Atomic Diamagnetism: Quasi Landau Spectrum near the Ionization Threshold

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The positions of quasi Landau resonances as a function of magnetic field strength are reported for the first time. These resonances were observed near the ionization limit in the even-parity channels of barium and strontium with use of two-photon laser spectroscopy. The change in the resonance positions as the field strength is varied and the energy spacings of the resonances agree well with hydrogenic semiclassical theory. Resonance profiles, but not positions, are found to be species dependent.

We have observed quasi Landau resonances[1,2] in the even-parity spectra of barium and strontisu near their first ionization thresholds using two-photon ionization spectroscopy. The resonances are observed as high as 100 cm^{-1} above the ionization limit and extend continuously below the limit as a periodic intensity modulation of the severely n- and l-mixed Rydberg levels to energies where individual diamagnetically shifted nl levels are identifiable. The positions of these resonances and their motion as B is varied are reported for the first time and are found to agree well with semiclassical calculations, for both atomic species observed. However, the profiles of the individual resonances in Sr differ radically from those of Ba. The resonances are observed in $M=0$ channels, whereas, in the original observations of Garton and Tomkins, they occur only in the $M=\pm 1$ spectra of the odd-parity series of Ba.

The potential seen by a highly excited atomic electron in the presence of a strong external magnetic field is a superposition of the spherically symmetric Coulomb potential V_C, due to the ionic core, and the cylindrically symmetric diamagnetic potential, $V_D = e^2B^2r^2\sin^2\theta/8mc^2$. Only parity and M, the projection of the angular momentum along the field axis, remain strict invariants. For high excitation or strong magnetic field, when $V_D \gg V_C$, the electron is trapped in a harmonic potential for motion perpendicular to the magnetic field, giving rise to approximately equally spaced, diffuse resonances in the photoabsorption spectrum of atoms in the energy region spanning the ionization limit. In this region, since the orbital frequency for electron motion in the Coulomb field along the magnetic field axis is much less than the cyclotron frequency ω_c, the adiabatic approximation is employed to obtain a two-dimensional hydrogenic model for the resonance positions.[3,4] Using the WKB approximation and the Bohr-Sommerfeld quantization condition, Edmonds obtained[3]

$$\int_{\rho_1}^{\rho_2}[2E+2/\rho-\alpha^2\rho^2]^{1/2}d\rho=(\tilde{n}+\tfrac{1}{2})\pi, \qquad (1)$$

for the energy spectrum of an electron with $M=0$ in a nearly planar orbit perpendicular to the field and passing through the ionic core. Here E is the electron energy with respect to the zero-field ionization limit, ρ is the radial coordinate in the plane of the orbit, ρ_1 and ρ_2 are the turning points of the classical motion, and $\alpha = eB/2mc$. Equation (1), which is expressed in atomic units, is numerically integrated to obtain energy E as a function of the quantum number $\tilde{n}$.

The WKB hydrogenic model which leads to Eq. (1) depicts the quasi Landau resonances as highly excited bound states characterized by a single quantum number $\tilde{n}$, with successively lower $\tilde{n}$ levels being raised across the ionization threshold as B is increased. Although the situation is clearly much more complicated, with each resonance below the limit comprising of many individual l- and n-mixed bound levels, we have found that Eq. (1) successfully describes the spacing and B dependence of the resonance energies.

A linearly polarized, pressure-tuned, nitrogen-laser–pumped dye laser with a linewidth of ~0.06 cm^{-1} was used to observe highly excited even-parity states of Sr I and Ba I via two-photon transitions out of the $ms^2\,{}^1S_0$ ground state, where $m=5$ for Sr I and $m=6$ for Ba I. Atomic vapor pressures of ~0.1 Torr were produced in a resistively heated oven with neon buffer gas at 10 Torr. The oven was placed inside a superconducting dipole magnet producing fields up to 40 kG. The two-photon transitions were detected with a space-charge–amplification ionization detector[5] with the applied electric field parallel to the magnetic field and <0.5 V/cm. Laser wave numbers were obtained using the Argonne National Laboratory 30-ft spectrograph and are accurate to ± 0.02 cm^{-1}.

Figure 1 presents spectra obtained near the first ionization threshold T_∞ for Sr I and Ba I. The value $T_\infty = 45\,932.19\ \text{cm}^{-1}$ for Sr I was obtained by Esherick,[6] and $T_\infty = 42\,034.85\ \text{cm}^{-1}$ for Ba I was obtained by Rubbmark, Borgstrom, and Bockasten.[7] For $B=0$, the spectrum near the limit is dominated by the $ms\,{}^1S_0$–$msnd\,{}^1D_2$ ($m=5$ for Sr and $m=6$ for Ba) Rydberg series converging to the threshold. When $B \geqslant 25$ kG, the quasi Landau resonances appear as a periodic intensity modulation in the spectrum spanning the region of the $B=0$ ionization limit. The noise level for Figs. 1(b) and 1(c) is the same as that seen in the $B=0$ data [Fig. 1(a)]. Complex, incompletely resolved line structure is seen above the noise level, extending up to and possibly across the limit. This fine structure is due to the multitudinous diamagnetically raised and mixed ***nl*** Rydberg levels. The sharp line structure is suppressed above the limit, presumably because the excited elec-

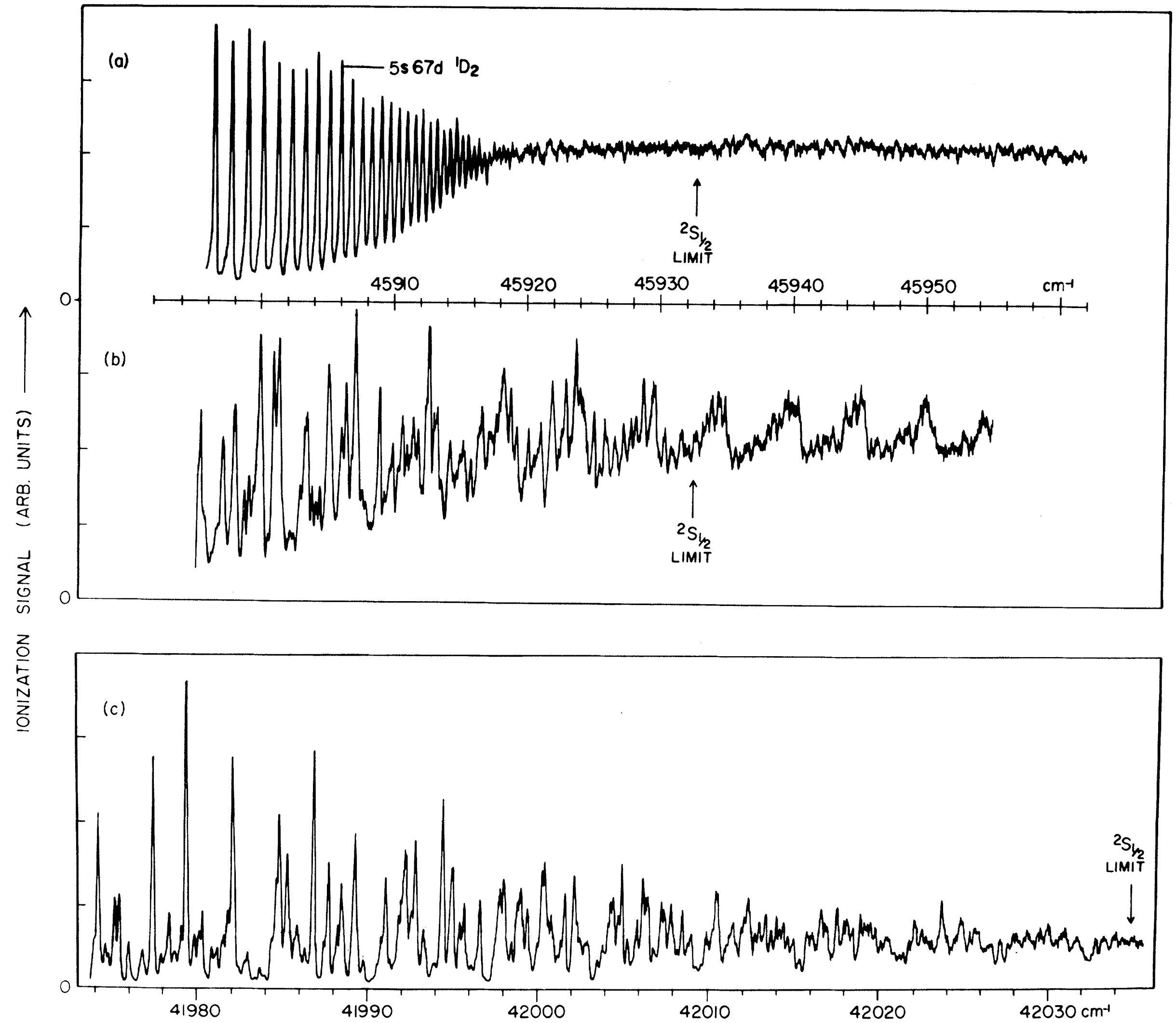

FIG. 1. Two-photon ionization spectra showing quasi Landau resonances in region of $B=0$ ionization threshold. (a) Sr I spectrum with $5snd\,{}^1D_2$ series converging to Sr II $5s^2S_{1/2}$, $B=0$. (b) Same as (a), but $B = 40.0 \pm 0.1$ kG, π polarization ($M=0$). (c) Ba I, $B = 40.1 \pm 0.1$ kG, $M=0$, showing intensity minima below the Ba II $6s^2S_{1/2}$ limit. (For $B = 40$ kG, $1.5\hbar\omega_c = 5.60\ \text{cm}^{-1}$.)

tron can autoionize by moving along the magnetic field direction. The structure below the limit is reproducible, provided that the field strength is accurately reproduced.

For π polarization ($M=0$) and $B=40$ kG, the intensity modulations in the Ba I spectrum below the limit were identifiable through the n-mixing region into the l-mixing region, where the $6s31s$ $^1S_0$ and $6s32s$ $^1S_0$ levels are still identifiable. The contrast of the resonances gradually decreases as E increases beyond the limit until, for $E>100$ cm^{-1}, their positions are no longer measurable at $B=40$ kG. No periodic intensity modulations were clearly observed in σ polarization, but any resonances present may have been obscured due to superposition of $M=0$, $+2$, and -2 components. No resonances were observed above the noise level for $B\leq 20$ kG.

The comparison of the 30.4 and 29.4 kG Sr I scans in Fig. 2 demonstrates the rapid movement of the resonances and the scrambling of the fine structure as B is varied. Data obtained at various field strengths, some as little as 0.3 kG apart, allowed the positions of individual resonances to be followed over the range $B=25$–40 kG.

A plot of the energies of the resonances as a function of $\tilde{n}$ is given in Fig. 3 for a few B values. The solid lines are calculated from numerical integration of Eq. (1). The position of a resonance was defined by its most identifiable feature: the intensity minimum for barium and the peak of the sawtooth-shaped profile for strontium. Uncertainties in these measurements are due mostly to the complicated fine structure superposed on the resonances, and are usually less than 0.5 cm^{-1}.

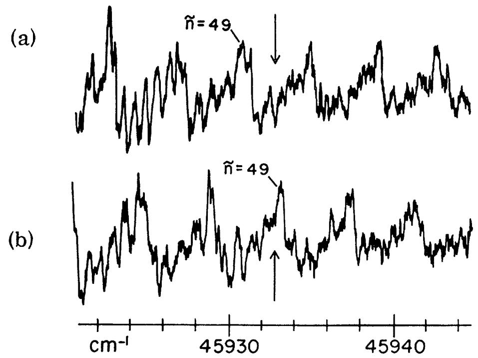

FIG. 2. Movement of quasi Landau resonances in Sr I as B is varied. The $\tilde{n}=49.0$ resonance is labeled for both (a) $B=29.4\pm0.2$ kG and (b) $B=30.4\pm0.1$ kG. For $B=30$ kG, $1.5\hbar\omega_c=4.20$ cm^{-1}. Arrows denote Sr II $5s\,^2S_{1/2}$ limit.

To compare these measurements to the results of Eq. (1), one resonance for each element at $B=40$ kG is assigned a value of $\tilde{n}$ such that the interpolated value of $\tilde{n}$ at the zero-field ionization limit agrees with that calculated from Eq. (1), which gives $\tilde{n}=44.58$ for $E=0$ and $B=40$ kG. Thus, for Sr the first peak above the zero-field limit at $B=40$ kG is assigned a value of $\tilde{n}=45.0$, while for Ba the first minimum above the limit at $B=40$ kG is assigned the same $\tilde{n}$ value. At the lowest energies in Ba, peaks rather than minima are measured, with the peaks being assigned half-integral $\tilde{n}$ values. The one datum point for each element whose $\tilde{n}$ value is defined to agree with Eq. (1) is marked with an arrow. With this definition for each element, the $\tilde{n}$ values of all the resonances are fixed for all field strengths, since the motion of each resonance can be unambiguously traced from 25 to 40 kG.

In agreement with the calculations of Starace[4] and O'Connell,[8] the spacing of the resonances at $E=0$ is found to be $1.5\hbar\omega_c$ for all values of B, in contrast to the value $\hbar\omega_c$ expected for a free electron in the magnetic field.[2] Also, the prediction[9] by Rau of a $B^{-1/3}$ scaling of the value of $\tilde{n}$ at the ionization threshold, which is implicit in Eq. (1)

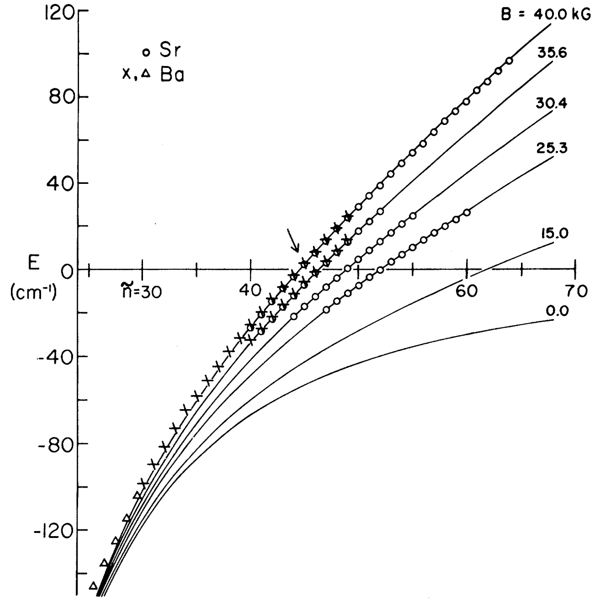

FIG. 3. Resonance positions as a function of $\tilde{n}$. Curves are obtained by numerical integration of Eq. (1). The two data points defined to agree with the theory are marked by the arrow. Circles, Sr I resonance peak positions; crosses, Ba I resonance minimum positions; triangles, Ba I peak positions. The lowest datum point corresponds to the $6s31s$ $^1S_0$ level.

for large $\tilde{n}$, is confirmed. The agreement between the data and the calculated curves in Fig. 3 over the full energy range observed is noteworthy since the adiabatic approximation is not valid over this entire range. Roughly, one would expect Eq. (1) to be invalid for energies for which $\omega_c < \omega_B$, where ω_B is the electron orbital frequency in a Bohr atom. For $B = 40$ kG, $\omega_c = \omega_B$ at $E \approx -46$ cm^{-1}. Below this energy, deviations between Eq. (1) and the data may be expected, although qualitative agreement is not surprising since, for $V_D \ll V_C$, Eq. (1) can be expanded to yield the usual hydrogenic energy levels with $n_C = \tilde{n} + \frac{1}{2}$ (where n_C is the usual principal quantum number), plus a diamagnetic-shift term having the correct $n_C{}^4B^2$ dependence, but with too large a coefficient. Indeed, the Ba I $6s31s\ {}^1S_0$ level has been assigned $\tilde{n} + \frac{1}{2} = 26.0$ by extrapolation from the $\tilde{n}$ assignment at the limit, while its effective quantum number at $B = 0$ is $n^* = 26.8$. Diamagnetic shifts of $6sns\ {}^1S_0$ levels with $n \leq 30$ were reported earlier and shown to agree with hydrogenic theory except for complications due to configuration interactions.[10]

While we have found that Eq. (1) successfully describes the coarse features of the spectrum in the region of $E = 0$, it is clear that much additional study is required before the strong-field mixing regime is understood. Particularly interesting would be a theoretical study of the species-dependent resonance profiles and an elucidation of the connection between $\tilde{n}$ and the zero-field quantum numbers. Further experimental work aimed at identifying the $B = 0$ Rydberg levels responsible for the observed fine structure and at testing for the appearance of resonances in $M = \pm 1, \pm 2$ spectra could be fruitful.

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Observation of Laser-Induced Penning and Associative Ionization in Li-Li Collisions

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We report the direct observation of ^{6}Li$^+$, ^{7}Li$^+$, ^{13}Li$_2{}^+$, ^{14}Li$_2{}^+$ arising from single collisions between beams of lithium atoms in the presence of a laser field. Although the relative intensity of *atomic* ions reflects the natural isotopic abundance, a pronounced enrichment of ^{13}Li$_2{}^+$ relative to ^{14}Li$_2{}^+$ is observed.

The enhancement of inelastic collision processes in the presence of an intense light field offers the possibility of controlling atomic interactions by external laser radiation. An important distinguishing feature of "laser-induced" collisions is that the energy absorbed from the field is nonresonant with respect to the separated collision partners. Several such processes have received theoretical treatment, including energy transfer,[1-11] charge transfer,[12-14] Penning ionization,[15] and collisional ionization.[16,17] Although two separate laboratories have observed a laser-induced energy-transfer collision,[18,19] other experimental results[20,21] have not found a simple