Downloaded from rsta.royalsocietypublishing.org

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Fast and Sensitive Nonlinear Processes: Bistability in CdHgTe

A. Miller and G. Parry

Phil. Trans. R. Soc. Lond. A 1984 **313**, 277-284
doi: 10.1098/rsta.1984.0107

Email alerting service

Receive free email alerts when new articles cite this article - sign up in the box at the top right-hand corner of the article or click **here**

To subscribe to *Phil. Trans. R. Soc. Lond. A* go to: **http://rsta.royalsocietypublishing.org/subscriptions**

This journal is © 1984 The Royal Society

Downloaded from rsta.royalsocietypublishing.org

Phil. Trans. R. Soc. Lond. A **313**, 277–284 (1984)
Printed in Great Britain

Fast and sensitive nonlinear processes: bistability in CdHgTe

By A. Miller and G. Parry†
Royal Signals and Radar Establishment, St Andrews Road, Great Malvern WR14 3PS, U.K.

We discuss the origin and characteristics of the very large intensity-dependent changes in the refractive index of the semiconductor cadmium mercury telluride at 10.6 μm. With continuous wave (c.w.) radiation and sample temperatures of 77 K nonlinear Fabry–Perot effects can be observed at powers as low as 1 mW. The Auger dominated recombination process and the thermal contributions to the refractive index make observations of optical bistability difficult with c.w. radiation. However, the use of a pulsed CO_2 laser and two-photon absorption of the radiation does lead to the observation of bistability at room temperature. We compare these results with the reported work on InSb.

1. Introduction

The alloy semiconductor $Cd_x Hg_{1-x} Te$ offers a variation of band gap with composition from 0 to 1.5 eV (Dornhaus & Nimtz 1976). This wide range thus offers energy gaps resonant with a number of important laser frequencies and in particular it is one of the few semiconductors suitable for the study of band-gap resonant optical nonlinearities in the CO_2 laser output band at around 10 μm (Hill *et al.* 1982). CdHgTe crystallizes in the zincblende structure so that small-gap alloys of this material have energy bands closely related to that of InSb, a semiconductor that shows extremely large band-filling optical nonlinearities at 5 μm (D. A. B. Miller *et al.* 1978, 1981). This makes CdHgTe an obvious choice as a material in which the band-gap resonant nonlinearities discovered in InSb and subsequently used to demonstrate various optically bistable and related devices (Walker *et al.*, this symposium) can be studied with the flexibility of varying the band-gap energy as a function of both alloy composition and temperature. In this paper we discuss the origin of large optical nonlinearities in CdHgTe at 10 μm, report nonlinear Fabry–Perot response under different experimental conditions and make some comparisons with InSb.

With the use of a low-power c.w. CO_2 laser, a $Cd_{0.23}Hg_{0.77}Te$ etalon at 77 K showed nonlinear features at input powers as low as 1 mW. With pulsed lasers, optical switching and bistability could be observed in the transmission of a room-temperature etalon. As predicted by theory, the resonant nonlinearity was found to be larger than for InSb for this smaller band gap semiconductor, but we also found that the carrier recombination mechanism and thermal properties of CdHgTe give an intensity-dependent etalon response quite distinct from that of InSb.

2. Optical nonlinearities in CdHgTe

In semiconductors, numerous mechanisms may result in optical nonlinearities (A. Miller *et al.* 1981; Jain 1982), and a number of different nonlinear effects have been observed in CdHgTe under a variety of different conditions. Initial interest centred on the stimulated spin-flip Raman

† Present address: Electronic and Electrical Engineering Department, University College London, Torrington Place, London WC1E 7JE, U.K.

The Royal Society is collaborating with JSTOR to digitize, preserve, and extend access to *Philosophical Transactions of the Royal Society of London. Series A, Mathematical and Physical Sciences.* www.jstor.org

Downloaded from rsta.royalsocietypublishing.org

process which can be described in terms of a third-order nonlinear susceptibility, $\chi^{(3)}$. Kruse *et al.* (1979) achieved tunable laser action by using a Q-switched CO_2 laser to pump a band-gap resonant sample of $Cd_{0.23}Hg_{0.77}Te$ at 4 K in a high magnetic field. Under similar conditions, but making use of two different laser frequencies, ω_1 and ω_2, resonant four-wave mixing of type $\omega_3 = 2\omega_1 - \omega_2$ could be observed when $\omega_2 - \omega_1$ equalled the energy separation of spin-up and spin-down states of a given Landau level, and a value of $\chi^{(3)} = 10^{-4}$ e.s.u.† was deduced for this spin-resonant mixing process (Kruse 1981).

More recently, interest has turned to phase conjugation by degenerate four-wave mixing (d.f.w.m.). Initially, high-power pulsed lasers were employed. Khan *et al.* (1980) observed 9% phase conjugate reflectivities of 10.6 μm radiation at 12, 77 and 295 K with n-type $Cd_xHg_{1-x}Te$ with $x = 0.216$ and 0.232. Although some of these conditions were band-gap resonant, interpretation was made in terms of a non-resonant nonlinearity arising from conduction-band non-parabolicity and values of $\chi^{(3)}$ in the range 5×10^{-8} to 4×10^{-7} e.s.u. were deduced. Independently, Jain & Steel (1980) achieved similar results at room temperature under band-gap resonant conditions. They concluded that this was consistent with a phase grating produced by the generated free-carrier plasma refraction with a value of $\chi^{(3)} \approx 5.4 \times 10^{-6}$ e.s.u. Limiting of the reflected d.f.w.m. pulses at 10% (Khan *et al.* 1980; Jain & Steel 1980) has been studied recently by Yuen & Becla (1983) in p-type CdHgTe under band-gap resonant conditions and interpreted in terms of the effect that bleaching the inter-band absorption has on the refractive index change. Also, non-degenerate four-wave mixing of type $\omega_3 = 2\omega_1 - \omega_2$ with two CO_2 lasers by Yuen (1982) in $Cd_{0.265}Hg_{0.735}Te$ at 2 °K (band gap 0.183 eV) showed a variation of $\chi^{(3)}$ with $\Delta\omega = \omega_1 - \omega_2$. For small $\Delta\omega$, $\chi^{(3)}$ was considered to be due to band non-parabolicity for free electrons generated by two-photon absorption, whereas for $\Delta\omega > 10\ \text{cm}^{-1}$, the origin of the nonlinearity was thought to be due to bound electrons with $\chi^{(3)} \approx 3 \times 10^{-8}$ e.s.u.

Much larger free carrier plasma nonlinearities, $\chi^{(3)} \approx 5 \times 10^{-2}$ e.s.u., were discovered by Jain *et al.* (1981, 1982) in d.f.w.m. experiments by lowering the temperature of $Cd_xHg_{1-x}Te$ ($x = 0.21$–0.23) to tune the band gap to the output of a c.w. CO_2 laser. A lower temperature also has the effect of increasing the Auger-dominated lifetime of the free carrier plasma, thus enhancing the magnitude of the nonlinearity. Similar d.f.w.m. experiments by Khan *et al.* (1981) gave $\chi^{(3)} \approx 3 \times 10^{-2}$ e.s.u.

Even larger optical nonlinearities were observed by Hill *et al.* (1982) in band-gap resonant self-defocusing studies of $Cd_{0.21}Hg_{0.79}Te$ at 175 K. Refractive index changes were found to vary according to $\Delta n = -7 \times 10^{-3} I^{\frac{1}{3}}$ (I in watts per square centimetre) and changes in beam profiles in the far-field were observable at 1 W cm^{-2} corresponding to an effective $\chi^{(3)} \approx 6$ e.s.u. at this intensity. This extremely large nonlinearity arises from both a free carrier plasma contribution and the effect of band filling, and the power dependence is explained by the role of Auger recombination limiting the accumulation of generated carriers as the intensity is increased (§3).

Preliminary evidence of nonlinear Fabry–Perot transmission in CdHgTe has been reported by Jain & Steel (1982) and Khan *et al.* (1983).

† 1 e.s.u. = 1 $cm^3\ erg^{-1} \equiv 1.4 \times 10^{-8}\ m^2\ V^{-2}$.

Downloaded from rsta.royalsocietypublishing.org

3. Band-gap resonant nonlinear refraction in CdHgTe

For photon energies less than the band gap energy of a semiconductor, a change in refractive index, Δn, may be expected to result when an excess density of electron–hole pairs, ΔN, is created. In CdHgTe, a large refractive index change can arise from the free carrier plasma. This can be adequately described by the Drude expression derived from standard dispersion theory (Jain & Klein 1979):

$$\sigma_p = -e^2/2\epsilon_0 n_0 m^* \omega^2, \tag{1}$$

where σ_p gives the refractive index change per electron–hole pair per unit volume, e is the electronic charge, ϵ_0 the free space dielectric constant, n_0 the linear refractive index, m^* the conduction band effective mass and ω the photon frequency. For a semiconductor with small band gap this is a particularly large effect because of the low effective mass and small photon energy. An additional important contribution to the nonlinear refraction for photon energies close to the band gap is filling of the conduction band states resulting in blocking of interband transitions. This effect has been analysed in some detail in InSb (D. A. B. Miller *et al.* 1981) and found to be consistent with the expression derived by using the Kramers–Kronig integration to relate the changes in refraction with changes in absorption:

$$\sigma_s = \sigma_p \left\{ \frac{2}{3\sqrt{\pi}} \frac{m^*}{m} \left(\frac{mP^2}{\hbar} \right) \frac{1}{kT} J\left(\frac{\hbar\omega - E_g}{kT} \right) \right\}, \tag{2}$$

where P is the momentum matrix element and

$$J(a) = \int_0^\infty \frac{\mathrm{d}x\, x^{\frac{1}{2}} e^{-x}}{(x-a)}.$$

Sub-band excitation can cause the creation of excess carriers either via band-tail states or by two-photon absorption. For the 77 K, $Cd_{0.23}Hg_{0.77}Te$ samples employed in the experiments described in §4, the photon energy is within 3.5 kT of the band gap energy for 10.6 μm radiation and should result in $\sigma_p \approx -1.1 \times 10^{-18}$ cm^3 and $\sigma_s \approx -1.2 \times 10^{-18}$ cm^3. The same composition at room temperature ($E_g = 0.2$ eV) is suitable for two photon excitation and should give $\sigma_p \approx -7.4 \times 10^{-19}$ cm^3 and $\sigma_s \approx -3.7 \times 10^{-19}$ cm^3 at 10.6 μm.

To a first approximation, these free carrier nonlinearities are proportional to the density of excess carriers so that

$$\Delta n = (\sigma_p + \sigma_s)\, \Delta N. \tag{3}$$

The density of carriers in a semiconductor under optical excitation is governed by the dynamic balance of generation, recombination and diffusion. Under the conditions employed in our experiments, diffusion can be neglected. Recombination in CdHgTe at both 77 and 295 K is dominated by Auger processes, which cause a strong concentration-dependence for the carrier lifetime. This therefore has a significant effect on the intensity dependence of the nonlinearity and in turn the form of the nonlinear Fabry–Perot characteristics. For our 77 K samples ($N_D - N_A = 1 \times 10^{15}$ cm^{-3}), the small-modulation Auger lifetime is about 2.5 μs and to a good approximation we should expect the refractive index change to be governed by

$$\Delta n = \gamma I^{\frac{1}{3}}, \tag{4}$$

where $\gamma = -5 \times 10^{-3}$ (cm^2 W^{-1})$^{\frac{1}{3}}$. This intensity dependence was confirmed in self-defocusing measurements in which the $I^{\frac{1}{3}}$ dependence is seen in the power dependence of far-field beam

Downloaded from rsta.royalsocietypublishing.org

widths (figure 1) (Hill *et al.* 1982). It has the effect of making optical bistability more difficult to achieve because spacing of the cavity orders with intensity becomes larger and the slope of the transmission curve with cavity intensity is shallower in all orders than for a nonlinearity in which the refractive index change is proportional to intensity (Miller & Parry 1984).

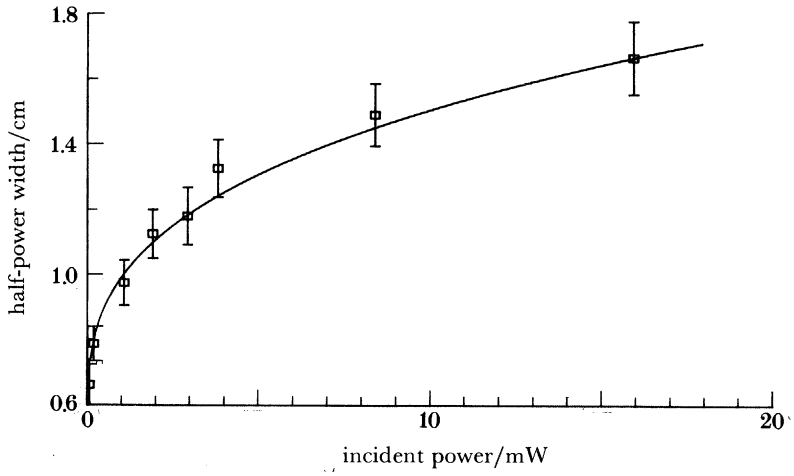

FIGURE 1. Power dependence of far-field beam widths for 10.6 μm radiation after transmission through 330 μm thick $Cd_{0.21}Hg_{0.79}Te$ at 175 K placed just beyond a beam waist of 200 μm (f.w.h.m.). The solid line shows a one-third power dependence for comparison (Hill *et al.* 1982).

4. Low-power nonlinear Fabry–Perot

We have found that a 200 μm thick 77 K etalon of $Cd_{0.23}Hg_{0.77}Te$ with polished faces gives a nonlinear response at a c.w. CO_2 laser power level less than 1 mW (75 μm f.w.h.m. spot diameter), in good agreement with the expected band-gap resonant nonlinearities (§3) (Parry *et al.* 1984). We have also studied a sample with a high-reflectivity metal coating applied to the rear surface to enhance the etalon finesse (Miller *et al.* 1984). This gave minimum and maximum reflectivities of 6 and 62% respectively in the 10.6 μm region. A 20:1 mark–space ratio chopper was placed before the sample to reduce thermal contributions. Figure 2 shows the total reflected power plotted against the input power at a wavelength of 10.48 μm for a given initial cavity tuning adjusted by making use of the small variation of optical thickness across the sample. The negative slope at 30–40 mW predicts an optical gain of 2.2. This nonlinear characteristic occurs at incident powers consistent with that predicted by a theoretical analysis of band-gap resonant nonlinearities including Auger recombination. An estimate of the nonlinear coefficient can be deduced from the incident intensities at which the maximum and minimum reflected powers occur, giving $\gamma = -3 \times 10^{-3}$ $(cm^2\ W^{-1})^{\frac{1}{3}}$, in good agreement with the calculated value of -5×10^{-3} (§3).

There are two principal difficulties in attaining optical bistability in CdHgTe with a c.w. laser. Auger recombination severely limits the size of the nonlinearity above about 10 W cm^{-2}, which causes higher-order resonances to occur at relatively high powers. The low thermal conductivity of CdHgTe means that these higher orders are difficult to reach. The second difficulty relates to the negative sign for the temperature coefficient of the refractive index. This is opposite to most other semiconductors and has the result that as a cavity approaches resonance due to the negative electronic nonlinearity, the rapidly increasing power absorbed

Downloaded from rsta.royalsocietypublishing.org

in the etalon pushes the cavity further into resonance. Thus even a small thermal contribution will always force the cavity into resonance. This is avoided in the case illustrated in figure 2 because the chopper allows the heat to dissipate between successive pulses. However, altering the initial tuning to give steeper nonlinear characteristics resulted in the etalon's always being swept into resonance (low reflectivity) by the thermal contribution. Nevertheless it should be possible to demonstrate differential gain, modulation transfer and optical logic functions at 10 μm with the characteristic shown in figure 2 if synchronously chopped beams are employed.

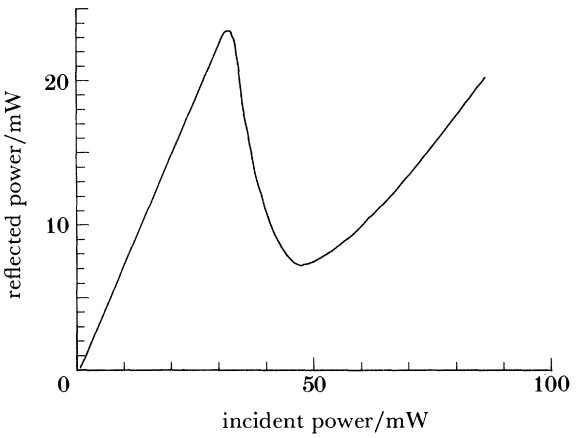

FIGURE 2. Input–output characteristic in reflection of 10.49 μm radiation with the use of a 208 μm thick $Cd_{0.23}Hg_{0.77}Te$ etalon at 77 K with a high reflectivity coating on the rear face (Miller *et al.* 1984).

5. TWO-PHOTON INDUCED OPTICAL SWITCHING AND BISTABILITY

The excitation of excess carriers by two-photon absorption has the attraction of causing refractive index changes at wavelengths where the linear absorption of the material is low. Optical switching by this mechanism has been reported recently in the transmission of a room-temperature InSb etalon at a few hundred kilowatts per square centimetre (Kar *et al.* 1983). The increase in band gap energy with temperature for CdHgTe makes the alloy composition used for the studies described in §4 ideal for room-temperature two-photon excitation. At room temperature $Cd_{0.23}Hg_{0.77}Te$ is intrinsic with concentration 1.5×10^{16} cm^{-3} and has a low-excitation Auger lifetime of 90 ns. An uncoated sample 200 μm thick was studied in transmission by using a short-cavity TEA CO_2 laser (Mathew *et al.* 1984). An estimated two-photon absorption coefficient of 5 cm MW^{-1} and the calculated nonlinear refraction (§3) predicts that 2.3×10^{16} cm^{-3} excess carriers (requiring a cavity intensity of *ca.* 100 kW cm^{-2}) should cause a single-pass phase change of π. At this density, the carrier lifetime will be approximately 20 ns, i.e. close to the laser pulse width.

Figure 3 shows the input pulse temporal profile (*a*) and transmitted profiles (*b–f*) for increasing peak incident intensities. Only the central region of the beam profiles were monitored with fast small-area detectors. Nonlinear Fabry–Perot effects are observed in these profiles at 100 kW cm^{-2} peak intensities as a dynamic modulation of the transmission as the intensity varies within the pulse. The maximum carrier concentration (and thus the maximum phase change) will occur after the peak of the input pulse owing to the effect of carrier lifetime. Thereafter the cavity will retrace its transmission state during the tail of the laser pulse on a timescale

Downloaded from rsta.royalsocietypublishing.org

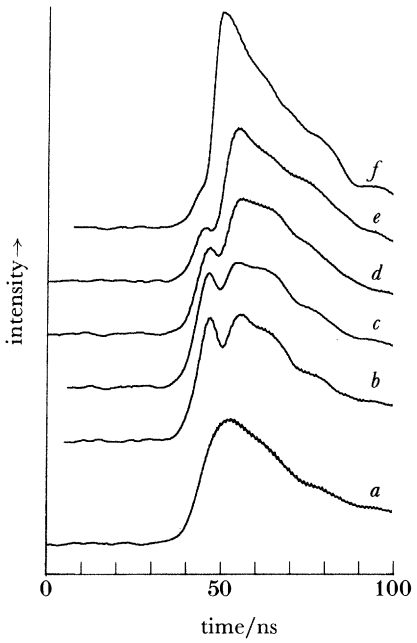

FIGURE 3. Transmitted 10.6 μm temporal (30 ns f.w.h.m.) pulse shapes for 208 μm thick, 300 K, $Cd_{0.23}Hg_{0.77}Te$ with polished faces compared with a typical input pulse (*a*). Peak incident intensities: (*b*) 116, (*c*) 130, (*d*) 165, (*e*) 323, and (*f*) 448 kW cm^{-2} (Mathew *et al.* 1984).

determined principally by the carrier recombination rate. At higher peak intensities, the first-order feature occurs earlier in the pulse and another feature appears at the peak of the pulse. At above 400 kW cm^{-2}, this second-order feature switches the transmission abruptly from low to high transmission in a time close to the resolution of the detection equipment. This switching is a rather complex dynamical effect due to the comparable pulse width and carrier lifetime.

Optical bistability was demonstrated by using longer duration pulses to give quasi-c.w. conditions (Craig *et al.* 1984). An uncoated sample at room temperature of the same composition and 363 μm in thickness was studied by using a hybrid TEA CO_2 laser producing 1.5 μs (f.w.h.m.) pulses. Figure 4*a* shows incident and transmitted temporal pulse profiles at 500 kW cm^{-2} peak intensity. In this case, a 100 μm diameter pinhole placed in contact with the exit face of the sample reduced the effect of etalon inhomogeneities. The plot of output against input signals (figure 4*b*) extracted from the temporal pulse shapes shows hysteresis due to optical bistability.

6. Conclusions

Low-temperature CdHgTe shows an extremely large optical nonlinearity at 10.6 μm for samples with band-gap energies close to the photon energy. This is significant for all-optical device applications in the 10 μm region because of the importance and widespread use of CO_2 lasers. As predicted, the nonlinearity is larger than that measured in InSb at 5 μm. However, CdHgTe has several disadvantages for optically bistable devices. It is appropriate here to compare these two materials.

1. CdHgTe is an alloy that is difficult to grow in a homogenous crystal. A Fabry–Perot is

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES
PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Downloaded from rsta.royalsocietypublishing.org

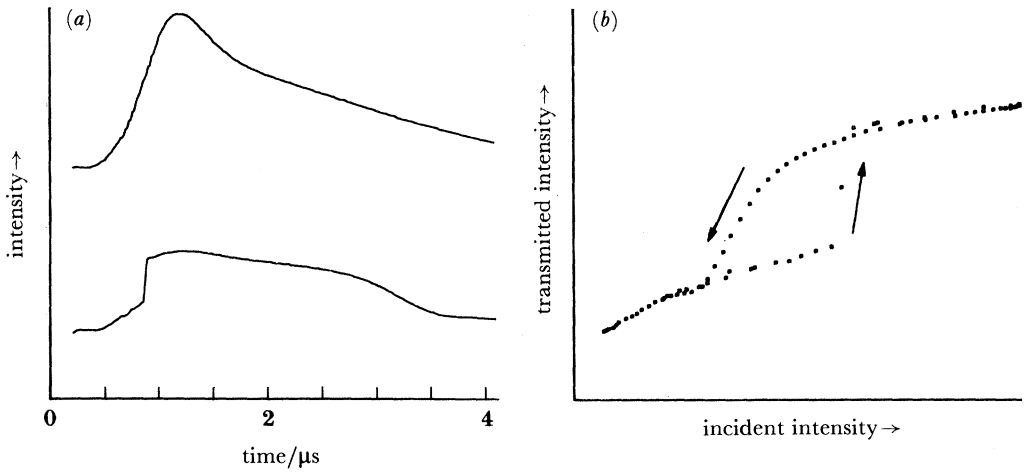

FIGURE 4. (*a*) Incident and transmitted temporal (1.5 μs f.w.h.m.) pulse shapes for 363 μm thick, 300 K, $Cd_{0.23}Hg_{0.77}Te$ with polished faces at 500 kW cm^{-2}. (*b*) Corresponding input–output characteristics showing optical bistability (Craig *et al.* 1984).

very sensitive to variations in refractive index with the result that etalon fringes can easily be washed out for moderate optical beam sizes unless the crystal is of very high quality.

2. The recombination mechanism at low temperature in CdHgTe is dominated by the non-radiative Auger mechanism, which is very dependent on carrier density, whereas low-temperature recombination in InSb is governed by a Shockley-Read process independent of carrier density at 77 K. Recombination in InSb at 77 K should however, become Auger-dominated once approximately 5×10^{15} cm^{-3} electron–hole pairs are excited.

3. The principal reason why c.w. optical bistability is very difficult to achieve in CdHgTe relates to the band-gap dependence with temperature. The negative refractive index change for both electronic and thermal contributions gives a positive feedback, which tends to sweep the cavity into resonance at a fixed input power. This is opposite to most other semiconductors in which the thermal refractive index coefficients are positive, and indeed the thermal effects will tend to stabilize the output on a steep nonlinear Fabry–Perot characteristic in these materials.

4. Pulsed inputs can overcome thermal problems and CdHgTe etalons behave in a similar way to InSb in two-photon induced optical bistability at room temperature. Carrier recombination in both materials is governed by Auger processes and the lifetimes are determined principally by the band-gap energies. A nonlinear refraction dependence according to $I^{\frac{2}{3}}$ results. Here CdHgTe has the advantage of the flexibility it offers in band-gap variation with composition to optimize the two-photon absorption coefficient, refractive index change per excited carrier and carrier lifetime.

We thank Mullard Limited for providing CdHgTe samples.

REFERENCES

Craig, D., Mathew, J. G. H., Kar, A. K. & Miller, A. 1984 (In preparation.)
Dornhaus, R. & Nimtz, G. 1976 In *Springer tracts in modern physics*, vol. 78, pp. 1–119, Berlin: Springer-Verlag.
Hill, J. R., Parry, G. & Miller, A. 1982 *Optics Commun.* **43**, 151–156.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES
PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Downloaded from rsta.royalsocietypublishing.org

Jain, R. K. 1982 *Opt. Engng* **21**, 199–218.
Jain, R. K., Giuliano, C. R., Klein, M. B., Lind, R. C. & Steel, D. G. 1981 In *Proceedings of the International Conference on Excited States and Multiresonant Nonlinear Optical Processes in Solids*, *Aussois* (Les Éditions de Physique, Orsay), pp. 4–5.
Jain, R. K. & Klein, M. B. 1979 *Appl. Phys. Lett.* **35**, 454–456.
Jain, R. K. & Steel, D. G. 1980 *Appl. Phys. Lett.* **37**, 1–3.
Jain, R. K. & Steel, D. G. 1982 *Optics Commun.* **43**, 72–77.
Kar, A. K., Mathew, J. G. H., Smith, S. D., Davis, B. & Prettl, W. 1983 *Appl. Phys. Lett.* **42**, 334–336.
Khan, M. A., Bennet,R. L. H. & Kruse, P. W. 1981 *Optics Lett.* **6**, 560–562.
Khan, M. A., Kruse, P. W. & Ready, J. F. 1980 *Optics Lett.* **5**, 261–263.
Khan, M. A., Kruse, P. W. & Wood, R. A. 1983 Presented at Conference on Lasers and Electro-Optics, May 1983, paper ThH6.
Kruse, P. W., Khan, M. A. & Ready, J. F. 1981 In *Wavefront distortions in power optics* (*S.P.I.E.* no. 293), pp. 183–189.
Kruse, P. W., Ready, J. F. & Khan, M. A. 1979 *Infrared Phys.* **19**, 497–506.
Mathew, J. G. H., Craig, A. & Miller, A. 1984 *Appl. Phys. Lett.* (In the press.)
Miller, A., Miller, D. A. B. & Smith, S. D. 1981 *Adv. Phys.* **30**, 697–800.
Miller, A. & Parry, G. 1984 *Opt. Quantum Electron.* **16**, 339–348.
Miller, A., Parry, G. & Daley, R. 1984 *IEEE Jl Quantum Electron.* **QE20**, 710–715.
Miller, D. A. B., Mozolowski, M. H., Miller, A. & Smith, S. D. 1978 *Optics Commun.* **27**, 133–136.
Miller, D. A. B., Seaton, C. T., Prise, M. E. & Smith, S. D. 1981 *Phys. Rev. Lett.* **47**, 197–200.
Parry, G., Miller, A. & Daley, R. 1984 In *Optical Bistability II* (ed. C. Bowden, H. Gibbs & S. McCall), pp. 289–296. New York: Plenum.
Yuen, S. Y. 1982 *Appl. Phys. Lett.* **41**, 590–592.
Yuen, S. Y. & Becla, P. 1983 *Optics Lett.* **8**, 356–358.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES