P H Y S I C A L   R E V I E W   L E T T E R S

Anderson Wall and Bloch Oscillations in Molecular Rotation

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We describe a universal behavior of linear molecules excited by a periodic train of short laser pulses under quantum resonance conditions. In a rigid rotor, the resonance causes an unlimited ballistic growth of the angular momentum. We show that the centrifugal distortion of rotating molecules eventually halts the growth, by causing Anderson localization beyond a critical value of the angular momentum—the Anderson wall. Its position solely depends on the molecular rotational constants and lies in the range of a few tens of $\hbar$. Below the wall, rotational excitation oscillates with the number of pulses due to a mechanism similar to Bloch oscillations in crystalline solids. We suggest optical experiments capable of observing the rotational Anderson wall and Bloch oscillations at near-ambient conditions with the help of existing laser technology.

The periodically kicked rigid rotor is a standard model in nonlinear dynamics studies [1,2]. The classical kicked rotor exhibits truly chaotic motion with an unbounded diffusive growth of the angular momentum J. In the quantum regime, this chaotic motion is either suppressed by a mechanism similar to Anderson localization in disordered solids [3,4], or the rotational excitation is enhanced due to the so-called quantum resonance [5,6]. Although these fundamental quantum phenomena have been theoretically studied for several decades already, there has not been a single experiment demonstrating Anderson localization in an actually rotating system. Several known experiments were done in a substitute system—cold atoms interacting with a pulsed standing light wave—imitating the dynamics of the planar kicked rotor [7–10]. An early proposal [11] on using microwave excited polar molecules as a testing ground for Anderson localization has never been realized (probably due to the complexity of the required field source). Recently [12,13], we drew attention to the fact that current technology used for laser alignment of molecules offers tools for exploring the dynamics of the periodically kicked quantum rotor in a molecular system. As a first step following our proposal [12], quantum resonance excited by a few-pulse train was observed in molecular nitrogen [14].

In this Letter, we present two qualitatively new rotational phenomena in linear molecules subject to moderately long resonant trains of laser pulses readily available nowadays. We show that the resonant growth of the molecular angular momentum is suppressed by centrifugal distortion, which causes Anderson localization beyond a critical value J_A of the angular momentum—the Anderson wall. Resonant rotational excitation below J_A exhibits beats similar to Bloch oscillations of electrons in crystalline solids subject to a dc electric field [15,16]. The amplitude of the oscillations grows with the intensity of the laser pulses, but is restricted by the Anderson wall. These phenomena are absent in the standard kicked rotor model or its atom optics implementation, but are universal

features of kicked molecules. We suggest several optical experiments on observing these phenomena. Our results are of importance for multiple current experiments employing resonant laser kicking for enhanced molecular alignment [17], isotope-selective excitation [14,18], and impulsive gas heating for Raman photoacoustics [19] and controlling high power optical pulse propagation in atmosphere [20].

We consider linear molecules interacting with short nonresonant linearly polarized laser pulses, as used in standard experiments on molecular alignment (for reviews see [21–23]). The laser field affects the molecular rotation via a Raman-type interaction [24,25]. The electric field of the pulse induces anisotropic molecular polarization, interacts with it, and tends to align the molecular axis along the laser polarization direction. An ultrashort laser pulse acts like a kick exciting molecular rotation, and the alignment is observed under field-free conditions after the pulse [26–28]. The laser-molecule interaction is given by

$$V = -\frac{1}{4}\Delta\alpha\cos^2\theta\sum_{n=0}^{N-1}\mathcal{E}^2(t-n\tau). \tag{1}$$

Here, $\mathcal{E}(t)$ is the temporal envelope of a single laser pulse, $\Delta\alpha = \alpha_\parallel - \alpha_\perp$, where $\alpha_\parallel$ and $\alpha_\perp$ are the polarizability along the molecular axis and perpendicular to it, N is the number of pulses, τ is the period of the train, and θ is the angle between the laser polarization direction and the molecular axis. We introduce the effective interaction strength $P = \Delta\alpha/(4\hbar)\int\mathcal{E}^2(t)dt$. It reflects the typical change of the molecular angular momentum (in units of $\hbar$) induced by a single laser pulse. The selection rules for the interaction are $\Delta J = 0, \pm2$ and $\Delta M_J = 0$, where J is the angular momentum and M_J its projection on the laser polarization direction.

The rotational levels of a linear molecule are [29]

$$E_J = BJ(J+1) - DJ^2(J+1)^2, \tag{2}$$

where B and D are the rotational and the centrifugal distortion constant, respectively. For low-lying rotational states one may

neglect the second term in Eq. (2), and the rotational dynamics is defined by a single parameter, the rotational revival time $t_{rev} = \pi\hbar/B$. Any rotational wave packet of a free rotor reproduces itself after integer multiples of the revival time. Short kicks separated in time by t_{rev} add constructively their actions, and therefore, the molecular angular momentum grows ballistically (linear) with N. This effect is called quantum resonance. As the molecule rotates faster, the centrifugal force pulls the atoms apart, and due to the increase of the moment of inertia, the molecule becomes detuned from the quantum resonance, with consequences described below.

A good way to understand the dynamics of a periodically driven quantum system is by looking at its quasienergy states (Floquet states) [30], the eigenstates of a one-cycle (pulse-to-pulse) evolution operator. The quasienergy eigenstate $|\chi_\varepsilon\rangle(t)$ reproduces itself after a one-period evolution up to a certain phase factor:

$$|\chi_\varepsilon\rangle(t + \tau) = e^{-i\varepsilon\tau/\hbar}|\chi_\varepsilon\rangle(t), \qquad (3)$$

where ε is the quasienergy eigenvalue. It was shown [4] that finding quasienergy states for the kicked rotor problem can be mapped onto solving a stationary Schrödinger equation for a one-dimensional tight-binding model, in which each site corresponds to a J state and the energy of the site is proportional to

$$T(J) = \tan\left[\tau(\varepsilon - E_J)/(2\hbar)\right]. \qquad (4)$$

For resonant kicking ($\tau = t_{rev}$), this quantity becomes

$$T_r(J) = \tan[\phi(J)]; \qquad \phi(J) = \frac{\pi}{2}\left[\frac{\varepsilon}{B} + \frac{D}{B}J^2(J+1)^2\right]. \quad (5)$$

Here, we omitted integer multiples of π in $\phi(J)$. Since $D/B \ll 1$ (see Table I), one may neglect the second term in $\phi(J)$ for small J, and Eq. (5) corresponds to a completely periodic lattice of identical sites. The energy spectrum of such a system has a band structure related to an unlimited propagation along the lattice, reflecting the quantum resonance. For large J, the second term in $\phi(J)$ becomes important. Since D/B is in general an irrational number, $T_r(J) = \tan[\phi(J)]$ behaves as a pseudorandom function for large enough J. Using speculations similar to those of Ref. [4], one may argue that this pseudorandomness leads

to a localization of the quasienergy states in J space, similar to electronic states localized in disordered solids, as predicted by Anderson in 1958 [3]. The localization is likely to start when the difference of phases $\Delta\phi(J) = \phi(J + 2) - \phi(J)$ for two neighboring coupled sites reaches the value of $\sim\pi/2$, on the halfway between zero and the π period of the tangent function in Eq. (5), i.e., at

$$J \equiv J_A \sim \frac{1}{2}\sqrt[3]{\frac{B}{D}}. \qquad (6)$$

The value of J_A—the "Anderson wall" separating delocalized and localized quasienergy states under resonant driving—is a distinct attribute of every molecule. In Table I, we show its estimated value for several molecules.

To test our arguments, we calculated the quasienergy states for $^{14}N_2$ molecules kicked periodically at the rotational revival time $t_{rev} = 8.38$ ps by laser pulses of $P = 3$. This value corresponds to 50 fs long pulses (full width at half maximum of the intensity envelope) of peak intensities of about 40 TW/cm^2, which is below the onset of ionization [17,34]. For the range of J states considered here, such a pulse is shorter than the rotational periods involved, and it acts as a delta pulse. We have verified that the finite duration of the pulses has no qualitative influence on the results presented below. The details of our numerical procedure are described in [13]. Figure 1 shows the absolute squares of the projection coefficients $|\langle J, 0|\chi_\varepsilon\rangle|^2$ for several representative quasienergy states. For low values of J, one can see extended states with an almost constant (in logarithmic scale) amplitude having a cutoff around $J_R \sim 20$. The extended character of these states provides the initial ballistic growth of the angular momentum under the condition of quantum resonance. For $J \geq J_A \sim 35$, the quasienergy states are localized

TABLE I. Rotational revival time t_{rev}, ratio D/B of the centrifugal distortion constant and the rotational one, and estimated position J_A of the Anderson wall, for typical linear molecules. All data are for the vibrational ground state. Rotational constants are taken from [31–33].

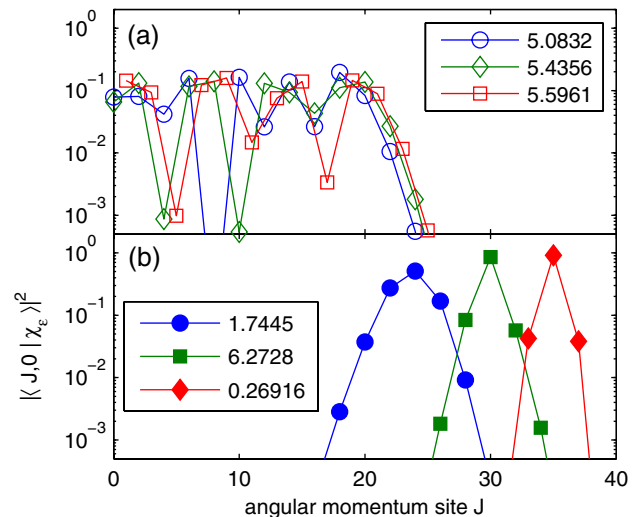|	t_{rev} in ps	D/B 10^6	estimated J_A
H_2	0.281	794	~5
N_2	8.383	2.90	~35
Cl_2	68.57	0.765	~55
CO_2	42.74	0.343	~70
Br_2	203.5	0.255	~80
OCS	82.22	0.214	~80

FIG. 1 (color online).   Selected quasienergy states $|\chi_\varepsilon\rangle$ (projected on the angular momentum states $|J, 0\rangle$), for resonantly kicked $^{14}N_2$ molecules (kick strength $P = 3$). Panel (a) shows extended states in the region of small angular momentum J, panel (b) shows localized states close to and above the Anderson wall. The legends depict the quasienergy values (in units of $\hbar/t_{rev}$) modulo 2π.

around single angular momentum sites, in good agreement with the above arguments on Anderson localization. There is a transition region $J_R < J < J_A$ in which the quasienergy states span over a couple of momentum sites. The cutoff position $J_R \lesssim J_A$ grows with P, while the position of the Anderson wall is P independent.

The structure of the quasienergy states leads to some general conclusions about the rotational dynamics of molecules subject to resonant kicking by laser pulses. If a molecule is initially in a low-lying rotational level, its wave function has reasonable overlap only with the extended quasienergy states, but not the localized states beyond the Anderson wall. Therefore, the molecule cannot be excited above $J = J_A$, even if driven by a very long pulse train. Moreover, as the initial rotational wave function overlaps only with a finite number of quasienergy states, the driven rotational dynamics is almost periodic, and the molecules should closely reconstruct their initial state many times for sufficiently long pulse trains [35]. If, however, the initial angular momentum is above J_A, we expect the wave function to be stuck to its initial momentum due to Anderson localization.

Figure 2 demonstrates how these general statements reveal themselves in the above example of resonantly kicked $^{14}N_2$ molecules. It shows the calculated angular momentum distribution as a function of the number of pulses for different initial molecular states $|J_0, 0\rangle$. The used numerical procedure is the same as in [36,37]. For a molecule initially at rest [Fig. 2(a)], one sees first a ballistic growth of the angular momentum, where each pulse shifts the distribution by about $3\hbar$. After eight pulses, the wave of excitation reaches $J_R \approx 20$, is reflected, and ballistically propagates back towards $J = 0$. After 16 pulses, the molecule (approximately) returns to the initial state, and the cycle restarts. For $0 < J_0 < J_R$ [Fig. 2(b)], there are two streams of rotational excitation, directed initially upwards and downwards, since in half of the angular space the direction of the kick coincides with the initial rotational velocity, whilst in the other half it is directed oppositely. Each of the streams oscillates with time between zero and J_R. For an initial state lying in the transition region, $J_R < J_0 < J_A$, [Fig. 2(c)], the amplitude of the oscillations of the angular momentum distribution is strongly reduced. Finally, if the initial state lies behind the Anderson wall [Fig. 2(d)], the laser pulses hardly lead to any excitation or deexcitation. The angular momentum distribution is stuck to the initial state (Anderson localization).

To reveal the physical origin of the oscillations observed for $J \lesssim J_R$, let us first consider the limit of weak pulses, $P \ll 1$. In this case, the potential of Eq. (1) couples only nearest neighbors in the J lattice, and the change of the rotational excitation from pulse to pulse is small. Therefore, our rotational system corresponds to a quantum particle moving continuously in the J lattice. In the absence of centrifugal distortion, the lattice sites are identical, and the quasienergy states are characterized by a continuous dimensionless quasimomentum $\vec{k}$ describing propagation of the Bloch wave function along the J lattice [38]. The
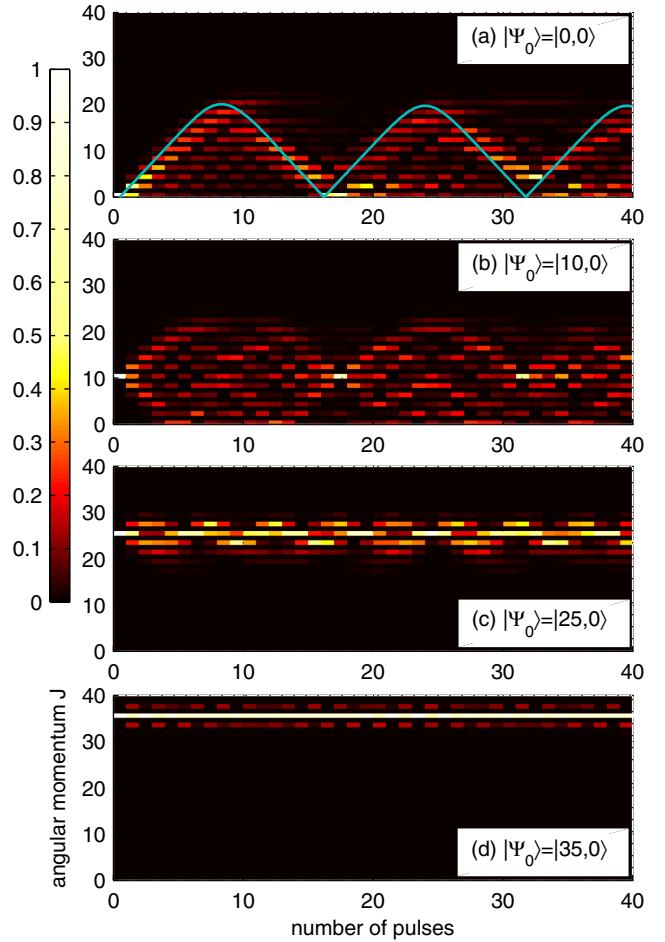

FIG. 2 (color online). Simulated angular momentum distribution as a function of the number of pulses, for resonantly kicked $^{14}N_2$ molecules. Each panel shows the result for a different initial state $|\Psi_0\rangle$. Pulses are 50 fs long, with kick strength $P = 3$. The solid line in panel (a) is a solution of the semiclassical model Eq. (7).

quasienergy spectrum of this "particle" has a band structure: $\varepsilon(k) = -\hbar P/(2t_{rev})\cos(2k)$. Due to the nonrigidity of the molecules, there is an additional "potential" $U(J)$, which is proportional to the single-cycle phase added to each rotational state by the centrifugal distortion correction of the rotational levels Eq. (2): $U(J) = -\pi(D/B)J^2(J + 1)^2$. Notice that this "potential" may be additionally controlled by a slight detuning of the train period from the resonant value. Analogous to solid state physics [15,16], one can derive semiclassical equations of motion for the quasimomentum k and the lattice coordinate J:

$$\frac{dk}{dn} = -\frac{dU(J)}{dJ}; \qquad \frac{dJ}{dn} = \frac{t_{rev}}{\hbar}\frac{d\varepsilon(k)}{dk} = P\sin(2k). \quad (7)$$

Here, the dimensionless time n is a continuous analog of the number of pulses. The first equation is Newton's second law, and the second one defines the group velocity of the Bloch waves. As we will show elsewhere, Eqs. (7) can also be obtained by the "ϵ-classics" approach [39], lifting the condition $P \ll 1$. Equations (7) are similar to the ones describing Bloch oscillations of electrons in crystalline

solids subject to a constant electric field [15,16], with the difference that here the accelerating "force" $F = -dU(J)/dJ$ is "coordinate" dependent. Since $U(J) < 0$, the "force" F initially leads to accelerated motion away from $J = 0$. However, eventually the solution for $J(n)$ oscillates due to Bragg reflection of the Bloch waves from the edge of the Brillouin zone of the J lattice. In Fig. 2(a), the solution for $J(n)$ in the domain $J \geq 0$ is plotted as a solid line. As initial quasimomentum, we chose $k(0) = \pi/4$, which corresponds to the initial growth rate $dJ/dn = P$ of the angular momentum (the typical change of J by a single pulse). The agreement between the exact quantum mechanical solution and the semiclassical model of Eq. (7) is very good.

The nonstationary rotational distribution can be measured with time-resolved coherent Raman scattering spectroscopy, especially utilizing schemes in which full spectral information is retrieved using a single femtosecond pulse [40]. Moreover, the evolution of the angular momentum distribution is exhibited in the time-dependent alignment signal, defined as the expectation value $\langle \cos^2 \theta \rangle (t)$. The deviation of this quantity from the isotropic value $1/3$ determines the laser induced anisotropy of the gas refraction index (birefringence), which can be measured by optical methods [21–23,41]. In Fig. 3, the simulated angular momentum distribution and alignment signal for resonantly kicked $^{14}N_2$ at room temperature are shown. The finite temperature is included in the simulation by calculating the signal for different initial states and summing the results, weighted by the corresponding Boltzmann factors (see [36,37]). No collisions are assumed to happen during the pulse train duration. Remarkably, the sawlike pattern seen in Fig. 2 survives quite well for the incoherent thermal initial state. Regarding the alignment
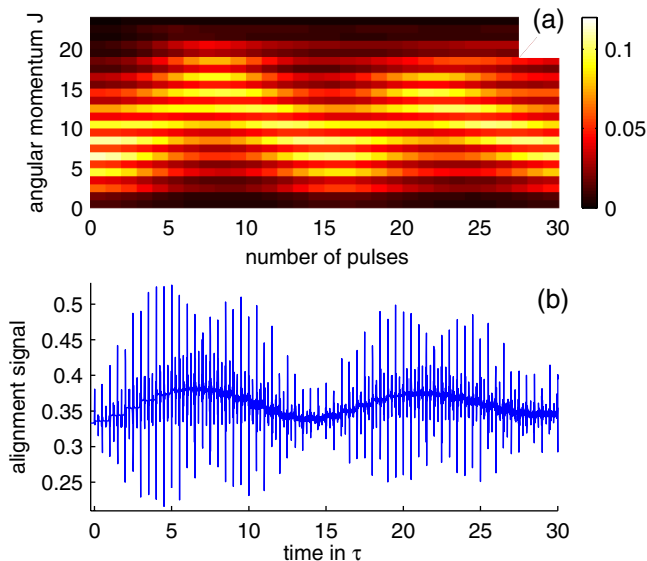

FIG. 3 (color online). Simulated (a) angular momentum distribution and (b) alignment signal $\langle \cos^2 \theta \rangle$ as a function of the number of pulses, for resonantly kicked $^{14}N_2$ at $T = 298$ K. Pulses are 50 fs long with kick strength $P = 3$. Nuclear spin statistics cause different intensities of even and odd J [29].

signal, one can see low-frequency oscillations of the baseline (time-averaged alignment). They can be attributed to the Bloch oscillations of the angular momentum distribution. The conditions used for our simulations shown in Fig. 3 are very close to the conditions of the experiment described in [17], where molecular alignment induced by a periodic train of laser pulses was measured. This experiment used 50 fs 800 nm pulses with peak intensities of 36 TW/cm^2, separated in time by $\sim$8.4 ps, interacting with molecular nitrogen at STP conditions. By comparing Fig. 3(b) with Fig. 2 from Ref. [17], we conclude that Cryan et al. had actually reached the reflection point J_R, but did not observe the Bloch oscillation itself, since their number of pulses was limited to eight. A decisive experiment to monitor a full Bloch oscillation cycle requires about 16 pulses. Such a pulse train was successfully demonstrated in [42]. The total length of this train is 125 ps, which is comparable to a single collision time at STP conditions. Potential detrimental decoherence effects can be avoided by a slight reduction of the pressure used in [17]. An alternative route to detect the Bloch oscillations of the angular momentum distribution is to measure the absorbed rotational energy as a function of the number of pulses. This can be done via the photoacoustic approach [43,44], which measures the heat dumped into the gas as a result of rotational relaxation. To demonstrate localization above the Anderson wall, one may use the optical centrifuge technique [45–48] to prepare molecules in high-J initial states.

Concluding, we described two interrelated phenomena—Anderson wall and Bloch oscillations—for linear molecules that are excited by a periodic train of short laser pulses under the condition of quantum resonance. These effects are rotational counterparts of quantum localization phenomena in solid state physics, and can be observed in common molecules (N_2, CO_2, OCS, …) using existing schemes for generating laser pulse trains and a variety of available detection methods. Such experiments may result in the first observation of Anderson localization in a material kicked rotor. As shown here, laser experiments on rotating molecules provide a new testing ground for quantum localization effects, in addition to semiconductor superlattices [49], cold atoms in optical lattices [50,51], and coupled photonic structures [52–56]. Most remarkably, these effects can be observed at near-ambient conditions. Finally, our predictions have immediate implications for numerous current experimental schemes using resonant laser kicking for molecular alignment [17], isotope-selective excitation [14,18], and impulsive gas heating for Raman photoacoustics [19] and controlling high power optical pulse propagation [20].

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