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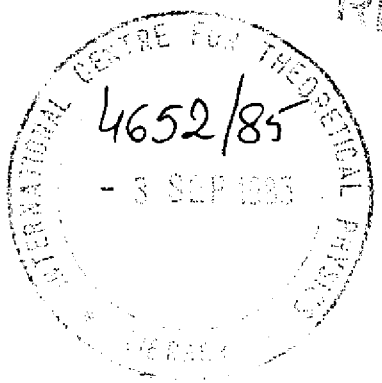

INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS

EXCITON DISTRIBUTION FUNCTION AND SECONDARY RADIATION IN POLAR SEMICONDUCTORS

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INTERNATIONAL ATOMIC ENERGY AGENCY

UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION

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EXCITON DISTRIBUTION FUNCTION AND SECONDARY RADIATION IN POLAR SEMICONDUCTORS *

C. Trallero Giner **
International Centre for Theoretical Physics, Trieste, Italy,

and

O. Sotolongo Costa
Department of Theoretical Physics, Havana University,
San Lazaro y L. Havana, Cuba.

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** Permanent address: Department of Theoretical Physics, Havana University, San Lazaro y L. Havana, Cuba.

ABSTRACT

An explicit non-equilibrium distribution function for excitons in the ground state n=1 in the case when the fundamental interaction is with acoustical phonons is calculated for polar semiconductors. Using it, a general expression for the secondary radiation cross-section (valid for Raman, hot and thermalized luminescence processes), is obtained. The results are applied to explain the temperature dependence of the 1LO and 2LO luminescence lines half-width in CdS single crystals.
The relative contributions of 3LO Raman and luminescence intensities and the variation of the secondary emission spectrum as function of exciton life-time are studied. Comparison with experimental results yields quantitative agreement.

I- INTRODUCTION

The study of secondary radiation of light by condensed matter is a powerful method of investigating elementary excitations within matter. There is a number of investigations dealing with this problem in polar semiconductors (see [1]).

It has been shown [2-6] that the excitonic model is a successful one to explain the process of secondary radiation in many polar semiconductors like CdS, ZnTe, CdSe, etc.

In CdS the excitonic spectrum of secondary radiation [2] and the relative intensities of 3LO-Raman and luminescence [3,4] were investigated. In general these processes of light scattering with participation of excitons as intermediate states of the crystal can be viewed in the following way:

a) Indirect creation of an exciton with kinetic energy $E \neq 0$ in the energetic state with principal quantum number n=1.

b) Exciton relaxation.

c) Indirect exciton annihilation with emission of a photon $\hbar\omega_s$ and at least one LO-phonon with energy $\hbar\omega_{LO}$. On the other hand, it is known [7] that the differential cross-section of the secondary emission radiated by an excited solid, is given by the formula:

$$\frac{d^2\sigma}{d\omega_s d\Omega} = \frac{V_0^2 \omega_s^2 n(\omega_s)}{(2\pi)^3 c^4 n(\omega_i)} W(\omega_s), \qquad (1)$$

where c is the velocity of light in vacuum, $n(\omega)$ the frequency-dependent refraction index, $W(\omega_s)$ the emission probability of a quantum of secondary radiation $\hbar\omega_s$ by unit of time and of solid angle Ω, when an incident light quantum $\hbar\omega_i$ is in the volume Vo with a refraction index equal to unity.

In terms of the exciton distribution function (EDF), $W(\omega_s)$ is given by:[8]

$$W(\omega_s) = \int P(E) W_i(E, \omega_s) dE \, . \qquad (2)$$

$W_i(E, \omega_s)$ is the annihilation probability per unit time and solid angle with photon emission of energy $\hbar\omega_s$ and emission of LO-phonons by an exciton having kinetic energy E, averaged through the angle between the directions of the exciton and photon momenta, and $P(E)dE$ is the number of excitons within the energy interval (E,E+dE). Although the EDF for polar semiconductors has been found when the exciton kinetic energy is greater than that of an optical phonon (i.e. $E > \hbar\omega_{LO}$)[8]; this problem up to now has not been solved for $E < \hbar\omega_{LO}$. This EDF, taking into account $W_i(E, \omega_s)$, gives the possibility to investigate the light scattering process by the above named semiconductors when the incident photon has an energy:

$$E_g - \Delta E < \hbar\omega_i < E_g - \Delta E + 2\hbar\omega_{LO} \, , \qquad (3)$$

here Eg is the gap and ΔE-the exciton binding energy. In the following, this is the energy interval that will be considered.

If the scattering processes are described by the relaxation time τ_s and the exciton annihilation by τ, the stationary population in the band is conditioned by the concurrence of both life-time and relaxation. If $\tau \gg \tau_s$, a thermal quasi-equilibrium of excito-

lons takes place leading to formation of thermalized luminescence If this condition is not fulfilled, the emission line tends to a Raman one with participation of real intermediate states.

In the case of thermalized luminescence, it is logical to expect from the EDF to be a Maxwell-Boltzmann one, whereas for the Raman line, the corresponding EDF must be a strong non-equilibrium one.

In section II we deal with the balance equation, obtaining an approximate expression of the EDF valid for both cases.

In section III an application of our results to experimental observations is made.

II. EXCITON DISTRIBUTION FUNCTION

The determination of the EDF is a complex problem, governed by the concrete physical conditions of the system. Let us neglect exciton-exciton interaction and consider that the exciton relaxation is conditioned only by the interaction with acoustical phonons (this is the case in enough clean polar semiconductors - often the exciton kinetic energy $E<\hbar\omega_{LO}$ at low temperatures). In the case of an isotropic EDF the balance equation is:

$$\frac{\partial P(E,t)}{\partial t} = W_{in} - W_{out} + W_{ex}\,\delta(E-E_0), \qquad (4)$$

where W_{ex} is the rate of radiative creation of a Wannier-Mott exciton with energy E_0, W_{in} represents all the contributions per unit time to the excitonic state with energy E, and conversely for W_{out}. Into the processes contained in W_{out} we have: 1) exciton scattering $w(E,E')$ from energy E to energy E' in the quantum states, performed by acoustical phonons, 2)phonon assisted transitions to states with n>1 (Wn), 3)exciton decay (Wd),4)LO-phonon assisted exciton annihilation with emission of a light quantum $\hbar\omega_s=\hbar\omega_f-k\hbar\omega_{LO}(K>1)\,(\frac{1}{\tau_R})$, 5)processes of non-radiative annihilation $(\frac{1}{\tau_{NR}})$. For the case of exciton-LA phonon interaction when the deformation potential model is used, it was shown [9] that the processes 2 and 3 for $E<\hbar\omega_{LO}$ are negligible. Similarly, this can be proved taking into account the piezoelectric model. Then we can write:

$$W_{out} = \int w(E,E')P(E,t)dE' + \frac{P(E,t)}{\tau}, \qquad (5)$$

In (5) the total lifetime of excitons $\frac{1}{\tau} = \frac{1}{\tau_R} + \frac{1}{\tau_{NR}}$ is governed by radiative and non radiative annihilation processes.

In the same way we may represent:

$$W_{in} = \int w(E',E)\,P(E',t)\,dE'. \qquad (6)$$

In the appendix a calculation of w(E,E') is performed for piezoelectric and deformation potential interaction hamiltonians. Using relations (4)-(6) and (A.16) from the appendix, the balance equation is transformed to:

$$\frac{\partial f(E,t)}{\partial t} = \int w(E,E')\left\{f(E',t) - f(E,t)\right\}dE' - \frac{f(E,t)}{\tau} + J(E_0)\,\delta(E-E_0), \qquad (7)$$

where

$$J(E_0) = W_{ex}\,\frac{e^{E_0/k_BT}}{\sqrt{E_0}}, \qquad (8)$$

$$f(E,t) = P(E,t)\,\frac{e^{E/k_BT}}{\sqrt{E}}\,. \qquad (9)$$

Noting that the exciton energy E is varied in a quite small amount after acoustical phonon absorption or emission, we may expand f(E',t) as:

$$f(E',t) = \sum_{n=0}^{\infty} \frac{d^n f}{dE^n}\,\frac{(E'-E)^n}{n!}\,. \qquad (10)$$

In the stationary case and retaining terms up to second order in (10), from (7) it follows that:

$$\frac{d^2 f}{dE^2} + \frac{a_1}{a_2}\,\frac{df}{dE} - \frac{f}{a_2 \tau} = -\,\frac{J(E_0)}{a_2}\,\delta(E-E_0), \qquad (11)$$

being:

$$a_n(E) = \frac{1}{n!}\int (E'-E)^n\, w(E,E')\,dE'\,. \qquad (12)$$

The coefficients $a_n(E)$ can be obtained by a direct integration from equation (A.9) and (A.13). It can be shown that for exciton kinetic energies $E>m u^2$, a_1 is negative.

It is clear that a direct solution of (11) is impossible. In order to make it possible we assume that the coefficients a_1 and a_2 are constants. Imposing that $P(E)\to 0$ for $E\to\infty$ and that the total number of particles in the stationary state is $N=W_{ex}\tau = \int P(E)\,dE$, we obtain

for the EDF the expression:

$$P(E) = W_{ex}\,\tau\, e^{-\left(\beta+\frac{1}{k_BT}\right)E}\sqrt{E}\left\{\frac{2}{\sqrt{\pi}}\left(\beta+\frac{1}{k_BT}\right)^{3/2} + \right.$$
$$+\frac{e^{\left(\beta+\frac{1}{k_BT}\right)E_0}}{2\sqrt{E_0}\,a_2\tau\sqrt{\left(\frac{a_1}{2a_2}\right)^2+\frac{1}{a_2\tau}}}\left[\Phi\left(\sqrt{E_0\left(\beta+\frac{1}{k_BT}\right)}\right) - \right. \qquad (13)$$
$$\left.\left. - \frac{2}{\sqrt{\pi}}\cdot\sqrt{\left(\beta+\frac{1}{k_BT}\right)E_0}\; e^{-\left(\beta+\frac{1}{k_BT}\right)E_0} - \Theta(E_0-E)\right]\right\},$$

where:

$$\beta = \frac{a_1}{2a_2} + \sqrt{\left(\frac{a_1}{2a_2}\right)^2+\frac{1}{a_2\tau}}\,, \qquad (14)$$

$\Phi(x)$ is the error function and $\Theta(x)$ the Heaviside's unit step function defined as:

$$\Theta(x) = \begin{cases} 1 & ; \quad x>0 \\ 0 & ; \quad x<0 \end{cases}\,. \qquad (15)$$

If τ is great enough, EDF (13) reduces to a Maxwell-Boltzmann one:

$$P(E) = \frac{2W_{ex}\tau}{\sqrt{\pi}\,(k_BT)^{3/2}}\sqrt{E}\,e^{-E/k_BT}\,. \qquad (16)$$

III- COMPARISON WITH EXPERIMENTAL RESULTS

In this section we will apply our theoretical result to the experimental observations reported in [2,3,4].

a) Half-width of 1LO and 2LO luminescence lines.

In [2] the half-width of 1LO and 2LO luminescence lines in CdS was measured as a function of temperature using an exciting radiation of wavelength λ=485.3 nm creating excitons with a kinetic energy $E\simeq\hbar\omega_{LO}$.

For the 1LO luminescence line the annihilation probability $W_1(E,\omega_s)$ is given by:

$$W_1(E,\omega_s) = \frac{2\pi}{\hbar} \sum_{\vec{q}} |M|^2 \delta(\hbar\omega_{LO} + \hbar\omega_s - E), \qquad (17)$$

where $\vec{q}$ is the phonon wavevector and M is the transition matrix - element given by:

$$M = \sum_b \frac{\langle f|H_{e-l}|b\rangle\langle b|H_{e-p}|i\rangle}{E_i - E_b + i\delta}, \qquad (18)$$

the sum in (18) runs for all virtual excitonic states. H_{e-l} (H_{e-p}) is the exciton-light (exciton-LO phonon) interaction hamiltonian. In [10] this matrix element was calculated. For the energy range we deal with, the influence of the continuous spectrum can be proved to be negligible. Then we have:

$$|M(E)|^2 = \frac{\hbar\omega_s}{E} \propto \Lambda^2 z^{10} \hbar^2 \omega_{LO}^3 \left[\sum_{n=1}^{4} \frac{1}{n^3} \frac{1}{(E - \hbar\omega_{LO} + \Delta E(\frac{1}{n^2} - 1))} \cdot \right.$$

$$\left. \cdot \left[\frac{P_n\left(\frac{m_2}{m_1}\frac{E}{\Delta E}\right)}{\left[\frac{m_2}{m_1}\frac{E}{\Delta E} + \left(\frac{n+1}{n}\right)^2\right]^{n+1}} - \frac{P_n\left(\frac{m_1}{m_2}\frac{E}{\Delta E}\right)}{\left[\frac{m_1}{m_2}\frac{E}{\Delta E} + \left(\frac{n+1}{n}\right)^2\right]^{n+1}} \right]^2, \qquad (19)$$

where:

$$P_1(y) = 1 \; ; \quad P_2(y) = y \; ; \quad P_3(y) = y^2 + \frac{1}{3}\left(\frac{4}{3}\right)^2 y \; ; \qquad (20)$$

$$P_4(y) = y^3 + \frac{11}{8} y^2 + \frac{1}{4}\left(\frac{5}{4}\right)^3 y,$$

m_1 (m_2) is the effective electron (hole) mass, and α and Λ are constants given in [10].

Using (1), (2) and (17) we obtain:

$$\frac{d^2\sigma}{d\omega_s d\Omega} = \frac{V_0^{-2}\omega_s^2 n(\omega_s)}{(2\pi)^2 c^4 n(\omega_k)\hbar} P(\hbar\omega_s + k\hbar\omega_{LO}) |M(k\hbar\omega_{LO} + \hbar\omega_s)|^2. \qquad (21)$$

Formula (21) permits us to obtain the thermal increase of 1LO luminescence line half-width. A detailed analysis of the experimental results lead us to conclude that in the sample used in the experiment τ is a big one, so we may use the Maxwell-Boltzmann -- EDF (16) in (21).

Concerning 2LO luminescence line, it can be seen that two phonon exciton annihilation probability is independent of energy as was shown in [2]. By this reason the differential cross-section can be written as:

$$\frac{d^2\sigma}{d\omega_s d\Omega} = \frac{V_0^2 \omega_s^2 n(\omega_s)}{(2\pi)^2 c^4 n(\omega_k)\hbar} |M|^2 P(\hbar\omega_s + 2k\omega_{LO}), \qquad (22)$$

where for this case the matrix element M is independent of energy.

In figure 1 the comparison of theoretical (using formulae (21) - and (22) without any adjustment) and experimental results is shown. The half-widths are given in units of $\hbar\omega_{LO}$ (for CdS $\hbar\omega_{LO}$ is equal to 0,038 eV).

b) Temporal evolution of exciton secondary emission spectrum

In [3] the experimental study of the secondary emission in CdS - single crystals, using samples with different free exciton life-time was reported. It was seen that the relation of 3LO luminescence integral intensity with 3LO Raman emission line is highly dependent of the relation between exciton life-time and acoustical

relaxation time τ_s. The crystals were excited at 2K by a laser -- line λ = 476,5 nm. In this case 1LO assisted exciton creation with kinetic energy $E \simeq \hbar\omega_{LO}$ is performed. This means that 3LO emission line is obtained by 2LO assisted exciton annihilation, whose probability, as we already know, is independent of energy. Then, for 3LO line intensity (I_{3LO}) we can use formula (22). From this formula it follows that the integral intensity I_Σ is proportional to the whole number of excitons in the band $W_{exc}\tau$.

In order to obtain I_{3LO}/I_Σ it is necessary to evaluate a_1 and a_2 which can be expressed approximately as:

$$a_1 \simeq -\frac{\Delta\varepsilon}{\tau_s} \quad ; \quad a_2 \simeq \frac{1}{2}\left(\frac{\Delta\varepsilon}{\tau_s}\right)^2, \tag{23}$$

which follows from (12). In (23) $\Delta\varepsilon$ is the characteristic energy of the acoustical phonons, equal to $2mu^2$. With this it is possible to show that:

$$\frac{I_{3LO}}{I_\Sigma} = \frac{A}{\frac{\tau}{\tau_s}\sqrt{1+2\frac{\tau_s}{\tau}}}, \tag{24}$$

being A a certain constant.

In table 1 a comparison of experimental values with those obtained from (24) is presented for different samples.

In figure 2 secondary emission spectra of CdS samples with different lifetimes of excitons, reported in [3] are presented and compared with results obtained from (22). As can be seen the EDF given by (13) satisfactorily reflects the temporal evolution of secondary emission spectrum as a function of exciton lifetime.

The observed emission spectrum in the region near 499 nm is due to the contribution of bound excitons which are not taken into account in our model.

In [4] a comparison of the integral luminescence intensity I_Λ with I_{3LO} as a function of $\frac{\tau}{\tau_s}$ was presented. This luminescence -- I_Λ reveals itself as a band of hot luminescente in the long wave-lenght part of the 3LO line. Its integral intensity is equal to:

$$I_\Lambda = I_\Sigma - I_{3LO}. \tag{25}$$

Then from (24) we have:

$$\frac{I_\Lambda}{I_{3LO}} = \frac{1}{A}\frac{\tau}{\tau_s}\sqrt{1+2\frac{\tau_s}{\tau}} - 1. \tag{26}$$

In figure 3 the comparison with experimental results is shown. The theoretical curve reveals that, as in the experiment, the increase of τ leads to a more effective exciton thermalization.

Finally, from the obtained results it is important to note that the formula (1) derived from the general theory of secondary radiation [7] and the EDF given by (12) may be used to investigate Raman scattering processes as well as hot and equilibrium luminescence for exciton kinetic energy $E \simeq \hbar\omega_{LO}$. The obtained EDF, expressed as a function of τ, τ_s and temperature reflects the above showed experimental facts that concurrence of acoustic relaxation and exciton lifetime determines exciton population in the band.

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APPENDIX

THE SCATTERING PROBABILITY w(E,E')

The exciton piezoelectric phonon interaction hamiltonian is:

$$H_{e\text{-}p}=\sum_{j,\nu,\vec{q}}\left(C_{j\nu\vec{q}}\,e^{i\vec{q}\cdot\vec{r}_j}\,b_{\nu\vec{q}}+C^{*}_{j\nu\vec{q}}\,e^{-i\vec{q}\cdot\vec{r}_j}\,b^{+}_{\nu\vec{q}}\right), \tag{A.1}$$

where $j=1,2$ corresponding respectively to electron and hole, $\vec{r}$ the radius vector, $b_{\nu\vec{q}}$ ($b^{+}_{\nu\vec{q}}$) are the annihilation (creation) operators of the acoustic phonon in the branch ν with wavevector $\vec{q}$, and

$$C_{j\nu\vec{q}}=(-1)^{j+1}\frac{2\pi e\,\beta_{\alpha,\mu\lambda}}{\varepsilon_{mn}\,q_m q_n}\left[\frac{\hbar}{2V_0\rho_0\omega_\nu(\vec{q})}\right]^{1/2}q_\alpha\left(e_{\nu\mu}(\vec{q})q_\lambda+e_{\nu\lambda}(\vec{q})q_\mu\right), \tag{A.2}$$

e is the electron charge, $\beta_{\alpha,\mu\lambda}$ the tensor of piezoelectric moduli, ε_{mn} the dielectric tensor, V_0 the normalization volume, ρ_0 the crystal density; ω_ν and $\vec{e}_\nu$ the frequency and polarization vector. Considering exciton wavefunctions for the initial and final states as being of the type:

$$\psi=\frac{1}{\sqrt{V_0}}\,e^{i\vec{K}\cdot\vec{R}}\,\frac{e^{-\rho/a}}{\sqrt{\pi a^3}}, \tag{A.3}$$

where $\vec{R}$ is the radius vector of the center of mass of the exciton, and a its Bohr's radius, and $\rho=|\vec{r}_1-\vec{r}_2|$.

The calculation of w(E,E') can be made through Fermi's golden rule, according to which the transition probability is:

$$W(E)=\frac{2\pi}{\hbar}\sum_f|\langle f|H_{e\text{-}p}|i\rangle|^2\,\delta(E_f-E_i) \tag{A.4}$$

Using (A.1)-(A.4) we obtain:

$$W(E)=\frac{e^2\bar{\gamma}\sqrt{2m}}{8\pi\hbar^2}\frac{1}{\sqrt{E}}\left\{\int_E^{(\sqrt{E'}+\sqrt{2mu^2})^2}dE'\,F\left(a\frac{E'-E}{\hbar u}\right)N\left(\frac{E'-E}{k_BT}\right)+\right.$$

$$\left.+\int_{(\sqrt{E'}-\sqrt{2mu^2})^2}^{E}dE'\,F\left(a\frac{E-E'}{\hbar u}\right)\left[1+N\left(\frac{E-E'}{k_BT}\right)\right]\right\}, \tag{A.5}$$

where:

$$F(z)=\left\{\left[1+\left(\frac{m_2}{m}z\right)^2\right]^{-2}-\left[1+\left(\frac{m_1}{m}z\right)^2\right]^{-2}\right\}^2 \tag{A.6}$$

$$\left.\begin{aligned}N(x)&=\left[e^x-1\right]^{-1}\\ m&=m_1+m_2\end{aligned}\right\}, \tag{A.7}$$

$\bar{\gamma}$ is defined as in [11] like the mean value of:

$$\sum_\nu\lim_{q\to 0}\frac{1}{\rho_0 u_\nu^2(\vec{q})}\left|\frac{2\pi\beta_{\alpha,\mu\lambda}}{\varepsilon_{mn}q_m q_n}q_\alpha\left[e_{\nu\mu}(\vec{q})q_\lambda+e_{\nu\lambda}(\vec{q})q_\mu\right]\right|^2 \tag{A.8}$$

with respect to the angles formed between the direction of $\vec{q}$ relative to the crystal axes. In [11,12] this value was found to be $\bar{\gamma}=\left(\frac{4\pi}{\varepsilon_0}\right)^2 2{,}68\cdot10^{-2}$ for CdS. The scattering probability by unit of time and energy is:

$$w(E,E')=\begin{cases}\dfrac{\bar{D}}{\sqrt{E}}N\left(\dfrac{E'-E}{k_BT}\right)F\left(a\dfrac{E'-E}{\hbar u}\right), & E\le E'\le(\sqrt{E}+\sqrt{2mu^2})^2\\[2ex] \dfrac{\bar{D}}{\sqrt{E}}\left[1+N\left(\dfrac{E-E'}{k_BT}\right)\right]F\left(a\dfrac{E-E'}{\hbar u}\right), & (\sqrt{E}-\sqrt{2mu^2})^2\le E'\le E,\end{cases} \tag{A.9}$$

here:

$$\overline{\overline{D}} = \frac{e^2 \bar{\gamma} \sqrt{2m}}{8\pi \hbar^2} . \qquad (A.10)$$

In the same way w(E,E') may be calculated using the deformation potential model, where the exciton-phonon interaction hamiltonian is:

$$H_{e-p} = \sum_{\vec{q},j} \left\{ C_{j\vec{q}} e^{i\vec{q}\cdot\vec{r}_j} b_{\vec{q}} + C^*_{j\vec{q}} e^{-i\vec{q}\cdot\vec{r}_j} b^+_{\vec{q}} \right\}, \qquad (A.11)$$

where:

$$C_{j\vec{q}} = (-1)^{j+1} \left(\frac{\hbar}{2V_0 \rho_0 u} \right)^{1/2} q^{1/2} E_j , \qquad (A.12)$$

and E_j is the deformation potential for electrons or holes.

In this case:

$$w(E,E') = \begin{cases} \frac{D}{\sqrt{E}} (E'-E)^2 N\left(\frac{E'-E}{k_B T}\right) F_d\left(a \frac{E'-E}{\hbar u}\right); & E \le E' \le \left(\sqrt{E} + \sqrt{2mu^2}\right)^2 \\ \frac{D}{\sqrt{E}} (E-E')^2 \left[1 + N\left(\frac{E-E'}{k_B T}\right)\right] F_d\left(a \frac{E-E'}{\hbar u}\right); & \left(\sqrt{E} - \sqrt{2mu^2}\right)^2 \le E' \le E , \end{cases} \qquad (A.13)$$

$$D = \sqrt{m}\, E_1^2 / (4\pi \sqrt{2}\, \rho_0 u^4 \hbar^4) , \qquad (A.14)$$

$$F_d(z) = \left\{ \left[1 + \left(\frac{m_2}{m} z\right)^2\right]^{-2} - \frac{E_2}{E_1} \left[1 + \left(\frac{m_1}{m} z\right)^2\right]^{-2} \right\}^2 . \qquad (A.15)$$

In both models (piezoelectric and deformation potential) it can be seen that:

$$w(E',E) = \sqrt{\frac{E}{E'}}\, e^{\frac{(E'-E)}{k_B T}}\, w(E,E') . \qquad (A.16)$$

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FIGURE CAPTIONS

Fig. 1: Broadening of exciton emission lines in units of $\hbar\omega_{LO}$ in CdS with temperature. Lines 1 and 2 were obtained using formulae (21) and (22) respectively. The half-widths are those at 1/e of the maximum intensity value.

Fig. 2: Exciton secondary emission spectra for CdS samples with different lifetimes. Dotted curves are theoretically predicted with formula (22). The samples are the corresponding ones to table 1.

Fig. 3: I_A / I_{3LO} vs. $s = \frac{\tau}{\tau_s}$. The dots represent experimental observations for different samples. The curve was obtained with formula (26).

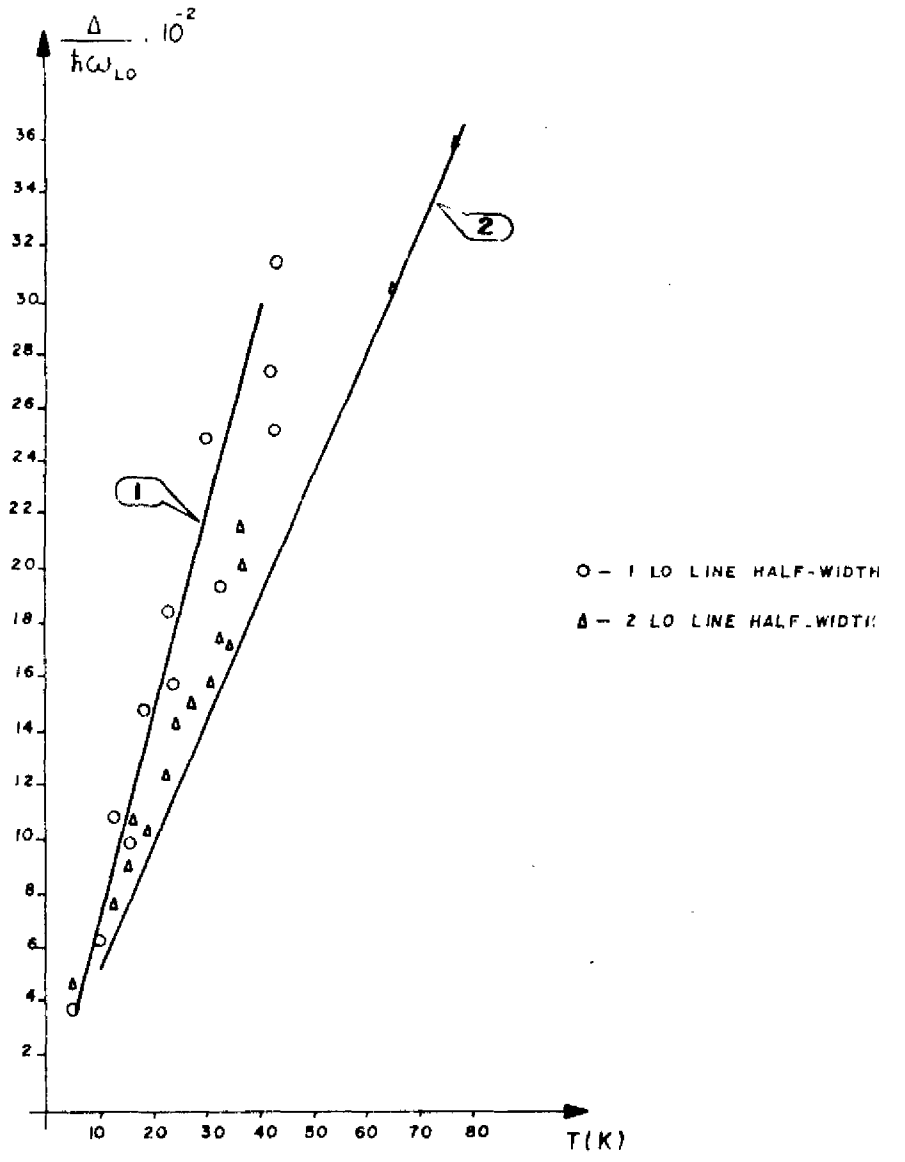
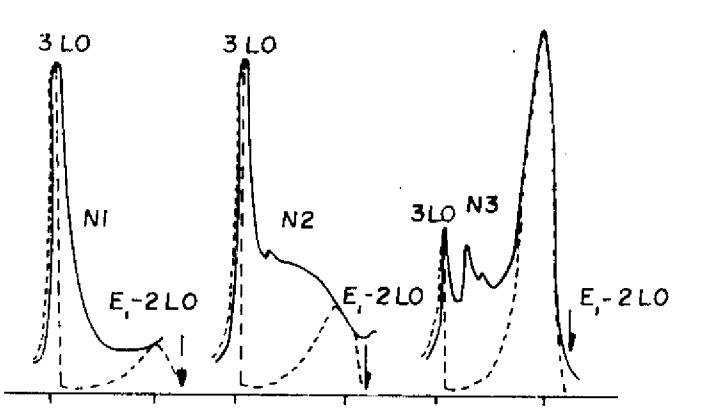


Fig.1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λ (nm)

Fig.2

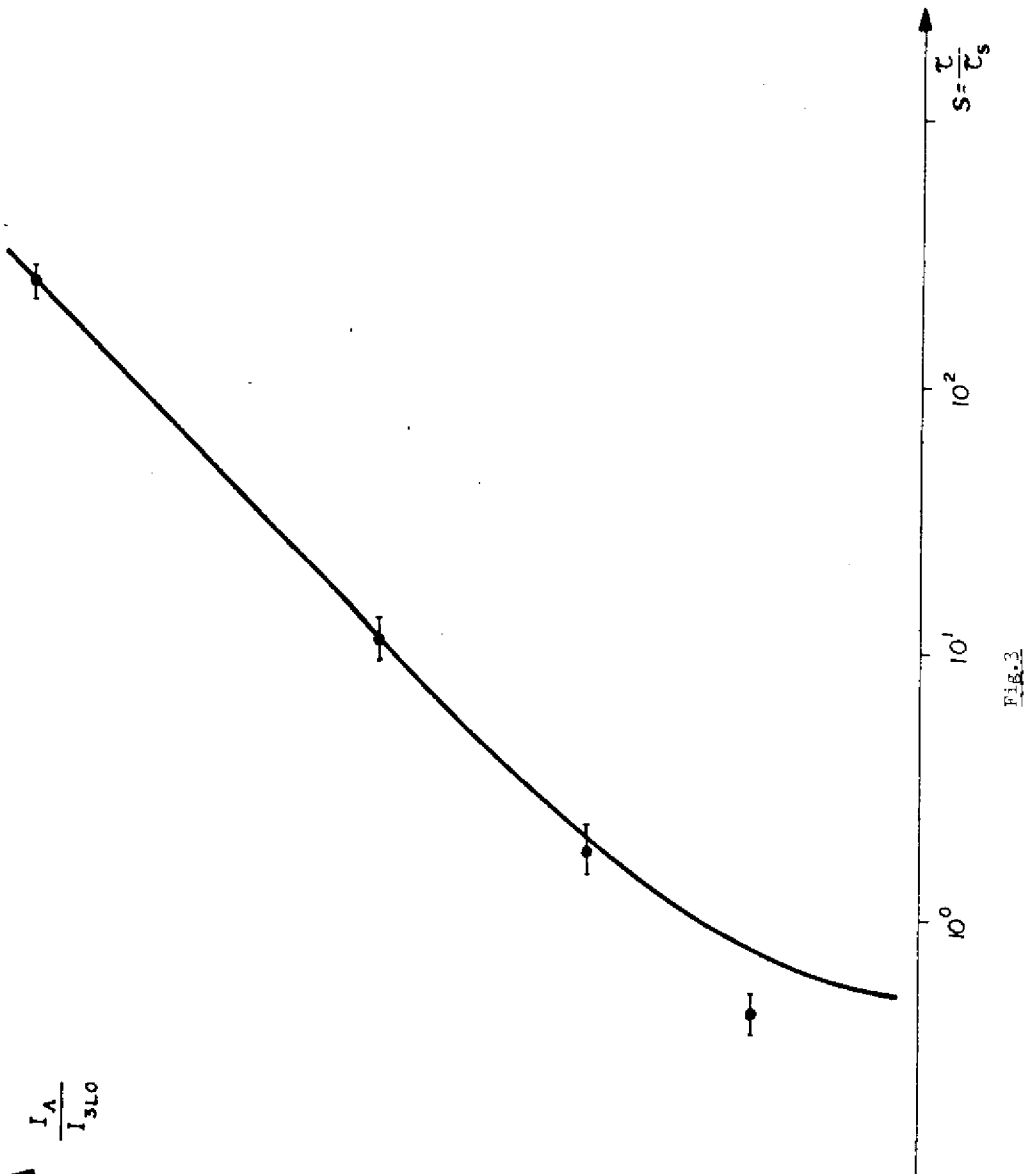
$\frac{I_A}{I_{3LO}}$
10^2
10^1
1
10^{-1}
$S=\frac{\tau}{\tau_s}$
10^2
10^1
10^0

Fig.3

SAMPLE	τ (s)	τ_s (s)	$\frac{I_{3LO}}{I_\Sigma}$ (EXPERIMENTAL)	$\frac{I_{3LO}}{I_\Sigma}$ (THEORETICAL)
N_1	$3 . 10^{-12}$	$3,5.10^{-12}$	0,6	0,6
N_2	$9 . 10^{-12}$		0,289	0,274
N_3	$4,5 . 10^{-12}$		$7,03.10^{-2}$	$6,8'.10^{-2}$
N_4	$1,6 . 10^{-12}$		$2,16.10^{-3}$	$2,05.10^{-3}$

Table I – COMPARISON OF $\frac{I_{3LO}}{I_\Sigma}$ OBTAINED FROM (24) WITH EXPERIMENTAL DATA τ, τ_s, I_{3LO} AND I_Σ ARE REPORTED IN [3] FOR DIFERENT SAMPLES. – THE PROPORTIONALITY CONSTANT A WAS ADJUSTED TO SAMPLE N_1 . –