

Hole-diffusion length and transport parameters of thin CdS films from a Schottky barrier

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The optoelectronic and transport properties of thin sprayed CdS films, having a thickness of less than $2\text{ }\mu\text{m}$ are reported. The use of two independent technics, surface photovoltage and photoelectrochemical measurements, led to a good agreement in the hole-diffusion length values. These range from 0.017 to $0.15\text{ }\mu\text{m}$ and behaved differently in two zones. A rapid increase of this parameter is observed below a film thickness of $0.4\text{ }\mu\text{m}$. Above this thickness, the value obtained is constant. Specific space-charge widths are expected because of the respective measurement conditions. The hole-diffusion length decreases as carrier density increases. The hole lifetime shows a regular decrease as the thickness factor increases, while the hole-diffusion coefficient and mobility patterns are similar to that of the photocurrent.

I. INTRODUCTION

The minority-carrier diffusion length, like the carrier density, is one of the main factors controlling the photoreactivity of a semiconductor. This parameter has been measured for most of the compounds belonging to this material group and commonly used in solar cell devices. As a result, single crystals have generally been easily characterized. The minority-carrier diffusion length values in this case are relatively high and can be determined with good precision. However, very low magnitudes are obtained with polycrystalline specimens. This fact, obviously, portrays the more precise configuration of the diffusion zone in the first case, and explains why its contribution in the total current can be neglected when very thin samples are used. Even in such cases, a complete characterization of a semiconductor under illumination requires a certain knowledge of this parameter.

Many measurement technics, mainly based on the spectral properties of the material are proposed in the literature for solid devices.¹⁻⁴ With solid-liquid heterojunctions, the Gartner approach is used in different ways from the classical photoelectrochemical method (PEC),^{5,6} to the differential photocurrent technic (DPC),⁷ the latter being the most recent method used. This variety shows that this model is still the one most commonly used to study this type of devices. Even though the previous investigators applied it for epitaxial layers, n/p homo- and heterojunctions,^{8,9} the importance of the surface photovoltage measurement (SPV) for polycrystalline thin semiconductor films has been emphasized by Charles *et al.*,¹⁰ since surface recombination does not influence the measured lifetime values. According to these authors, it has been tested successfully by several workers, especially with silicon.¹¹ Moreover, Tarricone *et al.*¹² have indicated that the SPV is a technic suitable for application to thin semiconductor films as a component of Schottky barriers. Since semiconductor-electrolyte junctions are basically considered as a Schottky barrier, some workers also confirmed the applicability of this technic to such junctions.¹³

Recently, the peculiar electro-optical properties of the sprayed CdS films within the entire range of thickness down to $1\text{ }\mu\text{m}$ were revealed.^{14,15} The behavior of the photocurrent

vis-a-vis this parameter was shown to depend strongly on the microstructure of the films whose configuration is influenced by the nature of the deposition substrate. But the main result obtained was the relatively high values of the absorption coefficient given by the thinnest samples, which decrease rapidly with increase in film thickness. One can also point out a similar behavior pattern observed in the case of the carrier density parameter. The two parameters have been used to study the maximum photocurrent obtained with films of about $0.3\text{ }\mu\text{m}$ in thickness deposited onto SnO_2 and ITO coated soda limes. Keeping in line with the optoelectronic characterization of these films, an attempted comparison between the minority-carrier diffusion length values given by the SPV measurements, and those obtained from the usual PEC technic is presented here and serves to monitor the behavior of this parameter in the thickness region of less than $2\text{ }\mu\text{m}$. Since the two technics are applied to the same liquid Schottky barrier, it seemed necessary to sum up the situation about the theoretical relationship governing each of them. The implications of the behavior of the minority-carrier diffusion length on the film's electrical properties are then analyzed subsequently.

II. THEORY

The comprehensive treatise of the Schottky barrier is based on the band model applied to a semiconductor material. This approach led Gartner¹⁶ to propose the following general expression of the photocurrent J_{ph} :

$$J_{\text{ph}} = q\phi(1 - e^{-\alpha W}) + q\phi[\alpha L_p e^{-\alpha W}/(1 + \alpha L_p)] + \rho(D_p/L_p) \quad (1)$$

The first term of the right-hand side of this expression corresponds to the current contribution of the space-charge region J_{sc} , and the second one, J_{qn} , to that of the quasineutral region; both of these characterize an illuminated semiconductor. The different parameters used have their usual significance: namely, q , the elementary charge; ϕ , the incident photon flux; α , the absorption coefficient of the semiconductor; W , the space-charge region width; L_p , the minority-carrier diffusion length.

Of course, Eq. (1) is valid only on the condition that certain hypotheses described widely in the literature be accepted.^{5,6} That presupposes that the third term on the right-hand side is negligible, and this is in fact reflected in most of the experimental cases. Equation (1) completely describes the photovoltaic charge transfer at the junction and directly corresponds to the processes occurring in the photoelectrochemical cell. It is commonly reduced to

$$J_{ph} = q\phi(1 - e^{-\alpha W}/1 + \alpha L_p). \quad (1a)$$

In this form, it easily enables determination of the hole diffusion length by two ways. The first one derives directly from Eq. (1a) by means of the quantum yield defined as $\eta = J_{ph}/q\phi$. Hence, L_p is deduced to

$$L_p = 1/\eta[(e^{-\alpha W}/1 - \eta) - 1]. \quad (2)$$

The use of Eq. (2) for the L_p calculation requires knowledge of the precise values of η and α .

The second way of determining L_p is by putting Eq. (2) into a logarithmic form such as

$$\ln(1 - \eta) = -\alpha W_0 V_s^{1/2} - \ln(1 - \alpha L_p), \quad (3)$$

where V_s is the surface potential of the semiconductor, and W_0 the space-charge width when $V_s = 1$ V. If the condition $\alpha L_p \ll 1$ is fulfilled, Eq. (3) is then simplified

$$\ln(1 - \eta) = -\alpha W_0 V_s^{1/2} - \alpha L_p. \quad (3a)$$

In this form, the L_p value can be determined by extrapolating the linear part of $\ln(1 - \eta)$ vs $V_s^{1/2}$, plots to $\ln(1 - \eta) = 0$. This method requires knowledge of the values of W and α .

The charge transfer process occurring in the surface photovoltage measurements implies that only the semiconductor is involved, the direct role of the interface being suppressed theoretically by the light chopping performance.^{8,9,17} The space-charge region is supposed to act just as a capacitance in which occurs a new charge redistribution under illumination. Then, respecting the required condition of low-light intensity, the main role in the charge transfer process is attributed to the quasineutral region. The total current in this case is limited to

$$J_{ph} = q\phi[\alpha L_p/(1 + \alpha L_p)]e^{-\alpha W}, \quad (4)$$

provided that the following conditions: $d \gg 2L_p$, $\alpha d \gg 1$, $\alpha W \ll 1$, and $W \ll L_p$ are fulfilled, d being the film thickness parameter. As described widely in the literature, the surface photovoltage produced can be measured either in the short-circuit conditions, where $J_{ph} = J_{cc}$ or, in the open-circuit conditions with the V_{oc} parameter. The two methods are equivalent since one has $V_{oc} = K \ln(J_{cc}/J_s + 1)$, where J_s is the saturation current, and $K = kT/q$, the thermal potential. The incident photon flux must, as required be weak, thus leading to $J_{cc} \ll J_s$. Hence the last general relation becomes

$$V_{oc} = K \cdot J_{cc}/J_s. \quad (5)$$

Determination of the hole-diffusion length by the surface photovoltage measurement is carried out by computing the incident photon flux value in the wavelength region slightly above the absorption threshold. This parameter is expected to vary with the light penetration depth α^{-1} . From Eq. (4), it can be expressed

$$\phi = J_{cc}/qL_p e^{-\alpha W}(\alpha^{-1} + L_p). \quad (6)$$

According to the condition $\alpha W \ll 1$, the term $J_{cc}/qL_p e^{-\alpha W} \simeq Cte$. Hence

$$\phi = Cte(\alpha^{-1} + L_p). \quad (6a)$$

The L_p value is hereby obtained by extrapolating the linear part of ϕ vs α^{-1} plots to zero intensity as a negative intercept. It is worth recalling here that the last relation is completely valid only if the reflectance of the semiconductor is almost constant in the working wavelength zone, and the trapping effect through the interface quite negligible.

III. EXPERIMENTAL DETAILS

The cadmium sulfide films used in this work have been prepared under precise conditions reported elsewhere.^{14,15} Nevertheless, it should be indicated that only the SnO₂-coated glasses (Corning 7056) served as deposition substrates. For the two technics of L_p determination, the same junction configuration was used. This involved the dipping of the film-electrode into alkaline medium ($pH = 12.5$) with the redox couple S^{-2}/S_x^{-2} ($E_0 = -0.72$ V/SCE), which is known to be corrosion-proof. This electrolyte has been prepared with 0.5 M S; 1 M NaS and 1 M NaOH. This medium absorbs optically down to 420 nm. Beyond this threshold, there are not optical disturbances involved in the working wavelength zone.

As far as the PEC measurements are concerned, the usual potentiostatic setup was utilized. The cell had a Pt wire as a counter electrode and a saturated calomel electrode (SCE) as the reference. A full light illumination was supplied by a Xe lamp ORIEL X 150 W simulator. However, the study was carried out with a monochromatic wavelength of 510 nm supplied by an optical Schott (DAL) filter put through the white light beam. The front of the CdS electrode was irradiated over an area of 0.3 cm² through the electrolyte. The light intensity, fixed at 0.26 mW cm⁻² was measured with a Kipp and Zonen thermopile. As mentioned above, the I - V characteristics revealed certain particularities in photoresponse. Thus, only three typical thickness values have been chosen to illustrate the investigation, namely 0.06, 0.3, and 1.7 μ m, in the working thickness zone. The L_p determination carried out this way required the calculation of the space-charge-width values in a manner similar to that of another study,¹⁵ using the Gartner approach adapted to thin polycrystalline semiconductor films with flat-band potential value $V_{fb} = -1.5$ V/SCE.

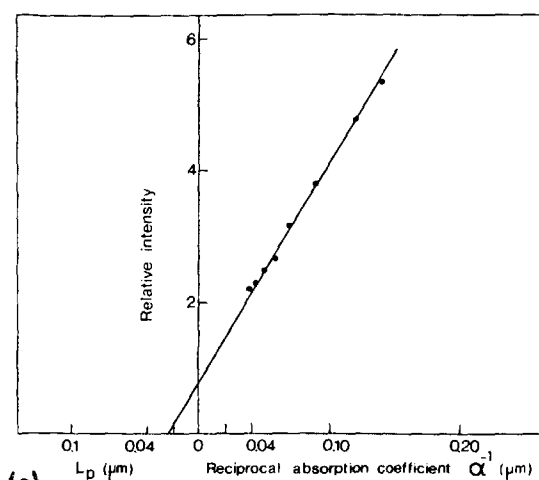
The liquid Schottky barrier contact as used here has been found to be more suitable and simpler in comparison with the Kelvin probe used by many workers^{18,19} in the SPV measurement, once the stability problem is solved. In this work, only the open-circuit conditions were applied. In this context, the last potentiostatic circuit was reduced to a simple electrical connection between the CdS electrode and the SCE reference via a Minisis 5000 Tacussel potentiometer for the control of the photovoltage fixed at $\Delta V = 15$ mV. This value was found to be convenient since the saturated photovoltage attained 600 mV. The adjustment of the incident photon flux to this photovoltage value needed to fit the intensity of the light beam. Here, the full light went through a

Jobin Yvon H25 monochromator which provides the desired wavelength. A regular wavelength step was chosen for scanning the whole working zone, ranging from 490 and 530 nm, that surrounds the absorption peak found at 516 nm. The incident light was modulated at a frequency of 20 Hz, in compliance with the theoretical requirement, with an Ortec Brookdeal 9479 chopper. However, no change in the potential value was noted in the measurement involving unmodulated light. This experimental effect has been explained by Moore¹⁹ who imputed it to the occurrence of a more direct contact at the junction. It may be also recalled that Dresner *et al.*²⁰ used unmodulated light with the same type of junction. In order to monitor the behavior of the incident photon flux according to Eq. (6a), the light penetration depths used in this part were calculated with the absorption coefficient values determined from the optical transmission data,¹⁴ knowing that the two parameters are correlated in the general expression $T = (1 - R)^2 e^{-\alpha d}$, where R is the reflection coefficient.

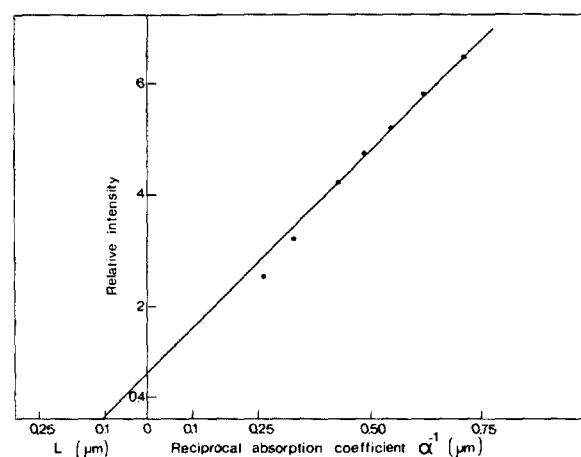
IV. RESULTS

The application of Eq. (6a) to the SPV measurements showed that the expected linearity depends partly on the CdS film thickness. It can be observed that the larger linear zone from around 0.20–2.5 μm corresponds with the thickest film as in Fig. 1(a). This zone is reduced by approximately 0.3–0.8 μm for the 0.3 μm thickness film in Fig. 1(b), and by 0.04–0.15 μm for the 0.06 μm thickness as in Fig. 1(c). In the case of the latter, this linear zone appeared to be sufficient to give an idea of the corresponding hole-diffusion length. A deviation must be expected from the linear law with the decrease in the thickness parameter, since the extension of the space-charge region and its electrical field could no longer be small compared to the minority-diffusion length, this being one of the main conditions for using the SPV technique. The intercept of the extrapolated linear part of the curves to zero photon flux emphasizes the dependence of the hole-diffusion length value on the film thickness as in Table I. However, the values obtained $0.026 < L_p < 0.15 \mu\text{m}$ remain in the usual range given in the literature for the polycrystalline CdS films, at least for the thickest specimens whose order of thickness has been more often studied.²¹

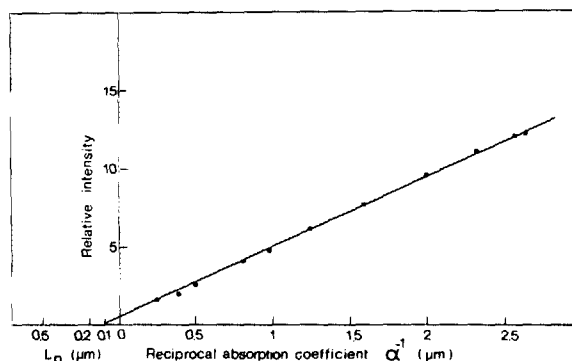
The calculation of the L_p values from Eq. (2) showed that the product αL_p in the three cases is inferior to unity. This fact evidently justifies the use of Eq. (3a) for the plotting of $\ln(1 - \eta)$ vs $V_s^{-1/2}$. In Fig. 2, a good linearity is observed from roughly -0.86 V/SCE towards anodic potentials, where the applicability of the Gartner approach was possible. Before this potential is attained, the influence of the hole-electron recombination and surface states is important. This explains the deviation from linearity. The vicinity of the linear zone of the curves relating to the film thicknesses of 0.06 and 1.7 μm is due to the close proximity of their respective slopes represented by the quantity αW_0 in Eq. (3a). This quantity has been found to be characteristic of thin sprayed CdS films and it varies with the thickness parameter.¹⁵ The maximum value of this term has been found to be close to unity for a film thickness of about 0.2 μm . As far as film thicknesses of 0.06 and 1.7 μm are concerned, almost the



(a)



(b)



(c)

FIG. 1. Determination of the hole-diffusion length by SPV measurement. Curve (a) related to $d = 0.06 \mu\text{m}$. Curve (b) related to $d = 0.30 \mu\text{m}$. Curve (c) related to $d = 1.70 \mu\text{m}$.

same value was obtained, to be precise, 0.65, and 0.60. Table I brings out the good agreement between the L_p values calculated from Eq. (2), and those deduced from the plotting of Eq. (3a). Obviously, a light deviation is unavoidable with the thinnest films, and this is certainly due to the precision limits of the technic used.

V. DISCUSSION

The concordance of the L_p values stemming from the two technics used in this work confirmed the behavior of this

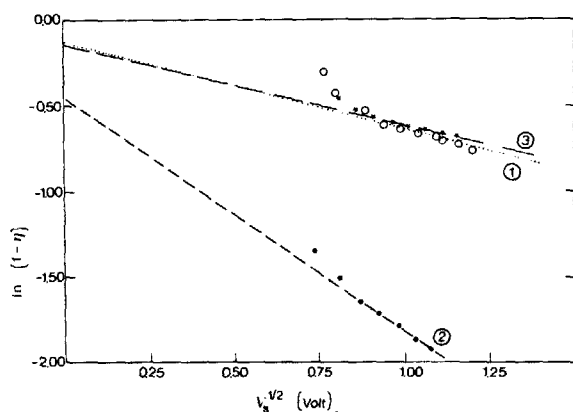


FIG. 2. Determination of the hole-diffusion length by PEC measurement. Curve (1) related to $d = 0.06 \mu\text{m}$. Curve (2) related to $d = 0.30 \mu\text{m}$. Curve (3) related to $d = 1.70 \mu\text{m}$.

parameter vis-a-vis film thickness. However, these results require some clarifications on the processes occurring in the semiconductor. An analysis of the relationship between the main parameters seems necessary.

A. Space-charge region width

Two situations can characterize the semiconductor electrode. At rest or static equilibrium in darkness, the space-charge region has an initial potential whose value is taken after the equalization of the Fermi levels at the junction. It is commonly named the built-in or diffusion potential V_{bi} . When the junction is irradiated and under a potential bias, this surface potential is expressed as $V_s = V_{bi} - V$, where V represents the new electrode potential value after dynamic equilibrium.²² In this case, one can expect the relation $V_s > V_{bi}$ to be fulfilled. As the space-charge region width is always proportional to the square root of the surface potential through the Schottky relation $W = W_0 V_s^{1/2}$, one can deduce $W > W'$ with $W' = W_0 V_{bi}^{1/2}$ knowing that $W_0 = (2\epsilon_s/qN_d)^{1/2}$ with ϵ_s the semiconductor permittivity, and N_d , the carrier density.

In the PEC measurements, the photocurrent values have been determined away from the static equilibrium found at -0.7 V/SCE , precisely at -0.5 V/SCE . Since one is dealing with low film thicknesses, the condition $W > L_p$ can be easily fulfilled. The photovoltage created in the SPV measurements usually amounts to some microvolts or millivolts in the higher cases. This does not modify sufficiently the equilibrium state in darkness. The space-charge width is then very close to W' and might be smaller than W . From the results obtained in plotting ϕ vs α^{-1} , the linearity

TABLE I. Minority diffusion-length values obtained from Gartner and SPV techniques: d —by calculation method; g —by graphic method.

d (μm)	L_p (Gartner) _{d} (μm)	L_p (Gartner) _{g} (μm)	L_p (SPV) (μm)
0.06	0.017	0.020	0.026
0.30	0.12	0.10	0.10
1.70	0.13	0.13	0.15

attested sufficiently that the required condition, $W' \ll L_p$ was fulfilled here. Now, since we know that the condition $W > W'$ might occur, a contradiction is no longer possible in the relationship between the space-charge width and the hole-diffusion length in this work, the first parameter magnitude appearing, as shown here, to depend on the respective technic used.

B. Relationship between L_p and N_d

An extension of the L_p determination to other film thicknesses gives a good idea of the behavior of this parameter. Two typical regions appear in Fig. 3. Below $0.4 \mu\text{m}$, a rapid increase of L_p contrasts with the decrease of N_d . No clear explanation of this fact can be given right now, even though the back interface impurities in the zone surrounding SnO_2 -CdS contact seem to be the possible cause, as well as for N_d and for α .^{14,15} Over this thickness value, the last interface is supposed to be out of the active region of the semiconductor under illumination. Beyond this zone, the homogeneity of the material led to a constant value of L_p and N_d , depicted by a plateau in this thickness zone for the two parameters.

L_p , N_d , and α are intimately related through the theory of Roosbroeck and Shockley.²³ A dependence has been noted previously between the first two parameters in the study of some semiconductors such as GaAs and GaP.^{24,25} To my knowledge, no result as has yet been published on this subject regarding CdS material. In Fig. 4, a downward trend of L_p values is noticeable, particularly down to a carrier concentration of $5 \times 10^{16} \text{ cm}^{-3}$. The strongly sloping part, at relatively small N_d , is in good agreement with the results reported for some other II-VI compounds.²⁶ This behavior was found to depend strongly on the nature and the concentration of the predominant dopant elements. Since these results were deduced from a controlled doping of the material, the shape of the curve obtained here strengthens the hypothesis of the gradual decrease of the concentration of photoionized donors with increase in sample thickness, as proposed elsewhere.¹⁵

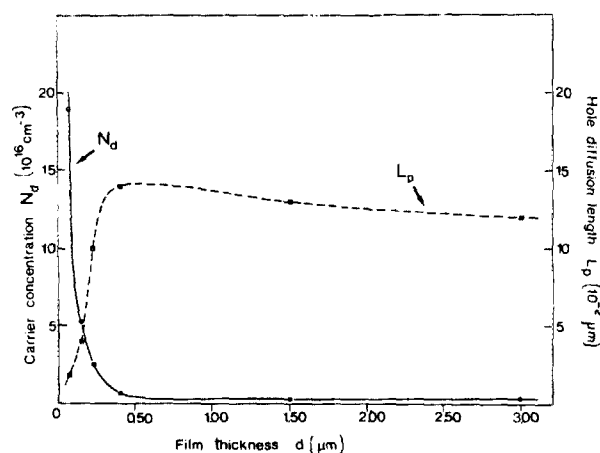


FIG. 3. Thickness dependence of L_p and N_d parameters of thin sprayed CdS films. Dotted line related to L_p evolution. Full line related to N_d evolution.

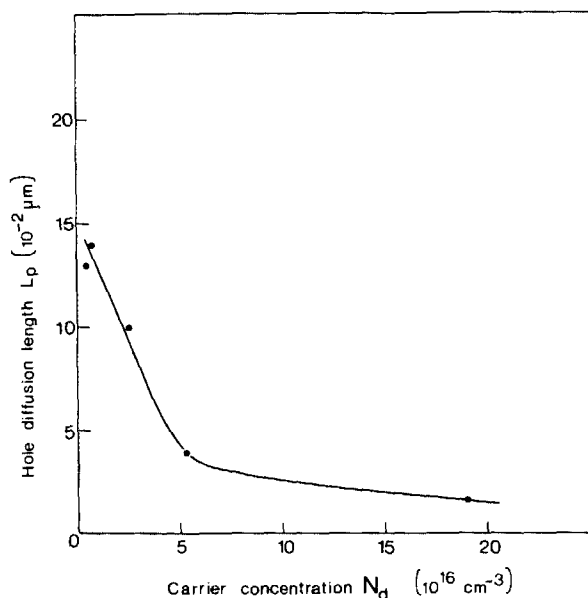


FIG. 4. Hole-diffusion length as a function of the carrier concentration of thin sprayed CdS films.

C. Hole lifetime and mobility

The evolution of L_p as it appears in Fig. 3 implies some changes in the transport properties of the CdS films. It is known that the hole-diffusion length is defined as $L_p = (D_p \tau_p)^{1/2}$, where τ_p is the hole-diffusion lifetime and D_p the hole-diffusion coefficient, which is correlated with the hole mobility μ_p through the Einstein relationship as $D_p = K \mu_p$, assuming that the temperature factor is constant. With the hypothesis of no recombination in the space-charge region, Card²⁷ established analytical expressions and graphic patterns concerning, at the same time, the hole-collection velocity S at the inner space-charge boundary, the carrier concentration N_d , and the hole lifetime τ_p . The application of these general results requires that the condition $V_{bi} > K$ be fulfilled. As a result the variation of N_d leads to that of S and τ_p as in Table II. One can notice here that the thinner the film, the higher the S value. This implies, in this case, that all the holes produced in the quasineutral region attain the inner boundary of the space-charge region. However, the high electron flux, due to both the space-charge region, and the photoionized impurities, immediately neutralizes their evolution and shortens their lifetime. When the film thickness increases, the quasineutral region becomes wider. More holes then are expected while fewer electrons come from the space-charge zone, since there are fewer back

TABLE II. Some transport properties of thin sprayed CdS films.

d (μm)	S 10^7 (cm s $^{-1}$)	τ_p 10^{-9} (s)	D_p 10^{-3} (cm 2 s $^{-1}$)
0.07	20	2.0	1.4
0.15	8.5	9.1	1.7
0.22	7.0	20.0	5.0
0.40	4.0	75.0	2.6
1.50	2.5	150.0	1.1
3.00	2.6	200.0	0.7

interface impurities ionized. The neutralization process and the recombination front shift towards the bulk because the surface electrical field attracts to a lesser extent the holes of the inner quasineutral region. Only some of them reach the space-charge boundary, thus leading to small S values. The holes which cross this boundary, safely reach the electrolyte, and thus have a greater lifetime. This process must go on until a certain limit is attained, this being the point where in the back, interface impurities become marginal. This occurs with thicker films, the L_p value being constant. At this stage, a stationary recombination speed becomes established.

When S and τ_p decrease regularly vs d , a different behavior of the diffusion coefficient is observed and this is consequently similar to that of the hole mobility. This behavior brings out the photoresponse property of these films previously reported.¹⁵ The maximum mobility value obtained at film thickness of roughly $0.3 \mu m$ in Fig. 5 seems to correspond with the maximum photocurrent, thus suggesting that this parameter probably has a role in the phenomenon. However a complete analysis of this subject also requires knowledge of the electron mobility μ_e . The μ_p values obtained here between 5×10^{-2} and $2.5 \times 10^{-1} cm^2 s^{-1} V^{-1}$ can not be compared, since there is no data in the literature on the CdS material at such a thickness zone. However, if one takes into account the ratio $\mu_e/\mu_p \simeq 6$ reported for this material,²⁸ the electron mobility values deduced seem compatible with those proposed by Siu *et al.*²⁹

VI. SUMMARY AND CONCLUSION

The characterization of very thin semiconductor films is sometimes difficult because the measurements are carried out at the limits of possibility. The usual technics therefore require some adaptations and more accuracy in these cases. This aspect is illustrated in this work by a lesser agreement of L_p values given by the thinnest samples from the two independent technics used. It is not claimed here that the values sought for each one of them have been attained. We are aware of the problems involved in trying to attain such a precise goal. However, this is some satisfaction from the fact that the same behavior of this parameter has been observed for both. This was our main objective. On this light, the val-

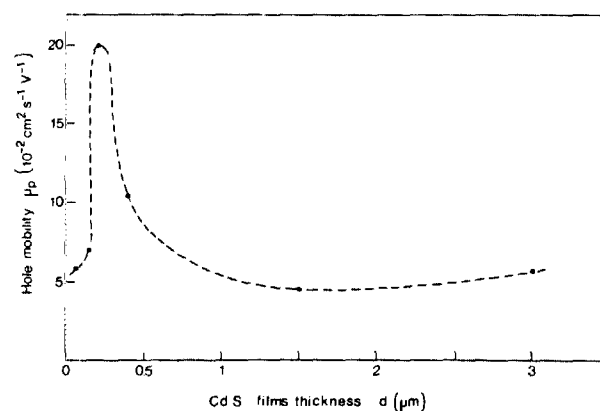


FIG. 5. Hole mobility as a function of thin sprayed CdS film thickness at ambient temperature, under monochromatic light ($\lambda = 510 nm$), with $0.26 mW cm^{-2}$ intensity.

ues of the other parameters deduced from L_p in the second part of this paper simply have an indicative significance, even though some concordance is found with those reported in the literature.

Taken as a whole, these results appear to characterize quite thinly sprayed CdS films. N_d , as well as L_p , both evolve rapidly but inversely where the back impurities are supposed to be most active. A quasiconstant value of the two parameters is observed when the film thickness is above $0.4\ \mu\text{m}$. The hole-mobility behavior is clearly similar to that of the photocurrent, at least in the increasing part.

Bearing in mind that our main purpose has been to complete the study of these films, we believe that more attention must be paid to the phenomena which can occur at the electron collector-semiconductor interfaces, when one is dealing with very thin films.

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