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Landé factor and lifetime measurements in even-parity Rydberg series of Pb I using time-resolved laser spectroscopy

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Abstract

Landé $g_J$ factors and lifetimes for the levels in the even-parity Rydberg series $6p^np$ $(n = 9–16)$ and $6p^nf$ $(n = 6–12)$ as well as for the levels from perturbing configurations of neutral lead have been measured using laser-induced fluorescence and Zeeman quantum-beat techniques. Two-step excitation was performed in a thermal atomic lead beam. Strong perturbations in the Rydberg series are manifested both in the $g_J$ factor and lifetime data. (Some figures in this article are in colour only in the electronic version)

1. Introduction

The investigation of atomic Rydberg states remains an active research topic in the areas of atomic physics and laser spectroscopy (Aymar et al 1996). Although the Rydberg states of alkali and alkaline earth elements have been extensively investigated and their spectra have been successfully analysed with multi-channel quantum defect theory (MQDT) (Aymar 1984), studies of heavy elements are still rare. Lead is the second heaviest stable element besides bismuth, and it has distinct characteristics in atomic structure and spectra. First, its ground configuration consists of two 6p electrons outside a full shell. Therefore its Rydberg series are composed of a p-electron ion core and a highly excited electron, so in this respect it is different from alkali earth Rydberg series with an s-electron ion core. Second, its orbital–spin coupling is strong enough so that its ground configuration and the excited configurations $6p^ns$ and $6p^np$ are adequately described by $jj$ coupling while the configurations with high-angular momentum electrons (e.g. $6p^{n-1}$ and $6p^{n-2}$) are close to $jk$ coupling. Third, lead is a high-$Z$ atom with strong configuration interaction and channel mixing effects. Therefore it is interesting to investigate the Rydberg states of the lead atom and test the validity of the
theoretical approaches to this atomic system. In addition, the investigation of the high-lying states of carbon group atoms offers a good chance to understand chemical reactions of excited atomic components.

In the previous work by Li et al (1998), the properties of the odd-parity levels in the 6pns and 6pnd Rydberg series were comprehensively studied by combining time-resolved laser spectroscopic lifetime measurements and relativistic Hartree–Fock calculations. The astrophysically important transitions connected with those odd-parity levels were also analysed by Biémont et al (2000). For the even-parity Rydberg series, a few spectroscopic energy level studies should be mentioned. By analysing the emission spectrum in the spectral range 1733–39 039 Å, Wood and Andrew (1968) determined the low-lying even-parity levels. In a following paper, the \(g_J\) factors of some low-lying even-parity levels were also investigated by Wood et al (1968). Three-photon resonance laser spectroscopy was first used by Young et al (1980) to investigate the even-parity Rydberg series. Ding et al (1989) studied the \(J = 0\) and 2 Rydberg series by means of two-photon resonance ionization spectroscopy. With a two-step excitation used in conjunction with a thermonic diode detector, Farooqi et al (1995) reported new data for the \(J = 0, 1\) and 2 even-parity levels. Recently, two-colour resonance ionization spectroscopy of \(J = 0, 1\) and 2 even-parity levels was applied to Pb I by Hasegawa and Suzuki (1996). The corresponding term energy values were analysed with the MQDT approach by the corresponding authors in the last three works mentioned above. In order to give more insight into the understanding of the properties of Rydberg states of Pb I, additional physical quantities are required. When more than one channel converges to a given limit, the energy values are not sufficient to derive the MQDT wavefunctions. Experimental quantities such as lifetimes, Landé \(g_J\) factors and hyperfine structures are required in order to obtain the complete wavefunctions (Aymar et al 1996, Luo Xingye et al 1994).

For the reasons mentioned above, we performed an experiment to determine the Landé \(g_J\) factors and lifetimes in the even-parity Rydberg states of Pb I using Zeeman quantum-beat and time-resolved laser spectroscopy. The radiative lifetimes of 25 levels and \(g_J\) factors of 28 levels were measured. The \(g_J\) factors were obtained from the analysis of the Zeeman quantum beats in the time-resolved fluorescence signal. This technique has been previously successfully used in Lund for the measurements of \(g_J\) factors in, e.g., barium (Grafström et al 1982) and ytterbium (Jiang Zhankui and Larsson 1991, Zerne et al 1996) Rydberg series.

2. Experimental set-up

A partial energy level diagram and excitation schemes of Pb I, which are relevant to the present experiments, are shown in figure 1. Two-step excitation was used to populate the even-parity Rydberg series. For the first step, 283.39 nm laser radiation was employed to induce the transition of a Pb atom from the ground state 6p\(^2(1/2, 1/2)\) to the intermediate 6p7s(1/2, 1/2)\(_1\) level, which has a lifetime of 6.0 ns (Biémont et al 2000). Another tunable laser pulse in temporal overlap with the first-step laser pulse was used to excite the atoms further to the selected even-parity levels. The fluorescence in the transition to 6p7s(1/2, 1/2)\(_0\) or 6p7s(1/2, 1/2)\(_1\) was chosen to record the exponential decay.

The experimental setup is shown in figure 2. A Continuum ND-60 dye laser operating with Rhodamine 6G was pumped by a Continuum NY-82 Nd:YAG laser (YAG denotes yttrium aluminium garnet). The radiation used to induce the transition from 6p\(^2(1/2, 1/2)\)\(_0\) to 6p7s(1/2, 1/2)\(_1\) was obtained by frequency doubling of the dye laser in a KDP crystal. A further tunable laser system of the same type (Nd:YAG laser pumped dye laser), which operated with different dyes (Coumarin 307, 102, 47, 120 and Stilbene 3), was used to produce laser radiation in the region 517–420 nm to induce the transitions from 6p7s(1/2, 1/2)\(_1\) to the
even-parity Rydberg levels. In order to make proper time overlap of the two laser beams, which is absolutely necessary for the short lifetime of the intermediate level, both Nd:YAG lasers were externally triggered by a Stanford Research Systems Model 535 digital delay generator. A resistively heated oven containing metallic lead produced the atomic beam of low collimation ratio in the vacuum chamber. The two laser beams were shone horizontally to cross at a small angle at the centre of the vacuum chamber, where the atomic beam was also encountered perpendicularly. Following the stepwise excitation, fluorescence light was detected perpendicular to both the laser beams and atomic beam by a RCA C31034 photomultiplier tube (PMT) after spectral selection with a grating monochromator. The signal was then recorded by a Tektronix DSA 602 transient signal analyser. Through a GPIB cable, the time-resolved signal could be transferred to a personal computer, where the analysis was
performed in direct connection with the experiments.

A set of Helmholtz coils was employed to generate a homogeneous magnetic field, which was oriented along the direction of fluorescence detection and parallel to the horizontal component of the Earth’s field. This field was applied to generate the Zeeman splitting in the investigated level. Another coil system was used to compensate for the vertical component of the Earth’s magnetic field. The Helmholtz coils were operated with a high stability constant-current power supply. The current can be monitored from the voltage drop across a precision resistor using a digital multi-meter (Hewlett Packard 3420A) and the stability was better than 5 parts in 10^5. The magnetic field was calibrated using Zeeman quantum beat measurements in the 6s6p^3P_1 level of Yb I, which has a g_J factor determined to be 1.4928 (Budick and Snir 1967, Baumann and Wendel 1968). Beside the lead oven in the vacuum chamber, another resistively heated oven containing metallic ytterbium was mounted, which provided the ytterbium atomic beam for the calibration of the magnetic field. The accuracy of the calibration was within 0.02%.

3. Measurement and results

The correct wavelength tuning for the first step was ensured by observing the fluorescence light from the 6p7s(1/2, 1/2)_1 level to the 6p^2(1/2, 3/2)_2 level at 409.6 nm. The laser for the second-step excitation has a linewidth of 0.2 cm^{-1} and a pulse duration of 8 ns. The wavelength tuning for the second-step excitation was calculated from the energy values of the Rydberg levels given in those references summarized in table 1.

As is well known, a degenerate atomic level will split into several sublevels in a magnetic field (the Zeeman effect). Zeeman sublevels can be coherently populated with a pulsed laser and a temporal modulation (Zeeman quantum beats) can be observed in the time-resolved fluorescence signal. In the present experiments, two linearly polarized laser beams have been utilized for the excitations. The first-step excitation laser was horizontally polarized and the second-step excitation laser was vertically polarized, which was chosen to be orthogonal to the magnetic field \( B \) to introduce \( \sigma \) transitions. The quantum beats in the detected fluorescence occur with a frequency \( \omega \) related to the magnetic field \( B \) by the equation \( \hbar \omega = 2g_J\mu_B B \), where \( \mu_B \) is the Bohr magneton. If the magnetic field strength and the beat frequency are measured, the Landé \( g_J \) factor can be determined. By sending light at the fundamental frequency of the dye laser, which provided the first-step transition, through the ytterbium beam and tuning to resonance to the 6s6p^3P_1 level of Yb I, the magnetic field could be calibrated through the quantum beats measurements of this level in real time. A typical recording of the fluorescence signal with Zeeman quantum beats is shown in figure 3. The frequency of the quantum beats in the detected fluorescence curve was obtained by a Fourier transform performed on a personal computer. During the measurements of Landé \( g_J \) factors the fluorescence decay curves were recorded with the magnetic field alternately in opposite directions by switching the current direction in the Helmholtz coils. The average of the two beat frequencies is independent of the horizontal component of the Earth’s magnetic field. Three magnetic fields of different strength were generated in the coils for the \( g_J \) factor measurements, and the mean values were given as the final results in table 2, where the quoted error bars reflect statistical scattering from different recordings.

In the lifetime measurements, a sufficiently strong magnetic field provided by the Helmholtz coils has been applied to wash out the Zeeman quantum beats. Lifetime values were obtained by fitting an exponential to the recorded fluorescence curves. A typical fluorescence curve with an exponential fitting is shown in figure 4. To check the effects of radiation trapping and collisions, the atomic density and the background pressure in the vacuum system were
### Table 1. Published energy levels relevant to this work.

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<th>Wood et al.a</th>
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*a, b, c, d, e, f* Young et al. (1987)
Table 1. (Continued.)

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* Designation according to Wood and Andrew (1968).
* Energy levels measured with emission spectroscopy from a discharge by Wood and Andrew (1968).
* Energy levels measured with three-photon excitation by Young et al (1980).
* The ‘best’ values we suggest.

---

Figure 3. Typical fluorescence signal with Zeeman quantum beats.

The measured lifetime values changed by one order of magnitude, and no systematic effects on the evaluated lifetime values were found. For highly excited Rydberg states, it has been reported that the blackbody radiation has a substantial effect on the radiative lifetimes (Gallagher and Cooke 1979, Theodosiou 1984, Vidolova-Angelova et al 1989, Galvez et al 1995). The present lifetime measurements were performed at room temperature, and the experimental results should be regarded as the room temperature lifetime values. A crucial point in a transient recording experiment is to ensure that the detector has a linear response. In the present experiment, the number of photons recorded in each transient was small enough to exclude the influence of a nonlinear effect of the detector. Four thousand shots have been accumulated for each recording to make a smooth curve. For each state, about 20 curves have been recorded for different experimental conditions. The mean values were taken as the final results. The measured lifetime values...
Table 2. Lifetime and $g_f$ factor results for states in neutral lead atoms.

<table>
<thead>
<tr>
<th>Levela</th>
<th>Laser λ2 (nm)vac</th>
<th>Lifetimes (ns)</th>
<th>$g_f$ factors</th>
</tr>
</thead>
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<tr>
<td></td>
<td></td>
<td>This work Exp.</td>
<td>Previous</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7p(1/2, 1/2)1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>7p(1/2, 1/2)0</td>
<td>—</td>
<td>—</td>
<td>52.85b</td>
</tr>
<tr>
<td>7p(1/2, 3/2)1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>7p(1/2, 3/2)2</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>8p(1/2, 1/2)1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>8p(1/2, 1/2)0</td>
<td>—</td>
<td>—</td>
<td>111.85b, 130c</td>
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<tr>
<td>8p(1/2, 3/2)1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>8p(1/2, 3/2)2</td>
<td>—</td>
<td>—</td>
<td>131.4c, 122(8)c</td>
</tr>
<tr>
<td>5f 1/2[5/2]1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>9p(1/2, 1/2)1</td>
<td>516.36</td>
<td>449(23)</td>
<td>583.4b</td>
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<tr>
<td>9p(1/2, 1/2)0</td>
<td>510.87</td>
<td>274(12)</td>
<td>198.7b</td>
</tr>
<tr>
<td>9p(1/2, 3/2)1</td>
<td>509.14</td>
<td>237(11)</td>
<td>299.8b, 250(20)c</td>
</tr>
<tr>
<td>9p(1/2, 3/2)2</td>
<td>509.09</td>
<td>240(10)</td>
<td>292b</td>
</tr>
<tr>
<td>6f 1/2[5/2]1</td>
<td>498.19</td>
<td>104(8)</td>
<td>110.1b</td>
</tr>
<tr>
<td>10p(1/2, 1/2)1</td>
<td>475.02</td>
<td>778(70)</td>
<td>976b</td>
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<tr>
<td>10p(1/2, 1/2)0</td>
<td>472.50</td>
<td>430(18)</td>
<td>325.6b</td>
</tr>
<tr>
<td>10p(1/2, 3/2)1</td>
<td>472.13</td>
<td>335(10)</td>
<td>405.0b</td>
</tr>
<tr>
<td>10p(1/2, 3/2)2</td>
<td>471.97</td>
<td>380(15)</td>
<td>373.7b</td>
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<tr>
<td>7f 1/2[5/2]1</td>
<td>466.58</td>
<td>155(6)</td>
<td>—</td>
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<td>7p 3/2, 1/2</td>
<td>460.35</td>
<td>55(3)</td>
<td>—</td>
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<tr>
<td>11p(1/2, 3/2)1</td>
<td>—</td>
<td>—</td>
<td>181.7b</td>
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<tr>
<td>11p(1/2, 1/2)0</td>
<td>453.91</td>
<td>980(80)</td>
<td>1271.0b</td>
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<tr>
<td>11p(1/2, 1/2)0</td>
<td>452.62</td>
<td>692(30)</td>
<td>485.6b</td>
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<tr>
<td>11p(1/2, 3/2)1</td>
<td>451.62</td>
<td>370(18)</td>
<td>170.5b</td>
</tr>
<tr>
<td>8f 1/2[5/2]1</td>
<td>450.24</td>
<td>158(5)</td>
<td>—</td>
</tr>
<tr>
<td>7p(3/2, 1/2)2</td>
<td>448.20</td>
<td>146(7)</td>
<td>—</td>
</tr>
<tr>
<td>12p(1/2, 1/2)1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>12p(1/2, 1/2)0</td>
<td>440.81</td>
<td>930(90)</td>
<td>—</td>
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<tr>
<td>12p(1/2, 3/2)1</td>
<td>440.33</td>
<td>650(50)</td>
<td>—</td>
</tr>
<tr>
<td>12p(1/2, 3/2)2</td>
<td>440.03</td>
<td>490(20)</td>
<td>—</td>
</tr>
<tr>
<td>9f 1/2[5/2]1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>7p(3/2, 3/2)1</td>
<td>434.03</td>
<td>103(5)</td>
<td>—</td>
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<tr>
<td>13p(1/2, 3/2)1</td>
<td>432.75</td>
<td>750(100)</td>
<td>—</td>
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<tr>
<td>13p(1/2, 1/2)1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>13p(1/2, 1/2)0</td>
<td>433.19</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>13p(1/2, 3/2)1</td>
<td>432.52</td>
<td>100(7)</td>
<td>—</td>
</tr>
<tr>
<td>10f 1/2[5/2]1</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>14p(1/2, 1/2)1</td>
<td>—</td>
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<td>—</td>
</tr>
<tr>
<td>14p(1/2, 1/2)0</td>
<td>—</td>
<td>—</td>
<td>—</td>
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<tr>
<td>14p(1/2, 3/2)1</td>
<td>427.67</td>
<td>1200</td>
<td>—</td>
</tr>
<tr>
<td>14p(1/2, 3/2)2</td>
<td>427.72</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>11f 1/2[5/2]1</td>
<td>—</td>
<td>—</td>
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are listed in Table 2 together with those previously known for comparison. The quoted error bars reflected the statistical scattering of the results from different measurements and a small allowance for residual systematic errors. For the levels 14p(1/2, 3/2)_1 and 15p(1/2, 3/2)_1, no error bars are given since the fluorescence signals are not strong enough to check effectively for systematic errors. The signals for the levels 14p(1/2, 3/2)_2, 15p(1/2, 3/2)_2, 16p(1/2, 3/2)_1 and 16p(1/2, 3/2)_2 are even weaker, so only \( g_J \) factor values were measured.

<table>
<thead>
<tr>
<th>Level(^a)</th>
<th>( \lambda_J ) (nm)</th>
<th>Lifetimes (ns)</th>
<th>( g_J ) factors</th>
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<tr>
<td></td>
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<td>Previous Cal.</td>
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<td>15p(1/2, 1/2(_1))</td>
<td>—</td>
<td>—</td>
<td>0.6667</td>
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<tr>
<td>15p(1/2, 3/2(_2))</td>
<td>424.40</td>
<td>—</td>
<td>1.1798(20)</td>
</tr>
<tr>
<td>15p(1/2, 1/2(_0))</td>
<td>—</td>
<td>—</td>
<td>1.5133(9)</td>
</tr>
<tr>
<td>15p(1/2, 3/2(_1))</td>
<td>423.99</td>
<td>1300</td>
<td>1.5000</td>
</tr>
<tr>
<td>12f 1/2[5/2(_2)]</td>
<td>—</td>
<td>—</td>
<td>0.8889</td>
</tr>
<tr>
<td>7p(3/2, 3/2(_2))</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>16p(1/2, 1/2(_1))</td>
<td>—</td>
<td>—</td>
<td>0.6667</td>
</tr>
<tr>
<td>16p(1/2, 1/2(_0))</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>16p(1/2, 3/2(_1))</td>
<td>421.25</td>
<td>—</td>
<td>1.5114(20)</td>
</tr>
<tr>
<td>16p(1/2, 3/2(_2))</td>
<td>420.97</td>
<td>—</td>
<td>1.0936(50)</td>
</tr>
</tbody>
</table>

\( g_J \) factors are computed from the lifetime data.

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**Table 2.** (Continued.)

**Figure 4.** Typical fluorescence signal with Zeeman quantum beats being washed out by a strong magnetic field.
4. Discussion

The theoretical $g_J$ values for the levels were calculated according to their $J$ values and designations. As can be seen in table 2, the calculations are quite close to the measured values, which indicates that the assigned coupling schemes to each channel are quite good. For pure $jj$ coupling the $g_J$ value of 6pnp(1/2,3/2)$_1$ is calculated to be 1.5. Our experimental $g_J$ values as a function of the effective principal quantum number $n^*$ are shown in figure 5. It is evident that the $g_J$ factors have been perturbed by the levels 6pnp(3/2,1/2)$_1$ and 6pnp(3/2,3/2)$_1$ and the same effect can also be seen for the lifetime values shown in figure 6. The MQDT analysis for this aspect is in progress.

It should be mentioned that the $g_J$ value of the 6p9p(1/2,3/2)$_2$ level given by Wood et al. (1968) is 0.89(10), while the present measurement gives a value of 1.16(10). As the 6p9p(1/2,3/2)$_2$ level is close to the level 6p6f1/2[5/2]$_2$, which has a $g_J$ factor measured to be 0.8901(30), it is reasonable to believe that it is a misinterpretation of the energy levels by Wood et al that causes the disagreement.

The available energy level positions from five papers are compiled in table 1. Some results do not agree with each other. According to our $g_J$ and wavelength values of the second step excitation, we reassigned some levels. A few points should be noted:

1. The energy level 6p11p(1/2,3/2)$_2$ determined by Farooqi et al (1995) as 57 318.71 cm$^{-1}$, should be denoted as 6p11p(1/2,1/2)$_1$, which is in agreement with the assignment by Ding et al (1989).
Figure 6. The experimental lifetime values versus effective principal quantum number $n^*$. The symbols —— represent the 6pnp ($1/2, 3/2)_1$ Rydberg series. The perturber levels 6p7p ($3/2, 1/2)_1$ and 6p7p ($3/2, 3/2)_1$ are represented by • and ▲.

(2) The energy of the 6p12p ($1/2, 1/2)_0$ level determined by Wood and Andrew (1968), Ding et al (1989) and Farooqi et al (1995) agrees mutually within 0.2 cm$^{-1}$ and the average of the results is 57 972.10 cm$^{-1}$, which is also supported by our measurements. The value for the same level given by Hasegawa and Suzuki (1996) is 57 968.05 cm$^{-1}$, but the reason for this disagreement is not clear.

(3) Wood and Andrew (1968) measured a level at 58 368.91 cm$^{-1}$ and they assigned it to be 6p13p ($1/2, 3/2)_2$. According to our $g_J$ value of this level, it should belong to 6p13p ($1/2, 1/2)_1$, which is confirmed by others (Ding et al 1989, Farooqi et al 1995, Hasegawa et al 1996).

(4) In the last column of table 1, ‘best’ energy level values are suggested by the authors, which are the mean values of the results compiled in the table except in the doubtful cases which have been pointed out above.

Acknowledgments

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References

Aymar M, Greene C H and Luc-Koenig E 1996 Rev. Mod. Phys. 68 1015