Laser-induced quantum chaos in 1-D crystals

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Abstract

We study the electronic band structure for a model one-dimensional periodic potential in the presence of a spatially homogeneous laser field. The statistical properties of the energy bands depend on the coupling between the crystal and the laser field, going from Poisson to Wigner-Dyson (GOE) and back to Poisson as the coupling increases. We argue that the classical dynamics of this system resembles that of a periodically driven pendulum. We find that the chaotic regime is not restricted solely to high-lying bands and should thus be of easier access to optical experiments.
In the past decade, many model systems exhibiting what conventionally became known as quantum chaos have been discussed in the literature. Among those, the kicked rotator and the Hydrogen atom in the presence of a uniform magnetic field have been studied in great detail, both theoretically and experimentally. Formally, one speaks of quantum chaotic behavior when the system dynamics in the classical limit is very irregular or completely chaotic. For a quantum system whose classical dynamics is regular one usually finds that energy levels are uncorrelated and therefore obey a Poissonian statistics. On the other hand, if the system is fully chaotic in the classical limit, its energy levels tend to repel each other strongly. In this case the statistical properties of the spectrum coincide with those drawn from the Gaussian ensembles of random matrix theory (RMT).

Recently, Mucciolo and coworkers showed that the electronic spectra of crystalline solids (e.g., Si) also show the universal signatures of quantum chaotic behavior. Their analysis was restricted to high-lying bands, well above the Fermi energy. Since crystals are fixed by nature, no external parameter exists to modify the system and therefore try to enhance the irregularity in its classical dynamics. This is in contrast with the two previously mentioned systems, where chaos is controlled by an external parameter, as for example the strength of a time-periodic impulse or the intensity of a magnetic field.

In this work we present a new model exhibiting spectral signatures of chaos based on electrons in a periodic potential. In our case a monochromatic laser field is the main responsible for the irregular electron dynamics. The intensity of the laser field can be regarded as an external parameter changing the behavior of the system. We believe that this mechanism could increase the chances of experimentally observing chaos in crystals by bringing universal statistics to low-lying bands as well. By looking at the energy level statistics of a very simple 1-D periodic potential we were able to identify three regimes. These suggest that the system dynamics moves from regular to chaotic and then back to regular as the field intensity is increased. Our formalism is based on the dressed band approach which we briefly review below.

According to Bloch’s theorem, the wave function for an electron in a 1-D periodic potential can be labelled by a momentum \( k, -\pi/d \leq k < \pi/d \), where \( d \) is the lattice parameter. This allows the Hamiltonian to be decoupled into well-defined \( k \) (reduced) components. Inclusion of the interaction with a monochromatic laser field leads to the following reduced Hamiltonian:

\[
H = H_k + H_\gamma + H_{int},
\]

where, in atomic units (\( \hbar = e = m = 1 \)),

\[
H_k = \frac{p^2}{2} + V(z)
= -\frac{1}{2} \left( \frac{d}{dz} + ik \right)^2 + \sum_\ell V(G_\ell) \exp(i G_\ell z),
\]

\[
H_\gamma = \omega \ a^+ a,
\]

and
\[ H_{\text{int}} = \alpha A p + \frac{\alpha^2}{2} A^2 . \]  

In the above equations, \( V(z) \) denotes the lattice potential, \( a^+ (a) \) is the creation (annihilation) field operator, \( \alpha = 1/137 \) is the fine structure constant, and \( A = \frac{1}{\sqrt{2\pi/\omega\Omega}} (a + a^+) \), where \( \Omega \) is the quantization volume. The periodicity of the lattice potential allows it to be written in terms of a reciprocal lattice sum over \( G_\ell = 2\pi \ell / d \), as indicated in Eq. (2). Here the field polarization is taken along the \( z \)-direction.

By performing adequate unitary transformations in terms of a basis of eigenstates \( |G, n\rangle \) of the momentum \( p \) and the field Hamiltonian \( H_\gamma \), \( H \) can be transformed into a Floquet Hamiltonian \( H_F \). One can then show that the spectrum of \( H \) follows from the diagonalization of the Bloch-Floquet matrix

\[ \langle G', n'|H_F - N_0\omega|G, n\rangle = \left[ \nu \omega + \frac{(G + k)^2}{2} \right] \delta_{G,G'}\delta_{n,n'} + J_n - n \left( \frac{\alpha A_0}{\omega} (G - G') \right) V_{G-G'} , \]  

where \( A_0 = \frac{2}{\sqrt{2\pi N_0/\omega\Omega}} \) is the amplitude of the classical vector potential and \( J_n \) is the Bessel function of order \( n \). It should be noted that \( n \) in the state \( |G, n\rangle \) denotes an integer (positive or negative) that describes the deviation from the average number of photons \( N_0 \), which is supposed to be large. Note that the matrix defined by Eq. (5) is time-reversal symmetric.

We present results for a model potential where \( V(G_\ell) = -\sigma \) for \( -3 \leq \ell \leq 3 \) and zero for other \( \ell \) values. This is a truncated Kronig-Penney attractive model. We take the energy unit to be \( \epsilon = G_1^2/2 \) and the potential strength \( \sigma = 0.25\epsilon \). The laser intensity is characterized by the dimensionless parameter \( x = \alpha A_0 G_1/\omega \). Equation (5) allows for a simple interpretation of the spectrum in this problem: In the absence of the laser field, the electronic energy levels consist essentially of the free-electron parabola folded into the first Brillouin zone (BZ), plus gaps opening around the crossing points \( (k = 0 \text{ and } \pm G_1/2) \). The gap amplitude and the distortion of the bands with respect to the free-electron parabola depend on the potential strength \( \sigma \). Inclusion of the field produces a dressing effect in the bands, which may be described by replica-bands translated by an amount \( n\omega \). The interaction \( H_{\text{int}} \) causes an anti-crossing whenever two noninteracting replica-bands cross. Clearly, this occurs for all \( k \) in the BZ; for a fixed value of \( \omega \), the gaps widen as the intensity \( x \) increases.

In Fig. 1 we show the spectrum obtained for \( -10 \leq n \leq 10, \omega = 0.3\epsilon \), and field intensity \( x = 10 \). We avoid regions at the extremes of the energy spectrum, where only replicas of the lowest or higher bands are present (due to the truncation in \( n \) adopted in our numerical treatment) and limit our analysis to the energy region \( 1 < E/\epsilon < 5 \), where a large number of anti-crossings occur. For the parameters chosen here, this energy region is representative of the complete basis, \( -\infty < n < \infty \). The laser frequency \( \omega \) is chosen to maximize the mixing between the dressed bands. Note that the spectrum in Fig. 1 reveals an intricate level repulsion structure which resembles those characteristic of quantum chaotic regimes in other systems.

A quantitative identification of quantum chaos consists primarily in unfolding the energy spectrum (to attain a constant density of states) and then analyzing the short and long-range statistical correlations present. For spinless, time-reversal symmetric systems with a complex
dynamics, RMT predicts that the distribution of nearest-neighbor level spacings (NNS) should follow closely the Wigner surmise for the Gaussian orthogonal ensemble (GOE),

\[ P_{\text{GOE}}(s) = \frac{\pi}{2} s e^{-\pi s^2/4}, \]  

where the level spacing \( s \) is given in units of the mean level spacing. On the other hand, the energy levels of most integrable systems are essentially uncorrelated (one important exception is the harmonic oscillator), in which case the NNS distribution is a simple exponential, namely,

\[ P_{\text{Poisson}}(s) = e^{-s}. \]  

Long-range correlations can be diagnosed through the least square deviation \( \Delta_3(L) \) of the number of levels in a given energy interval of length \( L \) from a constant slope.\[ When \( L \) is given in units of the mean level spacing, \( \Delta_3(L) \) is expected to fall between two extreme limits: linear \((L/15)\) for uncorrelated levels and constant \((1/12)\) for fully rigid, equally spaced spectra. The GOE prediction is an intermediate function which, for large \( L \), behaves as \( \Delta_{3,\text{GOE}}(L) \approx (1/\pi^2) \ln(2\pi L) - 0.007. \]

In Fig. 2 and 3 we present our results for the NNS distributions and \( \Delta_3 \), respectively, obtained for increasing values of the field intensity parameter \( x \). The data correspond to a sampling over 100 equidistant \( k \) values along \( 0 \leq k/G_1 \leq 0.5 \). The level density remained essentially constant on the energy interval considered and no unfolding was carried out. The resulting statistics involved over 5,200 calculated levels for each \( x \). Let us concentrate initially on Fig. 2. For \( x = 0 \) the uncorrelated character of the energy levels and the integrability of the Hamiltonian in Eq. (1) is reflected in the agreement between the data and the Poisson law. As \( x \) increases, a weak level repulsion appears. Note that \( x = 1 \) corresponds to a crossover regime in which neither of the distributions in Eqs. (7) and (6) agrees with the data. However, for \( x = 10 \) the Wigner surmise provides a very accurate fit, suggesting a chaotic regime at this intensity. Further increase in \( x \) causes the data to change, deviating from the Wigner surmise. For \( x = 1000 \), the spectrum tends again to a Poisson statistics, which reflects the exact integrability in the limit of infinite field intensity as well. It is important to note that the NNS is not a delta-function centered at \( \omega \) when \( x \to \infty \): The periodic potential, although relatively weak, folds the sequence of equidistant parabolas back onto the first BZ, causing the NNS to be Poissonian.

A similar interpretation goes for Fig. 3. The maximal spectral rigidity occurs around \( x = 10 \). In this figure, however, we see a marked deviation from either the Poisson \((x = 0\) and 1000\)) or the GOE \((x = 10)\) predictions beyond intervals of length \( L_{\text{max}} \approx 4 \). From Fig 4 we may notice a tendency of levels to form clusters of four. This effect is well known in the literature. In a semiclassical language, it indicates the breakdown of the universal regime and the appearance of system-dependent contributions, which are related to very short periodic orbits. We have checked that \( L_{\text{max}} \) is always approximately equal to the laser frequency \( \omega \) (in units of the mean level spacing). The periodic orbit associated to this frequency is the oscillation of the laser field in its phase space. The reason why this orbit is always the shortest possible one and its signature survives different values of \( x \) is related to the nature of our model. For the Kronig-Penney model there is only one bound state \((E < 0)\)
in the spectrum. As a result, in the energy range where the $\Delta_3$ was obtained, the electron classical motion can only be bounded (and therefore periodic) through the interaction with the laser field. This always leads to a periodicity $T > 2\pi/\omega$ and the intrinsic period of oscillation of the laser field remains the shortest one.

As noted before, for a 3-D periodic potential the electronic spectrum at low-symmetry regions of the BZ manifests quantum chaotic behavior. In the equivalent 1-D problem, no chaotic behavior is expected, since the electrons have only one degree of freedom ($z$) and the problem is classically integrable. The introduction of a laser field adds another degree of freedom to the system and makes the problem nonintegrable. As indicated by our numerical results, for high enough intensities of the laser field, it is plausible that this perturbation causes the electron motion to be strongly irregular, leading to chaos.

Although we treat the laser field in a second quantized form, our results are equivalent to those obtained from a semiclassical, time-dependent description where the field is fixed and can be written in the form $A_0 \cos(\omega t)$. Both descriptions yield the same spectrum (band structure) of energies and quasi-energies in the quantized and semi-classical descriptions, respectively. The only difference is that, in the semiclassical case, one formally incorporates the time-periodic vector potential to the crystal potential, resulting in a space-time (two-dimensional) BZ, but with matrix elements for the effective Hamiltonian similar to Eq. (5).

It is interesting to compare the problem of a laser-illuminated 1-D crystal to the kicked rotator. The dynamics in the latter is generated by a Hamiltonian in which the free rotator is subject to a time-periodic force whose amplitude is a function of the rotation angle ($2\pi$-periodic in $\theta$). In the classical counterpart of our model (and in the kicked rotator), an external field, space and time periodic, acts on a particle moving in one dimension. The difference between these two systems is that, in our case, space and time components are decoupled, allowing one of the potentials to be made negligible with respect to the other by varying the parameter $x$. Thus, the $x = 0$ limit corresponds to an electron in a 1-D crystal problem, and $x \to \infty$ corresponds to a free electron in the presence of the laser field. In both limits the problem is exactly integrable.

No such decoupling is possible in the kicked rotator. Our problem would be more closely related to a periodically kicked pendulum, in which case the kick amplitude $\lambda$ would be a control parameter similar to the laser intensity, and this system should present quantum chaos for intermediate values of $\lambda$. For $\lambda = 0$ and for $\lambda \to \infty$ the problem would be dominated by the gravitational field or by the kick force respectively, becoming integrable and therefore non-chaotic.

To our knowledge, no other system discussed in the literature has been led from regularity into chaos and than back to regularity by increasing a single control parameter. One might think that a similar situation would occur for the H atom in a magnetic field since the problem is exactly integrable in the limits of zero magnetic field and zero Coulomb potential. However, even in the limit of extremely strong magnetic field, the Coulomb interaction can not be neglected. In this limit the classical motion of the electron is strongly constrained in the field direction and the electronic distribution is no longer spherically symmetric. The electronic motion may be approximated by a 1-D truncated Coulomb interaction with the nucleus, which has a completely different spectrum from the Landau problem. Other simple model-Hamiltonians, such as the the spin-boson model, may display a variety of behaviors, but they require variations of several control parameters.
Finally, we discuss the experimental implications of our model. Contrary to the 3-D band structure problem, the chaotic behavior presented here is not limited to highly excited bands. Any region of the spectrum of the 1-D system displays the universal features under a laser field of adequate intensity. The same effect should be present in 3-D systems, even for bands close to the Fermi energy. The limitation on the long-range universal correlations imposed by short periodic orbits should become less stringent in higher dimensions due to the increase in the density of states. We believe that optical pump-probe techniques should be adequate to investigate the dressing effects presented here. They should reveal universal signatures in the frequency and momentum dependence of the dielectric function and the optical conductivity, as predicted by Taniguchi and Altshuler.

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FIG. 1. The electronic spectrum in a 1-D periodic potential under a laser field of intensity \( x = 10 \). Energy bands are shown as a function of the Bloch momentum \( k \).
FIG. 2. Histograms giving the nearest-neighbor spacing distributions for the energy spectra of Eq. (1) under increasing field intensity (from top to bottom). Solid and dashed lines correspond to GOE and Poisson statistics respectively. Note that, as the field intensity increases, the distribution evolves from Poisson-like into GOE-like, and then back to Poisson-like.
FIG. 3. Least square deviation statistics $\Delta_3(L)$ for the indicated values of the field intensity parameter $x$. The solid and dashed lines are the GOE and Poisson regimes, respectively.