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A photoconductive film evaluated as an optical resonator

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ABSTRACT

Oscillations in the spectral photoresponse of a semiconducting thin film with the period of geometrical cavity resonance are made visible by a wide overlap between the photosensitive interband range and the transparency range in CdS. This becomes possible due to enhanced absorption edge broadening in a polycrystalline film. A quantitative fit of the modulation depth of the oscillations in both transmittance and photoresponse is given in terms of resonator optics and semiconductor parameters.

INTRODUCTION

Intrinsic photoconductors (especially direct-gap materials) usually exhibit a sharp spectral cut-off due to the steep interband absorption edge. Thus observation of cavity effects due to coherent multiple reflections at the parallel air/photoconductor and photoconductor/substrate interfaces are restricted to a narrow region near to and below the bandgap, and can be neglected in the interband region $h\nu > E_g$ where the absorption coefficients α are too high to allow a high finesse of the cavity. In semiconductor injection lasers interband absorption is suppressed by population inversion and the cavity effect becomes visible through the axial modes (Saleh & Teich 1991). This cavity effect can also be used to design *resonant cavity enhanced photonic devices* with increased response at selected wavelengths and suppressed response at unwanted wavelengths (Uenlue 1995).

In polycrystalline and amorphous photoconductors the interband absorption edge is more gradual than in bulk material due to disorder. Thus the overlap between the photosensitive and the low-absorption ranges is extended and more eigenfrequencies of the resonator become observable. In this paper a study was made of optical transmittance and steady-state spectral photoresponse (PC) of CdS sputtered thin films on glass substrates.

EXPERIMENTAL

CdS polycrystalline films have been deposited by radio frequency sputtering on glass substrates. X-ray diffraction studies revealed the hexagonal structure of the films. The enhancement of the 002 peak indicated perpendicular orientation in agreement

with previous reports (Hill 1978). The crystallite size was estimated for the approximate formula

$$d = \frac{\lambda}{D \cos \theta}, \tag{1}$$

where λ is the X-ray wavelength, D is the angular FWHM line width and θ is the Bragg angle. The calculated crystallite size for the sample used for the experiment in Fig. 1 is $d = 35.6\,\text{nm}$. The thickness of the samples was typically 0.5 to 1.5 μm. The samples were contacted by evaporated dots of indium which is known to deliver Ohmic contacts. Optical transmittance was measured by a VARIAN CARY 5E spectrophotometer. Photoconductivity (PC) was excited by the chopped light from a tungsten halogen lamp spectrally resolved by a BENTHAM grating monochromator and detected by lock-in technique. A spectrally flat silicon thermopile was used as a reference detector (Neff 1996). The samples were mounted in a CRYOMECH closed-cycle optical cryostat reaching temperatures between 15 and 300°K.

EXPERIMENTAL RESULTS

Typical PC spectra are plotted in Fig. 1. The ordinate, the photocurrent per quant at a given biasing dc current, is proportional to the total nonequilibrium carrier concentration per unit quantum flux $\Delta n/Q_0 = \int_0^d \delta n(z)dz/Q_0$, where z is the coordinate of light propagation and d is sample thickness.

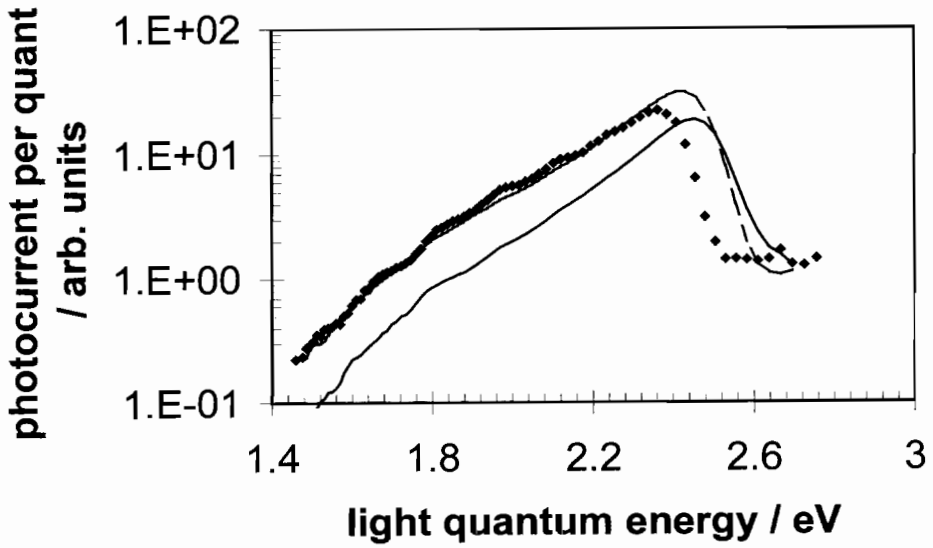

Fig. 1. Spectral photocurrent per quant for a 1.4 μm thick CdS sample on a glass substrate at $T = 300$°K (◆). In the spectra measured at $T = 170$°K (broken line) and $T = 75$°K (solid line), the data points are omitted for clearity.

The spectrum exhibits the following features:

Below the energy gap ($E_g(300°K) = 2.42\,eV$ for crystalline CdS), a very soft cut-on is observed which is extended over approximately 1 eV. From transmittance in that energy range the conclusion $\alpha d \ll 1$ can be drawn. Consequently the PC spectrum is proportional to the absorption spectrum $\Delta n(\hbar\omega) \propto \alpha(\hbar\omega)$, when $\Delta n/Q_0$ is the excitation spectrum of electron-hole pairs, and this feature has to be attributed to an exponential absorption edge or to absorption edge tailing. In disordered polycrystalline or amorphous materials absorption edges of that type have been attributed to density of states tails ("Urbach tail") and/or to decreasing mobility (mobility gap). In Fig. 1 for $1.8 \leq h\nu \leq 2.4\,eV$, the characteristic energy for decay to 1/e is $E_o = 240\,meV$ at 300°K. E_o is nearly independent of temperature with the tendency for a slight decrease with decreasing temperature.

The abrupt cut-off photocurrent near to the energy gap by more than one order of magnitude reflects the growing influence of surface recombination at the transition from bulk to surface excitation (Boer 1990). The surface recombination parameter $S = s\tau/L$ was evaluated and found to be 1.2. The temperature shift of this abrupt decrease reflects the decrease of the energy gap with temperature ($\partial E_g/\partial T = -4.1 \times 10^{-4}\,eV/K$).

As an additional new result, weak oscillations with a period of approximately 0.18 eV were observed at all temperatures. From the observation at all temperatures and from the large period, the typical hot-photoelectron mechanism of oscillatory PC due to emission of optical photons must be ruled out. Instead, from the comparison with the transmittance spectrum of the same sample in Fig. 2 (full line), it is evident that the oscillations are due to coherent multiple reflections as they

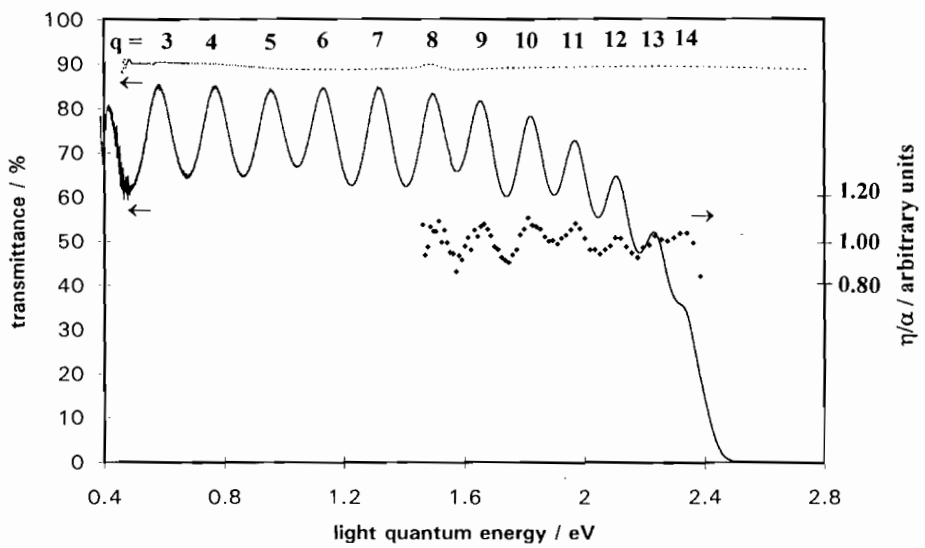

Fig. 2. Spectral transmittance of a polycrystalline CdS thin-film sample at $T = 300°K$ (solid line), q is the interference order. Dashed line, substrate transmittance. ◆, spectral photocurrent per unit absorption coefficient as processed by the procedure described in the text (right ordinate).

are observed in transmittance or reflectance for any thin-film sample with a highly reflecting surface. Because transmittance was measured at room temperature, the room temperature transmittance and photoconductive spectra were then compared.

At first glance the PC oscillations in Fig. 1 seem to be a small effect as compared with the oscillations in transmittance, but this is due to the logarithmic plot. For the discussion we should consider the photoeffect per absorbed quant rather than the photoeffect per incident quant. For that purpose, we first averaged the oscillations in the exponential part of the photocurrent spectrum by fitting the logarithm to a polynomial and assuming this average to be proportional to the absorption coefficient $\alpha(h\nu)$. Then in a new plot of the photocurrent divided by $\alpha(h\nu)$ (symbols ◆ in Fig. 2) the oscillations are more emphasized than in the original Fig. 1, and the modulation depth in this plot now compares with the modulation depth in the transmittance spectrum.

The coincidence between the oscillation periods in both types of measurement proves the cavity to be the origin of the oscillatory PC observed. Moreover, the oscillations in both effects are in phase because a high transmittance at a given wavelength is correlated with a high energy density in the cavity, as will be discussed.

Although effects of multiple reflections have been observed occasionally in photoconductors like CdS, to our knowledge, this is the first quantitative proof of such a coincidence extended over a large number of cavity resonances. The interface orders $q = 8$ to 14 are indicated in Fig. 2. In general, the disorder connected with exponential band-edge tailing will reduce the photosensitivity of any material. However, in CdS photoconductivity remains observable due to long majority carrier lifetime.

PARAMETERS EXTRACTED FROM THE CAVITY EFFECT

Refractive index

Processing the geometrical resonance in transmittance or reflectance spectra is used as a standard method for the determination of the refractive index $\bar{n}$ and of its dispersion $\bar{n}(\lambda)$. The presented results open a new approach to this quantity from photoelectric measurements. In both the transmittance and the photocurrent spectra, a decrease of the oscillation period with increasing interference order was observed, which reflects the dispersion of refractive index due to beginning interband transitions.

Following this concept, the observed refractive indices were fitted to a Sellmeir formula with a single oscillator representing the total interband absorption. The parameters have been fitted to the dispersion model proposed by Wemple and DiDomenico (1971). The good agreement with this model justifies our earlier approach when using the refractive index of bulk CdS for calculating the sample thickness.

Absorption edge

The averaged PC spectrum can be compared with absorption edges reported for CdS. In general this approach is possible as long as $\alpha d \ll 1$. We might call this

photoelectric determination of the absorption edge. Previously, this approach has been applied to broadening mechanisms of the absorption edge in mercury cadmium telluride (Herrmann 1992).

Photocurrent spectroscopy is used at large for studying quantum well structures. For comparison with directly measured absorption edges we need a reference point. For the plot in Fig. 3 we adjusted the photoelectric spectrum to $\alpha = 2000\,\mathrm{cm}^{-1}$ at $h\nu = 2\,\mathrm{eV}$. This was based on a very recent analysis of exponentially broadened absorption edges (Bogoboyashchiy 1997). The observation from Fig. 3 is a relatively high broadening energy of our samples, even in comparison with data reported for polycrystalline samples (Ward 1991). This observation has potential for optimizing optoelectronic devices. The single crystal data are given only for reference.

Quantitative analysis of the modulation depth in the spectra

The ratio of maximum and minimum transmittance in the spectrum $T(\hbar\omega)$ can be described by:

$$\frac{T_{\max}}{T_{\min}} = 1 + \left(\frac{2\mathcal{F}}{\pi}\right)^2 = \left(\frac{1+r}{1-r}\right)^2, \tag{2}$$

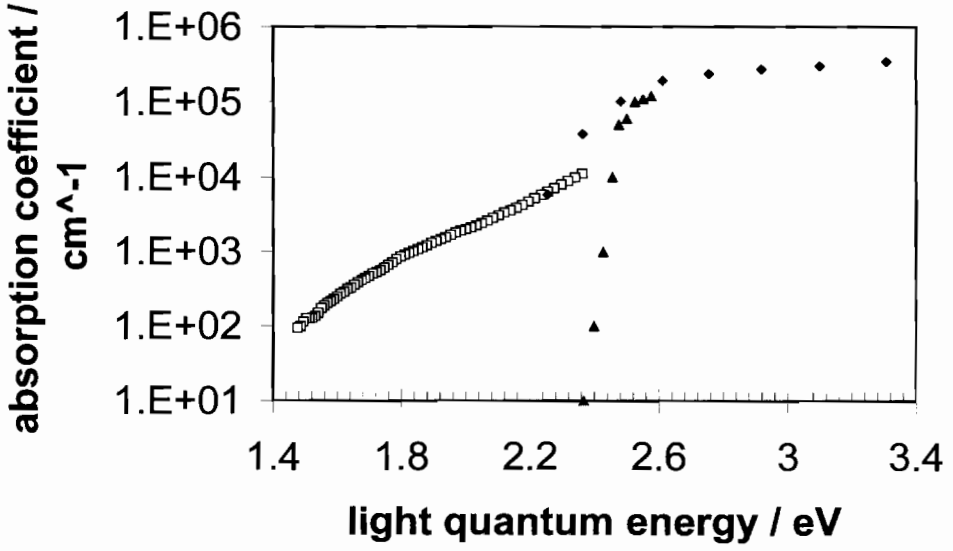

Fig. 3. Exponential interband absorption edge for single crystal and polycrystalline CdS as detected by transmittance and photoconductivity. ▲, absorption of a single crystal (Spiegelberg 1976), ◆, absorption of a polycrystalline film (Ward 1991), □, spectral photoconductivity (this paper) adjusted to $2000\,\mathrm{cm}^{-1}$ at $h\nu = 2\,\mathrm{eV}$, cavity effect not eliminated. All data at room temperature.

where the finesse $\mathcal{F}$ for a lossy resonator as given by Saleh and Teich (1991) is:

$$\mathcal{F} = \frac{\pi r^{1/2}}{1-r}, \quad \text{and} \qquad r^2 = R_1 R_2 e^{-2\alpha_s d} \tag{3}$$

is the intensity round-trip attenuation factor. The observed finesse of the CdS layer is relatively low, $\mathcal{F} = 0.93$ at $h\nu = 1.7\,\text{eV}$. Using the reflectivity at the air/CdS interface $R_1 = 0.188$ (obtained from the refractive index $\bar{n}_{\text{CdS}} = 2.53$ (Ward 1991)), the reflectivity at the CdS/substrate interface $R_2 = 0.0627$ (from $\bar{n}_{\text{substrate}} = 1.517$), and the sample thickness $d = 1.4\,\mu\text{m}$, from Eq. (3) we get the absorption coefficient of the film α_s used for the argument $\alpha d \ll 1$ above.

The decrease of the modulation depth when approaching the energy gap is a consequence of increasing absorption that reduces the finesse $\mathcal{F}$ of the resonator. The best evidence is obtained from the range near to the gap where the absorption coefficient reaches values of several $10^5\,\text{cm}^{-1}$ in CdS.

We can apply this same analysis to the PC spectrum, because the average energy density within the cavity, ρ, is oscillating with light frequency according to the same law on which Eq. (2) is based and in phase with the transmittance oscillations. This applies to a loss-free resonator; but in the limit of small resonator losses (weak absorption $\alpha d \ll 1$), it remains qualitatively valid.

The generation rate of non-equilibrium carriers can be written as follows:

$$g(z) = \left.\frac{\partial \delta n(z)}{\partial t}\right|_{\text{gen}} = \alpha \frac{I(z)}{\hbar\omega} = \alpha \frac{c\rho(z)}{\bar{n}\hbar\omega}, \tag{4}$$

where c is speed of light, I is intensity and $\rho(z)$ is energy density. For the photoresponse we need only $\Delta n = \int_0^d \delta n(z) dz$. The steady-state photocurrent is proportional to $\Delta n = g\tau$ with τ being the lifetime appearing in the linear recombination term $\partial \delta n/\partial t = -\delta n/\tau$ used for the case of linear response.

Thus, it is evident that the transmittance and the photocurrent spectra should be in phase as observed. The oscillations are not eliminated when normalizing the photocurrent to unit photon flux because we use the "spectrally flat" signal from a silicon thermopile as reference. This signal is being measured for the same photon energy range at the sample's position and of course it does not exhibit the cavity-related spectral oscillations.

In CdS, mainly the electrons contribute to photocurrent, thus we considered the continuity equation for non-equilibrium electrons. Since the light intensity dependence was found to be linear, we can approximate the recombination term as mentioned above and solve the continuity equation with the generation term according to Eq. (4) and with the surface recombination written into the boundary condition (Boer 1990). The results of this modeling can be summarized as follows. For weakly non-uniform excitation ($q = 8$, $\alpha d = 0.636$) the modulation depths of spectral transmittance $T(\hbar\omega)$ and photocurrent per absorption coefficient $\Delta n/\alpha$ are equal each other, in good agreement with the experimental results.

In reality, at resonance we have also a periodic spatial distribution of light intensity and consequently a periodic non-equilibrium carrier concentration within the sample due to the existence of a standing wave. This results in a phenomenon

known as light-induced grating. However, since this spatial nonuniformity cannot be detected in the photocurrent, its influence on the magnitude of the photocurrent can be neglected as long as we observe linear response, that means a linear dependence of the photocurrent on light intensity. In this common case of small non-equilibrium carrier concentration, the diffusion of non-equilibrium carriers from antinodes to nodes does not enhance recombination.

We also attempted to understand the additional experimental observation that the photocurrent oscillations die out faster than the transmittance oscillations when approaching the energy gap. For example, for $q = 8$ we found $T_{max}/T_{min} = 1.26 \approx (\Delta n/\alpha)_{max}/(\Delta n/\alpha)_{min} = 1.23$, and for $q = 12$, we found $T_{max}/T_{min} = 1.25$ but $(\Delta n/\alpha)_{max}/(\Delta n/\alpha)_{min} = 1.11$ (compare the two curves in Fig. 2). Most probably this is a case where diffusion does enhance recombination, but this time surface recombination is important. Thus, we attribute this stronger decrease of modulation depth in the PC spectrum to the increasing additional influence of surface recombination and explain it as follows. With increasing interference order, the generation rate $g(z)$ becomes more surface-like (more non-uniform) for two reasons: First the absorption coefficient increases (at the interference extremum with order $q = 12$ we have already $\alpha d = 1.26$) and then the wavelength of the standing wave inside the cavity decreases thus making the region with high intensity between the surface and the first node more narrow, and thus more sensitive to surface recombination. With surface recombination, the magnitude of the photocurrent is reduced due to carrier diffusion towards the front surface, which is reflected in the faster decrease of the PC modulation depth.

CONCLUSIONS

In conclusion the following results were obtained:

The cavity effect on intrinsic photoconductivity was analyzed in a CdS thin film on glass substrate and was used to evaluate features of the optical resonator as well as of the photoconductive film. A quantitative description of the modulation depth was obtained from a solution of the continuity equation for the non-equilibrium carrier concentration. The results have application potential for both solar-cell related materials research and for cavity enhanced optoelectronic devices.

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شريحة رقيقة ذات موصلية كهروضوئية تعمل كمهتز ذو رنين

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الخلاصة

تطابق مجال الحساسية الضوئية مع مجال النفاذيه في كبريتيد الكادميوم جعل من الممكـــن ظهور اهتزازات في طيف الاستجابة الضوئية في الشرائح الرقيقة شبه الموصلة. ويصبح هـذا ممكنا كنتيجة لوجود توسيع كبير في حافة الامتصاص في الشرائح متعددة البلورات. نعـــرض تطابق كمي لعمق الاهتزازات في كل من النفاذيه الاستجابة الضوئيـــة باســتخدام المعطيـــات الضوئية والشبه موصله للمهتز.