

LETTER TO THE EDITOR

Stabilisation approach to the quasi-Landau spectrum of the magnetised hydrogen atom

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Abstract. The stabilisation method is used to obtain quantum mechanically positions and widths of the quasi-Landau periodic-orbit-based resonances for the hydrogen atom in a strong uniform magnetic field. Diagonalisation of only small matrices is required despite the irregular character of the system studied. Characteristic properties of periodic-orbit-based resonances are discussed.

One of the most intriguing problems in the study of irregular systems is the relation between classical unstable periodic orbits (PO) and regularities in the very complex, congested spectra. These regularities appear as pronounced peaks in the Fourier transform (FT) of, at first glance, random spectra, as exemplified by the photoabsorption spectrum of the hydrogen atom in a strong uniform magnetic field B close to the ionisation limit (quadratic Zeeman effect—to be referred to as QZE) (Holle *et al* 1986, Main *et al* 1986). The positions of the peaks in the FT time domain spectra were shown to be strongly correlated (Main *et al* 1986, Wintgen 1987, Wintgen and Friedrich 1987) with the periods of the unstable PO embedded in the predominantly chaotic motion of the system near the ionisation limit ($E = 0$). The spectra themselves, on the other hand, show the Wigner-type nearest-neighbour level spacings distribution (Delande and Gay 1986, Wintgen and Friedrich 1986, Wunner *et al* 1986) postulated for systems that exhibit 'quantum chaos' (Bohigas *et al* 1984).

Correlations between quantum spectra and classical PO have been a subject of extensive study within the so-called semiclassical PO theory (Gutzwiller 1971, Balian and Bloch 1974, Berry and Tabor 1976). This theory predicts that the density of states has maxima at energies corresponding to the quantised values of actions calculated along PO. Du and Delos (1987, 1988) used this theory (with necessary, important modifications) to successfully simulate semiclassically the FT of the experimental QZE spectra.

The understanding of the role played by classical PO in the quantum spectra in the correspondence regime (as is the case for QZE) may be twofold. Utilising the time-dependent formalism (Heller 1978a, b), Reinhardt (1983) has shown that wavepacket propagation along the PO leads to time recurrences (with the PO period) in the dipole moment correlation function, which gives the FT of the quantum spectrum. Reinhardt

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applied this method to the famous quasi-Landau oscillations present in low-resolution spectra (Garton and Tomkins 1969) and showed them to be related to the period of the ρ_0 perpendicular to the B field. Previously (Starace 1973) the energy spacings of the quasi-Landau oscillations had been obtained semiclassically by quantisation of the action along this short-period ρ_0 . However, the absolute positions of the resonances obtained in this way are uncertain (Delande *et al* 1984).

Recently we have developed a fully quantum mechanical time independent picture of the phenomenon (Taylor *et al* 1988, Taylor and Zakrzewski 1988) based on resonance scattering theory. In short, regions of space near the ρ_0 are approximately dynamically decoupled from the generally chaotic remaining part of the phase space. In the correspondence regime a similar approximate dynamical decoupling of this region is valid for the quantum system. This allows us, in the spirit of the Feshbach partitioning theory (Feshbach 1962) to separate the Hilbert space into the part which in the position representation is localised in the vicinity of the ρ_0 (say P subspace) and the remaining part (Q), with P and Q being only weakly coupled. A ladder of ρ_0 -based resonances will be shown below to exist and will have large wavefunction amplitudes in the region spanned by P space (over the ρ_0). A small diagonalisation in the P space will give the ladder of resonances localised along the given ρ_0 .

Strictly speaking resonances in the pure sense may exist only in the continuum part of the spectrum. The high density of states just below the ionisation limit allows us to extend the same formalism below the B -field-shifted ionisation threshold. 'Resonances' obtained in the bounded part of the spectrum are to be understood as major configurations which in full diagonalisation mix with many others (which describe the off- ρ_0 chaotic part of the wavefunction) to create wavefunctions which are scarred (Heller 1984) in the ρ_0 region and have 'chaotic' random nodal patterns of low amplitude off the ρ_0 . The 'resonance' energy and its width define the energy interval where the exact wavefunctions resemble the resonance wavefunction over the region above the ρ_0 . Placing localised Gaussian basis sets along different ρ_0 to simulate the corresponding P subspaces and diagonalising the Hamiltonian we were able (Taylor and Zakrzewski 1988) to obtain quantum mechanically the peaks in the FT of the resulting eigenvalues in agreement with experiment (Main *et al* 1986) and big full-scale diagonalisations (Wintgen and Friedrich 1987).

The aim of the present letter is to show that the previously developed resonance formalism (Taylor *et al* 1988, Taylor and Zakrzewski 1988) is capable of reproducing quantum mechanically not only the level splittings (and FT peaks) but also the absolute positions of the resonance energy levels. This allows us to predict theoretically the positions of quasi-Landau oscillations in the low-resolution spectra and give for the first time an estimate of the resonance widths.

The Schrödinger equation for a hydrogen atom in a uniform magnetic field B in cylindrical coordinates (au) is

$$\left(\frac{1}{2}(p_\rho^2 + p_z^2) + \frac{m^2}{2\rho^2} + \frac{1}{8}\gamma^2\rho^2 - \frac{1}{r} \right) \psi = E\psi \quad (1)$$

where $r = (\rho^2 + z^2)^{1/2}$, γ is the magnetic field strength in au ($\gamma = B/B_0$, $B_0 = 2.35054 \times 10^5$ T) and m is the conserved z component of the angular momentum parallel to the field. Equation (1) may be expressed in semiparabolic coordinates $u = \sqrt{(r+z)}$, $v = \sqrt{(r-z)}$, as

$$[T_u + T_v - E(V_u^{\text{HO}} + V_v^{\text{HO}}) + \frac{1}{8}\gamma^2(V_u^{\text{NL}} + V_v^{\text{NL}}) - 2]\psi(u, v) = 0 \quad (2)$$

where

$$T_u = \frac{1}{2} \left(\frac{1}{u} \frac{\partial}{\partial u} u \frac{\partial}{\partial u} + \frac{m^2}{u^2} \right) \quad (3a)$$

$$V_u^{\text{HO}} = u^2 \quad (3b)$$

$$V_u^{\text{NL}} = u^4 v^2 \quad (3c)$$

and operators with subscript v are obtained by interchanging u and v . In the following we restrict ourselves to the $m = 0$ case only.

To obtain the resonance energies responsible for the low-resolution quasi-Landau feature we use the popular stabilisation method (Hazi and Taylor 1970). The method has been frequently described in detail (Tucker and Truhlar 1987 and references therein); in short, localised basis sets with a variable parameter are used for diagonalisation of the Schrödinger equation. By producing a stabilisation graph (i.e. the plot of eigenvalues as a function of the basis parameter) the resonance energies are found as the most stable eigenvalues. In the present calculation we use a basis constructed from the Gaussian functions:

$$\Phi(u, v) = A(\alpha, u_0, v_0) \exp[-\alpha(u - u_0)^2] \exp[-\alpha(v - v_0)^2] \quad (4)$$

where $A(\cdot, \cdot, \cdot)$ is a normalisation constant. This choice is motivated by the facts that the Gaussian basis functions can be easily centred on the desired region of space and that matrix elements needed can be easily evaluated (Taylor and Zakrzewski 1988). In equation (4) α (inversely proportional to the square of the Gaussians' width) is a variable parameter. In the calculations reported here we use five rows of Gaussians, one on top of the ρ_0 perpendicular to the B field (i.e. the one shown semiclassically to be responsible for low-resolution features) and two rows on each side of it with about 60 Gaussians per row. The Hamiltonian (1) is symmetric with respect to the z variable (and (2) is invariant under the interchange of u and v). This allows us to treat the even and odd z parity cases separately after constructing the appropriate symmetry-constrained basis functions from (4). Several diagonalisations of small (at most 200×200) matrices yield the stabilisation graphs, parts of which are presented in figures 1(a) and 1(b) for even and odd z parity correspondingly. As the experimental

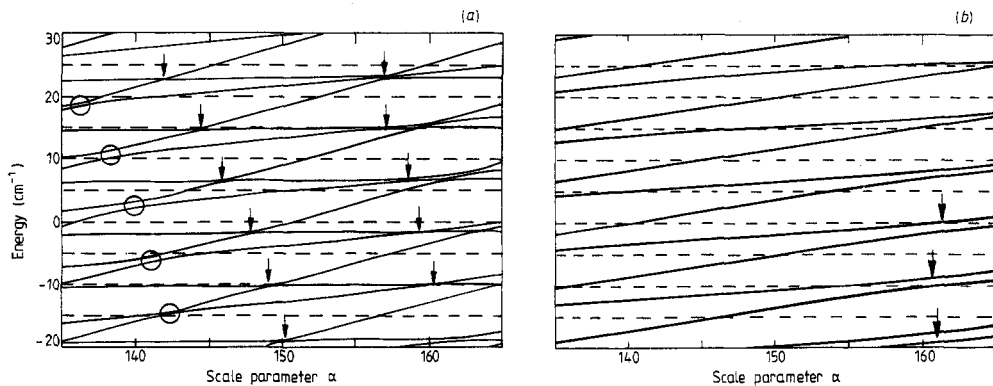


Figure 1. Stabilisation graphs for the QZE around the ionisation limit for even (a) and odd (b) z parity. Arrows in (a) indicate positions of avoided crossings from which the stable roots may be visually traced. Horizontal grid lines help us to see the slope of the stable roots. Circles indicate avoided crossings between 'excited' even resonance states. Indicated values of α are in au multiplied by a factor $\gamma^{-4/3}$, where $\gamma = 2.5526 \times 10^{-5}$.

spectrum for the $m = 0$ case has been obtained for even z parity (Holle *et al* 1986) let us discuss this case first.

Note the distinct ladder of almost equally spaced, almost horizontal lines (these will give the stable roots corresponding to the resonance energies) in figure 1(a). These lines go through avoided crossings for some values of α . That results from the fact that for different α the resonance may 'belong' to a different eigenvalue. From these avoided crossings the width of the resonances will be extracted (see below). Accurate determination of the resonance energies directly from the graph is impossible because a close inspection of the 'stable' eigenvalue lines reveals that they are slightly tilted upwards as a function of α . This behaviour is easily understood by realising that by placing the Gaussians only in the vicinity of the ρ_0 we produce an effective energy-independent well in the direction perpendicular to the ρ_0 . Therefore the energies obtained are shifted upwards by the zero-point energy of the motion perpendicular to the ρ_0 . The size of the well is determined by α and is independent of energy. Recall from elementary quantum mechanics that the zero-point energy in the square-well potential is inversely proportional to the square of the well's width; thus in our case it is proportional to α (compare equation (4)). Therefore for each stable root the relation

$$E_i(\alpha) = C\alpha + E_i \quad (5)$$

should hold. In (5) E_i are the correct unshifted resonance energies and C is a constant characterising the well and independent of the particular resonance energy level. Utilising (5) the extraction of the resonance energies becomes simple. For each stable root we take the energy values as a function of α (omitting points close to the avoided crossings where (5) is obviously invalid) and fit them to the straight line obtaining slope coefficients C_i and resonance energies E_i . According to (5) the slopes should be i -independent; we find that they differ for different roots by at most 5%. This small spread of C_i values around the average provides the internal consistency test of the validity of (5). Using the average slope C the stabilisation graph may be rotated and shifted downwards to cancel the influence of the perpendicular well; the resulting graph (for the even case) is presented in figure 2.

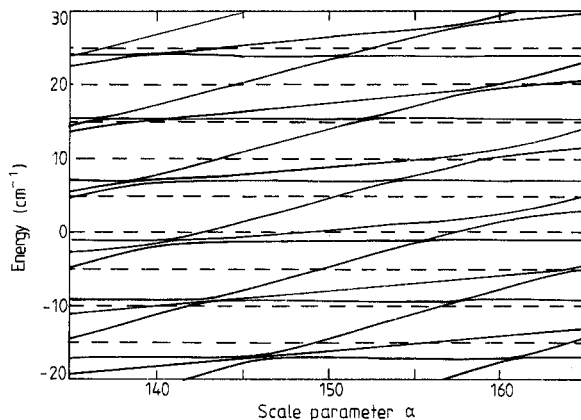


Figure 2. Even z -parity graph but with the zero-point energy of the perpendicular motion extracted using the procedure described in the text. Note the almost perfectly horizontal direction of the stable roots. Indicated values of α are in au multiplied by the $\gamma^{-4/3}$ factor.

It is worth stressing that the standard stabilisation procedure does not employ (5). Instead the basis is expanded in the desired direction (in our case perpendicular to the ρ_0) until the roots are fully independent of the basis parameter. This approach fails for the QZE as the presence of other ρ_0 disturbs the stabilisation graph when the basis is too large. The scheme devised, based on (5), is a novel twist of the method; its applicability to other problems is being investigated.

The ρ_0 -based resonances obtained in this way are depicted as upwards arrows in figure 3 for direct comparison with the experimental spectrum (courtesy of K Welge, from Holle *et al* 1986). The agreement between theory and experiment is indeed very good.

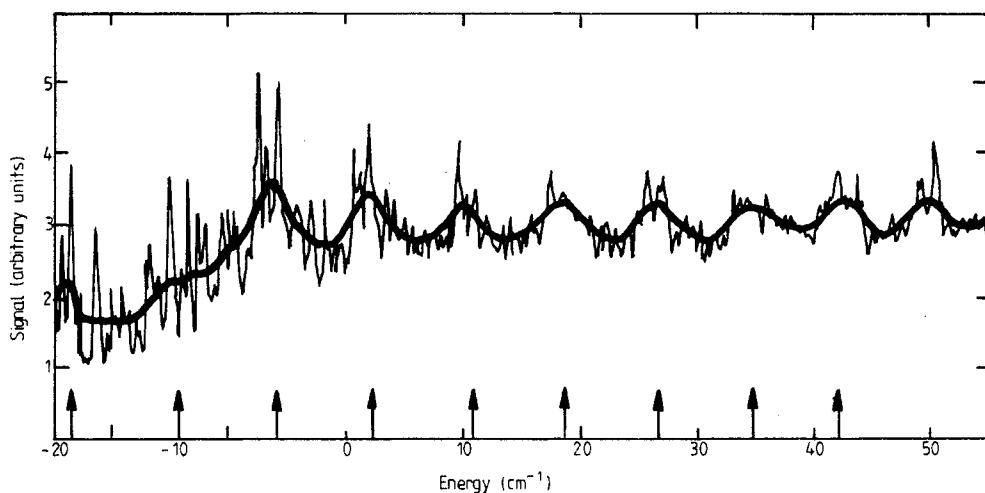


Figure 3. Experimental high-resolution (0.3 cm^{-1} , light line) and simulated (by convolution with a Gaussian of 2 cm^{-1} FWHM) low-resolution spectra of H atom in a 6 T magnetic field for $m=0$ (Holle *et al* 1986). Zero on the energy scale corresponds to the field-free ionisation threshold. Arrows pointing upwards indicate positions of calculated resonance energies.

The widths of the resonances are calculated using an analytic continuation method (Isaacson and Truhlar 1984). In the vicinity of the avoided crossing (figure 1(a)) two eigenvalues may be expressed as solutions of the quadratic equation $E^2 + p(\alpha)E + q(\alpha) = 0$. Taking points that belong to one eigenvalue going through the avoided crossing, one fits the p and q in the form of polynomials in α . The stable root in the complex plane, which may be expressed as $E = E_i - i\Gamma/2$, where Γ is the resonance width, is obtained from the stability condition $dE/d\alpha = 0$. For details we refer the reader to the original paper by Isaacson and Truhlar (1984). The widths obtained (with 30% accuracy) for resonances depicted in figure 2 grow with increasing energy and are in the range $0.4\text{--}1.0 \text{ cm}^{-1}$. We therefore conclude that in the low-resolution spectrum reproduced in figure 3 the widths of the peaks are dominated by the Gaussian width (2 cm^{-1}) used in the simulation of the low resolution. It is worth stressing that the widths obtained are also consistent with the higher (0.3 cm^{-1}) resolution spectrum (light line in figure 3).

An additional remark is in order. Here the resonance theory is used also for the part of the spectrum below the ionisation limit. In this energy region the 'resonance'

width has the meaning of the interval of energies centred on the 'resonance' energy. In this region some of the wavefunctions of the full problem corresponding to discrete eigenvalues will be scarred (Heller 1984), i.e. will have a relatively large amplitude in the vicinity of the ρ_0 . Therefore within the width there may be many high-resolution final total eigenstates whose wavefunctions, scarred by the ρ_0 , will lead to larger transition amplitudes.

Let us now discuss the odd z -parity resonances (figure 1(b)). The sizes of the avoided crossings show that the widths associated with these resonances are much larger than for the even parity resonances (compare figure 1(a)). In QZE the odd-parity states are not seen in the experiment with transitions to the odd z -parity final states as the excitation from the tightly bound $2p$ state is restricted to the region of space very close to the nucleus, where the odd-parity resonances have very small amplitude. On the other hand one may imagine the experiment leading to excitation of the same final states via some intermediate, more extended in space, state. In such a case we would predict the excitation of states scarred by the 'odd-parity' resonance, that in turn would lead to an appearance of the corresponding peak in the FT of the spectra for the odd-parity final states. However, the large widths of the odd z -parity resonances will make their contribution to the spectrum much less dominant than in the even-states case.

Secondly, a careful examination of figure 1(a) will reveal other less stable roots which also undergo avoided crossings. These states are more extended in the direction perpendicular to the ρ_0 as indicated by their bigger slope in α . They correspond, as may be verified by the wavefunction plots, to even 'excited' resonance states (with two nodes in the direction perpendicular to the ρ_0). Similarly to the odd-parity resonances, the large widths of the avoided crossings suggest associated large widths for this type of resonance. Therefore the ρ_0 gives rise to two kinds of resonance states, even and odd with respect to the ρ_0 symmetry. Also higher-excitation (in the perpendicular direction) resonance states exist, albeit their experimental significance becoming less important owing to their large width. We conjecture that this property is not specific for this particular ρ_0 but rather is a general property of ρ_0 -based resonances.

Let us also point out that full large-scale diagonalisations (Wintgen and Friedrich 1986, 1987, Delande and Gay 1986, Wunner *et al* 1986) stop just below the ionisation limit. Therefore, as far as we know, ours is the first fully quantum calculation of the low-resolution spectra close to and above the ionisation limit for laboratory B -field strengths (which are most interesting for 'quantum chaos' studies).

To conclude, we have used the well known (albeit novel in application to QZE) stabilisation technique to obtain quantum mechanically positions and widths of quasi-Landau oscillations close to the ionisation threshold. We introduced the 'energy correction' method (5) which may prove useful in other applications of the stabilisation method. We have shown that a single ρ_0 leads to a family of resonance states differing by the excitation of the perpendicular to the ρ_0 motion. Further study of properties of the ρ_0 -based resonances is being currently carried out. Similar methods can be used to obtain the other ρ_0 -based resonances, e.g. the one responsible for smaller-frequency oscillations observed for the odd-parity spectrum (Holle *et al* 1986). Lastly let us mention that the technique presented has proven to be useful in the study of more complicated systems, where full diagonalisation is virtually impossible. An example is the highly excited vibrational spectrum of H_3^+ (Gomes Llorente *et al* 1988).

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