Subharmonic generation in quantum systems

Martin Holthaus

Department of Physics, Center for Nonlinear Science, and Center for Free-Electron Laser Studies, University of California, Santa Barbara, CA 93106-9530

Michael E. Flatté

Institute for Theoretical Physics, University of California, Santa Barbara, CA 93106-4030

(April 19, 1993)

Abstract

We show how the classical-quantum correspondence permits long-lived subharmonic motion in a quantum system driven by a periodic force. Exponentially small deviations from exact subharmonicity are due to coherent tunneling between quantized vortex tubes which surround classical elliptic periodic orbits.

PACS numbers: 05.45.+b, 03.65.Sq, 42.65.Ky
When atoms interact with intense laser fields, extremely high harmonics can be generated. In recent experiments subjecting Ne atoms to 1-ps 10^{15}-W/cm^2 laser pulses, harmonics up to the 135th were detected [1].

The question then arises whether nonlinearities in the laser-matter interaction can also lead to the generation of subharmonics. Subharmonic motion occurs quite naturally in classical periodically-forced nonlinear oscillators, and recent investigations have shown that the correspondence principle extends much farther than previously believed [2–7]. The purpose of this Letter is to demonstrate how classical subharmonic motion survives in the corresponding quantum system.

To illustrate our considerations, we use the model of a particle in a one-dimensional triangular well interacting with an external periodic force [8,9]. We employ dimensionless “atomic units” such that the particle mass, the potential slope, and Planck’s quantum $\hbar$ are unity, and denote the strength and frequency of the driving force by $\lambda$ and $\omega$. The Hamiltonian is then given by

$$ H(p, x, t) = \frac{p^2}{2} + x + \lambda x \sin(\omega t) \quad (1) $$

for $x \geq 0$, with a hard wall at $x = 0$.

Fig. 1 shows a Poincaré surface of section for the classical version of this system with $\lambda = 0.4$ and $\omega = 0.92$, plotted in action-angle variables $(I, \varphi)$ of the undriven well. In the lower left corner, the elliptic island that originates from the primary 1:1 - resonance is visible. This island is organized around a stable (elliptic) periodic orbit which closes on itself after one period $T$ of the external force. In contrast to the chaotic motion of trajectories in the surrounding stochastic sea, motion inside this island is mainly regular. A major fraction of the extended phase space $\{(p, x, t)\}$ surrounding the periodic orbit is filled with invariant $T$-periodic vortex tubes [10], and a trajectory with an initial condition on such a tube remains confined to it for all times $t$. The closed curves inside the islands seen in Fig. 1 are sections of vortex tubes with the plane $t = 0$.

Disregarding small secondary resonances, the main other features of the Poincaré section
are the two large islands which belong to the 2:1 - resonance. In this case, the central periodic orbit closes only after two cycles of the driving field, and the surrounding vortex tubes are 2T-periodic. The appearance of two islands in Fig. 1 is simply a consequence of the fact that the classical flow is sampled once every period, or, expressed differently, that the projection of the 2T-periodic vortex tubes to the fundamental part of phase space \{(p, x, t), 0 \leq t < T\} consists of two disconnected pieces. Thus, “inside” the 2:1 - resonance there is a portion of phase space that sloshes with half the frequency of the external drive.

If the area of these islands is large enough to support quantum states, this subharmonic sloshing should have consequences for the corresponding quantum system. The scale for the classical-quantum comparison is set by Planck’s constant. Since $\hbar = 1$ in our units, the action $I_n = n + 3/4$ (starting from $n = 0$) corresponds semiclassically to the $n$-th quantum state. Thus Fig. 1 indicates that the 2:1 - resonance will strongly influence quantum states between (roughly) $n = 25$ and $n = 50$.

In general, the quantum dynamics of a system with a $T$-periodic Hamiltonian operator $H(p, x, t)$ should be investigated in terms of its Floquet states $u(x, t)$, i.e. the $T$-periodic eigensolutions of the equation

$$\left(H(p, x, t) - i\partial_t\right) u(x, t) = \varepsilon u(x, t) .$$

The Floquet states form a complete set in an extended Hilbert space of square integrable, $T$-periodic functions [11]. Their role in periodic systems is analogous to that of energy eigenstates in time-independent systems. The eigenvalues $\varepsilon$, which are called quasienergies [12,13], are defined up to an integer multiple of $\omega$ and can therefore be arranged in Brillouin zones, with the first zone ranging from $-\omega/2$ to $+\omega/2$.

Quasienergies for the driven triangular well are shown in Fig. 2 as functions of $\lambda$. Only those quasienergies that connect for $\lambda \rightarrow 0$ to the energies of the unperturbed stationary states from $n = 20$ to $n = 50$ have been displayed. The influence of the classical 2:1 - resonance is clearly visible: There are two “fans” of quasienergies that appear to behave almost identically with coupling strength, except that they differ by an amount very close
to $\omega/2 \pmod{\omega}$.

It is important to realize that such a spectral feature is not due to the specific triangular well potential, but solely to the presence of a resonance. Let $\chi_n(x)$ denote the energy eigenfunctions of the undriven well, and $E_n$ their energies. The existence of a classical 2:1 - resonance corresponds, in the quantum system, to an energy spacing which is close to $\omega/2$ in the vicinity of a certain state $n_0$. Thus, we also assume $E'_{n_0} = \omega/2$ (the prime denotes differentiation with respect to quantum number) and expand the wave function as

$$
\psi(x,t) = \sum_n c_n(t) \chi_n(x) \exp\left\{-i(n\omega/2 + E_{n_0})t\right\} .
$$

If only resonant terms are kept, one obtains for the coefficients $c_n(t)$ the equation

$$
i\dot{c}_n = \left(E_n - n\frac{\omega}{2} - E_{n_0}\right)c_n + V_{n,n+2}c_{n+2} + V_{n,n-2}c_{n-2}
$$

with $V_{n,m} = \lambda \langle n|x|m \rangle / 2$. This equation leads to two separate groups of states, since it decouples coefficients with odd from those with even indices. Expanding the energies $E_n$ quadratically around $E_{n_0}$ and assuming $V_{n,n+2} = V_{n,n-2} \equiv V_0$ to be a constant, standard techniques \cite{14,15} can be employed to express the resonant Floquet states in terms of Mathieu functions \cite{16}. For even $n-n_0$, the quasienergies are found to be

$$
\varepsilon_k(q) = E_{n_0} + \frac{1}{2}E''_{n_0}\alpha_k(q) \pmod{\omega}
$$

where $q = 2V_0/E''_{n_0}$, and $\alpha_k(q)$ is a characteristic value of the Mathieu equation that is associated with a $\pi$-periodic Mathieu function, i.e. one of those characteristic values usually denoted by $a_0, b_2, a_2, \ldots$ \cite{14}. If, however, $n-n_0$ is odd, the quasienergies are

$$
\varepsilon_k(q) = E_{n_0} + \frac{1}{2}E''_{n_0}\alpha_k(q) + \frac{1}{2}\omega \pmod{\omega}
$$

where now $\alpha_k(q)$ is one of the characteristic values $b_1, a_3, b_3, \ldots$, belonging to a $2\pi$-periodic Mathieu function. Despite the approximations, these formulae yield a quite good description of the numerically computed spectrum \cite{13}. Most importantly, the known asymptotic
behavior of the characteristic values \[\{16\}\] leads to an analytical estimate for the quasienergies in the strong driving regime. For instance, the difference between the two quasienergies indicated by the arrows in Fig. 2 is given, modulo $\omega$, by

$$\frac{\omega}{2} + \frac{1}{2} E'_{n_0} (b_1 - a_0) \approx \frac{\omega}{2} + E''_{n_0} 2^4 \sqrt{\frac{2}{\pi}} q^{3/4} \exp(-4\sqrt{q}). \quad (7)$$

Since $q$ is proportional to $\lambda$, we find that the deviation from $\omega/2$ becomes exponentially small with the square root of the coupling strength. For $\lambda = 0.4$, we estimate this deviation to be $4 \cdot 10^{-18} \omega$.

A coherent superposition of two Floquet states $u_1(x,t)$ and $u_2(x,t)$ with quasienergies $\varepsilon_1$ and $\varepsilon_2 = \varepsilon_1 + \omega/2$,

$$\psi = A_1 u_1 \exp\{-i\varepsilon_1 t\} + A_2 u_2 \exp\{-i(\varepsilon_1 + \omega/2)t\}, \quad (8)$$

will radiate at half the driving frequency. This occurs because the dipole $\langle \psi | x | \psi \rangle$ is not $T$-periodic — its shortest cycle time is $2T$. A spectrum like that in Fig. 2 thus suggests the possibility of subharmonic generation which, neglecting the exponentially small corrections \[\{7\}\], is independent of the precise value of the driving amplitude $\lambda$. This possibility survives even for pulses with a slowly varying amplitude, to which the wave function \[\{8\}\] can respond adiabatically.

We now interpret these results from a semiclassical point of view. Floquet states and quasienergies can be calculated approximately by means of semiclassical quantization rules which are similar to the Einstein-Brillouin-Keller (EBK) conditions \[\{17\}\]. For driven one-dimensional systems, like the particle in a triangular well, they can be written as

$$\oint_{\gamma_1} p dx = 2\pi \left( n_1 + \frac{1}{2} \right), \quad (9)$$

where the quantization path $\gamma_1$ winds around a $T$-periodic vortex tube in a plane of constant time $t$, and

$$\varepsilon = -\frac{1}{T} \int_{\gamma_2} (p dx - H dt) + n_2 \omega \quad (10)$$
with a \( T \)-periodic path \( \gamma_2 \) that lies on such a tube, and the integration extending over one period. The integer \( n_1 \) is the semiclassical quantum number; \( n_2 \) accounts for the mod \( \omega \) multiplicity of the quasienergies \( \varepsilon \). It is crucial to realize that the application of these rules requires the existence of \( T \)-periodic vortex tubes. They can be applied to vortex tubes that are merely perturbative (and, therefore, \( T \)-periodic) deformations of energy manifolds of the undriven system, or to the tubes of a 1:1 - resonance, but not to any other resonance. The first rule \((9)\) will always allow the selection of quantized vortex tubes and the construction of associated semiclassical wave functions, but the \( T \)-periodic boundary conditions of the second one \((10)\) are incompatible with, e.g., the \( 2T \)-periodic tubes of a 2:1 - resonance, so that these functions are no Floquet states.

Such a contradiction can only be resolved if a Floquet state is not associated with a single quantized \( 2T \)-vortex tube, but with both parts that result from its projection to the fundamental time interval. In this way, \( T \)-periodicity is restored. Because there are two equivalent quantized \( 2T \)-tubes, the Floquet states must appear in pairs, with almost the same probability density for both members of such a pair.

This reasoning is confirmed by a numerical computation of the exact Floquet states. Fig. 3 shows, for \( \lambda = 0.4 \), the probability density of one of the two states that belong to the marked quasienergies in Fig. 2. The density is concentrated along both realizations of the periodic orbit that bounces against the wall at \( x = 0 \) and is reflected. As expected, the density of the corresponding state in the second quasienergy fan is almost the same.

Conceptually, the situation encountered here is strongly reminiscent of a particle in a double well \[15\]. An attempt to calculate the eigenstates in a double well potential by applying the simple Bohr-Sommerfeld quantization separately to the two wells, without accounting for tunneling through the barrier, yields states which are strictly confined to the individual wells. The correct eigenstates appear in doublets that are delocalized over both wells (odd and even combinations). Analogously, the Floquet state shown in Fig. 3 is a member of the ground state doublet of the 2:1 - resonance; the other corresponding states in the two fans constitute the excited doublets. This also explains the \( \omega/2 \) difference in
quasienergies between states in a doublet. After one period $T$, the phases of the odd and even combinations differ by $\pi$.

In an ordinary double well, localized states can be realized by appropriate linear combinations of the eigenstates. In the same way, linear combinations of a Floquet-doublet lead to a density which is concentrated asymmetrically along only one of the two equivalent tubes. Such a density is $2T$-periodic, and that is why a superposition like (8) generates subharmonics. An example of a superposition of the 2:1 ground state doublet is shown in Fig. 4; the corresponding expectation value of the dipole operator is plotted in Fig. 5 for an interval of six periods of the driving force. It is obvious that this expectation value contains a strong subharmonic contribution; a Fourier analysis confirms that the subharmonic mode is by far the dominant one.

If there were no quantum mechanical communication between the two classically isolated vortex tubes, the dipole would be exactly $2T$-periodic. But there is quantum tunneling from one tube through the stochastic sea to its counterpart, and, as a consequence, the two corresponding quasienergies do not differ by exactly $\omega/2$. To complete the double well analogy, the deviation from this value has to be interpreted as the tunnel splitting; in particular, eq. (7) describes the tunnel splitting of the ground state doublet. Its remarkable exponential suppression with increasing driving strength could have important practical consequences.

The picture discussed so far can immediately be generalized to a primary $r:1$ - resonance. In such a case, there are $r$ disconnected $rT$-periodic vortex tubes, and the Floquet states are extended over all of them. Appropriate linear combinations, which localize the probability density along one of these tubes, lead to the generation of subharmonics with frequency $\omega/r$. The approximate analytic construction of the Floquet states then also requires the $r\pi$-periodic Mathieu functions.

From a practical point of view, the mechanism of subharmonic generation outlined in this Letter has several attractive features. It depends only on a property of the driven potential — the existence of a 2:1 - resonance — but neither on the exact potential shape
nor (neglecting the tunneling corrections) on the exact strength of the driving force. In addition, any linear superposition of Floquet states that contains both members of a doublet yields subharmonics. The particular example of a triangular well potential is realized in semiconductor heterojunctions [19], with characteristic energy spacings in the far-infrared regime. Now that harmonic generation in far-infrared driven heterostructures has been reported [20], it is both an experimental and a theoretical challenge to explore whether our mechanism of subharmonic generation can be exploited in such mesoscopic devices.

We thank S.J. Allen and M.S. Sherwin for discussions. M.E.F. was supported by the National Science Foundation under Grant No. PHY89-04035. M.H. acknowledges support from ONR grant N00014-92-J-1415 and the Alexander von Humboldt-Stiftung.
REFERENCES

[1] A. L’Huillier and P. Balcou, Phys. Rev. Lett. 70, 774 (1993).

[2] M.C. Gutzwiller, *Chaos in Classical and Quantum Mechanics* (Springer, New York, 1991).

[3] M.V. Berry and J.P. Keating, J. Phys. A 23, 4839 (1990).

[4] G. Tanner, P. Scherer, E.B. Bogomolny, B. Eckhardt, and D. Wintgen, Phys. Rev. Lett. 67, 2410 (1991).

[5] R. Aurich, C. Matthies, M. Sieber, and F. Steiner, Phys. Rev. Lett. 68, 1629 (1992).

[6] M.A. Sepúlveda, S. Tomsovic, and E.J. Heller, Phys. Rev. Lett. 69, 402 (1992).

[7] O. Bohigas, S. Tomsovic, and D. Ullmo, Phys. Rep. 223, 43 (1993).

[8] E. Shimshoni and U. Smilansky, Nonlinearity 1, 435 (1988).

[9] F. Benvenuto, G. Casati, I. Guarneri, and D.L. Shepelyansky, Z. Phys. B 84, 159 (1991).

[10] V.I. Arnold, *Mathematical Methods of Classical Mechanics* (Springer, New York, 1978).

[11] H. Sambé, Phys. Rev. A 7, 2203 (1973).

[12] Ya.B. Zel’dovich, Zh. Eksp. Teor. Fiz. 51, 1492 (1966) [Sov. Phys. JETP 24, 1006 (1967)].

[13] V.I. Ritus, Zh. Eksp. Teor. Fiz. 51, 1544 (1966) [Sov. Phys. JETP 24, 1041 (1967)].

[14] G.P. Berman and G.M. Zaslavsky, Phys. Lett. A 61, 295 (1977).

[15] M. Holthaus and M.E. Flatté, to be published.

[16] M. Abramowitz and I.A. Stegun (eds.), *Handbook of Mathematical Functions* (Dover, New York, 1972).

[17] H.P. Breuer and M. Holthaus, Ann. Phys. (N.Y.) 211, 249 (1991).
[18] F. Bensch, H.J. Korsch, B. Mirbach, and N. Ben-Tal, J. Phys. A 25, 6761 (1992).

[19] See, e.g., G. Bastard, Wave Mechanics Applied to Semiconductor Heterostructures (Les Editions de Physique, Les Ulis, 1988).

[20] W.W. Bewley, C.L. Felix, J.J. Plombon, M.S. Sherwin, M. Sundaram, P.F. Hopkins, and A.C. Gossard (submitted to Phys. Rev. B).
FIGURES

FIG. 1. Poincaré surface of section for the driven triangular well (1) for $\lambda = 0.4$ and $\omega = 0.92$, taken at $t = 0 \mod T$.

FIG. 2. Quasienergies for the driven triangular well with $\omega = 0.92$, as functions of $\lambda$. The arrows on the right margin indicate the ground state doublet of the 2:1-resonance.

FIG. 3. Probability density of a member of the ground state doublet of the 2:1-resonance, for two periods of the external force. Lines connect points of equal density.

FIG. 4. Probability density of a solution of the time-dependent Schrödinger equation that consists of a superposition of both members of the ground state doublet.

FIG. 5. Expectation value of the dipole operator for the wave function shown in Fig. 4, for $6T$. 