High harmonic spectroscopy of disorder-induced Anderson localization

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Exponential localization of wavefunctions in lattices, whether in real or synthetic dimensions, is a
fundamental wave interference phenomenon. Localization of Bloch-type functions in space-periodic
lattice, triggered by spatial disorder, is known as Anderson localization and arrests diffusion of clas-
sical particles in disordered potentials. In time-periodic Floquet lattices, exponential localization in
a periodically driven quantum system similarly arrests diffusion of its classically chaotic counterpart
in the action-angle space. Here we demonstrate that nonlinear optical response allows for clear
detection of the disorder-induced phase transition between delocalized and localized states. The
optical signature of the transition is the emergence of symmetry-forbidden even-order harmonics:
these harmonics are enabled by Anderson-type localization and arise for sufficiently strong disorder
even when the overall charge distribution in the field-free system spatially symmetric. The ratio of
even to odd harmonic intensities as a function of disorder maps out the phase transition even when
the associated changes in the band structure are negligibly small.

Disorder is an ubiquitous effect in crystals 11. The seminal work by Anderson 2 predicted that above a
critical disorder value, the electronic wavefunction will change from being delocalized across the lattice to
exponentially localized (insulating state) due to the inter-
ference of multiple quantum paths originating from the scattering with random impurities and defects. Anderson
localization is a fundamental wave phenomenon and thus
permeates many branches of physics: it has been observed in matter waves 3, light waves 4, and microwaves 5.
Anderson localization also finds direct analogues in peri-
dodically driven systems, with time-periodic dynamics
taking the role of space-periodic structure. While peri-
dodically driven classical systems can develop chaotic be-
haviour for sufficiently strong driving fields, leading to
delocalization of the original ensemble across the whole
phase space, their quantum counterpart shows exponen-
tial localization of the light-dressed states 6.

Dramatic changes in a wavefunction during transi-
tion from a delocalized to a localized state may lead to
changes in the nonlinear optical response of the system.
In this context, symmetry–forbidden harmonics of the
driving field are an appealing bellwether candidate. In-
deed, while even-order harmonics are known to be for-
bidden in systems with inversion symmetry 7, they are
also known to arise in such systems if and when charges
localize 8,9. The required symmetry breaking can then
be triggered by even a small asymmetry in the oscillating
electric field of the driving laser pulse. Such asymmetry is
natural in short laser pulses and is controlled by the phase
of the electric field oscillations under the pulse envelope,
i.e., the carrier-envelope phase (CEP) 10. Exponen-
tially localized states in symmetric multiple well poten-
tials appear to be particularly sensitive to even small field
asymmetries, leading to even harmonics in the nonlinear
response even for pulses encompassing tens of cycles 8.

High harmonic generation (HHG) is a powerful tool
for ultrafast spectroscopy 11,12. Extremely large
cohert bandwidth of harmonic spectra enables sub-
femtosecond resolution. HHG is a sensitive probe of Cooper minima 13, Auger decay 14, attosecond
dynamics of optical tunnelling 15,16 and the dynamics
of electron exchange 17 in atoms, ultrafast hole dynam-
ics 11,18,19, nuclear motion 21,22 in small molecules, and
eantio-sensitive electronic response in more com-
plex chiral molecules 24,25. In solids, high harmonic
spectroscopy has allowed observation of dynamical Bloch
oscillations, band structure tomography 26, probing
of defects in solids 29,30, sub-fs monitoring of core
excitons 31, optical measurement of the valley pseu-
dospins 32,33, tracking of van Hove singularities 35,
picometer resolution of valence band electrons 36, imag-
ing internal structures of a unit cell 37, monitoring of
light-driven insulator-to-metal transitions 38, and prob-
ing of topological effects 39,40.

In this work, we employ HHG to track phase tran-
sition between delocalized and localized states in the
Aubry-André (AA) system 41 (similar to that pro-
posed in Ref. 42)), where localization occurs only above
a critical value of disorder, already in one dimension.
This model captures the metal-to-insulator transition,
is a workhorse to study non-trivial topology, and has
been realized in optical lattices and photonic quasi-
crystals 43,44.

The model system is described by the following tight-
briding Hamiltonian,

\[ \hat{H} = -t_0 \sum_{j=1}^{L-1} \left( c_j^\dagger c_{j+1} + \text{h.c.} \right) + V \sum_{j=1}^{L} \cos(2\pi \sigma j) c_j^\dagger c_j, \]

where \( t_0 \) is the nearest neighbour hopping term, \( V \) is the
strength of the potential, \( c_j^\dagger \) and \( c_j \) are, respectively, the
fermionic creation and annihilation operators at site \( j \),

\[ \sigma = \begin{cases} 1 & \text{if } j \text{ even,} \\ 0 & \text{if } j \text{ odd.} \end{cases} \]
h.c. stands for the hermitian conjugate, \( L \) is the total number of lattice sites and \( \sigma \) determines the periodicity of the potential. A rational value of \( \sigma \) corresponds to a periodic potential and consequently to delocalized electronic wavefunctions. If \( \sigma \) is irrational, the potential becomes quasi-periodically disordered (for finite systems, this may also happen for rational \( \sigma \)). For a disordered potential, the system undergoes the localization phase transition at \( V/t_0 = 2 \). For \( V/t_0 > 2 \) all states are exponentially localized on one site, while for \( V/t_0 < 2 \) the states are delocalized.

Figures 1(a) and (b) show the eigenspectrum of our system, for \( \sigma = \frac{\sqrt{5}+1}{2} \), with 100 lattice sites, in the delocalized \( (V/t_0 = 1.9) \) and localized \( (V/t_0 = 2.1) \) phases, respectively. The hopping term \( t_0 = 0.26 \) eV and the lattice constant \( a_0 = 7.56 \) atomic unit of length \((\sim 0.4 \) nm\) are used throughout. The Fermi energy is \( E_F = -0.2 \) eV, so that red-colored states correspond to fully occupied valence band states, while the blue-colored states are unoccupied. Differences in the eigenspectrum for both phases are indiscernible. In contrast, the individual eigenstates of the system present a clear localization phase transition: Figs. 1(c) and (d) show the occupation number of the eigenstate with index=10 (other eigenstates show similar behaviour). The eigenstate is delocalized for \( V/t_0 = 1.9 \) but fully localized for \( V/t_0 = 2.1 \). These differences between the charge distribution in individual eigenstates disappear completely when we consider the fully-filled valence band, see Figs. 1(e) and (f): the differences between both phases are barely visible.

The question is: will the non-linear optical response be sensitive to the phase transition? To address this question, we consider a non-resonant, low-frequency field polarized along the 1D chain,

\[
F(t) = F_0 f(t) \cos(\omega t + \phi),
\]

with \( \phi \) the carrier-envelope phase and \( f(t) \) the sine-squared envelope with a full duration of 10 optical cycles. We include the laser-matter interaction via the time-dependent Peierls phase: \( t_0 \to t_0 e^{i\phi(t)} \), where \( A(t) \) is the field vector potential, \( F(t) = -eA(t)/dt \), and \( e \) is the electron charge. The carrier \( \omega = 0.136 \) eV is set well below the bandgap \( \Delta \approx 0.4 \) eV.

Two characteristic regimes describe laser-induced electron dynamics in such low-frequency fields.

In the localized phase, efficient resonant tunnelling between localized states at different sites, including transitions from the valence to the conduction band, becomes possible when the peak voltage between the adjacent sites \( F_{0a}a_0 \) approaches and/or exceeds the characteristic energy gap \( \Delta \), \( F_{0a}a_0 \approx \Delta \). This regime enables rapid energy gain by the system within a few laser cycles, allowing it to climb to the top of the energy scale [45]. In our case, its signature would be the emission of harmonics up to the maximum transition frequency of the system (harmonic 10), for all fields enabling resonant tunnelling (RT). The onset of this regime corresponds to \( F_{0,\text{RT}} \sim \Delta/a_0 \approx 0.4 \) MV/cm in our system. In the localized phase, exponential sensitivity of resonant tunnelling to positions of individual states and to the field strength create ideal conditions for symmetry breaking, leading to generation of even harmonics; the direction in which the symmetry is broken is controlled by the pulse CEP.

In the delocalized case, the delocalized states adiabatically follow oscillations of the low-frequency driving field, preventing symmetry breaking. The latter requires light-induced electron localization, which occurs when \( F_{0a}a_0 \omega \geq \Delta^2/4 \), i.e., when \( F_0 \geq 1 \) MV/cm in our system. Even harmonic generation should therefore only emerge around \( F_0 \approx 1 \) MV/cm.

Our numerical simulations below fully confirm all of
these expectations. The time-dependent Schrödinger equation is solved independently for the $m$ normalized eigenstates $|\psi_m(t = t_i)\rangle$ of the field-free Hamiltonian Eq. (1) that lie below $E_F$, $|\psi_m(t)\rangle = e^{-i\int_{t_i}^{t} H(t')dt'}|\psi_m(t = t_i)\rangle$, where $\hat{H}(t)$ is the Hamiltonian in Eq. (1) with the time-dependent Peierls substitution. The current from a single eigenstate $m$ is calculated as

$$j_m(t) = \langle \psi_m(t)|\hat{J}(t)|\psi_m(t)\rangle$$  \hspace{1cm} (3)$$

where the current operator is defined as

$$\hat{J}(t) = -iea_0 \sum_{j=1}^{L} \left( e^{-ia_0 c A(t)} c^\dagger_j c_{j+1} - e^{ia_0 c A(t)} c^\dagger_{j+1} c_j \right).$$  \hspace{1cm} (4)$$

The total current from all the valence band eigenstates $m$, i.e., the fully-filled valence band, is,

$$j(t) = \sum_m j_m(t).$$  \hspace{1cm} (5)$$

The harmonic spectra are then calculated from the Fourier transform of the time derivative of the total current. Prior to the Fourier transform, we apply an envelope to the current that coincides with the laser pulse envelope \cite{17}, to filter out emission after the end of the laser pulse. We consider 100 lattice sites and 0.01 atomic unit of time-step (~0.25 as) to get the converged spectra.

Figure 2 shows the HHG spectrum for different initial states and field strengths in the localized and delocalized phases. First, in Figs. 2(a) and (b), we consider a single valence band eigenstate as our initial state ($m = 10$, shown in Fig. 1(c,d)). The charge distribution of the initial state is strongly asymmetric in both phases, which breaks the left-right symmetry of the chain in the HHG process and leads to the appearance of even harmonics, both in the delocalized [Fig. 2(a)] and localized [Fig. 2(b)] phases.

However, the equilibrium initial state corresponds to the fully-filled valence band, with the charge distributed relatively evenly between all sites; the distribution is virtually identical between the two phases [Figs. 1(e) and (f)]. With this initial condition, the HHG spectra of the two phases, shown in Figs. 2(c) and (d) for a field strength of $F_0 = 0.6$ MV/cm, are now strikingly different.

Destructive interference from different initial states completely suppresses even harmonics in the delocalized phase [Fig. 2(c)], but they remain prominent in the localized phase [Fig. 2(d)]. This result follows from our previous discussion. The field strength $F_0 = 0.6$ MV/cm is above the threshold for resonant tunneling between localized states in our system ($F_0, RT = 0.4$ MV/cm), where the instantaneous field brings the energy levels into resonance, generating coherence between all sites. Resonant tunneling between the sites depends sensitively on the instantaneous field strength, leading to CEP-dependent

FIG. 2. (a,b) High harmonic spectra for the system in the (a, c, e, g, i) delocalized and (b, d, f, h, j) localized phase: (a, b) HHG from the $10^{th}$ eigenstate only for a field strength $F_0 = 0.6$ MV/cm. HHG from the fully-filled valence band for a field strength (c, d) $F_0 = 0.6$ MV/cm, (e, f) $F_0 = 2.2$ MV/cm, (e, f) $F_0 = 0.4$ MV/cm, and (i, j) $F_0 = 1.0$ MV/cm.
FIG. 3. High harmonic spectra in the delocalized (top panels, red curve) and the localized (lower panels, green curve) phase calculated for different carrier-envelope phases (CEP) of the field: (a,d) CEP=+π/2, (b,e) CEP=−π/2 and (c,f) the coherent superposition of CEP=+π/2 and CEP=−π/2.

symmetry breaking and the appearance of even harmonics in the localized phase. We find that even harmonics emerge already at \( F_0 \approx 0.4 \text{ MV/cm} \). As resonant tunnelling induces coherences between the localized sites, it enables population of the lowest band and generation of higher harmonics in the localized phase than in the delocalized phase [Figs. 2(c) and (d)]. In the latter, the system follows adiabatically the field oscillations and transitions to the highest states are suppressed.

The field strength used in Figs. 2(c) and (d) is below the threshold field for laser-induced localization in the delocalized phase, which is \( F_0 \approx 1 \text{ MV/cm} \) for our system. Therefore, the harmonic spectrum in the delocalized regime shows no sign of even harmonics and a smaller cut-off energy [Fig. 2(c)]. Even harmonics emerge as soon as the field amplitude crosses this threshold. At \( F_0 = 2.2 \text{ MV/cm} \) the harmonic spectra in both phases become very similar [Figs. 2(c) and (f)]. For this field strength, the cut-off is the same in both phases, and corresponds to the (Stark-shifted) maximum transition frequency of the system.

To confirm the origin of symmetry breaking and even harmonics emission, Fig. 3 shows the harmonic spectra for two values of the pulse carrier-envelope phase (CEP) shifted by \( \pi \), and their coherent superposition in Figs. 3(c) and (f). In the delocalized case, even harmonics are absent regardless of the CEP [Figs. 3(a) and (b)]. In the localized case, they are identical in both cases, Figs. 3(d) and (e), but with opposite phase: upon coherent addition even harmonics are completely washed out [Fig. 3(f)]. The reason is that the laser-induced asymmetry in the electron charge distribution at CEP=+π/2 is exactly opposite to that at CEP=−π/2, as graphically illustrated by the cancellation of the emission upon interference.

Figure 4 shows that the relative intensity of even harmonics does indeed track the metal to insulator (delocalized – localized) phase transition in the system. The figure plots the ratio of even to odd harmonics for different values of \( V/t_0 \), at a field strength of \( F_0 = 0.6 \text{ MV/cm} \), i.e., that which generates resonant tunneling in the localized phase but not laser-induced localization in the delocalized phase. The even–odd ratio increases dramatically at the phase transition \( V/t_0 = 2 \), showing that HHG, driven by a phase-stable CEP pulse, is able to map disorder-induced electron localization in a solid.

In conclusion, we have used high harmonic spectroscopy to track delocalized to localized phase transition in the Aubry-André system. For aperiodic potentials \( (\sigma = \sqrt{5+1}/2) \), the localized and delocalized phases show almost identical eigenspectra and site occupation numbers for a state with fully-filled valence band. Yet, high harmonic spectra between the two phases show striking differences, especially in the appearance of forbidden (even) harmonics. This effect is a consequence of dynamical symmetry breaking induced by the field but enabled by initial electron localization, which promotes resonant tunneling and leads to CEP-dependent symmetry breaking even in the low-frequency regime. Our work shows that the localisation-delocalisation phase transition, which can be driven by a very small modification of the on-site energy, can be effectively traced by HHG spectroscopy.

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[45] M. Ivanov, T. Seideman, P. Corkum, F. Ilkov, and P. Dietrich, Phys. Rev. A 54, 1541 (1996).
[46] P. Dietrich, M. Y. Ivanov, F. A. Ilkov, and P. B. Corkum, Phys. Rev. Lett. 77, 4150 (1996).
[47] M. Wu, S. Ghimire, D. A. Reis, K. J. Schafer, and M. B. Gaarde, Phys. Rev. A 91, 043839 (2015).