

Shaping Long-lived Electron Wavepackets to Create Customizable Optical Spectra

Rumen Dangovski¹, Nicholas Rivera¹, Marin Soljačić¹, Ido Kaminer²

¹Department of Physics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge 02139, Massachusetts, USA

²Department of Electrical Engineering, Technion – Israel Institute of Technology, Haifa 32000, Israel
rumenrd@mit.edu

Abstract: We introduce new shape-invariant electron wavepackets constructed via superpositions of states in the ionization continuum, enabling customizable optical emission spectra in the eV-keV range. Their shape-invariance is prolonged indefinitely in exchange for larger spatial spreads.

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The rich physics of bound electrons in atoms and molecules is typically limited by the discrete nature of the energy spectrum and by the ionization energy threshold. Going beyond the ionization energy usually involves electron states that are not bound in space—extended states—which have positive energy and are therefore not considered relevant for atomic physics phenomena. An approach to create states of positive energy that behave as bound states is by shaping the electron wavepacket as a superposition of extended states, thus creating a localized state. For example, in 1979 Berry and Balázs proposed a solution of the free-space Schrödinger equation that appears to be localized in space and whose shape remains time-invariant as with bound electron states [1]. In recent years, this idea has been extensively explored in the optics community, with paraxial and non-paraxial optical beams [2-4]. More generally, these kinds of optical beams, such as Bessel [5] and Airy [3] beams, are propagation-invariant wave functions that also accelerate in the absence of external force, and have the intriguing property of self-healing—restoring their original shape after encountering an obstacle [6]. Importantly, due to the mathematical analogies between optical wavepackets and electron wavepackets, similar concepts are applicable to electrons [7-8]. However, the well-known problem with all of the above time-invariant or propagation-invariant wavepackets is that their probability density is not square-integrable (hence without a physical interpretation as a probability density). To remedy this, wavepacket shaping in space and time offers localized and long-lived electron wavepackets in the continuum of energy levels, created from superpositions of extended states.

Here, we propose and study the shaping of shape-invariant wavepackets that simultaneously suppress their own diffraction, while enabling access to a customizable spectrum of transitions, ranging from the visible to the hard-X-ray, via radiative decay to bound states. We develop the analytic tools to maintain the shape-invariance of shaped electron states and the analytic tools to calculate the radiative transitions of such states into bound states. Our methods can be extended to a variety of potentials, including the transitions of free electrons illuminated by general time-dependent fields. Specifically, for attractive static potential, we derive the non-diffracting electron wavepackets and study their dynamics. For example, we find that a non-diffracting shape of the wavepacket also affects the behavior of the electron in an attractive potential. We show that the presence of a Coulomb potential changes the physics of the system drastically by allowing propagating wavepackets to decay to bound states through radiative capture. We monitor the “competition” between spontaneous emission and diffraction by developing a Fermi Golden Rule (FGR) formalism which quantifies the rate of decay by the excited wavepacket. We find that in all these cases, the electron state lifetime is limited by the diffraction dynamics of its wavepacket. We concisely refer to the constructed states, which are almost propagation-invariant and time-invariant, as being *quasi-shape-invariant*.

To illustrate quasi-shape-invariance we begin from the textbook example of the hydrogen atom, consisting of the Schrödinger equation with the Coulomb potential $V(r) = -e^2/4\pi\epsilon_0 r$. Fig. 1 presents the shaping of the electron wavepacket created from superpositions of eigenstates at positive energies, which are called the Whittaker functions $w_\kappa(x, t)$, whose particular form is described in [9], at dimensionless momentum

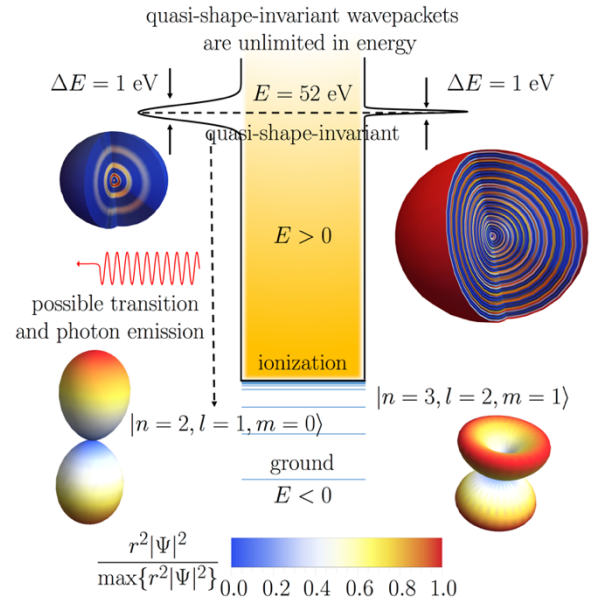


Figure 1: Shaping of electron states in the continuum of energy levels of the hydrogen atom creates localized and quasi-shape-invariant high-energy wavepackets. As opposed to the bound states, the electron states in the continuum of the energy spectrum are described by Whittaker functions, thus we call our wavepackets “Whittaker wavepackets.” The possibility of decay to bound states enables a photon emission with energy higher than the ionization threshold. We present truncated temperature maps of the typical shapes of the probability densities for both Whittaker wavepackets and bound states. As we decrease the energy spread ΔE , the wavepacket spreads out in space, hence the electron is more likely to be found farther from the origin.

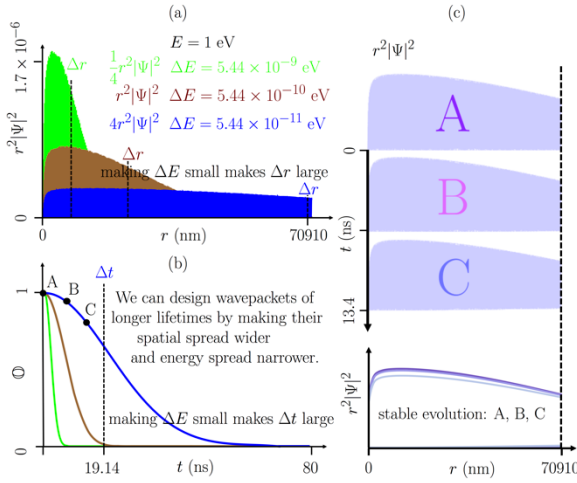


Figure 2: By shaping the (Whittaker) electron wavepacket we can prolong the lifetime to a desired duration in exchange for increasing the spatial spread. (a) As we decrease the energy spread ΔE , the probability density spreads out farther in space. Thus there is less probability to find the electron near the origin. (b) The benefit of making ΔE small, however, is that the diffraction lifetime also increases; $\Delta t = 19.14$ ns. (c) The envelope of the wavepacket at three points in time A, B, C (also marked in (b)), showing shape-invariant dynamics and slow diffraction over tens of ns.

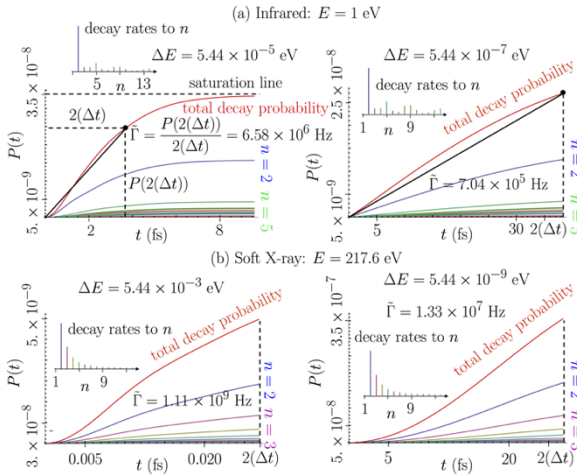


Figure 3: The radiative decay of the Whittaker wavepackets to bound states. (a) The decay probability reaches a saturation line. As time evolves, the electron is farther away from the origin, thus the effect of the Coulomb potential is reduced, and the instantaneous rate of decay converges to zero. The profile of decay rates to individual bound states follows an oscillatory pattern, with the highest decay rate to $n = 2$. (b) The profiles of the decay rates are monotonously decreasing in comparison to the profiles in (a). The dynamics and lifetime of the electron wavepacket are dominated by the diffraction and not by the radiative decay, since the diffraction lifetime is generally shorter than the radiative (FGR) lifetime.

$\kappa = ka_0$, distance $x = (2/a_0)r$ and time t for the Bohr radius a_0 . For simplicity, we focus on a spherically symmetric wavepacket with a Gaussian weighting over momentum space $\Psi_{E,\Delta E}(r, t)$, given as $\Psi_{E,\Delta E}(r, t) = \mathcal{N} \int_{-\infty}^{\infty} e^{-(\kappa-\mu)^2/2\sigma^2} w_{\kappa}(x, t) d\kappa$, where \mathcal{N} is a normalization constant, μ and σ are the mean and spread (standard deviation) of momentum. In SI units the energy E is parameterized as $E(\kappa) = \kappa^2(2e^2/4\pi\epsilon_0 a_0)$. We denote $E = E(\mu)$ and ΔE for the spread (standard deviation). We call the resulting wavepackets, which are superpositions of the eigenstates, “Whittaker wavepackets”

We now characterize their spatial and temporal dynamics. We can define a spatial spread Δr given by a standard deviation $\Delta r = \sqrt{\text{var}(\text{envelope}(\Psi(r, 0)))}$, and a diffraction lifetime Δt , also defined as a standard deviation $\Delta t = \sqrt{\text{var}(\mathbb{O}(t))}$ of an overlap function $\mathbb{O}(t) = |\int_0^{\infty} \Psi^*(r, 0)\Psi(r, t)r^2 dr|^2$. As we decrease the energy spread ΔE , then Δr increases (fig. 2 (a)) and Δt can become very large if ΔE is as small as possible (fig. 2(b)). Naturally, as this limit is taken, Δt increases without bound (fig. 2(c)).

Importantly, the physics of (hydrogenic) Whittaker wavepackets is different from that of free electrons, because of the spontaneous emission to bound states. To our knowledge, the entire concept of studying optical transitions of customizable electron states that are not just the conventional bound states has never been explored. In fig. 3 we show both qualitatively and quantitatively the properties of the total probability of decay, which is the sum of all probabilities of decay to single bound states. To calculate the radiative decay, we develop a FGR formula based on a QED formalism and the S -matrix approach [10], for which the infinitesimal probability of transition from the initial state $|i\rangle$ to the final state $|f\rangle$ through the emission of a photon with momentum \mathbf{k} and polarization λ , is given by $dP_{fi}(\mathbf{k}, \lambda) = (Vd^3\mathbf{k})/(2\pi)^3 |S_{fi}(\mathbf{k}, \lambda)|^2$; here $S_{fi} = \langle f | \exp(-i/\hbar \int_0^t \hat{H}_{\text{int}} dt') | i \rangle$ is the matrix element of the time-ordered unitary evolution operator of the EM interaction Hamiltonian. Up to first order we compute the universal expression $S_{fi}(\mathbf{k}, \lambda) = -\frac{e}{m_e} \sqrt{\frac{\hbar}{2\epsilon_0 \omega_k V}} \int_0^t dt' e^{i\omega_k t'} \hat{\epsilon}_{\mathbf{k}\lambda} \cdot \int d^3\mathbf{x} \langle f | \mathbf{x} \rangle e^{-i\mathbf{k}\cdot\mathbf{x}} \langle i | \mathbf{x} \rangle (t')$ for the photon’s frequency ω_k and its polarized direction $\hat{\epsilon}_{\mathbf{k}\lambda}$.

Since the radiative lifetime is much longer than the diffraction lifetime Δt , the customizable parameters Δt and Δr determine the stability of the quasi-shape-invariant Whittaker wavepackets. For example, a Whittaker wavepacket with a fixed diffraction lifetime $\Delta t = 53$ as (compared to the shortest X-ray pulse duration measured in [11]) allows for the following customizable optical spectra: 1. A soft X-ray ($E = 200$ eV) wavepacket with energy spread $\Delta E = 0.033$ eV and spatial spread $\Delta r = 0.72$ nm; 2. A hard X-ray ($E = 10$ keV) wavepacket with energy spread $\Delta E = 66$ μ eV and spatial spread $\Delta r = 5.1$ nm. We also studied Whittaker wavepackets with lifetimes on the order of nanoseconds, which require spread Δr on the order of microns.

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