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Optical bistability observed in amorphous semiconductor films

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ABSTRACT

The optical properties of amorphous self-supporting $GeSe_2$ films show strong non-linear behaviour under the influence of relatively low-intensity c.w. laser irradiation. Discontinuities, bistability and, in special circumstances, oscillation in the optical properties have been found. The importance of thermal effects is experimentally proved. A theoretical model is constructed which explains some of the experimental findings. Evidence is presented, however, that, as well as thermal effects, photostructural changes also play an important role in the bistability and oscillatory phenomena.

§ 1. INTRODUCTION

In recent years there has been great interest in optical bistability, which may have potential applications in integrated optics. The effect was first discussed by Szöke, Daneu, Goldhar and Kurnit (1969), and first observed in vapour by Gibbs, McCall and Venkatesan (1976). More recently optical bistability has been found in different crystalline semiconductors (Gibbs, McCall, Venkatesan, Gossard, Passner and Wiegmann 1979, Miller, Smith and Johnston 1979). A review of the present experimental and theoretical situation is given by Abraham and Smith (1982).

Recently we reported that transmission oscillations occur in amorphous semiconductors under the influence of a c.w. laser beam of fixed intensity. This phenomenon was observed both in a-$GeSe_2$ films (Hajtó, Zentai and Kósa Somogyi 1977) and in bulk samples of As_2S_3 (Hajtó and Ewen 1978). The aim in this paper is to report a comprehensive study of non-linear optical phenomena in amorphous *self-supporting* $GeSe_2$ films. The oscillation of the optical properties in self-supporting films occurs only in very restricted circumstances. On the other hand, optical bistability and hysteresis was found. The experiments summarized in this paper help to clarify the underlying mechanism which causes non-linear optical phenomena.

Previously suggested mechanisms for crystalline semiconductors (Gibbs *et al.* 1979, Miller, Seaton, Prise and Smith 1981) cannot operate in our case. This is evident from the fact that if the film is crystallized (e.g. by an intense

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laser beam) the observed non-linearities disappear. Therefore we confine our attention to other types of non-linear interaction between the laser field and the amorphous material.

Recently Fazekas (1981) has proposed a theoretical model in order to explain this peculiar non-linear behaviour of semiconductor films. The model is based on the cooperative mechanism of charge disproportionation of defect centres in an amorphous network. In this model electronic processes are dominant and thermal effects are neglected.

In the present paper we consider an alternative explanation in which laser heating produces the observed optical anomalies. In our model the laser beam is considered as a source of local heating only, and the direct non-linear coupling between the laser field and the electronic and atomic structure of the medium is neglected. This is the opposite limit to that considered by Fazekas (1981) but, as we shall demonstrate, our model is also able to explain the observed anomalies in the optical properties of the films.

We emphasize, however, that we do not think that thermal effects are the only source of the observed phenomena. We present in this paper experimental evidence that another effect, namely laser-induced photostructural changes, also plays an important role. It is probable that all three of the effects described above (electronic, thermal and photostructural) are acting simultaneously under the influence of illumination.

The paper is organized as follows: in § 2 we describe the experimental arrangement, § 3 contains the experimental results on self-supporting $GeSe_2$ films, the theoretical considerations are presented in § 4, and § 5 contains some final conclusions.

§ 2. Experimental arrangement

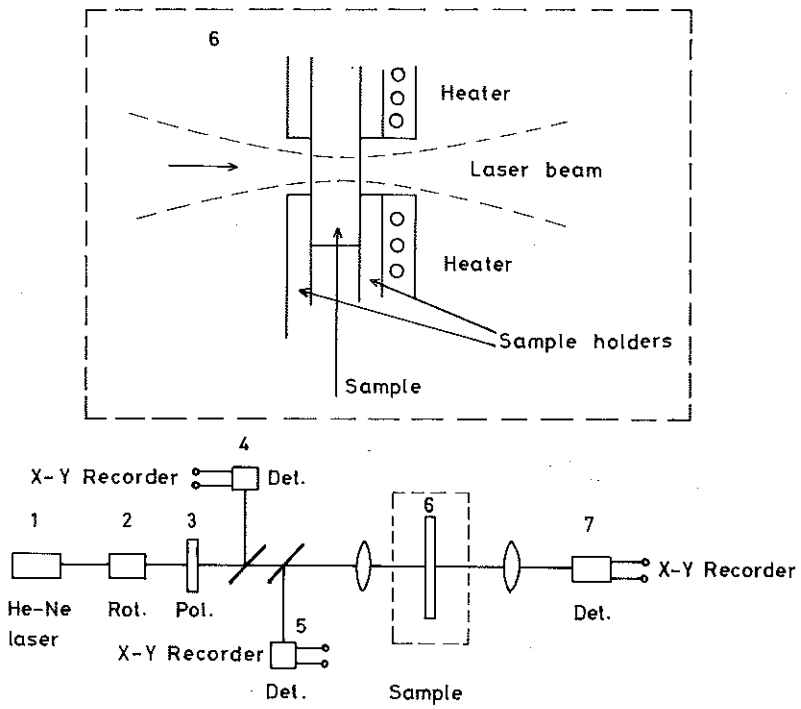
The a-$GeSe_2$ films were prepared by vacuum evaporation from a Ta boat at a pressure of 2×10^{-6} Torr ($2{\cdot}7 \times 10^{-4}$ Pa) onto glass substrates using polycrystalline $GeSe_2$ ingots as an evaporation source. For the measurements the films were removed from the glass substrates using an ultrasonic bath in order to reduce the effects of multiple light reflections and of mechanical stresses occurring at the $GeSe_2$–substrate interface.

For the measurements the films were placed in a sample holder, as shown in the inset of fig. 1. This arrangement allowed us to carry out optical measurements and regulate the temperature of the films. In the following, by the 'film temperature', T_F, we mean the temperature of the sample holder. We note that, because of absorption of the laser light, the temperature of the illuminated spot (spot temperature T_S) may differ from the 'film temperature' T_F.

For illumination we used a linearly polarized c.w. He–Ne laser beam which was focused on a Gaussian optical spot between 35 and 200 μm in diameter. The intensity of the incident laser beam was regulated using a combination of a Spectra-Physics polarization rotator (Model 310-21) and a sheet polarizer plane (2 and 3 in fig. 1).

The intensity of the incident, reflected and transmitted light was measured using Si photodetectors (4, 5 and 7 in fig. 1) and their signals were displayed on a Hewlett-Packard (Model 7046-A) X–Y recorder.

Fig. 1



Experimental arrangement. The inset shows the specimen in the sample holder.

§ 3. Results

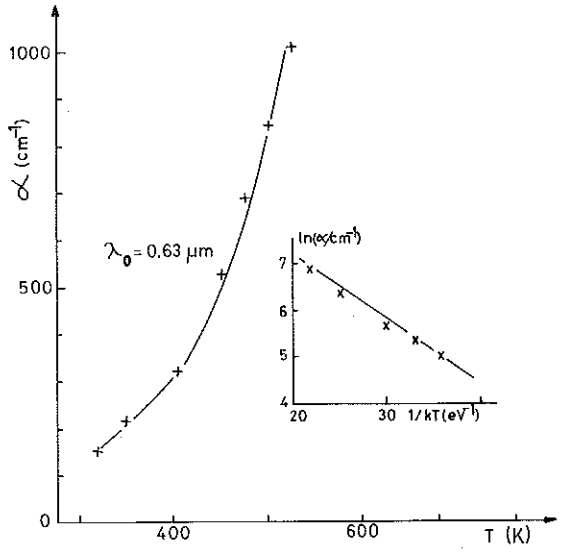
3.1. *Temperature dependence of absorption coefficient α of amorphous $GeSe_2$ films*

In this section we present measurements of the temperature dependence of the optical properties carried out at a low measuring light intensity where non-linear effects are negligible. The absorption coefficient α was determined from the reflected and transmitted intensity of a He–Ne laser beam ($\lambda_0 = 6328$Å) at a power density of 10^{-1} W cm^{-2}.

The photon energy of the He–Ne laser light ($h\nu = 1{\cdot}96$ eV at $\lambda_0 = 6328$ Å) used to induce the non-linear optical phenomena is less than the value of the optical energy gap of $GeSe_2$ films ($E_g = 2{\cdot}1$ eV ; Hajtó and Füstöss-Wégner 1980). In this energy range (below the band-gap energy) the optical absorption coefficient (10^2 cm$^{-1} < \alpha < 10^4$ cm^{-1}) varies exponentially with the photon energy (Hajtó 1980), which is the typical Urbach behaviour of chalcogenide glasses (Mott and Davis 1979). At a fixed wavelength in the Urbach region, α is strongly temperature-dependent above room temperature (Mott and Davis 1979). In this temperature region α can be described by the Urbach rule,

$$\alpha = \alpha_0 \exp\left(-\frac{E_g - h\nu}{k_B T}\right); \qquad (1\,a)$$

Fig. 2



The temperature dependence of the absorption coefficient α.

this rule was also obeyed in our experiments. A 6 μm thick amorphous $GeSe_2$ film was used for measuring α. Figure 2 shows that α is a strongly non-linear function of temperature. As shown in the inset, the Urbach rule is satisfied with $\alpha_0 = 2 \times 10^4$ cm^{-1} (at $\lambda_0 = 6328$ Å) and $E_g - h\nu = 0{\cdot}138$ eV in the temperature range 300–500 K. From the measurements the refractive index n could also be determined. We found, for temperatures of 300–500 K, that

$$n = n_0 + \beta T, \tag{1 b}$$

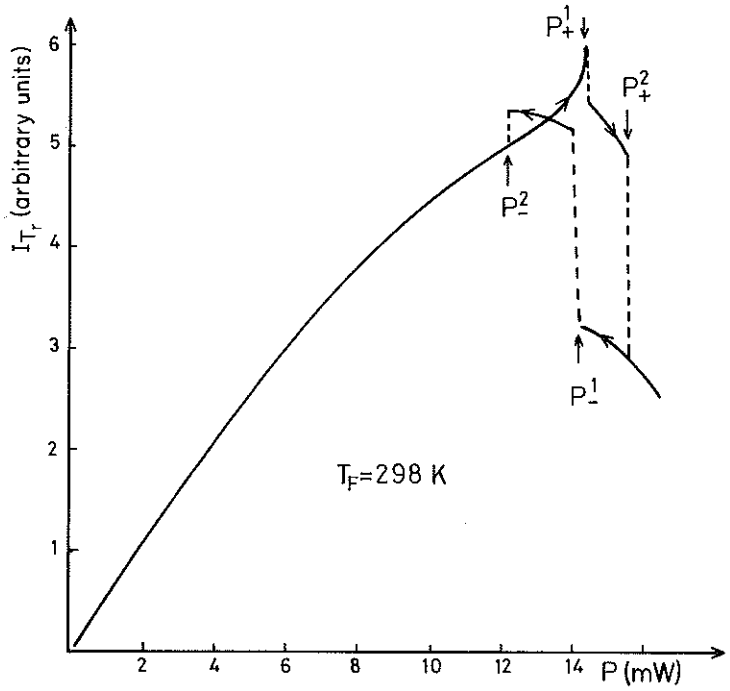
with $n_0 = 2{\cdot}50$ and $\beta = 2{\cdot}56 \times 10^{-4}$ K^{-1}, i.e. the refractive index is a linear function of temperature. This is in accordance with other experimental results (Kastner 1973). We emphasize that both α and n are *continuous* functions of temperature and that no discontinuity was found in the optical constants when the measuring light intensity was kept at a very low value. However, on increasing the incident light intensity, other types of optical effect started to develop ; these are described in the sections below.

3.2. *Laser-induced optical discontinuity and optical bistability in amorphous $GeSe_2$ films*

We have measured the actual transmission of a c.w. He–Ne laser beam focused onto a self supporting amorphous $GeSe_2$ film as a function of the incident laser intensity. Our optical system (air–sample–air configuration) is a very simple one, no external mirrors being used to form a Fabry–Perot system.

In this section we present the experimental results on the transmitted light intensity to demonstrate the non-linear nature of the optical phenomena. The simultaneous transmission and reflection measurements (necessary for calculating the optical constants) will be presented in § 4.3, where the experimental results are compared with our model. All measurements presented in this section were carried out by changing the incident laser intensity at a constant rate ($\sim 0{\cdot}1$ mW s^{-1}). The recorded values of the transmitted light intensity (I_{Tr}) as a function of the incident laser intensity are shown in fig. 3 for $T_{\mathrm{F}} = 298$ K.

Fig. 3

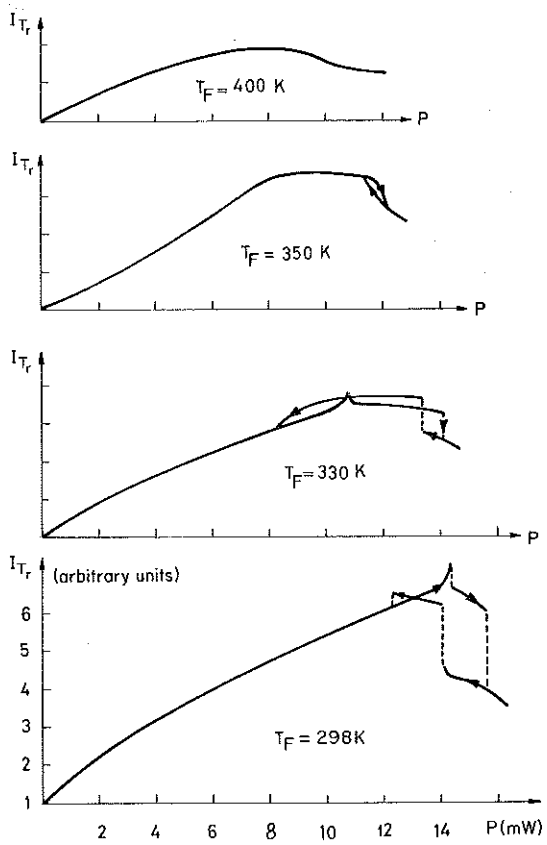


Transmitted light intensity as a function of the incident laser intensity in a-$GeSe_2$ film ($L = 6$ μm, laser spot diameter = 207 μm).

A first discontinuity was observed at 14·4 mW incident laser power (P_+^1), characterized by a sharp increase and subsequent discontinuous decrease in the transmitted signal. On increasing further the incident laser power, a switch to the dark state was observed, i.e. a large decrease in the transmitted signal at 15·6 mW (P_+^2).

Starting from the dark state and decreasing the laser power, the transmitted light showed a hysteresis, e.g. the transition to the bright state (higher transmitted values) occurs at 14·2 mW (P_-^1), which is lower than P_+^2. As a consequence the transmitted light intensity has two different stable values at a fixed laser power between P_+^2 and P_-^1, i.e. optical bistability was found. On further decreasing the laser power, a second discontinuity was observed at 12·3 mW (P_-^2), where the hysteresis disappeared.

Fig. 4

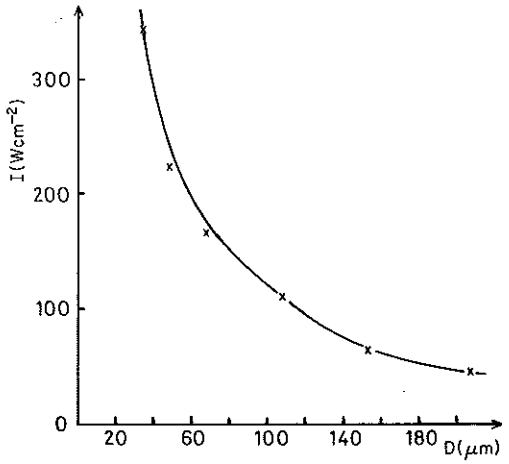


Transmitted light intensity as a function of the laser intensity at different film temperatures T_F.

We found that the shape of the recorded curves varied somewhat with experimental conditions such as the temperature of the films, laser spot diameter and the rate of increase of the intensity. The last of these effects is discussed in § 3.3. The effect of the film temperature is shown in fig. 4. Increasing the film temperature T_F by using the sample heater, as described in § 2, the critical laser powers for producing the optical discontinuities decreased. Similarly, the amplitudes of the optical discontinuities decreased with increasing film temperature. A decrease in the hysteresis $(P_+^2 - P_-^1)$ with increasing film temperature was also observed. The optical discontinuities and the optical bistability disappeared when the film temperature was increased above 500 K.

We have measured the critical laser power P_-^1 (at which the first switching from the dark to the bright state occurs) as a function of the incident laser beam diameter. We chose this discontinuity (P_-^1) because it seemed to be the most reproducible one ; i.e. it does not depend strongly on the rate of decrease of the intensity. The critical laser flux density (P_-^1 divided by the area of the laser spot) as a function of the laser spot diameter is shown in fig. 5.

Fig. 5



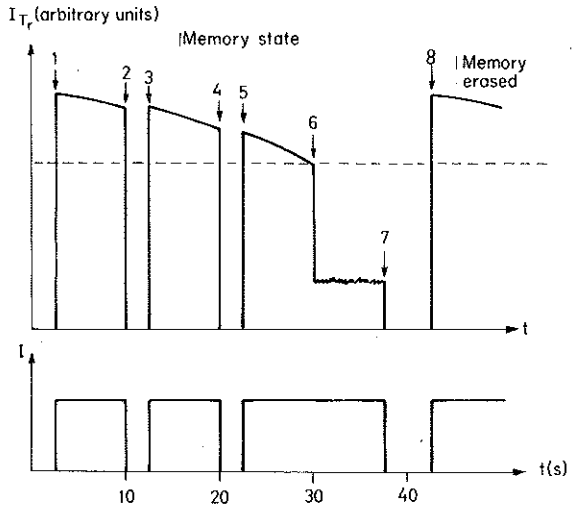
Critical laser flux density as a function of incident laser beam diameter.

Assuming a pure electronic excitation effect for producing optical bistability (Fazekas 1981), one would expect the switch to the dark state to occur at a constant electric field produced by the laser, i.e. at constant laser flux density independent of the laser beam diameter. Figure 5 shows that this is not the case for the a-$GeSe_2$ films. The laser flux density necessary for producing the optical switching effect increases with decreasing laser beam diameter. This experimental observation indicates the importance of thermal effects in connection with the laser-induced non-linear phenomena in a-$GeSe_2$ films. The increase in the spot temperature T_S at a given laser flux density is less when the beam diameter is decreased because of greater heat conduction. Consequently, the laser flux density must be higher to produce a given spot temperature.

3.3. *Photostructural changes*

As mentioned in the previous section, the shape of the transmission curves depends on the rate of increase of the intensity. We established that this dependence can be traced back to another effect, namely that the transmission changes, even under the influence of a *constant* laser power. The details of this effect are shown in fig. 6. In this experiment we recorded the transmitted light intensity at a fixed laser power, which was lower than the critical intensity at which the optical switching occurs to the dark state. In the first period of illumination a continuous decrease in the transmission was observed. This decrease cannot be attributed simply to the increase in the spot temperature, because it possesses a ' memory ' behaviour. Switching off the light (points 2 and 4 in fig. 6) and keeping the sample in the dark for a while does not restore the initial transmission value. On switching the light on again (points 3 and 5 in fig. 6), the transmission continues to decrease from the same value as before. This is a memory state, which must be connected with photostructural changes

Fig. 6



Time dependence of the transmitted light intensity at a fixed laser power ($P = 11$ mW, laser beam diameter $= 207$ μm).

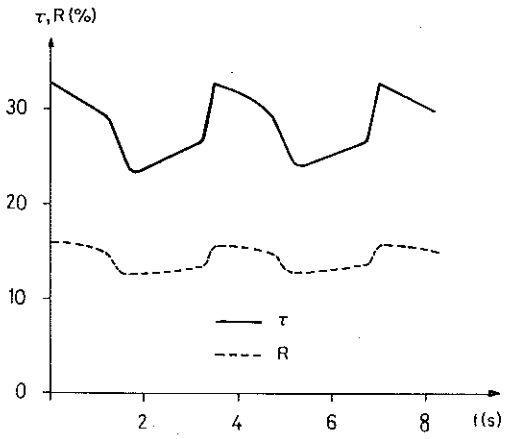
(de Neufville 1976). Different aspects of the laser-induced structural changes in a-$GeSe_2$ were studied recently by Griffiths, Espinosa, Remeika and Phillips (1982) and by Hajtó, Jánossy and Forgács (1982).

When the transmission decreases to a critical value (point 6 in fig. 6), a discontinuous switching to the dark state occurs. We observed that switching off the light in the dark state (point 7 in fig. 6) causes the ' memory ' to be erased ; the initial transmission value is restored (point 8 in fig. 6) and the whole cycle recommences. This fact indicates that in the dark state a different type of photostructural change takes place which erases the structure of the amorphous network developed in the bright period (above the dashed line in fig. 6).

3.4. *Laser-induced oscillatory phenomena*

We found experimentally that, for an appropriate laser power, the dark state is unstable and it switches back spontaneously to the bright one. In this case periodic oscillation of the optical properties occurs (see fig. 7). Such oscillations had already been found and reported (Hajtó *et al.* 1977) in $GeSe_2$ films on a silica substrate. The differences between self-supporting samples and samples on a substrate are the following. In the latter case a much higher laser intensity (~ 2 kW cm^{-2}) was necessary to produce oscillations, but above a critical laser field oscillations always occur. In the present case oscillation was found only in a rather narrow intensity range which corresponds more or less to the observed hysteresis (see fig. 3). In this range the intensity is of the order of 40 W cm^{-2}, i.e. smaller by a factor of 50 than the intensity for samples on a substrate. As can be seen from fig. 7, the frequency of the oscillation is of the order of few seconds, while for samples on a substrate it is of the order of hundredths of a second (Hajtó 1980).

Fig. 7



Oscillation of the transmitted and reflected light intensity at a fixed laser power ($P = 14{\cdot}5$ mW, laser beam diameter $= 207$ μm).

§ 4. Theoretical considerations

4.1. *General remarks*

To explain the experimental observations described in the previous section, one has to consider the possible non-linear interactions between the laser field and the amorphous film. We suggest that there are three types of such interactions that may play an important role in the present phenomena.

Thermal effects

As the chalcogenide glasses are poor thermal conductors, the laser beam may produce a significant local temperature rise in the illuminated spot. In turn, the absorption coefficient α, the value of which determines the dissipation of the light field in the sample—and thus the temperature rise, is strongly temperature-dependent (see § 3.1). A simple quantitative treatment of the thermal effects is given in § 4.2, where it is shown that this feedback between the value of α and the temperature rise can give rise to a highly non-linear behaviour of the optical characteristics of the film and may produce discontinuities and bistability in the transmitted light intensity. The dependence of the critical laser intensities as a function of the spot size (see § 3.2) suggests the importance of thermal effects.

Photostructural changes

As is well known from the literature (de Neufville 1976, Tanaka 1980), and also as shown by our observations, laser irradiation causes structural changes in the amorphous network (see § 3.3). These structural changes, which are connected with displacements of atoms, modify the optical constants of the substance. As a result, the absorption coefficient at a given spot depends not only on the local temperature, but also on the history of the amorphous network within that spot.

The ' memory effect ' described in § 3.3 indicates that the photostructural changes in a-$GeSe_2$ are not restricted to a short initial period of the illumination. The observations can be interpreted by assuming that the stationary configuration of the structure is a function of the illuminating laser intensity and the temperature. The stationary configuration, obtained by a long illumination at a fixed laser intensity and at a *fixed* spot temperature, can be altered by changing one or both of these parameters. We note that in the experiments the spot temperature is *not* fixed. In this case it is not necessary, at a constant laser intensity, for a stationary state to be established, i.e. oscillation can occur. This point is discussed further in § 4.4.

' Conventional ' non-linearity

There may also be a ' conventional ' non-linearity in the system, arising from the interaction of the laser field with the electronic structure of the amorphous material. Such an interaction was investigated by Fazekas (1981), who considered the effect to be due to charged and neutral defect centres and demonstrated that the interaction between these centres may lead to a discontinuous change in the electronic structure at a critical laser intensity.

Even if this laser-induced ' phase transition ' was not the origin of the observed discontinuities, one has to consider the possibility that the absorption coefficient may depend explicitly (i.e. not through thermal effects) on the laser intensity.

In our opinion all three effects described above are important in the observed phenomena. However, the measurements carried out up to now are not sufficient to enable their relative importance to be properly estimated. In the following section we consider quantitatively the effect of the thermal component only. This is the opposite limit to that considered by Fazekas (1981) but, as we shall demonstrate, our model is also able to explain some of the observed optical anomalies.

4.2. *Quantitative treatment of the thermal effects*

Let us consider a plane parallel, self-supporting film of thickness L of a given substance. The substance can be characterized by a complex refractive index $\hat{n}=n+i\kappa$, where κ is related to the absorption coefficient by $\alpha=(4\pi/\lambda_0)\kappa$. In this treatment n and α are considered to be single-valued, continuous functions of the temperature. We investigate the case of normal incidence of the light beam and, for simplicity, replace the Gaussian intensity profile of the laser beam by a uniform intensity, I, which is cut off at a radius r_0.

The transmission and reflection coefficients of the film can be calculated by Abelès' method (Born and Wolf 1968). In the limit $\kappa \ll n$, the transmission and reflection coefficients are respectively

$$\tau = 1/(n_1^2\,\Gamma + n_2^2/\Gamma - 2n_1n_2\cos 2\delta), \tag{2}$$

$$R = n_1n_2(\Gamma + 1/\Gamma - 2\cos 2\delta)\tau, \tag{3}$$

where

$$n_1=(n+1)^2/4n, \quad n_2=(n-1)^2/4n$$

$$\Gamma=\exp(\alpha L), \qquad \delta=\frac{2\pi}{\lambda_0}nL.$$

The energy dissipated per unit time and unit area, W, can be determined from the energy conservation law :

$$W = ID, \quad D = 1 - (R + \tau). \tag{4}$$

To determine the temperature distribution in the sample, it is necessary to know how the dissipated heat is conducted away from the spot. We consider two limiting cases.

(1) The heat is conducted away in the direction normal to the surfaces by the surrounding air. The heat flow can be taken to be proportional to $T_S - T_F$. The time dependence of the spot temperature T_S is then governed by the equation

$$\frac{\partial T_S}{\partial t} = \frac{I}{cL} D(T_S) - k(T_S - T_F), \tag{5}$$

where c is the specific heat of the substance and k is a constant.

(2) In the opposite limit we assume that heat conduction takes place only in the plane of the sample. The temperature is assumed to be fixed (T_F) at a radius R. The heat conduction equation in this case is

$$\frac{\partial T}{\partial t} = \begin{cases} \dfrac{I}{cL} D(T(r)) - \lambda \left(\dfrac{\partial^2 T}{\partial r^2} + \dfrac{1}{r} \dfrac{\partial T}{\partial r} \right), & r < r_0 \\ - \lambda \left(\dfrac{\partial^2 T}{\partial r^2} + \dfrac{1}{r} \dfrac{\partial T}{\partial r} \right), & r > r_0, \end{cases} \tag{6}$$

where r_0 is the radius of the laser beam and λ the heat conductivity. At $r = r_0$ we require T and $\partial T/\partial r$ to be continuous.

For $r > r_0$ the solution can be written as

$$T = A \ln (R/r) + T_F. \tag{7}$$

For $r < r_0$, eqn. (6) can be solved, for example by expanding T and D in powers of r. This gives

$$\left.\begin{aligned} T(r) &= T_0 - Br^2 + \ldots \\ D(T(r)) &= D(T_0) + \left.\frac{\partial D}{\partial T}\right|_{T=T_0} (T - T_0) + \ldots \\ &= D(T_0) - B \left.\frac{\partial D}{\partial T}\right|_{T=T_0} r^2 + \ldots, \end{aligned}\right\} \tag{8}$$

where $T_0 = T(0)$. Taking into account that $R \gg r_0$, it can be assumed that the temperature drop within the spot is not too large, so that fourth-and higher-order terms in r can be neglected. In this case

$$T(r_0) \approx T_0 - Br_0^2, \quad T(r_0) \approx -2Br_0$$

and the continuity conditions give

$$B = \frac{T_0 - T_F}{r_0^2 [1 + 2 \ln (R/r_0)]}.$$

Equations (6) and (8) yield

$$\frac{\partial T_0}{\partial t}=\frac{I}{cL}D(T_0)-\frac{\lambda}{r_0^2[1+2\ln(R/r_0)]}(T_0-T_F).$$

Identifying T_0 with the ' spot temperature ' T_S, we have the same equation as in case (1), eqn. (5), with

$$k=\frac{\lambda}{r_0^2[1+2\ln(R/r_0)]}. \tag{9}$$

In a real experiment both types of heat conduction occur simultaneously. However, as we have seen, in both limits the same mathematical equation describes the problem. We now investigate the properties of this equation, writing it in the form

$$\tau_0\frac{\partial T}{\partial t}=\eta I D(T)-(T-T_F) \tag{10}$$

(from now on we omit the subscript S from T_S). η is a constant determined by the geometry.

We are interested in stationary solutions, when

$$\eta I D(T)=T-T_F. \tag{11}$$

To solve eqn. (11) explicitly, the temperature dependences of α and n have to be known. As discussed in § 3.1, for amorphous semiconductors these can be given as (see eqns. (1 *a*) and (1 *b*))

$$\alpha(T)=\alpha_0\exp\left(-(E_g-h\nu)/k_BT\right],$$
$$n(T)=n_0+\beta T.$$

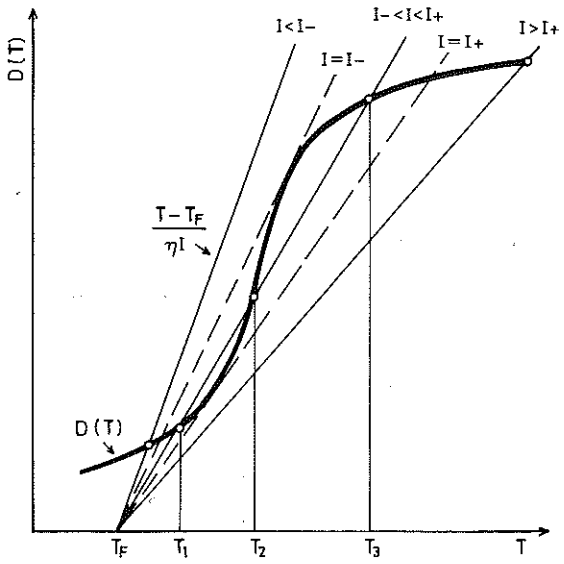
To demonstrate how discontinuities and bistability can occur, let us first neglect the temperature dependence of the refractive index. A schematic curve representing $D(T)$ for this case is given in fig. 8. At low temperatures, where $\exp(\alpha L)-1\ll 1$, $D(T)$ is proportional to $\alpha(T)$. At high temperatures where $\exp(\alpha L)\gg 1$, $D(T)$ tends to a saturation value, $1-[(n-1)/(n+1)]^2$ (i.e. all light entering the sample is absorbed). Equation (11) can be solved graphically by determining the intercepts of $D(T)$ and the straight line $(T-T_F)/\eta I$. As can be seen from fig. 8, if $I<I_-$ or $I>I_+$, there is only one solution for T. However, in the range $I_-<I<I_+$ there are three solutions.

To decide whether a given solution is stable or not, one has to consider what happens if the temperature deviates slightly from its equilibrium value. If the sign of $\partial T/\partial t$ is such that the temperature approaches the equilibrium value, the solution is stable ; in the opposite case it is unstable. From eqn. (10) we deduce the condition of stability to be

$$\frac{\partial}{\partial T}\left(D(T)-\frac{T-T_F}{\eta I}\right)<0. \tag{12}$$

This inequality is satisfied for the solutions denoted by T_1 and T_3 in fig. 8. Consequently, in the intensity range $I_-<I<I_+$ there are two stable solutions. The first solution, T_1, corresponds to a cold and transparent state of the film ; this happens when the laser intensity is increased from 0. The state becomes unstable at $I=I_+$, where a discontinuous transition takes place to a warm and

Fig. 8



Determination of the spot temperature at different laser intensities ; n is considered to be constant.

strongly absorbing state. When the intensity is decreased from a value larger than I_+, the film remains in this ' warm ' state down to a second critical intensity, I_- ($I_- < I_+$). At this intensity the film transforms back to the ' cold ' state.

The behaviour described above is typical of optical bistability (Abraham and Smith 1982). Note, however, that in the present mechanism the reflection at the boundaries does not play an important role, and that the switching is from the transparent to the dark state when the intensity is increased. These features are the opposite of what is found in conventional optical bistability.

In the above considerations the variation of the refractive index with temperature has been neglected. In fig. 9 we demonstrate that this variation, although weak, may itself also lead to bistability. Keeping α a non-zero constant, D oscillates with increasing T because of the temperature dependence of the term $\cos 2\delta$ in eqns. (2) and (3). As shown in fig. 9, in this case there is again an intensity range in which two stable spot temperatures belong to a given I. (At higher intensities further bistable ranges exist.) In this second mechanism of bistability the reflections of the light beam at the boundaries play an important role. However, we emphasize that the bistability is again caused by the feedback between the temperature rise of the spot and the temperature dependence of the optical constants.

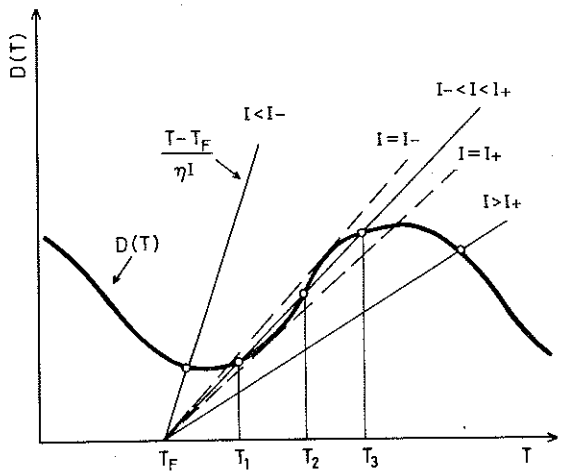
In fig. 10 we present schematically the transmission coefficient τ as a function of laser intensity when only α (fig. 10 (a)) and only n (fig. 10 (b)) depends on the temperature. In both cases the ' dynamical transmission coefficient ',

defined as $\partial\tau/\partial I$, becomes infinite at the critical intensities. It can be shown that near the critical intensities

$$\frac{\partial\tau}{\partial I}\sim(I-I_c)^{-1/2}. \tag{13}$$

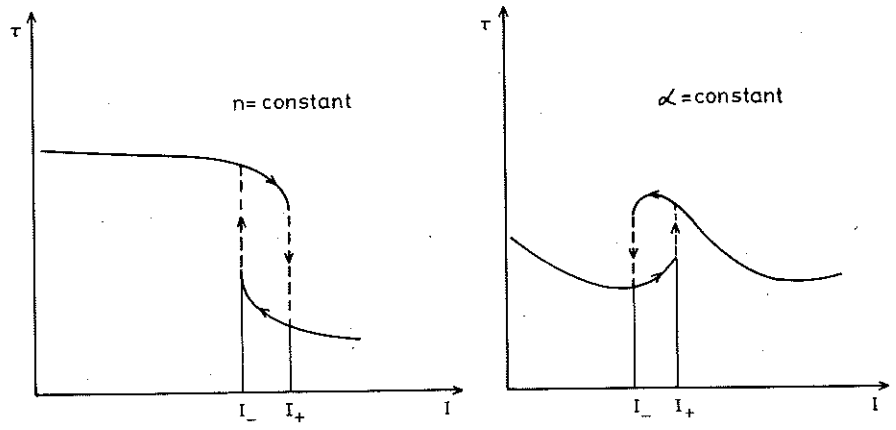
However, the direction of the jump of τ in one case is the opposite of that in the other.

Fig. 9



Determination of the spot temperature at different laser intensities ; α is considered to be constant.

Fig. 10



(*a*) (*b*)

Transmission coefficient as a function of laser intensity ; (*a*) $n=$constant, (*b*) $\alpha=$constant.

In a real situation, when both α and n are temperature dependent, interference between the two effects leads to different complicated shapes and singularities in the curves of transmission versus incident intensity (see the next section).

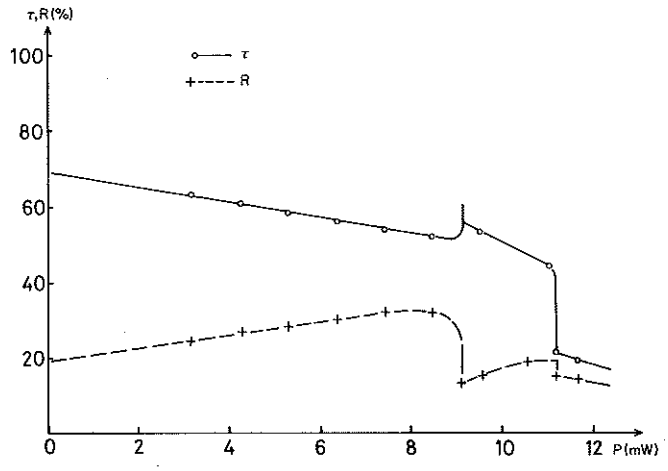
4.3. *Application of the model to a-$GeSe_2$ films*

In this section we present some calculated transmission and reflection curves as a function of light intensity in which we utilized the experimentally determined $\alpha(T)$ and $n(T)$ values of a-$GeSe_2$ films (see § 3.1). The shape of the curves also depends on the thickness of the films. The problem arises that the term $\cos 2\delta$ (see eqns. (2) and (3) in § 4.2) is very sensitive to the exact value of the thickness. Therefore in the calculation we used the relation $\delta = \delta_0 + 2(\pi/\lambda_0)L\beta T$ and δ_0 was considered to be an independent parameter. The calculation showed that the shape of the curves is very sensitive to the value of δ_0. This may explain the experimental finding that in samples with slightly different thicknesses the detailed shapes of the curves of transmission versus intensity were rather diverse.

In fig. 11 the experimentally determined transmission (τ) and reflection (R) coefficients are plotted against incident laser power. For comparison, the curves of τ and R versus laser power calculated according to the thermal model (see § 4.2) are shown in fig. 12. The value of δ_0 was chosen such that the calculated and measured τ and R should be the same at zero intensity. Although there is no quantitative agreement between the curves in figs. 11 and 12, the model reflects the non-linear behaviour and, in particular, the discontinuities both in transmission and reflection.

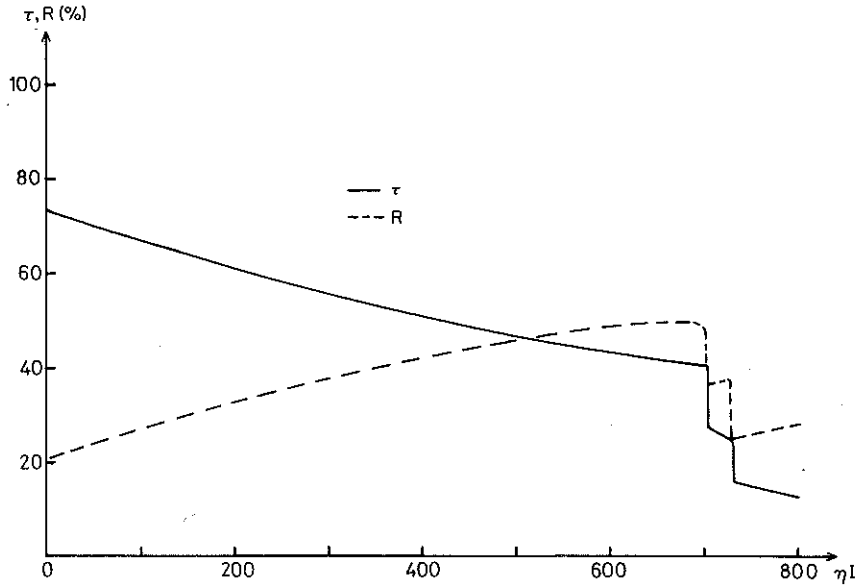
In fig. 13 the experimentally determined values of the absorption coefficient α are compared with the theoretically calculated values. There are two sharp increases in α at the observed optical discontinuities, both in the experimental

Fig. 11



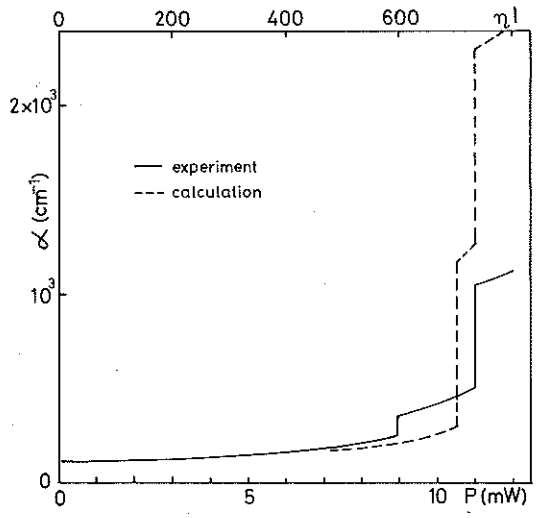
Experimentally determined τ and R values as a function of laser power for a laser beam diameter of 160 μm.

Fig. 12



Calculated τ and R values as a function of laser intensity using $\delta_0 = 2{\cdot}21$. (ηI are given in degrees Kelvin.)

Fig. 13



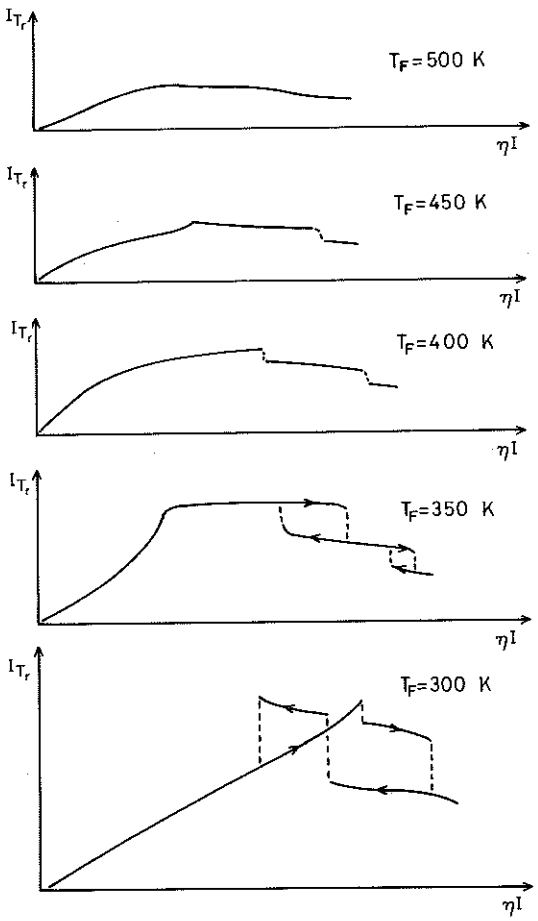
Absorption coefficient α as a function of laser power.

and theoretical curves. However, the model predicts a higher increase in α than was found experimentally.

The model predicts that the critical laser intensity I_c at which the optical switching occurs depends strongly on the spot size. Considering heat conduction to occur only in the plane of the film (see § 4.2, case (2)), we would expect from eqn. (9) that $I_c \sim 1/r_0^2[1 + 2 \ln (R/r_0)]$. This is in accordance with the experimental observation that I_c increases with decreasing laser spot size (see § 3.2).

The model allows us also to investigate the influence of the film temperature. Figure 14 shows the calculated transmitted light intensity I_{Tr} as a function of laser intensity at different film temperatures T_F. In these calculations δ_0 was chosen such that the room-temperature curve should resemble the experimental curve presented in fig. 3. One can see the same tendencies in

Fig. 14



Calculated transmitted light intensity as a function of film temperature using $\delta_0 = 2{\cdot}85$.

fig. 14 as in the experimentally recorded curves (fig. 4) : that both the amplitude of the discontinuities and the hysteresis decrease with increasing film temperature. We made no attempt to determine the absolute value of the incident laser intensity in the theoretical calculations described above ; this would require knowledge of the value of the term η introduced in eqn. (10).

Considering only the thermal effect, the model is able to reproduce the general trends of the observed phenomena. However, the influence of the increase in film temperature is more pronounced in the experiments than is predicted by our thermal model.

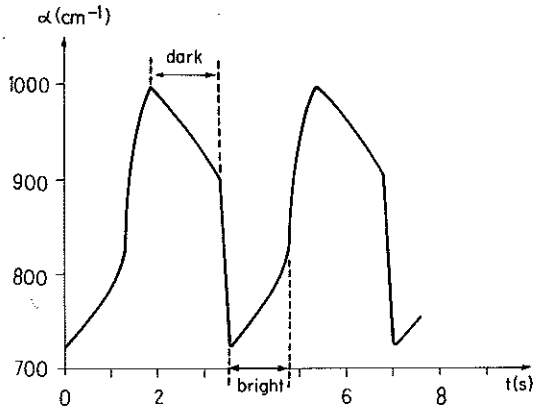
It is clear that we cannot obtain quantitative agreement between experiment and theory without taking into account other effects produced by the irradiation (see, for example, points (2) and (3) in § 4.1). This is particularly true for oscillatory phenomena (see § 3.4) which could not be explained by a purely thermal model.

4.4. *Considerations concerning the oscillatory phenomena*

We suggest that the occurrence of oscillation under certain experimental conditions (see § 3.4) at a fixed laser intensity can be explained by a combination of thermal effects and photostructural changes. To see more precisely how the oscillation occurs, we calculated the oscillation of the absorption coefficient α from the recorded transmission and reflection curves in fig. 7. The result is shown in fig. 15. In the bright state of oscillation (lower α values), α is increasing. The phenomenon has already been discussed (see § 3.3) and has turned out to be connected with photostructural changes. As α increases due to the photostructural change, the spot temperature also increases due to the higher dissipation. This can lead to a discontinuity in the same way as for increasing laser intensity.

In the dark state (higher α values), where the spot temperature is supposed to be higher, the laser irradiation causes a continuous *decrease* in the absorption

Fig. 15



Calculated oscillation of the absorption coefficient using the experimental data presented in fig. 7.

coefficient. At the elevated temperature the direction of the slow photostructural change is reversed, and the spot temperature decreases continuously. This decrease can lead to a discontinuous switch to the bright state (lower α values).

A model for photostructural changes in chalcogenide glasses was presented by Kolobov, Kolomiets, Konstantinov and Lyubin (1981), based on the double well potential description proposed by Tanaka (1980). The model predicts that, on raising the spot temperature, the direction of the photostructural change may reverse. A quantitative treatment of this problem will be given by us in a forthcoming paper.

As mentioned above, there are marked differences in the oscillatory phenomena in samples with and without substrates (self-supporting). The role of the substrate may be that it enables the heat produced by the laser to be conducted away more efficiently. Consequently, thermal effects are reduced and the material can tolerate a much higher laser field without crystallizing. The higher laser field enhances the photostructural effects rather than the thermal effects.

§ 5. Conclusion

We have shown that amorphous semiconductor films display a rich variety of non-linear optical phenomena. We have observed a new type of optical bistability in amorphous $GeSe_2$ films which cannot be explained by the same mechanism (Miller *et al.* 1981, Gibbs *et al.* 1979) used for crystalline semiconductors.

The non-linear behaviour in the present case may be attributed to at least three different mechanisms, namely thermal effects, photostructural changes and electronic non-linearity. Considering only electronic non-linearity, as did Fazekas (1981), one can explain the discontinuity in the optical properties but cannot account for the memory effect and hysteresis. The purely thermal model presented in this paper explains the discontinuity, the hysteresis and the fact that the critical laser power densities depend on the laser beam diameter. On the other hand, the memory effect and the corresponding oscillations cannot be explained.

In order to explain quantitatively all the observations, a model is needed which combines the three mechanisms (thermal, photostructural andel ectronic) acting simultaneously during the illumination. In view of this, further optical measurements are planned in order to separate the photostructural changes and electronic non-linearity from the thermal effect.

Finally, we emphasize that amorphous semiconductor films could provide simple and easily accessible elements in some optical applications. Because of the optical singularities (see, for example, fig. 3 and eqn. (13)), the films can be used as a new type of optical transistor.

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