Diamagnetism of the Hydrogen Atom in the Quasi-Landau Regime

A. Holle, G. Wiebusch, J. Main, B. Hager, H. Rottke, and K. H. Welge
Fakultät für Physik, Universität Bielefeld, D-4800 Bielefeld, Federal Republic of Germany
(Received 31 March 1986)

H-atom diamagnetism has been studied in the quasi-Landau regime with final magnetic states $|m_l^f=0\rangle$, $|m_l^f=+1\rangle$, $|m_l^f=0,+2\rangle$, selectively excited by resonant two-photon absorption. All spectra exhibit detailed characteristic line structures. For $|m_l^f=+1\rangle$ a new quasi-Landau resonance is discovered with a modulation spacing of $0.64\hbar\omega_c$ at $E=0$, while the other final states are modulated with $1.5\hbar\omega_c$. The new resonance can be explained theoretically by a quasibound classical trajectory which lies *outside* the $z=0$ symmetry plane.

PACS numbers: 32.80.–t, 32.60.+i

Diamagnetism of the hydrogen atom is still an essentially open problem of fundamental importance, theoretically as well as experimentally, in one-electron atomic physics (see Kleppner, Littmann, and Zimmermann[1]). At laboratory magnetic fields theory has made substantial progress in the l- and low-n-mixing regimes, well below the ionization limit.[1–3] However, in the particularly interesting quasi-Landau regime with strong-field mixing, no general theoretical solution yet exists, because even the simple Hamiltonian containing only the Coulomb and diamagnetic interactions,

$$H=p^2/2-1/r+\tfrac{1}{8}\alpha^2B^2\rho^2 \text{ (a.u.)},$$

does not allow separation of the Schrödinger equation. In this regime, the problem has only been treated by confinement of the electron to the two dimensions perpendicular to the magnetic field, i.e., to the $z=0$ plane.[4,5] This, however, accounts only for the quasi-Landau spacing, $\gamma\hbar\omega_c$ ($\omega_c=eB/m$), which has been observed in previous experiments with the characteristic modulation factor $\gamma_0=1.5$ at threshold ($E=0$).[6–12] These treatments, of course, do not provide any information on details of the spectrum, and thus on the atomic structure and dynamics in the quasi-Landau regime. A more sophisticated quantum-mechanical treatment has recently been presented.[13] First experiments with the hydrogen atom, naturally of fundamental interest, have recently been reported, including the observation of ($\gamma_0=1.5$)-type quasi-Landau oscillations.[14] We report here more detailed work with surprising results. New and unexpected spectral structures have been observed and theoretically analyzed, significantly modifying our understanding of atomic diamagnetism under strong-field-mixing conditions.

In the experiment, the atoms are excited in a collimated atomic beam at the center of magnetic field by two photons (vuv, vacuum ultraviolet; uv, ultraviolet), $H(n=1)+\text{vuv}\rightarrow H(n=2)+\text{uv}\rightarrow H^*$, as described previously.[15] The atomic beam, with its axis parallel to the field, is crossed at the field center by two pulsed, tunable laser beams (vuv and uv) that are perpendicular to each other, to define an effective excitation zone of ~ 0.5 mm in diameter. The beam intersection point was located between two flat, parallel, fine-mesh grid electrodes (7 mm apart) with their surfaces perpendicular to the magnetic field axis. A surface-barrier diode located 30 mm behind the second electrode served to monitor the ionization of H^* atoms by electron detection. It was operated at ground potential, while the two electrodes were both kept at -6 kV, with a small rest field ($F<1$ V/cm) maintained between them. The electrode arrangement served, first, to shield the excitation region electrically and, second, to drag electrons produced by spontaneous or field ionization towards the detector. Note that the rest field was sufficiently weak so that electric-field effects remained small and unnoticeable. The signal from the surface-barrier detector was observed through a time gate of ~ 5 μs width, started at the time of the laser pulse, to discriminate against detection of H^* atoms ionized later.

At the magnetic field strengths employed ($B\leqslant 6$ T), the Lyman-α transition $(n=1)\rightarrow(n-2)$ is fully governed by the Paschen-Back effect yielding three p-orbital-type magnetic sublevels $|m_l^i=0,\pm1\rangle$. Individual $|m_l^i\rangle$ levels were excited selectively by the vuv laser radiation of sufficiently high resolution of 0.1–0.3 cm^{-1} with linear polarization parallel (π) or perpendicular (σ) to the field. The second excitation step was carried out with the uv laser radiation π or σ polarized. With π polarization ($\Delta m_l=0$), pure final states are excited, that is $|m_l^f\rangle=|-1\rangle$, $|0\rangle$, $|+1\rangle$. With σ polarization ($\Delta m_l=\pm1$) both $m_l^f=\pm2$ and $m_l^f=0$ final states can be excited to yield the mixed final state $|m_l^f=0,-2\rangle$ from $|m_l^i=-1\rangle$ or $|m_l^f=0,+2\rangle$ from $|m_l^i=+1\rangle$. The relative probability of the competing transitions to $|m_l^f|=2$ and $m_l^f=0$ is about 4:1.

Here we have investigated spectra with final states $m_l^f=0$, $+1$, and $+2$. Results are shown in Fig. 1 (light lines), obtained at $B=6$ T. In Figs. 1(a) and 1(b), the final states are purely $|m_l^f=0\rangle$ or $|m_l^f=+1\rangle$, while spectrum 1(c) consists of a 4:1 mixture of $|m_l^f=+2\rangle$ and $|m_l^f=0\rangle$ states. $E=0$ refers to the field-free ionization potential of the atom. Also la-

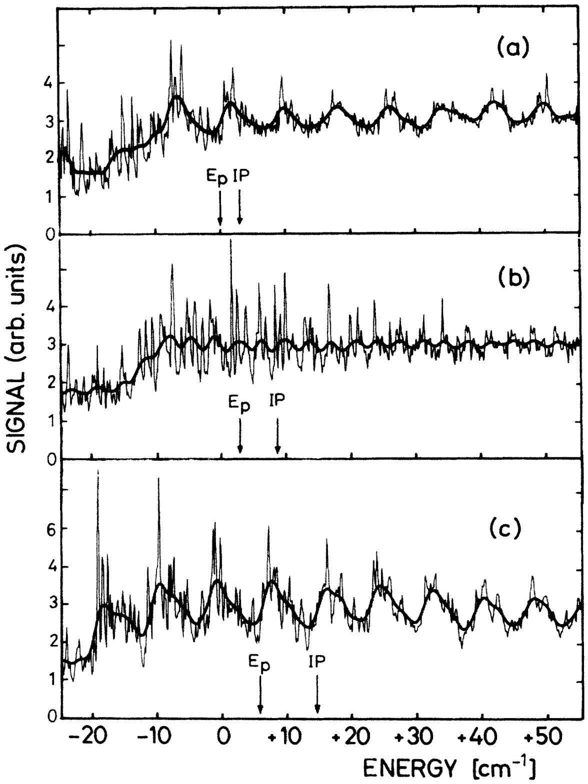

FIG. 1. Two-photon resonant excitation spectra (light lines) of the hydrogen-atom Balmer series around the ionization limit in a magnetic field of strength $B=6$ T, excited through individually selected magnetic substates $|m_l^i=0\rangle$ and $|m_l^i+1\rangle$ of the $n=2$ state to final states $|m_l^f\rangle$ of even parity: (a) $|m_l^f=0\rangle$, (b) $|m_l^f=+1\rangle$, (c) $|m_l^f=+2\rangle$, plus some admixture ($\sim 25\%$ of $|m_l^f=0\rangle$. Resolution $\simeq 0.3$ cm^{-1}. Heavy-line curves: Gaussian average of measured spectra with 2 cm^{-1} FWHM.

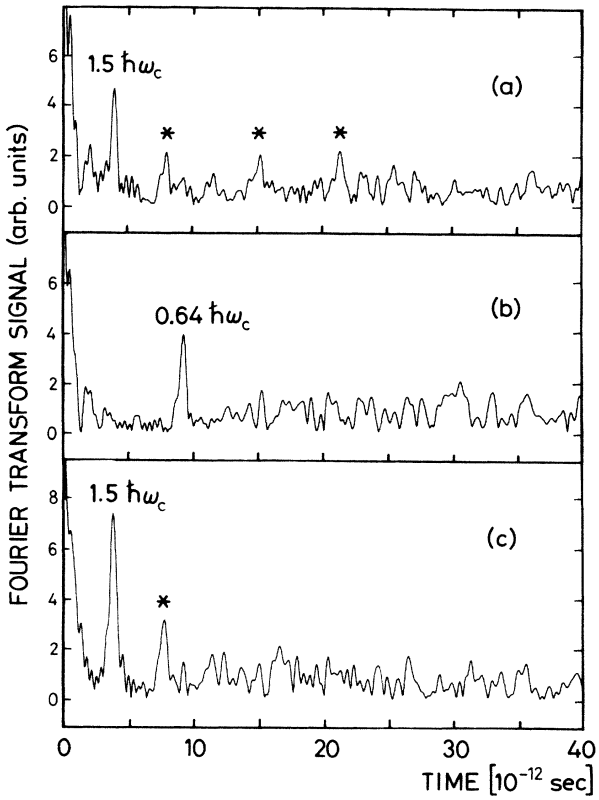

FIG. 2. Fourier transformation (absolute value) of corresponding measured spectra in Fig. 1.

beled are the paramagnetic interaction energy $E_p = \alpha m_l B/2$ and the ionization threshold IP given by the quantum-mechanical zero-point energy of the two-dimensional harmonic oscillator in the Landau limit, $\mathrm{IP}=\alpha(|m_l|+m_l+1)B/2$.[16]

In measuring the spectra, the uv wavelength was scanned in steps of 0.16 cm^{-1}, with the signal in each step accumulated over twenty laser shots at a 10-Hz pulse repetition rate. Each spectrum represents the sum of six independently repeated runs, thus generating confidence in the details of the spectral structures within a precision of $\cong 0.3$ cm^{-1}, almost exclusively from the uv-laser bandwidth ($\cong 0.3$ cm^{-1}). The spectra exhibit individually characteristic line structures superimposed on continuum background. Many of the lines below as well as above the ionization threshold are as narrow as 0.3 cm^{-1}, which corresponds to a spontaneous-ionization lifetime of at least 2×10^{-11} s.

The $|m_l^f=0\rangle$ and $|m_l^f=+2,0\rangle$ spectra [Figs. 1(a) and 1(c)] clearly show quasi-Landau periodicities in the line structures with the previously known characteristic modulation spacing factor $\gamma_0=1.5$. This periodicity is also exhibited by the smooth, heavy-line curves, which were obtained by convolution of the measured spectra theoretically with a 2-cm^{-1} FWHM Gaussian profile. While the measured $|m_l^f=+1\rangle$ spectrum [Fig. 1(b), light line] shows no obvious periodicity, application of the Gaussian smearing procedure reveals a new and fully unexpected quasi-Landau periodicity (heavy line) with $\gamma_0=0.64\pm 0.01$ at $E=0$. The error estimate of ± 0.01 results from a linear fit to the spacing of all resonances throughout the observed energy range. To check this surprising finding, we repeated the experiments at other field strengths ($B=5$ and 4 T), and still obtained $\gamma_0=0.64\pm 0.01$ for the $|m_l^f=+1\rangle$ spectra. This largely excludes experimental artifacts, including the electric-field-mixing effect, where $\gamma_0=0.5$ would be expected [Fig. 1(b)].[17] The decrease of the average signal height at negative energies is attributed to the inefficient field ionization in the weak field. Additional support for this new quasi-Landau resonance is derived from the dependence of γ on the excitation energy. For the spectra $|m_l^f=0\rangle$ and $|m_l^f=+2,0\rangle$ we find from linear fitting $\gamma(E)=-3\times 10^{-3}E+1.5$ (with E in units of inverse centimeters), whereas for

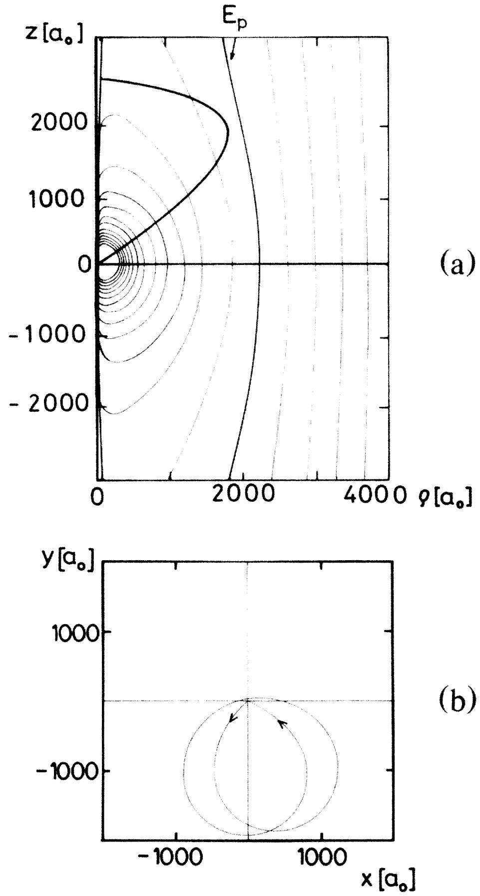

FIG. 3. Closed classical trajectory of electron motion corresponding to $\gamma_0 = 0.64\hbar\omega_c$ quasi-Landau resonance with azimuthal angle $\theta \cong 54°$ and recurrence time $t_{\text{rec}} = 9.34\times10^{-12}$ sec, theoretically calculated for excitation energy E_p. Initial starting conditions at $t=0$: $\rho(t=0)\cong 0$, $z(t=0)\cong 0$, i.e., the Coulomb center. Trajectory projections (a) onto (ρ, z) and (b) onto $(x,y,z=0)$ surfaces.

$|m_f^f = +1\rangle$, $\gamma(E) = -6\times10^{-4}E + 0.64$, so that the slopes also differ.

In addition, we took a Fourier transformation of the spectra over the measured spectral range from -35 to $+60$ cm^{-1}. The Fourier spectra (Fig. 2) show pronounced resonances at times $t_{\text{res}} = 3.95\times10^{-12}$ s and $t_{\text{res}} = 9.34\times10^{-12}$ s. Setting $2\pi\hbar/t_{\text{res}} = \gamma_{\text{res}}\hbar\omega_c$, one obtains the corresponding periodicities $\gamma_{\text{res}} = 1.51$ and $\gamma_{\text{res}} = 0.64$, in agreement with the directly observed γ_0 factors at $E=0$.

Although the quasi-Landau spacing is energy dependent, the γ_{res} from the Fourier transformation are in good agreement with γ_0 because the spectral range is relatively narrow and the energy dependence correspondingly small. The difference between γ_{res} and γ_0 is within the precision of the experiments. Note that some features in the Fourier spectra, particularly those marked by asterisks in Figs. 2(a) and 2(c), are very likely not due to experimental fluctuations but rather represent genuine resonances. If this is true, it indicates the existence of even more quasi-Landau states with corresponding γ factors other than $\gamma_0 = 1.5$ and 0.64. Preliminary results of experiments now in progress to investigate this highly interesting question, support this conjecture.

In analogy to the semiclassical interpretation which attributes the $\gamma_0 = 1.5$ quasi-Landau resonances to an electron motion confined to the $z=0$ plane, i.e., $\theta = 90°$ azimuthal angle, the $\gamma_0 = 0.64$ resonance indicates the existence of a kind of quasi-Landau oscillatoin with the electron moving *outside* of this special plane, that is, with a component of the motion *along* the z coordinate. To understand the $\gamma_0 = 0.64$ quasi-Landau oscillation theoretically, we solved the classical Hamilton equations, as suggested by Reinhardt,[5] seeking a closed trajectory $\{\rho(t), z(t)\}$ on which the electron starts at time $t=0$ as close as $m_l^f = 1$ allows to the origin $\{\rho(t=0)\cong 0, z(t=0)\simeq 0\}$ in an azimuthal direction $\theta(t=0)\neq 90°$, and returns to the origin periodically in a time t_{rec}. We chose the initial condition at the origin to model an electron excited in the Coulomb field with the initial wave packet at the proton hardly perturbed by the long-range part of the potential. At $E=E_p$ and $B=6$ T we find that indeed one, and only one closed trajectory exists with $\gamma_0 = 0.64$. It starts and returns to the origin under an azimuthal angle of about 54° and has a calculated recurrence time $t_{\text{rec}} = 9.34\times10^{-12}$ s, in exact agreement with the experimental t_{res} from the Fourier spectrum. Figure 3 illustrates the trajectory in projections onto the (ρ, z) and $(x,y,z=0)$ surfaces.

In summary, the diamagnetic spectra of the Balmer series of the hydrogen atom around the ionization threshold with selected final magnetic states $|m_l^f = 0\rangle$, $|m_l^f = +1\rangle$, and $|m_l^f = 0, +2\rangle$ were investigated in detail. Even beyond the ionization limit, the quasi-Landau resonances exhibit distinct line substructures, depending characteristically on the nature of the final state. Aside from the known quasi-Landau resonance with $\gamma_0 = 1.5$, a new type with $\gamma_0 = 0.64$ has been discovered, which is rationalized theoretically by a classical motion of the electron in a closed trajectory *outside* of the $(z=0)$ plane with an azimuthal angle $\theta \cong 54°$ at the origin. This will modify the view of atomic diamagnetism in the strong-field-mixing regime significantly. Indeed, we suspect that even more quasi-Landau states will be found.

We wish to thank Professor Andrä for the magnet, and W. P. Reinhardt, K. T. Lu, D. Wintgen, and J. T. Broad for fruitful discussions.

---

[1]The field of highly excited atoms in external fields has re-

cently been extensively reviewed; see, for example, D. Kleppner, M. G. Littmann, and M. L. Zimmermann, in *Rydberg States of Atoms and Molecules,* edited by R. F. Stebbings and F. B. Dunning (Cambridge Univ. Press, Cambridge, England, 1983), pp. 73–116; C. W. Clark, K. T. Lu, and A. F. Starace, in *Progress in Atomic Spectroscopy, Part C,* edited by H. J. Beyer and H. Kleinpoppen (Plenum, New York, 1984), pp. 247–320; J. C. Gay, *ibid.,* pp. 177–246; J. Phys. (Paris), Colloq. **43**, C2 (1982) ("Atomic and Molecular Physics Close to the Ionization Threshold in High Fields").

[2]C. W. Clark and K. T. Taylor, J. Phys. B **15**, 1175 (1982).

[3]G. Wunner, to be published.

[4]A. R. Edmonds, J. Phys. (Paris), Colloq. **31**, C4-71 (1970); A. F. Starace, J. Phys. B **6**, 585 (1973); A. R. P. Rau, Phys. Rev. A **16**, 613 (1977); J. A. C. Gallas, F. Gerck, and R. F. O'Connell, Phys. Rev. Lett. **50**, 324 (1980).

[5]W. P. Reinhardt, J. Phys. B **16**, 635 (1983), and private communication.

[6]W. R. S. Garton and F. S. Tomkins, Astrophys. J. **158**, 839 (1969).

[7]K. T. Lu, F. S. Tomkins, and W. R. S. Garton, Proc. Roy. Soc. London, Ser. A **362**, 421 (1978).

[8]N. P. Economou, R. R. Freeman, and P. F. Liao, Phys. Rev. A **18**, 2506 (1978).

[9]J. C. Castro, M. L. Zimmermann, R. G. Hulet, and D. Kleppner, Phys. Rev. Lett. **45**, 1780 (1980).

[10]R. J. Fonk, F. L. Roesler, D. H. Tracy, and F. S. Tomkins, Phys. Rev. A **21**, 861 (1980).

[11]J. C. Gay, D. Delande, and F. Biraben, J. Phys. B **13**, L729 (1980).

[12]W. R. S. Garton, F. S. Tomkins, and H. M. Crosswhite, Proc. Roy. Soc. London, Ser. A **373**, 189 (1980).

[13]D. Wintgen and H. Friedrich, to be published and private communication.

[14]A. Holle and K. H. Welge, in *Proceedings of the Seventh International Conference on Laser Spectroscopy, Maui, Hawaii, June 1985,* edited by T. W. Haensch and Y. R. Shen, Springer Series in Optical Science Vol. 49 (Springer-Verlag, New York, 1985).

[15]H. Rottke, H. Zacharias, and K. H. Welge, in *Laser Techniques for Extreme Ultraviolet Spectroscopy,* edited by T. J. McIlrath and R. R. Freeman, AIP Conference Proceedings No. 90 (American Institute of Physics, New York, 1982), pp. 402–421; H. Rottke and K. H. Welge, Phys. Rev. A **33**, 301 (1986).

[16]L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Addison-Wesley, Reading, Mass., 1965), 2nd ed., Sect. 111.

[17]K. T. Lu, F. S. Tomkins, H. M. Crosswhite, and H. Crosswhite, Phys. Rev. Lett. **41**, 1034 (1978).