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Optical bistability in CuCl

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Optical bistability due to biexcitons in CuCl is discussed with respect to its physical origin. We show that switching times and switching intensities are functions of the photon energy of the exciting light beam.

I. Introduction

Optical bistability was first observed by Gibbs *et al.* (1976) using a Fabry–Perot etalon filled with sodium vapour. Although this material shows saturable absorption, it turned out that the intensity dependence of the refractive index was extremely important for the observation of optical bistability. Since the use of vapour requires Fabry–Perot resonators with a long 'round-trip' time, research switched to more dense materials like liquids and solids. In semiconductors, optical nonlinearities may be very important. Near resonances, optical bistable behaviour was first observed in the infrared and visible spectral region in InSb and GaAs by Miller *et al.* (1979) and Gibbs *et al.* (1979), respectively. In both materials, bistability is due to the saturation of an absorption (band to band or exciton transitions) and the resulting intensity dependence of the refractive index. In both cases, switch-on times of the devices are limited in principle by the cavity round-trip time, which is in the picosecond range. Since the index change depends on the density of the quasi-particles excited throughout the process, the switch-off time is limited by the lifetime of the particles, which is in the nanosecond range. Although this time constant may be diminished by doping, it would be particularly important to look for nonlinear optical devices with switch-off times in the picosecond timescale.

Recently, Hanamura (1981) has studied the transient behaviour of the dielectric function in semiconductors like CuCl. As will be discussed in more detail, it can be described by a three-level system, in which virtual transitions are responsible for some nonlinearities. Therefore, the lifetimes of the excited quasi-particles do not influence the switching times, which are then only limited by the round-trip time in the cavity. In addition, in CuCl, the band gap is about 3.38 eV, which leads to an optical bistability in a new spectral region around 3.186 eV (at about 390 nm). To understand the physical origin of this bistability, we will first give some insights into the band structure of quasi-particles in CuCl.

II. Absorption and dispersion anomalies in CuCl

Copper chloride is a zinc-blende-type material (T_d) with a direct band gap at the centre of the Brillouin zone. It has a lower conduction band of point group symmetry Γ_6 and two uppermost valence bands of Γ_7 and Γ_8.

This band structure gives rise to two exciton series called Z_{12} and Z_3, respectively, having the symmetry properties

$$Z_3: \Gamma_6 \otimes \Gamma_7 = \Gamma_2 \oplus \Gamma_5,$$
$$Z_{12}: \Gamma_6 \otimes \Gamma_8 = \Gamma_3 \oplus \Gamma_4 \oplus \Gamma_5.$$

For CuCl, the Z_3 series is the exciton series of the lowest energy and only its ground state will be considered here.

Two excitons may couple together to give rise to a bound state: the biexciton or excitonic molecule. The symmetry of the biexciton ground state with energy E_{bi} is given by the antisymmetric product of the Bloch functions of two electrons and two holes:

$$\Gamma_{env} \otimes (\Gamma_6 \times \Gamma_6)^- \otimes (\Gamma_7 \times \Gamma_7)^- = \Gamma_1.$$

Because of these symmetries, optical dipole transitions are allowed between the crystal ground state and the transverse Γ_5 exciton state, and between Γ_5 exciton states and the Γ_1 biexciton ground state. All other one-photon transitions are forbidden. However, biexcitons may be excited by two-photon absorption. More details of this system can be seen in a recent review article by Grun *et al.* (1982).

Owing to the strong coupling between the Γ_5 transverse exciton and the electromagnetic radiation field, eigenstates of the interacting system are polaritons. Their dispersion $E_t(\boldsymbol{Q}_t)$ is given at low intensities of excitation by Hopfield's one-oscillator model, which depends on four parameters: the transverse and longitudinal exciton energy E_T and E_L, the exciton effective mass m_{ex} and the background dielectric constant ϵ_b. The polariton dispersion is directly related to the refractive index by the relation $n = \hbar c Q_t / E_t(\boldsymbol{Q}_t)$.

We have determined the excitonic polariton dispersion, using hyper-Raman scattering (Phach *et al.* 1978). By a self-consistent analysis of the spectral positions of the emission lines, we could obtain the different parameters that describe the polariton dispersion (Hönerlage *et al.* 1978).

What happens when such a crystal is excited by an intense laser beam with a photon energy $\hbar\omega_p$? When the laser is tuned close to the exciton resonance, an exciton population is created and, since the transition between excitons and biexcitons is dipole allowed, a test beam with frequency ω_t may be absorbed if $\hbar\omega_t = E_{bi} - E_T$. This corresponds to an induced oscillator strength at this energy and gives rise to an anomaly in the polariton dispersion curve. In the absence of any damping, a gap is induced, the magnitude of which is roughly proportional to the number of excitons created by the pump beam. If this pump beam is detuned from the exciton resonance, we obtain qualitatively a similar behaviour: an anomaly of the dispersion, due to the two-photon $(\hbar\omega_t, \hbar\omega_p)$ absorption to the biexciton ground state, is created at $E_t = E_{bi} - \hbar\omega_p$. This new resonance shifts when the photon energy of the pump beam is tuned. In this case, the magnitude of the gap is proportional to the number n_p of polaritons in the mode of frequency ω_p (May *et al.* 1979; Märtz *et al.* 1980).

To describe these phenomena quantitatively, our group have developed a theoretical model to calculate the dispersion of a three-level system, by using the density matrix formalism (Bigot *et al.* 1984; Hönerlage *et al.* 1984). Typical results are shown in figure 1. The pump beam is fixed at an energy of 3.184 eV and has an intensity of 10^{15} photons per cubic centimetre. The dispersion of a test beam is considerably changed at the energy 3.1695 eV, which corresponds to the induced absorption from the exciton to the biexciton state, as discussed above. A second anomaly at 3.188 eV is due to two-photon absorption to the biexciton state that uses one photon of the pump beam and one of the test beam. At very high intensities, a quasi-resonance appears

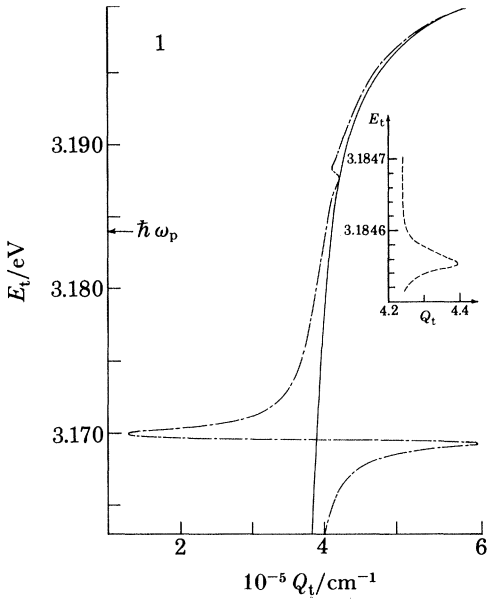

FIGURE 1. The chained curve represents the polariton dispersion of CuCl, when excited at $\hbar\omega_p = 3.184$ eV by a pump beam with a photon density $n_p = 10^{15}$ cm^{-3}, the damping constant being $\hbar\Gamma = 0.2$ meV. The full line gives the result for a one-oscillator model without damping and with $n_p = 0$. The inset shows the pseudoresonance around the energy of the pump beam for $n_p = 10^{16}$ cm^{-3}

also near the energy of the pump beam. It is due to a periodic variation of the exciton and biexciton populations.

We have discussed till now the change of the dispersion of a test beam in the presence of a pump beam; however, the pump beam can also change its own dispersion, especially when the crystal is excited near half the biexciton energy. In this case, the pump beam approaches the upper anomaly of figure 1 and creates biexcitons by two-photon absorption. The resulting dispersion is shown in figure 2. We observe a strong variation of the dispersion around $\frac{1}{2}E_{bi}$. Outside the biexciton resonance, biexcitons are only virtually created, but the variation of the dispersion is still quite strong.

By hyper-Raman scattering (Grun *et al.* 1983) and non-degenerated four-wave mixing (Hönerlage *et al.* 1983), we have been able to study the energy and intensity dependence of these anomalies for different polarizations of the light sources. We have concluded that the nonlinear refractive index is, at the maximum, of the order of 4×10^{-5} cm^2 kW^{-1}. It is therefore comparable to the results found in GaAs.

After this discussion on the physical origin of these nonlinearities, let us now consider the optical bistability, which has been predicted for CuCl by Koch *et al.* (1981) and Hanamura (1981) and observed experimentally by our group (Levy *et al.* 1983) and by Peyghambarian *et al.* (1983).

III. Optical bistability

We have worked with a single exciting beam, using the auto-renormalization shown in figure 2 as the source of nonlinearity. When the laser frequency is tuned through the biexciton resonance, exciting polaritons are subject to this anomaly. Therefore, one might hope that this process could lead to optical bistability if enough feedback is provided. When

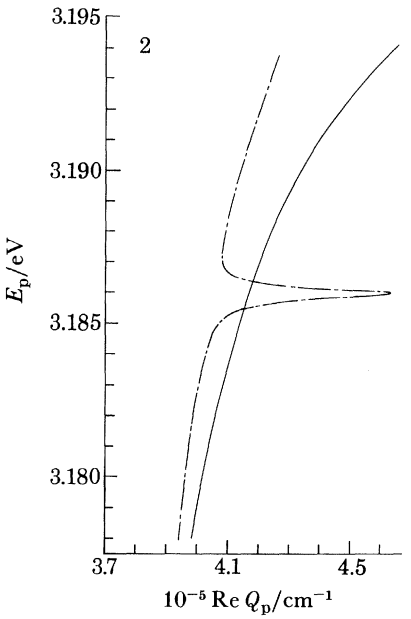

FIGURE 2. Dispersion $E_p(Q_p)$ in CuCl in a one-oscillator model when a single beam ($E_t = E_p$) is considered: full line, without renormalization; chained line, our model with $n_p = 10^{15}$ cm^{-3}.

biexcitons are created only virtually in the process, the switching time of the device should not depend on the lifetimes of the elementary excitations and therefore may be very short.

A CuCl sample of 10–30 μm thickness is placed between mirrors that provide an optical feedback. Since the sample is in direct contact with the mirrors, this is also the length of the Fabry–Perot etalon (F.P.), which is excited perpendicular to its surface. The mirrors are thin glass plates coated with platinum films. Their reflectivity is 90 %. Platinum has been chosen since most other metals react chemically with CuCl. When using natural or directly coated surfaces, no sufficient feedback was obtained. Although the mirrors are highly reflective, only a maximum intensity variation of 40 % is obtained. This is due to the absorption of the crystal near the 1S exciton line and to a lack of parallelism of the crystal surfaces. The F.P. and the sample are cooled down to low temperatures.

Figure 3 shows our experimental arrangement. A XeCl laser (Lambda Physik) pumps a dye laser containing a diluted solution of αNND in ethanol. The emission is amplified in a second dye cell also filled with αNND and pumped by the same excimer laser. Great care is taken to keep the intensity of the super-radiant emission small compared to the laser emission. The shape of the pulses is kept well-defined in time with a width of about 3 ns (fwhm). The laser emission has a spectral width of 0.05 meV. After passing through a diaphragm, neutral density filters and a glass polarizer, the beam is split into two parts by a glass plate.

One part of the beam is focused onto the F.P. containing the crystal, in a spot of 100 μm diameter. Power intensity can be varied up to 50 MW cm^{-2}. The transmission of the laser pulses through the F.P is detected by a fast photocell and is analysed in time by an oscilloscope operating at gigahertz frequencies. The overall time resolution is less than 500 ps. The transmission spectrum could equally well be analysed by a spectrograph and an O.M.A. system. The outer part of the beam passes, after an optical delay of 1.9 ns, through neutral density filters and is then detected by the same photocell. This beam is used as a reference for the shape

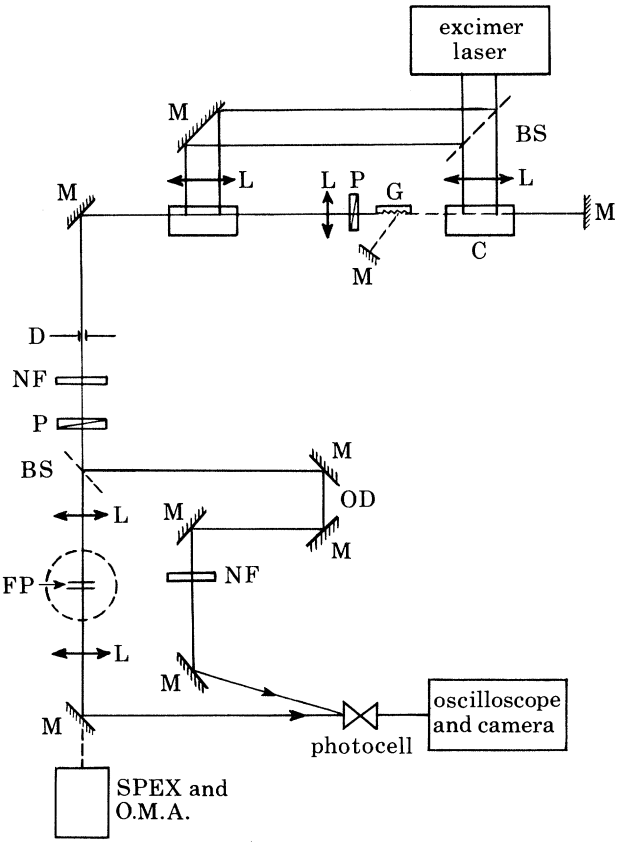

FIGURE 3. Experimental arrangement: M, mirror; BS, beam splitter; L, lens; C, dye cell; P, polarizer; D, diaphragm; NF, neutral density filter; F.P., Fabry–Perot; G, grating; OD, optical delay; O.M.A., optical multichannel analyser; SPEX, spectrograph.

of the pulse. The signal, observed on the oscilloscope is then photographed with a camera, so that single shots may be analysed.

Figure 4 shows the transmitted and reference laser pulses analysed by the oscilloscope when the F.P. is excited with a photon energy close to the biexciton energy; δ is the time delay between transmitted and reference pulses. At this energy and intensity of excitation, the transmitted pulse is clearly deformed compared to the reference pulse. The intensity of the transmitted pulse is now analysed in detail.

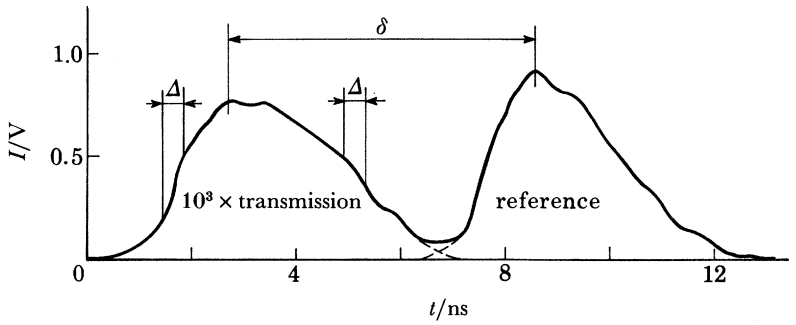

FIGURE 4. Picture of the transmitted and incident dye laser pulses (the transmitted pulse is attenuated by a factor of 1000 by the F.P). Δ, time resolution (*ca.* 500 ps); δ, optical delay between the two pulses.

Figure 5 shows the output against input intensity curves for different maximum intensities of excitations. Open circles correspond to increasing intensities and crosses to decreasing intensities. Figure 5*a* corresponds to the figure when the bistability is clearly observed with a 'switch-on' intensity of 15 MW cm^{-2} and a 'switch-off' intensity of 5 MW cm^{-2}. The switching point stays at a fixed intensity when the maximum intensity is decreased (figure 5*b*). In figure 5*d*, no bistability is observed since $I_{max} < 15$ MW cm^{-2} and, in figure 5*e*, we had no sample inside the cavity. Both corresponding curves are linear.

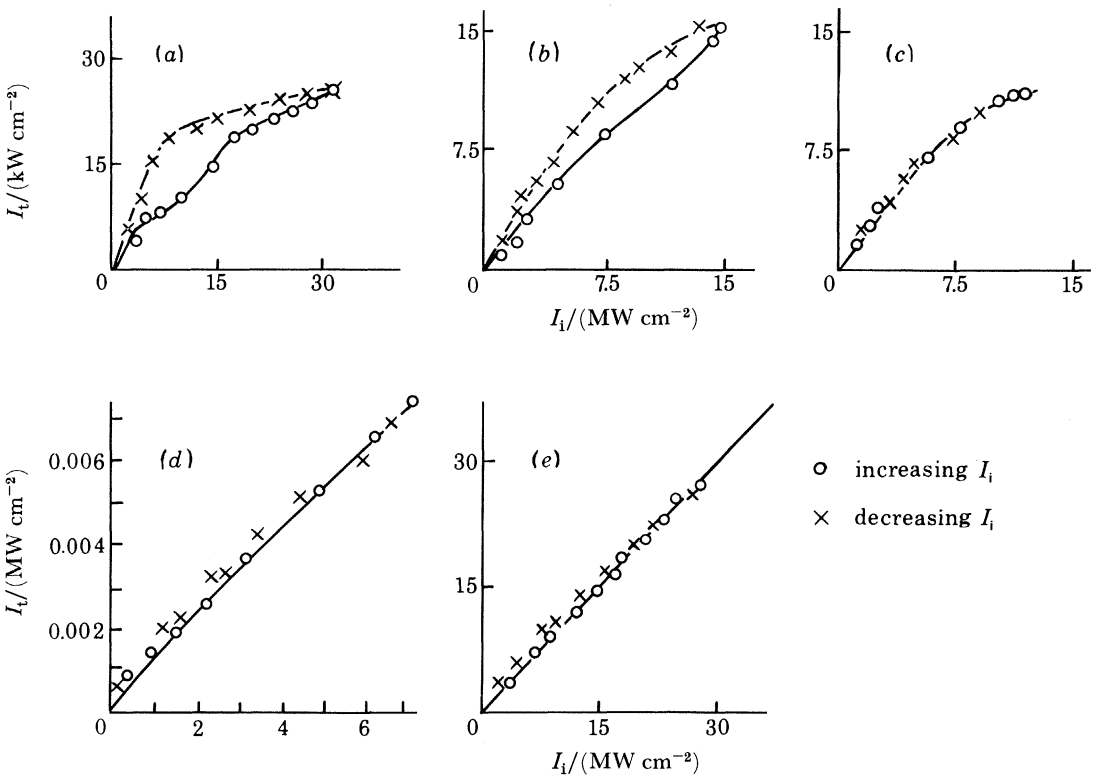

FIGURE 5. Output (I_t) against input (I_i) intensity curves for different maximum intensities (*a*)–(*d*), (*e*) no sample inside the cavity.

Let us now investigate the dependence of the hysteresis loop on the photon energy of the exciting beam. In figure 6, we have plotted the areas of the bistability loop as a function of the photon energy of the exciting beam. This quantity is proportional to the energy stocked inside the F.P. cavity. Here we obtain a structure with two maxima. The first maximum is situated at an energy corresponding to a maximum of transmission of the F.P. at 3.1855 eV. The second maximum is situated near $\frac{1}{2}E_{bi} = 3.186$ eV. This doublet structure depends on the position of the maximum of transmission of the F.P. relative to $\frac{1}{2}E_{bi}$. In this case they are well separated, but we can also obtain excitation spectra with one maximum. In any case, we have shown that the switching behaviour is different depending on whether the crystal is excited at the biexciton resonance or not.

Figure 7*a* shows a hysteresis cycle that is typically observed when we excite near the first maximum. The 'switch-on' and 'switch-off' times are not resolved by the apparatus and they

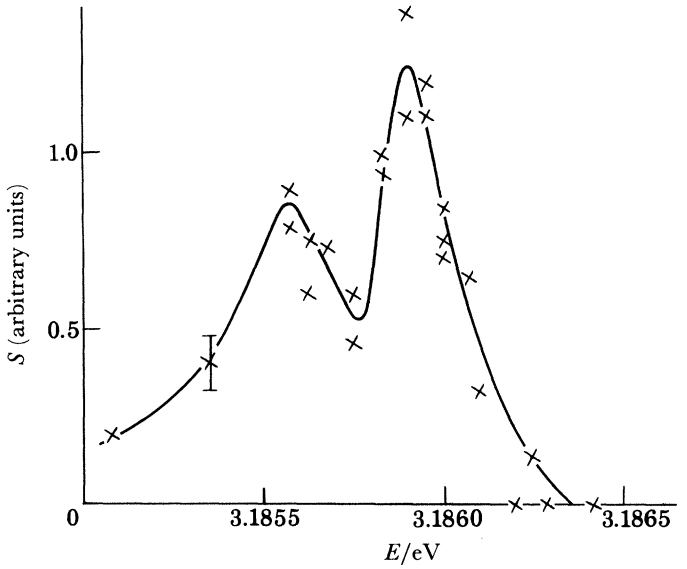

FIGURE 6. Areas of the bistability loop as a function of the photon energy of the exciting beam.

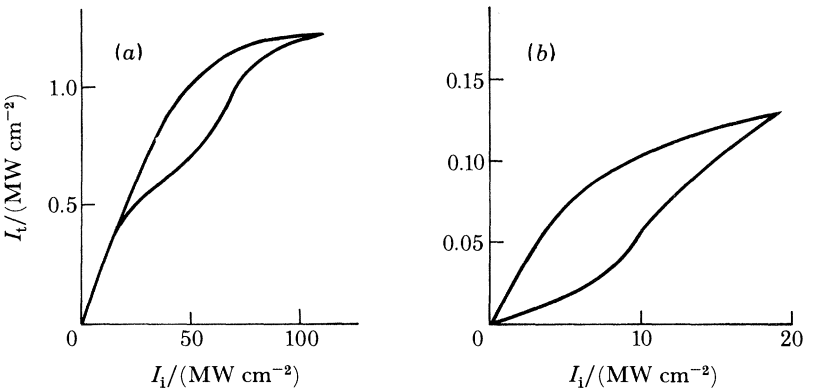

FIGURE 7. (*a*) A typical loop when exciting near the first maximum of figure 6, i.e. 3.1855 eV. (*b*) Hysteresis loop when exciting near the second maximum of figure 6, i.e. 3.186 eV.

are both below 500 ps. This can be concluded from the fact that increasing and decreasing pulses follow the same straight line near the origin (Hönerlage *et al.* 1983).

If, however, the energy is near $\frac{1}{2}E_{bi}$, the hysteresis cycle (figure 7*b*) has a completely different shape than the one shown in figure 7*a*. As our pulses are not long enough, we estimate the commutation time from the 'on' to the 'off' state to be of the order of 3 ns.

These results lead us to suppose that, in the first case, the commutation time is small because biexcitons are created only virtually. In the second, biexcitons are really created. The index change may then be due to the number of excitons and biexcitons created. This would imply that their radiative lifetime affects the switch-off time of the device.

We have used our theoretical model discussed in connection with figure 2 to calculate the transmission characteristic of a 10 μm thick sample in a F.P with a coefficient of reflection of 95%.

Near $\frac{1}{2}E_{bi}$ we obtain an important hysteresis loop, as shown in figure 8*a*. If we tune the energy

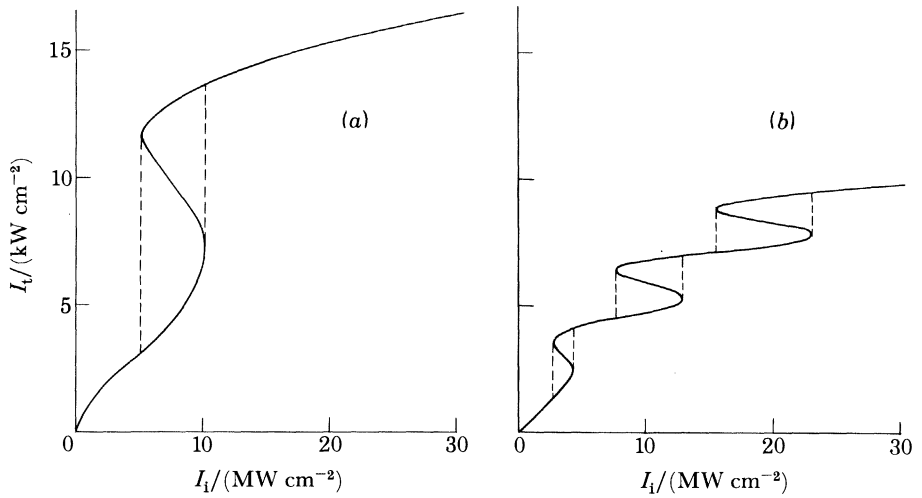

FIGURE 8 (*a*) Calculated hysteresis cycle for a photon energy of 3.185 eV. (*b*) Calculated hysteresis cycles for a photon energy of 3.1855 eV.

of the exciting beam closer to $\frac{1}{2}E_{\mathrm{bi}}$, more F.P. fringes are then covered. A typical result is shown in figure 8*b*. Here many hysteresis loops overlap and only the envelope may be observed experimentally. Using this model, we can qualitatively explain the excitation spectrum given in figure 6. A more detailed study is under way.

CONCLUSION

In conclusion, we have given experimental evidence for optical bistability due to biexcitons in CuCl at both 1.9 K and 60 K. We have shown that the phenomenon is strongly resonant at half the biexciton energy, which allows us to state that the nonlinearity is due to biexciton two-photon absorption. Working with virtual states, the switching times are shorter than the experimental time resolution, which is 500 ps. However, the required input intensity is rather high (*ca.* 15 MW $\mathrm{cm^{-2}}$). At the resonance, switching times are longer.

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