LIFE-TIME FOR THE MOLECULAR PREDISSOCIATION

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Dedicated to Prof. Yoshinori MORIMOTO
for his 60-th Birthday (Kyoto, January 2011)

As a model for molecular predissociation in the Born-Oppenheimer approximation (see, e.g., [KMSW, MaSo], we consider the following semiclassical 2×2 matrix Schrödinger operator,

$$P = \begin{pmatrix} P_1 & 0 \\ 0 & P_2 \end{pmatrix} + hR(x, hD_x) \tag{0.1}$$

with,

$$P_j := -h^2\Delta + V_j(x) \quad (j = 1, 2),$$

where $x = (x_1, \dots, x_n)$ is the current variable in $\mathbb{R}^n$ $(n \geq 1)$, $h > 0$ denotes a semiclassical parameter, and $R(x, hD_x) = (r_{j,k}(x, hD_x))_{1 \leq j,k \leq 2}$ is a formally self-adjoint 2×2 matrix of first-order semiclassical pseudodifferential operators.

Assumption 1. *The potentials V_1 and V_2 are smooth and bounded on $\mathbb{R}^n$, and satisfy,*

(0.2) $V_1(0) > 0$; 0 *is a non-trapping energy for* V_1;

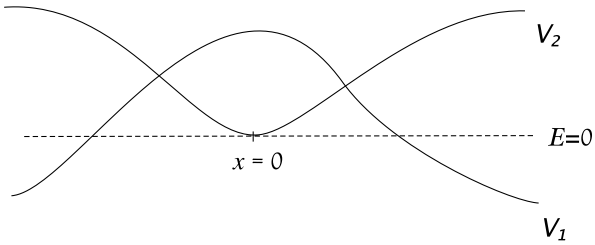
(0.3) V_1 *has a strictly negative limit as* $|x| \to \infty$

(0.4) $V_2 \geq 0$; $V_2^{-1}(0) = \{0\}$; $\mathrm{Hess} V_2(0) > 0$; $\liminf\limits_{|x| \to \infty} V_2 > 0$.

(Here, the fact that 0 is a non-trapping energy for V_1 means that, for any $(x, \xi) \in p_1^{-1}(0)$, one has $|\exp tH_{p_1}(x, \xi)| \to +\infty$ as $t \to \infty$, where $p_1(x, \xi) := \xi^2 + V_1(x)$ is the symbol of P_1, and $H_{p_1} := (\nabla_\xi p_1, -\nabla_x p_1)$ is the Hamilton field of p_1.)

In particular, at energy 0, V_2 admits a unique non degenerate well at $x = 0$, and this well is included in the island $\ddot{O}$ defined as the bounded open set :

$$\ddot{O} = \{x \in \mathbb{R}^n; V_1(x) > 0\}. \tag{0.5}$$



We are interested in the study of the resonances of P near the energy level $E = 0$. From a physical point of view, in this context resonances correspond to unstable molecules, that is, molecules that will dissociate within a final time (see, e.g., [Kl]). Such a time T (called the life-time of the molecule) is related to the imaginary part of the corresponding resonance ρ, through the formula: $T = |\mathrm{Im}\, \rho|^{-1}$. It is therefore of great interest to obtain good estimates on the location of the resonances, and in particular on the size of their imaginary part (width).

In order to study the resonances of P, we assume:

Assumption 2. *The potentials V_1 and V_2 extend to bounded holomorphic functions near a complex sector of the form, $\mathcal{S}_{R_0,\delta} := \{x \in \mathbb{C}^n\,;\, |\mathrm{Re}\, x| \geq R_0\,,\, |\mathrm{Im}\, x| \leq \delta |\mathrm{Re}\, x|\}$, with $R_0, \delta > 0$ (R_0 arbitrarily large). Moreover V_1 tends to its limit at ∞ in this sector and $\mathrm{Re}\, V_2$ stays away from 0 in this sector.*

Assumption 3. *The symbols $r_{j,k}(x,\xi)$ for $(j,k) = (1,1), (1,2), (2,2)$ extend to holomorphic functions near,*

$$\widetilde{\mathcal{S}}_{R_0,\delta} := \mathcal{S}_{R_0,\delta} \times \{\xi \in \mathbb{C}^n\,;\, |\mathrm{Im}\, \xi| \leq \max(\delta \langle \mathrm{Re}\, x\rangle, \sqrt{M_0})\},$$

with,

$$M_0 > \sup_{x\in\mathbb{R}^n} \min(V_1(x), V_2(x)).$$

and, for any $\alpha \in \mathbb{N}^{2n}$, they satisfy

$$\partial^\alpha r_{j,k}(x,\xi) = \mathcal{O}(\langle \mathrm{Re}\, \xi\rangle) \quad \textit{uniformly on } \widetilde{\mathcal{S}}_{R_0,\delta}. \tag{0.6}$$

We also define the cirque Ω as,

$$\Omega = \{x \in \mathbb{R}^n; V_2(x) < V_1(x)\}, \tag{0.7}$$

and we consider the Agmon distance d associated to the pseudo-metric $\min(V_1, V_2))_+ dx^2$. Then, using results from [HeSj1, Pe], it can be shown that the function $\varphi(x) := d(x, 0)$ is C^∞ on $\ddot{O}\backslash\Omega$, and is C^1 near Ω.

We set,

$$S = d(0, \partial\ddot{O}), \tag{0.8}$$

and we consider the ball $B_S = \{x \in \ddot{O}, \varphi(x) < S\}$ and its closure $\bar{B}_S$.

Assumption 4. *At any point $x \in \partial\Omega \cap \bar{B}_S$ we have $\nabla V_1 \neq \nabla V_2$. Moreover, for any $x \in B_S$ the unique geodesic of minimal length joining x to 0 intersects $\partial\Omega$ at a finite number of points and the intersection is tranversal at each of these points.*

Finally we consider the points of $\bar{B}_S \cap \partial\ddot{O}$, that is the points of the boundary of the island that are joined to the well by a minimal geodesic included in the island (these points are called points of type 1 in [HeSj2]). Near those points, we define the caustic set $\mathcal{C}$ as the closure of the set of points $x \in \ddot{O}$ with $\varphi(x) = S + d(x, \partial\ddot{O})$. As in [HeSj2], we assume

Assumption 5. *The points of type 1 form a submanifold Γ, and $\mathcal{C}$ has a contact of order exactly two with $\partial\ddot{O}$ along Γ.*

We denote by n_Γ the dimension of Γ.

Now we consider the following assumption on the minimal geodesic joining the well 0 to the sea (this assumption is to insure that there is an interaction between the two Schrödinger operators P_1 and P_2).

Assumption 6. *There exists at least one point x_1 of type 1 for which we have the ellipticity condition $r_{12}(x, i\nabla\varphi(x)) \neq 0$ at every points x where the minimal geodesic γ joining x_1 to the well 0 crosses the boundary of the cirque $\partial\Omega$.*

In the following, we denote by $\Lambda_0 := \{e_1 < e_2 \leq e_3 \leq \dots\}$ the non-decreasing sequence of eigenvalues of the harmonic oscillator $\Delta + \frac{1}{2}\langle V_2''(0)x, x\rangle$. Then, our result is,

Theorem 0.1. *Under Assumption 1 to Assumption 5, for any $C \in (0, \infty)\backslash\Lambda_0$ and for any $h > 0$ sufficiently small, the resonances of P in $[0, Ch] - i[0, Ch \ln \frac{1}{h}]$ consist in a finite set $\{\rho_1, \dots, \rho_m\}$ (where $m \geq 1$ is such that $e_m < C < e_{m+1}$), such that,*

- *For all j, ρ_j admits a real asymptotic expansion as $h \to 0_+$, of the form,*

$$\rho_j \sim e_j h + \sum_{\ell \geq 1} \rho_{j,\ell} h^{\ell/2}, \quad (\rho_{j,\ell} \in \mathbb{R});$$

- *For all j there exists a constant $m_j \in \mathbb{R}$, such that ,*
$$|\mathrm{Im}\,\rho_j| = \mathcal{O}(h^{m_j} e^{-2S/h}),$$
uniformly as $h \to 0_+$;

If in addition Assumption 6 is satisfied, then,
$$\mathrm{Im}\,\rho_1 = -h^{(1-n_\Gamma)/2} f(h) e^{-2S/h},$$
where $f(h)$ admits an asymptotic expansion of the form,
$$f(h) \sim \sum_{0\leq \ell' \leq \ell} f_{\ell,\ell'} h^\ell (\ln h^{-1})^{\ell'},$$
with $f_{0,0} > 0$. In particular, there exists a constant $C_0 > 0$ such that,
$$|\mathrm{Im}\,\rho_1| \geq \frac{1}{C_0} h^{(1-n_\Gamma)/2} e^{-2S/h},$$
for all $h > 0$ small enough.

The proof of this theorem mainly relies on two ingredients:

- The construction of an asymptotic solution of the equation $Pu = \rho u$ in a neighborhood of the island $\ddot{O}$;
- Some propagation arguments in order to compare the true out-going solution with the asymptotic one.

Construction of an asymptotic solution

The asymptotic solution is constructed starting from a neighborhood of the well 0, where a WKB construction is available (see [HeSj1]), of the form,
$$\begin{pmatrix} ha_1(x,h) \\ a_2(x,h) \end{pmatrix} e^{-\varphi(x)/h}$$
with $a_j(x,h) \sim \sum_{k\geq 0} a_{j,k}(x) h^k$ $(j = 1,2)$.

Then, we can follow any minimal geodesic of d, until we meet the boundary of the cirque $\partial\Omega$. At that point, we generalize the constructions of [Pe] (in order to take into account the pseudodifferential operator $R(x, hD_x)$), and obtain a formal solution involving Weber functions.

After such a point (still following a minimal geodesic), we recover a WKB expression of the form,
$$\begin{pmatrix} b_1(x,h) \\ hb_2(x,h) \end{pmatrix} e^{-\varphi(x)/h},$$
with,
$$b_j(x,h) \sim \sum_{0\leq k\leq \ell} b_{j,k,\ell}(x) h^\ell \left(\ln \frac{1}{h}\right)^k \quad (j = 1,2).$$

Finally, when arriving near the boundary of the island $\partial\ddot{O}$, we use the constructions of [FLM], that are based on an Airy representation of the solution (as in [HeSj2]), but, in absence of analyticity in this region, are valid at a distance of order $(h \ln h^{-1})^{2/3}$ only from $\partial\ddot{O}$.

Propagation arguments

In order to compare the asymptotic solution with the actual one, we first use Agmon estimates inside $\ddot{O}$ (up to a distance of order $(h \ln h^{-1})^{2/3}$ from $\partial\ddot{O}$), as in [HeSj2, FLM]. Then, a refined microlocal propagation argument in h-dependent domains permits, as in [FLM], to estimates the difference in any ball centered at a point of type 1, and of radius $C(h \ln h^{-1})^{2/3}$ ($C > 0$ arbitrarily large). Thanks to this, and to a representation of the imaginary part of ρ by means of the solution, the result can be obtained. The lower bound on the width comes from the fact that, when Assumption 6 is satisfied, one can follow the ellipticity of the various symbols involved in the previous constructions. In particular, the ellipticity of the symbol a_2 is transmitted to the symbol b_1 when crossing the boundary of the cirque.

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