Angular Focusing, Squeezing, and Rainbow Formation in a Strongly Driven Quantum Rotor

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Semiclassical catastrophes in the dynamics of a quantum rotor (molecule) driven by a strong time-varying field are considered. We show that for strong enough fields, a sharp peak in the rotor angular distribution can be achieved via a time-domain focusing phenomenon, followed by the formation of rainbowlike angular structures. A strategy leading to the enhanced angular squeezing is proposed that uses a specially designed sequence of pulses. The predicted effects can be observed in many processes, ranging from molecular alignment (orientation) by laser fields to heavy-ion collisions, and the trapping of cold atoms by a standing light wave.

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The driven rotor is a standard model in classical and quantum nonlinear dynamics studies [1]. Increasing interest in the problem has arisen because of a recent atom optics realization of the quantum δ-kicked rotor [2], and novel experiments on molecule orientation (alignment) by strong laser fields [3–6]. A strong enough laser field creates the so-called pendular molecular states [7] that are hybrids of field-free rotor eigenstates. By adiabatically turning on the laser field, it is possible to trap a molecule in the ground pendular state, thus leading to molecular alignment. The only way to reach a considerable degree of alignment in this approach is by increasing the intensity of the field. However, many applications may require only a transient molecular alignment (orientation), when the molecular angular distribution becomes extremely squeezed at some predetermined moment of time. It is well known that a physically related problem of squeezed states generation in a harmonic system may be solved by a proper time modulation of the driving force (parametric resonance excitation). Behavior of a rotor in general, strong, time-varying fields is a much less-studied problem, although it is understood that the long-persisting beats in the molecular angular distribution may be induced by short laser pulses [8–14].

In the present paper, we analyze generic features in the dynamics of a quantum rotor (molecule) driven by strong pulses, and present a strategy for efficient squeezing of the rotor angular distribution by a sequence of pulses of moderate intensity.

We start with a generic Hamiltonian:

$$\hat{H} = \frac{\hat{L}^2}{2I} + V(\theta, t), \tag{1}$$

where $\hat{L}$ is the angular momentum operator, and I is the momentum of inertia of the rotor. For a linear molecule having a permanent dipole moment μ, and driven by a linearly polarized field, the interaction potential is

$$V(\theta, t) = -\mu \mathcal{E}(t)\cos(\theta), \tag{2}$$

where $\mathcal{E}(t)$ is the field amplitude (e.g., of a half-cycle pulse), and θ is the polar angle between the molecular axis and the field direction. In the absence of interaction with a permanent dipole moment, the external field couples with the induced molecular polarization. For nonresonant laser fields, this interaction, averaged over fast optical oscillations, is (see, e.g., [7,15])

$$V(\theta, t) = -1/4\mathcal{E}^2(t)[(\alpha_{\parallel} - \alpha_{\perp})\cos^2(\theta) + \alpha_{\perp}]. \tag{3}$$

Here $\alpha_{\parallel}$ and $\alpha_{\perp}$ are the components of the polarizability, parallel and perpendicular to the molecular axis, and $\mathcal{E}(t)$ is the *envelope* of the laser pulse. Although the two above forms of $V(\theta, t)$ may lead to different physical consequences (i.e., orientation vs alignment), the effects we will present are more or less insensitive to the choice of interaction. Therefore, we chose $V(\theta, t)$ from Eq. (2) for the following presentation. Moreover, for the sake of clarity, we will focus mainly on the simplest model of a two-dimensional rigid rotor, which contains almost all of the physics, and we will only briefly mention the additional effects that appear in 3D. The detailed study of the three-dimensional case will be published elsewhere [16]. It is worth mentioning that 2D rotor is a standard model in nonlinear dynamics studies, and it also describes behavior of cold atoms trapped in pulsed optical lattices [2].

By introducing dimensionless time $\tau = t\hbar/I$, and interaction strength $\varepsilon = \mu\mathcal{E}(t)I/\hbar^2$, the Hamiltonian can be written as

$$\hat{H} = -\frac{1}{2}\frac{\partial^2}{\partial\theta^2} - \varepsilon(\tau)\cos(\theta).$$

The wave function of the system can be expanded in the eigenfunctions of a free rotor

$$\Psi(\theta, \tau) = \frac{1}{\sqrt{2\pi}}\sum_{n=-\infty}^{+\infty} c_n(\tau)e^{in\theta}.$$

In the absence of the field, the wave function takes the form

$$\Psi(\theta, \tau) = \frac{1}{\sqrt{2\pi}}\sum_{n=-\infty}^{+\infty} c_n(0)e^{-in^2\tau/2 + in\theta}. \tag{4}$$

Despite a simple form of Eq. (4), the wave function exhibits extremely rich space-time dynamics. In particular, it shows periodic behavior in time with the period $T_{\rm rev} = 4\pi$ (full revival) and a number of fractional revivals at $\tau = p/sT_{\rm rev}$ and (p and s are mutually prime numbers) [17]. An analytical solution valid for a general time-dependent field is unknown even for this simplest model. Much effort has been devoted to the case of extremely short field pulses (δ kicks) (see, e.g., [1]—and references therein). In general, as a result of a single kick applied to the rotor at $\tau = \tau_k$, the coefficients c_n transform as

$$c_n(\tau_k + 0) = \sum_{m=-\infty}^{+\infty} i^{n-m} J_{n-m}(P) c_m(\tau_k - 0), \qquad (5)$$

where

$$P = \int_{-\infty}^{+\infty} \varepsilon(\tau)\, d\tau,$$

and $J_n(P)$ is the Bessel function of the nth order. The result of multiple kicks applied at different times can be obtained by combining transformations (5) after each kick with a free evolution according to Eq. (4) between the kicks. If the kicks are applied periodically to the system with the period $T_{\rm rev}$, the system does not show chaotic behavior, and the energy accumulates quadratically with time (the so-called "quantum resonance" [2,18,19]). It is, therefore, quite natural to examine potential accumulation of angular squeezing of the rotor wave function under the "quantum resonance" excitation. In this case, because of the exact quantum revivals at the free-evolution stages, the effect of N kicks of a magnitude P is equivalent to the action of a single strong pulse of strength NP (see, e.g., [18]). In Fig. 1, we show numerically calculated time evolution of the probability density $|\Psi(\theta, \tau)|^2$ after a relatively strong kick of a magnitude $P = 85$ applied at $\tau = 0$. Initially the rotor was in the ground s state $[c_n(0) = \delta_{n0}]$. For the chosen values of τ, several distinct phenomena can be seen in these plots. First of all, the wave function shows an extreme narrowing in the region of small θ after some delay following the kick (Fig. 1b). The physics of this effect may be understood with the help of the following semiclassical arguments. Consider an ensemble of randomly oriented classical rotors subject to a kick. The angular velocity of a rotor located at the angle θ is

$$\omega(\theta) = -P \sin(\theta) \qquad (6)$$

just after the kick, assuming negligible initial velocity. For rotors from the region of small $\theta \ll 1$, the acquired velocity is linearly proportional to their initial angle, so that all of them arrive at the focal point $\theta = 0$ at the same time

$$\tau_f = 1/P. \qquad (7)$$

This phenomenon is quite similar to the focusing of light rays by a thin optical lens. For $P \gg 1$, the shape of the distribution at the focusing time τ_f is dictated by the aberration mechanism [deviation of the $\cos(\theta)$ potential from the parabolic one], and it is P independent. We consider the orientation factor $O = \langle 1 - \cos(\theta)\rangle$ (where angular brackets mean averaging over the state of rotor) as a measure of the rotor orientation. For large enough P, the time-dependent orientation factor (for the initial s state) may

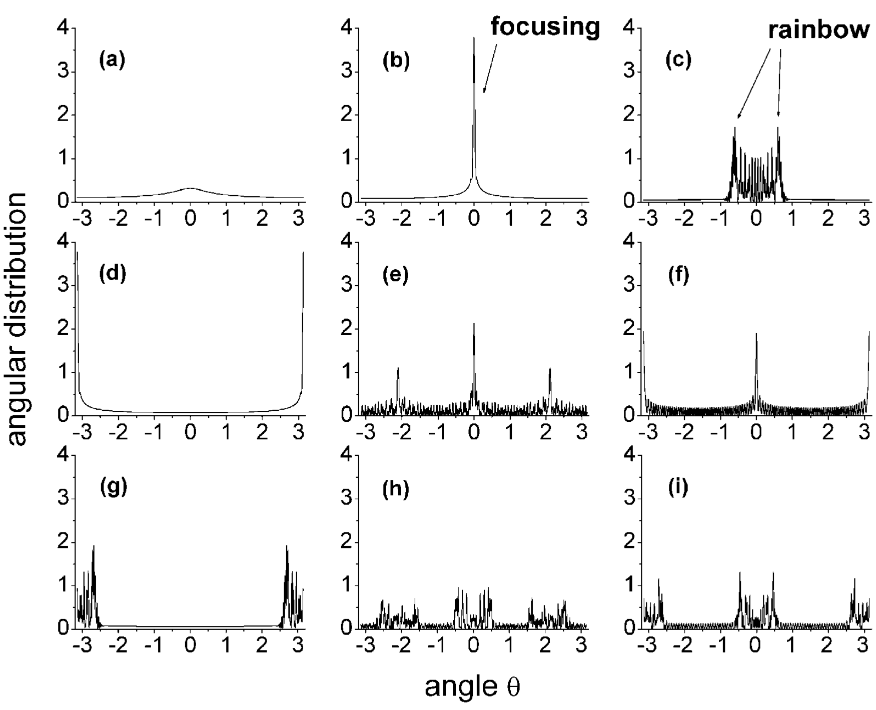

FIG. 1. Angular distribution of a quantum rotor excited by a strong δ kick ($P = 85$). The graphs correspond to (a) $\tau = 0.5\tau_f$, (b) $\tau = \tau_f$, (c) $\tau = 2\tau_f$, (d) $\tau = \tau_f + T_{\rm rev}/2$, (e) $\tau = \tau_f + T_{\rm rev}/3$, (f) $\tau = \tau_f + T_{\rm rev}/4$, (g) $\tau = 1.8\tau_f + T_{\rm rev}/2$, (h) $\tau = 1.8\tau_f + T_{\rm rev}/3$, and (i) $\tau = 1.8\tau_f + T_{\rm rev}/4$, respectively.

be easily estimated by averaging over the initially uniform classical ensemble of rotors having the velocity distribution of Eq. (6): $O(\tau) = 1 - J_1(P\tau)$. Here $J_1(x)$ is Bessel function of the first order. The minimal value $O \approx 0.418$ of the orientation factor is, in fact, achieved in the post-focusing regime, at $\tau \approx 1.84\tau_f$. As seen in Fig. 1c, a new phenomenon can be observed in the angular probability distribution just after the focusing. Sharp singularlike features are formed in the distribution, which are moving with time. Each of these features has a typical asymmetric shape, with pronounced oscillations on one side and an abrupt drop down on the other side. Again, the origin of this effect can be traced in the time evolution of a classical ensemble of initially motionless rotors.

After a kick applied at $\tau = 0$, the motion of the rotors is described by

$$\theta = \theta_0 - P\sin(\theta_0)\tau \pmod{2\pi}, \tag{8}$$

where θ_0 is the initial angle. For $\tau < \tau_f$ Eq. (8) represents a one-to-one mapping $\theta(\theta_0)$ (see Fig. 2a). At $\tau = \tau_f$ the curve $\theta(\theta_0)$ touches the horizontal axis (Fig. 2b). At $\tau > \tau_f$ the angle θ_0 becomes a multivalued function of θ (Figs. 2c and 2d). The classical time-dependent angular distribution function of the ensemble is given by

$$f(\theta, \tau) = \sum_a \frac{f(\theta_0^a, \tau = 0)}{|d\theta/d\theta_0^a|}. \tag{9}$$

The summation in (9) is performed over all possible branches of the function $\theta_0(\theta)$ defined by Eq. (8). It follows immediately from Eq. (9) that even for a smooth initial distribution, $f(\theta, \tau)$ may exhibit a singular behavior near the angles where $d\theta/d\theta_0^a \to 0$. The quantum nature of the rotor motion replaces the classical singularities by sharp maxima in the probability distribution with the Airy-like shape typical to rainbow phenomena. Indeed, this effect is similar to the formation of caustics in the wave optics [20], and rainbow-type scattering in optics and quantum mechanics [21–23]. We should stress, however, that the long-time asymptotic regimes are radically different for the classical and truly quantum motion of the rotor. Thus, contrary to the classical limit, in which the caustics exist forever, they gradually disappear in the quantum case because of the overall decay of the initial rotational wave packet. On even longer time scale, another quantum phenomenon can be seen, namely revivals and fractional revivals of the initial classical-like motion. Figures 1e–1i show several examples of fractional foci and rainbows in the angular distribution, which is a purely quantum effect.

As we have demonstrated, a mere application of δ kicks at the condition of "quantum resonance" does not lead to accumulated angular squeezing, and the orientation is saturated at some finite asymptotic level. Here we suggest an excitation scheme that exhibits the desired accumulation property. As previously mentioned, the wave function of the rotor reaches the state of the maximal orientation (i.e., minimal O value) after a certain delay $\Delta\tau_1$ following the application of the first kick at $\tau = \tau_1 = 0$. We suggest to apply the second kick at $\tau_2 = \Delta\tau_1$. Immediately after the second kick, the system will keep the same probability density distribution. On the other hand, $\tau = \tau_2$ will no longer be a stationary point for $O(\tau) = \langle 1 - \cos(\theta)\rangle(\tau)$. The orientation factor $O(\tau)$, and its derivative are continuous and periodic functions of time in the course of a free evolution. Therefore, $O(\tau)$ will reach a new minimum at some point $\tau_2 + \Delta\tau_2$ in the interval $[\tau_2, \tau_2 + T_{\mathrm{rev}}]$. Clearly, the new minimal value of the orientation factor is smaller than the previous one. By continuing this way, we will apply short kicks at iterative time instants $\tau_{k+1} = \tau_k + \Delta\tau_k$. By construction of this pulse sequence, the squeezing effect will accumulate with time, in contrast to the quantum resonance excitation. This is demonstrated by Fig. 3, which shows calculated sequences $\{\Delta\tau_k\}$ and $\{\langle 1 - \cos(\theta)\rangle(\tau_k)\}$ for a rotor initially in the s state and being kicked by pulses with $P = 3$. The logarithm of the orientation factor gradually decreases, without any sign of saturation.

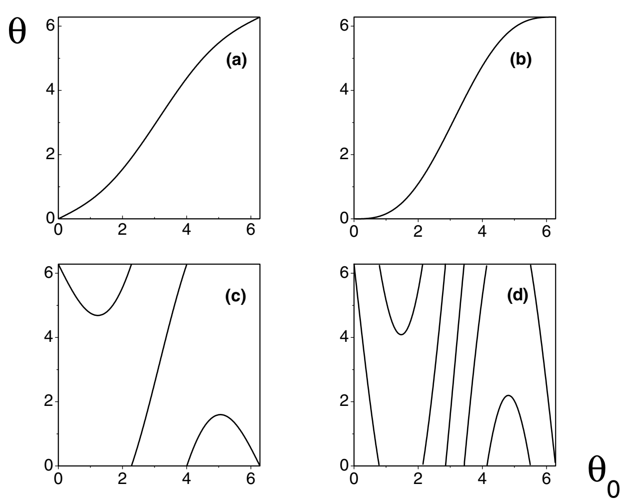

FIG. 2. Classical map representing the final angle θ as a function of initial angle θ_0 for (a) $\tau = 0.5\tau_f$, (b) $\tau = \tau_f$, (c) $\tau = 3\tau_f$, and (d) $\tau = 10\tau_f$.

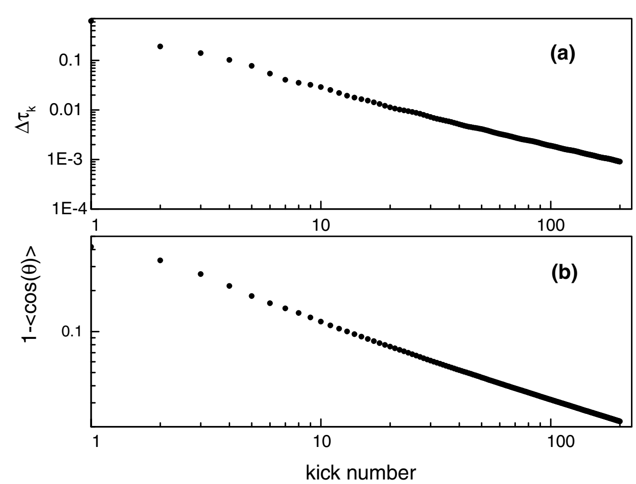

FIG. 3. Accumulative angular squeezing. Graphs are shown in double logarithmic scale.

At the stage of a well-developed squeezing ($O \ll 1$), the $\cos(\theta)$ potential may be approximated by a parabolic one. It can be easily shown (both classically and quantum mechanically) that in this limit our strategy provides the following recurrent relationships for the time intervals $\Delta\tau_k$, successive values of the angular variance $u_k = \langle\theta^2\rangle_k \approx 2O_k$, and normalized variance of the angular velocity $w_k = \langle(-id/d\theta)^2\rangle_k/P^2$:

$$\begin{aligned} \Delta\tau_k &= \frac{u_k}{u_k + w_k}, \\ u_{k+1} &= u_k - \frac{u_k^2}{u_k + w_k}, \\ w_{k+1} &= w_k + u_k. \end{aligned} \tag{10}$$

For large k, the last two finite-difference equations may be replaced by a system of coupled differential equations. The latter has an exact solution providing the following asymptotics: $\langle\theta^2\rangle_k \propto 1/\sqrt{k}$ and $\Delta\tau_k \propto 1/k$. This result is in good agreement with the numerically observed power-laws behavior of graphs 3a and 3b, and it describes correctly their slopes at $k \gg 1$. We note that in contrast to the wave optics (in which the size of the focal spot is diffraction limited), our system may be, in principle, "unlimitedly" squeezed in angle. We also note that a quasiperiodic sequence of kicks applied at $\tau_{k+1} = \tau_k + \Delta\tau_k + T_{\rm rev}$ provides the same squeezing scenario for a quantum rotor. The introduction of the $T_{\rm rev}$ shift between pulses may be useful in the practical realizations of the scenario to avoid the overlap between short excitation pulses of a finite duration.

All the described phenomena are rather common under general conditions of a strong excitation of a quantum rotor. The results of direct quantum numerical simulations we performed for various interaction types and shapes $\epsilon(\tau)$ of the strong field in the three-dimensional case are in a good agreement with the above quasiclassical arguments even after thermal averaging [16]. The most spectacular additional features observed in the 3D problem are the *glory*-type singularities [21] at $\theta = 0, \pi$ in the rotor angular distribution. They appear in the post-focusing regime, when the newly emerging branches of the map (8) cross the lines $\theta = 0, \pi$.

The predicted effects may be observed in a wide range of systems with strongly driven rotational degrees of freedom. Possible examples range from heavy-ion collitions (when highly excited wave packets of nuclear rotational states are produced [9]) to molecules subject to strong laser pulses, and cold atoms trapped by standing light waves. The dynamics of the last system is formally related to that one of the driven rotor [2]. The spectacular features described in our paper may be observed in the spatial distribution of an atomic ensemble driven by pulsed optical lattices. Moreover, the related squeezing approaches may find application in atom lithography of ultrahigh resolution [24]. In the case of molecules, the considered effects may reveal themselves in the angular distribution of fragments produced by intense laser-field molecular interaction. The most direct evidence can be achieved in a two-pulse experiment, in which the first strong nonresonant pulse attempts to orient the molecular ensemble, while the second short delayed pulse creates fragment ions.

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[1] F. Haake, *Quantum Signatures of Chaos* (Springer-Verlag, Berlin, 1991).

[2] F. L. Moore *et al.,* Phys. Rev. Lett. **75**, 4598 (1995).

[3] D. Normand, L. A. Lompre, and C. Cornaggia, J. Phys. B **25**, L497 (1992).

[4] P. Dietrich *et al.,* Phys. Rev. A **47**, 2305 (1993).

[5] G. R. Kumar *et al.,* Phys. Rev. A **53**, 3098 (1996).

[6] J. Karczmarek *et al.,* Phys. Rev. Lett. **82**, 3420 (1999).

[7] B. Friedrich and D. Herschbach, Phys. Rev. Lett. **74**, 4623 (1995); J. Phys. Chem. **99**, 15 686 (1995).

[8] C. H. Lin, J. P. Heritage, and T. K. Gustafson, Appl. Phys. Lett. **19**, 397 (1971); J. P. Heritage, T. K. Gustafson, and C. H. Lin, Phys. Rev. Lett. **34**, 1299 (1975).

[9] L. Fonda, N. Mankoč-Borštnik, and M. Rosina, Phys. Rep. **158**, 159 (1988).

[10] P. M. Felker, J. Phys. Chem. **96**, 7844 (1992).

[11] T. Seideman, J. Chem. Phys. **103**, 7887 (1995); **106**, 2881 (1997).

[12] T. Seideman, Phys. Rev. Lett. **83**, 4971 (1999).

[13] J. Ortigoso *et al.,* J. Chem. Phys. **110**, 3870 (1999).

[14] L. Cai, J. Marango, and B. Friedrich, Phys. Rev. Lett. **86**, 775 (2001).

[15] R. W. Boyd, *Nonlinear Optics* (Academic Press, Boston, 1992).

[16] I. Sh. Averbukh, R. Arvieu, and M. Leibscher (to be published).

[17] I. Sh. Averbukh and N. F. Perelman, Phys. Lett. A **139**, 449 (1989); Sov. Phys. JETP **69**, 464 (1989).

[18] F. M. Izrailev *et al.,* in *Proceedings of the Conference on Stochastic Behavior in Classical and Quantum Hamiltonian Systems, Como, Italy* (Springer-Verlag, Berlin, 1977), p. 334.

[19] F. M. Izrailev and D. L. Shepelyansky, Dokl. Akad. Nauk SSSR **249**, 1103 (1979); Sov. Phys. Dokl. **24**, 996 (1979).

[20] Yu. A. Kravtsov and Yu. I. Orlov, *Caustics Catastrophes and Wave Fields,* Springer Series on Wave Phenomena, 2nd ed., Vol. 15 (Springer-Verlag, Berlin, 1999).

[21] K. W. Ford and J. A. Wheeler, Ann. Phys. **7**, 259 (1959).

[22] M. V. Berry, Adv. Phys. **25**, 1 (1976).

[23] S. D. Bosanac, J. Chem. Phys. **95**, 5732 (1991).

[24] For a recent review of atom lithography, see, e.g., Special Issue on Nanomanipulation of Atoms, edited by D. Meschede and J. Mlynek [Appl. Phys. B **70** (2000)].