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A Time-Dependent Perturbative Analysis for a Quantum Particle in a Cloud Chamber

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Abstract. We consider a simple model of a cloud chamber consisting of a test particle (the α-particle) interacting with two quantum systems (the atoms of the vapor) initially confined around $a_1, a_2 \in \mathbb{R}^3$. At time zero, the α-particle is described by an outgoing spherical wave centered in the origin and the atoms are in their ground state. We show that, under suitable assumptions on the physical parameters of the system and up to second order in perturbation theory, the probability that both atoms are ionized is negligible unless a_2 lies on the line joining the origin with a_1. The work is a fully time-dependent version of the original analysis proposed by Mott in 1929.

1. Introduction

The classical limit of quantum mechanics is a widely studied subject in mathematical physics and many detailed results on the asymptotic regime $\hbar \to 0$ are available (see e.g., [20] and references therein). In most cases the limit $\hbar \to 0$ for the time-dependent Schrödinger equation relative to a given quantum system is studied only for suitable chosen, "almost classical" states, namely WKB or coherent states. Roughly speaking, these results guarantee that if one chooses an almost classical initial state then for $\hbar \to 0$ its propagation remains close to a classical propagation at any (not too long) later time. The problem arises when one considers a situation in which the initial state of the system is genuinely non-classical , e.g., a superposition state, and nevertheless the system exhibits, at later times, a classical behavior. Examples of this situation are the localization effect in chiral molecules or the suppression of the interference fringes for a heavy particle in a two-slit experiment. It is clear that the emergence of such classical behavior cannot be understood if one insists to consider the limit $\hbar \to 0$ for the isolated quantum system. It is worth mentioning that the problem has some relevance from the conceptual point of view. In fact it was already raised in the earliest debate on the foundation of

Quantum Mechanics (see e.g., [5]). The accepted explanation in these cases is based on the consideration of the interaction of the quantum system with an environment, and in particular on the decoherence effect produced by the environment. It is important to stress that the decoherence effect must be proved in each situation, starting from specific models of system plus environment and introducing suitable assumptions on the parameters of the model. Many results in this direction have been obtained in the physical literature (see e.g., the reviews [13,17] and references therein) but only few mathematical results are available.

Here, we want to focus on a problem of a different kind, raised by Mott [18] in 1929, concerning the explanation of the straight tracks left by an α-particle in a cloud chamber. According to quantum mechanics [8,12] the α-particle, isotropically emitted by a radioactive source, is initially described by a spherical wave function and then interacts with the atoms of the vapor surrounding the radioactive source. The observed tracks are the macroscopic manifestation of ionizations of the atoms induced by the α-particle and "it is a little difficult to picture how it is that an outgoing spherical wave can produce a straight track; we think intuitively that it should ionise atoms at random throughout space" [18]. The explanation proposed by Mott was based on a simple model describing the α-particle in interaction with only two atoms. Exploiting time-independent perturbation arguments, he concluded that the probability that both atoms are ionized is negligible unless the two atoms and the center of the spherical wave lie on the same line. In such rather indirect sense, this would explain why we see straight tracks in the experiments. We will remark on this aspect in Sect. 3. Notice that one typically speaks of tracks due to the decay of an α-particle. A standard theoretical examination of the meaning of tracks left by an α-particle should rely on the reduction postulate where the cloud of vapor is assumed to act as a macroscopic apparatus measuring positions (or better tracks) instantaneously localizing the particle at the place where the ionization takes place.

We mention that the problem is also discussed in [15] and later in [2], and some further elaborations on the subject can be found in [4,6,7,9,14,21].

In this paper, we reconsider the three-particle model of a cloud chamber. Under suitable assumptions on the parameters of the model, which will be specified later, we give a proof of Mott's result through a fully time-dependent analysis and up to second order in perturbation theory. We remark that in our model a crucial assumption is the choice of a semiclassical initial state for the α-particle, i.e., a spherical wave in the short wavelength limit. In this sense our result should be considered as an example of semiclassical analysis in presence of an environment. The method of the proof is rather elementary and it basically relies on stationary and non stationary phase arguments for the estimate of the oscillatory integrals appearing in the perturbative expansion. The work extends to the three-dimensional case the result obtained in [10] for the simpler one-dimensional case, where the spherical wave reduces to the coherent superposition of two wave packets with opposite average momentum.

2. Description of the Model

In this section, we describe the model, i.e., the Hamiltonian, the initial state, the assumptions on the physical parameters, and introduce some notation.

Let us first introduce the Hamiltonian. We consider a three-particle non relativistic, spinless quantum system in dimension three, made of a particle with mass M (the α-particle) and two other particles with mass m which play the role of electrons in two model-atoms with fixed nuclei. More precisely we describe such electrons as particles subject to an attractive point interaction placed at fixed positions $a_1, a_2 \in \mathbb{R}^3$, with $a_1 \neq 0, a_2 \neq 0$, and $a_1 \neq a_2$. Moreover we assume that the interaction between the α-particle and each atom is given by a smooth two-body potential V. We denote by R the position coordinate of the α-particle and by r_1, r_2 the position coordinates of the two electrons. The Hamiltonian of the system in $L^2(\mathbb{R}^9)$ is formally written as

$$H = H_0 + \lambda H_1 \tag{2.1}$$

$$H_0 = K_0 + K_1 + K_2 \tag{2.2}$$

$$H_1 = V(\gamma^{-1}(R - r_1)) + V(\gamma^{-1}(R - r_2)) \tag{2.3}$$

where K_0 denotes the free Hamiltonian for the α-particle

$$K_0 = -\frac{\hbar^2}{2M}\Delta_R, \tag{2.4}$$

$\lambda > 0$ is a coupling constant and $K_j, j = 1, 2$, is the Schrödinger operator in $L^2(\mathbb{R}^3)$ with an attractive point interaction of strength $-(4\pi\gamma)^{-1}, \gamma > 0$, placed at a_j. We recall that the operator K_j is by definition a non trivial selfadjoint extension of the free Hamiltonian restricted on $C_0^\infty(\mathbb{R}^3 \backslash \{a_j\})$. In the Appendix I, (Sect. 7) we collect some basic facts on this kind of Hamiltonians while for a complete treatment we refer to [1]. Here we only specify the spectrum

$$\sigma_p(K_j) = \{E_0\}, \quad E_0 = -\frac{\hbar^2}{2m\gamma^2}, \quad \sigma_c(K_j) = \sigma_{ac}(K_j) = [0, \infty) \tag{2.5}$$

and the proper and generalized eigenfunctions, respectively given by

$$\zeta_j(r) = \frac{1}{\gamma^{3/2}}\zeta^0(\gamma^{-1}(r - a_j)), \quad \zeta^0(x) = \frac{1}{\sqrt{2\pi}}\frac{e^{-|x|}}{|x|} \tag{2.6}$$

$$\phi_j(r, k) = e^{ik\cdot a_j}\phi^0(\gamma^{-1}(r - a_j), \gamma k),$$

$$\phi^0(x, y) = \frac{1}{(2\pi)^{3/2}}\left(e^{iy\cdot x} - \frac{1}{1 - i|y|}\frac{e^{-i|y||x|}}{|x|}\right) \tag{2.7}$$

The parameter γ has the physical meaning of a scattering length and it characterizes the effective range of the point interaction. From (2.6) it is also clear that γ is a measure of the linear spread of the ground state, i.e., of the atoms.

The unperturbed Hamiltonian H_0 is obviously selfadjoint and bounded from below in $L^2(\mathbb{R}^9)$ and moreover the smoothness assumption on the interaction potential V (see Theorems 1, 2) guarantees that the perturbed Hamiltonian H is also selfadjoint and bounded from below on the same domain of H_0 (see e.g., [19]). In particular this implies that the evolution problem associated with the Hamiltonian H is well posed.

We choose the initial state in the product form

$$\Psi_0(R, r_1, r_2) = \psi(R)\zeta_1(r_1)\zeta_2(r_2) \tag{2.8}$$

where ζ_j is defined in (2.6) and $\psi(R)$ is a spherical wave defined as follows. Let us consider a gaussian wave packet localized in space around the origin, with standard deviation γ and mean momentum $P_0 > 0$ along the direction $\hat{u} \in S^2$. Integrating over the unit sphere S^2, one obtains

$$\psi(R) = \frac{\mathcal{N}_\varepsilon}{\varepsilon\gamma^{3/2}} f(\gamma^{-1}R) \int_{S^2} \mathrm{d}\hat{u}\, e^{\frac{i}{\varepsilon}\hat{u}\cdot\frac{R}{\gamma}}, \; f(x) = e^{-\frac{|x|^2}{2}} \tag{2.9}$$

where $\varepsilon > 0$ is the dimensionless parameter

$$\varepsilon \equiv \frac{\hbar}{P_0\gamma} \tag{2.10}$$

and $\mathcal{N}_\varepsilon$ is the normalization factor chosen to guarantee that $\|\psi\|_{L^2(\mathbb{R}^3)} = 1$, i.e.,

$$\mathcal{N}_\varepsilon = \frac{1}{4\pi^{7/4}\left(1 - e^{-\frac{1}{\varepsilon^2}}\right)^{1/2}} \tag{2.11}$$

Notice that $\mathcal{N}_\varepsilon \to (4\pi^{7/4})^{-1}$ for $\varepsilon \to 0$. By an elementary integration we also get

$$\psi(R) = \frac{4\pi\mathcal{N}_\varepsilon}{\gamma^{1/2}} \frac{e^{-\frac{R^2}{2\gamma^2}}}{|R|} \sin(\varepsilon^{-1}\gamma^{-1}|R|) \tag{2.12}$$

We remark that the characteristic length γ appears in the definition of the Hamiltonian as well as in the initial state in such a way that the range of the interaction between the α-particle and the atoms, the linear dimension of the atoms and the localization in space of the spherical wave are all of order γ. As it will become clear further on, this is a crucial ingredient for the proof of our result.

We also notice that the initial state (2.8) coincides with the one considered by Mott. Namely the α-particle is emitted as an outgoing spherical wave and the atoms are in their ground state.

Let us describe the hypotheses on the physical parameters of the model. We assume

$$\varepsilon \ll 1 \tag{2.13}$$

Moreover there are two positive constants $c < C < \infty$, independent of ε, such that

$$c\varepsilon < \frac{\gamma}{|a_j|} < C\varepsilon, \quad j = 1, 2 \tag{2.14}$$

$$c\varepsilon < \frac{m}{M} < C\varepsilon \tag{2.15}$$

Condition (2.13) means that the wavelength $\hbar P_0^{-1}$ associated to the initial state of the α-particle is much smaller than the linear dimension of the atoms and the range of the interaction, which means that we are in a semi-classical regime for the α-particle. In (2.14) we assume that $|a_1|$, $|a_2|$ are macroscopic distances with respect to the characteristic length γ and in (2.15) we require that the mass ratio is small. Finally we tacitly assume

$$c\varepsilon < \lambda_0 \equiv \frac{\lambda}{Mv_0^2} < C\varepsilon \tag{2.16}$$

where $v_0 = P_0 M^{-1}$. Condition (2.16) is necessary in order to make reasonable the application of our perturbative techniques, even if it is not strictly required for the proof of our results. The above assumptions (2.13), (2.14), (2.15) have some relevant physical implications. In particular from (2.13), (2.15) one sees that the binding energy of the atoms is small compared to the kinetic energy of the α-particle

$$\frac{2|E_0|}{Mv_0^2} = \frac{M}{m}\left(\frac{\hbar}{P_0\gamma}\right)^2 \equiv \frac{M}{m}\varepsilon^2 \tag{2.17}$$

Furthermore the assumptions (2.13) and (2.14) imply two relations among the characteristic times of the system which will be relevant in what follows. In particular, we define the flight times to the atoms of the α-particle

$$\tau_j = \frac{|a_j|}{v_0}, \quad j = 1, 2 \tag{2.18}$$

the characteristic "period" of the atoms

$$T_a = 2\pi\frac{\hbar}{|E_0|} = 4\pi\frac{m\gamma^2}{\hbar} \tag{2.19}$$

and the transit time of the α-particle in the region where the atom are localized

$$T_t = \frac{\gamma}{v_0} \tag{2.20}$$

Then one has

$$\frac{T_t}{\tau_j} = \frac{\gamma}{|a_j|}, \quad j = 1, 2 \tag{2.21}$$

$$\frac{T_t}{T_a} = \frac{1}{4\pi}\frac{M}{m}\frac{\hbar}{P_0\gamma} \equiv \frac{1}{4\pi}\frac{M}{m}\varepsilon \tag{2.22}$$

i.e., the transit time T_t is small with respect to the flight times τ_j but it is comparable with the characteristic period of the atoms T_a. This means that the α-particle can "see" the internal structure of the atoms.

Let us introduce some notation to streamline the presentation.

$$\hat{a}_j = \frac{a_j}{|a_j|}, \quad j = 1, 2 \tag{2.23}$$

$$\omega(y) = \frac{1}{2}(1 + y^2), \quad y \in \mathbb{R}^3 \tag{2.24}$$

$$\mathfrak{a} = \frac{\hbar t}{M\gamma^2}, \quad \mathfrak{b}_j = \frac{\hbar \tau_j}{M\gamma^2}, \quad \mathfrak{c}_j = \frac{\hbar^2 t}{P_0 \gamma m \gamma^2}\omega(y_j), \quad y_j \in \mathbb{R}^3, \quad j = 1, 2 \tag{2.25}$$

$$h(\xi, y) = \frac{1}{(2\pi)^{3/2}} \int \mathrm{d}x\, e^{-i\xi\cdot x}\overline{\phi^0}(x, y)\zeta^0(x), \quad \xi, y \in \mathbb{R}^3 \tag{2.26}$$

$$g(\xi, y) = \widetilde{V}(\xi)h(\xi, y) \tag{2.27}$$

where

$$\tilde{F}(q) = \frac{1}{(2\pi)^{3/2}} \int \mathrm{d}x\, e^{-iq\cdot x}F(x) \tag{2.28}$$

denotes the Fourier transform of F. Moreover, for $n, m \in \mathbb{N}$, we denote

$$\|u\|_{W^{1,n}_m} = \sum_{0 \le |\alpha| \le n} \|{<}x{>}^m D^\alpha u\|_{L^1(\mathbb{R}^3)} \tag{2.29}$$

where ${<}x{>}^2 = 1 + x^2, x = (x_1, x_2, x_3) \in \mathbb{R}^3, \alpha = (\alpha_1, \alpha_2, \alpha_3) \in \mathbb{N}^3, |\alpha| = \alpha_1 + \alpha_2 + \alpha_3$ and

$$D^\alpha u = \frac{\partial^{\alpha_1}}{\partial x_1^{\alpha_1}} \frac{\partial^{\alpha_2}}{\partial x_2^{\alpha_2}} \frac{\partial^{\alpha_3}}{\partial x_3^{\alpha_3}} u \tag{2.30}$$

Finally $\mathcal{C}_k$ denotes a positive numerical constant, depending on $k \in \mathbb{N}$ and, possibly, on the dimensionless parameters (2.25).

It is important to notice that, for any fixed $t > \tau_2$ and $y_j \in \mathbb{R}^3$, the quantities $\mathfrak{a}, \mathfrak{b}_j, \mathfrak{c}_j$ are all of order one. In fact it is sufficient to notice that

$$\mathfrak{a} = \frac{\hbar}{P_0\gamma} \frac{|a_2|}{\gamma} \frac{t}{\tau_2}, \quad \mathfrak{b}_j = \frac{\hbar}{P_0\gamma} \frac{|a_j|}{\gamma}, \quad \mathfrak{c}_j = \frac{M}{m} \left(\frac{\hbar}{P_0\gamma}\right)^2 \frac{|a_2|}{\gamma} \frac{t}{\tau_2}\omega(y_j) \tag{2.31}$$

and to use (2.13), (2.14), and (2.15).

3. Result

We are now in a position to formulate our result. We are interested in the computation of the probability that both atoms are ionized at time $t > 0$. An exact computation obviously requires the complete knowledge of the state $\Psi(t)$ of the system, which is not available. Following the original strategy of Mott we shall limit to consider the second order approximation $\Psi_2(t)$ of the state $\Psi(t)$ which, iterating twice Duhamel's formula, is given by

$$\Psi_2(t) = e^{-\frac{i}{\hbar}tH_0}\hat{\Psi}_2(t) \tag{3.1}$$

$$\hat{\Psi}_2(t) = \Psi_0 - i\frac{\lambda}{\hbar}\int_0^t \mathrm{d}t_1 e^{\frac{i}{\hbar}t_1 H_0} H_1 e^{-\frac{i}{\hbar}t_1 H_0}\Psi_0$$
$$-\frac{\lambda^2}{\hbar^2}\int_0^t \mathrm{d}t_1 e^{\frac{i}{\hbar}t_1 H_0} H_1 e^{-\frac{i}{\hbar}t_1 H_0}\int_0^{t_1} \mathrm{d}t_2\, e^{\frac{i}{\hbar}t_2 H_0} H_1 e^{-\frac{i}{\hbar}t_2 H_0}\Psi_0 \tag{3.2}$$

Therefore we shall study the probability that both atoms are ionized up to second order in perturbation theory, i.e.

$$\mathcal{P}(t) = \int \mathrm{d}R\mathrm{d}k_1\mathrm{d}k_2 \left| \int \mathrm{d}r_1\mathrm{d}r_2 \overline{\phi_1}(r_1,k_1)\overline{\phi_2}(r_2,k_2)\hat{\Psi}_2(R,r_1,r_2,t)\right|^2 \tag{3.3}$$

Our main result is the characterization of the ionization probability $\mathcal{P}(t)$ for a fixed time $t > \tau_2$ and it is summarized in Theorems 1 and 2 below. In Theorem 1 we consider the case in which a_2 is not aligned with a_1 and the origin and we show that the ionization probability decays faster than any power of ε.

Theorem 1. *Let us fix $t > \tau_2, |a_1| < |a_2|, \hat{a}_1 \cdot \hat{a}_2 < 1$ and let us assume (2.13), (2.14), and (2.15), $V \in \mathcal{S}(\mathbb{R}^3)$. Then for any $k \in \mathbb{N}$ there exists $\mathcal{C}_k > 0$ such that*

$$\mathcal{P}(t) \le \left(\frac{\lambda t}{\hbar}\right)^4 \mathcal{N}_\varepsilon^2 2\mathcal{C}_k \|\widetilde{V}\|^4_{W_k^{1,k}} \left[\left(1 - \frac{|a_1|}{|a_2|}\right)^{-2k} + (1 - \hat{a}_1\cdot\hat{a}_2)^{-k}\right]\varepsilon^{2k-2} \tag{3.4}$$

We notice that the estimate (3.4) is still meaningful (for some values of k) if the angle between a_1 and a_2 is of order ε^p with $0 < p < 1$, while it gives no information for $p \ge 1$. In the latter case the second atom lies inside a small cone of aperture proportional to ε, apex in the position a_1 of the first atom and axis on the line joining the origin and a_1. This situation is considered in Theorem 2 where we compute the leading term of the asymptotic expansion for $\varepsilon \to 0$ of the ionization probability. We explicitly mention that we do not consider some intermediate situations, like the case of an angle proportional to $\varepsilon|\log\varepsilon|$.

Theorem 2. *Let us fix $t > \tau_2, |a_1| < |a_2|, \hat{a}_1 \cdot \hat{a}_2 = \cos\chi_\varepsilon$, where $\chi_\varepsilon \in [0, \chi_0\varepsilon], \chi_0 > 0$, and let us assume (2.13), (2.14), and (2.15), $V \in \mathcal{S}(\mathbb{R}^3)$. Then, at the leading order for $\varepsilon \to 0$, we have*

$$\mathcal{P}(t) \sim \varepsilon^2(\varepsilon^{-1}\lambda_0)^4\left(\varepsilon^{-1}\frac{\gamma}{|a_1|}\right)^4 \mathcal{N}_\varepsilon^2 \int_{\mathbb{R}^9} \mathrm{d}x\mathrm{d}y\mathrm{d}z \left|\int_{\mathbb{R}^2} \mathrm{d}\eta_1\mathrm{d}\eta_2 F(\eta_1,\eta_2;x,y,z)\right|^2 \tag{3.5}$$

where the function F is independent of ε and will be specified during the proof.

In the following remarks we briefly comment on the above results.

Remark 3.1. The estimate (3.4) is valid for t larger but of the same order of τ_2, while it loses its meaning for $t \to \infty$. This is only due to the method we use for the proof, based on second order perturbation theory. A non-perturbative approach or a more detailed perturbative analysis should provide an estimate which is uniform in time. We also remark that in Theorem 2 we limit ourselves to the computation of the leading term, without making any attempt to estimate the remainder. Such leading term is small for $\varepsilon \to 0$ being proportional, as expected, to the solid angle that the atoms subtend at the origin. It would be interesting to extend the results given here at any order in perturbation theory and in particular to verify that the leading term has the same behavior for $\varepsilon \to 0$ at all orders.

Remark 3.2. We recall that the normalization factor $\mathcal{N}_\varepsilon$ goes to $(4\pi^{7/4})^{-1}$ for $\varepsilon \to 0$ (see (2.11)). Moreover from (2.10), (2.14), and (2.16) one has that

$$\left(\frac{\lambda t}{\hbar}\right)^4 = \left(\frac{\lambda_0}{\varepsilon}\frac{|a_2|}{\gamma}\frac{t}{\tau_2}\right)^4 \tag{3.6}$$

is proportional to ε^{-4} for any t larger but of the same order of τ_2. This means that it is sufficient to take any integer $k > 4$ in (3.4) to conclude that the ionization probability estimated in Theorem 1 is much smaller than the one computed in Theorem 2. We underline this point since it allows to understand the results in Theorems 1, and 2 on the basis of the original physical argument given by Mott, which can be described as follows. At time zero the spherical wave starts to propagate in the chamber and at time τ_1 it interacts with the atom in a_1. If, as result of the interaction, such atom is ionized then a localized wave packet emerges from a_1 with momentum along the direction $\overline{Oa_1}$. In order to obtain also ionization of the atom in a_2 the localized wave packet must hit the atom in a_2 (at time τ_2) and this can happen only if a_2 approximately lies on the line $\overline{Oa_1}$. It should be stressed that such physical behavior is far from being universal and it strongly depends on our assumptions on the physical parameters of the model.

Remark 3.3. Finally we observe that our result states that one can only observe straight tracks in a cloud chamber. With this we do not mean that there is any focusing of the support of the wave packet of the α-particle along a classical straight trajectory, corresponding to the observed track. In fact the solution of the Schrödinger equation with Hamiltonian (2.1) and initial datum (2.8) has the form

$$\begin{aligned}\Psi(R,r_1,r_2,t) = \mathcal{F}_{00}(R,t)\zeta_1(r_1)\zeta_2(r_2) &+ \int \mathrm{d}k_1 \mathcal{F}_{c0}(R,k_1,t)\phi_1(r_1,k_1)\cdot\zeta_2(r_2)\\ &+ \int \mathrm{d}k_2 \mathcal{F}_{0c}(R,k_2,t)\phi_2(r_2,k_2)\cdot\zeta_1(r_1)\\ &+ \int \mathrm{d}k_1\mathrm{d}k_2 \mathcal{F}_{cc}(R,k_1,k_2,t)\phi_1(r_1,k_1)\phi_2(r_2,k_2)\end{aligned} \tag{3.7}$$

where the four probability amplitudes $\mathcal{F}_{00}, \mathcal{F}_{c0}, \mathcal{F}_{0c}, \mathcal{F}_{cc}$ are localized in different regions of the configuration space of the whole system and therefore describe not interfering "quantum histories". If one interprets double ionization as the only case of macroscopic ionization, giving then rise to an observable track, one can say, in a pictorial language, that is along the track the expected value of the position of the α particle in the states in which the track (as an observable) has expectation one.

Let us outline the strategy of the proof of Theorems 1 and 2. The starting point is the following, more convenient, representation formula for the ionization probability

$$\mathcal{P}(t) = \frac{\lambda^4 t^4}{\hbar^4}\frac{\mathcal{N}_\varepsilon^2}{\varepsilon^2}\int \mathrm{d}x\mathrm{d}y_1\mathrm{d}y_2 \left|\mathcal{G}^\varepsilon_{12}(x,y_1,y_2,t) + \mathcal{G}^\varepsilon_{21}(x,y_1,y_2,t)\right|^2 \tag{3.8}$$

where for $l, j = 1, 2, j \neq l$ one has

$$\mathcal{G}^\varepsilon_{lj}(x,y_1,y_2,t) = \int_{S^2}\mathrm{d}\hat{u}\int_0^1 \mathrm{d}\alpha\int_0^\alpha \mathrm{d}\beta\int \mathrm{d}\eta\mathrm{d}\xi G_{lj}(\alpha,\beta,\eta,\xi;x,y_1,y_2,t)e^{\frac{i}{\varepsilon}\Theta_{lj}(\hat{u},\alpha,\beta,\eta,\xi;x,y_1,y_2,t)} \tag{3.9}$$

and dropping the parametric dependence on x, y_1, y_2, t

$$\Theta_{lj}(\hat{u},\alpha,\beta,\eta,\xi) = \hat{u}\cdot(x+\mathfrak{a}(\alpha\eta+\beta\xi)) - \mathfrak{b}_j\hat{a}_j\cdot\eta - \mathfrak{b}_l\hat{a}_l\cdot\xi + \mathfrak{c}_j\alpha + \mathfrak{c}_l\beta \tag{3.10}$$

$$G_{lj}(\alpha,\beta,\eta,\xi) = g(\eta,y_j)g(\xi,y_l)f(x+\mathfrak{a}(\alpha\eta+\beta\xi))e^{i\phi(\alpha,\beta,\eta,\xi)} \tag{3.11}$$

$$\phi(\alpha,\beta,\eta,\xi) = x\cdot(\eta+\xi) + \frac{\mathfrak{a}}{2}(\alpha\eta^2+\beta\xi^2+2\alpha\eta\cdot\xi) \tag{3.12}$$

In (3.8) and (3.9), we have denoted by x, y_1, y_2 the rescaled position of the α-particle and the rescaled momenta of the electrons respectively, while α, β play the role of rescaled time variables. Moreover we recall that f is the gaussian defined in (2.9).

The proof of (3.8) is a long but straightforward computation and it is postponed to the Appendix II (Section 8).

Due to formula (3.8), we are reduced to the analysis of the two oscillatory integrals $\mathcal{G}^\varepsilon_{lj}$ corresponding to the possible graphs in the second order perturbative expansion. In particular $\mathcal{G}^\varepsilon_{12}$ describes the graph in which the atom in a_1 is ionized before the atom in a_2 and $\mathcal{G}^\varepsilon_{21}$ the opposite case. Since we always assume $|a_1| < |a_2|$, we expect that the contribution of $\mathcal{G}^\varepsilon_{21}$ is negligible. In fact, in Sect. 4 we shall see that the phase Θ_{21} has no critical points and then, by standard integration by parts, we shall prove that the contribution of the oscillatory integral $\mathcal{G}^\varepsilon_{21}$ is bounded by a constant times ε^k, for any $k \in \mathbb{N}$.

The estimate of the term $\mathcal{G}^\varepsilon_{12}$ is more delicate and we have to distinguish the non aligned and the aligned case. It turns out that the phase Θ_{12} has no critical points in the first case and then the contribution of $\mathcal{G}^\varepsilon_{12}$ is bounded by a constant times ε^k, for any $k \in \mathbb{N}$. This will be proved in Sect. 5, concluding also the proof of Theorem 1.

In Sect. 6, we consider the aligned case, where the phase Θ_{12} has a manifold of critical points parametrized by a vector in $\mathbb{R}^2$. By a careful application of the stationary phase method to $\mathcal{G}^{\varepsilon}_{12}$, we compute the leading term of the asymptotic expansion for $\varepsilon \to 0$ and then we also conclude the proof of theorem 2.

4. Estimate of $\mathcal{G}^{\varepsilon}_{21}$

In this section, we shall prove that the the contribution of the oscillatory integral $\mathcal{G}^{\varepsilon}_{21}$ is negligible for any orientation of the unit vectors $\hat{a}_1, \hat{a}_2$.

Proposition 4.1. *Let us fix $t > \tau_2, |a_1| < |a_2|$ and let us assume* (2.13), (2.14), (2.15)*, $V \in \mathcal{S}(\mathbb{R}^3)$. Then for any $k \in \mathbb{N}$ there exists $\mathcal{C}_k > 0$ such that*

$$\int dx\, dy_1\, dy_2\, |\mathcal{G}^{\varepsilon}_{21}(x, y_1, y_2)|^2 \leq \mathcal{C}_k \|\widetilde{V}\|^4_{W^{1,k}_k} \left(1 - \frac{|a_1|}{|a_2|}\right)^{-2k} \varepsilon^{2k} \tag{4.1}$$

Proof. The crucial point is that the gradient of the phase

$$\Theta_{21} = \hat{u} \cdot (x + \mathfrak{a}(\alpha\eta + \beta\xi)) - \mathfrak{b}_1 \hat{a}_1 \cdot \eta - \mathfrak{b}_2 \hat{a}_2 \cdot \xi + \mathfrak{c}_1 \alpha + \mathfrak{c}_2 \beta \tag{4.2}$$

does not vanish in the integration region. To see this it is sufficient to compute

$$\begin{aligned} \sum_{k=1}^{3} \left[\left(\frac{\partial \Theta_{21}}{\partial \eta_k}\right)^2 + \left(\frac{\partial \Theta_{21}}{\partial \xi_k}\right)^2 \right] &= (\mathfrak{a}\alpha\hat{u} - \mathfrak{b}_1\hat{a}_1)^2 + (\mathfrak{a}\beta\hat{u} - \mathfrak{b}_2\hat{a}_2)^2 \\ &\geq (\mathfrak{a}\alpha - \mathfrak{b}_1)^2 + (\mathfrak{a}\beta - \mathfrak{b}_2)^2 \\ &\equiv \mathfrak{a}^2 \left[\left(\alpha - \frac{\tau_1}{t}\right)^2 + \left(\beta - \frac{\tau_2}{t}\right)^2 \right] \end{aligned} \tag{4.3}$$

In the region $\{(\alpha, \beta) \in \mathbb{R}^2 | 0 \leq \alpha \leq 1, 0 \leq \beta \leq \alpha\}$ the r.h.s. of (4.3) takes its minimum in (α_0, β_0), with $\alpha_0 = \beta_0 = \frac{\tau_1 + \tau_2}{2t}$, then

$$\sum_{k=1}^{3} \left[\left(\frac{\partial \Theta_{21}}{\partial \eta_k}\right)^2 + \left(\frac{\partial \Theta_{21}}{\partial \xi_k}\right)^2 \right] \geq \Delta^2_{21} \tag{4.4}$$

where

$$\Delta_{21} = \frac{\hbar}{\sqrt{2} M \gamma^2}(\tau_2 - \tau_1) \equiv \frac{\hbar}{\sqrt{2} P_0 \gamma} \frac{|a_2|}{\gamma} \left(1 - \frac{|a_1|}{|a_2|}\right) \tag{4.5}$$

Notice that, under the assumptions (2.13), (2.14), (2.15) and $|a_1| < |a_2|$, there exists a positive constant c_0, independent of ε, such that $\Delta_{21} > c_0$ for any $\varepsilon > 0$. The estimate (4.4) allows to control $\mathcal{G}_{21}$ using standard non stationary phase methods [3,11,16]. In fact, recalling the identity

$$a e^{ib} = -i div \left(e^{ib} \frac{\nabla b}{|\nabla b|^2} a\right) + i e^{ib} div \left(\frac{\nabla b}{|\nabla b|^2} a\right) \tag{4.6}$$

and performing k integration by parts we have

$$\int \mathrm{d}\eta \mathrm{d}\xi G_{21} e^{\frac{i}{\varepsilon}\Theta_{21}} = (i\varepsilon)^k \int \mathrm{d}\eta \mathrm{d}\xi (L^k G_{21}) e^{\frac{i}{\varepsilon}\Theta_{21}} \tag{4.7}$$

where the operator L acts on the variables $\zeta = (\zeta_1, \ldots, \zeta_6) \equiv (\eta_1, \eta_2, \eta_3, \xi_1, \xi_2, \xi_3)$ as follows

$$LG_{21} = \sum_{j=1}^{6} u_j \frac{\partial G_{21}}{\partial \zeta_j}, \quad u_j = \frac{1}{|\nabla_\zeta \Theta_{21}|^2} \frac{\partial \Theta_{21}}{\partial \zeta_j} \tag{4.8}$$

and moreover

$$L^k G_{21} = \sum_{j_1,\ldots,j_k=1}^{6} u_{j_1}, \ldots, u_{j_k} D^k_{\zeta_{j_1},\ldots,\zeta_{j_k}} G_{21} \tag{4.9}$$

In (4.9) we have denoted by $D^k_{\zeta_{j_1},\ldots,\zeta_{j_k}}$ the derivative of order k with respect to $\zeta_{j_1}, \ldots, \zeta_{j_k}$. From (4.9), (4.8), (4.4), (4.5) we easily get the estimate

$$\begin{aligned} |\mathcal{G}^\varepsilon_{21}| &\le \varepsilon^k \int_{S^2} \mathrm{d}\hat{u} \int_0^1 \mathrm{d}\alpha \int_0^\alpha \mathrm{d}\beta \int \mathrm{d}\eta \mathrm{d}\xi \left| L^k G_{21} \right| \\ &\le 4\pi \frac{\varepsilon^k}{\Delta^k_{21}} \int_0^1 \mathrm{d}\alpha \int_0^\alpha \mathrm{d}\beta \int \mathrm{d}\eta \mathrm{d}\xi \left| \sum_{j_1,\ldots,j_k=1}^{6} D^k_{\zeta_{j_1},\ldots,\zeta_{j_k}} G_{21} \right| \end{aligned} \tag{4.10}$$

If we square (4.10), integrate w.r.t. the variables x, y_1, y_2 and use Schwartz inequality we find

$$\begin{aligned} &\int \mathrm{d}x \mathrm{d}y_1 \mathrm{d}y_2 |\mathcal{G}^\varepsilon_{21}|^2 \\ &\le 4\pi^2 \frac{\varepsilon^{2k}}{\Delta^{2k}_{21}} \sup_{\alpha,\beta} \left[\int \mathrm{d}\eta \mathrm{d}\xi \left(\int \mathrm{d}x \mathrm{d}y_1 \mathrm{d}y_2 \left| \sum_{j_1,\ldots,j_k=1}^{6} D^k_{\zeta_{j_1},\ldots,\zeta_{j_k}} G_{21} \right|^2 \right)^{1/2} \right]^2 \end{aligned} \tag{4.11}$$

From the definition of G_{21} (see (3.11)), we have

$$\begin{aligned} &\sum_{j_1,\ldots,j_k=1}^{6} |D^k_{\zeta_{j_1},\ldots,\zeta_{j_k}} G_{21}| \\ &\le \mathcal{C}_k \sum_{i_1=1}^{k} |D^{i_1}_\eta g(\eta, y_1)| \sum_{i_2=1}^{k} |D^{i_2}_\xi g(\xi, y_2)| \\ &\quad \cdot \sum_{i_3=1}^{k} \left| D^{i_3}_x f(x + \mathfrak{a}(\alpha\eta + \beta\xi)) \right| \sum_{i_4=1}^{k} (|x| + \mathfrak{a}|\eta| + \mathfrak{a}|\xi|)^{i_4} \end{aligned} \tag{4.12}$$

The last term in (4.12) can be easily estimated as follows

$$\begin{aligned}
&\sum_{i_4=1}^{k}(|x|+\mathfrak{a}|\eta|+\mathfrak{a}|\xi|)^{i_4}\\
&\quad\le \sum_{i_4=1}^{k}(|x+\mathfrak{a}(\alpha\eta+\beta\xi)|+2\mathfrak{a}|\eta|+2\mathfrak{a}|\xi|)^{i_4}\\
&\quad\le \sum_{i_4=1}^{k}\left(\sqrt{2}4\mathfrak{a}^2<x+\mathfrak{a}(\alpha\eta+\beta\xi)><\eta><\xi>\right)^{i_4}\\
&\quad\le \left(\sum_{i_4=1}^{k}\mathfrak{a}^{2i_4}2^{5i_4/2}\right)<x+\mathfrak{a}(\alpha\eta+\beta\xi)>^k<\eta>^k<\xi>^k \qquad (4.13)
\end{aligned}$$

where $<x>^2=1+x^2, x\in\mathbb{R}^3$. Hence

$$\begin{aligned}
\sum_{j_1,\dots,j_k=1}^{6}\left|D^k_{\zeta_{j_1},\dots,\zeta_{j_k}}G_{21}\right| &\le \mathcal{C}_k<\eta>^k\sum_{i_1=1}^{k}\left|D^{i_1}_\eta g(\eta,y_1)\right|<\xi>^k\\
&\cdot\sum_{i_2=1}^{k}\left|D^{i_2}_\xi g(\xi,y_2)\right|<x+\mathfrak{a}(\alpha\eta+\beta\xi)>^k\sum_{i_3=1}^{k}\left|D^{i_3}_x f(x+\mathfrak{a}(\alpha\eta+\beta\xi))\right| \quad (4.14)
\end{aligned}$$

Moreover, recalling the definition (2.27), we get

$$\begin{aligned}
\sum_{j_1,\dots,j_k=1}^{6}\left|D^k_{\zeta_{j_1},\dots,\zeta_{j_k}}G_{21}\right| \le \mathcal{C}_k<\eta>^k\sum_{i_1=1}^{k}\left|D^{i_1}_\eta\widetilde{V}(\eta)\right|<\xi>^k\sum_{i_2=1}^{k}\left|D^{i_2}_\xi\widetilde{V}(\xi)\right|\\
\cdot<x+\mathfrak{a}(\alpha\eta+\beta\xi)>^k\sum_{i_3=1}^{k}\left|D^{i_3}_x f(x+\mathfrak{a}(\alpha\eta+\beta\xi))\right|\\
\times\sum_{i_4=1}^{k}\left|D^{i_4}_\eta h(\eta,y_1)\right|\sum_{i_5=1}^{k}\left|D^{i_5}_\xi h(\xi,y_2)\right| \qquad (4.15)
\end{aligned}$$

Using (4.15) in estimate (4.11) we find

$$\begin{aligned}
&\int \mathrm{d}x\mathrm{d}y_1\mathrm{d}y_2|\mathcal{G}^\varepsilon_{21}|^2\\
&\quad\le \frac{\varepsilon^{2k}}{\Delta^{2k}_{21}}\mathcal{C}_k\left\{\int \mathrm{d}\eta<\eta>^k\sum_{i_1=1}^{k}\left|D^{i_1}_\eta\widetilde{V}(\eta)\right|\int \mathrm{d}\xi<\xi>^k\right.\\
&\quad\cdot\sum_{i_2=1}^{k}|D^{i_2}_\xi\widetilde{V}(\xi)|\left[\int dy_1\left(\sum_{i_3=1}^{k}\left|D^{i_3}_\eta h(\eta,y_1)\right|\right)^2\right.\\
&\quad\left.\left.\times\int \mathrm{d}y_2\left(\sum_{i_4=1}^{k}\left|D^{i_4}_\xi h(\xi,y_2)\right|\right)^2\right]^{1/2}\right\}^2
\end{aligned}$$

$$\leq \frac{\varepsilon^{2k}}{\Delta_{21}^{2k}} \mathcal{C}_k \|\widetilde{V}\|_{W_k^{1,k}}^4 \left[\sup_\eta \int \mathrm{d}y \left(\sum_{m=1}^{k} |D_\eta^m h(\eta, y)| \right)^2 \right]^2$$

$$\leq \frac{\varepsilon^{2k}}{\Delta_{21}^{2k}} \mathcal{C}_k \|\widetilde{V}\|_{W_k^{1,k}}^4 \left[\sum_{m=1}^{k} \sup_\eta \left(\int \mathrm{d}y \left| D_\eta^m h(\eta, y) \right|^2 \right)^{1/2} \right]^4 \tag{4.16}$$

It remains to show that the last term in the r.h.s. of (4.16) is finite. From the definition (2.26) we have

$$D_\eta^m h(\eta, y) = \frac{(-i)^m}{(2\pi)^{3/2}} \int \mathrm{d}x e^{-i\eta\cdot x} x_1^{m_1} x_2^{m_2} x_3^{m_3} \zeta_0(x) \overline{\phi^0}(x, y) \tag{4.17}$$

where $m_1 + m_2 + m_3 = m$. The integral kernel $\overline{\phi^0}(x, y)$ defines a bounded operator in $L^2(\mathbb{R}^3)$, with norm less or equal to one. This fact directly follows from (7.9), in Appendix I. Hence

$$\int \mathrm{d}y \left| D_\eta^m h(\eta, y) \right|^2 \leq \frac{1}{(2\pi)^3} \int \mathrm{d}x \left| x_1^{m_1} x_2^{m_2} x_3^{m_3} \zeta_0(x) \right|^2 < \infty \tag{4.18}$$

Taking into account inequality (4.18)) in (4.16), we conclude the proof. □

5. Estimate of $\mathcal{G}_{12}^\varepsilon$ in the Case $\hat{a}_1 \cdot \hat{a}_2 < 1$

Following the same line of the previous section, we prove that the contribution of $\mathcal{G}_{12}^\varepsilon$ to the ionization probability is negligible provided that the two unit vectors $\hat{a}_1$ and $\hat{a}_2$ are not parallel. Of course, the estimate shall crucially depend on the angle between the two unit vectors.

Proposition 5.1. *Let us fix $t > \tau_2, \hat{a}_1 \cdot \hat{a}_2 < 1$ and let us assume* (2.13), (2.14), (2.15), *$V \in \mathcal{S}(\mathbb{R}^3)$. Then for any $k \in \mathbb{N}$ there exists a strictly positive constant $\mathcal{C}_k$ such that*

$$\int \mathrm{d}x \mathrm{d}y_1 \mathrm{d}y_2 \left| \mathcal{G}_{12}^\varepsilon(x, y_1, y_2) \right|^2 \leq \mathcal{C}_k \|\widetilde{V}\|_{W_k^{1,k}}^4 \left(1 - \hat{a}_1 \cdot \hat{a}_2\right)^{-k} \varepsilon^{2k} \tag{5.1}$$

Proof. As in the case of Proposition 4.1, we consider the phase

$$\Theta_{12} = \hat{u} \cdot (x + \mathfrak{a}(\alpha\eta + \beta\xi)) - \mathfrak{b}_2 \hat{a}_2 \cdot \eta - \mathfrak{b}_1 \hat{a}_1 \cdot \xi + \mathfrak{c}_2 \alpha + \mathfrak{c}_1 \beta \tag{5.2}$$

and we show that its gradient is strictly different from zero in the integration region. In fact

$$\sum_{k=1}^{3} \left[\left(\frac{\partial \Theta_{12}}{\partial \eta_k} \right)^2 + \left(\frac{\partial \Theta_{12}}{\partial \xi_k} \right)^2 \right] = (\mathfrak{a}\alpha\hat{u} - \mathfrak{b}_2 \hat{a}_2)^2 + (\mathfrak{a}\beta\hat{u} - \mathfrak{b}_1 \hat{a}_1)^2 \tag{5.3}$$

The r.h.s. of (5.3), considered as a function of the variables $(\alpha \geq 0, \beta \geq 0)$, takes its minimum in $(\alpha_1, \beta_1) = (\frac{\mathfrak{b}_2}{\mathfrak{a}} \hat{u} \cdot \hat{a}_2, \frac{\mathfrak{b}_1}{\mathfrak{a}} \hat{u} \cdot \hat{a}_1)$ when $\hat{u} \cdot \hat{a}_2 \geq 0, \hat{u} \cdot \hat{a}_1 \geq 0$ and in $\alpha = 0$ and/or $\beta = 0$ when $\hat{u} \cdot \hat{a}_2 < 0, \hat{u} \cdot \hat{a}_1 < 0$, respectively. Hence if

$\hat{u} \cdot \hat{a}_i < 0$ for at least one i then the r.h.s. of (5.3) is larger than $\min\{\mathfrak{b}_1^2, \mathfrak{b}_2^2\}$. Let us consider the case $\hat{u} \cdot \hat{a}_1 \geq 0, \hat{u} \cdot \hat{a}_2 \geq 0$. We have

$$\sum_{k=1}^{3} \left[\left(\frac{\partial \Theta_{12}}{\partial \eta_k} \right)^2 + \left(\frac{\partial \Theta_{12}}{\partial \xi_k} \right)^2 \right] \geq \mathfrak{b}_2^2 (\hat{u} \cdot \hat{a}_2 \hat{u} - \hat{a}_2)^2 + \mathfrak{b}_1^2 (\hat{u} \cdot \hat{a}_1 \hat{u} - \hat{a}_1)^2$$
$$\begin{cases} \geq \min\{\mathfrak{b}_1^2, \mathfrak{b}_2^2\} \left[2 - (\hat{u} \cdot \hat{a}_2)^2 - (\hat{u} \cdot \hat{a}_1)^2 \right] & \text{if } \hat{u} \cdot \hat{a}_2 \geq \frac{1}{\sqrt{2}}, \hat{u} \cdot \hat{a}_1 \geq \frac{1}{\sqrt{2}} \\ \geq \min\{\mathfrak{b}_1^2, \mathfrak{b}_2^2\} \frac{1}{2} \quad \text{if } \hat{u} \cdot \hat{a}_i < \frac{1}{\sqrt{2}} & \text{for at least one } i \end{cases} \tag{5.4}$$

Let us denote $\theta_i = \arccos \hat{u} \cdot \hat{a}_i$. From the convexity of $\cos^2 x$ for $x \in [0, \pi/4]$ it follows that

$$\frac{1}{2} \cos^2 \theta_1 + \frac{1}{2} \cos^2 \theta_2 \leq \cos^2 \frac{\theta_1 + \theta_2}{2} = \frac{1}{2} + \frac{1}{2} \cos(\theta_1 + \theta_2) \leq \frac{1}{2}(1 + \hat{a}_1 \cdot \hat{a}_2) \tag{5.5}$$

Then for $\hat{u} \cdot \hat{a}_2 \geq \frac{1}{\sqrt{2}}, \hat{u} \cdot \hat{a}_1 \geq \frac{1}{\sqrt{2}}$ we have

$$2 - (\hat{u} \cdot \hat{a}_2)^2 - (\hat{u} \cdot \hat{a}_1)^2 \geq 1 - \hat{a}_1 \cdot \hat{a}_2 \tag{5.6}$$

which implies

$$\sum_{k=1}^{3} \left[\left(\frac{\partial \Theta_{12}}{\partial \eta_k} \right)^2 + \left(\frac{\partial \Theta_{12}}{\partial \xi_k} \right)^2 \right] \geq \Delta_{12}^2 \tag{5.7}$$

where

$$\Delta_{12} = \frac{1}{\sqrt{2}} \min\{\mathfrak{b}_1, \mathfrak{b}_2\} \sqrt{1 - \hat{a}_1 \cdot \hat{a}_2} = \frac{\hbar}{\sqrt{2} P_0 \gamma} \frac{\min\{|a_1|, |a_2|\}}{\gamma} \sqrt{1 - \hat{a}_1 \cdot \hat{a}_2} \tag{5.8}$$

We notice that, under the assumptions (2.13), (2.14), (2.15) and $\hat{a}_1 \cdot \hat{a}_2 < 1$, the square modulus of the phase gradient remains strictly larger than zero.

From now on the proof proceeds exactly in the same way as in the previous Proposition 4.1 and we omit the details. □

Proof of Theorem 1. From (3.8) and taking into account Propositions 4.1, 5.1 we immediately get the proof. □

Remark 5.1. The same kind of estimate proved in Proposition 5.1 is also valid for $\mathcal{G}_{21}^{\varepsilon}$ which means that Theorem 1 could be proved without the assumption $|a_1| < |a_2|$. On the other hand the estimate given in Proposition 4.1 is crucial for the analysis of the stationary case of the next section.

6. The Stationary Case

Here, we consider the case $\hat{a}_1 \cdot \hat{a}_2 = 1 - O(\varepsilon^q)$, with $q \geq 2$. We shall see that the phase of the oscillatory integral $\mathcal{G}^{\varepsilon}_{12}$ has stationary points when exactly this case occurs. This implies that the ionization probability $\mathcal{P}(t)$ is not negligible for ε small as in the previous situation and the leading term of its asymptotic expansion in powers of ε can be computed.

Throughout this section we shall fix the unit vectors $\hat{a}_1, \hat{a}_2$ as follows

$$\hat{a}_1 = (0,0,1), \quad \hat{a}_2 = (\sin\chi_\varepsilon, 0, \cos\chi_\varepsilon) \tag{6.1}$$

where $\chi_\varepsilon \in [0, \chi_0\varepsilon], \chi_0 > 0$. Moreover, in order to characterize the asymptotic behavior of $\mathcal{G}^{\varepsilon}_{12}$, we introduce a convenient decomposition of the unit sphere S^2. More precisely we define $\Gamma_{\bar\theta}$ as the portion of S^2 inside a cone with apex in the origin, axis parallel to $\hat{a}_1$, aperture $\bar\theta, 0 < \bar\theta < \frac{\pi}{2}$, and we denote $\bar\Gamma_{\bar\theta} = S^2 \backslash \Gamma_{\bar\theta}$. For any choice of $\hat{a}_1, \hat{a}_2$ as in (6.1) the corresponding decomposition of $\mathcal{G}^{\varepsilon}_{12}$ in a non-stationary part (denoted with the label n) and a stationary part (denoted with the label s) is

$$\mathcal{G}^{\varepsilon}_{12} = \mathcal{G}^{\varepsilon,n}_{12} + \mathcal{G}^{\varepsilon,s}_{12} \tag{6.2}$$

$$\mathcal{G}^{\varepsilon,n}_{12} = \int_{\bar\Gamma_{\bar\theta}} \mathrm{d}\hat{u} \int_0^1 \mathrm{d}\alpha \int_0^\alpha \mathrm{d}\beta \int \mathrm{d}\eta d\xi G^{\varepsilon}_{12} e^{\frac{i}{\varepsilon}\Theta} \tag{6.3}$$

$$\mathcal{G}^{\varepsilon,s}_{12} = \int_{\Gamma_{\bar\theta}} \mathrm{d}\hat{u} \int_0^1 \mathrm{d}\alpha \int_0^\alpha \mathrm{d}\beta \int \mathrm{d}\eta d\xi G^{\varepsilon}_{12} e^{\frac{i}{\varepsilon}\Theta} \tag{6.4}$$

where

$$G^{\varepsilon}_{12} = G_{12}\, e^{i\delta_\varepsilon} \tag{6.5}$$

$$\delta_\varepsilon = -\frac{\sin\chi_\varepsilon}{\varepsilon}\,\mathfrak{b}_2\eta_1 + \frac{1-\cos\chi_\varepsilon}{\varepsilon}\,\mathfrak{b}_2\eta_3 \tag{6.6}$$

$$\Theta = \hat{u}\cdot(x + \mathfrak{a}(\alpha\eta + \beta\xi)) - \mathfrak{b}_1\xi_3 - \mathfrak{b}_2\eta_3 + \mathfrak{c}_2\alpha + \mathfrak{c}_1\beta \tag{6.7}$$

We shall analyze the asymptotic behavior of the two oscillatory integrals $\mathcal{G}^{\varepsilon,n}_{12}$ and $\mathcal{G}^{\varepsilon,s}_{12}$ separately. We first show that the phase Θ has no stationary points in $\bar\Gamma_{\bar\theta}$ and then the contribution of $\mathcal{G}^{\varepsilon,n}_{12}$ is negligible.

Proposition 6.1. *Let us fix $t > \tau_2, \hat{a}_1, \hat{a}_2$ as in (6.1) and let us assume (2.13), (2.14), (2.15), $V \in \mathcal{S}(\mathbb{R}^3)$. Then for any $k \in \mathbb{N}$ there exists $\mathcal{C}_k > 0$ such that*

$$\int dx\,dy_1\,dy_2\,|\mathcal{G}^{\varepsilon,n}_{12}(x,y_1,y_2)|^2 \leq \mathcal{C}_k \|\widetilde{V}\|^4_{W^{1,k}_k}\left(\frac{\varepsilon}{\sin\bar\theta}\right)^{2k} \tag{6.8}$$

Proof. If we denote $\hat{u} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta) \in \bar\Gamma_{\bar\theta}$, we have

$$\begin{aligned}\sum_{k=1}^3\left[\left(\frac{\partial\Theta}{\partial\eta_k}\right)^2 + \left(\frac{\partial\Theta}{\partial\xi_k}\right)^2\right] &= \mathfrak{a}^2(\alpha^2+\beta^2) + \mathfrak{b}_1^2 + \mathfrak{b}_2^2 - 2\mathfrak{a}(\mathfrak{b}_1\beta + \mathfrak{b}_2\alpha)\cos\theta \\ &\geq \mathfrak{a}^2(\alpha^2+\beta^2) + \mathfrak{b}_1^2 + \mathfrak{b}_2^2 \\ &\quad -2\mathfrak{a}(\mathfrak{b}_1\beta + \mathfrak{b}_2\alpha)\cos\bar\theta \end{aligned} \tag{6.9}$$

The r.h.s. of (6.9) takes its minimum in $(\alpha_2, \beta_2) = (\frac{\mathfrak{b}_2}{\mathfrak{a}} \cos\bar{\theta}, \frac{\mathfrak{b}_1}{\mathfrak{a}} \cos\bar{\theta})$, then

$$\sum_{k=1}^{3} \left[\left(\frac{\partial \Theta}{\partial \eta_k} \right)^2 + \left(\frac{\partial \Theta}{\partial \xi_k} \right)^2 \right] \geq \Delta^2 \tag{6.10}$$

where

$$\Delta = \sqrt{2} \min\{\mathfrak{b}_1, \mathfrak{b}_2\} \sin\bar{\theta} = \sqrt{2} \frac{\hbar}{P_0 \gamma} \frac{\min\{|a_1|, |a_2|\}}{\gamma} \sin\bar{\theta} \tag{6.11}$$

Exploiting estimate (6.10), (6.11) it is now straightforward to obtain (6.8) proceeding exactly as in the proof of Proposition 4.1. □

Remark 6.1. We notice that the estimate (6.8) is still meaningful if we choose the angle $\bar{\theta}$ proportional to ε^d, with $0 < d < 1$. This in particular means that only a small fraction of the unit sphere, of area proportional to ε^2 around the direction $\hat{a}_1$, can give a non trivial contribution to the ionization probability.

Let us consider the oscillatory integral $\mathcal{G}_{12}^{\varepsilon,s}$. It turns out that the phase Θ has a manifold of critical points in the integration region, parametrized by a vector in $\mathbb{R}^2$. Therefore we fix the variables $(\eta_1, \eta_2) \in \mathbb{R}^2$ as parameters and we write $\mathcal{G}_{12}^{\varepsilon,s}$ in the form

$$\mathcal{G}_{12}^{\varepsilon,s} = \int d\eta_1 d\eta_2 \mathcal{I}^{\varepsilon}(\eta_1, \eta_2) \tag{6.12}$$

$$\mathcal{I}^{\varepsilon}(\eta_1, \eta_2) = \int_{\Omega} dq G_{12}^{\varepsilon}(q; \eta_1, \eta_2) e^{\frac{i}{\varepsilon}\Theta(q; \eta_1, \eta_2)} \tag{6.13}$$

where

$$\Omega = \left\{ q \equiv (\hat{u}, \alpha, \beta, \eta_3, \xi) \,|\, \hat{u} \in \Gamma_{\bar{\theta}}, \alpha \in [0,1], \beta \in [0, \alpha], \eta_3 \in \mathbb{R}, \xi \in \mathbb{R}^3 \right\} \tag{6.14}$$

In the next lemma we show that for each value of the parameters (η_1, η_2) the phase in (6.13) has one, non degenerate stationary point. It is relevant that the value of the phase and of the Hessian of the phase at the critical point do not depend on (η_1, η_2).

Lemma 6.2. *For each* $(\eta_1, \eta_2) \in \mathbb{R}^2$ *the phase* $\Theta(q; \eta_1, \eta_2), q \in \Omega$, *has exactly one critical point*

$$q_0 \equiv \left(\hat{u}^0, \alpha^0, \beta^0, \eta_3^0, \xi_1^0, \xi_2^0, \xi_3^0 \right) \tag{6.15}$$

where

$$\hat{u}^0 = (0,0,1), \quad \alpha^0 = \frac{\mathfrak{b}_2}{\mathfrak{a}}, \quad \beta^0 = \frac{\mathfrak{b}_1}{\mathfrak{a}}, \quad \eta_3^0 = -\frac{\mathfrak{c}_2}{\mathfrak{a}}, \tag{6.16}$$

$$\xi_1^0 = -\frac{x_1 + \mathfrak{b}_2 \eta_1}{\mathfrak{b}_1}, \quad \xi_2^0 = -\frac{x_2 + \mathfrak{b}_2 \eta_2}{\mathfrak{b}_1}, \quad \xi_3^0 = -\frac{\mathfrak{c}_1}{\mathfrak{a}} \tag{6.17}$$

and $x = (x_1, x_2, x_3) \in \mathbb{R}^3$. *Moreover*

$$\Theta^0 \equiv \Theta(q_0; \eta_1, \eta_2) = x_3 + \frac{\mathfrak{b}_1 \mathfrak{c}_1}{\mathfrak{a}} + \frac{\mathfrak{b}_2 \mathfrak{c}_2}{\mathfrak{a}} \tag{6.18}$$

$$|D^2 \Theta^0| \equiv |D_q^2 \Theta(q_0; \eta_1, \eta_2)| = \mathfrak{a}^4 \mathfrak{b}_1^4 \tag{6.19}$$

Proof. In order to compute the critical points of the phase (6.7) as a function of $q \in \Omega$ it is convenient to write $\hat{u} = (\mu, \nu, \sqrt{1-\mu^2-\nu^2})$, where $(\mu,\nu) \in \mathbb{R}^2$ with $\mu^2+\nu^2 \le \sin^2\bar{\theta}$. Therefore

$$\Theta(q;\eta_1,\eta_2) = \mu w_1 + \nu w_2 + \sqrt{1-\mu^2-\nu^2}\,w_3 - \mathfrak{b}_2\eta_3 - \mathfrak{b}_1\xi_3 + \mathfrak{c}_2\alpha + \mathfrak{c}_1\beta \tag{6.20}$$

where we have introduced the short hand notation

$$w = (w_1, w_2, w_3), \quad w_j = x_j + \mathfrak{a}(\alpha\eta_j + \beta\xi_j) \tag{6.21}$$

By an explicit computation, one finds that the critical points are solutions of the system

$$\frac{\partial\Theta}{\partial\mu} = w_1 - \frac{\mu w_3}{\sqrt{1-\mu^2-\nu^2}} = 0 \tag{6.22}$$

$$\frac{\partial\Theta}{\partial\nu} = w_2 - \frac{\nu w_3}{\sqrt{1-\mu^2-\nu^2}} = 0 \tag{6.23}$$

$$\frac{\partial\Theta}{\partial\alpha} = \mathfrak{a}\mu\eta_1 + \mathfrak{a}\nu\eta_2 + \mathfrak{a}\sqrt{1-\mu^2-\nu^2}\,\eta_3 + \mathfrak{c}_2 = 0 \tag{6.24}$$

$$\frac{\partial\Theta}{\partial\beta} = \mathfrak{a}\mu\xi_1 + \mathfrak{a}\nu\xi_2 + \mathfrak{a}\sqrt{1-\mu^2-\nu^2}\,\xi_3 + \mathfrak{c}_1 = 0 \tag{6.25}$$

$$\frac{\partial\Theta}{\partial\eta_3} = \mathfrak{a}\sqrt{1-\mu^2-\nu^2}\,\alpha - \mathfrak{b}_2 = 0 \tag{6.26}$$

$$\frac{\partial\Theta}{\partial\xi_1} = \mathfrak{a}\mu\beta = 0 \tag{6.27}$$

$$\frac{\partial\Theta}{\partial\xi_2} = \mathfrak{a}\nu\beta = 0 \tag{6.28}$$

$$\frac{\partial\Theta}{\partial\xi_3} = \mathfrak{a}\sqrt{1-\mu^2-\nu^2}\,\beta - \mathfrak{b}_1 = 0 \tag{6.29}$$

First we notice that α and β cannot be zero, otherwise from (6.26), (6.29) one would have $\mathfrak{b}_2 = \mathfrak{b}_1 = 0$. Then from (6.27), (6.28) we have $\mu = \nu = 0$ and from (6.26), (6.27) we have $\alpha = \frac{\mathfrak{b}_2}{\mathfrak{a}}, \beta = \frac{\mathfrak{b}_1}{\mathfrak{a}}$. Exploiting the remaining equations it is now trivial to find the unique solution (6.16), (6.17). Furthermore the value of the phase at the critical point (6.18) is easily obtained. For the proof of (6.19) we need the second derivatives of the phase evaluated at the critical point

$$\frac{\partial^2\Theta}{\partial\mu^2} = -w_3, \quad \frac{\partial^2\Theta}{\partial\mu\partial\nu} = 0, \quad \frac{\partial^2\Theta}{\partial\mu\partial\alpha} = \mathfrak{a}\eta_1, \quad \frac{\partial^2\Theta}{\partial\mu\partial\beta} = \mathfrak{a}\xi_1, \quad \frac{\partial^2\Theta}{\partial\mu\partial\eta_3} = 0$$

$$\frac{\partial^2\Theta}{\partial\mu\partial\xi_1} = \mathfrak{b}_1, \quad \frac{\partial^2\Theta}{\partial\mu\partial\xi_2} = 0, \quad \frac{\partial^2\Theta}{\partial\mu\partial\xi_3} = 0$$

$$\frac{\partial^2\Theta}{\partial\nu^2} = -w_3, \quad \frac{\partial^2\Theta}{\partial\nu\partial\alpha} = \mathfrak{a}\eta_2, \quad \frac{\partial^2\Theta}{\partial\nu\partial\beta} = \mathfrak{a}\xi_2, \quad \frac{\partial^2\Theta}{\partial\nu\partial\eta_3} = 0$$

$$\frac{\partial^2\Theta}{\partial\nu\partial\xi_1} = 0, \quad \frac{\partial^2\Theta}{\partial\nu\partial\xi_2} = \mathfrak{b}_1, \quad \frac{\partial^2\Theta}{\partial\nu\partial\xi_3} = 0$$

$$\begin{aligned}
&\frac{\partial^2\Theta}{\partial\alpha^2}=0, \quad \frac{\partial^2\Theta}{\partial\alpha\partial\beta}=0, \quad \frac{\partial^2\Theta}{\partial\alpha\partial\eta_3}=\mathfrak{a}, \quad \frac{\partial^2\Theta}{\partial\alpha\partial\xi_j}=0\\
&\frac{\partial^2\Theta}{\partial\beta^2}=0, \quad \frac{\partial^2\Theta}{\partial\beta\partial\eta_3}=0, \quad \frac{\partial^2\Theta}{\partial\beta\partial\xi_1}=0, \quad \frac{\partial^2\Theta}{\partial\beta\partial\xi_2}=0, \quad \frac{\partial^2\Theta}{\partial\beta\partial\xi_3}=\mathfrak{a}\\
&\frac{\partial^2\Theta}{\partial\eta_3^2}=0, \quad \frac{\partial^2\Theta}{\partial\eta_3\partial\xi_j}=0, \quad \frac{\partial^2\Theta}{\partial\xi_j\partial\xi_k}=0
\end{aligned} \tag{6.30}$$

The computation of the Hessian is now a tedious but straightforward exercise and it is omitted for the sake of brevity. □

We are now ready to conclude the proof of theorem 2.

Proof of Theorem 2. Exploiting the stationary phase theorem [3,11,16] and the previous lemma, we find the leading term of the asymptotic expansion of (6.13) for $\varepsilon \to 0$

$$\mathcal{I}^\varepsilon(\eta_1,\eta_2) \sim \frac{(2\pi\varepsilon)^4}{\mathfrak{a}^2\mathfrak{b}_1^2} e^{\frac{i}{\varepsilon}\Theta^0} G_{12}(q_0;\eta_1,\eta_2) e^{i\delta_0} e^{i\frac{\pi}{4}\mu_0} \tag{6.31}$$

where μ_0 denotes the signature of the Hessian matrix at the critical point and moreover

$$\delta_0 = -\lim_{\varepsilon\to 0}\frac{\sin\chi_\varepsilon}{\varepsilon}\mathfrak{b}_2\eta_1 \tag{6.32}$$

In particular $\delta_0 \neq 0$ if $\chi_\varepsilon = O(\varepsilon)$ and $\delta_0 = 0$ if $\chi_\varepsilon = O(\varepsilon^q), q > 1$, or $\chi_\varepsilon = 0$. From (6.12) we also obtain

$$\mathcal{G}_{12}^{\varepsilon,s} \sim \frac{(2\pi\varepsilon)^4}{\mathfrak{a}^2\mathfrak{b}_1^2} e^{\frac{i}{\varepsilon}\Theta_2^0} \int \mathrm{d}\eta_1\mathrm{d}\eta_2 G_{12}(q_0;\eta_1,\eta_2) e^{i\delta_0} e^{i\frac{\pi}{4}\mu_0} \tag{6.33}$$

We notice that the integrand in (6.33) is a function of x (position of the α-particle), y_j (momentum of the jth ionized atom) and η_1,η_2. Hence we denote

$$F(\eta_1,\eta_2;x,y_1,y_2) \equiv (2\pi)^4 G_{12}(q_0;\eta_1,\eta_2) e^{i\delta_0} e^{i\frac{\pi}{4}\mu_0} \tag{6.34}$$

and, taking into account (3.8), Proposition 4.1, (6.2), Lemma 6.2 and (6.33), we find

$$\mathcal{P}(t) \sim \left(\frac{\lambda t}{\hbar}\right)^4 \frac{\mathcal{N}_\varepsilon^2}{\mathfrak{a}^4\mathfrak{b}_1^4}\varepsilon^6 \int \mathrm{d}x\mathrm{d}y\mathrm{d}z \left|\int \mathrm{d}\eta_1\mathrm{d}\eta_2 F(\eta_1,\eta_2;x,y,z)\right|^2 \tag{6.35}$$

Using the definition of $\mathfrak{a}, \mathfrak{b}_1, \tau_1$ in (6.35), we easily get formula (3.5). It remains to show that the integral in (6.35) is finite. From the definitions (3.11), (2.27), (2.26) and the boundedness of $h(\xi,y)$ we have

$$|F(\eta_1,\eta_2;x,y,z)| \le c\left|\widetilde{V}\left(\eta_1,\eta_2,\eta_3^0\right)\right|\left|\widetilde{V}\left(\xi_1^0,\xi_2^0,\xi_3^0\right)\right||f(w_0)| \tag{6.36}$$

where

$$\begin{aligned}
&\eta_3^0 = -\varepsilon\frac{M}{m}\omega(z), \quad \xi_1^0 = -\varepsilon^{-1}\frac{\gamma}{|a_1|}x_1 - \frac{\tau_2}{\tau_1}\eta_1, \quad \xi_2^0 = -\varepsilon^{-1}\frac{\gamma}{|a_1|}x_2 - \frac{\tau_2}{\tau_1}\eta_2,\\
&\xi_3^0 = -\varepsilon\frac{M}{m}\omega(y), \quad w_0 = x_3 - \varepsilon^2\frac{M}{m}\frac{|a_1|}{\gamma}\omega(y) - \varepsilon^2\frac{M}{m}\frac{|a_2|}{\gamma}\omega(z)
\end{aligned} \tag{6.37}$$

We recall that f and $\omega(y)$ are defined in (2.9), (2.24), respectively, and $x = (x_1, x_2, x_3) \in \mathbb{R}^3$. Then we write

$$\begin{aligned}\int \mathrm{d}x\mathrm{d}y\mathrm{d}z \left| \int \mathrm{d}\eta_1 \mathrm{d}\eta_2 F(\eta_1, \eta_2; x, y, z) \right|^2 & \\ \leq c \int \mathrm{d}x_1 \mathrm{d}x_2 \mathrm{d}y \mathrm{d}z \left(\int \mathrm{d}\eta_1 \mathrm{d}\eta_2 |\widetilde{V}(\eta_1, \eta_2, \eta_3^0)||\widetilde{V}\left(\xi_1^0, \xi_2^0, \xi_3^0\right)| \right)^2 \int \mathrm{d}x_3 |f(w_0)|^2 \end{aligned} \tag{6.38}$$

Using the Schwartz inequality in the integral with respect to (η_1, η_2) we have

$$\begin{aligned}\int \mathrm{d}x\mathrm{d}y\mathrm{d}z \left| \int \mathrm{d}\eta_1 \mathrm{d}\eta_2 F(\eta_1, \eta_2; x, y, z) \right|^2 \leq c \int \mathrm{d}z \int \mathrm{d}\eta_1 \mathrm{d}\eta_2 <\eta_1>^4 <\eta_2>^4 \\ \times |\widetilde{V}(\eta_1, \eta_2, \eta_3^0)|^2 \int \mathrm{d}y \int \mathrm{d}x_1 \mathrm{d}x_2 \left| \widetilde{V}\left(\xi_1^0, \xi_2^0, \xi_3^0\right) \right|^2 \int \mathrm{d}x_3 |f(w_0)|^2 \end{aligned} \tag{6.39}$$

The last integral is finite due to the assumptions on V and this concludes the proof of the theorem. □

As we already pointed out in Remark 3.1, an explicit estimate of the remainder in the expansion of (6.13) goes beyond the scope of this paper. We notice that a complete asymptotic expansion, with an estimate of the rest, for $\varepsilon \to 0$ of $\mathcal{I}^\varepsilon(\eta_1, \eta_2)$ can be obtained following the general analysis of [11] where, in Theorem 2.4, the case of oscillatory integrals depending on several parameters is discussed in detail.

Here, we want just to outline an alternative possible strategy to evaluate the rest based on an elementary manipulation of the integral and exploiting the specific form of the phase. We define

$$\boldsymbol{p} = (\mu, \nu, \alpha, \beta), \quad \boldsymbol{k} = (\eta_3, \xi_1, \xi_2, \xi_3) \tag{6.40}$$

$$\begin{aligned} B(\boldsymbol{p}; \eta, \eta_2) = \mu(x_1 + \mathfrak{a}\alpha\eta_1) + \nu(x_2 + \mathfrak{a}\alpha\eta_2) + \sqrt{1 - \mu^2 - \nu^2}\, x_3 \\ + \mathfrak{c}_2\alpha + \mathfrak{c}_1\beta \end{aligned} \tag{6.41}$$

$$\boldsymbol{A}(\boldsymbol{p}) = \left(\sqrt{1 - \mu^2 - \nu^2}\mathfrak{a}\alpha - \mathfrak{b}_2, \mu\mathfrak{a}\beta, \nu\mathfrak{a}\beta, \sqrt{1 - \mu^{-}\nu^2}\mathfrak{a}\beta - \mathfrak{b}_1 \right) \tag{6.42}$$

Then, dropping the dependence on the parameters η_1, η_2, we have

$$\mathcal{I}^\varepsilon = \int_D \mathrm{d}\boldsymbol{p} e^{\frac{i}{\varepsilon} B(\boldsymbol{p})} \int \mathrm{d}\boldsymbol{k} G_{12}^\varepsilon(\boldsymbol{p}, \boldsymbol{k}) e^{\frac{i}{\varepsilon}\boldsymbol{A}(\boldsymbol{p})\cdot\boldsymbol{k}} \tag{6.43}$$

where D is the domain of integration corresponding to the variables $\boldsymbol{p}$.

We introduce the following linear change of coordinates $\boldsymbol{p} = (\mu, \nu, \alpha, \beta) \to \boldsymbol{z} = (z_1, z_2, z_3, z_4)$

$$\mu = \frac{\varepsilon}{\mathfrak{b}_1} z_2, \quad \nu = \frac{\varepsilon}{\mathfrak{b}_1} z_3, \quad \alpha = \frac{\mathfrak{b}_2}{\mathfrak{a}} + \frac{\varepsilon}{\mathfrak{a}} z_1, \quad \beta = \frac{\mathfrak{b}_1}{\mathfrak{a}} + \frac{\varepsilon}{\mathfrak{a}} z_4 \tag{6.44}$$

and denote $\boldsymbol{p} = L_\varepsilon \boldsymbol{z}$. Hence

$$\mathcal{I}^\varepsilon = \frac{\varepsilon^4}{\mathfrak{a}^2 \mathfrak{b}_1^2} \int_{D_\varepsilon} \mathrm{d}\boldsymbol{z} e^{\frac{i}{\varepsilon} B(L_\varepsilon \boldsymbol{z})} \int \mathrm{d}\boldsymbol{k} G_{12}^\varepsilon(L_\varepsilon \boldsymbol{z}, \boldsymbol{k}) e^{\frac{i}{\varepsilon}\boldsymbol{A}(L_\varepsilon \boldsymbol{z})\cdot\boldsymbol{k}} \tag{6.45}$$

where D_ε is the domain of integration corresponding to the variables $\boldsymbol{z}$. We notice that

$$B(L_\varepsilon \boldsymbol{z}) = \Theta^0 - \varepsilon \boldsymbol{k}^0 \cdot \boldsymbol{z} + \Lambda_\varepsilon(\boldsymbol{z}), \tag{6.46}$$

$$\boldsymbol{A}(L_\varepsilon \boldsymbol{z}) = \varepsilon \boldsymbol{z} + \boldsymbol{\Gamma}_\varepsilon(\boldsymbol{z}) \tag{6.47}$$

where

$$\boldsymbol{k}^0 = -\left(\frac{\mathfrak{c}_2}{\mathfrak{a}}, \frac{x_1 + \mathfrak{b}_2 \eta_1}{\mathfrak{b}_1}, \frac{x_2 + \mathfrak{b}_2 \eta_2}{\mathfrak{b}_1}, \frac{\mathfrak{c}_1}{\mathfrak{a}} \right) \tag{6.48}$$

and $\Lambda_\varepsilon(\boldsymbol{z}), \boldsymbol{\Gamma}_\varepsilon(\boldsymbol{z})$ are explicitly known functions of order ε^2 for $\varepsilon \to 0$. We also notice that D_ε reduces to $\mathbb{R}^4$ for $\varepsilon \to 0$. Taking into account (6.46), (6.47), we have

$$\mathcal{I}^\varepsilon = \frac{\varepsilon^4}{\mathfrak{a}^2 \mathfrak{b}_1^2} e^{\frac{i}{\varepsilon}\Theta^0} \int_{D_\varepsilon} \mathrm{d}\boldsymbol{z} e^{-i\boldsymbol{k}^0 \cdot \boldsymbol{z}} e^{\frac{i}{\varepsilon}\Lambda_\varepsilon(\boldsymbol{z})} \int \mathrm{d}\boldsymbol{k} G_{12}^\varepsilon(L_\varepsilon \boldsymbol{z}, \boldsymbol{k}) e^{i\boldsymbol{z}\cdot\boldsymbol{k}} e^{\frac{i}{\varepsilon}\boldsymbol{\Gamma}_\varepsilon(\boldsymbol{z})\cdot\boldsymbol{k}} \tag{6.49}$$

From the above formula one sees that the expansion of $\mathcal{I}^\varepsilon$ for $\varepsilon \to 0$, with an explicit remainder, is reduced to the Taylor expansion of the integrand in the r.h.s. of (6.49) and then to the estimate of the error done if the domain D_ε is replaced by $\mathbb{R}^4$. Finally, by suitable integration by parts, one can show that each term of the expansion and the remainder are summable w.r.t. the other variables (η_1, η_2 and then x, y, z).

We plan to follow this strategy in further work where the analysis of the model will be carried out at any order in perturbation theory.

7. Appendix I

We recall here the definition and the main properties of the Schrödinger operator with an attractive point interaction, which is used as model Hamiltonian for the two "atoms" (for further details and for the proofs we refer to [1]).

We fix for simplicity $\hbar = 1, m = 1/2$ and define domain and action of the Schrödinger operator $\mathcal{K}_\alpha$ in $L^2(\mathbb{R}^3)$ with a point interaction placed at the origin and strength $\alpha < 0$ as follows

$$D(\mathcal{K}_\alpha) = \left\{ u \in L^2(\mathbb{R}^3) | u = w^\lambda + qG^\lambda, w^\lambda \in D(-\Delta), q \in \mathbb{C}, w^\lambda(0) = \left(\alpha + \frac{\sqrt{\lambda}}{4\pi}\right) q \right\} \tag{7.1}$$

$$\mathcal{K}_\alpha u = -\Delta w^\lambda - \lambda q G^\lambda \tag{7.2}$$

where $D(-\Delta)$ is the domain of the free Hamiltonian, $\lambda > (4\pi\alpha)^2$ and $G^\lambda(x) = \frac{e^{-\sqrt{\lambda}|x|}}{4\pi|x|}$. The operator defined in (7.1), (7.2) is selfadjoint and bounded from below, with spectrum explicitly given by

$$\sigma_p(\mathcal{K}_\alpha) = \{-(4\pi\alpha)^2\}, \quad \sigma_c(\mathcal{K}_\alpha) = \sigma_{ac}(\mathcal{K}_\alpha) = [0, \infty) \tag{7.3}$$

The unique proper eigenfunction corresponding to the negative eigenvalue and a set of generalized eigenfunctions corresponding to the absolutely continuous spectrum are respectively given by

$$\zeta_\alpha(x) = (-\alpha)^{1/2} \frac{e^{4\pi\alpha|x|}}{|x|} \tag{7.4}$$

$$\phi_\alpha(x,k) = \frac{1}{(2\pi)^{3/2}} \left(e^{ik\cdot x} + \frac{1}{4\pi\alpha + i|k|} \frac{e^{-i|k||x|}}{|x|} \right) \tag{7.5}$$

We notice that for $\alpha = -(4\pi)^{-1}$ the expressions (7.4), (7.5) reduce to $\zeta^0(x)$, $\phi^0(x,k)$ (defined in (2.6), (2.7)). Exploiting ζ_α and ϕ_α one obtains an eigenfunction expansion theorem for the operator $\mathcal{K}_\alpha$. More precisely one defines the map

$$u \to \{u_0, \hat{u}\} \tag{7.6}$$

$$u_0 = \int \mathrm{d}x u(x)\zeta_\alpha(x), \quad \hat{u}(k) = \int \mathrm{d}x u(x)\overline{\phi_\alpha}(x,k) \tag{7.7}$$

for $u \in L^2(\mathbb{R}^3)$ sufficiently smooth and then one can prove the following statements.

(1) The map (7.6), (7.7) extends to a unitary operator

$$U_\alpha : u \in L^2(\mathbb{R}^3) \to \{u_0, \hat{u}\} \in \mathbb{C} \oplus L^2(\mathbb{R}^3) \tag{7.8}$$

where the last integral in (7.7) should be now intended in the L^2-sense, i.e., $\hat{u}$ is by definition the limit in $L^2(\mathbb{R}^3)$ for $N \to \infty$ of the sequence $\int_{|x|<N} \mathrm{d}x u(x)\overline{\phi_\alpha}(x,\cdot)$. In particular one has

$$\|u\|^2_{L^2(\mathbb{R}^3)} = |u_0|^2 + \|\hat{u}\|^2_{L^2(\mathbb{R}^3)} \tag{7.9}$$

Moreover for any $u \in L^2(\mathbb{R}^3)$ the following inversion formula holds

$$u(x) = u_0\zeta_\alpha(x) + \int \mathrm{d}k\hat{u}(k)\phi_\alpha(x,k) \tag{7.10}$$

where the two terms of the sum in r.h.s. of (7.10) are orthogonal.

(2) $U_\alpha\mathcal{K}_\alpha U_\alpha^{-1}$ is a diagonal operator , i.e. for $u \in D(\mathcal{K}_\alpha)$

$$(\mathcal{K}_\alpha u)_0 = -(4\pi\alpha)^2\, u_0, (\widehat{\mathcal{K}_\alpha u})(k) = k^2\hat{u}(k) \tag{7.11}$$

which in particular means that the pure point and the absolutely continuous subspaces associated to $\mathcal{K}_\alpha$ are explicitly characterized as follows

$$L^2_{pp}(\mathbb{R}^3) = \{u \in L^2(\mathbb{R}^3) | \hat{u} = 0\}, \quad L^2_{ac}(\mathbb{R}^3) = \{u \in L^2(\mathbb{R}^3) | u_0 = 0\} \tag{7.12}$$

(3) For any $u \in L^2(\mathbb{R}^3)$ the unitary propagator reads

$$\left(e^{-it\mathcal{K}_\alpha}u\right)(x) = e^{-i(4\pi\alpha)^2 t}u_0\zeta_\alpha(x) + \int \mathrm{d}ke^{-itk^2}\hat{u}(k)\phi_\alpha(x,k) \tag{7.13}$$

where again the two terms of the sum in the r.h.s. of (7.13) are orthogonal for any $t > 0$.

In principle the above statements can be derived exploiting general results about eigenfunction expansions for selfadjoint operators. On the other hand in the case of the operator $\mathcal{K}_\alpha$ the eigenfunctions $\zeta_\alpha, \phi_\alpha$ are explicitly known in terms of elementary functions and therefore all the above properties can be directly checked. In particular it is a nice exercise to verify that U_α is unitary and (7.10) holds, by simply mimicking the standard proof of Plancherel's theorem for the Fourier transform in L^2. Moreover a direct computation shows that the two terms of the sum in the r.h.s. of (7.10) are orthogonal. Formulae (7.11), (7.12) can be directly checked exploiting the definition of $\mathcal{K}_\alpha$ and the representation (7.13) for the unitary propagator follows from (7.11) and the fact that U_α is unitary.

8. Appendix II

Here, we give a proof of the representation formula (3.8). The relevant object to compute is the probability amplitude in (3.3)

$$\mathcal{F}(R,k_1,k_2,t)=\int \mathrm{d}r_1\mathrm{d}r_2\overline{\phi_1}(r_1,k_1)\overline{\phi_2}(r_2,k_2)\hat{\Psi}_2(R,r_1,r_2,t) \tag{8.1}$$

As we already pointed out in Appendix I, formula (8.1) defines $\mathcal{F}(R,\cdot,\cdot,t)$ as the limit in $L^2(\mathbb{R}^6)$ for $N,M\to\infty$ of the sequence $\int_{|r_1|<N}\mathrm{d}r_1\int_{|r_2|<M}\mathrm{d}r_2\overline{\phi_1}(r_1,\cdot)\overline{\phi_2}(r_2,\cdot)\hat{\Psi}_2(R,r_1,r_2,t)$.

We notice that

$$e^{\frac{i}{\hbar}tH_0}H_1e^{-\frac{i}{\hbar}tH_0}=W_1(t)+W_2(t) \tag{8.2}$$

$$W_j(t)=e^{\frac{i}{\hbar}tK_0}e^{\frac{i}{\hbar}tK_j}V_je^{-\frac{i}{\hbar}tK_0}e^{-\frac{i}{\hbar}tK_j} \tag{8.3}$$

where V_j denotes the multiplication operator by

$$V_j(R,r_j)=V(\gamma^{-1}(R-r_j)) \tag{8.4}$$

Using (8.2) we rewrite the r.h.s. of (8.1) in the more convenient form

$$\begin{aligned}\mathcal{F}(R,k_1,k_2,t)=&\int \mathrm{d}r_1\mathrm{d}r_2\overline{\phi_1}(r_1,k_1)\overline{\phi_2}(r_2,k_2)\Psi_0(R,r_1,r_2)\\&-i\frac{\lambda}{\hbar}\int_0^t\mathrm{d}t_1\int\mathrm{d}r_1\mathrm{d}r_2\overline{\phi_1}(r_1,k_1)\overline{\phi_2}(r_2,k_2)\\&\times\left[\left(W_1(t_1)+W_2(t_1)\right)\Psi_0\right](R,r_1,r_2)\\&-\frac{\lambda^2}{\hbar^2}\int_0^t\mathrm{d}t_1\int_0^{t_1}\mathrm{d}t_2\int\mathrm{d}r_1\mathrm{d}r_2\overline{\phi_1}(r_1,k_1)\overline{\phi_2}(r_2,k_2)\\&\times\left[\left(W_1(t_1)+W_2(t_1)\right)\left(W_1(t_2)+W_2(t_2)\right)\Psi_0\right](R,r_1,r_2)\end{aligned} \tag{8.5}$$

We observe that the operator $W_j(t)$ acts non trivially only on the variable R and r_j. Exploiting this fact and the orthogonality relation (see Appendix I)

$$\int \mathrm{d}r\overline{\phi_j}(r,k)\zeta_j(r)=0,\quad j=1,2 \tag{8.6}$$

we obtain

$$\mathcal{F} = \mathcal{F}_{12} + \mathcal{F}_{21} \tag{8.7}$$

$$\begin{aligned}&\mathcal{F}_{lj}(R, k_1, k_2, t)\\ &\quad = \frac{\lambda^2}{\hbar^2} \int_0^t \mathrm{d}t_1 \int_0^{t_1} \mathrm{d}t_2 \int \mathrm{d}r_1 \mathrm{d}r_2 \overline{\phi}_1(r_1, k_1)\overline{\phi}_2(r_2, k_2) \left(W_j(t_1)W_l(t_2)\Psi_0\right)(R, r_1, r_2)\end{aligned} \tag{8.8}$$

where $l, j = 1, 2, j \neq l$. Due to the specific factorized form of the initial state of the system, we have

$$W_j(t_1)W_l(t_2)\Psi_0 = e^{\frac{i}{\hbar}(t_1+t_2)|E_0|} e^{\frac{i}{\hbar}t_1 K_0} e^{\frac{i}{\hbar}t_1 K_j} V_j \zeta_j e^{\frac{i}{\hbar}t_2 K_l} V_l \zeta_l e^{-\frac{i}{\hbar}t_2 K_0} \psi \tag{8.9}$$

Moreover, according to the eigenfunction expansion theorem recalled in Appendix I, the propagator generated by K_j is

$$\left(e^{\frac{i}{\hbar}tK_j}(V_j(R, \cdot)\zeta_j)\right)(r) = e^{\frac{i}{\hbar}tE_0} c_j^0(R)\zeta_j(r) + \int \mathrm{d}k e^{\frac{i}{\hbar}tE(k)} \hat{V}_j(R, k)\phi_j(r, k) \tag{8.10}$$

where

$$c_j^0(R) = \int \mathrm{d}r V_j(R, r)\zeta_j^2(r) \tag{8.11}$$

$$\hat{V}_j(R, k) = \int \mathrm{d}r \overline{\phi}_j(r, k) V(\gamma^{-1}(R - r))\zeta_j(r) \tag{8.12}$$

$$E(k) = \frac{\hbar^2 k^2}{2m} \tag{8.13}$$

Therefore

$$\int \mathrm{d}r \overline{\phi}_j(r, k) \left(e^{\frac{i}{\hbar}tK_j}(V_j(R, \cdot)\zeta_j)\right)(r) = e^{\frac{i}{\hbar}tE(k)} \hat{V}_j(R, k) \tag{8.14}$$

Exploiting (8.9) and (8.14), formula (8.8) reduces to

$$\begin{aligned}\mathcal{F}_{lj}(R, k_1, k_2, t) = \frac{\lambda^2}{\hbar^2} \int_0^t \mathrm{d}t_1 \int_0^{t_1} \mathrm{d}t_2 e^{\frac{i}{\hbar}t_1(E(k_j)+|E_0|)} e^{\frac{i}{\hbar}t_2(E(k_l)+|E_0|)}\\ \times \left(e^{\frac{i}{\hbar}t_1 K_0} \hat{V}_j(\cdot, k_j) e^{-\frac{i}{\hbar}t_1 K_0} e^{\frac{i}{\hbar}t_2 K_0} \hat{V}_l(\cdot, k_l) e^{-\frac{i}{\hbar}t_2 K_0} \psi\right)(R)\end{aligned} \tag{8.15}$$

We notice that the r.h.s. of (8.12) can be more conveniently written in terms of the Fourier transform $\widetilde{V}$ of the interaction potential as follows

$$\hat{V}_j(R, k_j) = e^{-ik\cdot a_j} \gamma^{3/2} \int \mathrm{d}\xi e^{i\gamma^{-1}(R-a_j)\cdot\xi} g(\xi, \gamma k_j) \tag{8.16}$$

where $g(\xi,y)$ has been defined in (2.27). From (8.16) and the explicit expression of the free propagator we have

$$\left(e^{\frac{i}{\hbar}t_2K_0}\hat{V}_l(\cdot,k_l)e^{-\frac{i}{\hbar}t_2K_0}\psi\right)(R)$$
$$= e^{-ik_l\cdot a_l}\gamma^{3/2}\int \mathrm{d}\xi g(\xi,\gamma k_l)e^{i\frac{\hbar t_2}{2M\gamma^2}\xi^2+i\frac{R}{\gamma}\cdot\xi-i\frac{a_l}{\gamma}\cdot\xi}\psi\left(R+\frac{\hbar t_2}{M\gamma}\xi\right) \tag{8.17}$$

and

$$\left(e^{\frac{i}{\hbar}t_1K_0}\hat{V}_j(\cdot,k_j)e^{-\frac{i}{\hbar}t_1K_0}e^{\frac{i}{\hbar}t_2K_0}\hat{V}_l(\cdot,k_l)e^{-\frac{i}{\hbar}t_2K_0}\psi\right)(R)$$
$$= e^{-ik_l\cdot a_l-ik_j\cdot a_j}\gamma^3\int \mathrm{d}\xi\mathrm{d}\eta g(\eta,\gamma k_j)g(\xi,\gamma k_l)$$
$$\cdot\, e^{i\left(\frac{\hbar t_1}{2M\gamma^2}\eta^2+\frac{R}{\gamma}\cdot\eta+\frac{\hbar t_2}{2M\gamma^2}\xi^2+\frac{R}{\gamma}\cdot\xi+\frac{\hbar t_1}{M\gamma^2}\eta\cdot\xi\right)}e^{i\left(-\frac{a_j}{\gamma}\cdot\eta-\frac{a_l}{\gamma}\cdot\xi\right)}\psi$$
$$\times\left(R+\frac{\hbar t_1}{M\gamma}\eta+\frac{\hbar t_2}{M\gamma}\xi\right) \tag{8.18}$$

Finally we consider the time-dependent phase factor in (8.15). We notice that

$$\frac{t_1}{\hbar}(E(k_j)+|E_0|)+\frac{t_2}{\hbar}(E(k_l)+|E_0|)=\frac{\hbar}{m\gamma^2}\left(t_1w(\gamma k_j)+t_2w(\gamma k_l)\right) \tag{8.19}$$

Taking into account (8.18), (8.19), (2.9), and rescaling the time variables according to $t_1=t\alpha, t_2=t\beta$, we can rewrite (8.15) as follows

$$\mathcal{F}_{lj}(R,k_1,k_2,t)=\frac{\lambda^2t^2}{\hbar^2}\frac{\mathcal{N}_\varepsilon}{\varepsilon}\gamma^{3/2}e^{-ik_l\cdot a_l-ik_j\cdot a_j}$$
$$\times\int_{S^2}\mathrm{d}\hat{u}\int_0^1\mathrm{d}\alpha\int_0^\alpha\mathrm{d}\beta\int\mathrm{d}\xi\mathrm{d}\eta g(\eta,\gamma k_j)g(\xi,\gamma k_l)f$$
$$\times\left(\frac{R}{\gamma}+\frac{\hbar t}{M\gamma^2}(\alpha\eta+\beta\xi)\right)$$
$$\cdot\, e^{i\left(\frac{\hbar t}{2M\gamma^2}\alpha\eta^2+\frac{R}{\gamma}\cdot\eta+\frac{\hbar t}{2M\gamma^2}\beta\xi^2+\frac{R}{\gamma}\cdot\xi+\frac{\hbar t}{M\gamma^2}\alpha\eta\cdot\xi\right)}$$
$$\cdot\, e^{i\left[\frac{1}{\varepsilon}\hat{u}\cdot\left(\frac{R}{\gamma}+\frac{\hbar t}{M\gamma^2}(\alpha\eta+\beta\xi)\right)-\frac{a_j}{\gamma}\cdot\eta-\frac{a_l}{\gamma}\cdot\xi+\frac{\hbar t}{m\gamma^2}(\omega(\gamma k_j)\alpha+\omega(\gamma k_l)\beta)\right]} \tag{8.20}$$

We observe that

$$\frac{\hbar t}{M\gamma^2}=\mathfrak{a} \tag{8.21}$$

is of order one for $\varepsilon\to 0$ and this means that the first exponential in the integral in (8.20) has a slowly oscillating phase for $\varepsilon\ll 1$. On other hand

$$\frac{\hbar t}{m\gamma^2}=\frac{M}{m}\mathfrak{a} \tag{8.22}$$

is proportional to ε^{-1} and therefore the last exponential in the integral in (8.20) has a rapidly oscillating phase for $\varepsilon\ll 1$. Denoting $R=\gamma x, k_j=\gamma^{-1}y_j$ and using the notation (2.25), we find

$$\mathcal{F}_{lj}(\gamma x, \gamma^{-1}y_1, \gamma^{-1}y_2) = \frac{\lambda^2 t^2}{\hbar^2}\frac{\mathcal{N}_\varepsilon}{\varepsilon}\gamma^{3/2}e^{-ik_l\cdot a_l - ik_j\cdot a_j}$$
$$\times \int_{S^2} d\hat{u}\int_0^1 \mathrm{d}\alpha \int_0^\alpha \mathrm{d}\beta \int \mathrm{d}\xi\mathrm{d}\eta g(\eta, y_j)g(\xi, y_l)f(x+\mathfrak{a}(\alpha\eta+\beta\xi))$$
$$\times e^{i\left[x\cdot(\eta+\xi)+\frac{\mathfrak{a}}{2}(\alpha\eta^2+\beta\xi^2+2\alpha\eta\cdot\xi)\right]}e^{\frac{i}{\varepsilon}[\hat{u}\cdot x+\hat{u}\cdot\mathfrak{a}(\alpha\eta+\beta\xi)-\mathfrak{b}_j\hat{a}_j\cdot\eta-\mathfrak{b}_l\hat{a}_l\cdot\xi+\mathfrak{c}_j\alpha+\mathfrak{c}_l\beta]}$$
$$\equiv \frac{\lambda^2 t^2}{\hbar^2}\frac{\mathcal{N}_\varepsilon}{\varepsilon}\gamma^{3/2}e^{-ik_l\cdot a_l-ik_j\cdot a_j}\mathcal{G}^\varepsilon_{lj}(x, y_1, y_2, t) \tag{8.23}$$

where in the last line we have used (3.9), (3.10), (3.11), (3.12). From (3.3), (8.1) and (8.23) we obtain

$$\mathcal{P}(t) = \gamma^{-3}\int \mathrm{d}x\mathrm{d}y_1\mathrm{d}y_2 \left|\mathcal{F}_{12}(\gamma x, \gamma^{-1}y_1, \gamma^{-1}y_2)+\mathcal{F}_{21}(\gamma x, \gamma^{-1}y_1, \gamma^{-1}y_2)\right|^2$$
$$= \frac{\lambda^4 t^4}{\hbar^4}\frac{\mathcal{N}_\varepsilon^2}{\varepsilon^2}\int \mathrm{d}x\mathrm{d}y_1\mathrm{d}y_2\left|\mathcal{G}^\varepsilon_{12}(x, y_1, y_2)+\mathcal{G}^\varepsilon_{21}(x, y_1, y_2)\right|^2 \tag{8.24}$$

and this concludes the proof of (3.8).

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