Observation of Biexcitonic Optical Bistability and Optical Limiting in CuCl

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Experimental evidence for the occurrence of optical bistability using the biexciton two-photon resonance in CuCl is presented. These observations agree with recent theoretical predictions that require an intensity of 10 MW/cm^2 and 10-μm-thick etalons with 90% reflective coatings. Earlier much less demanding predictions neglected two important physical effects: unsaturable background absorption at the biexciton from the tail of the exciton resonance and broadening of the biexciton resonance with intensity.

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It is well established that biexcitons, which are the bound state of two excitons, can be generated directly by two-photon absorption in some direct–band-gap semiconductors.[1-4] In CuCl, excitation of biexcitons gives rise to an intensity-dependent refractive index. Recently, Koch and Haug[5] and also, independently, Hanamura[6] have predicted the possibility of using this nonlinearity to obtain optical bistability. By using a density-matrix formalism, Hanamura suggested that the switching time for the bistability could lie in the picosecond regime when operating slightly off resonance. This is due to the virtual formation of biexcitons with the two-photon excitation detuned from resonance. The fast response of the system is interesting in terms of both the physics of the process as well as its potential application for picosecond optical logic and signal processing. Koch and Haug obtained the complex dielectric function near the biexciton resonance from a Green's-function formalism and suggested a bistable behavior for a thin CuCl slab. They neglected the background absorption due to the exciton tail and the broadening of the biexciton resonance with intensity, and predicted bistability with a 1-μm-thick CuCl etalon with natural reflecting surfaces using a switch-on intensity of only 0.1 MW/cm^2. Sarid, Peyghambarian, and Gibbs[7] included both of the above factors in their calculations and suggested that bistability was possible only with much thicker samples (on the order of 10 μm), 90% reflective coatings, and intensities on the order of 10 MW/cm^2.

In this paper, experimental observations of optical bistability and optical limiting in CuCl are presented. These results[8] agree with the theoretical predictions of Sarid, Peyghambarian, and Gibbs.

High-purity bulk single crystals of CuCl were grown by direct reaction of pure Cu metal and CCl_4 in a sealed and evacuated silica tube.[9] These single crystals were evaporated onto fused silica substrates, which were dielectrically coated yielding 90% reflectivity. A similar dielectric mirror was placed on the CuCl film to complete the fabrication of the etalon. The thickness of the CuCl films (between 8 and 12 μm) produced this way could be measured by a film-thickness monitor with a crystal oscillator. The etalons were mounted on the cold finger of an Air Products cryostat and were cooled to liquid-He temperature. The input laser beam was provided by a Hanna-type dye laser[10] containing a mixture of 20% BBQ and 80% BPBD dye (to move the peak of the amplified spontaneous emission away from the operating frequencies) pumped by a Molectron UV-400 nitrogen laser. Pulses of 4 ns duration and less than 30 GHz linewidth at 10 Hz repetition rates were easily tuned from 380 to 392 nm. The laser beam was focused to a 20- to 25-μm-diam spot on the sample with a 32-mm–focal-length microscope objective. An Instrument Technology, Inc. vacuum photodiode with 0.1 ns rise time was used to detect both input and output pulses. The pulses were displayed on a Tektronix 7104 oscilloscope, with an amplifier rise time of 0.6 ns.

Hysteresis loops were obtained with the oscilloscope using the x-y display mode. The input pulse was optically delayed before striking the detector, so that the photodiode signal consisted of a time-resolved output and input pulse pair (time traces as in Figs. 1–4). This two-pulse signal was then sent "backwards" through an HP 15104A adder; one output of the adder was connected directly to the oscilloscope x axis and the other output was cable delayed and connected to the y axis. The oscilloscope internal time delay was used to synchronize the timing so that the etalon output versus input yielded a straight line in the absence of the sample (see Fig. 4).

Figure 1 shows optical limiting action in a 10-

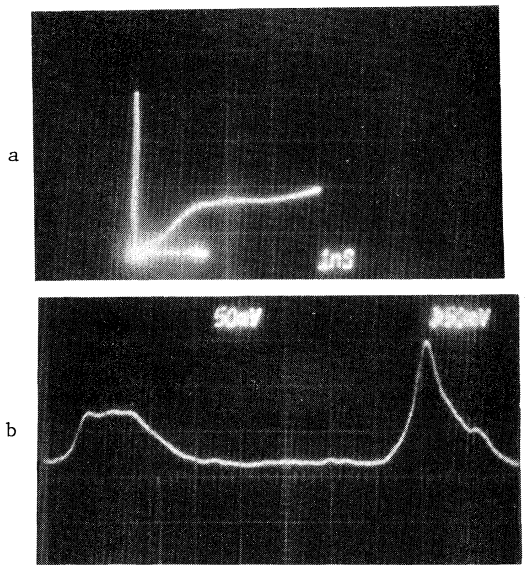

FIG. 1. Limiting action in a 10-μm-thick CuCl etalon. (a) Output vs input intensity. (b) Input pulse (right) and output pulse (left) vs time.

μm-thick CuCl etalon at 7 K with about 15 MW/cm^2 intensity. In Fig. 1(a) the output versus input intensity is plotted, and Fig. 1(b) shows both output (first pulse) and input (second pulse) versus time. This behavior was observed only in a very small frequency range near the biexciton resonance ($\simeq$ 3.186 eV). Changing the frequency by a fraction of a millielectronvolt caused the limiting action to disappear. Optical bistability was not observed in this etalon, presumably because the finesse was too low.

Figures 2 and 3 show optical bistability in a 12-μm-thick etalon at 17 K with about 7 to 14 MW/cm^2 intensity. The operating frequency corresponded to about 3.187 eV (the biexciton resonance, which is 3.1861 eV at 4 K, shifts to higher energies at higher temperatures). The laser frequency was changed by a fraction of a millielectronvolt in the sequence of Figs. 2(a)–2(c). Bistability was seen only in a narrow frequency range [Fig. 2(b)]. Detuning the laser by a slight amount to lower frequency (thereby moving it further from the Fabry-Perot peak) caused the switch-on to occur at higher intensity; detuning it further by a fraction of a millielectronvolt resulted in the disappearance of bistability. Similar bistable behavior was observed when the laser frequency was held fixed and the Fabry-Perot–laser detuning was varied by moving across the sample to regions of differing thicknesses. The Fabry-Perot peak transmission frequency was

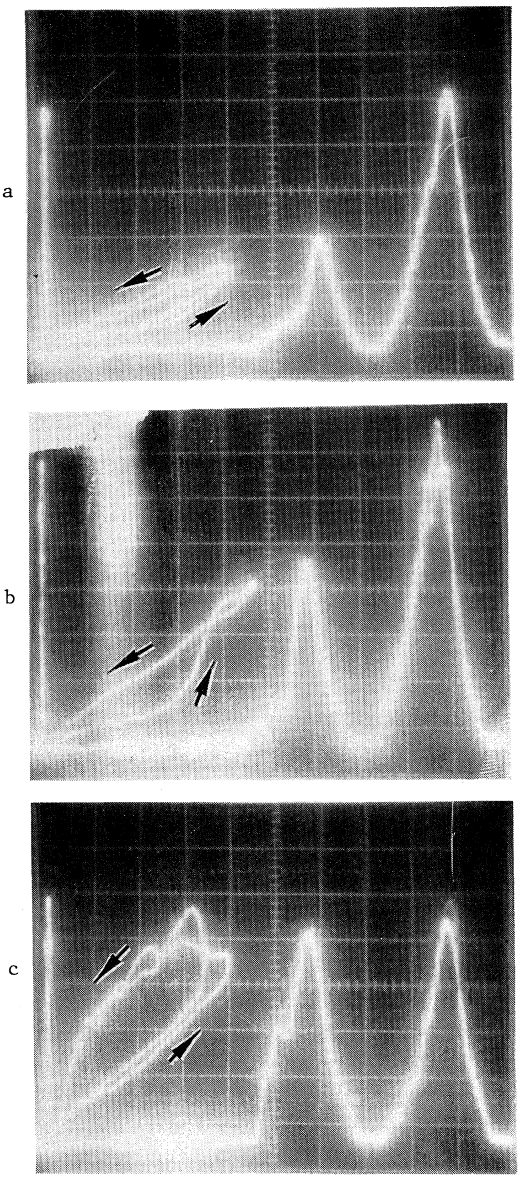

FIG. 2. Switch-on in a 12-μm-thick CuCl etalon. The laser frequency is gradually increased (corresponding to a fraction of a millielectronvolt change) in the sequence of photographs (a)–(c). Each photograph shows hysteresis loop, output pulse (center), and input pulse (right). Note that only (b) shows switch-on. Switch-off is not resolved in (b) as the input dye-laser pulse falls too steeply. The time scale is 5 ns/div.

located on the high-energy side of the laser frequency and both were on the low-energy side of the biexciton resonance. This is consistent for biexcitonic bistability, since the index increases with increasing intensity on the low-energy side of the biexciton resonance (see Fig. 2 of Ref. 7). The switch-off is not well resolved in Fig. 2(b)

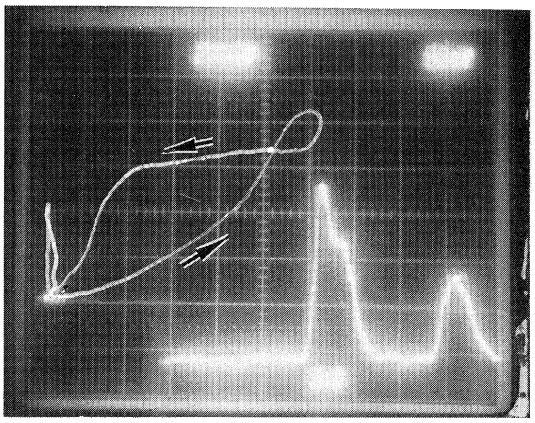

FIG. 3. Switch-off in a 12-μm-thick CuCl etalon. When the input dye-laser pulse (right) is adjusted for a slow falloff, switch-off in the output pulse (center) is clearly seen. The time scale is 5 ns/div.

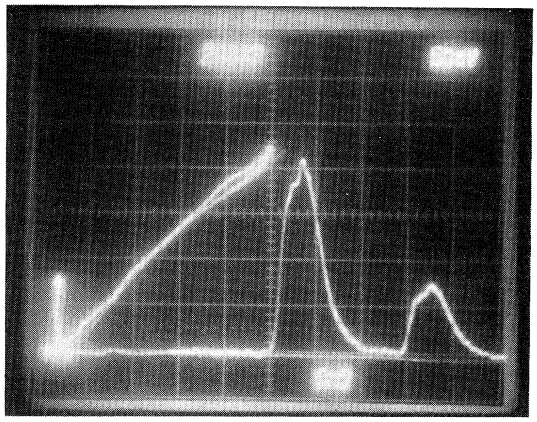

FIG. 4. Output pulse (center) vs input pulse (right) shows no hysteresis (left) in the absence of a sample.

because of the fast fall time of the input pulse (detection limited by the oscilloscope response time). However, when the input pulse was adjusted to fall more slowly, the switch-off was more apparent as shown in Fig. 3. Figure 4 demonstrates that the output versus input yields a straight line when the sample is removed (no hysteresis). A faster detection system would permit a study of switching times as a function of detuning and hopefully produce more nearly steady-state hysteresis loops.

It should be emphasized that the unsaturable background absorption plays a deterministic role in any type of optical bistable system whether the origin of the nonlinearity is excitonic, biexcitonic, band filling, etc. The background absorption lowers the finesse of the cavity (thereby destroying the feedback) and brings about a higher switching intensity requirement. In the case of biexcitons in CuCl, the exciton tail acts like an unsaturable background absorption at the biexciton resonance, causing the switching intensity to be about 10 MW/cm^2, two orders of magnitude larger than that in the absence of any background absorption. Also, the broadening of the biexciton resonance with intensity forces the operating point to move further off resonance because of the increase in absorption. This lowers the nonlinearity (producing a smaller phase shift) and causes the calculated bistability to disappear for a 1-μm-thick sample,[7] in contrast with predictions which ignore this effect.[5] To increase the nonlinearity, thicker samples (≥ 10 μm) must be used.[7] This is consistent with our experimental measurements in which we fail to observe bistability with a 1-μm-thick CuCl film coated with 90% reflective mirrors.

In summary, we present experimental observation of subnanosecond optical bistability and optical limiting action using the biexciton two-photon resonance in CuCl.[11] Theoretical calculations and experimental results agree that 10 MW/cm^2 is needed for biexciton optical bistability in CuCl, much higher than initially predicted. Nonetheless, if the switching times are indeed 1 ps, a device with a wavelength-in-the-material (λ/n_0) transverse dimension would need only several femtojoules of energy to switch. This corresponds to only a few thousand photons—a number close to the statistical limit[12] (i.e., the minimum number of photons needed to avoid occasional failure of switching due to statistical fluctuations). Thus both the physics of the process and device possibilities motivate further studies of the switching times as a function of detuning.

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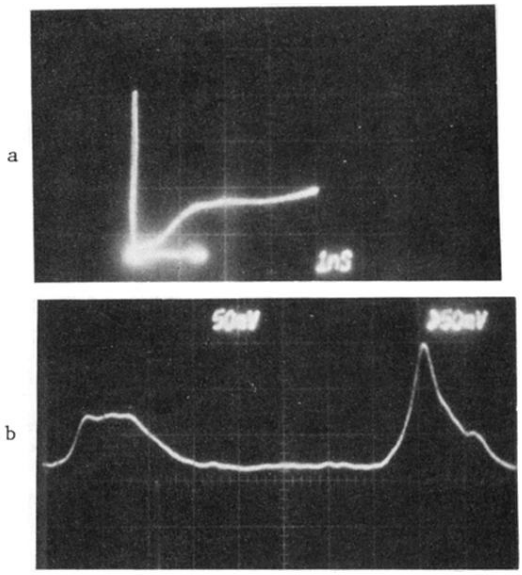
[1]G. M. Gale and A. Mysyrowicz, Phys. Rev. Lett. 32, 727 (1974).

[2]R. W. Svorec and L. L. Chase, Solid State Commun.

20, 353 (1976).
[3]A. Marvani, J. L. Oudor, E. Batifol, and D. S. Chemla, Phys. Rev. Lett. 41, 1372 (1978).
[4]L. L. Chase, N. Peyghambarian, G. Grynberg, and A. Mysyrowicz, Opt. Commun. 28, 189 (1979).
[5]S. W. Koch and H. Haug, Phys. Rev. Lett. 46, 450 (1981).
[6]E. Hanamura, Solid State Commun. 38, 939 (1981).
[7]D. Sarid, N. Peyghambarian, and H. M. Gibbs, Phys. Rev. B 28, 1184 (1983).
[8]These results were reported by N. Peyghambarian *et al.*, in Proceedings of the Optical Bistability Conference, Rochester, 15–17 June 1983 (to be published).
[9]B. Batlog, J. P. Remeika, and A. Jayaraman, Bull. Am. Phys. Soc. 5, 268 (1980).
[10]D. C. Hanna, P. A. Karkkainen, and R. Wyatt, Opt. Quant. Electron. 7, 115 (1975).
[11]Observation of optical bistability in CuCl has also been reported by B. Honerlage, J. Y. Bigot, and R. Levi, in Proceedings of the Optical Bistability Conference, Rochester, 15–17 June, 1983 (to be published).
[12]S. L. McCall and H. M. Gibbs, in *Optical Bistability*, edited by C. M. Bowden, M. Ciftan, and H. R. Robl (Plenum, New York, 1981).



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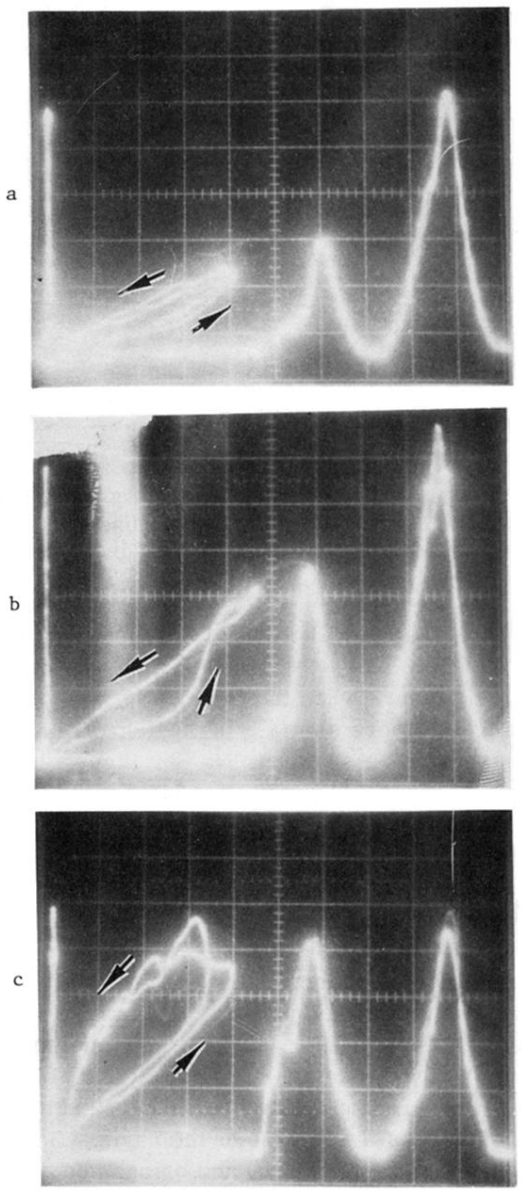
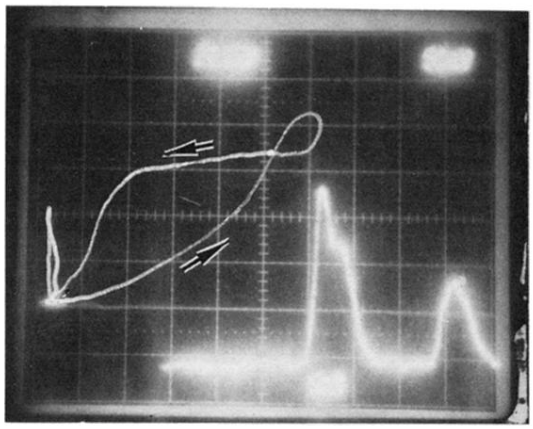


FIG. 2. Switch-on in a 12-μm-thick CuCl etalon. The laser frequency is gradually increased (corresponding to a fraction of a millielectronvolt change) in the sequence of photographs (a)–(c). Each photograph shows hysteresis loop, output pulse (center), and input pulse (right). Note that only (b) shows switch-on. Switch-off is not resolved in (b) as the input dye-laser pulse falls too steeply. The time scale is 5 ns/div.



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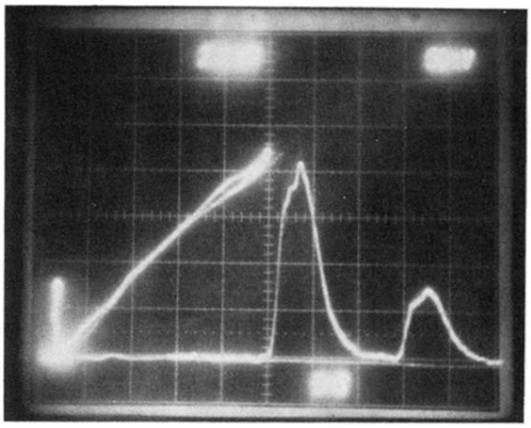

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