Motional Stark Effect on Li Vapor Photoabsorption in High Magnetic Fields

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Calculation of the motional Stark effect for $B \sim 50$ kG shows it to become dominant along Rydberg series of light elements at $T \sim 1000$°C before the diamagnetic shift changes the spectrum from Rydberg to Landau type, thus accounting for the $\frac{1}{2}\hbar\omega_c$ spacing observed in Li. This spacing is preserved when both of the competing effects—motional Stark and Landau modulation—are strong. The spectrum appears to depend critically on the relative position of the thresholds for the onset of the two effects.

The study of quasi-Landau levels near the ionization threshold of atomic photoabsorption spectra in strong magnetic fields[1] has recently taken a new turn when extended to Li.[2] The level spacing in Li has been found equal to one Lorentz unit (1 L.U. $= \mu_0 B = \frac{1}{2}\hbar\omega_c$) instead of the value $\frac{3}{2}\hbar\omega_c = 3\mu_0 B = 3$ L.U. found previously for other elements.[1] Also the strong l mixing induced by the magnetic field was found to include l values of both parities thus violating the traditional selection rule. As mentioned in Ref. 2, this violation might be caused by the electric field resulting from the thermal motion of Li atoms across the magnetic field. However, the new spacing $\frac{1}{2}\hbar\omega_c$ was not accounted for. This Letter demonstrates how the motional Stark field causes not only parity-forbidden m-nonconserving transitions but also the new spacing in Li.

A perturbation treatment and a matrix diagonalization will show the motional Stark effect to become predominant in Li as the principal quantum number increases from $n \sim 20$ to $n \sim 30$, that is, just *before* the diamagnetic shift overtakes the Rydberg line interval, leading to the appearance of quasi-Landau levels. The picture that emerges will probably remain fragmentary pending development of a theory of the onset of the Landau spectrum. We aim here at directing attention to the contrasting evidence from different spectra, to the opportunity for experimentation with combined magnetic and electric fields, and to the apparently critical role of the thresholds for dominance of diamagnetic or of motional Stark effect.

The presence of a thermal velocity $\vec{V}_{th}$ and of the resulting field $\vec{E} = \vec{V}_{th} \times \vec{B}/c$ breaks both the axial symmetry about $\vec{B}$ and the inversion symmetry at $\vec{r} = 0$, thus allowing changes of both the magnetic quantum number $m = \vec{l} \cdot \hat{B}$ and the orbital parity $(-1)^l$. The linear Zeeman spacing of orbital levels, $\mu_0 B$, can then reemerge as a dominant feature in the spectrum near threshold. (The g factor is omitted here as we deal with the orbital motion only. For Li in a strong field the linear Zeeman effect reduces to its Paschen-Back limit in which orbit and spin remain uncoupled. The spin does not flip and does not influence the spectrum.) This phenomenon is amplified by the particularly high value of V_{th} for Li.

The nonconservation of $m = \vec{l} \cdot \hat{B}$ is documented by the analysis of the l-mixing satellites of the $2s \rightarrow 21p$ σ_+ transition in Li shown in Fig. 1. The calculation diagonalizes the energy matrix, with diamagnetic and motional Stark contributions, in a base set consisting of the Rydberg states with $n = 21$, $l + m$ even, and $m = -1$, 0, 1, 2, and 3. Recall that photoabsorption of σ_+-polarized light from the $m = 0$ ground state would excite only $m = 1$ states in the absence of motional Stark mixing. Also the quantum defects of Li are ~ 0.05 for

p states and negligible for $l>1$. Accordingly the calculated eigenvalues remain in the vicinity of the unperturbed $21p$ level. This level is indicated by σ_0 in Fig. 1; the $22p$ level lies 22 cm^{-1} higher. The diamagnetic splitting of the $m=1$ group of levels is seen to spread over ~3 cm^{-1}, but the levels with $m=2$ and 3 lie clearly outside the $m=1$ group. Thus the motional Stark effect exceeds the diamagnetic splitting here, while both of them remain well below the separation of successive Rydberg lines.

A perturbation-theory estimate serves to illustrate the results shown in Fig. 1 and to interpret the lower intensity of the levels with $m\neq 1$. The electron's energy due to the motional Stark effect is $e\vec{V}_{th}\times\vec{B}\cdot\vec{r}/c=m_eV_\perp\omega_c x$, where the x axis is chosen along $\vec{V}_{th}\times\vec{B}$ and $V_\perp=|\vec{V}_{th}\times\hat{B}|$. The amplitude ratio of optical transitions to states with $m\neq 1$ and $m=1$ is then given by first-order perturbation theory as the ratio of the matrix element of the perturbation energy, $m_eV_\perp\omega_c x$, to the energy difference of successive Zeeman levels with $|\Delta m|=1$, namely, $\mu_0B=\frac{1}{2}\hbar\omega_c$. (This energy difference should actually be corrected for the nondegeneracy of s, p, d levels; see below.) To evaluate this matrix element, set $x=(x^2+y^2)^{1/2}\times\cos\varphi$. The matrix element of $(x^2+y^2)^{1/2}$ diagonal in n is $\sim a_0n^2$, where a_0 = Bohr radius, while $(m|\cos\varphi|m')=\frac{1}{2}\delta_{m',m\pm1}$. The relative probability of transitions to $m\neq 1$ is then given by the squared ratio

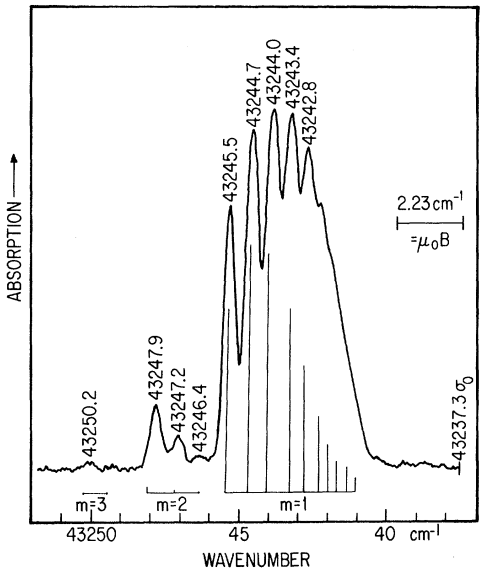

FIG. 1. Li absorption spectrum for $n=21$ manifold in the magnetic field. The curves are densitometer traces from photoabsorption measurements (Ref. 3) taken at vapor pressure 0.1 mm, for magnetic field strength $B=47.8$ kG. The calculated values of positions corresponding to different nominal m components, $m=1$, 2, or 3 are marked. The vertical lines represent calculated values of the square of the eigenvector component belonging to $l=1$, $m=1$ allowed transitions. Experimental line centers, the $B=0$ position ($\sigma_0=43\,237.31$ cm^{-1}) of $n=21$, and the magnitude of linear Zeeman shift (μ_0B) for $B=47.8$ kG are indicated. The absorption features with $m=3$ (and 2) become far more prominent in spectra at higher pressures.

$$P=\frac{P_{n=21,m=1\pm1}}{P_{n=21,m=1}}\sim\left[\frac{m_eV_\perp\omega_c(n,l,m|x|n,l+1,m\pm1)}{\frac{1}{2}\hbar\omega_c}\right]^2_{n=21,m=1}\sim\left[\frac{m_e}{M_{Li}}\frac{kT}{R}n^4\right]_{n=21}\sim10^{-1},\tag{1}$$

where we have set $\frac{1}{2}M_{Li}V_\perp^2\sim kT$ and $R=\hbar^2/2m_ea_0^2=13.6$ eV.

This probability becomes large in Li as n increases from ~20 to ~30, i.e., just before the diamagnetic shift and l mixing also become important by approaching the separation of Rydberg levels in fields of 40–50 kG. The m mixing due to the motional Stark effect redistributes the intensities of the observable lines more rapidly, thus accounting for the observed collapse of the Li spectrum at lower n than for other spectra. Note for comparison that in the case of Na the ratio (1) would be reduced by a factor $M_{Na}/M_{Li}\sim3$. Further, the larger value of the quantum defect requires us to increase the level separation indicated by $\frac{1}{2}\hbar\omega_c$ in Eq. (1) to $\frac{1}{2}\hbar\omega_c+R(2/n^3)\Delta\mu$ which allows for the nondegeneracy of levels with different l. Setting $\Delta\mu=\mu_p-\mu_d$, this additional term increases the denominator in the brackets of (1) by only ~$\frac{1}{3}$ for Li but by a factor of 2 for Na. In aggregate the value of P would then be ~10 times smaller for Na than for Li. (Experiments with Na are hindered by its weak photoabsorption near threshold.)

Only a weak structure remains observable by the time the quasi-Landau spectrum of Li gets established at $n>30$. At this point the m mixing should be essentially complete as the value of P, Eq. (1), approaches unity. (Note that the Stark effect mixes l values of opposite parity, whereas parity conservation has a main role in the formation of quasi-Landau states.[1]) The quasi-Landau mechanism alone would now yield a series of

levels with different quantum numbers n_L spaced by $3\mu_0 B = \frac{3}{2}\hbar\omega_c$ for each value of m. The linear Zeeman shift of orbital levels would displace the series with different m by $\mu_0 B = \frac{1}{2}\hbar\omega_c$ for each unit of m. Accordingly the ensemble of all Landau levels would be represented by $(3n_L + m)\mu_0 B$, forming the two-dimensional array shown in Fig. 2. While photoabsorption from the $m = 0$ ground state in the σ_+ polarization would reach only the $m = 1$ levels of this array in the absence of the motional Stark effect, this effect couples now all adjacent columns strongly for Li in the Landau range. Superposition of the columns yields then the uniform level spacing of $\frac{1}{2}\hbar\omega_c$, but only under the remarkable condition that the spacing remains *unaffected by the strong coupling* of the columns. It will now be shown that the spacing is indeed preserved in a plausible approximation.

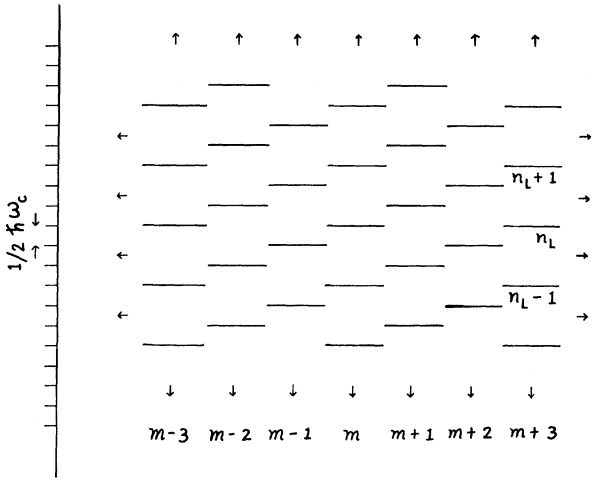

FIG. 2. Array of levels $E_{n_L m} = (m + 3n_L)\mu_0 B$.

To this end we express the magnetic Hamiltonian in the Landau range as a matrix whose diagonal elements represent the array of Fig. 2 while the off-diagonal elements couple the levels of adjacent columns. This coupling is not sharply restricted in vertical range but it presumably depends on the difference $|n_L' - n_L|$ of two levels—for the large values of n_L of interest—rather than on n_L and n_L' separately. Thus we write

$$(n_L m|H|n_L'm') \sim (3n_L + m)\mu_0 B\delta_{n_L n_L'}\delta_{mm'} + f(|n_L - n_L'|)(\delta_{m,m'-1} + \delta_{m,m'+1}). \tag{2}$$

Provided further that this approximation holds over ranges of n_L and m larger than $f(|n_L - n_L'|)/\mu_0 B$, the matrix (2) varies, under translation in m or n_L, as

$$\begin{aligned}(n_L m|H|n_L'm') &= (n_L, m+1|H|n_L', m'+1) - \mu_0 B\,\delta_{n_L n_L'}\delta_{mm'} \\ &= (n_L + 1, m|H|n_L' + 1, m) - 3\mu_0 B\,\delta_{n_L n_L'}\delta_{mm'}.\end{aligned} \tag{3}$$

Given an eigenvector $\psi(n_L, m)$ of this matrix,

$$\sum_{n_L' m'}(n_L m|H|n_L'm')\psi(n_L', m') = E\psi(n_L, m), \tag{4}$$

one sees that $\psi(n_L, m+1)$ is also an eigenvector $E + \mu_0 B$ and $\psi(n_L + 1, m)$ is an eigenvector with eigenvalue $E + 3\mu_0 B$. The translational property, Eq. (2), thus implies invariance of the level spacing.

Several lines of investigation emerge now as promising. Firstly, of course, the interpretation in terms of motional Stark effect should be verified and extended by studying the absorption spectra of several elements under the combined influence of a strong magnetic field and of a weak external electric field, in the range of 50–300 V/cm. Supporting, if complementary, evidence has emerged anyhow from an independent study of the combined influences of the motional Stark and diamagnetic effects upon the $n = 9$ levels of He in very strong fields.[4]

Secondly one may study further the motion of an electron in the combined potential of the Coulomb, magnetic, and electric (motional or external) fields. This potential may be represented as the sum of a Coulomb and of a parabolic term with the axis of the parabola shifted outside the nucleus:

$$V = -e^2/r + \tfrac{1}{8}m_e\omega_c^2(x^2 + y^2) - m_e V_\perp \omega_c x = -e^2/r + \tfrac{1}{8}m_e\omega_c^2[(x - V_\perp/\omega_c)^2 + y^2] - 2m_e V_\perp^2, \tag{5}$$

$V_\perp$ being replaced by $eE/m_e\omega_c$ for a dc field of strength E. Note that the strength of the motional Stark effect, represented by $V_\perp$, has only a secondary influence on the potential (5). The last term of this expression only shifts all levels equally, while the ratio $V_\perp/\omega_c$—which represents the relative strength of Stark and Zeeman effects—shifts only the axis but not the strength of the parabolic potential.

Finally, we have seen that the onset of the motional Stark effect depends for small quantum defect on the ratio of the Stark and linear Zeeman shifts, which is independent of B and increases as $\langle x^2+y^2\rangle \sim a_0^2 n^4$; on the other hand, the onset of the quasi-Landau spectrum depends on the ratio of the quadratic Zeeman shift $(e^2/8mc^2) \times B^2\langle x^2+y^2\rangle$, and of the interval of Rydberg levels, which increases as n^7. The Stark effect becomes dominant at n values for which the ratio (1) exceeds unity [with $V_\perp$ replaced by $eE/m_e\omega_c$ and $\frac{1}{2}\hbar\omega_c$ increased by $R(2/n^3)\Delta\mu$ as appropriate]. Similarly the Landau structure emerges when the product $[B/(50\text{ kG})]^2\ (n/29)^7$ exceeds unity. The two phenomena thus set in at comparable values of n but either onset may be advanced or retarded by experimental parameters, such as the mass and quantum defect of the element. Thus the intensity of photoabsorption is modulated by the Landau-level periodicity $\frac{3}{2}\hbar\omega_c$ for $n \gtrsim 30$ and $B = 47$ kG in Ba and in other elements, with only a trace of the additional periodicity $\frac{1}{2}\hbar\omega_c$,[5] while only the latter modulation appears in Li. The amplitude ratio of the two modulations is thus very large or very small in the Landau range for the atoms investigated this far. The two modulations might well coexist with comparable amplitudes depending on experimental parameters such as $V_\perp$. Alternatively, extreme values of their amplitude ratio might result quite generally depending on whether the motional Stark or the Landau effect dominates the spectrum at a lower value of n. A theoretical study of this situation may be worthwhile as a prototype of spectral phenomena reflecting the critical onset of two or more competing effects.

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Calculation of Parity-Nonconserving Optical Rotation in Atomic Bismuth

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Using results of the Weinberg-Salam model, we have calculated in lowest order the magnitude of the optical rotation parameter $\mathrm{Im}(E1/M1)$ in atomic bismuth for the four allowed $M1$ transitions from the ground state within the configuration $6s^2 6p^3$. Results based on Dirac-Hartree-Fock orbitals are found to vary significantly according to the form used for the electric dipole operator in the $E1$ matrix element. This is interpreted as evidence for large correlation effects.

The existence of weak neutral currents and associated prediction of parity-nonconserving (PNC) effects in atoms have stimulated active research, both theoretical and experimental.[1] Except for the experiment by Barkov and Zolotorev,[2] efforts to detect PNC optical rotation in atomic bismuth[2-5] have yielded results essentially inconsistent with that predicted by recent theoretical calculations.[6-8] It seems apparent, however, that in addition to being inherently difficult the calculations are strongly model dependent and may be sensitive to small variations in electron wave functions. To help resolve these difficulties we have reexamined the problem with an *ab initio*

