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Abstract: The ionization of hydrogen can be treated by classical theory when the initial quantum number is large, and the photon energy is small. Classically, the electron motion is stochastic for high intensities and the resulting diffusion can lead to ionization. However, Casati et al. (Phys. Rev. Lett. **57**, 823 (1986)) have found that the ionization threshold is often higher than the threshold for classical stochasticity. We present here a heuristic explanation: classical stochasticity will be suppressed when the phase space area escaping through classical cantori each period of the electric field, is small compared to Planck's constant. We obtain a scaling law which agrees remarkably well with the numerical results of Casati et al.

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Introduction

A quantum perturbation calculation of multi-photon ionization rates of excited atoms is impractical when the number of photons involved is large. Only recently have numerical calculations become possible for the simplest models.[1] For this reason semi-classical or classical theories which are simpler to implement numerically, and which lead to simple analytical predictions, are of great value.[2,3] Furthermore, some predictions based on classical theory have been found to agree well with experiments.[3]

In this paper we study the one-dimensional model for the ionization process given by the Hamiltonian

$$H(p,x,t) = {}^{p^2}/_{2} - {}^{1}/_{x} + \varepsilon\, x \cos(\omega t)\,, \qquad x>0 \tag{1}$$

This model has obtained great success[3] in comparison with experiments,[4] basically because electronic states highly elongated along the field direction are the easiest to ionize, and these states are only weakly coupled to those corresponding to the other dimensions. Here ε is the electromagnetic field strength and ω its frequency, in atomic units.

It is easier to treat bounded motion in this model using the coordinates (I,θ) which are the action-angle variables for $\varepsilon=0$ (Ref. 3):

$$H(I,\theta,t) = +{}^{1}/_{2I^2} - \varepsilon I^2 \sum_{m=-\infty}^{\infty} \kappa(m)\, m^{-5/3} \cos(m\theta-\omega t)$$

$$\kappa(m) = m^{2/3} J_m'(m) \tag{2}$$

The function κ actually varies little with m: $\kappa(1) \approx 0.322$, $\kappa(2) \approx 0.355$, and $\kappa \to ({}^{4}/_{3})^{1/3}/\Gamma({}^{1}/_{3}) \approx 0.41$ as $m \to \infty$. We have changed the sign of H to conform with the usual convention of positive kinetic energy.

We first discuss the classical behaviour of this system. For $\varepsilon = 0$, the system is integrable and every orbit lies on an invariant torus $I =$ constant (thinking of t modulo $2\pi/\omega$, with $\theta(t) = \theta_0 + t/I^3 \bmod 2\pi$). For $\varepsilon > 0$, there is an $I_c(\varepsilon)$ such that for $I < I_c$ the motion is dominated by invariant tori which are deformations of the tori $I =$ constant, while for

$I > I_c$ there are no such invariant tori and many orbits are free to increase arbitrarily in I.[5,6] We say the atom ionises if I gets to infinity. There is a highest invariant torus which makes ionisation impossible for all initial conditions below it, while a large fraction of initial conditions above it will ionise. I_c decreases as ε increases, so we can turn this round to say that for initial action I there is a threshold field strength $\varepsilon_c(I)$ below which ionisation is impossible and above which it may be possible.

Numerical simulations of the quantum system can show significant deviations from the classical threshold, however.[1,7] In this paper we propose a heuristic explanation based on the concepts of partial barriers and turnstiles.[8,9] We predict an elevation in the quantum threshold for ionization above the classical one, or more precisely a threshold above which quantum effects should not be important and classical diffusion can lead to ionization. Our prediction is that significant ionisation will occur if and only if the quantities ΔW given in equations (14) and (7) both exceed 1.

$\Delta W > \hbar$ Criterion

First we review the results of Refs. 8-9 about partial barriers and turnstiles. Orbits of a $1^1/_2$ degree of freedom Hamiltonian system can be obtained from the principle of stationary action

$$W = \int I d\theta - H(I,\theta,t)dt \equiv \int L(\theta,\dot{\theta},t)dt \ , \qquad (3)$$

where L is the Lagrangian, $\dot{\theta} = \partial H/\partial I$, and we assume $\partial^2 H/\partial I^2 > 0$. An orbit $[I(t),\theta(t)]$ of H corresponds to a path $\theta(t)$ for which for all t_1 and t_2, W is stationary with respect to all variations of $\theta(t)$ fixing $\theta(t_1)$ and $\theta(t_2)$. Periodic or quasi-periodic orbits can be labelled by a winding ratio ν, which is the ratio of the forcing frequency to the orbit frequency. Periodic orbits with winding ratio p/q correspond to paths satisfying $\theta(t) = \theta(t+2\pi q/\omega) - 2\pi p$ for which W, integrated over one period, is stationary with respect to variations in the class of periodic paths.

For each p/q there are at least two periodic orbits. One can be

obtained as a minimum of W. Given such a minimum, $\theta(t)$, then $\theta(t+2\pi k/\omega) - 2\pi m$ is another minimum for any integers k and m, though it corresponds to the same orbit phase-shifted. Between two such minima, W must have another critical point called a minimax (generically a saddle). $\Delta W_{p/q}$ is defined as the difference in action between the minimax and minimizing orbits. Generically the minimizing orbit is regular hyperbolic and the minimax orbit is either elliptic or inversion hyperbolic.

Aubry[10] and Mather[11] prove that there exist minimizing orbits for each irrational winding number too, forming either invariant tori, or invariant Cantor sets called "cantori." In the latter case there is a minimax orbit in each orbit of gaps of the cantorus.[12] ΔW_ν is the maximum of the differences in action for minimax orbits in the gaps of the cantorus.

The major barriers to transport in classical Hamiltonian systems appear to be the cantori. A "partial barrier" can be constructed from a cantorus, and the only way orbits can cross the barrier is by leaking through its "turnstile". The important quantity governing the rate of crossing is the turnstile flux which is the phase space area crossing the partial barrier in one forcing period. This flux is equal to the sum of the ΔW_ν for each family of gaps in the cantorus (typically there is only one family). Thus, the most important cantori are the ones with smallest ΔW.

We propose, as we suggested in Ref. 8, that classical predictions of transport will be reasonably good when $\Delta W \gg \hbar$ for all cantori, but quantum effects will suppress transport to the level of quantum tunnelling when $\Delta W \leq \hbar$. Heuristically, one knows that the minimum spreading of a wavepacket in phase space is of order $\hbar$, and that quantum mechanics ignores structure on finer scales than this. Semi-classical wavefunctions are associated with classical tori whose action is an integer multiple of $\hbar$ (with a half integer Maslov index correction). When the classical tori are destroyed we know of no theory for determining semi-classical eigenstates; however, if the holes in the classical tori are of a scale smaller than $\hbar$, then they should be quantally undetectable. The appropriate measure of the size of holes in a cantorus is the flux ΔW; it has the units of phase space area, as does $\hbar$.

This idea is supported by the computations of Brown and Wyatt[13] for a model similar to (1). They show that a wave packet initially localized inside of a cantorus of flux 1.1$\hbar$, remains localized for at least several times the classical escape time. Similarly Radons, Geisel, and Rubner[14] show that cantori of the standard map with $\Delta W < \hbar$ can be as effective barriers as tori.

Estimating ΔW_ν

One could evaluate the desired ΔW's numerically, and this is the only accurate approach in general; however, quite good predictions of ΔW's can be obtained analytically. There are two tractable cases in which we can do this.

When tori have just broken up, the ΔW's of the cantori can be obtained from those of nearby periodic orbits using a scaling law based on renormalisation. In this case properties of the resonances can be measured numerically if desired, but also can often be estimated simply by first order perturbation theory. This applies when the residues of the periodic orbits are not too large. The residue of a periodic orbit is $R=(2-\lambda-1/\lambda)/4$, where $\lambda, 1/\lambda$ are the multipliers of the orbit. When the orbit is linearly stable,

$$R = \sin^2(\Omega T/2) \quad ,$$

where T is the period of the orbit and Ω is the frequency of small oscillations about it.[6]

The second tractable limit is near the separatrix between trapped and unbounded motion. Here the ΔW's of all orbits are much the same, and can be estimated by the flux escaping through the separatrix itself. This can be perturbatively calculated using the Melnikov technique.

ΔW Near Breakup

Near break-up of invariant tori, numerical evidence[15,16] and an approximate renormalisation theory[17,18] suggest that the cantorus with the smallest ΔW in any range of frequency always has a noble frequency, i.e. of the form $(\gamma p + p')/(\gamma q + q')$, with $\gamma = (1 + \sqrt{5})/2$ and $pq' - p'q = \pm 1$. Indeed, out of all frequencies between p/q and p'/q', ΔW usually is smallest at this noble or the analogous one with p/q and p'/q' interchanged.

Renormalisation theory suggests that the torus of winding ratio $\nu = (\gamma p+p')/(\gamma q+q')$ breaks when the residues of the p/q and p'/q' resonances

(or more accurately a weighted combination of them) pass through about $R^* \approx 0.25008$. Furthermore, a scaling law can be obtained for the cantorus flux ΔW_ν, when $R_{p/q} > R^*$:

$$\Delta W_\nu \approx C \Delta W_{p/q}(R^*) \, (\log R_{p/q}/R^*)^\zeta$$

$$\zeta \approx 3.0117 \, , \quad C \approx 0.37 \qquad (4)$$

Here $\Delta W_{p/q}(R^*)$ is the action difference between the minimax and minimising p/q orbits, evaluated at the critical parameter for the breakup of the torus.

This scaling law is expected to be valid for any system provided $R_{p/q}$ is close enough to R^*, and is derived as follows. There appears to be a universal one-parameter family T^*_μ which governs the breakup of noble tori.[19] For this family, $\log R_{p/q}$ appears to depend almost linearly on parameter μ. For each factor of $\delta \approx 8.72$ in μ, ΔW_ν grows by a factor of $\alpha\beta \approx 4.337$. This gives rise to the power law with exponent $\zeta = \log \alpha\beta / \log \delta$. The constant C was determined by the observation that for T^*_μ, $\Delta W_\nu(R_{p/q} = 1)$ is within 1% of $\Delta W_{p/q}(R^*)$ (figures 4.5.1.7, 4.5.1.8 of Ref. 16). This also agrees well with the value obtained for the p/1 resonances of the standard map.[8]

If the residues of the p/q and p'/q' resonances are not of the same order then one can include the effect of both, e.g. by using a weighted average $[1/\gamma \log R_{p/q} + 1/\gamma^2 \log R_{p'/q'}]$ instead of $\log R_{p/q}$. This can be derived from an approximate renormalisation theory.[18]

ΔW near the Separatrix

A separatrix divides phase space into two regions, trapped and untrapped motion; it is formed from the stable and unstable manifolds of a hyperbolic periodic orbit. In an integrable system these two manifolds join smoothly, but typically they intersect transversally and the resulting complicated oscillations lie at the root of stochasticity.

The orbits of points where the two manifolds cross are homoclinic

to the periodic orbit; there is a minimizing, M, and a minimax, S, homoclinic orbit. As before, the flux between this orbit pair is given by their difference in action:[9]

$$\Delta W_S = \int \left(I d\theta - H(I,\theta,t) dt \right) \Big|_M^S \tag{5}$$

By Stokes' theorem this can be written as an integral over any surface, σ, in the three dimensional space (I,θ,t) that connects the two orbits (thus, σ is a ribbon):

$$\Delta W_S = \int_\sigma \left[dI \wedge d\theta - dH \wedge dt \right] \tag{6}$$

To obtain an approximate formula for ΔW, let $H = H_0(I,\theta) + H_1(I,\theta,t)$, and take as an approximate surface, σ_0, the unperturbed separatrix which is a surface of constant H_0, cross the time interval $[0,T]$. The separatrix can be parameterized by a time t_0, the time along the unperturbed flow. Let $z_S(t_0) = (I,\theta)(t_0)$ be the separatrix point reached after following the unperturbed flow $dI/dt_0 = -\partial H_0/\partial\theta$, $d\theta/dt_0 = \partial H_0/\partial I$ for a time t_0 from some reference point on the separatrix. The surface σ_0 is swept out by $z_S[t_0+t]$ as t ranges from $-\infty$ to ∞ and t_0 from 0 to T. The first term in Eq. (6) vanishes because the unperturbed flow lies in the surface σ_0, while the second becomes

$$dH \wedge dt = \left(\frac{dH}{dI}\frac{dI}{dt_0} + \frac{dH}{d\theta}\frac{d\theta}{dt_0} \right) dt_0 \wedge dt$$

$$= -\{ H_1(z_S(t_0+t),t),\, H_0(z_S(t_0+t)) \}\, dt_0 \wedge dt \tag{7}$$

where $\{\,,\,\}$ is the Poisson Bracket. To obtain the flux ΔW, first integrate along the ribbon σ_0 by letting t range from $-\infty$ to ∞, obtaining

$$M(t_0) \equiv \int_{-\infty}^{\infty} dt\ \{H_1,H_0\}(z_s(t_0+t),t) \tag{8}$$

Here M(t) is called the Melnikov function;[20] it is periodic with period T. Then the outward flux across the unperturbed separatrix in one forcing period is

$$\int_0^T dt\ \max(0,\ M(t)) \quad , \tag{9}$$

and can be easily computed for model systems. Note that this is an exact formula for the flux across the unperturbed separatrix, but ΔW for the actual homoclinic orbits may be slightly different.

Mather[12] proved in the case of twist maps that ΔW is continuous as one approaches a separatrix. Thus Eq. (9) can be used to estimate the flux through cantori near the separatrix.

Application to Ionisation

Now we apply these ideas to the ionisation problem, first computing ΔW near breakup.

For $\varepsilon=0$, orbits with action I have winding ratio $I^3\omega$. The most important resonances are the primary ones with winding ratio m = 1,2,3... The m^{th} resonance term in H is important when $m\dot{\theta} \approx \omega$, i.e. for $I \approx I_m \equiv (m/\omega)^{1/3}$. For $I_1 \leq I << \varepsilon^{-1/4}$, the most important cantori are expected to be those with rotation numbers $m + 1/\gamma^2$ and $m + 1/\gamma$. Their ΔW's can be estimated from the properties of the resonances of winding numbers m and m+1, which can easily be evaluated by first order perturbation theory. For $I < I_1$, there are no primary resonances and higher order perturbation theory or numerical evaluation would be required. So we will restrict attention to $I \geq I_1$. Unfortunately, most experiments, real and numerical, only just get into this regime, but there are sufficient comparisons that can be made.

The lowest order approximation for motion in and near to the m^{th} resonance is obtained by neglecting all other terms in the sum but the m^{th}. Expanding around I_m and performing a canonical transformation to new variables

$$P=(I-I_m)/m,\ Q=m\theta-\omega t$$

gives the pendulum Hamiltonian

$$K(P,Q) = {}^3/_2\ (\omega^2 m)^{2/3}\ P^2 - \varepsilon\kappa/(m\omega^{2/3})\cos Q \qquad (10)$$

The periodic orbits of the m^{th} resonance correspond to the equilibria of K. They have period $T = 2\pi m/\omega$. For the stable one, nearby orbits oscillate with the frequency $\Omega^2 = 3\varepsilon\kappa(\omega^2/m)^{1/3}$. For ε small this gives the residue

$$R_m \approx 3\pi^2\kappa\varepsilon\ (m^5/\omega^4)^{1/3} \qquad (11)$$

Hence the $m \pm 1/\gamma^2$ tori break when

$$\varepsilon > \varepsilon_c \equiv (R^*/3\pi^2\kappa)(\omega^4/m^5)^{1/3} \tag{12}$$

This is essentially the same result as predicted by the resonance overlap criterion.[3]

In order to estimate ΔW_ν for the cantorus we need $\Delta W_m(R^*)$. This can be easily obtained to first order in ε. Using the (P,Q) coordinates to evaluate W by Eq. (3), there is no contribution from the first term since the stable and unstable orbits both occur at P=0. However, their energies differ by twice the height of the potential, thus $\Delta W_m = T\, 2\varepsilon\kappa/(m\omega^{2/3})$. Evaluating this at $R_m = R^*$ yields

$$\Delta W_m(R^*) = 4R^*/[3\pi(\omega m^5)^{1/3}] \tag{13}$$

Thus we predict that ΔW_ν for $\nu = m \pm 1/\gamma^2$ is

$$\Delta W_\nu = \frac{4CR^*}{3\pi(\omega m^5)^{1/3}}\left[\log\left\{\frac{3\pi^2\kappa\varepsilon}{R^*}\left(\frac{m^5}{\omega^4}\right)^{1/3}\right\}\right]^{\zeta}$$

$$\approx \frac{0.039}{(\omega m^5)^{1/3}}\left[\log\left\{49\varepsilon\left(\frac{m^5}{\omega^4}\right)^{1/3}\right\}\right]^{3.0} \tag{14}$$

provided R is of order R^*.

For the case $I > \varepsilon^{-1/4}$, the residues are large, and the ΔW's are best approximated by the flux through the separatrix. For this calculation it is most convenient to use the (p,x) coordinates of the original Hamiltonian (1). There are two branches to the $\varepsilon = 0$ separatrix, which meet at ($p=0$, $x\to\infty$):

$$p_s(t_0) = \pm\left[{}^3/_4\,|\,t_0\,|\,\right]^{-1/3} \quad , \tag{15}$$

for $t_0 > 0$ or $t_0 < 0$, respectively. The Melnikov function, Eq. (7), has a contribution from each branch

$$M(t_0) = 2\varepsilon\int_{-t_0}^{\infty} p_s(t_0+t)\cos\omega t\, dt \quad . \tag{16}$$

Evaluating this, and substituting the result into Eq. (7) gives

$$\Delta W_S = 8\pi\varepsilon\kappa(\infty)/(\sqrt{3}\,\omega^{5/3})$$

$$= 5.9\,\varepsilon\omega^{-5/3} \qquad (17)$$

Equations (14) and (17) for ΔW are sketched in Fig. 1 for fixed ω and ε, as a function of m. The separatrix result applies in the limit $m\to\infty$, and in the figure we assume that ΔW_γ smoothly approaches this result.

The criterion $\Delta W > \hbar = 1$, can be applied to our problem to obtain two conditions on the field strength for significant ionisation. First, the field strength must be large enough (or the frequency small enough) so that the separatrix flux, Eq.(17), is larger than $\hbar$. Secondly, the electron must be initially in a state for which Eq.(14) is larger than $\hbar$. This will occur near the classical threshold unless the coefficient of the logarithm in Eq.(14) is small compared to $\hbar$.

In the experiments the atoms are initially put in an eigenstate of the unperturbed system with $I = (m/\omega)^{1/3} = n\hbar$. Thus the threshold $\Delta W = \hbar$ will be significantly elevated above the classical one when the winding ratio

$$\omega n^3 \geq \sqrt{n/25} \qquad (18)$$

The simple scaling law (14) requires $\omega n^3 \geq 1$, as we consider only the primary resonances m = 1,2,..., and the major cantori between them. Of course ΔW could be computed numerically for smaller frequencies. Unfortunately, experiments so far have been limited to $\omega n^3 < 1.1$, so we can not compare the results with our theory. However, there are two numerical experiments of Casati et al.[1,7] with which we can make a comparison.

(i) In Ref. 1 the probability of ionisation is calculated for a wave packet initially localized at $n_0 = 66$, in a field with $n_0^4\varepsilon = 0.05$. Apart from single photon ionisation at high frequency, they find significant ionisation only in the frequency range $0.4 < n_0^3\omega < 2$. For this field strength, eq. (14) predicts that the turnstiles for $I > n_0$ are all bigger than

$\hbar$ iff $0.3 < n_0^3\omega < 1.6$ (remember, we choose frequencies of the form $m \pm 1/\gamma^2$ since these are the most important cantori). So indeed we find just an interval of frequencies over which ionisation is likely to be significant, which matches fairly well with the observations, though it is slightly pessimistic. Actually our predicted lower limit should not be relied upon, as it has $m < 1$, where we did not calculate ΔW.

(ii) In ref. 7 a wave packet initially localized at $n_0 = 66$ is allowed to evolve in a field with $n_0^4\varepsilon = 0.03$ and $n_0^3\omega = 1.2$. The distribution function is observed to spread to the range $60 < n < 75$, and then to leak out from this range via a tail with $n > 75$. The $m=1$ resonance occupies the range over which the initial spreading occurs, and cantori below the resonance have very small ΔW's: Eq. (14) (though not reliable for $n^3\omega<1$) predicts that $\Delta W \approx 0$ for $n < 60$. Moving up from the $m=1$ resonance, the first significant cantorus has frequency γ, and occurs at $n \approx 72$. Equation (6) predicts that $\Delta W_\gamma \approx \hbar$ and so this will be an effective barrier. However $\Delta W > \hbar$ for $n > 74$. Thus any part of the wavepacket that escapes across γ can be expected to classically diffuse, as observed.

Casati et al. propose an alternative explanation in terms of a "quantum delocalisation threshold"

$$\varepsilon_q = 0.41\omega^{7/6}/n$$

but its predictions do not seem to agree so well with the numerical results. In case (i) it predicts an ionisation range of $n_0^3\omega < 0.99$, and in case (ii) it predicts a localisation range of $n < 137$. Since this quantum delocalisation threshold and ours have different functional dependence, they can not both be right. Experiments at larger values of ωn^3, either numerical or physical, might reveal which, if either, is right.

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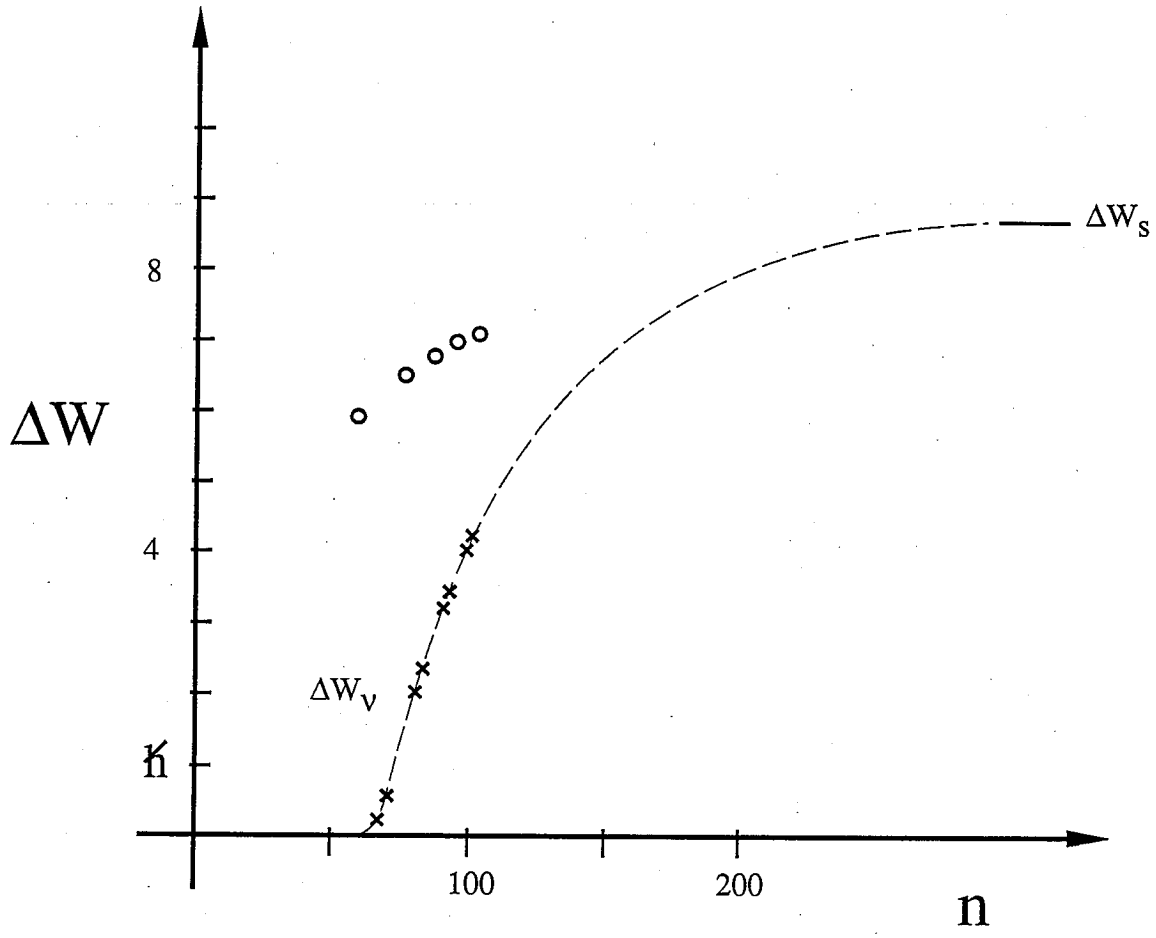
Figure

Sketch of ΔW as a function of the action $I = n\hbar$ for fixed $\omega = 4.2\times10^{-6}$ and $\varepsilon = 1.6\times10^{-9}$, corresponding to example (ii). The circles represent ΔW_m for $m=1,2,...5$, and the crosses represent ΔW_ν, Eq. (14), for $\nu = m\pm\sigma^{-2}$, the most resistant cantori. Eq. (14) is valid for $n\hbar < \varepsilon^{-1/4} \approx 160$ or R_m/R^* not too large ($R_m/R^* \approx 20$ at $n = 130$). Both ΔW_m and ΔW_ν asymptote to ΔW_s, Eq. (17), for $n\hbar >> 160$.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