Exact Results for Ionization of Model Atomic Systems

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We review recent rigorous results concerning the ionization of model quantum systems by time periodic external fields. The systems we consider consist of a single particle (electron) with a reference Hamiltonian $H_0 = -\Delta + V_0(x)$ ($x \in \mathbb{R}^d$) having both bound and continuum states. Starting from an initially localized state $\psi_0(x) \in L^2(\mathbb{R}^d)$, the system is subjected for $t \geq 0$ to an arbitrary strength time-periodic potential $V_1(x,t) = V_1(x, t + 2\pi/\omega)$. We prove that for a large class of $V_0(x)$ and $V_1(x,t)$ the wavefunction $\psi(x,t)$ will delocalize as $t \to \infty$, i.e. the system will ionize. The only exceptions are cases where there are time-periodic bound states of the Floquet operator associated to $H_0 + V_1$. These do occur (albeit rarely) when V_1 is not small. For spatially rapidly decaying V_0 and V_1, $\psi(x,t)$ is generally given, for very long times, by a power series in $t^{-1/2}$ which we prove in some cases to be Borel summable. For the Coulomb potential $V_0(x) = -b|x|^{-1}$ in $\mathbb{R}^3$, we prove ionization for $V_1(x,t) = V_1(|x|)\sin(\omega t - \theta)$, $V_1(|x|) = 0$ for $|x| > R$ and $V_1(x) > 0$ for $|x| \leq R$. For this model, if ψ_0 is compactly supported both in x and in angular momentum, $\mathbf{L}$, we obtain that $\psi(x,t) \sim O(t^{-5/6})$ as $t \to \infty$.

I. INTRODUCTION

The ionization of atoms or dissociation of molecules subjected to external time-dependent fields is an issue of central importance in atomic physics. There exist a variety of methods for treating this problem, including perturbation theory (Fermi's golden rule), numerical integration of the time-dependent Schrödinger equation, semi-classical phase-space analysis, Floquet theory and complex dilations[1,10–12,14,20,23]. Still, there are very few rigorous results proving or disproving ionization by a periodic field of arbitrary strength and frequency, even for the simplest systems with both bound and continuum states. Such results are clearly desirable from both a theoretical and practical point of view. Numerical results are also difficult since delocalization of the wavefunction of the electron creates truncation errors at the boundary of the computational domain (absorbing potentials can help, but may also cause difficult to detect errors).

We present here a review of recent rigorous results for this problem in the context of non-relativistic quantum mechanics where the field causing the ionization is treated classically[1]. Most of these results are derived in detail in[3–8,18]. We give here the ideas of the proofs in a unified and simple form.

Our analysis is based on the study of the long time behavior of the solution of the Schrödinger equation in d dimensions (in units such that $\hbar = 2m = 1$),

$$i\partial_t \psi(x,t) = \Big(-\Delta + V_0(x) + V_1(x,t) \Big)\psi(x,t); \quad \psi(0,x) = \psi_0(x) \in L^2(\mathbb{R}^d) \qquad (\text{I.1})$$

Here, $x \in \mathbb{R}^d$, $t \geq 0$, $V_0(x)$ is a binding potential having both bound and continuum states, and V_1 is a time-periodic field of *zero average*,

$$V_1(x,t) = \sum_{j=1}^{\infty} \Big(\Omega_j(x)e^{ij\omega t} + c.c. \Big) \qquad (\text{I.2})$$

representing the external forcing.

A. Ionization

Our primary interest is whether the system ionizes under the influence of the forcing $V_1(x,t)$, as well as to determine the rate of ionization if it occurs. Ionization corresponds to delocalization of the wavefunction as $t \to \infty$. In particular we say that the system, e.g.

a Hydrogen atom, will completely ionize if the probability of finding the electron in any bounded spatial region $B \subset \mathbb{R}^d$ goes to zero as time becomes large, i.e.

$$\lim_{t \to \infty} \int_B |\psi(x,t)|^2 \, dx = 0. \tag{I.3}$$

At a heuristic level, complete ionization is expected since the potential V_1 constantly imparts energy, eventually overcoming V_0 whatever the relative strengths of the two is. Generically, ψ should thus decay. But exceptions do exist, and will be discussed.

By simple triangle inequality arguments, it is seen that ionization occurs for any initial condition $\psi(x,0) = \psi_0(x)$ in L^2 if this is the case on a dense set. For this reason, we typically take ψ_0 to be compactly supported since this allows for a more detailed analysis of the ionization process.

B. Overview

The basic strategy followed to prove ionization is to first show that the time Laplace transform of the wave function $\hat{\psi}(x,p)$, is regular and analytic for Re $p > 0$ and bounded for $p \in i\mathbb{R}$. We then show that, after subtraction of a few explicit terms, the Riemann-Lebesgue lemma applies and $\psi(x,t)$, the inverse Laplace transform of $\hat{\psi}(x,p)$, decays as $t \to \infty$, implying ionization.

The rest of the paper is organized as follows. We first review the common elements in our approach outlined above and then describe various models we studied using these methods. We start with simple one-dimensional systems where $V_0 = -2\delta(x)$ subjected to various types of forcing, including the dipole one, $V_1(x,t) = Ex \cos \omega t$. We then discuss higher dimensional systems in which $V_0(x)$ and $V_1(x,t)$ have compact support in $\mathbb{R}^d$. Finally, we describe our results for the most difficult and interesting system, the Hydrogen atom, where $V_0 = -b/|x|$, $x \in \mathbb{R}^3$.

II. GENERAL FRAMEWORK

A. Laplace reformulation of the problem

The time Laplace transform of $\psi(x,t)$,

$$\hat{\psi}(x,p) = \int_0^\infty e^{-pt} \psi(x,t) dt$$

converts the asymptotic problem (I.3) into an analytical one, which turns out to be easier to study. The Laplace transform is well defined for $\mathrm{Re}\,p > 0$ due to the existence of a continuous unitary propagator for the solution of (I.2).

After writing the transformed equation (I.1) in suitable integral forms for $\mathrm{Re}\,p \geq 0$ and using weighted L^2 spaces, the question of ionization becomes a question of sufficient regularity of the resolvent of a compact operator for $p \in \overline{\mathbb{H}}$. Here $\mathbb{H}$ denotes the right half complex plane and $\overline{\mathbb{H}}$ is its closure.

To study the regularity question, we formulate it as a Fredholm alternative problem. The transformations needed for that are relatively straightforward in simple models but can be quite challenging to find when V_0 is the Coulomb potential, the slow decay of which leads to substantial difficulties. The regularity of the resolvent translates into regularity of the solution via the Fredholm alternative. The question then becomes one of existence of acceptable solutions of an associated homogeneous problem. These solutions turn out to be closely related to eigenfunctions of the Floquet operator.

1. The Laplace space equations

Subtracting first a few asymptotic terms from ψ in (I.1) in order to ensure decay in the dual variable p and Laplace transforming the resulting equation, we obtain the following system of linear differential-difference equations

$$(H_0 + \sigma + n\omega)\hat{y}_n = y_n^{[0]} - \sum_{j \in \mathbb{Z}} \Omega_j(x)\hat{y}_{n-j} \tag{II.1}$$

where $p = i(\sigma + n\omega)$, $H_0 = -\Delta + V_0(x)$ is the reference Hamiltonian, $\hat{y}_n(x, \sigma) = \hat{y}(x, in\omega + i\sigma)$ for $n \in \mathbb{Z}$, $y_n^{[0]}$ is related to ψ_0, and σ may be restricted to $\mathrm{Re}\,\sigma \in [0, \omega)$. It is often more convenient to study the dependence on σ for $\mathrm{Re}\,\sigma \in (-\epsilon, \omega)$ and fixed n. This avoids a separate analysis of $n = 0$, σ close to 0, and of $n = -1$ and σ close to ω, needed to study the behavior of the solution near $p = 0$. The corresponding homogeneous system, used in the sequel, is

$$(H_0 + \sigma + n\omega)v_n = -\sum_{j \in \mathbb{Z}} \Omega_j(x)v_{n-j} \tag{II.2}$$

This homogeneous problem is always highly overdetermined. In the case V_1 is compactly supported in space, we obtain that for all $n < 0$, $v_n = 0$ outside the support of V_1, implying simultaneous Dirichlet and Neumann conditions for the associated differential equations.

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Generically no nonzero solution is possible, and in that case the Fredholm alternative ensures sufficient regularity in p to show complete ionization. However, proving that the homogeneous system has indeed no nonzero solution is sometimes delicate, particularly in higher dimensional problems.

B. Integral form of Laplace space equations

For inverting the differential operators, it is convenient to add a suitable purely imaginary term of the form $-i\beta(p)X_{B_R}(x)$, where $X_{B_R}(x)$ is the characteristic of the ball of radius R to both sides of the equation. (The choice of $X_{B_R}(x)$ and of $\beta(p)$ will be explained as necessary in the examples considered.) Eq. (II.1) becomes

$$\mathcal{A}_{\beta,n}\hat{y}_n \equiv (H_0 + \sigma + n\omega - i\beta X_{B_R}(r))\hat{y}_n = -i\beta X_{B_R}(r)\hat{y}_n + y_n^{[0]} - \sum_{j\in\mathbb{Z}}\Omega_j(x)\hat{y}_{n-j} \qquad \text{(II.3)}$$

C. Compact operator formulation

Inverting now the differential operators in (II.3), after possibly some further simple transformations, the system is brought to the form

$$Y = \mathfrak{T}Y_0 + \mathfrak{C}Y \qquad \text{(II.4)}$$

where $Y = \{y_n\}_{n\in\mathbb{Z}}$, $Y_0 = \{y_n^{[0]}\}_{n\in\mathbb{Z}}$, and $\mathfrak{C}$ and $\mathfrak{T}$ are compact operators. The exact form of $\mathfrak{C}$ and the associated Hilbert space will depend on the problem and will vary from case to case; for example, in the Coulomb case, it is given in (V.2). The properties of the solution of (I.1) will follow from those of (II.4) on which we focus from now on.

Applying now the Fredholm alternative, we see that existence of a sufficiently regular solution of (II.4) is equivalent to the *absence* of a regular solution of the homogeneous system corresponding to (II.4).

$$v = \mathfrak{C}v \qquad \text{(II.5)}$$

The operator $\mathfrak{C}$ is shown to be sufficiently regular: it is analytic in a suitable parameter. This is simply $\sqrt{p}$ in problems where the potential has sufficient decay in x. For the Coulomb potential, an extended parameter space needs to be introduced.

In essence, analyticity in the modified parameter ensures that the solution of (II.4) is sufficiently regular for the contour of integration of the inverse Laplace transform to be

pushed all the way to the imaginary line, where the Riemann-Lebesgue lemma implies decay of the wave function.

With a few simple exceptions, we do not present the proofs in this review; for proofs we refer to our articles, in particular[8] and[4].

D.   Connection with Floquet theory

Our analysis connects with Floquet theory in a number of ways, that we briefly sketch. Let K be the quasi-energy operator in Floquet theory

$$(Ku)(x, \theta) = \left(-i\frac{\partial}{\partial \theta} - \Delta + V_0(x) + V_1(x, \theta) \right) u(x, \theta); \ x \in \mathbb{R}^d, \ \theta \in S^1_{2\pi/\omega} \qquad \text{(II.6)}$$

Then, letting

$$u(x, \theta; \sigma) = \sum_{n \in \mathbb{Z}} y_n^{[1]}(x; \sigma) e^{in\omega\theta} \qquad \text{(II.7)}$$

be the solution of the eigenvalue equation

$$Ku = -\sigma u \qquad \text{(II.8)}$$

we get an equation for the $y_n^{[1]}$ which is identical to the homogeneous part of equation (II.1). (However, the functional spaces are generally different.) Solutions of (II.8) with $u \in L^2(\mathbb{R}^d \times S^1_{2\pi/\omega})$ correspond to L^2 eigenfunctions of K.

Complete ionization clearly requires the absence of such solutions, *i.e.*, of a point spectrum of (II.8), $\sigma_p(K)$. Otherwise, if $u(x, \theta)$ is an eigenfunction of K, then $e^{i\sigma t}u(x, t)$ would be a space localized solution of the Schrödinger equation.

In fact, we show that the converse is also true in the models we considered.

Theorem 1 *In our examples, we show that (II.5) has no nontrivial solutions,* i.e. *ionization is complete,* **iff** $\sigma_p(K) = \emptyset$.

Remark 2 Theorem 1 rules out the existence of singular continuous spectrum of K in the models we study. Under assumptions of smoothness and sufficiently fast spatial decay of V_0 and V_1, Galtbayar, Jensen and Yajima[12] obtained absence of singular continuous spectrum as well as an asymptotic power series for ψ in powers of $t^{-1/2}$ as $t \to \infty$, assuming $\sigma_p(K) = \emptyset$. Möller and Skibsted proved in[16] the equivalence of absence of point spectrum and ionization for a large class of systems subject to periodic fields, with weaker decay conditions, for dilation-analytic potentials.

The main problem for us is finding $\sigma_p(K)$. In most cases $\sigma_p(K)$ is empty, but, as shown in the examples below, there are situations when it is not, and in this latter case, complete ionization does not occur.

III. ONE DIMENSIONAL MODELS WITH $V_0(x) = -2\delta(x)$

The zero range (delta–function) attractive potential is much used in the literature to model short range attractive potentials. In time dependent settings it captures qualitative and sometimes even quantitative features of more realistic models. In one dimension, the unperturbed Hamiltonian H_0 has, in suitable units, the form

$$H_0 = -\frac{d^2}{dx^2} - 2\,\delta(x), \quad -\infty < x < \infty. \tag{III.1}$$

and belongs to the class $K_1{}^9$. H_0 has a single bound state

$$u_b(x) = e^{-|x|} \tag{III.2}$$

with energy $-\omega_0 = -1$. It also has continuous uniform spectrum on the positive real line, with generalized eigenfunctions

$$u(k, x) = \frac{1}{\sqrt{2\pi}} \left(e^{ikx} - \frac{1}{1 + i|k|} e^{i|kx|} \right), \quad -\infty < k < \infty$$

and energies k^2.

Note 3 It turns out that for delta-potential models it suffices to look at the projection of the wave function on the bound state,

$$\theta(t) = \langle \psi, u_b \rangle = \int_{-\infty}^{\infty} u_b(x)\psi(x, t)dx$$

A. Parametric forcing

Parametric forcing simply means

$$V_1(x, t) = -2\,\eta(t)\delta(x); \quad \eta(t) = \sum_{j=1}^{\infty} \left(C_j e^{i\omega jt} + c.c. \right) \tag{III.3}$$

The following condition turns out to be crucial to the nature of $\sigma_p(K)$.

Genericity condition (g). Consider the right shift operator T on $l_2(\mathbb{N})$ given by

$$T(C_1, C_2, ..., C_n, ...) = (C_2, C_3, ..., C_{n+1}, ...)$$

We say that $\mathbf{C} \in l_2(\mathbb{N})$ is *generic with respect to T* if the Hilbert space generated by all the translates of $\mathbf{C}$ contains the vector $e_1 = (1, 0, 0, ...,)$ (which is the kernel of T):

$$e_1 \in \bigvee_{n=0}^{\infty} T^n \mathbf{C} \tag{III.4}$$

(where the right side of (III.4) denotes the closure of the space generated by the $T^n \mathbf{C}$ with $n \geq 0$.) This condition is generically satisfied, and is obviously weaker than the "cyclicity" condition $l_2(\mathbb{N}) \ominus \bigvee_{n=0}^{\infty} T^n \mathbf{C} = \{\mathbf{0}\}$, which is also generic[17].

A case which satisfies (III.4), (but fails the cyclicity condition) corresponds to η being a trigonometric polynomial, namely $\mathbf{C} \not\equiv \mathbf{0}$ but $C_n = 0$ for all large enough n. (We can in fact replace $\mathbf{e}_1$ in (III.4) by $\mathbf{e}_k$ with any $k \geq 1$.)

Under this genericity condition, we prove that complete ionization occurs.

A simple example which *fails* (III.4) is $C_n = -r\lambda^n$ for $n \geq 1$, for some $\lambda \in (0, 1)$. In this case the space generated by $T^n \mathbf{C}$ is one-dimensional. The associated η is

$$\eta(t) = 2r\lambda \frac{\lambda - \cos(\omega t)}{1 + \lambda^2 - 2\lambda \cos(\omega t)} \tag{III.5}$$

For this case we prove that there are values of r and λ for which ionization is incomplete and $\psi(x, t)$ does not delocalize as $t \to \infty$.

The precise results are given below.

Theorem 4 *Under the assumption (g) the survival probability $P(t)$ of the bound state u_b, $|\theta(t)|^2$ tends to zero as $t \to \infty$.*

Theorem 5 *For $\psi_0(x) = u_b(x)$ there exist values of λ, ω and r in (III.5), for which $|\theta(t)| \not\to 0$ as $t \to \infty$.*

For the proof of these theorems, see[4].

1.   *Harmonic η*

In[6] we investigated in detail the case when η is harmonic,

$$\eta(t) = r \sin \omega t$$

Sketch of proof of ionization. In this case the Laplace transform of $\theta(t)$ is clearly independent of x, and (II.5) is just an infinite scalar difference system of the form

$$\sqrt{\sigma + n\omega}\, v_n = v_n - \frac{1}{2}v_{n+1} - \frac{1}{2}v_{n-1} \tag{III.6}$$

The proof that v_n is zero if $n < 0$ and subsequently showing that (II.5) has no nonzero solution are then particularly simple. Multiplying both sides of (III.6) by $\overline{v_n}$ and summing over n, which in the associated Hilbert space must converge, we get

$$\sum_{n \in \mathbb{Z}} \sqrt{\sigma + n\omega}\, v_n \overline{v_n} = \sum_{n \in \mathbb{Z}} v_n \overline{v_n} - \frac{1}{2}\sum_{n \in \mathbb{Z}} v_{n+1}\overline{v_n} - \frac{1}{2}\sum_{n \in \mathbb{Z}} v_{n-1}\overline{v_n} \tag{III.7}$$

The second and third terms on the right side of (III.7) are complex conjugates of each-other, so the right side of (III.7) is real. For $\operatorname{Im}\sigma \neq 0$, we see that $\operatorname{Im}(\sqrt{\sigma + n\omega}\, v_n \overline{v_n})$ has the same sign as $\operatorname{Im}\sigma$. The left side cannot be real and hence the equation has only zero solutions.

In the (most interesting) case $\operatorname{Im}\sigma = 0$, we see that for $n < 0$ the sign of $\operatorname{Im}(\sqrt{\sigma + n\omega}\, v_n \overline{v_n})$ does not depend on n, and for $n \geq 0$, $\sqrt{\sigma + n\omega}\, v_n \overline{v_n}$ is real. Clearly, since $\sqrt{n\omega}$ is purely imaginary for negative n and $|v_n|^2$ is purely real, we must have $v_n = 0$ for all $n < 0$. Induction shows that $v_n = 0$ for all n. From this result and regularity considerations, complete ionization follows.

The proofs in more complex models follow completely different lines.

2. *Dependence of ionization rates on the parameters in the harmonic case*

We also obtained a detailed picture of how the decay of $\theta(t)$ depends on r and ω, when $\psi_0 = u_b$. For all r, ω, $\theta(t)$ can be (uniquely) written in the form

$$\theta(t) = e^{-\gamma(r;\omega)t + it}F_\omega(t) + \sum_{m=-\infty}^{\infty} e^{(mi\omega - i)t}h_m(t) \tag{III.8}$$

where F_ω is periodic of period $2\pi\omega^{-1}$ and the functions $h_m(t)$ are equal to the Borel sums of their asymptotic series in powers of $t^{-1/2}$. The decomposition is the time-periodic analog of a resonance expansion, with the added information about Borel summability. The infinite sum in (III.8) is convergent for all $t > 0$, and rapidly so if t is large. For large t we have

$$\sum_{m=-\infty}^{\infty} e^{(mi\omega - i)t}h_m(t) = O(r^2 t^{-3/2})$$

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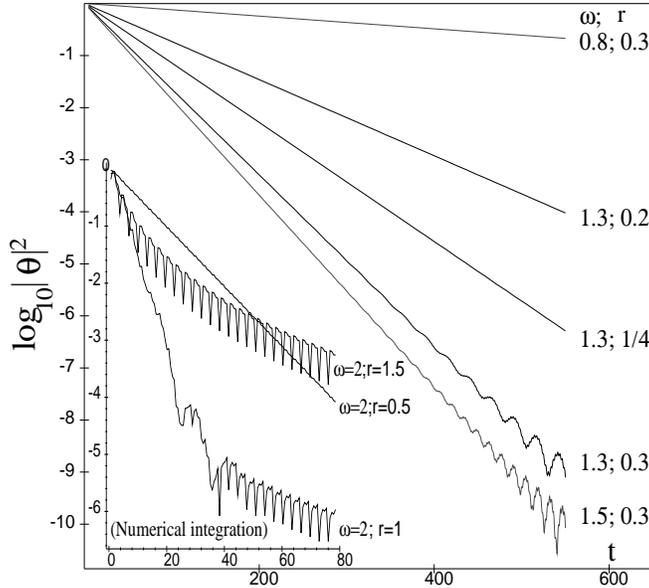
FIG. 1. Plot of $\log_{10} |\theta(t)|^2$ vs. time in units of ω_0^{-1} for several values of ω and r.

For small r and ω^{-1} not too close to an integer we get an exponential decay of $|\theta(t)|^2$, with a decay exponent $\Gamma = 2\gamma(r,\omega) \sim r^{2(1+\lfloor \omega^{-1} \rfloor)}$ where $\lfloor \omega^{-1} \rfloor$ is the integer part of ω^{-1}. For $\omega > 1$, this corresponds to $\Gamma \sim \Gamma_F$, the Fermi golden rule constant. At times large compared to Γ^{-1}, $|\theta(t)|^2$ decays as t^{-3}. The picture becomes much more complicated when r is large and/or ω^{-1} is an integer. In particular there is no monotonicity in $|\theta(t)|$ as a function of r, see Fig. 1.

The ionization results for this very simple delta function model compares relatively well with actual experimental ones obtained for Rydberg atoms, using an effective value of r, see Fig. 2.

B. Non-parametric delta-function forcing

Using the same H_0 as in (III.1) we considered in[18] two different forcings,

$$V_1^{(1)}(x,t) = 2\delta(x-a)r\sin\omega t \tag{III.9}$$

and

$$V_1^{(2)}(x,t) = 2\left[\delta(x-a) - \delta(x+a)\right]r\sin\omega t \tag{III.10}$$

where $a \in \mathbb{R}$ is a new parameter. When $a = 0$, (III.9) reduces to the previous case. The spatial part of V_1 in (III.10) suggests a dipolar type forcing. (The actual dipole forcing is

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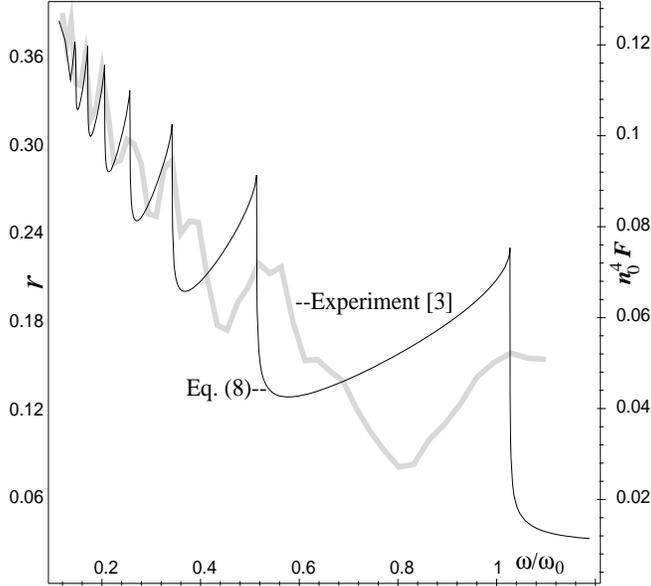
FIG. 2. Threshold amplitudes for 50% ionization vs. ω/ω_0, calculated from the delta function model, and the experiment, see[6] for details.

considered in §III C.)

We prove in both cases that the survival probability goes to zero as $t \to \infty$ for almost all values of r, ω and a. The decay is initially exponential, followed by a t^{-3} law if ω is not close to resonances and r is small; otherwise, the exponential term is not visible and Fermi's golden rule fails. We further show that there are exceptional sets of parameters r, ω and a for which the survival probability $|\theta(t)|^2$ never decays to zero, corresponding to the Floquet operator having a bound state. These values form a two-dimensional manifold in the parameter space. We show examples of decay of $|\theta(t)|^2$, and of lack of decay, in Fig. 3.

This lack of decay is presumably due to the exceptional nature of the delta function forcings in one dimension. In fact, we show similar behavior even in the absence of a binding potential, i.e., $V_0 = 0, V_1 = V_1^{(2)}$, permitting a free particle to be trapped by the harmonically oscillating delta function potential.

C.   Dipole forcing

A common model of ionization in the physical literature is the dipole model. The most realistic model of (monochromatic) radiation would be to take $V_1(x,t) = F(kx - \omega t)$ with

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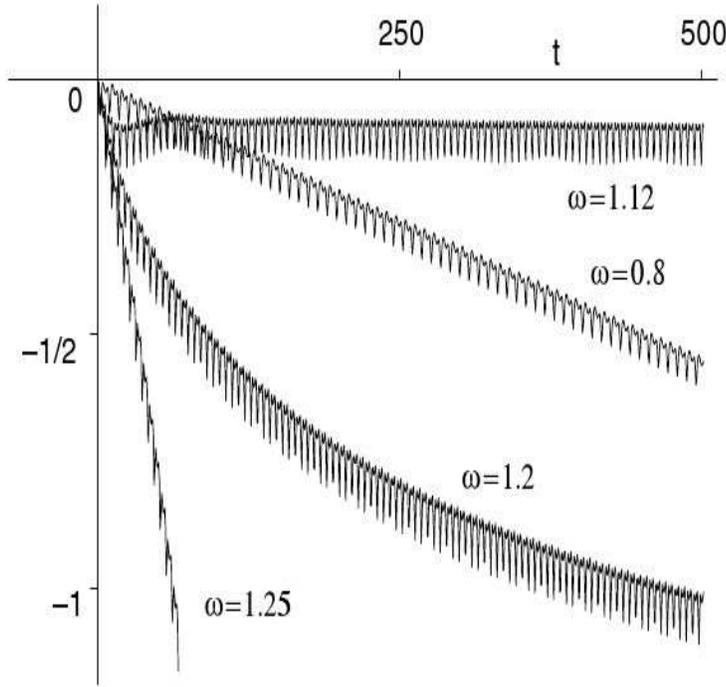
FIG. 3. Plot of $\log_{10} |\theta(t)|^2$ vs. time for $V_1^{(2)}$, for $a = 0.59$, $r = 1$ and several values of ω, including a critical one $\omega \approx 1.12$, for which there is no decay. See[18] for details.

$F(\theta)$ a periodic function. By Taylor expanding about $x = 0$ to linear order and using a gauge transformation to remove the constant term we obtain the frequently used dipole model. The model has the same H_0 as in(III.1) and $V_1(x,t) = E(t)x$ with $E(t)$ being $2\pi/\omega$-periodic. This approximation is valid when the wavelength of $F(kx - \omega t)$ is large compared to the size of the bound state.

In the dipole case, the analysis was simplified considerably by using the Zak transform:

$$\mathcal{Z}[\psi](x, \sigma, t) = \sum_{j \in \mathbb{Z}} e^{i\sigma(t + 2\pi j/\omega)} \psi(x, t + 2\pi j/\omega) \tag{III.11}$$

The Zak transform is related to the Laplace transform in a fairly straightforward manner via the Poisson Summation formula:

$$\mathcal{Z}[\psi](x, \sigma, t) = \frac{\omega}{2\pi} \sum_{n \in \mathbb{Z}} \hat{\psi}(x, -i(\sigma + n\omega)) e^{-in\omega t} \tag{III.12}$$

The key property of the Zak transform is that it commutes with periodic operators, as well as with the periodic parts of other integral operators. This is useful for the following reason.

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In the time domain, one can show that $\psi(0,t)$ satisfies (modulo a gauge transformation) the integral equation:

$$\psi(0,t) = [e^{i\partial_x^2 t}\psi_0](0,t) + \sqrt{\frac{i}{\pi}} \int_0^t \exp\left(\frac{i(c(t) - c(t-s))^2}{4s}\right) \psi(0,t-s)\frac{ds}{\sqrt{s}}$$

where $c(t)$ is periodic and $c''(t) = E(t)$. The Zak transform "commutes" with the integral operator in the following manner:

$$\mathcal{Z}[\psi](0,t) = \mathcal{Z}([e^{i\partial_x^2 t}\psi_0])(0,t)$$
$$+ \sqrt{\frac{i}{\pi}} \int_0^\infty \exp\left(\frac{i(c(t) - c(t-s))^2}{4s}\right) \mathcal{Z}[\psi](0,t-s)\frac{ds}{\sqrt{s}}$$

It is shown in[5] that this integral operator is compact after Zak transformation, and that therefore the same theory as described in Section II B and II C applies. This formulation also allows us to prove a result analogous to Theorem 1.

Using Theorem 1, we can then provide a general condition for ionization to hold:

Theorem 6 *Suppose $E(t)$ is a trigonometric polynomial. Then for any $\psi_0(x) \in L^2(\mathbb{R})$ complete ionization occurs, i.e. (I.3) holds. If $\psi_0(x) \in L^1(\mathbb{R}) \cap L^2(\mathbb{R})$, then $\|\psi(x,t)\|^2_{L^2([-L,L])}$ approaches zero at least as fast as t^{-1}.*

1. Sketch of the proof of ionization

The approach to proving Theorem 6 is as follows. By Theorem 1, if ionization fails, it fails because $\sigma_p(K) \neq \emptyset$ which in turn implies the existence of a nonzero eigenvector u of the quasi-energy operator K from Section II D. To prove ionization, we need to prove that this cannot occur.

The proof that no such $u := u(x,\theta)$ exists is performed in the following manner. It is fairly straightforward to construct time-periodic solutions of (II.8) $u_L(x,\theta)$ and $u_R(x,\theta)$ which are valid for $x \leq 0$ and $x \geq 0$ respectively, and to show that this set of solutions is complete. Of course, for u to be continuous, we need that $u_L(0,\theta) = u_R(0,\theta)$.

The technical details of the proof consist of showing that one cannot continuously match $u_L(x,\theta)$ to $u_R(x,\theta)$ at $x = 0$. The specific method of proving this is to analytically continue $u(0,\theta)$ in the complex θ plane. By analytically continuing $u_R(0,\theta)$, we obtain decay conditions in a certain region of the complex θ-plane. By analytically continuing $u_L(0,\theta)$ in θ we

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obtain decay conditions in a complementary region of the complex plane. The two decay conditions can be combined to show that $u(0, \theta)$ is bounded in the entire complex θ-plane, and is therefore constant (and zero). The details are contained in[5] (Section 2).

The requirement that $E(t)$ be a trigonometric polynomial is needed because that implies $u(0, \theta)$ has finite exponential order. Using the finite exponential order is a key step in the proof, making use of the Phragmen-Lindelof theorem.

IV. HIGHER DIMENSIONAL MODELS WITH COMPACTLY SUPPORTED POTENTIALS

We consider now the case when $x \in \mathbb{R}^d$, $d = 1, 2, 3$, with $V_0(x)$ and $V_1(x, t)$ supported in a compact domain D,[7], with

$$V_0(x) = \chi_D(x)V_0(x); \quad \Omega_j(x) = \chi_D(x)\Omega_j(x) \tag{IV.1}$$

where $\chi_D(x)$ is the characteristic function of D.

We first show the following convenient condition of ionization.

Theorem 7 *In the setting (IV.1), if (II.5) has a nontrivial solution, then*

$$v_n(x) = 0 \quad \text{for all} \ \ n < 0 \ \text{and} \ x \notin D \tag{IV.2}$$

Proof. Assume there exists a nontrivial solution in the corresponding $\mathcal{H}$ to the system

$$(-\Delta + \sigma + n\omega)v_n = -V_0 v_n - \sum_{j \in \mathbb{Z}} \Omega_j(x) v_{n-j} \tag{IV.3}$$

We multiply (IV.3) by $\bar{v}_n$, integrate over a ball B containing D, sum over n (the sum converges in our space) and take the imaginary part of the resulting expression. Noting that

$$\overline{\sum_{j,n \in \mathbb{Z}} \Omega_j(x) v_{n-j} \bar{v}_n} = \sum_{j,n \in \mathbb{Z}} \Omega_{-j} \bar{v}_{n-j} v_n = \sum_{j,n \in \mathbb{Z}} \Omega_j \bar{v}_{n+j} v_n$$

$$= \sum_{j,m \in \mathbb{Z}} \Omega_j(x) \overline{v_m} v_{m-j} \tag{IV.4}$$

so the sum (IV.4) is real, we get

$$0 = \mathrm{Im} \left(-\sigma \sum_{n \in \mathbb{Z}} \|v_n\|^2 + \int_B \sum_{n \in \mathbb{Z}} dx \bar{v}_n \Delta v_n \right)$$

$$= -\mathrm{Im}\sigma \sum_{n \in \mathbb{Z}} \|v_n\|^2 + \frac{1}{2i} \int_{\partial B} \left(\sum_{n \in \mathbb{Z}} \bar{v}_n \nabla v_n - v_n \nabla \bar{v}_n \right) \cdot \mathbf{n} \, dS \tag{IV.5}$$

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We take $d = 3$ (the analysis is simpler in one or two dimensions). It is convenient to decompose v_n using spherical harmonics; we write

$$v_n = \sum_{l \geq 0, |m| \leq l} R_{n,l,m}(r) Y_l^m(\theta, \phi). \tag{IV.6}$$

The last integral in (IV.5), including the prefactor, then equals

$$-8\pi\, i\, r_B^2 \sum_{n \in \mathbb{Z}} \sum_{m,l} \left[\overline{R}_{n,m,l} R'_{n,m,l} - \overline{R'}_{n,m,l} R_{n,m,l} \right]$$

$$= -8\pi\, i\, r_B^2 \sum_{n \in \mathbb{Z}} \sum_{m,l} W[\overline{R}_{n,m,l}, R_{n,m,l}] \tag{IV.7}$$

where r_B is the radius of B and $W[f, g]$ is the Wronskian of f and g. On the other hand, since V and Ω are compactly supported in B, we have outside of B

$$\Delta v_n - (\sigma + n\omega) v_n = 0 \tag{IV.8}$$

Then, by (IV.6), $R_{n,l,m}$ satisfy for $r > r_B$ the equation

$$R'' + \frac{2}{r} R' - \frac{l(l+1)}{r^2} R = (\sigma + n\omega) R \tag{IV.9}$$

where we have suppressed the subscripts. Let $g_{n,l,m} = r R_{n,l,m}$. Then for the $g_{n,l,m}$ we get

$$g'' - \left[\frac{l(l+1)}{r^2} + (\sigma + n\omega) \right] g = 0 \tag{IV.10}$$

thus

$$\overline{R} R' = \frac{\overline{g} g'}{r^2} - \frac{|g|^2}{r^3} \tag{IV.11}$$

and

$$r^2 W[\overline{R}, R] = W[\overline{g}, g] =: W_n. \tag{IV.12}$$

Multiplying (IV.10) by $\overline{g}$, the conjugate of (IV.10) by g and subtracting, we get for $r > r_B$,

$$W'_n = (\sigma - \overline{\sigma}) |g|^2 = 2i |g|^2 \mathrm{Im}\, \sigma \tag{IV.13}$$

Remark 8 *Simple estimates using the resolvent of the free Laplacian[7] imply that, for some c_n,*

$$v_n(x) = \frac{e^{-\kappa_n |x|}}{|x|} \left(c_n(\theta, \phi) + O(|x|^{-1}) \right) \text{ as } |x| \to \infty \tag{IV.14}$$

where

$$\kappa_n = \sqrt{-ip} = \sqrt{\sigma + n\omega} \text{ (when } p \in \mathbb{H}, \ \kappa_n \text{ is in the fourth quadrant)} \tag{IV.15}$$

Let us consider two cases of (IV.3).

Case (i): $\operatorname{Im}\sigma < 0$. By Remark 8 we have

$$g \sim C e^{-\kappa_n r}(1 + o(1)) \quad \text{as} \quad r \to \infty \tag{IV.16}$$

There is a one-parameter family of solutions of (IV.10) satisfying (IV.16) and the asymptotic expansion can be differentiated[22]. We assume, to get a contradiction, that there exist n for which $g_n \neq 0$. For these n we have, using (IV.16), differentiability of this asymptotic expansion and (IV.15) that

$$\frac{1}{2i} \lim_{r \to \infty} |g_n|^{-2} W_n = -\operatorname{Im}\kappa_n > 0. \tag{IV.17}$$

It follows from (IV.13) and (IV.17) that $\frac{1}{2i}W_n$ is strictly positive for all $r > r_B$ and all n for which $g_n \neq 0$. This implies that the last term in (IV.5) is a sum of positive terms which shows that (IV.5) cannot be satisfied.

Case (ii): $\operatorname{Im}\sigma = 0$. For $n > 0$ there exists only one solution g of (IV.10) which decays at infinity (cf. Remark 8 and the discussion in Case (i)), and since (IV.10) has real coefficients this g must be a real as well (up to a multiplicative constant); therefore we have $W_n = 0$ for $n \geq 0$.

For $n < 0$, we use Remark 8 (and differentiability of the asymptotic expansion as in Case (i)) to calculate the Wronskian W_n of $g, \bar{g}$ in the limit $r \to \infty$: $W_n = |c_n|^2(1 + o(1))$. Since for $\operatorname{Im}\sigma = 0$, W_n is constant, cf. (IV.13), it follows that W_n is exactly equal to $|c_n|^2$. Thus, using (IV.5) and (IV.7) we have

$$v_n(x) = 0 \quad \text{for all} \quad n < 0 \text{ and } |x| > r_B \tag{IV.18}$$

Outside D we have $\mathfrak{O}v_n = 0$, where $\mathfrak{O}$ is the elliptic operator $-\Delta + \sigma + n\omega$. The proof follows immediately from (IV.18), by standard unique continuation results[13,15,21] (in fact, $\mathfrak{O}$ is analytic hypoelliptic). $\square$

To go from Theorem 7 to complete ionization requires further work and further assumptions. A particular such example is the parametric forcing of a well of arbitrary shape.

Theorem 9 *Let*

$$V_0(x) = V_D \,\chi_D(x); \quad V_1(x,t) = 2\Omega_D \,\chi_D(x) \sin \omega t \tag{IV.19}$$

where V_D and Ω_D are arbitrary nonzero constants. Then, starting with $\psi_0(x) \in H^2(\mathbb{R}^3)$, the system will completely ionize.

A. Sketch of proof of Theorem 9

It is convenient to Fourier transform the system (IV.3) in x. In view of (IV.2), for $n < 0$, $v_n = 0$ outside D. We then have, for $n < 0$,

$$\check{y}_n := \int_{\mathbb{R}^3} v_n e^{-ik\cdot x} dx = \int_D v_n e^{-ik\cdot x} dx \tag{IV.20}$$

and

$$-k^2 \check{y}_n = -k^2 \int_{\mathbb{R}^3} v_n e^{-ik\cdot x} dx = \int_{\mathbb{R}^3} \Delta v_n e^{-ik\cdot x} dx = \int_D \Delta v_n e^{-ik\cdot x} dx \tag{IV.21}$$

For the setting (IV.19) and $n < -1$, (IV.3) reads

$$(k^2 + \sigma + n\omega)\check{y}_n = -V_D \check{y}_n + i\Omega_D (\check{y}_{n+1} - \check{y}_{n-1}) \tag{IV.22}$$

Remark 10 *For $n \leq -1$, the functions $\check{y}_n$ are entire of exponential order one; more precisely, if B is a ball containing D we have*

$$|\check{y}_n(k)| \leq \sqrt{\text{Vol}(D)} \; e^{|k|r_B} \; \|v_n\|_{L^2(D)} \tag{IV.23}$$

Proof. This follows immediately from the definition of $\check{y}$. (See also[24] for a comprehensive characterization of the Fourier transform of a compactly supported distribution.)

Proposition 11 *The generating function*

$$Y(k, z) = \sum_{m \geq 0} \check{y}_{-m-2}(k) z^m \tag{IV.24}$$

is entire in k and analytic in z for $|z| < 1$.

A straightforward calculation shows that Y satisfies the equation

$$MY - z\frac{\partial Y}{\partial z} - i\beta \left(z - \frac{1}{z} \right) Y = i\beta \check{y}_{-1} + i\beta \frac{\check{y}_{-2}}{z} \tag{IV.25}$$

where

$$M = \omega^{-1}(k^2 + \sigma - 2\omega + V_D) \tag{IV.26}$$

and $\beta = \Omega_D/\omega$. The solution of (IV.25) is

$$Y = z^M e^{-i\beta(z+z^{-1})} \left[C(k) - i\beta \int_0^z e^{i\beta(s+s^{-1})} \left(\frac{\check{y}_{-1}}{s^{M+1}} + \frac{\check{y}_{-2}}{s^{M+2}} \right) ds \right] \tag{IV.27}$$

where the integral follows a path in which 0 is approached along the negative imaginary line.

Remark 12 *Proposition 11 implies $C(k) \equiv 0$.*

Proof. It is easy to check that otherwise the limit of $Y(k,z)$ as $z \to 0$ along $i\mathbb{R}^-$ would not exist. $\square$

Thus

$$Y(k,z) = -i\beta z^M e^{-i\beta(z+z^{-1})} \int_0^z e^{i\beta(s+s^{-1})} \left(\frac{\check{y}_{-1}(k)}{s^{M+1}} + \frac{\check{y}_{-2}(k)}{s^{M+2}} \right) ds \tag{IV.28}$$

Let

$$F(M) = \oint_{\mathcal{C}} \frac{e^{i\beta(s+s^{-1})}}{s^M} ds \tag{IV.29}$$

Proposition 13 *We have*

$$\check{y}_{-1}(k)F(M+1) + \check{y}_{-2}(k)F(M+2) = 0 \tag{IV.30}$$

Proof. This follows immediately from the discussion above. $\square$

Proposition 14 *For every large $N \in \mathbb{N}$, $F(z)$ has exactly one zero of the form $z_N = N + o(1)$. For large N we have $F(1 + z_N) \neq 0$.*

Proposition 15 *Relation (IV.30), with $\check{y}_{-1}(k), \check{y}_{-2}(k)$ entire of exponential order one (cf. Remark 10) implies*

$$\check{y}_{-1}(k) = \check{y}_{-2}(k) = 0 \ \forall k \in \mathbb{C}^3 \tag{IV.31}$$

and then

$$y_n(x) = 0 \ \forall n \in \mathbb{Z} \text{ and almost all } x \in \mathbb{R}^d \tag{IV.32}$$

V. THE COULOMB POTENTIAL

We now consider the case of the Coulomb potential, $V_0(x) = -b/r$, $r = |x|$ with V_1 compactly supported in space,[8]. Writing $\psi = \psi_0 e^{-t} + y$ in (I.1) (in order to ensure decay in the dual variable p), and Laplace transforming the resulting equation, we obtain (II.3) with $H_0 = H_C = -\Delta - b/|x|$. As is well known, the eigenvalues of H_C are $E_n = -b/n^2, n \in \mathbb{N}$ where the multiplicity of E_n is n^2.

We proceed as in §II B where $\chi_{B_R}(x)$ is the characteristic function of $B_R = \{x : |x| < R\}$ and

$$\beta = \beta(p) = \begin{cases} c \text{ if } |\text{Im } p| \in [0, p_c] \\ 0 \text{ otherwise} \end{cases} \tag{V.1}$$

where $p_c > b^2/4$ (the ground state eigenvalue of $-H_c$). (The artificially created discontinuity of the modified problem at $|\mathrm{Im}p| = p_c$ poses no problem, since p_c is arbitrary).

The following Hilbert space is natural here: $\mathcal{H} = \{\{v_n\}_{n\in\mathbb{Z}} =: Y, \ v_n \in L^2(B_R)\}$ with

$$\|Y\|^2 = \sum_{n\in\mathbb{Z}}(1 + |n|)^{4/3}\|v_n\|^2_{L^2(B_R)} < \infty \tag{V.2}$$

The large $|n|$ decay of v_n required by this norm translates into decay for large $|p|$ of $v_n(x, \sigma)$ (where in accordance with ref.[8] we have set $\sigma = -ip_1$) needed to analyze its inverse Laplace transform.

The operator $\mathfrak{C} : \mathcal{H} \to \mathcal{H}$ in (II.4) is given here by

$$\{\mathfrak{C}Y\}_n = \chi_{B_R}(x)\mathfrak{R}_{\beta,n}\chi_{B_R}(x)\left[-i\beta v_n(\sigma, x) - \sum_{j\in\mathbb{Z}}\Omega_j(x)v_{n-j}(\sigma, x)\right] \tag{V.3}$$

where $\mathfrak{R}_{\beta,n}$ denotes the resolvent $\mathcal{A}_{\beta,n}^{-1}$.

As mentioned before, the term $-i\beta\chi_R(x)$ is introduced to ensure invertibility of the operators; the space-compactness of $\chi_{B_1}(r)$ is needed for the sandwiched operator $\chi_{B_1}\mathfrak{R}_{\beta,n}\chi_{B_1}$ to be compact. The poles of the resolvent $\mathfrak{R}_{\beta,n}$, are pushed into the left-half p -domain. However, the eigenvalues close to 0 of H_C correspond to large orbitals and a compact perturbation has a small effect on them. As a result, the poles of $\mathfrak{R}_{\beta,n}$ accumulate tangentially towards the positive imaginary axis at $p = 0$, and $p = 0$ remains an essential singularity (though now an integrable one). The analysis at 0 is delicate, since detailed knowledge of the nature of the singularity is needed in the final decay estimates of the inverse Laplace transform.

With $Y = \{\hat{y}_n(x, \sigma)\}_{n\in\mathbb{Z}}$ and $\tilde{Y}^{[0]} = \{\mathfrak{R}_{\beta,n}\chi_{B_1}y_n^{[0]}\}_{n\in\mathbb{Z}}$ we get, as in the general case,

$$Y = \tilde{Y}^{[0]} + \mathfrak{C}Y \tag{V.4}$$

Compactness of $\mathfrak{R}_{\beta,n}$ ultimately translates into compactness of $\mathfrak{C}$ in $\mathcal{H}$. The proof is delicate, since norm-decay in n is related to space decay of the Coulomb potential, which in turn is very slow.

To describe the essential singularity of $\mathfrak{R}_{\beta,n}$ at the origin, we extend the parameter space from σ to $X = (\lambda, Z)$, $\lambda = \sqrt{\sigma}$, $Z = e^{i\pi b/(2\lambda)}$. We rewrite $\mathfrak{C}$ in a suitable way in the extended parameter space and show that it is analytic in X in a neighborhood of $\overline{\mathbb{D}_\epsilon} \times \overline{\mathbb{D}_1}$ where $\mathbb{D}_a$ is the disk of radius a. This analyticity is inherited by $Y = (I - \mathfrak{C})^{-1}Y_0$ as long as $(I - \mathfrak{C})^{-1}$

exists. Analyticity in X provides all the information needed to show that ψ decays like $t^{-5/6}$ for large t, for ψ_0 compactly supported in x and angular momentum $\mathbf{L}$.

Ruling out nonzero $\mathbf{v}$ for Im $\sigma < 0$ is relatively straightforward and relies on self-adjointness arguments in the Hilbert space $\mathcal{H}$. However, for $\sigma \in [0, \omega) \subset \mathbb{R}$ as before, analyzing the Floquet equation (II.2) and determining that there is no nonzero solution in $\mathcal{H}$ is a difficult problem. Unlike in one-dimensional problems, we do not have exact representation of solutions to work with. Further, an infinite system of coupled linear differential equations, one equation for each temporal Fourier mode (recalling (II.2)), is involved. An important intermediate step is the following result, similar to Theorem 7.

Theorem 16 *If there exists a nonzero solution of (II.2) $\mathbf{v} \in \mathcal{H}$, then $\mathbf{v}$ has the further property that for $r > 1$,*

$$v_n = 0 \ for \ all \ \ n < 0 \tag{V.5}$$

Cauchy-Kowaleski type of arguments show that for a nonzero solution $\mathbf{v}$, there must exist some integer n_0 so that either (a) v_{n_0} or (b) $\partial v_{n_0}/\partial r$ is nonzero at $r = 1$, the edge of support of V_1.

Using Theorem 16, the asymptotics of a non zero solution v_n to (II.2) as $n \to -\infty$ can be extracted by using a novel rigorous WKB method. The heuristic argument is presented in the following when Ω_j in (II.2) are spherically symmetric and

$$\Omega_j = 0 \text{ for } |j| > M \text{ and } \Omega_j > 0 \text{ on their support } \{x : |x| \leq 1\}$$

We use a spherical harmonics representation

$$v_{n_0-k} = \frac{Y_{l,m}}{r} g_{n_0-k}(r)$$

in equation (II.2) and note that

$$\mathcal{L}_k g_{n_0-k} \equiv g''_{n_0-k} + \left[(n_0 - k)\omega + \sigma + \frac{b}{r} - \frac{l(l+1)}{r^2}\right] g_{n_0-k} = -\sum_{j=-M}^{M} \Omega_j(r) g_{n_0-k-j}$$

In either of the cases (a) or (b), we substitute into the above equation the following ansatz for the asymptotics for $k \gg 1$, $r = O(1)$:

$$g_{n_0-k}(r) = \frac{c_*}{\Gamma(2k/M + 1)} \exp\left[k \log f_0(r) + \sum_{j=1}^{M} k^{1-j/M} f_j(r)\right]$$

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and demand that the error terms of order $O(k^{2-2j/M})$ vanish for $j = 0, .., M$. This leads to $(M + 1)$ first order differential equations for f_j. To leading order

$$f_0(r) = \left[\int_r^1 \sqrt{\Omega_{-M}(s)} ds\right]^{2/M}$$

(we recall the assumption of positivity of Ω_j). The expressions for $f_j(r)$ for $j \geqslant 1$ are more complicated and involve arbitrary constants to be determined from the information for small k at $r = 1$ and are different for cases (a) and (b). Because of the presence of $l(l + 1)r^{-2}$ term in $\mathcal{L}_k$, the formal error is $O(r^{-2})$, which is $O(k^2)$ when $r = O(k^{-1})$. Therefore, we seek uniform asymptotics in the revised form

$$g_{n_0-k}(r) \sim c_* \exp\left[k \log f_0(r) + \sum_{j=1}^{M} k^{1-j/M} f_j(r)\right] \frac{G(\alpha kr)}{\Gamma(2k/M + 1)}, \qquad (\text{V.6})$$

where $\mathfrak{s}(r) = \int_r^1 \sqrt{\Omega_{-M}(s)}$ and $\alpha = 2\sqrt{\Omega_{-M}(0)}/\mathfrak{s}(0)$. Then, if $\alpha kr = \zeta = O(1)$, we find to leading order $G(\zeta) \sim G_0(\zeta)$ where

$$G_0'' - 2G_0' - \frac{l(l + 1)}{\zeta^2} G_0 = 0$$

We require $G_0(\zeta) \sim 1$ as $\zeta \to \infty$. This leads to

$$G_0(\zeta) = \sqrt{\frac{2}{\pi}} e^\zeta \zeta^{1/2} K_{l+\frac{1}{2}}(\zeta)$$

Thus for nonzero g_{n_0-k}, the constant multiple in (V.6) is nonzero. On the other hand, the asymptotic behavior as $\zeta \downarrow 0$, $G_0(\zeta) \sim c_* \zeta^{-l}$ implying a singular behavior of g_{n_0-k}/r at $r = 0$. This singularity is inconsistent with $\mathbf{v} \in \mathcal{H}$ since standard elliptic theory implies that every component $v_n(x)$ should be in the Sobolev space H^2 and therefore continuous for $x \in \mathbb{R}^3$. Thus, no nonzero solution exists in the space $\mathcal{H}$ for the Floquet problem.

While the heuristic argument presented above is relatively simple, its mathematical justification is not. It requires very delicate analysis and is quite involved. Thus far, rigorous justification has been done[8] only for $M = 1$, which results in the following theorem.

Theorem 17 *For $V_1(t, x) = \Omega(r) \sin(\omega t - \theta)$, where $r = |x|$, with $\Omega(r) = 0$ for $r > 1$, $\Omega(r) > 0$ for $r \leq 1$ and $\Omega(r) \in C^\infty[0, 1]$, there is no nonzero solution in $\mathcal{H}$ and therefore ionization always occurs. Furthermore, if ψ_0 is spatially compactly supported and has finitely many spherical harmonic components, then $\|\psi(x, t)\|_{L^2(B_R)}^2 = O(t^{-5/3})$ for large t,*

Remark 18 The condition $\Omega(1^-) \neq 0$ simplifies the arguments but these could accommodate an algebraically vanishing Ω.

Remark 19 The analysis likely extends to systems with H_C replaced by

$$H_W = -\Delta - b/r + W(r)$$

where b may be zero and $W(r) = O(r^{-1-\epsilon})$ for large r and is in $L^\infty(\mathbb{R}^3)$.

VI. ACKNOWLEDGMENTS

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