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To cite this version:
N. Piccioli, R. Le Toullec. Exciton-phonon interaction in GaSe. Journal de Physique, 1989, 50 (23), pp.3395-3406. <10.1051/jphys:0198900500230339500>. <jpa-00211150>

HAL Id: jpa-00211150
https://hal.archives-ouvertes.fr/jpa-00211150
Submitted on 1 Jan 1989

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Exciton-phonon interaction in GaSe

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(Reçu le 21 juin 1989, accepté sous forme définitive le 17 août 1989)

Abstract. — The study of the temperature dependence of the optical properties of GaSe confirms the dominating role of the homopolar phonon at 16.7 meV. The quantitative study of the energy levels variations, near the fundamental absorption edge, yields a deformation potential value of $\varepsilon_d = 5.5$ eV $\cdot \text{Å}^{-1}$, in good agreement with the value deduced from the band calculations by Schlüter ($\varepsilon_d = 5.8$ eV $\cdot \text{Å}^{-1}$). The quantitative study of the temperature dependence of the full width at half maximum height of the first excitonic level exhibits a contribution parallel to the c-axis in the kinetic mass which has to be taken into account in the exciton — phonon interaction; the value of the kinetic mass (0.81 m) which provides the better agreement with the experiment is located between $M_{\parallel}$ and $M_\perp$.

1. Introduction.

Optical and transport properties of the lamellar III-VI compound GaSe have been reported in a number of papers. We have shown previously [1] that in the case of samples with small amounts of defects, the low temperature absorption coefficient can be quantitatively analysed using a model deduced from Elliott's theory [2]. This model leads to an asymmetrical lorentzian absorption shape for the exciton. Comparison with our experimental results has enabled us to determine the excitonic parameters such as the exciton binding energy $R$, the full width at half maximum $\Gamma_n$ and the energy gap $\sigma_G$.

We have extended this study by varying the sample temperature from 10 K to 504 K. The results presented in this paper concern particulary the thermal evolution of the energy gap.
\[ \sigma_G \] and the full width at half maximum \( \Gamma_1 \) of the first excitonic peak. Following band structure calculations \([3, 4]\), the width of the gap, represented by the distance between the \( \Gamma^-_4 \) and \( \Gamma^+_3 \) levels, is expected to be strongly sensitive to any process which might modify inter-layer distances. In the case of GaSe, there is a symmetrical compression mode which modulates the inter-layer space along the c-axis : the \( \text{A}_1' \) homopolar phonon with a room temperature energy of 16.70 meV (134.6 cm\(^{-1}\)). Figure 1 shows the motion of atoms in this mode. The contribution of this phonon to the variation of the energy level \( \text{versus} \) temperature is expected to be predominant, which is precisely what was observed by Schmid and Voitchovsky \([5]\) through their study of the hole mobility \( \text{versus} \) temperature. In the present work, we show that this phonon indeed dominates electron-phonon and exciton-phonon interactions at a variable temperature. In section 2, we present our experimental results showing the evolution of the absorption coefficient \( K_\perp \) \( \text{versus} \) temperature for the case where the electric vector of the radiation is polarised perpendicular to the c-axis (\( E_\perp \ c \)). In section 3, we discuss the variation of the exciton energy levels \( \sigma_n \) and the energy gap \( \sigma_G \) with temperature. In the last section, we show the major contribution to the exciton-phonon interaction to be that of the homopolar phonon, as compared to that of the exciton-acoustical phonon, from the interpretation of the full width at half maximum \( \Gamma_1 \) of the first excitonic peak and its variation with temperature.

![Fig. 1. — Motion of atoms in the symmetrical compression mode of the interlayer gap along the c axis. • Ga ; O Se.](image)

2. Experimental results.

Transmission measurements were performed on a T.800 triple grating monochromator, using a quartz-iodine incandescent lamp as the light source. This apparatus has a resolution which is comparable to that of a 2.4 m monochromator, with a stray-light rejection level improved by several orders of magnitude. The slit widths are 170 \( \mu \)m which corresponds to a resolution limit of \( d\sigma = 1 \text{ cm}^{-1} \). The minimum measurable transmission is then \( T_{\min} = 5 \times 10^{-8} \) \([1, 6]\).

Figure 2 shows some of our results obtained at 10 K, 78 K, 130 K, 296 K, and 504 K. The experimental points for \( K_\perp \) are deduced from our transmission measurements, and the theoretical curves from calculations using reference \([1]\). One can see that the first two excitonic lines are still very well defined at 78 K, while at 130 K, the \( n = 2 \) level is hardly detectable.
3. Energy level variation versus temperature.

The temperature coefficient for an electronic level $\sigma_n$ measured at a given temperature is due to two different contributions [7, 8]:

$$ \frac{\partial \sigma_n}{\partial T} \bigg|_P = \frac{1}{V} \frac{\partial V}{\partial T} \bigg|_P \sigma_n + \frac{\partial \sigma_n}{\partial T} \bigg|_V. $$

(1)
The first term represents the temperature dependance of the volume due to the phonon-phonon interaction while the second term is related to the electron-phonon interaction which we are studying.

a) PHONON-PHONON INTERACTION. — The first contribution can be written as:

\[
\frac{1}{V} \left. \frac{\partial V}{\partial T} \right|_P V \left. \frac{\partial \sigma_n}{\partial V} \right|_T = \alpha \cdot \sigma_d
\]

\[
\alpha = \frac{1}{V} \left. \frac{\partial V}{\partial T} \right|_P : \text{thermal expansion coefficient}
\]

\[
\sigma_d = V \left. \frac{\partial \sigma_n}{\partial V} \right|_T = \left. \frac{\partial \sigma_n}{\partial P} \right|_T V \left. \frac{\partial P}{\partial V} \right|_T = -\frac{1}{\chi} \left. \frac{\partial \sigma_n}{\partial P} \right|_T
\]

\(\sigma_d\) : deformation potential for the \(\sigma_n\) band

\(\chi = -\frac{1}{V} \left. \frac{\partial V}{\partial P} \right|_T : \) isothermal compressibility.

For a uniaxial crystal such as GaSe:

\[
\alpha = 2 \alpha_\perp + \alpha_\parallel \quad \text{and} \quad \chi = 2 \chi_\perp + \chi_\parallel
\]

where \(\perp\) and \(\parallel\) designate the directions perpendicular and parallel to the c-axis [7].

\(\alpha_\parallel\) and \(\alpha_\perp\) were measured by Aliev et al. [9] at temperatures up to 400 K.

\(\chi_\parallel\) and \(\chi_\perp\) have been calculated using the elastic constants obtained through Brillouin scattering [10, 11]. The results are:

\[
\chi_\parallel = 24.9 \times 10^{-3} \text{ GPa}^{-1}; \quad \chi_\perp = 5 \times 10^{-3} \text{ GPa}^{-1}.
\]

The pressure coefficient of the band gap \(dE_G/dP\) was measured by Besson et al. at 300 K [12]:

\[
\left. \frac{dE_G}{dP} \right|_T = (-4.2 \pm 0.3) \times 10^{-2} \text{ eV.GPa}^{-1}.
\]

The deformation potential of the band gap is obtained as:

\[
E_d = -\frac{1}{\chi} \left. \frac{\partial E_G}{\partial P} \right|_T = (1.2 \pm 0.1) \text{ eV} \quad \text{or} \quad \sigma_d = (9680 \pm 807) \text{ cm}^{-1}.
\]

Assuming this potential to be independent of temperature, one can write the first contribution as:

\[
\Delta \sigma_G^{(1)}(T) = \sigma_d \int_0^T \alpha \, dT.
\]

Table I shows values of this integral up to 400 K, which we have extrapolated to 500 K.

| Table I. — Variation of \(\int_0^T \alpha \, dT\) versus temperature. |
|-------------|---------|---------|---------|---------|---------|---------|---------|---------|
| \(T\) (K)   | 60      | 80      | 100     | 150     | 200     | 250     | 300     | 400     |
| \(\int_0^T \alpha \, dT\) \((10^{-4})\) | 2.79    | 6.18    | 10.85   | 26.17   | 42.40   | 57.90   | 72.87   | 101.04  |
b) ELECTRON-PHONON INTERACTION. — The second contribution to equation (1) comes from the interactions of electrons or holes with phonons. Schmid and Voitchovsky [5] have shown the hole mobility in GaSe to be limited by the interaction between the charge carriers and the longitudinal homopolar phonon $A_1^i$. These authors conclude that the variation of the energy gap can be interpreted in terms of the same interaction:

$$\Delta E_G^{(2)}(T) = \frac{8 \ln 2}{\pi} g^2_v \sqrt{\frac{\hbar^2 Q^2}{2 m^*_v}} \sqrt{\hbar \Omega_h n_h}$$  \hspace{1cm} (4)$$

where $v$ refers to the valence band, $m^*_v$ is the density of states effective mass, $\hbar \Omega_h$ is the phonon energy and $n_h$ is the occupation number:

$$n_h = \left[ \exp \left( \frac{\hbar \Omega_h}{k_B T} \right) - 1 \right]^{-1}. \hspace{1cm} (5)$$

Using reference [13], $Q$ is defined as:

$$Q = \left[ \frac{3 V_{BZ}}{4} \right]^{1/3} \hspace{1cm} (6)$$

where $V_{BZ}$ is the volume of the first Brillouin zone (Fig. 3) and $g_v$ is the coupling constant. The calculated value for $Q$ is $0.67 \times 10^8$ cm$^{-1}$. In the case of intraband interactions between carriers and homopolar optical phonons, $g_v$ is defined by:

$$g^2_v = \frac{\varepsilon_d^2 m^*_v^{3/2}}{2^{3/2} \pi h M_i N_i (\hbar \Omega)^{3/2}}$$ \hspace{1cm} (7)$$

where $M_i$ is the reduced ionic mass, $N_i$ is the number of cells per unit volume and $\varepsilon_d$ is the deformation potential for a unit displacement of the atoms in a normal co-ordinate system.

Fig. 3. — First Brillouin zone in GaSe.

In order to calculate $\Delta E_G^{(2)}$, one has to know the electron and hole effective masses. These are given in references [14, 15] as:

$$\frac{m_{h_i}^*}{m_{h_f}^*} = 4 ; \quad \frac{m_{e_\perp}^*}{m_{e_\parallel}^*} = 0.57 ; \quad \frac{m_{h_i}^*}{m_{e_\perp}^*} = 4.7.$$
Our own experimental measurements of the dielectric constants $\varepsilon_{0z}$ and $\varepsilon_{0\parallel}$ [16] and of magneto-absorption [17, 18] have enabled us to determine the reduced mass of the exciton as:

$$m^*_z = \frac{m^*_e \cdot m^*_h}{m^*_e + m^*_h} \approx 0.12 m$$

where $m$ is the free electron effective mass.

From the above data, we can deduce the electron effective masses:

$$m^*_e = 0.15 m; \quad m^*_e' = 0.26 m$$

the hole effective masses:

$$m^*_h = 0.71 m; \quad m^*_h' = 0.18 m$$

and the density of states effective mass for the valence band:

$$m^*_v = (m^*_e^{2/3} \cdot m^*_h^{2/3})^{1/3} \approx 0.45 m.$$  

The variation of $h\Omega_h$ versus temperature is assumed to be linear and is deduced from the values found by Allakhverdiev et al. [19], at 8 K and 295 K. $M_i^{-1}$ is defined by Schmid [13] as:

$$M_i^{-1} = \sum_a m_a^{-1} |e'_a|^2$$  

where the summation is over the number of atoms in the unit cell with the normalisation condition that:

$$\sum_a |e'_a|^2 = 1$$

$M_i^{-1}$ is calculated using a rigid half-layer model so that:

$$e'_{z1} (\approx e'_{z2}) = e'_{z4} (\approx e'_{z8})$$

$$e'_{z2} (\approx e'_{z6}) = e'_{z3} (\approx e'_{z7}).$$

The $e'_a$ coefficients have been calculated by Polian [20] who founds $e'_{z1}/e'_{z2} = 2.23$.

The number of unit cells per unit volume is given by:

$$N_i = \frac{2}{a^2 c(3)^{1/2}}$$

with

$$a = 3.755 \text{ Å} \quad \text{and} \quad c = 15.946 \text{ Å}.$$  

The calculation leads to $M_i N_i = 0.66 \text{ g.cm}^{-3}$, with $\varepsilon_d$ still to be determined. Schlüter [4] has deduced $\varepsilon_d = 5.8 \text{ eV} \cdot \text{Å}^{-1}$ from band structure calculations, while Schmid and Voitchovsky [5] propose $\varepsilon_d = 6.6 \text{ eV} \cdot \text{Å}^{-1}$ from mobility calculations. The variation of the energy gap versus temperature is given by:

$$\sigma_G(T) = \sigma_G(0) - (\Delta \sigma_G^{(1)} + \Delta \sigma_G^{(2)}).$$  

The energies of the excitonic levels are given by:

$$\sigma_n(T) = \sigma_G(T) - \frac{R}{n^2}$$
where the binding energy of the exciton is given by reference [1] as:

\[
R = R_0 F(A) \frac{m^*_\perp}{m} \frac{1}{\varepsilon_0 \perp \cdot \varepsilon_0 f}
\]  

(11)

where \( R_0 = 109737.3 \text{ cm}^{-1} \) (Rydberg wave number) and \( F(A) \) is a function of the anisotropy parameter:

\[
A = \frac{\varepsilon_0 \perp m^*_\perp}{\varepsilon_0 f m_f}.
\]  

(12)

In order to take into account the temperature dependence of \( R \), the electron and hole effective masses were made to vary as the energy gap. A linear variation was assumed for the dielectric constants using the low temperature and room temperature values. Table II shows the values of parameters used for the calculation of \( R \). Our experimental results for the temperature dependence of the excitonic levels and the energy gap are shown in figure 4. The calculated curve is obtained for \( \varepsilon_d = 5.5 \text{ eV.Å}^{-1} \) and \( \sigma_G(0) = 17184 \text{ cm}^{-1} \). For \( T \geq 200 \text{ K} \), we observe a linear variation of the energy levels with a coefficient \( d\sigma_G/dT = -4 \text{ cm}^{-1}.\text{K}^{-1} \) \((-0.5 \text{ meV.K}^{-1})\) in good agreement with the results of reference [5].

![Fig. 4. Temperature dependence of the energy levels: the circles are directly deduced from experimental transmission coefficient spectra; the squares are deduced from the adjustment between the absorption coefficient spectra and theoretical curves using Elliott's theory; the solid lines are the calculated curves.](image-url)
Table II. — Temperature dependence of the different parameters used to calculate $R$.

<table>
<thead>
<tr>
<th>$T$ (K)</th>
<th>$\varepsilon_{0\perp}$</th>
<th>$\varepsilon_{0|}$</th>
<th>$\frac{m^*}{m}$</th>
<th>$\frac{m^f}{m}$</th>
<th>$A$</th>
<th>$F(A)$</th>
<th>$R$ (cm$^{-1}$)</th>
</tr>
</thead>
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<td>0.12</td>
<td>0.101</td>
<td>2.06</td>
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</tr>
<tr>
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<td>6.04</td>
<td>0.12</td>
<td>0.101</td>
<td>2.05</td>
<td>0.7693</td>
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</tr>
<tr>
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<td>0.1008</td>
<td>2.00</td>
<td>0.7768</td>
<td>153</td>
</tr>
<tr>
<td>500</td>
<td>10.74</td>
<td>6.30</td>
<td>0.1122</td>
<td>0.0962</td>
<td>1.99</td>
<td>0.7784</td>
<td>142</td>
</tr>
</tbody>
</table>

4. Variation of $\Gamma_1$ versus temperature.

The full width at half maximum $\Gamma_1$ of the $n = 1$ level is due to three contributions:

$$\Gamma_1 = \Gamma_0 + \Gamma_{ac} + \Gamma_{op}$$  \hspace{1cm} (13)

where $\Gamma_0$ is the low temperature limiting value of $\Gamma_1$ resulting from the exciton scattering on the native defects in the crystal, $\Gamma_{ac}$ is the contribution of the exciton-acoustic phonon interaction and $\Gamma_{op}$ is the exciton-optical phonon contribution.

In the case of GaSe samples where absorption spectra show the first three excitonic levels to be at low temperature, i.e. for crystals with few defects, $\Gamma_0$ is found to be 4 cm$^{-1}$.

The $\Gamma_{ac}$ contribution is obtained from Toyozawa’s calculation [21]. Providing that the deformation potential of the band gap is:

$$E_d = \frac{2(C_c - C_v)}{3}; \quad \text{then} \quad \Gamma_{ac} = \frac{2M^2E_d^2}{\pi U_L \hbar^3 \rho k_B T}$$

where $M$ is the exciton mass, $U_L$ is the longitudinal sound velocity and $\rho$ the sample density. In the case of GaSe: $U_L = 2.61 \times 10^5$ cm.s$^{-1}$ and $\rho = 5.135$ g.cm$^{-3}$. Taking now the upper values for $M$ ($M = M_{\perp} = 0.86$ m), for $E_d$ (1.3 eV) and for $T$ (500 K), the upper limit for $\Gamma_{ac}$ is 0.37 cm$^{-1}$ which is negligible.

The optical phonon which is taken into account in the $\Gamma_{op}$ computation is the homopolar phonon and is used in the computation of $\sigma_{\|}(T)$. This phonon plays the major role in GaSe [5]. Camassel [22] has also proposed the homopolar phonon (115 cm$^{-1} = 14$ meV) to be the main contribution to the exciton-phonon interaction in InSe.

To compute this interaction, one may write following Toyozawa [20]:

$$\frac{1}{\tau_h} = \frac{2}{\hbar} \sum_q |\beta_q|^2 [(\hbar n + 1) \delta(E_K - E_K - q - \hbar \Omega_h) + \hbar n \delta(E_K - E_K + q + \hbar \Omega_h)]$$  \hspace{1cm} (14)

where $q$ and $\hbar \Omega_h$ are respectively the wave vector and the longitudinal homopolar phonon energy.

The transition matrix element $\beta_q$ is given by [13]:

$$\beta_q = \left[ \frac{\hbar}{2 M_i N_i V \Omega_h} \right]^{1/2} \varepsilon_d.$$  \hspace{1cm} (15)
For transitions close to $K = 0$, one obtains (see Appendix):

$$\Gamma_h = \frac{M^{3/2} \varepsilon_d^2}{2^{1/2} M_1 N_1 \hbar (\hbar \Omega_h)^{1/2}} \bar{n}_h$$

(16)

where $\varepsilon_d$, $M_1 N_1$, $\hbar \Omega_h$ and $\bar{n}_h$ were determined in the calculation of $\Delta \sigma_G^{(2)}$. The temperature dependence is shown in figure 5 for two values of $M$:

$$M = M_\perp = m_{e_\perp}^* + m_{h_\perp}^* = 0.86 \, m \quad \text{(curve 1)}$$

$$M = 0.81 \, m \quad \text{(curve 2)}$$

which is the value which provides better agreement with experiment. The difference in these two values of $M$ is larger than the error on $M_\perp$ which can be deduced from different results on the effective masses [14, 15]. The exciton mass which contributes to the exciton-homopolar phonon interaction is thus not purely perpendicular but contains a parallel contribution.

---

**Fig. 5.** — Contribution of the homopolar phonon to the full width at half maximum of the first excitonic peak; bars: experimental results; dashed line: calculated curve with excitonic mass $M = M_\perp = 0.86 \, m$; solid line: calculated curve with $M = 0.88 \, M_\perp + 0.12 \, M_\parallel = 0.81 \, m$.

5. Conclusions.

We have investigated the temperature dependence of the direct absorption edge of GaSe. From comparison of the experimental spectra with Elliott's model, we can account for the
temperature variation of the full width at half maximum of the \( n = 1 \) exciton peak. This analysis confirms the strong interaction of the exciton with the 134.6 cm\(^{-1}\) homopolar phonon. The value of the deformation potential that we find (\( \varepsilon_d = 5.5 \text{ eV.Å}^{-1} \)) confirms the value calculated by Schlüter (\( \varepsilon_d = 5.8 \text{ eV.Å}^{-1} \)). To obtain the best fit between \( \Gamma_1 \) and the calculated curve, we had to fix the value of the kinetic mass of the exciton between the one of \( M_1 \) and the one of \( M_1 \).

**Acknowledgments.**

We appreciate stimulating and helpful discussions with J. M. Besson. We wish to thank A. Chevy for providing high-quality GaSe samples.

**Appendix.**

**Calculation of \( \Gamma_h \).**

\[
\frac{1}{\tau_h} = \frac{2 \pi}{\hbar} \sum_q |\beta_h|^2 \left\{ (\bar{n}_h + 1) \delta (E_K - E_{K - q} - \hbar \Omega_h) + \bar{n}_h \delta (E_K - E_{K + q} + \hbar \Omega_h) \right\}.
\]

**HYPOTHESES : CONDITIONS OF THE CALCULATION.** — \( \hbar \Omega_h \) and \( \bar{n}_h \) are independent of \( q \)

\[
E_K = \frac{\hbar^2 K^2}{2 M}; \quad E_{K \pm q} = \frac{\hbar^2}{2 M} (K^2 + q^2 \pm 2 Kq \cos \theta)
\]

\[
\beta_h = \left[ \frac{\hbar}{2 M N \Omega_h} \right]^{1/2}
\]

\[
\frac{1}{\tau_h} = \frac{M \varepsilon_d^2}{(2 \pi)^2 \hbar M \Omega_h} \times \left[ (\bar{n}_h + 1) \int_{q_m}^{q_M} \delta \left( q^2 - 2 Kq \cos \theta + 2 \frac{M \Omega_h}{\hbar} \right) q^2 \sin \theta \, d\theta \, d\varphi \, dq + \bar{n}_h \int_{q_m'}^{q_M'} \delta \left( q^2 + 2 Kq \cos \theta - 2 \frac{M \Omega_h}{\hbar} \right) q^2 \sin \theta \, d\theta \, d\varphi \, dq \right]^{1/2}
\]

\( q_M, q_m, q_M' \) and \( q_m' \) are the integration limits respectively for the emission and the absorption of the homopolar phonon (\( q, \hbar \Omega_h \)). The integration over \( \varphi \) gives \( 2 \pi \) and one of the properties of the \( \delta \) functions allows the integration over \( \theta \):

\[
\int_a^b f(x) \delta (g(x)) \, dx = \sum_{x_i} \frac{f(x_i)}{\left| \frac{dg}{dx} \right|_{x_i}} \quad \text{if} \quad g(x_i = 0) \quad \text{with} \quad a < x_i < b.
\]

Consequently if :

\[
f(\theta) = q^2 \sin \theta \quad \text{and} \quad g(\theta) = q^2 \mp 2 Kq \cos \theta \mp 2 \frac{M \Omega_h}{\hbar}
\]
then the result of the two integrations over $\theta$ is $1/2 \, Kq$. Finally:

$$
\frac{1}{\tau_h} = \frac{M \varepsilon_d^2}{2 M_i N_i \pi \hbar (h\Omega)} \left[ \frac{\bar{n}_h + 1}{2 K} \int_{q_m}^{q_M} dq + \frac{\bar{n}_h}{2 K} \int_{q_m}^{q_M} dq \right].
$$

Consequently:

$$
\frac{1}{\tau_h} = \frac{M \varepsilon_d^2}{8 \pi M_i N_i \pi \hbar (h\Omega)} \left[ \frac{\bar{n}_h + 1}{K} \left( q_M^2 - q_m^2 \right) + \frac{\bar{n}_h}{K} \left( q_M^2 - q_m^2 \right) \right].
$$

DETERMINATION OF THE INTEGRATION LIMITS.

1) Emission terms. — The solutions of the equation $q^2 - 2 \, Kq \cos \theta + \frac{2 \, M \Omega_h}{\hbar} = 0$ are:

$$
q = K \cos \theta \pm \left[ K^2 \cos^2 \theta - \frac{2 \, M \Omega_h}{\hbar} \right]^{1/2}
$$

with no solution for $K^2 < \frac{2 \, M \Omega_h}{\hbar}$. Otherwise:

$$
q_M = K + \left[ K^2 - \frac{2 \, M \Omega_h}{\hbar} \right]^{1/2}; \quad q_m = K - \left[ K^2 - \frac{2 \, M \Omega_h}{\hbar} \right]^{1/2}.
$$

2) Absorption term. — The solutions of the equation $q^2 + 2 \, Kq \cos \theta - \frac{2 \, M \Omega_h}{\hbar} = 0$ are:

$$
q = - K \cos \theta \pm \left[ K^2 \cos^2 \theta + \frac{2 \, M \Omega_h}{\hbar} \right]^{1/2}.
$$

Therefore:

$$
q_M = K + \left[ K^2 + \frac{2 \, M \Omega_h}{\hbar} \right]^{1/2}; \quad q_m = - K + \left[ K^2 + \frac{2 \, M \Omega_h}{\hbar} \right]^{1/2}.
$$

Around $K = 0$, only the absorption term is taken into account. One obtains:

$$
\Gamma_h = \frac{\hbar}{\tau_h} = \frac{M^{3/2} \varepsilon_d^2}{2^{1/2} \pi M_i N_i \pi \hbar (h\Omega_h)^{1/2} \bar{n}_h}.
$$

References