M P E J

Mathematical Physics Electronic Journal

ISSN 1086-6655
Volume 13, 2007

Paper 4
Received: May 4, 2006, Revised: Jan 16, 2007, Accepted: Jul 20, 2007
Editor: D. Bambusi

Normal forms for conical intersections in quantum chemistry.

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Abstract: *In quantum chemistry, the dynamics of heavy molecules is described by Schrödinger equations with energy level crossings. These crossings generate energy transfers at leading order between the modes as was earlier noticed by Landau and Zener in the 30's. The mathematical analysis of these transfers relies on the use of normal forms. Here, we analyze the implications of Y. Colin de Verdière's recent result on normal forms in the context derived by G. Hagedorn for molecular propagation and extend these normal forms to codimension 5 crossings.*

1 Introduction

The seek of normal forms for systems presenting eigenvalues crossings has known great improvements thanks to the two articles of Yves Colin de Verdière [2] and [3] which give a general classification of generic crossings between two eigenvalues of multiplicity 1. This problem of eigenvalues crossings appears in different areas of mathematics and physics; we focus here on what is known in quantum chemistry as *conical intersections*' question.

The dynamics of a molecule with N_n nuclei and N_e electrons is described by the time- dependent Schrödinger equation

$$i\varepsilon\partial_t\Phi^\varepsilon = H^\varepsilon_{mol}\Phi^\varepsilon, \;\; \Phi^\varepsilon_{t=0} = \Phi^\varepsilon_0, \tag{1}$$

with initial data $\Phi^\varepsilon_0 \in L^2(\mathbf{R}^{3(N_e+N_n)})$ and where H^ε_{mol} is an essentially self-adjoint molecular Hamiltonian. The parameter ε is the square-root of the ratio of the electronic mass on the average mass of the nuclei and is supposed to be small: one is concerned with heavy molecules. The molecular Hamiltonian H^ε_{mol} can be written as

$$H^\varepsilon_{mol} = -\frac{\varepsilon^2}{2}\Delta_q + H_e(q),$$

where $q \in \mathbf{R}^{3N_n}$ describes the nuclear configuration. The electronic Hamiltonian $H_e(q)$ takes into account the kinetics of the electrons and the interactions between electrons and nuclei; it is supposed to depend smoothly on q. We consider $\sigma(q)$ a closed subset of the spectrum of $H_e(q)$ which is the union of two eigenvalues of the same multiplicity k and which is isolated from the rest of the spectrum. Then, time-dependent Born-Oppenheimer theory (as carried out by H. Spohn and S. Teufel for example in [16]) gives the following: for initial data in the range of the spectral projector associated to $\sigma(q)$, one obtains an approximation of $\Phi^\varepsilon(t)$ by solving a $2k$ equations' system of the form

$$\left\{\begin{array}{l} i\varepsilon\partial_t\psi^\varepsilon(q,t) = \left(-\frac{\varepsilon^2}{2}\Delta_q + V(q)\right)\psi^\varepsilon(q,t), \\ \psi^\varepsilon(\cdot,0) = \psi_0^\varepsilon \in L^2(\mathbf{R}^d, \mathbf{C}^{2k}), \end{array}\right. \tag{2}$$

where $V(q)$ is a $2k \times 2k$ hermitian matrix. This type of analysis dates back to the '20s and is originally assigned to M. Born, V. Fock and R. Oppenheimer.
We focus here on the case where the two eigenvalues may not be separated because there exists some q_0 such that $\lambda_1(q_0) = \lambda_2(q_0)$. In [11] G. Hagedorn derives normal forms for matrix-valued potentials $V(q)$ in such a context. Let us shortly recall his argument.

One classically associates to H^ε_{mol} its symmetry group G and the subgroup H of unitary elements of G. Two cases occur, either $G = H$, either H is a subgroup of index 2 of G.

- When $G = H$, standard group representation theory applies and one associates to each of the eigenvalues λ_1 and λ_2 a unique representation of G. These representations are either unitarily equivalent or not, whence two different cases.
- If $H \neq G$, one uses the theory of corepresentations (see [18] or [14]). According to [14], there exist three types of corepresentations and one associates to λ_1 and to λ_2 corepresentations of one of these three types. If these corepresentations are of the same type, they are either unitarily equivalent or not: whence six different situations. If not, they are of different types, and we have to deal with three new situations. This gives nine different cases.

Finally, we are left with eleven different cases. For each of them, assuming that *the eigenvalues are of minimal multiplicity*, G. Hagedorn derives normal forms which are diagonal as soon as the representations or corepresentations associated with the eigenvalues are not unitarily equivalent, i.e. in seven of the derived cases. The four remaining normal forms obtained are not diagonal, the potential is of the form

$$V(q) = v(q)\mathrm{Id} + V_\ell\left(\phi(q)\right), \quad \ell \in \{2, 3, 3', 5\},$$

with $v \in \mathcal{C}^\infty(\mathbf{R}^d, \mathbf{R})$, ϕ smooth and vector-valued and where the matrix V_ℓ is defined by:

- **Codimension two crossing**: $V_2(\phi) = \begin{pmatrix} \phi_1 & \phi_2 \\ \phi_2 & -\phi_1 \end{pmatrix}$,
- **Codimension three crossing**: $V_3(\phi) = \begin{pmatrix} \phi_1 & \phi_2 + i\phi_3 \\ \phi_2 - i\phi_3 & -\phi_1 \end{pmatrix}$,

$$\text{or } \; V_{3'}(\phi) = \begin{pmatrix} \begin{pmatrix} \phi_1 & \phi_2 + i\phi_3 \\ \phi_2 - i\phi_3 & -\phi_1 \end{pmatrix} & \mathbf{0} \\ \mathbf{0} & \begin{pmatrix} \phi_1 & \phi_2 - i\phi_3 \\ \phi_2 + i\phi_3 & -\phi_1 \end{pmatrix} \end{pmatrix},$$

• **Codimension five crossing**:

$$V_5(\phi) = \begin{pmatrix} \phi_1 \mathbf{1} & \begin{pmatrix} \phi_2 + i\phi_3 & \phi_4 + i\phi_5 \\ -\phi_4 + i\phi_5 & \phi_2 - i\phi_3 \end{pmatrix} \\ \begin{pmatrix} \phi_2 - i\phi_3 & -\phi_4 - i\phi_5 \\ \phi_4 - i\phi_5 & \phi_2 + i\phi_3 \end{pmatrix} & -\phi_1 \mathbf{1} \end{pmatrix}.$$

For these four matrices, the eigenvalues are $\pm \mid \phi \mid$, therefore there are eigenvalues crossings for $V(q)$ if $\phi(q) = 0$. This crossing is said to be *generic* at the point (q^*, p^*) if it satisfies the assumptions

$$\mathrm{d}\phi(q^*) \ \textit{is of maximal rank}, \tag{3}$$
$$\mathrm{d}\phi(q^*)p^* \neq 0. \tag{4}$$

The assumption (3) describes the geometric structure of the crossing set $\{\phi(q) = 0\}$: it is a submanifold of codimension 2 for $\ell = 2$, of codimension 3 for $\ell = 3, 3'$ and of codimension 5 for $\ell = 5$. This explains why it is usual to refer to these crossings as to codimension 2, 3 and 5 crossings and enlightens the choice of the index ℓ we made. Assumption (4) implies that at such points (q^*, p^*) of the crossing set pass exactly two Hamiltonian trajectories: one for each eigenvalue of $\frac{|p|^2}{2} + V(q)$. Moreover, these curves are transverse to the crossing set (see [6] and the discussion in Section 2 below). Therefore, this assumption has a dynamical interpretation which clearly appears when applied to wave packets as in [11]: the wave packets arrive at the crossing point q^* with the speed p^* transversally to the crossing set. We set

$$N(2) = N(3) = 2, \ \ N(3') = N(5) = 4$$

so that the wave functions $\psi^\varepsilon(t)$ belong to $\mathbf{C}^{N(\ell)}$ and V to $\mathbf{C}^{N(\ell)\times N(\ell)}$.

We are interested in the eventual transitions between the two energy bands which may occur at a crossing point. Of course, the transition coefficient is a quantitative information of great interest. This transition coefficient has been computed on model systems since the '30s by Landau and Zener [13],[19]. For Schrödinger equation, the evolution of gaussian wave packets through crossings has been studied by G. Hagedorn [11]. In this paper G. Hagedorn uses the fact that the gaussian wave packet are microlocalized on classical trajectories and uses linearization along these trajectories to reduce to some model problem close to those studied by Landau and Zener. When dealing with less localized data, one crucially needs normal forms in some larger open set. In [4]–[6], where the propagation of Wigner measure of families of solution to (2) is studied, the authors use normal forms which are *2-microlocal normal forms* in the sense that they hold at a distance of order $\sqrt{\varepsilon}$ of the crossing but give enough quantitative information to go back to the initial model. More precisely, through change of coordinates and of unknown, one reduces to a system of the form

$$\frac{\varepsilon}{i}\partial_s u^\varepsilon = \begin{pmatrix} s\,\mathbf{1} & G \\ G^* & -s\,\mathbf{1} \end{pmatrix} u^\varepsilon, \tag{5}$$

where G is an operator which commutes with s and ∂_s. The main interest of the normal form approach is that the asymptotic behavior of such functions u^ε as ε goes to 0 is well understood (see Proposition 7 in [6] and Section 9 below).

Today, in the spirit of the earlier work of Braam and Duistermaat [1], the more elaborated normal forms available are those of Y. Colin de Verdière [2] and [3]. They are *microlocal normal forms* since they hold locally in the phase space in a neighborhood of a crossing point. The point here is that if one wants to translate the information obtained on the normal form in the original system of coordinates, one has to revisit the proofs of [2]-[3] in order to find quantitative information. This is one of our purpose here. The other issue of this paper is to extend these normal forms in order to cover all the cases described above. Indeed, if codimension 2 crossing enters in the symmetric case discussed in [2], codimension 3 in the corank-2 hyperbolic case studied in [3], codimension 5 is not discussed in Colin de Verdière's articles. Thus, we extend normal forms to codimension 5 case. In Section 3, we give applications of this result and we emphasize that it is used in [8] and [9] for proving the convergence of an algorithm describing the evolution of Wigner transform through conical intersections.

2 The normal form theorem

The phase space $\mathbf{R}^{2d+2}$ has the structure of the cotangent space $T^*\mathbf{R}^{d+1}$. We denote by $(p,\tau)\in\mathbf{R}^{d+1}$ the dual variables of $(q,t)\in\mathbf{R}^{d+1}$. The space $T^*\mathbf{R}^{d+1}$ is endowed with a symplectic structure given by the 2-form

$$\omega = \mathrm{d}\alpha = \mathrm{d}p\wedge\mathrm{d}q + \mathrm{d}\tau\wedge\mathrm{d}t$$

where α is the Liouville 1-form $\alpha = p\,\mathrm{d}q + \tau\mathrm{d}t$. If f is a smooth scalar function on $T^*\mathbf{R}^{d+1}$, we call *Hamiltonian vector field* associated with f the vector field H_f on $T\left(T^*\mathbf{R}^{d+1}\right)$ defined by: $\forall\rho=(q,t,p,\tau)\in T^*\mathbf{R}^{d+1}$, $H_f(\rho)$ is the vector of $T_\rho\left(T^*\mathbf{R}^{d+1}\right)$

$$H_f(\rho) = \nabla_p f(\rho)\cdot\nabla_q + \partial_\tau f(\rho)\,\partial_t - \nabla_q f(\rho)\cdot\nabla_p - \partial_t f(\rho)\,\partial_\tau.$$

It is well known that the *characteristic set* and the *classical trajectories* of the equation are of great importance for describing the evolution of the solution ψ^ε. The characteristic set Σ is the subset of the phase space $\mathbf{R}^{2d+2}$

$$\Sigma := \left\{\left(\tau + v(q) + \frac{|p|^2}{2}\right)^2 - |\phi(q)|^2 = 0\right\}.$$

It is above these points that the energy may concentrate. The *crossing set* is then defined as the subset of the characteristic set where eigenvalues cross

$$S := \left\{\tau + v(q) + \frac{|p|^2}{2} = 0,\ \ \phi(q) = 0\right\}.$$

By (3), this set is a submanifold of codimension $\ell+1$ of the phase space $T^*\mathbf{R}^{d+1}$ and by (4), the symplectic form $\omega_{|S}$ is of corank $\ell-1$.
The classical trajectories are the Hamiltonian trajectories $(\rho^\pm)$ associated with the eigenvalues

$$\lambda^\pm(q,p,\tau) = \tau + v(q) + \frac{|p|^2}{2} \pm |\phi(q)|.$$

They satisfy the system

$$\left\{\begin{array}{l}\dot\rho^\pm_s = H_{\lambda^\pm}(\rho^\pm_s),\\ \rho^\pm_{s\ |s=0} = \rho_0.\end{array}\right. \tag{6}$$

Outside the crossing set, $\lambda^\pm$ are smooth functions and the trajectories are smooth and well-defined. If one considers now $\rho_0 \in S$ where (3) and (4) are satisfied, one can prove that for each mode $\pm$, the vectors $H_{\lambda^\pm}(\rho_s^\pm)$ have limits above S as s go to 0^+ or 0^- which are transverse to S. More precisely, there exist on $T_{\rho_0}S$ two vectors $H(\rho_0)$ and $H'(\rho_0)$ such that $\omega(H(\rho_0), H'(\rho_0)) \neq 0$ and

$$H(\rho_0) = \lim_{s\to 0^-} H_{\lambda^+}(\rho_s^+) = \lim_{s\to 0^+} H_{\lambda^-}(\rho_s^-), \quad H'(\rho_0) = \lim_{s\to 0^+} H_{\lambda^+}(\rho_s^+) = \lim_{s\to 0^-} H_{\lambda^-}(\rho_s^-). \tag{7}$$

Heuristically, this explains the following fact: for each mode, a unique curve $\rho^\pm$ passes in ρ_0 and the ingoing curves for the mode $\pm$ smoothly continue in the outgoing ones for the other mode $\mp$ (see [5] and [4] for a rigorous proof). We choose a neighborhood Ω of ρ_0 where (3) and (4) are satisfied and, in Ω, we denote by $J^{\pm,in}$ the set consisting in all the classical trajectories for the mode $\pm$ arriving to a point of $S\cap\Omega$ and by $J^{\pm,out}$ the set of all the trajectories arising from a point of $S\cap\Omega$. Because of the continuation properties of the curves $(\rho_s^\pm)$, the sets

$$J = J^{+,in} \cup J^{-,out}, \quad J' = J^{-,in} \cup J^{+,out}$$

are smooth codimension ℓ submanifolds of $T^*\mathbf{R}^{d+1}$.

In the following, for $a \in \mathcal{C}_0^\infty(\mathbf{R}^{2d+2})$ we denote by $\mathrm{op}_\varepsilon(a)$ the semi-classical pseudo-differential operator of symbol a defined on $L^2(\mathbf{R}^{d+1})$ with Weyl quantization by

$$\mathrm{op}_\varepsilon(a) f(q,t) = \int_{\mathbf{R}^{d+1}} a\left(\frac{q+q'}{2}, \frac{t+t'}{2}, \varepsilon p, \varepsilon\tau\right) \mathrm{e}^{ip\cdot(q-q')+i\tau\cdot(t-t')} f(q',t') \frac{\mathrm{d}q'\mathrm{d}p\,\mathrm{d}t'\mathrm{d}\tau}{(2\pi)^{d+1}}, \quad f\in L^2(\mathbf{R}^d).$$

The function f is said to satisfy the property (P) *microlocally* in $\Omega \subset T^*\mathbf{R}^{d+1}$ if for all $a \in \mathcal{C}_0^\infty(\Omega)$, $\mathrm{op}_\varepsilon(a) f$ satisfies (P).

In the following, we shall use *canonical transforms* and *Fourier integral operators* on which we shortly recall some basic facts. A canonical transform is a local change of symplectic coordinates, i.e. a local diffeomorphism which preserves the symplectic structure. One can associate with a canonical transform a unitary bounded operator of $L^2(\mathbf{R}^{d+1})$ compatible with the change of coordinates. Let us explain now this point. Once given the canonical transform κ in some open set Ω, one builds a path $\mathcal{C}^1$, $\delta \mapsto \kappa(\delta)$, $\delta\in[0,1]$ linking Id to κ. The fact that $\kappa(\delta)$ preserves the symplectic structure of $T^*\mathbf{R}^{d+1}$ yields that $\frac{\mathrm{d}}{\mathrm{d}\delta}\kappa(\delta)\circ\kappa(\delta)^{-1}$ is a Hamiltonian vector field on $T(T\mathbf{R}^{d+1})$ above Ω. Therefore, there exists a smooth function U such that $\kappa(\delta)$ solves

$$\frac{\mathrm{d}}{\mathrm{d}\delta}\kappa(\delta) = H_U \kappa(\delta), \quad \kappa(0) = \mathrm{Id}, \quad \kappa(1) = \kappa.$$

Observe that finding U or finding κ are equivalent questions; we will use this fact in the proof of normal forms (see Section 4).
For $f\in L^2(\mathbf{R}^{d+1})$, define $f^\varepsilon(\delta)$ by

$$\frac{\varepsilon}{i}\partial_\delta f^\varepsilon(\delta) = \mathrm{op}_\varepsilon(U) f^\varepsilon(\delta), \quad f^\varepsilon(0) = f, \quad \delta\in[0,1].$$

Then, the operator $Kf := f^\varepsilon(1)$ satisfies the following formula known as Egorov's Theorem

$$\forall a \in \mathcal{C}_0^\infty(\mathbf{R}^{2d+2}), \quad K^*\mathrm{op}_\varepsilon(a)\, K = \mathrm{op}_\varepsilon(a\circ\kappa) + O(\varepsilon^2) \text{ in } \mathcal{L}(L^2(\mathbf{R}^{d+1})). \tag{8}$$

The operator K is called *Fourier Integral Operator* associated with κ. The reader will find in [15] a complete analysis of Fourier Integral Operator, the presentation chosen here is the one of [5]. Observe that solutions of (2) are in $L^2_{loc}(\mathbf{R}, L^2(\mathbf{R}^d))$ so that (8) applies to such functions since a is compactly supported. We emphasize that this construction is local and we finish these preliminaries by some notations: we denote by $O_S(N)$ a function which vanishes up to the order $N-1$ on S and if $\ell \in \{2,3,3',5\}$ and if $\tilde z = (z_1,\cdots,z_{\ell-1}) \in \mathbf{R}^{\ell-1}$ we denote by $V_\ell(s,z')$ the matrix

$$V_\ell(s,\tilde z) = V_\ell(s,z_1,\cdots,z_{\ell-1}).$$

with the convention $\mathbf{R}^{3'-1} = \mathbf{R}^2$.

Theorem 1 *Consider $\rho_0 = (q_0,t_0,p_0,\tau_0) \in S$ such that (3) and (4) hold in a neighborhood Ω of ρ_0. Then, there exists a local canonical transform κ from a neighborhood of ρ_0 into some neighborhood $\tilde\Omega$ of 0,*

$$\kappa : (q,t,p,\tau) \mapsto (s,z,\sigma,\zeta), \quad \kappa(\rho_0) = 0.$$

There exist a Fourier integral operator K associated with κ and an invertible matrix-valued symbol $A_\varepsilon = A_0 + \varepsilon A_1 + \varepsilon^2 A_2 + \dots$ such that if ψ^ε is a family of solutions to (2) for some initial data ψ^ε_0 uniformly bounded in $L^2(\mathbf{R}^d)$, then

$$v^\varepsilon = K^* \mathrm{op}_\varepsilon\left((A_\varepsilon)^{-1}\right)\psi^\varepsilon$$

satisfies for all $\phi \in \mathcal{C}_0^\infty(\tilde\Omega)$,

$$\mathrm{op}_\varepsilon(\phi)\, \mathrm{op}_\varepsilon\Big(-\sigma + V_\ell\big(s, \tilde z + \gamma_\varepsilon(z,\zeta)\big)\Big)\, v^\varepsilon = O(\varepsilon^\infty) \tag{9}$$

in $L^2(\mathbf{R}^{d+1})$ where $\gamma_\varepsilon \in \mathcal{C}_0^\infty\left(\tilde\Omega, \mathbf{R}^{\ell-1}\right)$, $\gamma_\varepsilon = \gamma_0 + \varepsilon\gamma_1 + \varepsilon^2\gamma_2 + \cdots$ with

$$\text{for } \ell = 2,\ \ \gamma_0 = 0 \ \text{ and } \text{ for } \ \ell \geq 2,\ \ \gamma_\varepsilon = O(|\tilde z|^2).$$

Moreover,

$$J^{\pm,in} = \{\sigma \mp s = 0,\ \ \tilde z = 0,\ \ s \leq 0\}, \tag{10}$$

$$J^{\pm,out} = \{\sigma \pm s = 0,\ \ \tilde z = 0,\ \ s \geq 0\}. \tag{11}$$

Let $\pi_\ell(q,p)$ be the orthogonal projection on the hyperplane $T(q,p)$ of $\mathbf{R}^\ell$ orthogonal to $\mathrm{d}\phi(q)p$, then if $\kappa(q,t,p,\tau) = (s,z,\sigma,\zeta)$, $\tilde z$ are the coordinates of $|\mathrm{d}\phi(q)p|^{-1/2}\pi_\ell(q,p)(\phi(q))$ in an orthonormal basis $\mathcal{B}$ of $T(q,p)$ up to $O_S(2)$ so that

$$|\tilde z|^2 = |\mathrm{d}\phi(q)p|^{-1}|\pi_\ell(q,p)(\phi(q))|^2. \tag{12}$$

Besides

$$\sigma = -|\mathrm{d}\phi(q)p|^{-1/2}\left(\tau + \tfrac{|p|^2}{2} + v(q)\right) + O_S(2), \tag{13}$$

$$s = -|\mathrm{d}\phi(q)p|^{-1/2}\,\tfrac{\mathrm{d}\phi(q)p}{|\mathrm{d}\phi(q)p|}\cdot\phi(q) + O_S(2). \tag{14}$$

Finally, for $\ell = 5$, we have the following matrix-valued relation

$$\exists\alpha \in \{+1,-1\},\ \ \forall y \in \mathbf{R}^5,\ \ (A_0^*)_{|S} V_\ell(y)\,(A_0)_{|S} = \alpha V_5\left(-\frac{\mathrm{d}\phi(q)p}{|\mathrm{d}\phi(q)p|}\cdot y, \tilde z(y)\right) + O_S(1), \tag{15}$$

where $\tilde z(y)$ are the coordinates of $\pi_5(q,p)(y)$ in $\mathcal{B}$.

Remark 1 1- *We emphasize that these normal forms are microlocal, they simply hold microlocally in Ω which is an open subset of the phase space.*
2- *The matrix-valued relation (15) is important for systems presenting eigenvalues of multiplicity* 2. *Indeed, this equation allows to describe the change of polarization which occurs inside one of the modes when passing through a crossing point as in [4] where this relation is replaced by equation (9) of [4] in the 2-microlocal normal form.*

This Theorem is directly inspired by Colin de Verdière ideas in [2] and [3]. His articles are devoted to general systems presenting codimension 2 and 3 crossings between eigenvalues of multiplicity 1. Thus, our evolution equation appears as a special case. If we set

$$P = \tau + v(q) + \frac{|p|^2}{2} + V_\ell\left(\phi(q)\right), \quad P_0 = -\sigma + V_\ell(s, \tilde{z}), \quad \tilde{z} \in \mathbf{R}^{\ell-1}$$

and apply Theorem 3 of [2] in the case of codimension 2 crossing and Theorem 4 of [3] for codimension 3 crossings, we obtain the following:

Proposition 1 *For $\ell = 2, 3$ and assuming (3) and (4) in some point (q^*, p^*) of the singular set S, there exists a Fourier integral operator K, a symbol of order* 0 *denoted by $A_\varepsilon : T^*\mathbf{R}^{d+1} \to Gl(2, \mathbf{C})$ (a gauge transform) and a vector-valued symbol denoted*

$$\gamma_\varepsilon \sim \sum_{j=0}^{\infty} \gamma_j(z, \zeta)\, \varepsilon^j$$

with $\gamma_\varepsilon = O(|\tilde{z}|^2)$ and such that

$$K^* \mathrm{op}_\varepsilon(A_\varepsilon^*) \mathrm{op}_\varepsilon(P) \mathrm{op}_\varepsilon(A_\varepsilon) K = \mathrm{op}_\varepsilon(P_0) + \mathrm{op}_\varepsilon\left(V_\ell(0, \gamma_\varepsilon)\right) + O(\varepsilon^\infty). \tag{16}$$

This gives (9) for $\ell = 2, 3$. When $\ell = 3'$ (2) is a system of two decoupled systems corresponding to the case $\ell = 3$. Applying Proposition 1 for $\ell = 3$, one gets (9) for $\ell = 3'$. We are going to prove that this is also true for $\ell = 5$ up to some sign in front of P_0 (see Proposition 6).

It remains also to prove (10)-(15): the statement of Colin de Verdière's Theorems do not give this quantitative interpretation of the new symplectic coordinates (s, z, σ, ζ). To get these formula, one need to follow his proofs and check carefully the change of variables and the transformations of the Hamiltonian.

In the next sections, we first give an example of use of Theorem 1 (Section 3). Then, we develop the proof of Theorem 1 in the case $\ell = 5$ by following Colin de Verdière's method. This will also explain how to get equations (10)-(15) in the cases $\ell = 2, 3, 3'$.

3 Application

For simplicity, we suppose here $\ell \in \{2, 3\}$, $\phi(q) = q$ and $v(q) = 0$. We denote by $\Pi^\pm(q)$ the eigenprojectors associated with the eigenvalue $\pm|q|$ of $V_\ell(q)$ and we suppose that the projections of the initial data on each modes, $\phi^\varepsilon_{0,+} = \Pi^+(q)\psi^\varepsilon_0$ and $\phi^\varepsilon_{0,-} = \Pi^-(q)\psi^\varepsilon_0$ concentrate respectively on (q_0^+, p_0^+) and (q_0^-, p_0^-) with $q_0^\pm \neq 0$.

The Hamiltonian trajectories arising from these points of the phase space have been precisely calculated in [7]. It is proved in Proposition 1 of [7] that under the assumptions

$$q_0^\pm \wedge p_0^\pm = 0,$$
$$|p_0^-|^2 > 2|q_0^-|, \ \ p_0^- \cdot q_0^- < 0,$$
$$p_0^+ \cdot \frac{q_0^+}{|q_0^+|} + \sqrt{|p_0^+|^2 + 2|q_0^+|} = -p_0^- \cdot \frac{q_0^-}{|q_0^-|} - \sqrt{|p_0^-|^2 - 2|q_0^-|} := t^*,$$

the curves $(q^\pm(t), p^\pm(t))$ reach S at the same time t^* with a non-zero speed p^* and one can choose $(q_0^\pm, p_0^\pm)$ so that they reach S at the same point $(0, p^*)$. Indeed, one has

$$q^+(t) = -\frac{t^2}{2}\frac{q_0^+}{|q_0^+|} + tp_0^+ + q_0^+, \ \ p^+(t) = -t\frac{q_0^+}{|q_0^+|} + p_0^+,$$
$$q^-(t) = \frac{t^2}{2}\frac{q_0^-}{|q_0^-|} + tp_0^- + q_0^-, \ \ p^-(t) = t\frac{q_0^-}{|q_0^-|} + p_0^-.$$

Therefore, if $r^* > t^*/2$ and $\omega \in \mathbf{S}^{d-1}$ the data

$$q_0^+ = t^*(r^* - t^*/2)\,\omega, \ \ p_0^+ = (-r^* + t^*)\,\omega, \ \ q_0^- = t^*(r^* + t^*/2)\,\omega, \ \ p_0^- = (-r^* - t^*)\,\omega$$

generate such trajectories. Besides, observe that there exists $t_1 > t^*$ such that during (t^*, t_1) both trajectories do not meet S again.
We are concerned on describing the weak limits of the position/impulsion probability densities

$$n^\varepsilon_{pos}(q,t) := |\psi^\varepsilon(q,t)|^2 \mathrm{d}q \ \text{ and } \ n_{imp}(p,t) := (2\pi)^{-d}|\widehat{\psi}^\varepsilon(p,t)|^2 \mathrm{d}p$$

for $t \in (t^*, t_1)$.
For the sake of concreteness, we choose

$$\psi_0^\varepsilon = \varepsilon^{-\beta d/2}\Phi\left(\frac{q-q_0^+}{\varepsilon^\beta}\right)\exp\left(\frac{i}{2\varepsilon}(p_0^+\cdot\frac{q_0^+}{|q_0^+|})|q-\varepsilon^{\alpha^+}x_0^+|^2\right)E^+(q)$$
$$+\varepsilon^{-\beta d/2}\Psi\left(\frac{q-q_0^-}{\varepsilon^\beta}\right)\exp\left(\frac{i}{2\varepsilon}(p_0^-\cdot\frac{q_0^-}{|q_0^-|})|q-\varepsilon^{\alpha^-}x_0^-|^2\right)E^-(q)$$

where

$$0 < \alpha^\pm \leq 1/2, \ \ 0 < \beta < 1/2, \ \ x_0^+, x_0^- \in \mathbf{R}^d,$$

and Φ and Ψ are smooth compactly supported functions of $\mathbf{R}^d$, E^+ (resp. E^-) is a smooth bounded function such that $||E^\pm||_{\mathbf{C}^2} = 1$ and that on the support of $\Phi\left(\frac{q-q_0^+}{\varepsilon^\beta}\right)$ (resp. of $\Psi\left(\frac{q-q_0^-}{\varepsilon^\beta}\right)$) we have $\Pi^+E^+ = E^+$ (resp. $\Pi^-E^- = E^-$). We shall focus on both situations $x_0^+ \neq x_0^-$ and $x_0^+ = x_0^-$. We set

$$r_0^+ = p_0^+ \cdot \frac{q_0^+}{|q_0^+|}, \ \ r_0^- = p_0^- \cdot \frac{q_0^-}{|q_0^-|}, \ \ c^{+,in} = \|\Phi\|_{L^2}, \ \ c^{-,in} = \|\Psi\|_{L^2}$$

and for simplicity, we suppose

$$\eta_0^\pm := -r_0^\pm(q_0^\pm \wedge x_0^\pm) \neq 0$$

where

- if $\ell = 2$, $\forall (q,p) \in \mathbf{R}^2 \times \mathbf{R}^2$, $q \wedge p = q_1 p_2 - q_2 p_1$.
- if $\ell = 3$, $\forall (q,p) \in \mathbf{R}^3 \times \mathbf{R}^3$, $q \wedge p = (q_2 p_3 - p_3 q_2, q_3 p_1 - p_3 q_1, q_1 p_2 - q_2 p_1)$.

The result below applies to a larger class of initial data (see Remark 3 in Section 9).

Proposition 2 *The position/impulsion densities tends to $n_{pos}(q,t)$ and $n_{imp}(p,t)$ respectively. For $t \in [0, t^*)$,*

$$\begin{aligned} n_{pos}(q,t) &= c^{+,in}\delta(q - q^+(t)) + c^{-,in}\delta(q - q^-(t)), \\ n_{imp}(p,t) &= c^{+,in}\delta(p - p^+(t)) + c^{-,in}\delta(p - p^-(t)). \end{aligned}$$

For $t \in (t^*, t_1)$,

$$\begin{aligned} n_{pos}(q,t) &= c^{+,out}\delta(q - q^+(t)) + c^{-,out}\delta(q - q^-(t)), \\ n_{imp}(p,t) &= c^{+,out}\delta(p - p^+(t)) + c^{-,out}\delta(p - p^-(t)), \end{aligned}$$

where the coefficients $c^{+,out}$ and $c^{-,out}$ are given by

	$c^{+,out}$	$c^{-,out}$
$\alpha^-, \alpha^+ < 1/2$	$c^{+,in}$	$c^{-,in}$
$\alpha^+ < \alpha^- = 1/2$	$c^{+,in} + T(\eta_0^-)c^{-,in}$	$c^{-,in}\left(1 - T(\eta_0^-)\right)$
$\alpha^- < \alpha^+ = 1/2$	$c^{+,in}\left(1 - T(\eta_0^+)\right)$	$c^{-,in} + T(\eta_0^+)c^{+,in}$
$\begin{cases} \alpha^+ = \alpha^- = 1/2 \\ \eta_0^+ \neq \eta_0^- \end{cases}$	$c^{+,in}\left(1 - T(\eta_0^+)\right) + T(\eta_0^-)c^{-,in}$	$c^{-,in}\left(1 - T(\eta_0^-)\right) + T(\eta_0^+)c^{+,in}$

with

$$T(\eta) = \mathrm{e}^{-\pi \frac{|\eta|^2}{|p^*|}}.$$

If $\alpha^+ = \alpha^- = 1/2$ and $\eta_0^+ = \eta_0^- := \eta_0$, there exists $\gamma_0 \in \mathbf{R}^+$ and $\lambda_0 \in \mathbf{R}$ such that

$$\begin{cases} c^{+,out} = c^{+,in}\left(1 - T(\eta_0)\right) + T(\eta_0)c^{-,in} + \gamma_0 \cos(\lambda_0), \\ c^{-,out} = c^{-,in}\left(1 - T(\eta_0)\right) + T(\eta_0)c^{+,in} - \gamma_0 \cos(\lambda_0). \end{cases}$$

Besides, for any $\lambda \in \mathbf{R}$, if one turns Ψ into $\mathrm{e}^{i\lambda}\Psi$, then one has

$$\begin{cases} c^{+,out} = c^{+,in}\left(1 - T(\eta_0)\right) + T(\eta_0)c^{-,in} + \gamma_0 \cos(\lambda_0 - \lambda), \\ c^{-,out} = c^{-,in}\left(1 - T(\eta_0)\right) + T(\eta_0)c^{+,in} - \gamma_0 \cos(\lambda_0 - \lambda). \end{cases} \tag{17}$$

The transition coefficient $T(\eta)$ is the Landau-Zener coefficient between the modes which has been already described in numerous articles. The term $\gamma_0 \cos(\lambda_0 - \lambda)$ illustrates a more intricate coupling between modes which cannot be described by quadratic quantities. We emphasize the fact that the multiplication by an appropriate phase factor of one of the component of the initial data is enough to annihilate this coupling (see (17)). The proof of this Proposition is sketched in Section 9.

4 Steps of the proof of Theorem 1

We follow here Colin de Verdière's strategy that we apply to codimension 5 case with a special care to keep the information about the change of variables in order to prove (10)-(15). We choose $\ell = 5$ so that we have now

$$P = \tau + v(q) + \frac{|p|^2}{2} + V_5\left(\phi(q)\right), \quad P_0 = -\sigma + V_5(s, \tilde{z}), \quad \tilde{z} = (z_1, z_2, z_3, z_4) \in \mathbf{R}^4.$$

We consider *the dispersion relations*

$$g = \left(\tau + v(q) + \frac{|p|^2}{2}\right)^2 - |\phi(q)|^2, \quad g_0 = \sigma^2 - s^2 - |\tilde{z}|^2.$$

We have $\det(P) = g^2$, $\det(P_0) = g_0^2$. The proof consists in four steps.

1st. Step: This first step relies on the analysis of the geometry of the crossing set. One crucially uses that the vector fields H and H' are transverse to S. We build the germ of the canonical transform κ: the canonical transform will not be modified above S in the following steps of the proof. We will also prove here the equations (10)-(14). This analysis is purely scalar, in the sense that one only works with the dispersion relations; one proves the following:

Proposition 3 *Near ρ_0, there exist a canonical transform $\kappa_0 : (t, q, \tau, p) \mapsto (s, z, \sigma, \zeta)$ and a non-zero function e_0 such that $g \circ \kappa_0 = e_0 g_0 + O_S(3)$ and such that (12)-(14) hold for κ_0.*

Remark 2 *Observe that equations (13)-(14) yield that there exist positive constants k and k' such that*

$$H = k\partial_{\sigma+s}, \quad H' = k'\partial_{s-\sigma}.$$

2nd. Step: The second step is more analytic: a Birkhoff normal form allows to ameliorate step by step the remainder term of Proposition 3. This is done by solving homological equations. Then, a formal normal form is obtained for the dispersion relation.

Proposition 4 *Near ρ_0, for any $N \in \mathbf{N}$, $N \geq 4$, there exist*

- *a local canonical transform $\kappa_1 : (q, t, p, \tau) \mapsto (s, z, \sigma, \zeta)$,*
- *a smooth function $\gamma = \gamma(z, \zeta)$ which is polynomial of degree N in $\tilde{z}$ with coefficients in $\mathcal{C}^\infty(S)$ and such that $\gamma = O(|\tilde{z}|^2)$ as $\tilde{z}$ goes to 0,*
- *a smooth function $e_1 \neq 0$*

such that (12)-(14) hold for κ_1 and

$$g \circ \kappa_1 = e_1\left(\sigma^2 - s^2 - |\tilde{z} + \gamma(z, \zeta)|^2\right) + O_S(N). \tag{18}$$

3rd. Step: At this stage of the proof, one begins to work with the matrix structure of P: one finds the gauge transform (the classical symbol A_0 of A in (9)) and one proves (15). We denote by $\mathbf{H}$ the field of quaternions and by $h(\tilde{z})$ its elements,

$$h(\tilde{z}) = \begin{pmatrix} z_1 + iz_2 & z_3 + iz_4 \\ -z_3 + iz_4 & z_1 - iz_2 \end{pmatrix}, \quad \tilde{z} = (z_1, z_2, z_3, z_4) \in \mathbf{R}^4.$$

We will use the following properties of the quaternions which come from straightforward computations:

(i) $h(\tilde{z})h(\tilde{z})^* = |\tilde{z}|^2 \mathrm{Id}, \quad h(\tilde{z}) + h(\tilde{z})^* = 2z_1 \mathrm{Id}$.

(ii) $h(\tilde{z})h(\tilde{y}) = h(z_1 y_1 - z' \cdot y', z_1 y' + y_1 z' - z' \wedge y')$ with $z' = (z_2, z_3, z_4)$, $y' = (y_2, y_3, y_4)$.

(iii) $h(\tilde{z})h(\tilde{y})^* + h(\tilde{y})h(\tilde{z})^* = 2\,\tilde{y} \cdot \tilde{z}$.

We shall consider matrices of $\mathbf{H}^{2,2}$ which are 2×2 matrices of quaternions and that we identify to elements of $\mathbf{C}^{4,4}$. We focus on the subspace of the matrices of $\mathbf{H}^{2,2}$ which are Hermitian elements of $\mathbf{C}^{4,4}$; they are of the form

$$M = \begin{pmatrix} a\,\mathrm{Id} & h(f) \\ h(f)^* & b\,\mathrm{Id} \end{pmatrix}. \tag{19}$$

Besides, we observe that

(iv) if $A \in \mathbf{H}^{2,2}$ and if M is of the form (19), then the matrix $A^*M + MA$ also is of the form (19).

We prove the formal normal form for the matrix-valued Hamiltonian.

Proposition 5 *There exists a matrix $A_0 \in \mathbf{H}^{2,2}$, a canonical transform κ_2 and a smooth function $\gamma_0(z,\zeta)$ such that*

$$(A_0^* P A_0) \circ \kappa_2 = \pm\Big(-\sigma + V_5\big(s, \tilde{z} + \gamma_0(z,\zeta)\big)\Big). \tag{20}$$

Moreover (12)-(15) hold for κ_2 and $\gamma_0(z,\zeta) = O(|\tilde{z}|^2)$.

Observe that the $\pm$ sign in front of the symbol above is not a problem since we deal with the equation $\mathrm{op}_\varepsilon(P)\psi^\varepsilon = 0$. Of course, it would always be possible to modify s and σ so that this sign is $+$ but this would modify (12)-(14).

4th. Step: Finally, one has to quantify (20) and to ameliorate the rests in order to pass from a classical formal normal form to a semi-classical one. One proves the semi-classical normal form.

Proposition 6 *For any $j \in \mathbf{N}^*$, there exists a Fourier integral operator K_j and some matrices $A_0, \cdots A_{j-1} \in \mathbf{H}^{2,2}$ such that in $\mathcal{L}(L^2(\mathbf{R}^d))$*

$$(K_j^\varepsilon)^* \,\mathrm{op}_\varepsilon({A_\varepsilon^{(j)}}^*)\, \mathrm{op}_\varepsilon\,(P)\; \mathrm{op}_\varepsilon(A_\varepsilon^{(j)})\, K_j^\varepsilon = \mathrm{op}_\varepsilon(\pm P_0) + \mathrm{op}_\varepsilon(\Gamma_\varepsilon^{(j)}) + \varepsilon^j \mathrm{op}_\varepsilon(T_j) + O(\varepsilon^{j+1}), \tag{21}$$

where T_j is of the form (19) and where

$$A_\varepsilon^{(j)} = A_0 + \varepsilon A_1 + \cdots + \varepsilon^{j-1} A_{j-1},$$
$$\Gamma_\varepsilon^{(j)} = \Gamma_0 + \varepsilon \Gamma_1 + \cdots + \varepsilon^{j-1}\Gamma_{j-1}, \quad \Gamma_k = \begin{pmatrix} 0 & h(\gamma_k(z,\zeta)) \\ h(\gamma_k(z,\zeta))^* & 0 \end{pmatrix}, \quad 1 \le k \le j-1.$$

Besides (10)–(15) hold.

This proposition comes from the resolution of matrix-valued homological equations defining matrices $\Gamma_\varepsilon^{(j)}$ and T_j. At this stage of the proof, one crucially uses property *(iv)*. Then (21) and the relations on H and H' of Remark 2 yield the equations of J and J' (10) and (11).

5 The germ of the canonical transform

Proof of Proposition 3: We denote by ϕ_0 the function

$$\phi_0 = \tau + v(q) + \frac{|p|^2}{2}.$$

Consider $\rho_0 \in S$ where (3) and (4) are satisfied in a neighborhood U of ρ_0. In the following, we have $\rho = (q,t,p,\tau)$ which varies in $S \cap U$. Let us denote by $TS_{|\rho}^{\perp}$ the orthogonal of $TS_{|\rho}$ for the symplectic form ω. We first build a symplectic basis of $T_\rho S^{\perp}$, whence local coordinates on S that we extend to coordinates (s,z,σ,ζ) in U such that

$$S = \{g_0 = 0\} = \{s = 0,\ \sigma = 0,\ \tilde{\zeta} = 0\}.$$

If we find such coordinates, then $TS_{|\rho}^{\perp}$ will be generated by ∂_σ, ∂_s and ∂_{ζ_j}, $1 \leq j \leq 4$ and $TS_\rho \cap TS_{|\rho}^{\perp}$ by ∂_{ζ_j}, $1 \leq j \leq 4$. We observe that

$$TS_{|\rho}^{\perp} = \mathrm{Vect}\,(H_{\phi_i}(\rho),\ \ 0 \leq i \leq 5)\,;$$

thus, we will try to find s, σ, z, ζ such that the vectors ∂_σ, ∂_s and ∂_{ζ_j} are linear combinations of the $H_{\phi_i}(\rho)$ on $T_\rho(T\mathbf{R}^{d+1})$. We also want that the vectors $H(\rho)$ and $H'(\rho)$ of (7) which are tangent to J and J' respectively in ρ are mapped on $H_{\sigma-s}$ and $H_{\sigma+s}$ (see Remark 2). Therefore, it will be convenient to set

$$-\sigma + s = \sqrt{2}\,\eta, \quad -\sigma - s = \sqrt{2}\,y \tag{22}$$

and we will manage so that $H(\rho) = -k\partial_y$ and $H'(\rho) = k'\partial_\eta$ for some $k, k' > 0$.
Throughout the paper, s, σ, η and y will be linked by (22).

Of course, this is not enough and we also ask that in the new coordinates the function g becomes $e_0 g_0 = e_0(2y\eta - |\tilde{z}|^2)$ for some non zero smooth function e_0. For $\rho \in S$, we consider the linear map $M(\rho)$ on $T_\rho(T^*\mathbf{R}^{d+1})$ associated to the differential of the Hamiltonian vector field $H_g(\rho)$. We observe that if we already have $S = \{\sigma = s = 0,\ \tilde{z} = 0\}$, we only need to prove that in the new basis of the tangent $(\partial_y, \partial_z, \partial_\eta, \partial_\zeta)$ the matrix of $M(\rho)$ is of the form $2e_0M_0$ where $M_0\partial_y = \partial_y$, $M_0\partial_\eta = -\partial_\eta$, $M_0\partial_{z_k} = \partial_{\zeta_k}$ for $1 \leq k \leq 4$, $M_0\partial_{z_j} = 0$ for $j \notin \{1, \cdots, 4\}$ and $M_0\partial_{\zeta_i} = 0$ for all i. For this reason, we will analyze the linear map $M(\rho)$, observing that in the coordinates (q,t,p,τ), we have

$$M(\rho)\delta\rho = 2(\mathrm{d}\phi_0(\rho)\delta\rho)H_{\phi_0}(\rho) - 2\sum_{j=1}^{5}(\mathrm{d}\phi_j(\rho)\delta\rho)H_{\phi_j}(\rho).$$

An explicit basis of $TS_{|\rho}^{\perp}$: We set

$$\Omega(q,p) = \frac{\mathrm{d}\phi(q)p}{|\mathrm{d}\phi(q)p|} \in \mathbf{R}^5, \quad r = |\mathrm{d}\phi(q)p|.$$

Then, writing $H_{\lambda^\pm} = H_{\phi_0} \pm \frac{\phi(q)}{|\phi(q)|} \cdot H_\phi$, we get that if $\rho_s^\pm$ solves (6) with $\rho_0^\pm = \rho$,

$$H_{\lambda^\pm}(\rho_s^\pm) \underset{s\to 0^+}{\longrightarrow} H_{\phi_0}(\rho) \pm \Omega(q,p) \cdot H_\phi(\rho),$$
$$H_{\lambda^\pm}(\rho_s^\pm) \underset{s\to 0^-}{\longrightarrow} H_{\phi_0}(\rho) \mp \Omega(q,p) \cdot H_\phi(\rho).$$

Therefore
$$H(\rho) = H_{\phi_0}(\rho) - \Omega(q,p) \cdot H_\phi(\rho), \quad H'(\rho) = H_{\phi_0}(\rho) + \Omega(q,p) \cdot H_\phi(\rho)$$
where $H_\phi = (H_{\phi_1}, H_{\phi_2}, H_{\phi_3}, H_{\phi_4}, H_{\phi_5})$.
We consider now (v_k), $1 \le k \le 4$ an orthonormal basis of the hyperplane normal to Ω in $\mathbf{R}^5$ and we set
$$Y_k = v_k \cdot H_\phi,\ 1 \le k \le 4.$$
The six vectors $H(\rho_0)$, $H'(\rho_0)$, $Y_1, \cdots, Y_4$ are a basis of $TS^\perp_{|\rho}$ and $Y_1, \cdots, Y_4$ generates $TS_{|\rho} \cap TS^\perp_{|\rho}$. Besides
$$M(\rho)H(\rho) = rH(\rho), \quad M(\rho)H'(\rho) = -rH'(\rho), \quad M(\rho)Y_k(\rho) = 0,\ 1 \le k \le 4. \tag{23}$$
In view of

$$\omega(H, H') = 2r, \quad \omega(Y_k, Y'_k) = 0, \quad k, k' \in \{1, 4\}$$
$$\omega(Y_k, H) = \omega(Y_k, H') = 0, \quad k \in \{1, 4\},$$

we can modify these vectors to get a symplectic basis of $TS^\perp_{|\rho}$ by setting

$$e_1(\rho) = -(2r)^{-1/2}\, H, \quad f_1(\rho) = (2r)^{-1/2}\, H', \quad f_{k+1} = r^{-1/2} Y_k, \quad k \in \{1, 4\}.$$

Choosing moreover

$$e_{k+1} = r^{1/2} \left(-\mathrm{d}\phi(q)^{-1} v_k, 0, 0, (\mathrm{d}\phi(q)^{-1} v_k) \cdot \nabla v(q) \right), \quad k \in \{1, 4\},$$

the family $(e_k, f_k)_{1 \le k \le 5}$ is a symplectic family of $T_{|\rho}(T^*\mathbf{R}^{d+1})$.

Symplectic coordinates near ρ: We now extend this family in a symplectic basis of $T_\rho(T^*\mathbf{R}^{d+1})$. There exists local coordinates $(z', \tilde{\zeta}, \zeta') \in \mathbf{R}^{d-4} \times \mathbf{R}^4 \times \mathbf{R}^{d-4}$ on S such that

$$(f_2, \cdots, f_5) = \partial_{\tilde{\zeta}}, \quad \text{and} \quad \omega_{|S} = \mathrm{d}z' \wedge \mathrm{d}\zeta'.$$

Then, if we consider the symplectic form $\tilde{\omega}$ defined on $T_\rho(T^*\mathbf{R}^{d+1})$ for $\rho \in S$ by

$$\tilde{\omega} = f_1^* \wedge e_1^* + \cdots + f_5^* \wedge e_5^* + \mathrm{d}\zeta' \wedge \mathrm{d}z',$$

and extended to $T(T^*\mathbf{R}^{d+1})$ by stating that $\tilde{\omega}$ is invariant along the Hamiltonian trajectories of f_2, f_3, f_4, f_5, we have
$$\forall \rho \in S, \quad \omega = \tilde{\omega} \ \text{ on } \ T_\rho(T^*\mathbf{R}^{d+1}).$$
We use the Weinstein's Theorem (see [17]) which says that if two symplectic forms on $T^*\mathbf{R}^{d+1}$ are equal on a submanifold S, then there exists a local diffeomorphism Φ such that $\mathrm{d}\Phi = \mathrm{Id}$ and $\Phi^*\tilde{\omega} = \omega$.

Applying this theorem, we find local symplectic coordinates $(y, \tilde{z}, z', \eta, , \tilde{\zeta}, \zeta')$ near ρ such that

$$\partial_\eta = f_1, \;\; \partial_y = e_1, \;\; \partial_{\tilde{\zeta}} = (f_2, \cdots, f_4), \;\; \partial_{\tilde{z}} = (e_2, \cdots, e_4).$$

Expressions of the vector fields H and H': By the definition of s and σ, if $\rho \in S$, we have

$$\frac{1}{\sqrt{2r}} H = -e_1 = -\partial_y = \frac{1}{\sqrt{2}}(\partial_s + \partial_\sigma), \;\; \frac{1}{\sqrt{2r}} H' = f_1 = \partial_\eta = \frac{1}{\sqrt{2}}(\partial_s - \partial_\sigma).$$

Expression of $\tilde{z}$, σ and s in coordinates (q, t, p, τ): Consider $\delta\rho = (\delta q, \delta t, \delta p, \delta\tau) \in T_\rho(T^*\mathbf{R}^{d+1})$ for $\rho \in S$. We have for $1 \leq k \leq 4$,

$$\mathrm{d}z_k \, \delta\rho = \omega(e_{k+1}, \delta\rho) = r^{-1/2} \, \omega(Y_k, \delta\rho) = r^{-1/2} \, v_k \cdot (\mathrm{d}\phi(q)\delta q).$$

Besides, if $f(q,p) = |\mathrm{d}\phi(q)p|^{-1/2} \pi_5(q,p) \, (\phi(q))$, the expression of $\mathrm{d}f(q,p)\delta\rho$ in the basis $(v_k)_{1\leq k\leq 4}$ is

$$\mathrm{d}f \delta\rho = r^{-1/2} \left(v_1 \cdot (\mathrm{d}\phi(q)\delta q), \cdots, v_4 \cdot (\mathrm{d}\phi(q)\delta p) \right) + O\left(|\phi(q)|\right),$$

where we used $r = |\mathrm{d}\phi(q)p|$. Hence the relation stated in Theorem 1 for $\tilde{z}$ in terms of the coordinates (q,p). Similarly,

$$\mathrm{d}\sigma \, \delta\rho = -\omega(\partial_s, \delta\rho) = -\frac{1}{2\sqrt{r}} \omega(H + H', \delta\rho) = -r^{-1/2} \mathrm{d}(\tau + v(q) + \frac{|p|^2}{2})\delta\rho,$$
$$\mathrm{d}s \, \delta\rho = \omega(\partial_\sigma, \delta\rho) = \frac{1}{2\sqrt{r}} \omega(H - H', \delta\rho) = -r^{-1/2} \Omega \cdot \omega(H_\phi, \delta\rho) = -r^{-1/2} \Omega \cdot \mathrm{d}\phi(q)\delta\rho.$$

Hence (13)-(14).
Moreover, in this new system of coordinates, in view of $g = 0$, $\mathrm{d}g = 0$ and (23) the dispersion relation is

$$g = r(2y\eta - |\tilde{z}|^2) + O_S(3).$$

6 The formal normal form for the dispersion relation

Proof of Proposition 4: The idea is to ameliorate the rest term step by step. Proposition 3 yields the existence of a canonical transform κ_3, of functions $r_3 \in O_S(3)$ and e_3, $0 < e_3 < 1$ such that

$$g \circ \kappa_3 = e_3 g_0 + r_3.$$

We suppose that for $N \geq 3$ we have built

- a canonical transform κ_N,
- a function e_N with $0 < e_N < 1$,
- a function $r_N \in O_S(N)$,
- a polynomial γ_N of degree $N-1$ with smooth coefficients in $\mathcal{C}^\infty(S)$ such that $\gamma_N = O(|\tilde{z}|^2)$ and $\gamma_3 = 0$

such that

$$g \circ \kappa_N = e_N \left(\sigma^2 - s^2 - |\tilde{z} + \gamma_N(z,\zeta)|^2\right) + r_N.$$

Following the notations of [3], we introduce the spaces $\mathcal{H}_N$ of homogeneous functions of degree N with respect to $(y, \eta, \tilde{z})$, $\tilde{z} \in \mathbf{R}^4$. We use the following lemma

Lemme 1 *Consider $\rho \in \mathcal{H}_N$. There exists $U \in \mathcal{H}_N$, $W \in \mathcal{H}_{N-1}$ an homogeneous polynomial of degree $N-1$ with respect to the variables $\tilde{z}$ with coefficients in $\mathcal{C}^\infty(S)$, $V \in \mathcal{H}_{N-2}$ and $f \in \mathcal{H}_N$ an homogeneous polynomial of degree N with respect to the variable $\tilde{z}$ with coefficients in $\mathcal{C}^\infty(S)$, such that*

$$\{U + W, g_0\} + V g_0 + \rho = f + O_S(N+1). \tag{24}$$

The proof of this lemma is similar to the one of Lemma 5 in [3] (see also Lemma 2 in [2]).

Consider ρ the term of order N of the Taylor expansion of r_N on $\tilde{z}$. We apply the lemma to the function ρ. We find U, V, W, f satisfying (24) which will help us to build the canonical transform κ_{N+1} and the function e_{N+1}.
We define $\chi(\delta)$ the family of canonical transforms such that

$$\frac{\mathrm{d}}{\mathrm{d}\delta}\chi(\delta) = H_{U+W} \circ \chi(\delta), \quad \chi(0) = \mathrm{Id}.$$

We consider

$$\lambda(\delta) = [(1 + \delta V)(g_0 + \delta r_N)] \circ \chi(\delta).$$

Then

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}s}\lambda(\delta) &= (\{U + W, g_0\} + \rho + V g_0) \circ \chi(\delta) \\ &\quad + \delta\{U + W, V g_0 + r_N + \delta V r_N\} \circ \chi(\delta) + 2\delta V r_N + (r_N - \rho) \circ \chi(\delta).\end{aligned}$$

We observe that $r_N - \rho \in O_S(N+1)$, $V r_N \in O_S(2N-2) \subset O_S(N+1)$ since $N \geq 3$ and that $\{U, V g_0 + r_N\} \in O_S(2N-2) \subset O_S(N+1)$. Besides in the bracket $\{W, V g_0 + r_N\}$, there is no derivations in y, η but only in the other variables, therefore $\{W, V g_0 + r_N\} \in O_S(2N-2) \subset O_S(N+1)$.
We get

$$\frac{\mathrm{d}}{\mathrm{d}\delta}\lambda(\delta) = f \circ \chi(\delta) + O_S(N+1).$$

Integrating between $\delta = 0$ and $\delta = 1$, we obtain

$$[(1 + V)(g_0 + r_N)] \circ \chi(1) = g_0 + \tilde{f} + O_S(N+1),$$

where $\tilde{f}$ is an homogeneous polynomial of degree N in $\tilde{z}$ with coefficients in S (we just use Taylor expansion at S). We set $\kappa_{N+1} = \kappa_N \circ \chi(1)$ and we have

$$\begin{aligned}g \circ \kappa_{N+1} &= e_N\,(g_0 + r_N) \circ \chi(1) - e_N \left(2\tilde{z} \cdot \gamma_N + |\gamma_N|^2\right) \circ \chi(1) \\ &= e_{N+1} \left(g_0 + \tilde{f} + 2\tilde{z} \cdot \tilde{\gamma}_N - |\tilde{\gamma}_N|^2\right) \circ \chi(1) + O_S(N+1),\end{aligned}$$

where we have modified the coefficients of the polynomial γ_N. Since $\tilde{f}$ is homogeneous of degree N, we can write

$$g_0 + \tilde{f} + 2\tilde{z} \cdot \tilde{\gamma}_N - |\tilde{\gamma}_N|^2 = \sigma^2 - s^2 - |\tilde{z} - \gamma_{N+1}|^2 + O_S(N+1),$$

with γ_{N+1} a polynomial of degree N in $\tilde{z}$ with smooth coefficients in $\mathcal{C}^\infty(S)$.
Finally, one observes that equations (12)-(14) are preserved since $\mathrm{d}\chi(1) = \mathrm{Id}$ above S. $\diamondsuit$

7 The gauge transform

Proof of Proposition 5: Consider the function γ defined in Proposition 4 and satisfying equation (18). We set

$$\tilde{P}_0 := \begin{pmatrix} \sqrt{2}\,\eta\,\mathrm{Id} & h(\tilde{z}+\gamma(z,\zeta)) \\ h(\tilde{z}+\gamma(z,\zeta))^* & \sqrt{2}\,y\,\mathrm{Id} \end{pmatrix}.$$

We have

$$\det(P\circ\kappa_1) = \det\tilde{P}_0 + O_S(\infty).$$

We now linearize $P\circ\kappa_1$: we write $P\circ\kappa_1 = Q + O_S(2)$ where Q is a linear map from $\mathbf{R}^6_{y,\eta,\tilde{z}}$ on the set of matrix of the form (19). We have

$$\det Q = \left(2\eta y - |\tilde{z}|^2\right)^2.$$

The crucial lemma is the following:

Lemme 2 *Consider $Q(y,\eta,\tilde{z})$ a linear function of the form (19) and such that* $\det Q = 2\eta y - |\tilde{z}|^2$*. Then, there exists a matrix $\tilde{A}_0 \in \mathbf{H}^{2,2}$ and a canonical transform κ_2 which preserves y,η and $|\tilde{z}|^2$ such that,*

$$\tilde{A}_0^*\,(Q\circ\kappa_2)\,\tilde{A}_0 = \pm\begin{pmatrix} \sqrt{2}\,\eta\,\mathrm{Id} & h(\tilde{z}) \\ h(\tilde{z})^* & \sqrt{2}\,y\,\mathrm{Id} \end{pmatrix},$$

Besides, κ_2 preserves (12)-(14).

We postpone the proof of this lemma at the end of the section.

Using this lemma, we have

$$\tilde{A}_0^*(P\circ\kappa_1\circ\kappa_2)\tilde{A}_0 = \pm\tilde{P}_0 + O_S(2) \ \text{ and } \ \det\left(\tilde{A}_0^*(P\circ\kappa_1\circ\kappa_2)\tilde{A}_0\right) = \det\tilde{P}_0 + O_S(\infty).$$

Then we apply a path method similar to the one of [2] (Lemma 4) between $\tilde{P}_0$ and $\tilde{P} = \tilde{A}_0^*(P\circ\kappa_1\circ\kappa_2)\tilde{A}_0$ which are both of the form (19).
First, by using the Morse-Bott lemma exactly as in Lemma 4 of [2], one builds a path $\tilde{P}_\tau$ form $\tilde{P}_0$ to $\tilde{P} = \tilde{P}_1$, $\tau\in[0,1]$ such that

$$\det\tilde{P}_\tau = \det\tilde{P}_0.$$

Then, one finds B_τ such that

$$B_\tau^*\tilde{P}_\tau + \tilde{P}_\tau B_\tau = -\frac{\mathrm{d}}{\mathrm{d}\tau}\tilde{P}_\tau, \tag{25}$$

with B_τ of the form

$$B_\tau = \begin{pmatrix} \lambda\,\mathrm{Id} & h(f) \\ h(g)^* & -\lambda\,\mathrm{Id} \end{pmatrix}.$$

Finally, one considers A_τ such that

$$\frac{\mathrm{d}}{\mathrm{d}\tau}A_\tau = B_\tau A_\tau, \ \ (A_\tau)_{|\tau=0} = \mathrm{Id}.$$

$A_\tau \in \mathbf{H}^{2,2}$ and satisfies $A_\tau^* \tilde{P}_\tau A_\tau = \tilde{P}_0$.
We close the proof of Proposition 5 by choosing $A_0 = A_1 \tilde{A}_0$. The fact that one can solve (25) comes from precise computations on quaternions, using *(i), (ii), (iii)* and following exactly the same steps that in the proof of Lemma 4 in [2].
Besides, in view of (20), (13) and (14), we obtain that for $\tau = -\frac{|p|^2}{2} - v(q)$ and for $\phi(q) = 0$,

$$\forall \delta q \in \mathbf{R}^d,\ \ (A_0^*)_{|S} V_\ell \left(\mathrm{d}\phi(q)\delta q\right)(A_0)_{|S} = \pm |\mathrm{d}\phi(q)p|^{-1/2}\, V_\ell\left(-\frac{\mathrm{d}\phi(q)p}{|\mathrm{d}\phi(q)p|}\cdot \mathrm{d}\phi(q)\delta q, \delta\tilde{z}\right),$$

where $\delta\tilde{z}$ are the coordinates of $\pi_\ell(q,p)\left(\mathrm{d}\phi(q)\delta q\right)$. Since the range of $\mathrm{d}\phi(q)$ is $\mathbf{R}^5$, we obtain (15). It just remains to prove Lemma 2.

Proof of Lemma 2: We follow the ideas of the proof of Lemma 3 in [2]. We write

$$Q(\eta, y, 0) = \eta M_1 + y M_2,$$

where M_1 and M_2 are constant non zero matrices of the form (19) such that moreover

$$\det M_1 = \det M_2 = 0.$$

Therefore, M_1 and M_2 are of rank 2 and there exists $t_j \in \mathcal{C}^\infty(S, \mathbf{R}^*)$, $\omega_j \in \mathcal{C}^\infty(S, \mathbf{S}^3)$ such that

$$M_j = \begin{pmatrix} t_j\,\mathrm{Id} & h(\omega_j) \\ h(\omega_j)^* & 1/t_j\,\mathrm{Id} \end{pmatrix}.$$

This yields that M_j has a sign: M_j is positive if $t_j > 0$ and negative is $t_j < 0$. Besides, since $\det\left(yM_1 + \eta M_2\right) = 2\,y\eta$, we get

$$\left(\frac{t_2}{t_1} + \frac{t_1}{t_2}\right) - 2\,\omega_1 \cdot \omega_2 = 2. \tag{26}$$

The vectors of $\operatorname{Ker} M_j$ are of the form

$$V = \begin{pmatrix} X \\ -t_j h(\omega_j)^* X \end{pmatrix} \ \text{ or } \ V = \begin{pmatrix} -t_j^{-1} h(\omega_j) X \\ X \end{pmatrix},\ \ X \in \mathbf{R}^2.$$

We consider

$$V_1 = \begin{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \\ h(-t_1\omega_1)^* \begin{pmatrix} 1 \\ 0 \end{pmatrix} \end{pmatrix},\ \ W_1 = \begin{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix} \\ h(-t_1\omega_1)^* \begin{pmatrix} 0 \\ 1 \end{pmatrix} \end{pmatrix},$$

a basis of $\operatorname{Ker} M_1$ and

$$V_2 = \begin{pmatrix} h(-t_2^{-1}\omega_2) \begin{pmatrix} 1 \\ 0 \end{pmatrix} \\ \begin{pmatrix} 1 \\ 0 \end{pmatrix} \end{pmatrix},\ \ W_2 = \begin{pmatrix} h(-t_2^{-1}\omega_2) \begin{pmatrix} 0 \\ 1 \end{pmatrix} \\ \begin{pmatrix} 0 \\ 1 \end{pmatrix} \end{pmatrix},$$

a basis of $\operatorname{Ker} M_2$. These vectors are smooth functions on S.

We denote by f_1 (resp. f_2) the quadratic form associated with M_1 (resp. M_2), then the quadratic form $\tilde f$ associated with Q satisfies $\tilde f = \eta f_1 + y f_2$. Finally, we denote by $\tilde\phi$ the bilinear form associated with $\tilde f$. We have

$$\begin{array}{ll} \tilde f(V_1) = y f_2(V_1), & \tilde f(W_1) = y f_2(W_1), \\ \tilde f(V_2) = \eta f_1(V_2), & \tilde f(W_2) = \eta f_1(W_2), \\ \tilde\phi(V_1, V_2) = 0, & \tilde\phi(W_1, W_2) = 0. \end{array}$$

We observe that by use of (iii) and of (26), we have

$$f_1(V_2) = f_1(W_2) = 2\, t_1^{-1}, \;\; f_2(V_1) = f_2(W_1) = 2\, t_2^{-1}.$$

Therefore, modifying V_1, V_2, W_1, W_2 by a constant so that

$$|f_1(W_2)| = |f_1(V_2)| = |f_2(W_1)| = |f_2(V_1)| = 1,$$

the matrix of $\tilde\phi$ in the basis (V_1, W_1, V_2, W_2) is

$$\tilde A_0^* Q(\eta, y, 0) \tilde A_0 = \begin{pmatrix} y \begin{pmatrix} \varepsilon & 0 \\ 0 & \varepsilon \end{pmatrix} & \mathbf{0} \\ \mathbf{0} & \eta \begin{pmatrix} \varepsilon' & 0 \\ 0 & \varepsilon' \end{pmatrix} \end{pmatrix},$$

with $\varepsilon, \varepsilon' \in \{-1, +1\}$ and $\tilde A_0$ is a smooth matrix defined on S and valued in $\mathbf{H}^{2,2}$.
We come back now to $Q(\eta, y, \tilde z)$. There exists $a, b \in \mathcal{C}^\infty(S, \mathbf{R}^4)$ and $M \in \mathcal{C}^\infty(S, \mathbf{R}^{4,4})$ such that

$$\tilde A_0^* Q(y, \eta, \tilde z) \tilde A_0 = \begin{pmatrix} (\varepsilon\, y + \tilde z \cdot a)\, \mathrm{Id} & h(M\tilde z) \\ h(M\tilde z)^* & (\varepsilon'\, \eta + b \cdot \tilde z)\, \mathrm{Id} \end{pmatrix}.$$

The relation on the determinant yields

$$a = 0, \; b = 0, \; \varepsilon\varepsilon' = 1, \; |M\tilde z|^2 = |\tilde z|^2.$$

Up to some $\pm$ sign, we can suppose that $\varepsilon = \varepsilon' = 1$. Besides, $M \in O_4(\mathbf{R})$; thus $M\tilde z$ satisfies the same geometric property than $\tilde z$. By modifying $\tilde\zeta$, z' and ζ' one can turn $M(z', \zeta', \tilde\zeta)\tilde z$ into $\tilde z$. The canonical transform used for that purpose preserves y, η and $|\tilde z|^2$ and thus equations (12)-(15).$\diamondsuit$

8 The semi-classical normal form

Proof of Proposition 6: For simplicity, we work with the plus sign in Proposition 5.
We argue by induction. We first observe that for $j = 1$, (21) holds. Indeed, if K_1 is a Fourier integral operator associated with the canonical transform κ_2 of Proposition 5, then

$$K_1 \,\mathrm{op}_\varepsilon(A_0^*) \,\mathrm{op}_\varepsilon(P) \,\mathrm{op}_\varepsilon(A_0)\, K_1^* = \mathrm{op}_\varepsilon(P_0 + \Gamma_0) + \varepsilon \,\mathrm{op}_\varepsilon(T_1) + O(\varepsilon^2) \;\text{ in }\; \mathcal{L}(L^2(\mathbf{R}^d)),$$

with $A_0 \in \mathbf{C}^{4,4}$, $\Gamma_0 = \begin{pmatrix} 0 & h(\gamma(z,\zeta)) \\ h(\gamma(z,\zeta))^* & 0 \end{pmatrix}$ and T_1 of the form (19).

Let us suppose now that (21) holds for some $j \geq 1$ and let us build K_{j+1}^ε, A_j and Γ_j in order to get the statement for the index $j+1$.

We decompose the hermitian matrix T_j of $\mathbf{H}^{2,2}$ into the sum of a diagonal matrix and of a matrix $\tilde\Gamma_j$ with 0 on the diagonal:

$$\text{if } \; T_j = \begin{pmatrix} a\,\mathrm{Id} & h(\gamma_j) \\ h(\gamma_j)^* & b\,\mathrm{Id} \end{pmatrix} \; \text{ we set } \; \tilde\Gamma_j := \begin{pmatrix} \mathbf{0} & h(\gamma_j) \\ h(\gamma_j)^* & \mathbf{0} \end{pmatrix}.$$

In the following, the matrices named 'Γ' will have the same form.
We then claim that there exists a function U and a matrix $\tilde A_j$ of $\mathbf{C}^{4,4}$ such that

$$\{U, P_0\} + \tilde A_j^* P_0 + P_0 \tilde A_j = \tilde\Gamma_j - T_j. \tag{27}$$

We postpone the proof of this claim at the end of the section. We are now provided with U for building the canonical transform and of $\tilde A_j$, a matrix of $\mathbf{C}^{4,4}$.
We consider canonical transforms which are a perturbation of identity: $\chi^\varepsilon(\delta)$ for $\delta \in [0,1]$ satisfies

$$\frac{\mathrm{d}}{\mathrm{d}\delta}\chi^\varepsilon(\delta) = H_{1+\varepsilon^j U} \circ \chi^\varepsilon(\delta), \;\; \chi^\varepsilon(0) = \mathrm{Id}.$$

If $K^\varepsilon(\delta)$ is a Fourier integral operator associated to $\chi^\varepsilon(\delta)$, we have by (8)

$$\frac{\mathrm{d}}{\mathrm{d}\delta}\left(K^\varepsilon(\delta)^*\mathrm{op}_\varepsilon(a)K^\varepsilon(\delta)\right) = \varepsilon^j K^\varepsilon(\delta)^*\mathrm{op}_\varepsilon\Big(\{U,a\}\Big)K^\varepsilon(\delta) + O(\varepsilon^{j+1}) \;\text{ in }\; \mathcal{L}\left(L^2(\mathbf{R}^d,\mathbf{C}^4)\right).$$

We set $A_j = A_0\tilde A_j$ and we define

$$B^\varepsilon(\delta) = \\ K^\varepsilon(\delta)^*\left[K_j^{\varepsilon *}\mathrm{op}_\varepsilon((A_\varepsilon^{(j)})^* + \delta\,\varepsilon^j A_j^*)\;\mathrm{op}_\varepsilon(P)\;\mathrm{op}_\varepsilon(A_\varepsilon^{(j)} + \delta\,\varepsilon^j A_j)\,K_j^\varepsilon - (1-\delta)\,\varepsilon^j\,\mathrm{op}_\varepsilon(T_j)\right]K_\varepsilon(\delta).$$

Since (21) holds for the index j, we have in $\mathcal{L}\left(L^2(\mathbf{R}^d,\mathbf{C}^4)\right)$

$$B^\varepsilon(0) = \mathrm{op}_\varepsilon(P_0) + \mathrm{op}_\varepsilon\left(\Gamma_\varepsilon^{(j)}\right) + O(\varepsilon^{j+1}),$$
$$B^\varepsilon(1) = (K_{j+1}^\varepsilon)^*\,\mathrm{op}_\varepsilon(A_\varepsilon^{(j+1)^*})\,\mathrm{op}_\varepsilon(P)\,\mathrm{op}_\varepsilon(A_\varepsilon^{(j+1)})\,K_{j+1}^\varepsilon,$$

where we have set

$$K_{j+1}^\varepsilon = K_j^\varepsilon K^\varepsilon(1).$$

We observe that

$$B_\varepsilon(\delta) = K^\varepsilon(\delta)^*\left[\mathrm{op}_\varepsilon\left(P_0 + \Gamma_\varepsilon^{(j)} + \varepsilon^j\delta(\tilde A_j^* P_0 + P_0\tilde A_j) - (1-\delta)\varepsilon^j\,T_j\right)\right]K^\varepsilon(\delta) + O(\varepsilon^{j+1}),$$

in $\mathcal{L}\left(L^2(\mathbf{R}^d,\mathbf{C}^4)\right)$, where we have used the definition of A_j and symbolic calculus. Therefore in $\mathcal{L}\left(L^2(\mathbf{R}^d,\mathbf{C}^4)\right)$

$$\frac{\mathrm{d}}{\mathrm{d}\delta}B_\varepsilon(\delta) = \varepsilon^j K^\varepsilon(\delta)^*\left[\mathrm{op}_\varepsilon\left(\{U, P_0+\Gamma_0\} + \tilde A_j^* P_0 + P_0\tilde A_j + T_j\right)\right]K^\varepsilon(\delta) + O(\varepsilon^{j+1}). \tag{28}$$

The matrix

$$\Gamma(\delta) = K^\varepsilon(\delta)^*\mathrm{op}_\varepsilon\left(\{U,\Gamma_0\} + \tilde\Gamma_j\right)K^\varepsilon(\delta)$$

also is a hermitian matrix of $\mathbf{H}^{2,2}$ with 0 on the diagonal. Thus, integrating (28) between $\delta = 0$ and $\delta = 1$, we get in view of (27)

$$B^\varepsilon(1) = B^\varepsilon(0) + \varepsilon^j \mathrm{op}_\varepsilon(\Gamma_j) + O(\varepsilon^{j+1})$$

where $\Gamma_j = \int_0^1 \Gamma(\delta)\,\mathrm{d}\delta$ is a hermitian matrix of $\mathbf{H}^{2,2}$ with 0 on the diagonal. This gives (21) for the index $j+1$.

Let us prove now that we can solve the homological matrix-valued equation (27). We follow the strategy of Lemma 5 in [2] or of Lemma 6 in [3] and apply Proposition 5 to $P_0 + \delta(T_j - \tilde{\Gamma}_j)$ for $\delta \in [0, \delta_0]$, $\delta_0 \ll 1$. This is possible if the crossing for the symbol $P_0 + \delta(T_j - \tilde{\Gamma}_j)$ has the same symplectic structure that the one of P_0, i.e. if the corank of the symplectic form ω above the crossing set $S_\delta = \left\{\det\big(P_0 + \delta(T_j - \tilde{\Gamma}_j)\big) = 0\right\}$ is constant and equal to 4. This geometric assumption is fulfilled since $\tilde{\Gamma}_j - T_j$ is diagonal. Then, applying Proposition 5 to $P_0 + \delta(T_j - \tilde{\Gamma}_j)$, differentiating with respect to δ and plugging $\delta = 0$, one solves the matrix-valued homological equation (27).

For concluding the proof, it remains to prove equations (10) – (11). We observe that the image by the canonical transform of the Hamiltonian trajectories associated with the old system are those of the new one. Since $S = \{\sigma = s,\ \tilde{z} = 0\}$, the hamiltonian trajectories of the new system passing through S are included in $\{\sigma^2 = s^2,\ \tilde{z} = 0\}$. Therefore

$$J \cup J' \subset \{\sigma^2 = s^2,\ \ \tilde{z} = 0\}.$$

Because of the property of H and H' with respect to $\partial_{\sigma+s}$ and $\partial_{\sigma-s}$, the definition of $J^{\pm,in}$ and $J^{\pm,out}$ allows to identify each part of J and J'. One obtains

$$J^{\pm,in} \subset \{s < 0\},\ \ J^{\pm,out} \subset \{s > 0\},$$
$$J \subset \{\sigma - s = 0\},\ \ J' \subset \{\sigma + s = 0\}.$$

Hence, by dimension considerations, we get equations (10) – (11). $\diamondsuit$

9 Application: Analysis of the interactions between modes for microlocalized initial data

In this section, for proving Proposition 2, we make intensive use of results of [7]. However, our purpose is different since we are concerned with the interaction between both modes. It is well-known that it is not possible to calculate directly the evolution of the position/impulsion density but that it is useful to consider the microlocal energy density which is the Wigner transform. More precisely, when there are conical intersections and for this special potential, one uses two-scale Wigner transform (see [5] and [7])

$$W_\varepsilon^{(2)}(q,p,t,\tau,\eta) =$$
$$(2\pi)^{-d}\delta\left(\eta - \frac{q\wedge p}{\sqrt{\varepsilon}}\right) \otimes \int_{\mathbf{R}^d} \psi_0^\varepsilon\left(q - \varepsilon\frac{v}{2}, t - \varepsilon\frac{s}{2}\right) \otimes \overline{\psi}_0^\varepsilon\left(q + \varepsilon\frac{v}{2}, t + \varepsilon\frac{v}{2}\right) \mathrm{e}^{ip\cdot v + it\cdot s}\mathrm{d}v\,\mathrm{d}s.$$

This two-scale Wigner transform is tested against matrix-valued compactly supported test functions in the variables (q,t,p,τ,η) which is compatible with the fact that $\psi^\varepsilon \in L^2_{loc}(\mathbf{R}^{d+1})$. Any weak limit μ of this quantity is a positive Radon measure called *two-scale Wigner measure* of ψ^ε for the submanifold $\{q\wedge p=0\}$. The additional variable η encounters the spread of the wave packet with respect to the scale $\sqrt{\varepsilon}$ around the classical trajectories passing through crossing points. Indeed, these curves are included in the set $\{q\wedge p=0\}$. We point out that the position density (resp. the impulsion density) is the projection of $W^{(2)}_\varepsilon$ on the space of the variables (q,t) (resp. (p,t)). Besides, we observe that the initial data ψ^ε_0 is ε- oscillating, i.e.

$$\overline{\lim_{\varepsilon\to 0}}\int_{\tau+|p|>R/\varepsilon} |(\widehat{\phi\psi^\varepsilon_0})(\tau,p)|^2 \mathrm{d}p\,\mathrm{d}\tau \underset{R\to+\infty}{\longrightarrow} 0 \;\; \forall\phi\in\mathcal{C}_0^\infty(\mathbf{R}^{d+1}),$$

and that the same property holds for the solution ψ^ε. Therefore, the weak limits of the position (resp. impulsion) density satisfy

$$n_{pos}(q,t)=\int_{p,\tau,\eta}\mu(q,t,\mathrm{d}p,\mathrm{d}\tau,\mathrm{d}\eta),\;\; n_{imp}(q,t)=\int_{q,\tau,\eta}\mu(\mathrm{d}q,t,p,\mathrm{d}\tau,\mathrm{d}\eta).$$

Therefore, we will focus on the measures μ.

According to [7], outside crossings, the 2-scale Wigner measures μ decompose as $\mu=\mu^+\Pi^+ + \mu^-\Pi^-$, the measures $\mu^\pm$ are scalar, supported on $\{(q,p)=(q^\pm(t),p^\pm(t))\}\times\{\lambda^\pm(q,p,\tau)=0\}$, absolutely continuous with respect to the Lebesgue measure $\mathrm{d}t$ and propagate along classical trajectories. Therefore, after a standard computation on the initial data, we obtain that on $[0,t^*)\times\mathbf{R}^{2d+1}$

$$\mu^\pm(q,t,p,\tau,\eta)=c^{\pm,in}\delta\left(\lambda^\pm(q,p,\tau)\right)\otimes\delta\left(q-q^\pm(t)\right)\otimes\delta\left(p-p^\pm(t)\right)\otimes\nu^{\pm,in}(\eta)\,\mathrm{d}t,$$

where $q^\pm(t)$ are the classical trajectories arising from $(q_0^\pm,p_0^\pm)$ and $\nu^{\pm,in}$ is a positive Radon measure on $\mathbf{R}\cup\{\infty\}$ if $\ell=2$ and on $\mathbf{R}^3\cup\{\infty\}$ if $\ell=3$ with

$$\text{if } \;\alpha^\pm=1/2,\;\; \nu^{\pm,in}=\delta(\eta-\eta_0^\pm),\;\; \text{if }\;\alpha^\pm<1/2,\;\; \nu^{\pm,in}=\delta\left(\eta-\infty\right).$$

This gives the first part of Proposition 2. Besides, there exist measures $\nu^{\pm,out}$ such that for $t\in(t^*,t_1)$,

$$\mu^\pm(q,t,p,\tau,\eta)=\nu^{\pm,out}(\eta)\otimes\delta\left(\lambda^\pm(q,p,\tau)\right)\otimes\delta\left(q-q^\pm(t)\right)\otimes\delta\left(p-p^\pm(t)\right)\,\mathrm{d}t$$

and it remains to calculate the measures $\nu^{\pm,out}$ since the coefficients $c^{\pm,in/out}$ are given by

$$c^{\pm,in/out}=\int_\eta \nu^{\pm,in/out}(\mathrm{d}\eta).$$

For describing what happens near the crossing set, we use the normal form of Theorem 1 and we study $v^\varepsilon(s,z)$ which solves microlocally near $(0,t^*,p^*,\tau^*=-|p^*|^2/2)$

$$\frac{\varepsilon}{i}\partial_s u^\varepsilon=\begin{pmatrix} s\,\mathrm{Id} & \sqrt{\varepsilon}\,K\\ \sqrt{\varepsilon}K & -s\,\mathrm{Id}\end{pmatrix}u^\varepsilon, \tag{29}$$

where the operator K has different forms depending on ℓ

$$\ell=2:\quad K=\frac{z_1}{\sqrt{\varepsilon}},\qquad\qquad \ell=3:\quad K=\frac{z_1+iz_2}{\sqrt{\varepsilon}}+\frac{1}{\sqrt{\varepsilon}}\mathrm{op}_\varepsilon\left(\gamma_\varepsilon(z,\zeta)\right).$$

By Proposition 7 in [6] we have

Proposition 7 *There exist* $\omega^\varepsilon = \omega^\varepsilon(z) = \begin{pmatrix}\omega_1^\varepsilon\\ \omega_2^\varepsilon\end{pmatrix}$ *and* $\alpha^\varepsilon = \alpha^\varepsilon(z) = \begin{pmatrix}\alpha_1^\varepsilon\\ \alpha_2^\varepsilon\end{pmatrix}$ *such that for any* $R>0$ *and* $\chi \in \mathcal{C}_0^\infty\left(B(0,R^2)\right)$, *families* $(\chi(KK^*)\omega_1^\varepsilon)$, $(\chi(K^*K)\omega_2^\varepsilon)$, $(\chi(KK^*)\alpha_1^\varepsilon)$ *and* $(\chi(K^*K)\alpha_2^\varepsilon)$ *are bounded in* $L^2(\mathbf{R}^d_z)$ *and such that as* ε *goes to* 0, *we have in* $L^2(\mathbf{R}^d_z)$

$$\text{for } s<0,\quad \left\{\begin{array}{rcl} \chi(KK^*)v_1^\varepsilon(s,z) &=& \chi(KK^*)\mathrm{e}^{i\frac{s^2}{2\varepsilon}}\left|\frac{s}{\sqrt{\varepsilon}}\right|^{i\frac{KK^*}{2}}\alpha_1^\varepsilon + o(1),\\ \chi(K^*K)v_2^\varepsilon(s,z) &=& \chi(K^*K)\mathrm{e}^{-i\frac{s^2}{2\varepsilon}}\left|\frac{s}{\sqrt{\varepsilon}}\right|^{-i\frac{K^*K}{2}}\alpha_2^\varepsilon + o(1),\end{array}\right.$$

$$\text{for } s>0,\quad \left\{\begin{array}{rcl} \chi(KK^*)v_1^\varepsilon(s,z) &=& \chi(KK^*)\mathrm{e}^{i\frac{s^2}{2\varepsilon}}\left|\frac{s}{\sqrt{\varepsilon}}\right|^{i\frac{KK^*}{2}}\omega_1^\varepsilon + o(1),\\ \chi(K^*K)v_2^\varepsilon(s,z) &=& \chi(K^*K)\mathrm{e}^{-i\frac{s^2}{2\varepsilon}}\left|\frac{s}{\sqrt{\varepsilon}}\right|^{-i\frac{K^*K}{2}}\omega_2^\varepsilon + o(1),\end{array}\right.$$

Moreover

$$\begin{pmatrix}\omega_1^\varepsilon\\ \omega_2^\varepsilon\end{pmatrix} = \begin{pmatrix} a(KK^*) & -\overline{b}(KK^*)K\\ b(K^*K)K^* & a(K^*K)\end{pmatrix}\begin{pmatrix}\alpha_1^\varepsilon\\ \alpha_2^\varepsilon\end{pmatrix}, \tag{30}$$

$$\text{with } a(\lambda) = \mathrm{e}^{-\pi\frac{\lambda}{2}},\ \ a(\lambda)^2 + \lambda|b(\lambda)|^2 = 1. \tag{31}$$

We then use (10) and (11) to identify $\nu^{+,in}$ (resp. $\nu^{-,in}$) as the two-scale Wigner measure of α_1^ε (resp. α_2^ε) for $\{\tilde z = 0\}$ and $\nu^{+,out}$ (resp $\nu^{-,out}$) as the one of ω_2^ε (resp. ω_1^ε).
In the situation ($\alpha^\pm < \alpha^\mp = 1/2$) or ($\alpha^\pm = 1/2$ and $\eta_0^+ \neq \eta_0^-$), the two in-coming measures are mutually singular. Therefore, arguing as in [5], the measure of $\omega_1^\varepsilon = a(KK^*)\alpha_1^\varepsilon - \overline{b}(KK^*)K\alpha_2^\varepsilon$ is the sum of the measure of each term. We obtain

$$\nu^{-,out} = a(|\eta|^2)^2\nu^{+,in} + |\eta|^2|b(|\eta|^2)|^2\nu^{-,in}.$$

Similarly, we get

$$\nu^{+,out} = |\eta|^2|b(|\eta|^2)|^2\nu^{+,in} + a(|\eta|^2)^2\nu^{-,in}.$$

Equations (31) allow to conclude.
When $\alpha^\pm < 1/2$ both incident measures are localized in $\eta = \infty$ and one can then prove that the propagation along classical trajectories hold (see again [5]), whence

$$\nu^{\pm,out} = \nu^{\pm,in}.$$

It remains to consider the most interesting case when $\alpha^\pm = 1/2$ and $\eta_0^+ = \eta_0^-$. We get that $\nu^{\pm,out}$ is localized above $\eta_0^+ = \eta_0^- := \eta_0$ and we have to take into account the joint measure θ between α_1^ε and α_2^ε which is also supported above η_0. We obtain

$$\begin{aligned}\nu^{+,out} &= |\eta_0|^2|b(|\eta_0|^2)|^2\nu^{+,in} + a(|\eta_0|^2)^2\nu^{-,in} + 2\mathcal{R}e\left(\eta_0 a(|\eta_0|^2)b(|\eta_0|^2)\theta\right),\\ \nu^{-,out} &= a(|\eta_0|^2)^2\nu^{+,in} + |\eta_0|^2|b(|\eta_0|^2)|^2\nu^{-,in} - 2\mathcal{R}e\left(\eta_0 a(|\eta_0|^2)b(|\eta_0|^2)\theta\right).\end{aligned}$$

In view of (31), there exists $(\rho_0,\lambda_0) \in \mathbf{R}^+ \times [0,2\pi]$ such that

$$\eta_0 a(|\eta_0|^2)b(|\eta_0|^2)\theta = T(\eta_0)^{\frac12}\left(1 - T(\eta_0)\right)^{\frac12}\rho_0\,\mathrm{e}^{i\lambda_0}\delta(\eta - \eta_0).$$

One then observes that if one multiplies the minus component of the initial data by $\mathrm{e}^{i\lambda}$, one turns α_2^ε into $\mathrm{e}^{i\lambda}\alpha_2^\varepsilon$ and θ into $\mathrm{e}^{-i\lambda}\theta$, whence the result with

$$\gamma_0 = T(\eta_0)^{1/2}\left(1 - T(\eta_0)\right)^{1/2}\rho_0.\ \diamondsuit$$

Remark 3 *The above results apply as soon as the initial data has the same two-scale Wigner measure than our example, which gives a larger class of initial data.*

References

[1] P. Braam, H. Duistermaat: Normal forms of real symmetric systems with multiplicity. *Indag. Math.*; *N.S.*, **4**(4), p. 407-421 (1993).

[2] Y. Colin de Verdière: The level crossing problem in semi- classical analysis. I. The symmetric case. Proceedings of the International Conference in Honor of Frédéric Pham (Nice, 2002). *Ann. Inst. Fourier (Grenoble)*, **53** (2003), no. 4, p. 1023–1054.

[3] Y. Colin de Verdière : The level crossing problem in semi- classical analysis. II. The Hermitian case. *Ann. Inst. Fourier (Grenoble)*, **54** (2004), no. 5, p. 1423–1441.

[4] C. Fermanian Kammerer : Wigner measures and molecular propagation through generic energy level crossings, *Reviews in Mathematical Physics*, **15**, p. 1285-1317 (2003).

[5] C. Fermanian Kammerer, P. Gérard: Mesures semi-classiques et croisements de modes. *Bull. Soc. math. France*, **130**, N^{o}1, (2002), p.123–168.

[6] C. Fermanian Kammerer, P. Gérard: A Landau-Zener formula for non-degenerated involutive codimension 3 crossings. *Ann. Henri Poincaré*, **4**, (2003), p.513-552.

[7] C. Fermanian Kammerer, C. Lasser: Wigner measures and codimension two crossings. *Jour. Math. Phys.* **44**, 2, p. 507-527 (2003).

[8] C. Fermanian Kammerer, C. Lasser: An algorithm for quantum propagation through energy level crossings. *Proceedings of the EDP meeting – Forges-les-Eaux*, (2005).

[9] C. Fermanian Kammerer, C. Lasser: Propagation through generic energy level crossings: A surface hopping semi-group. *Prépublication de l'Université Cergy-Pontoise*, 1/07 – Mars 2007.

[10] P. Gérard, P. A. Markowich, N. J. Mauser, F. Poupaud: Homogenization Limits and Wigner Transforms. *Comm. Pure Appl. Math.*, **50**, (1997), 4, p.323-379 and Erratum : Homogenization limits and Wigner transform. *Comm. Pure Appl. Math.*, **53**, (2000), p.280-281.

[11] G. A. Hagedorn: Molecular Propagation through Electron Energy Level Crossings. *Memoirs of the A. M. S.*, **111**, N^o 536, (1994).

[12] L. Hörmander: The analysis of linear Partial Differential Operators III. *Springer-Verlag*, (1985).

[13] L. Landau: *Collected papers of L. Landau*, Pergamon Press, (1965).

[14] J. Lever, R. Shaw: Irreducible Multiplier Corepresentations and Generalized Inducing, *Comm. Math. Phys.* **38**, 257-277 (1974).

[15] D. Robert: Autour de l'approximation semi–classique, *Birkhaüser*, (1983).

[16] H. Spohn, S. Teufel: Adiabatic decoupling and time-dependent Born-Oppenheimer theory, *Commun. Math. Phys.* **224**, p. 113-132 (2001).

[17] A. Weinstein: Symplectic manifolds and their Lagrangian submanifolds, *Adv. Math.* **6**, p. 329-346 (1971).

[18] E. P. Wigner: *Group Theory and its Application to the Quantum Mechanics of Atomic Spectra.* New York, Academic Press, 1959

[19] C. Zener: Non-adiabatic crossing of energy levels, *Proc. Roy. Soc. Lond.* **137**, (1932), p. 696-702.

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