Zeno-clocking the Auger decay

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A tenet of time-resolved spectroscopy is faster laser pulses for shorter timescales. Here we suggest turning this paradigm around, and slow down the system dynamics via repeated measurements, to do spectroscopy on longer timescales. This is the principle of the quantum Zeno effect. We exemplify our approach with the Auger process, and find that repeated measurements increase the core-hole lifetime, redistribute the kinetic energy of Auger electrons, and alter entanglement formation. We further provide an explicit experimental protocol for atomic Li, to make our proposal concrete.

Time and motion are essential entities to man’s awareness of Nature’s changes. As such, they have been continually scrutinised by scholars, not seldom to undermine or negate their meaningfulness. A notable example was that of Zeno of Elea [1], who believed in the deceit of the ordinary perception of change and movement. In a famous conceptual paradox, he argued that an arrow should not move, since at any instant it is observed, it is at rest.

In the Copenhagen interpretation of quantum mechanics, when a system is subject to measurement, its state is reduced. This leads to a quantum version of Zeno’s arrow paradox [2]: If an unstable system is measured upon frequently enough, it will not be able to decay. The quantum Zeno effect (QZE) has since been realized in the laser-induced dynamics of two-level ions [3, 4], and in the decay of ultracold atomic gases [5, 6]. However, it has not been yet directly observed in natural decay processes.

Here we take a step in bridging this gap, by proposing a protocol to measure the QZE in Auger decaying atoms (see Fig. 1). The Auger decay is a fundamental atomic process [7–9] by which an inner shell vacancy (a core hole) relaxes by emitting a secondary electron. The energy of the emitted electron is characteristic of each atomic species. Due to the short lifetime of the core hole and the non-local nature of the interactions involved, theoretical modelling of the Auger process is challenging and, until recently, mostly performed in the energy domain [10, 12–17] (see however [14, 18]). Yet, due to progress in ultrafast spectroscopy, real-time studies of the Auger decay are coming of age [19–22], to e.g. probe electron correlations at the few fs timescale or to use core-hole lifetimes as a clock for timing atomic processes [23].

In this work, we i) demonstrate the QZE by real-time simulations of Auger decay in a model atomic system. The QZE is induced after initial core-ionization using a train of \( \pi \)-pulses to drive the transition from the Auger decaying state to a higher quasi-stable level (see Fig. 1). We show how the field acts as a local projective measurement of the valence electron density, and that by increasing the pulses intensity and decreasing the delay between them, the measurement frequency is increased and the QZE enhanced. We ii) explicitly consider the Li atom and (briefly) the hollow Li\(^{+} \) ion, and find an increased lifetime due to QZE that should be clearly experimentally detectable. We use our results to gauge the range of experimentally controllable parameters where the QZE should be observable in atomic Auger decay. We also iii) show how the QZE influences the Auger lineshape and the formation of entangled continuum states, and address the effects of treating the laser field with and without the rotating wave approximation (RWA) [24]. Finally, the scope of spectroscopy in systems with dynamics slowed down via the QZE is addressed in the end.

**Model atomic system and external fields.** We consider a model atomic system, where a core electron has already been ejected and does not interact with the system left behind (sudden ejection limit [11]). The atom...
has two spinless electrons, which enter the Auger decay, and is exposed to a classical time-dependent light-field $E(t)$ treated in the dipole approximation ($t$ labels time). Our choice of a spinless model is computationally convenient, while fully retaining the qualitative aspects of Auger physics compared to the spinful case [15]. In an atomic-orbital picture, the Hamiltonian characterising the QZE in the Auger decay takes the form

$$ H(t) = H_a + H_c + H_{int} + H_p(t). $$

In Eq. 1, the atomic Hamiltonian $H_a$ is modeled in terms of the four relevant atomic states: $|c\rangle$, $|1\rangle$, $|2\rangle$ and $|3\rangle$. Here $|c\rangle$ is the core state (initially already empty) and $|1\rangle$ and $|2\rangle$ are the levels entering the Auger decay without QZE. These three states are taken with orbital-s symmetry. State $|3\rangle$, a valence state of higher energy, is of p-symmetry and initially empty; however, if populated (via e.g. optical absorption), it also undergoes an Auger decay. Thus, the atomic Hamiltonian is

$$ H_a = \sum_i \epsilon_i \hat{n}_i + \sum_{i<j} U_{ij} \hat{n}_i \hat{n}_j, $$

where $i, j \in \{c, 1, 2, 3\}$, $\hat{n}_i = c_i^\dagger c_i$ creates a spinless electron in level $|i\rangle$ with orbital energy $\epsilon_i$, and $U_{ij}$ is the strength of the intratomic Coulomb interactions.

The term $H_c$ of Eq. 1 describes the Auger continuum states, grouped in two regions $S$ and $P$, respectively corresponding to states with s- and p-symmetry. According to the Auger selection rules, $S (P)$ provides the emission channel for the decay from level $|2\rangle$ (level $|3\rangle$). Thus, $H_c = \sum_{\ell \in \mathcal{L}} H_{c_{\ell}}$, where we have defined $\mathcal{L} = S \cup P$. A constant density of states is assumed for each of the two continuum regions (we verified that our results are not sensitive to the actual energy dependence of the density of states), spanning a finite energy interval centered at the Auger energy $\epsilon_{2(3)}^A = \epsilon_1 + \epsilon_{2(3)} - \epsilon_c$ for region $S (P)$. Finally, bound and continuum states interact via $H_{int} = \sum_{\ell \in \mathcal{L}} U_{c_{\ell}c_{\ell}} c_{\ell}^\dagger c_{\ell} + h.c.$, where $\nu_\ell = 2 \ (3)$ when $\ell \in S (P)$. Here, for simplicity, all interactions with Coulomb integrals with two or more indexes in the continuum are neglected.

We choose the atom-light interaction term $H_p(t)$ according to the following physical scenario: levels 1 and 2 are strongly bound, and the laser frequency $\omega$ is in resonance with the transition $|2\rangle \leftrightarrow |3\rangle$. Thus, $H_p(t) = \Omega f(t) \sin(\omega t) \left(c_1^\dagger c_2 + c_3^\dagger c_1\right)$, where $f(t)$ is an envelope function (due to which the field starts to be applied only at time $t > 0$), $\Omega = \mathcal{E} \Delta$ is the Rabi frequency of the transition, $\Delta = (2|x|3)$ the dipole transition element, and $\mathcal{E}$ the field strength of the laser. We further assume that $\omega$ is smaller than the system’s ionization potential [25].

At initial time $t = 0$, the electrons are in levels $|1\rangle$ and $|2\rangle$, there is no electron in $|c\rangle$ and no light field is applied. We solved exactly the Schrödinger equation $i\partial_t |\psi(t)\rangle = H(t)|\psi(t)\rangle$ with the Lanczos algorithm [29] to obtain the local electron densities $n_1(t)$, $n_2(t)$, $n_3(t)$ (where $n_i = \langle \psi(t) | \hat{n}_i | \psi(t) \rangle$) to monitor the Auger decay. The Auger lineshape was calculated via the time-dependent orbital occupation $A(\epsilon_f, t) = \langle \psi(t) | \hat{n}_i | \psi(t) \rangle$ of the continuum states.

Quantum Zeno protocol for Auger decay. To hinder the Auger decay in time via the QZE, we need to periodically bring the system back to its initial state. To “freeze” the decay, the time $\Delta t$ between return events (measurements) has to be small (ideally zero) compared to the lifetime $\tau$ of the process [27].

We start the discussion with a reduced version of the system of Eq. 1, where only state $|12\rangle$ is subject to Auger decay (i.e $U_{c_{13}} = 0$), but the field still drives the transition $|12\rangle \leftrightarrow |13\rangle$. The suggested QZE protocol is as follows [30][31]: At time $t = 0$ we send in a square pulse of duration $t_\pi = \pi/\Omega$ (a so-called $\pi$-pulse), after which the probability $P_{13}$ of finding the system in state $|13\rangle$ is given by $P_{13}(t_{\pi}) = P_{12}(0)(\Omega^2/((\Omega^2 + \delta^2)^2)$. Here $\delta = \omega - \Delta$ is the detuning from resonance, $\Delta = E_{13} - E_{12}$ the energy separation of the states, and $P_{12}(0)$ the probability that the atom is initially in state $|12\rangle$. At this point, a local projective measurement of the form $M = \gamma |13\rangle\langle 13|$ can in principle be performed in a time $t_m$. After the measurement, another $\pi$-pulse transfers the system back with probability $P_{12}(t_{\pi} + t_{m} + t_{\pi}) = P_{12}(0)(\Omega^2/((\Omega^2 + \delta^2)^2)$. In zero field $P_{12}(2t_{\pi} + t_{m}) = P_{12}(0)$, and the system would return to its original state, where the Auger decay can take place. In this sense, the scheme acts as a projective measurement: the wave function is collapsed onto the subspace where there is one electron in the level $|2\rangle$, but only when the Auger decay has not occurred yet. However, this is not really needed, since one can measure the dipole radiation induced by the oscillations $2 \leftrightarrow 3$.

To prevent Auger decay, we should consider $N$ measurements in $[0, T]$, with $T = N\delta t$ and $\delta t$ the time between measurements. For our protocol, $\delta t$ is bounded by the single measurement time: $\delta t > 2t_{\pi} + t_{m}$. At small enough times, the probability for the system to be in its initial state at the beginning of the first measurement is $P_{12}(\delta t) \approx 1 - (\delta t/\tau_{\Delta}^2)$, where $1/\tau_{\Delta}^2 = \langle H^2 \rangle - \langle H \rangle^2$ defines the Zeno time $\tau_{\Delta}$ [27]. Since the measurement preserves $P_{12}$, the chance of remaining in $|12\rangle$ at time $T$ is $P_{12}(T) = P_{12}(\delta t)^N \approx [1 - (T/N\tau_{\Delta}^2)^2]^N$. For “infinitely” many measurements, $\lim_{N \to \infty} P_{12}(T) = e^{-T^2/N\tau_{\Delta}^2} \to 1$.

The physical parameters suitable for the QZE protocol in the case of the Auger decay are constrained by the following observations: i) The measurement time must be significantly (in the limiting case, “infinitely”) shorter than the Auger lifetime $\tau$; since $t_{\pi} = \pi/\Omega$, this corresponds to having $\Omega^{-1} \ll \tau$. In principle, this is accomplished by increasing the field strength $\mathcal{E}$, because $\Omega = \mathcal{E} \Delta$. ii) However, the frequency to be used cannot be larger than the ionization potential; even so, for high intensities the multiphoton ionization (MPI) rate also becomes of importance. iii) At the same time, to have an
efficient population transfer between states $|12\rangle$ and $|13\rangle$, ideally we need to be in the weak coupling regime $\Omega \ll \omega$. iv) Often both states connected by the measuring field undergo (Auger) decay with comparable lifetimes, thus introducing an element of variance from the usual QZE dynamics as just described.

Can the conditions above be met during the Auger decay of a realistic atom? This is indeed the case: as shown next, the QZE turns out to be clearly measurable, either in the time-domain [19] or as a narrowing of the Auger spectral linewidth.

**Auger decay in Lithium, with and without QZE.** We considered two model systems, Li and Li$^+$, with the respective benefits of relatively long lifetimes and large transition energy. For the Li atom, we associate the atomic configurations $1s(2s^2S)^2S^o$ and $1s(2s2p^3P)^2P^o$ with the states $|12\rangle$ and $|13\rangle$ in our model. Since the 1s electron is frozen during the decay, the problem corresponds to an effective two-particle system that can be modelled via Eq. (1). The configurations $1s2s^2$ and $1s2s2p$ have respective lifetimes of 17.6 fs and 174 fs dominated by Auger decay [28], and the transition between the states is optically accessible by resonant driving with a field of frequency $\omega = 2.5$ eV [28]. This process is associated with the transition $|12\rangle \leftrightarrow |13\rangle$ in the model system.

The decay dynamics of Li is shown in the top panel of Fig. 2. With no external field the Auger transition from the state $1s2s^2$ (associated in our model with the states $|c\rangle$) happens with a lifetime $\tau = 17.0$ fs. Driving the transition $1s2s^2 \leftrightarrow 1s2s2p$ with a field of intensity $I = 5.1$ TW/cm$^2$ and a measurement time $t_m = 0.32$ fs, the lifetime of the state $1s2s^2$ is extended to 32.7 fs, and further to 35.3 fs by increasing $I$ to 20.4 TW/cm$^2$.

We can also analyze the decay via the occupation $A(\epsilon, t)$ of the electrons emitted into the continuum, and detect how the Auger spectral peaks arise in time (Fig. 2, bottom panel). Without external field, $A$ has a single peak in the long-time limit. Conversely, when measurements are performed (nonzero field), there are two peaks, resulting from the decay of the $1s2s^2$ and $1s2s2p$ levels. Each peak is split by the dynamical Stark effect into two subpeaks separated by $\Delta \epsilon = \Omega$, for a total of four peaks.

As further evidence we briefly consider hollow Li$^+$. For the configurations $2s^21S_0$ and $2s2p^1P_1$, which differ in energy by $\omega = 4.1$ eV [22], a field of intensity $I = 210$ TW/cm$^2$ increases the lifetime of state $2s^2$ from 3.3 to 4.7 fs (see Supplemental Material, SM, for further detail). Overall, the Li and Li$^+$ results are a clear proof of concept that it is possible to clock (and slow down) the Auger decay via QZE. Although we found only a slowing down of the Auger transition, the effect is large enough to be clearly measurable either in the time-domain [19] or as a narrowing of the spectral linewidth.

**QZE-vs-Auger trends.** We now assess the role of the lifetimes $\tau_1$ and $\tau_2$, their level spacing $\omega = E_2 - E_1$, and...
the Rabi frequency $\Omega$ ($\Omega^2$ is proportional to the field intensity $I$) in QZE, and start by considering $\tau_2 = \infty$. If $\omega$ is large enough that the RWA can be invoked, our protocol permits to extend $\tau_1$ to many times its unperturbed value. In the regime $\Omega/\omega \ll 1$ the RWA and the full field give the same dynamics. However, when $\Omega/\omega \approx 1$ the two approaches differ significantly (see Fig. 3), and the RWA overestimates the increase of the lifetime. However, even for these parameters the lifetime can be extended enough for the effect to be clearly measurable. For $\tau_2$ finite but larger than $\tau_1$ similar results are observed. In the RWA it is possible to significantly extend $\tau_1$, but now with an expected upper bound $\tau_2$. For strong fields compared to $\omega$ we again find differing results between RWA and full field dynamics, with the RWA overestimating the effective lifetime: The RWA by construction completely misses the protocol dependence on $\omega$, so only the ratios $\tau_1/\Omega$ and $\tau_2/\Omega$ play a role.

As shown in Fig. 3 our protocol performs best for weak coupling $\Omega \ll \omega$, where the effective lifetime can be significantly enhanced. This also clarifies why we don’t observe a full halt of the decay in Li and Li$^+$: the reason is a combination of the fast decay times ($\tau_{12} \approx 17.3$ fs in Li and $\tau_{12} \approx 3.4$ fs in Li$^+$) and the small transition energies ($\omega = 2.5$ eV in Li and $\omega = 4.1$ eV in Li$^+$). The short lifetimes require a high intensity for $\Omega^{-1}$ to be comparable with $\tau$, but the high intensities make $\Omega$ comparable $\omega$. This prevents $\tau_{12}$ to be extended beyond its value for $\Omega \approx \omega$. Although measurable already for Li and Li$^+$, the QZE should be more pronounced in systems with longer lifetimes and greater transition energies.

In summary, the transition energy (related to the applicability of RWA) and the lifetimes of the primary and measurement levels, all have a great influence on the occurrence of the QZE in the Auger decay. At least in the weak intensity limit, to maintain the applicability of RWA, one could use, instead of square pulses, pulses where the intensity and frequency can be changed as a function of time that are known to improve population transfer in e.g. NMR spectroscopy and quantum information.

Auger decay, QZE, and Entanglement. Our treatment does not keep track of the primary (core) photo-electron, and thus misses a description of the entanglement between photo- and Auger electrons, as possibly measurable by e.g. coincidence experiments. However, we can still explore how/if the QZE affects entanglement formation in the continuum during the Auger decay. We used mode concurrence as a convenient entanglement measure. In Fig. 4 we show the concurrence matrix $C_{\ell\ell'}$ without and with external fields. With no field there is a single Auger peak at $\epsilon_a \approx 35$, and there is concurrence between this state and all other $k$-points. With the field there are two peaks at $\epsilon_a \approx 35$ and 45 that are split due to the Stark effect. In this case there is concurrence within each continuum, but also between the different continua, suggesting that in general there is an interesting interplay between QZE, Auger decay, and entanglement formation in the continuum.

Conclusions and outlook. We showed that the Auger lifetime of an atom can be increased due to the quantum Zeno effect, when a measurement based on bound-bound transitions is repeatedly performed during the Auger decay (for bound-continuum transitions see SM). As concrete example we considered the Li atom, showing that the physical parameter values to be used are within experimental reach.

More in general, there is nothing especially unique about the QZE and Auger process, and it should be possible to perform the same procedure with other natural (or not) decay phenomena, be it radiative or not. On speculative grounds, one can envisage types of pump-probe experiments where, after an initial Hamiltonian quench, the ensuing relaxation dynamics can be studied on artificially longer timescales, thanks to repeated measurements which induce the quantum Zeno effect, and thus slow down the system’s time evolution.

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