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Explanation of the laser-induced oscillatory phenomenon in amorphous semiconductor films

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ABSTRACT

A new model is presented which explains the recently observed oscillatory phenomena in self-supporting a-GeSe$_2$ films.

The model is based on a combination of thermal and photoinduced effects. It can also be used for predicting the experimental conditions for observing light-induced bistability and oscillatory phenomena in other amorphous semiconductor films.

§ 1. INTRODUCTION

In recent years considerable interest has developed in connection with non-linear optical properties of condensed matter. Of special interest are the non-linear optical phenomena that occur at relatively low levels of light intensity, as these effects may find applications in integrated optics. Perhaps the most promising effect is the intrinsic optical bistability in crystalline semiconductors observed by Gibbs, McCall, Venkatesan, Gossard, Passner and Wiegmann (1979) and Miller, Smith and Johnston (1979) (see also Abraham and Smith 1982).

In the past few years it has been reported that strongly non-linear optical phenomena occur in amorphous evaporated GeSe$_2$ films. The first observations were made on a-GeSe$_2$ films on silica substrates. It was found that under the influence of a continuous focused He–Ne laser beam, the transmission and reflection coefficients of the film show periodic oscillations in time, above a threshold ~ 2 kW cm^{-2} (Hajtó, Zentai and Kósa Somogyi 1977, Hajtó and Apai 1980, Hajtó 1980).

More recently, the optical behaviour of self-supporting a-GeSe$_2$ films has been studied (Hajtó, Jánossy and Forgács 1982). In this case non-linearity occurs at much lower intensity levels (~ 50 W cm^{-2}). Oscillation of the optical properties occurs only in a narrow intensity range. Bistability and hysteresis were also found, without placing the sample in an optical resonator.

Different mechanisms have been suggested to explain this peculiar optical behaviour. Fazekas (1981) proposed that the observed anomalies are due to a

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collective phenomenon of the charged and neutral defects present in evaporated a-$GeSe_2$ films. Recently Phillips (1982) suggested that the oscillatory behaviour is closely connected to the laser-induced reversible microcrystallization, which was observed in a-$GeSe_2$ with the help of Raman spectroscopy (Griffiths, Espinosa, Remeika and Phillips 1982).

Recently (Hajtó and Jánossy 1983) an alternative interpretation has been considered in which laser heating produces the optical anomalies. This model gives a natural explanation of the differences in the behaviour of self-supporting samples and those on substrates. In the latter case the heat is conducted away much more efficiently from the illuminated spot and consequently in this case a much higher laser field is necessary to produce the same thermal effect. The pure thermal model explains the optical bistability, in terms of a mixture of 'absorptive' and 'dispersive' types of bistability (Abraham and Smith 1982). Furthermore it accounts for the observation that the critical laser power densities depend on the beam diameter. On the other hand it does not explain the oscillatory behaviour.

It was suggested by us that the oscillations can be explained by taking into account the photostructural changes induced by the laser beam in chalcogenide glasses (see, for example, Tanaka 1980). The aim of the present paper is to treat the combined effect of the laser heating and photostructural changes quantitatively and to show how these effects can produce oscillations. We confine ourselves to *reversible* photostructural changes (Tanaka 1980).

The photostructural changes are usually much slower than the thermal effects. Thus the pure thermal model (Hajtó and Jánossy 1983) is relevant for experiments in which the intensity is scanned fast enough to prevent significant photostructural changes. In the present paper we discuss the opposite limit, i.e. what happens at *fixed* intensity. In the theoretical description, presented in § 2, we consider for the sake of simplicity only the changes of the absorption coefficient. In reality the refractive index is also temperature dependent and shows photo-induced changes (Tanaka 1980, Hajtó *et al.* 1982). This fact adds a number of complications to the phenomenon. We think however that our simplified description reflects the main features of the underlying mechanism. Also we show in the discussion (§ 3) this mechanism should also lead to oscillations in other chalcogenide amorphous semiconductors, provided that the relevant parameters (wavelength of light, temperature and thickness of the film) are properly chosen.

§ 2. THEORETICAL CONSIDERATIONS

2.1. *Thermal effects*

A previous treatment of thermal effects was based on the strong temperature dependence of the absorption coefficient α (at fixed wavelength) which is observed in most amorphous semiconductors above room temperature (Mott and Davis 1979). This dependence can be described by Urbach's rule,

$$\alpha = \alpha_0 \exp\left[-\left(\frac{E_g - h\nu}{kT}\right)\right]. \tag{1}$$

The central problem (discussed by Hajtó and Jánossy (1983)) was to determine temperature rise in the illuminated spot due to the dissipation of the laser beam. This was calculated by assuming that the heat loss is due to the difference in temperature between the illuminated spot and that of the film (the 'spot temperature' T and the 'film temperature' T_F). Thus the spot temperature was determined from the equation

$$\tau_0 \frac{\partial T}{\partial t} = \eta J D(T) - (T - T_F). \tag{2}$$

where η and τ_0 are constants determined by the thermal constants of the material and geometry, J is the incident laser flux, D is the dissipation coefficient (= dissipation/incident flux) which depends on α and thus on T. D is simply related to the transmission and reflection coefficients (Q and R respectively) as

$$D = 1 - Q - R. \tag{3}$$

For self-supporting samples,

$$\left.\begin{aligned} Q &= 1/[n_1^2 \Gamma + (n_2^2/\Gamma) - 2n_1 n_2 \cos 2\delta], \\ R &= n_1 n_2 [\Gamma + (1/\Gamma) - 2 \cos 2\delta] Q, \end{aligned}\right\} \tag{4}$$

where $n_1 = (n+1)^2/4n$, $n_2 = (n-1)^2/4n$ and

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

where L is the sample thickness and n the refractive index.

The stationary value of the spot temperature can be determined from the equation

$$\eta J D(T) = T - T_F, \tag{5}$$

together with the stability condition

$$\frac{\partial}{\partial T}\left(D(T) - \frac{(T - T_F)}{\eta J} \right) < 0. \tag{6}$$

Discussion of the solutions of the above equations can be found in Hajtó and Jánossy (1983). It is shown there that, at certain critical intensities, 'thermal run-away' occurs. This is the origin of the discontinuities observed when the intensity is scanned.

To describe what happens at fixed intensity, the *photoinduced* processes must also be considered. In order to make the description quantitative, we use the theory of photostructural changes presented by Kolobov, Kolomiets, Konstantinov and Lyubin (1981). In § 2.2 we briefly summarize this theory.

2.2. *Photostructural changes*

A detailed description of photostructural changes in amorphous chalcogenide semiconductors has been given by Tanaka (1980). According to Tanaka (1980) these structural changes are due to photoinduced displacements of chalcogen atoms. A fraction of these atoms can sit in two different equilibrium positions. On a configurational coordinate diagram these positions correspond to the minima of the double-well potential. One of the minima represents the ground state and the other can be considered as a metastable excited state. Transitions from one configuration to the other occur through photoinduced and—near the glass transition temperature—pure thermal processes. According to Kolobov *et al.* (1981), the photoinduced transitions consist of two steps: a thermally activated small displacement of the atom and an electronic excitation by the incoming photons, which is followed by a relaxation to the new equilibrium position.

Kolobov *et al.* (1981) found the population kinetics are governed by the equation

$$\frac{dN}{dt} = -\left[\frac{1}{\tau} + J(\sigma_1 + \sigma_2)\right] N + J\sigma_1, \tag{7}$$

where N is the number of units in the metastable state divided by the total number of sites, and σ_1 and σ_2 are the cross-sections of the transitions from the ground state to the metastable state and for the inverse transitions, respectively. The temperature dependence of the cross-sections can be expressed as

$$\sigma_i = K_i \exp(-E_i/kT), \quad i = 1, 2, \tag{8}$$

where $K_i \sim 1/T$. The activation energies E_i are of the order of tenths of electron-volts.

In eqn. (7), τ is the relaxation time to the ground state via pure thermal transitions. This relaxation is usually very slow, except near the glass-transition temperature T_g, and it can often be neglected.

The stationary solution of eqn. (7) is

$$\bar{N} = \frac{\sigma_1}{1/J\tau + (\sigma_1 + \sigma_2)} = \frac{1}{1/q + 1 + f \exp(-\Delta E/kT)}$$

with $f = K_2/K_1$, $\Delta E = E_2 - E_1$ and $q = J\tau K_1 \exp(-E_1/kT)$. Equation (7) can be rewritten as

$$\frac{dN}{dt} = \frac{\bar{N} - N}{\tau_s}, \quad \text{with} \quad \frac{1}{\tau_s} = J(\sigma_1 + \sigma_2) + \frac{1}{\tau}. \tag{9}$$

The next problem to be considered is the connection between the value of N and the optical parameters of the substance. For the sake of simplicity we assume that the photoinduced processes influence only the value of α_0, otherwise the Urbach rule (eqn. (1)) remains valid. Furthermore we take α_0 proportional to N. With this assumption we derive from eqn. (9) an equation describing the kinetics of photostructural changes:

$$\frac{d\alpha_0}{dt} = \frac{\bar{\alpha}_0 - \alpha_0}{\tau_s}, \tag{10}$$

where $\bar{\alpha}_0$ is the stationary value of α_0:

$$\bar{\alpha}_0(T, J) = \frac{A}{1/q + 1 + f \exp(-\Delta E/kT)}. \tag{11}$$

To sum up, we have the following picture of photostructural change in amorphous semiconductors. The atomic configuration in amorphous materials is not so strictly determined as in crystalline solids, and a large number of metastable configurations can exist in them. In amorphous chalcogenide materials interaction with light quanta can cause certain atoms to move from one equilibrium position to another, thus altering the structure.

In our description, the instantaneous structure of the amorphous network is characterized from the optical point of view by the value of α_0 ; α_0 represents the fraction of atoms in the metastable state. Under the influence of laser irradiation the network approaches an equilibrium structure in which the number of transitions from the ground state to the metastable state equals the number of inverse transitions. This is characterized by the equilibrium value of α_0, $\overline{\alpha_0}(T, J)$.

We note that the simplifications made in deriving the exact form of $\overline{\alpha_0}(T, J)$ (eqn. (11)) are not essential in describing the oscillatory phenomenon. The important point is that there exists an equilibrium value of α_0 $(\overline{\alpha_0})$ which is determined by the temperature, and in some cases by the light intensity also. However, $\overline{\alpha_0}$ does not depend on the previous history of the illuminated spot, but the value of α_0 itself does depend on the previous history, and at a given moment it can be larger or smaller than the equilibrium value at a given temperature and intensity. In the first case, $(\alpha_0 > \overline{\alpha_0})$, α_0 decreases (photobleaching), in the second $(\alpha_0 < \overline{\alpha_0})$ α_0 increases (photodarkening). Thus whether darkening or bleaching occurs under the influence of the laser beam depends on the history of the given spot. This fact was proved experimentally by Averyanov, Kolobov, Kolomiets and Lyubin (1981).

Finally we estimate the energy change of the system associated with photostructural changes. Tanaka (1980) deduced from volume change measurements that during photostructural processes $N = 10^{19}$–10^{20} cm^{-3} atoms are moved. Furthermore, according to experiments, in a-GeSe$_2$ a typical relaxation time for photostructural changes at room temperature and an intensity level of ≈ 50 W cm^{-2} is a few seconds. Suppose that the energy released or absorbed in one atomic event is not more than 1 eV. The energy change per second per cm^3 due to photostructural changes under these conditions does not exceed 1 W cm^{-3}. However, the energy production due to normal absorption $J\alpha$, under the same conditions, is $\approx 5 \times 10^3$ W cm^{-3}, where we have taken $\alpha = 10^2$ cm^{-1}. Consequently, when determining the laser-induced heating, the energy change associated with photostructural changes can be neglected.

2.3. *Determination of the stationary state and its stability*

According to the considerations presented in §§ 2.1 and 2.2, the spot temperature T and α_0 can be considered as independent variables which satisfy the equations

$$\tau_0 \frac{\partial T}{\partial t} = \eta J D(\alpha_0, T) - (T - T_F), \tag{12}$$

$$\frac{\partial \alpha_0}{\partial t} = \frac{\overline{\alpha_0} - \alpha_0}{\tau_s}. \tag{13}$$

The stationary solutions (i.e. at $\partial T/\partial t = \partial \alpha_0/\partial t = 0$) of eqns. (12) and (13) are

$$\eta J D(\alpha_0, T) = T - T_F, \tag{14}$$

$$\alpha_0 = \overline{\alpha_0}(T, J). \tag{15}$$

For the present analysis it is very important to note that under the present experimental conditions the thermal relaxation time τ_0 is much smaller than the structural relaxation time τ_s (this was inferred from experimental observations). As already mentioned, we found τ_s to be a few seconds at $J \sim 50$ W cm^{-2}. However, measurements with a chopped laser beam showed another relaxation process with a time constant of a few milliseconds. A rough estimation of the parameters involved in eqn. (12) showed that this latter process can be identified with the establishment of a pseudo-thermal equilibrium, associated with the actual value of α_0.

As a consequence of the large difference between τ_0 and τ_s, we can assume that after switching on the laser, a pseudo-thermal equilibrium is first quickly established which corresponds to the actual value of α_0. As α_0 is changed by the laser beam, the spot temperature follows 'adiabatically' the pseudo-equilibrium value corresponding to the actual structure. The main point, however, is that this pseudo-equilibrium is not necessarily stable against thermal fluctuations (with fixed α_0).

To derive the condition of thermal stability let us denote by $T^{(s)}$ the temperature of the pseudo-equilibrium with a given α_0 (i.e. $T^{(s)}$ is a solution of eqn. (14), while eqn. (15) is not necessarily satisfied). If by a thermal fluctuation T is changed to $T^{(s)} + \delta T$, from eqn. (12) we obtain

$$\tau_0 \frac{\partial \delta T}{\partial t} = \frac{\partial}{\partial T}[\eta J D - (T^{(s)} - T_F)]\delta T.$$

As τ_0 is positive, δT relaxes to zero if

$$\frac{\partial}{\partial T}[\eta J D - (T^{(s)} - T_F)] < 0. \tag{16}$$

This condition can also be expressed in a more convenient form. Let us examine how $T^{(s)}$ changes if α_0 changes to $\alpha_0 + \delta\alpha_0$. From eqn. (14),

$$\delta[\eta J D(\alpha_0, T^{(s)}) - (T^{(s)} - T_F)] = \eta J \frac{\partial D}{\partial \alpha_0}\delta\alpha_0 + \frac{\partial}{\partial T} \times [\eta J D - (T^{(s)} - T_F)]\delta T^{(s)} = 0.$$

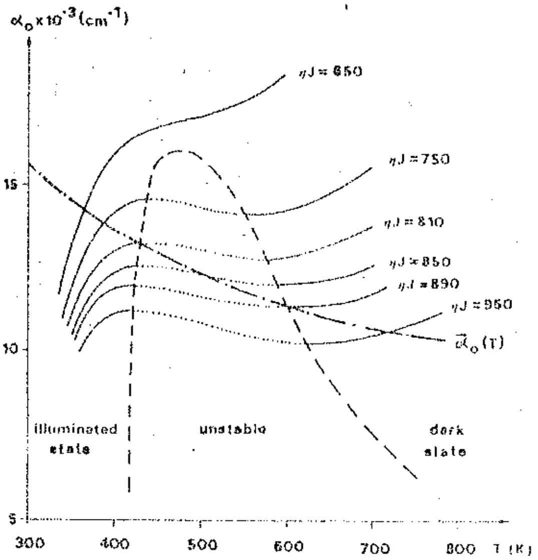
As can be seen from eqns. (3) and (4), $\partial D/\partial\alpha_0 > 0$ (i.e. for a higher absorption coefficient, there is a higher dissipation if all other parameters are fixed). Consequently the stability condition (16) can be written in the form

$$\frac{\delta\alpha_0}{\sigma T^{(s)}} > 0. \tag{17}$$

In fig. 1 α_0 is plotted against $T^{(s)}$ for different laser intensities (different values of ηJ). The parameters used were $L = 6$ μm, $E_g - h\nu = 0{\cdot}138$ eV, $T_F = 300$ K, $n = 2{\cdot}50$, $\cos 2\delta = 0$ (the temperature dependence of n is neglected). These parameters correspond to the data of a-GeSe$_2$ self-supporting films (Hajtó and Jánossy 1983). The dotted lines, where $\delta\alpha_0/T^{(s)} < 0$, correspond to unstable solutions. It can be seen from fig. 1 that there is a region on the α_0, T diagram where stable solutions do not exist. The unstable region separates solutions which correspond to the 'illuminated' and 'dark' states, respectively.

The stationary solution can be found by plotting $\bar{\alpha}_0(T)$ on the same diagram as the $\alpha_0(T)$ curves. As a given intensity the intercept of $\alpha_0(T)$ and $\bar{\alpha}_0(T)$ determines the stationary solution. If $\bar{\alpha}_0(T)$ crosses the unstable region there

Fig. 1



Graphical determination of the stationary state. The continuous and dotted lines represent the stable and unstable parts of the $\alpha_0(T)$ curves. The dashed line marks the boundary between the stable and unstable regions. The stationary state corresponds to the intercept of $\alpha_0(T)$ and $\overline{\alpha_0}(T)$. Oscillation occurs when the intercept is in the unstable region.

is an intensity interval where the intercept corresponds to an unstable solution ; in this case oscillation occurs.

To illustrate this we chose the parameters involved in Kolobov's theory in an arbitrary manner ($\overline{\alpha_0}(0)=2\times10^4$ cm^{-1}, $E_1=0{\cdot}15$ eV, $E_2=0{\cdot}2$ eV, $f=2$), and pure thermal relaxation is neglected ($1/\tau=0$). As shown in fig. 1, with these parameters oscillation occurs in the intensity range $810<\eta J<890$.

In § 2.4 we analyse in detail the behaviour of the film in different intensity ranges, assuming that the film was previously annealed.

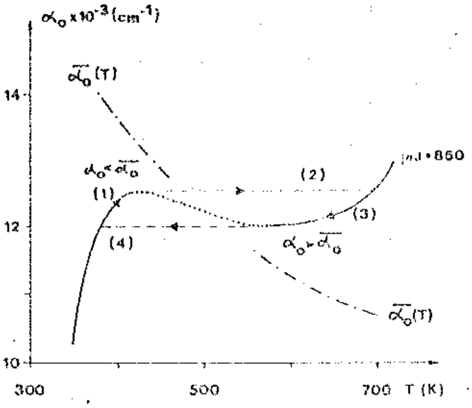
2.4. *Mechanism of oscillation*

According to our considerations, three different intensity ranges can be distinguished.

(1) The intensity is smaller than the lower threshold for oscillation. In this case only photodarkening takes place and a stationary state is established in the illuminated side.

(2) When no stable stationary solution exists, we use the time-dependent equations (12) and (13) to calculate the time variation of α_0 and T. Because the pseudo-thermal equilibrium is established much faster than the structural equilibrium, we can regard the α_0, T point (characterizing the state of the spot) as moving along the α_0 ($T^{(s)}$) curve corresponding to fixed intensity. It always moves in the direction in which the absolute value of $\overline{\alpha_0}-\alpha_0$ decreases. The different stages of the oscillation are illustrated in fig. 2. Let us start with a

Fig. 2



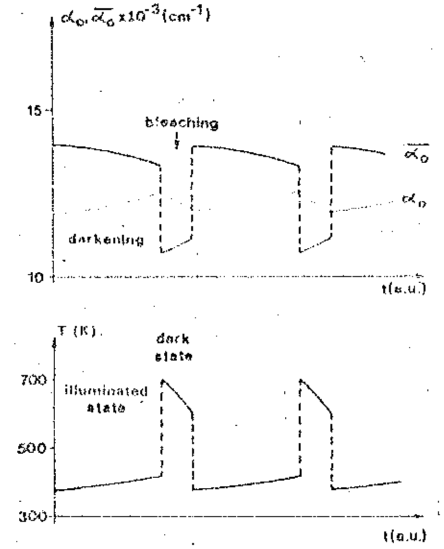
Different stages of oscillation : (1) photodarkening, (2) switch to the dark state, (3) photobleaching, (4) switch to the illuminated state.

sample for which at the beginning α_0 is small. The α_0, T' point is on the illuminated branch of the curve. As α_0 is smaller than $\overline{\alpha_0}$, photodarkening occurs and the point moves towards the unstable region. When it reaches the boundary of the unstable region a discontinuous switch takes place. According to our analysis this switch is fast, and there is no time for photostructural changes to take place, i.e. α_0 remains the same. Graphically this means that the spot 'moves' horizontally until it reaches a stable pseudo-equilibrium temperature on the dark side. Along the dark branch of the curve α_0 is larger than $\overline{\alpha_0}$, so photobleaching occurs and the α_0, T' point again moves towards the unstable region. At the boundary the next switch takes place to the illuminated state, again fast enough to avoid any significant change in α_0. Thus α_0 again becomes smaller than α_0 and the whole cycle recommences.

The variations in α_0, $\overline{\alpha_0}$ and T' during the cycle are presented in fig. 3 for $\eta J = 850$. The corresponding oscillation in the transmission and reflection coefficients are shown in fig. 4, where for comparison experimentally recorded curves are also shown. Considering the simplifications we have made in our model, quantitative agreement of the experimental curves with the calculated ones cannot be expected, but the similarity in the general features of the curves is obvious.

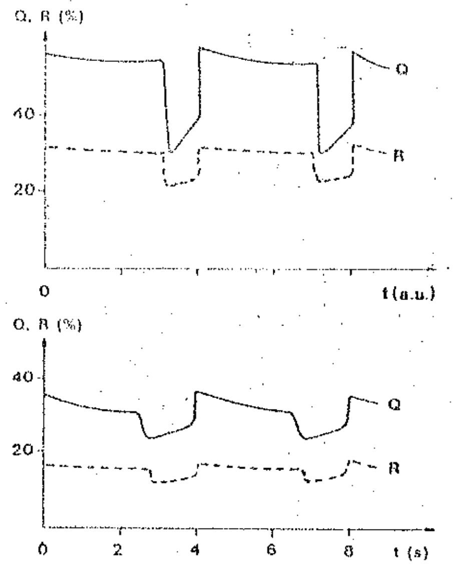
(3) If the intensity is higher than the upper limit for oscillation, photodarkening occurs first, followed by a switch to the dark state. In the dark state photobleaching occurs but is not sufficient to switch the spot back to the illuminated state ; a stationary state is established. We note that if the laser beam is switched off and after some time is turned on again, the thermal equilibrium corresponding to the illuminated state is first established (because before switching on the laser, $T' = T_F$). The stationary state is reached again through photodarkening, discontinuity and photobleaching. This effect has been observed in a-GeSe$_2$ films (Hajtó and Jánossy 1983, § 3.3, fig. 6).

Fig. 3



Variations of α_0, $\overline{\alpha_0}$ and T during oscillation for $\eta J = 850$.

Fig. 4



(*a*) Calculated transmission and reflection oscillations for $\eta J = 850$. (*b*) Experimentally registered curves for 6 μm thick self-supporting $GeSe_2$ films. Laser power 13·5 mW, spot diameter 207 μm.

§ 3. Discussion

In § 2 we showed that a combination of thermal and photostructural effects leads to highly non-linear behaviour and to oscillatory phenomenon in chalcogenide amorphous semiconductor films. Our theory explains the main features of observations made of self-supporting a-GeSe$_2$ films (Hajtó *et al.* 1982), namely that oscillations occur in a narrow intensity range; the oscillations consist of four different steps; at lower intensities 'memory effect' photodarkening is observed, at higher intensities the stationary state is reached through photodarkening and discontinuity, and this process is repeated every time the light is switched off. We believe that more detailed agreement could be obtained by taking into account the changes in the refractive index. A further improvement may also be obtained if the thermal relaxation of α_0 could be taken into account, which may have some significance in the dark (high-temperature) phase of the oscillation.

In principal the same theory should hold for GeSe$_2$ samples adhering to a substrate, the main difference being that higher intensities are needed, and as a consequence the oscillations are faster. Indeed, the recorded transmission and reflection oscillations show similar features of self-supporting samples (Hajtó 1980, Hajtó and Apai 1980). A remarkable difference, however, is that the curves obtained with GeSe$_2$ films on substrates are very noisy, especially in the dark state.

In some cases the noise becomes so strong that it is hard to distinguish it from the oscillations. This makes it difficult to carry out systematic measurements with such samples.

As an explanation of this fact we suggest, following Phillips (1982), that the noise is due to laser-induced reversible microcrystallization, and it 'simply reflects the noise of $N \geqslant 10^3$ microcrystallites randomly switching from the ordered crystalline to disordered glassy configurations' (Phillips 1982). The same effect is much weaker in self-supporting films because of the much smaller light intensity.

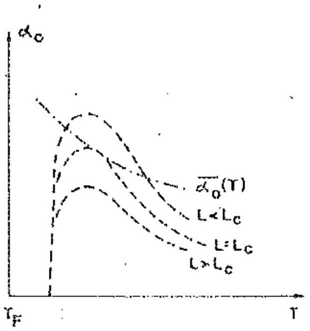
Finally, we note that according to our model oscillation should occur in any amorphous semiconductor film provided that

(1) the sample thickness, wavelength of the light and film temperature are properly chosen, and
(2) $\overline{\alpha_0}(T)$ is a decreasing function of T.

To show this, consider any material for which Urbach's rule is valid. As a first step the boundary curve between the stable and unstable regions has to be determined. This curve depends only on the thickness L, the film temperature, T_F and $E_g - h\nu$. Thus if L, T_F and (by choosing the wavelength properly) $E_g - h\nu$ are chosen to have the same value as for the curves presented in fig. 1, the same boundary curve is obtained.

The condition for oscillation is that $\overline{\alpha_0}(T)$ should pass through the unstable region. This condition is, of course, not automatically satisfied for a given thickness. However, as $\alpha_0(T)$ at fixed T is inversely proportional to L (D depends only on $\alpha_0 L$, see eqns. (1), (3) and (4)), $\alpha_0(T)$ should necessarily intersect the boundary curve if the thickness is small enough (see fig. 5). Thus if the film is thinner than a critical value of L, oscillation should occur. The above consideration may serve as a guide to finding oscillatory phenomenon in other chalcogenide materials.

Fig. 5



Boundary curves between stable and unstable regions for different thicknesses. If $L<L_c$ the $\overline{\alpha_0}(T)$ curve passes through the unstable region and oscillation occurs.

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