the setup shown in Fig. 3. The Pockels cell,
driven by a function generator, modulated the intensity of the laser beam. As a linear medium, we utilized a lithium fluoride (LiF) prism (as in [6]), whereas the nonlinear medium was a suspension of polystyrene microspheres (refractive index \( n = 1.59 \)) in ethanol. The value of the refractive index for LiF at 5145 Å is \( n = 1.3910 \) and for ethanol \( n = 1.3737 \). The microspheres (latex) are supplied in an aqueous solution. In order to obtain a good index matching, we substituted ethanol for water in order to raise the low-intensity effective refractive index of the nonlinear medium. We dehydrated the microsphere solution and added the quantity of ethanol needed to obtain the desired concentration of particles. This allowed us to analyze the reflectivity as a function of the physical parameters of the medium, and to determine experimentally the response of the device as a function of the nonlinear coefficient \( n_2 \). This was accomplished by shining an Ar cw laser beam (5145 Å) at an angle smaller than the critical angle, upon the interface. By recording the intensity of the reflected beam as the angle of incidence increases, we obtained a plot of the reflectivity vs. angle of incidence (similar to [6]) from which we determined the critical angle itself. Under the cited conditions the angle \( \gamma \) was 90°. Fixing the apparatus at this angle we then varied the incident power in order to obtain the reflectivity curve as a function of the incident intensity.

The results of this measurement for increasing incident intensity are displayed in Fig. 4 where the continuous curve is a fit to the experimental points. The main feature here is the appearance of two shoulders in the reflectivity, which is in a qualitative agreement with [5, 6]. Looking for bistability, we plotted the output power versus the input power for the increasing-decreasing power intensity cycle. In our first attempts, we found no bistability. Varying the concentration in the range 0.0025 to 0.5 in the radii of the microspheres from 0.0865 to 0.265 μm, we obtained hysteresis for the conditions reported in Fig. 5. The experimental points reported there were obtained using two power meters as photodetectors to plot values of the output versus the input power. The hysteresis behaviour was detectable in a small range (± 20%) around 0.1915 μm for the microsphere radius and 0.025 for concentration. Under the cited conditions, the nonlinear coefficient \( n_2 \) was 2.42 \( \times 10^{-2} \) cm²/MW. The curve shown in Fig. 5 was obtained with a total of 20 min of scanning time, and displays a pronounced hysteresis which would have disappeared if either of the two states had been unstable.

At this stage, it would be premature to draw firm conclusions as to the nature of the mechanism giving rise to the stable hysteresis observed here at nonlinear interfaces. We believe, however, that a possible explana-

![Fig. 4. Experimental measurement of the reflectivity versus (normalized) incident intensity power for \( \Psi = 9^\circ \)](image)

![Fig. 5. Diagram of output versus incident power obtained by plotting the responses of two power meters](image)
linear interaction (i.e. the dependence of the refractive index not only on the local field but also on the fields in the neighboring area). Comparing our experiment with that of [6], we note that this suggestion is consistent with the variation of parameters used by us to obtain the desired effect, since all of them — viscosity of liquid, concentration and radii of microspheres — significantly affect the diffusion of particles. This new factor (which could also be valid for some other, much faster mechanisms, like diffusion of carriers in some semiconductors [14]) may result in significant change of the near-interface processes. In addition to true bistability, it could also give rise to a long-propagating nonlinear surface wave [15] prohibited at a "purely" Kerr-nonlinear interface [5, 16].

In conclusion, we have observed stable hysteretic reflection of light at a glass-latex nonlinear interface, which demonstrates the possibility of cw bistability at nonlinear interfaces and indicates the importance of the fine details of the nonlinear mechanism.

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