Spectroscopic analysis of erbium-doped silicon and ytterbium-doped indium phosphide

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Chapter 6

Zeeman studies of the 4f intrashell transitions of ytterbium in indium phosphide

Abstract

Zeeman measurements in magnetic fields up to 16 T have been performed on the no-phonon lines labelled #3, #4 and #8 of the spectrum of ytterbium impurities in indium phosphide. The luminescence lines show well-observable splittings and a strong polarization effect and changes in intensities were observed. Also a complete change of the spectrum after some time of illumination was detected. An energy level diagram is presented that satisfactorily explains the magnetic field effect and the relative intensities of the photoluminescence lines and is consistent with experiments described in the literature.

6.1 Introduction

Ytterbium in indium phosphide is one of the best-investigated systems of this kind [6.1–6.7]. Ytterbium in its trivalent state, Yb\(^{3+}\) (4f\(^{13}\)), most probably placed substitutionally on an indium place in the InP lattice, always shows a characteristic luminescence spectrum. This is presented in figure 5.3 for temperatures of 4 and 40 kelvin. The lines with labels #2, #3, #4 and #8, at 10064, 10018, 9982.5 and 9920.5 cm\(^{-1}\) or 993.6, 998.2, 1001.8 and 1008.0 nm, respectively, are usually interpreted as zero-phonon transitions. The origin of other lines present in the spectrum, #5 – #7 and #9, is not completely established; they are described as phonon replica's [6.3] or assigned to a non-cubic centre [6.6]. Line #1 is attributed to a trigonal Yb\(^{3+}\)In – X\(_{p}\) centre [6.3]. The zero-phonon lines were shown to result from a cubic defect centre and arise from transitions between the spin–orbit levels \(^2F_{5/2}\) and \(^2F_{7/2}\). Due to the cubic crystal field the lower \(^2F_{7/2}\) level is split in three sublevels, of \(\Gamma_6\), \(\Gamma_7\) and \(\Gamma_8\).
symmetry type, respectively, and the higher $^2F_{5/2}$ level into two sublevels, with $\Gamma_7$ and $\Gamma_8$ symmetry label. Zeeman measurements [6.3], which show an eightfold splitting of line #3 in high magnetic fields, confirm that the origin of lines #2, #3, #4 and #8 is a cubic Yb centre. Following this study an energy level diagram for the $4f$ sublevels of the Yb$^{3+}$ ion, as shown in figure 6.1, has been established. This scheme displays an unusual reversal of the $\Gamma_7$ and $\Gamma_8$ sublevels at the excited state, which cannot be explained by the crystal-field calculations [6.7], and is in contrast to the predictions of a point charge model for an ytterbium atom on a substitutional cation site [6.8]. Furthermore, in this scheme one cannot explain the relative intensities of the lines #3, #4 and #8 in photoluminescence and the surprising increase of the luminescence intensity of line #3 observed for high magnetic fields, as will be discussed in section 6.5.

In chapter 5 the intensity, temperature and stress dependence of the luminescence spectrum and the EPR spectrum are discussed and it is concluded that two models can best explain all these effects, these are also depicted in figure 6.1.

![Energy level diagram of InP:Yb with level assignment according to Masterov et al. [6.1], Aszodi et al. [6.3], and model 1 of chapter 5. The zero-phonon transitions observed in the photoluminescence spectra are indicated.](image)

Figure 6.1 The energy level diagram of InP:Yb with the level assignment according to Masterov et al. [6.1], Aszodi et al. [6.3], and model 1 of chapter 5. The zero-phonon transitions observed in the photoluminescence spectra are indicated.
6.2 Experimental method

Two samples, kindly provided by F. Scholz and B. Lambert, were used in this study. One crystal has been grown by metal-organic chemical vapour deposition (MOCVD). The total ytterbium concentration was $10^{18}$ atoms/cm$^3$. The measured surface was a <100> plane. The other crystal has been grown by the high-pressure gradient freeze synthesis method. By this method, ytterbium was diluted in indium phosphide with a concentration of about $10^{17}$ atoms/cm$^3$. The measured surface of this sample was off-axis oriented. Results of examining the sample by x-ray are shown in figure 6.2.

![Laue patterns of the two indium phosphide samples](image1)

**Figure 6.2** The Laue patterns of the two indium phosphide samples. The left pattern is nearly invariant under a rotation of $2\pi/4$ following from the four-fold symmetry of indium phosphide about a [100] axis. The right one shows that the crystal is multi-crystalline and not aligned in a main direction.

6.3 Theoretical analysis of the Zeeman splitting

Ytterbium in the 3+ charge state, missing one electron in the 4f-shell, has electronic configuration $4f^{13}$ resulting in orbital moment $L = 3$ and spin $S = 1/2$. Spin–orbit interaction leads to two states: the $^2F_{5/2}$ state with $J = 5/2$ and sixfold degeneracy, and the $^2F_{7/2}$ state with $J = 7/2$ and eightfold degeneracy. In a cubic crystal field the degeneracy is partially lifted. The ground state $^2F_{7/2}$ splits in a doublet $\Gamma_6$, a doublet $\Gamma_7$ and a quartet $\Gamma_8$; the excited state $^2F_{5/2}$ splits in a doublet $\Gamma_7$ and a quartet $\Gamma_8$. In a magnetic field the remaining degeneracy is lifted completely. For the sextet the magnetic quantum number $m_j$ can have the following values: $+5/2$, $+3/2$, $+1/2$, $-1/2$, $-3/2$ and $-5/2$; these 6 states are called $|5/2, +5/2>$, $|5/2, +3/2>$,
The magnetic moment in the direction of the magnetic field (z) is $g \mu_B$, with $\mu_B$ the Bohr-magneton, $\mu_B = e \hbar / 2m_e$, and $g$ depending on $g_L$ and $g_s$, by

$$g = \frac{1}{2} \left( g_s + g_L \right) - \frac{L(L+1) - S(S+1)}{2J(J+1)} (6.1)$$

For $g_s = 2$ and $g_L = 1$ this formula transforms to the well-known formula for the $g$ value of Landé:

$$g = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} (6.2)$$

The Landé formula gives $g = 6/7$ for the $^2F_{5/2}$ state, and $g = 8/7$ for the $^2F_{7/2}$ state [6.9]. The splitting in a small magnetic field is summarised in table 6.1.

**Table 6.1 Sublevels, eigenvectors and magnetic properties of the $^2F_{5/2}$ and $^2F_{7/2}$ states in a cubic crystal field. The magnetic energy ($\Delta E$) of the sublevels, in a small field, is given in cm$^{-1}$/T. $m$ is the average value of the magnetic quantum value $m_J$ and $\mu$ is defined as equal to $m_J$ with the lowest (absolute) value.**

<table>
<thead>
<tr>
<th>Level</th>
<th>Sublevel</th>
<th>Eigenvector</th>
<th>$\mu$</th>
<th>$m$</th>
<th>$\Delta E$ (cm$^{-1}$/T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^2F_{5/2}$</td>
<td>$\Gamma_7a$</td>
<td>$\frac{1}{6} \sqrt{30}</td>
<td>5/2,+5/2&gt; - (1/6) \sqrt{30}</td>
<td>5/2,-3/2&gt;$</td>
<td>$-3/2$</td>
</tr>
<tr>
<td></td>
<td>$\Gamma_7b$</td>
<td>$\frac{1}{6} \sqrt{30}</td>
<td>5/2,-5/2&gt; - (1/6) \sqrt{30}</td>
<td>5/2,+3/2&gt;$</td>
<td>$+3/2$</td>
</tr>
<tr>
<td></td>
<td>$\Gamma_8a$</td>
<td>$\sqrt{30}</td>
<td>5/2,+5/2&gt; + (1/6) \sqrt{6}</td>
<td>5/2,-3/2&gt;$</td>
<td>$-3/2$</td>
</tr>
<tr>
<td></td>
<td>$\Gamma_8b$</td>
<td>$\sqrt{30}</td>
<td>5/2,-5/2&gt; + (1/6) \sqrt{6}</td>
<td>5/2,+3/2&gt;$</td>
<td>$+3/2$</td>
</tr>
<tr>
<td></td>
<td>$\Gamma_8c$</td>
<td>$</td>
<td>5/2,+1/2&gt;$</td>
<td>$+1/2$</td>
<td>$+1/2$</td>
</tr>
<tr>
<td></td>
<td>$\Gamma_8d$</td>
<td>$</td>
<td>5/2,-1/2&gt;$</td>
<td>$-1/2$</td>
<td>$-1/2$</td>
</tr>
<tr>
<td>Level</td>
<td>Sub-level</td>
<td>Eigenvector</td>
<td>(\mu)</td>
<td>(m)</td>
<td>(\Delta E) (cm(^{-1}/T))</td>
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</tr>
<tr>
<td>(^2F_{7/2})</td>
<td>(\Gamma_{6a})</td>
<td>((1/6)\sqrt{15}</td>
<td>7/2, +7/2&gt; + (1/6) \sqrt{21}</td>
<td>7/2, -1/2&gt;)</td>
<td>-1/2</td>
</tr>
<tr>
<td></td>
<td>(\Gamma_{6b})</td>
<td>((1/6)\sqrt{15}</td>
<td>7/2, -7/2&gt; + (1/6) \sqrt{21}</td>
<td>7/2, +1/2&gt;)</td>
<td>+1/2</td>
</tr>
<tr>
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<td>(\Gamma_{7a})</td>
<td>((1/2) \sqrt{3}</td>
<td>7/2, +5/2&gt; - (1/2)</td>
<td>7/2, -3/2&gt;)</td>
<td>-3/2</td>
</tr>
<tr>
<td></td>
<td>(\Gamma_{7b})</td>
<td>((1/2) \sqrt{3}</td>
<td>7/2, -5/2&gt; - (1/2)</td>
<td>7/2, +3/2&gt;)</td>
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<td></td>
<td>(\Gamma_{8a})</td>
<td>((1/6)\sqrt{21}</td>
<td>7/2, +7/2&gt; - (1/6) \sqrt{15}</td>
<td>7/2, -1/2&gt;)</td>
<td>-1/2</td>
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<tr>
<td></td>
<td>(\Gamma_{8b})</td>
<td>((1/6)\sqrt{21}</td>
<td>7/2, -7/2&gt; - (1/6) \sqrt{15}</td>
<td>7/2, +1/2&gt;)</td>
<td>+1/2</td>
</tr>
<tr>
<td></td>
<td>(\Gamma_{8c})</td>
<td>((1/2)</td>
<td>7/2, +5/2&gt; + (1/2) \sqrt{3}</td>
<td>7/2, -3/2&gt;)</td>
<td>-3/2</td>
</tr>
<tr>
<td></td>
<td>(\Gamma_{8d})</td>
<td>((1/2)</td>
<td>7/2, -5/2&gt; + (1/2) \sqrt{3}</td>
<td>7/2, +3/2&gt;)</td>
<td>+3/2</td>
</tr>
</tbody>
</table>

When a magnetic field along the \(z\) axis is applied, in the limit of a weak field (the Zeeman splitting being small compared to the crystal-field splitting), the energy shifts in a field \(B\) are given by \(\Delta E = g\mu_B B\), and \(m\) the average value of the magnetic quantum value \(m_J\). The \(\Gamma_7\) level will split into two and the \(\Gamma_8\) level into four, non-equidistant, lines. The parameter \(\mu\) is defined as equal to \(m_J\) with the lowest (absolute) value. The parameter \(\mu\) is also given in figure 6.3 for the different sublevels.

In a strong magnetic field the Zeeman energy is no longer small compared to the crystal field splitting. The magnetic energy, \(g\mu_B B\), is added to the diagonal elements of the corresponding matrix. The eigenvalues of this matrix supply the shifts of the energies.

When an atom is placed in a cubic crystal field with a strong magnetic field in a random direction the treatment becomes more complicated.

For the experiments considered in this chapter, the expected maximum Zeeman splitting for the \(\Gamma_{8a} - \Gamma_{8b}\) states in the field \(B = 16\ T\) is \(= 16\ \text{cm}^{-1}\) which is still small compared to the \(F_{5/2} - F_{7/2}\) spin–orbit splitting, \(= 10,000\ \text{cm}^{-1}\).

In this case the energy levels in one multiplet are calculated with the complete Hamiltonian of the crystal field plus the magnetic field:

\[
H = \frac{WxO_4}{F(4)} + \frac{W(1-|x|)}{F(6)} + g\mu_B B. \quad (6.3)
\]

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The vector $\mathbf{B}$ will be $B(\alpha_1 \mathbf{e}_x + \alpha_2 \mathbf{e}_y + \alpha_3 \mathbf{e}_z)$, with $(\alpha_1 \mathbf{e}_x + \alpha_2 \mathbf{e}_y + \alpha_3 \mathbf{e}_z)$ a unit vector, $\alpha_1$, $\alpha_2$, $\alpha_3$ are the cosines of the direction of the field. The complete Hamiltonian is now

$$H = \frac{W_x O_4}{F(4)} + \frac{W(1 - |x|)O_6}{F(6)} + g\mu_B B(\alpha_1 m_x + \alpha_2 m_y + \alpha_3 m_z).$$

Figure 6.3 A possible energy level diagram of Yb$^{3+}$ in InP in a magnetic field with the value of $\mu$ given for all the sublevels.
Not only $O_4$ and $O_6$ but also $m_x$, $m_y$ and $m_z$ are operators represented by matrices. $m_z$ is a matrix with only diagonal elements with the value of $m$. $m_x$ and $m_y$ are matrices with only values unequal to zero next to the diagonal with the values:

$$m_z(m_{-1}, m) = -m_z(m_{m}, m) = \frac{1}{2} \sqrt{(J - m)(J + m + 1)}.$$  \hspace{1cm} (6.5)

$$m_y(m_{-1}, m) = -m_y(m_{m}, m) = -\frac{i}{2} \sqrt{(J - m)(J + m + 1)}.$$  \hspace{1cm} (6.6)

So for $J = 5/2$ these matrices will be:

$$m_z = \frac{1}{2} \begin{pmatrix}
0 & \sqrt{5} & 0 & 0 & 0 & 0 \\
\sqrt{5} & 0 & 2\sqrt{2} & 0 & 0 & 0 \\
0 & 2\sqrt{2} & 0 & 3 & 0 & 0 \\
0 & 0 & 3 & 0 & 2\sqrt{2} & 0 \\
0 & 0 & 0 & 2\sqrt{2} & 0 & \sqrt{5} \\
0 & 0 & 0 & 0 & \sqrt{5} & 0
\end{pmatrix}$$  \hspace{1cm} (6.7)

$$m_y = \frac{i}{2} \begin{pmatrix}
0 & -\sqrt{5} & 0 & 0 & 0 & 0 \\
\sqrt{5} & 0 & -2\sqrt{2} & 0 & 0 & 0 \\
0 & 2\sqrt{2} & 0 & -3 & 0 & 0 \\
0 & 0 & 3 & 0 & -2\sqrt{2} & 0 \\
0 & 0 & 0 & 2\sqrt{2} & 0 & -\sqrt{5} \\
0 & 0 & 0 & 0 & 0 & \sqrt{5}
\end{pmatrix}$$  \hspace{1cm} (6.8)

$$m_x = \frac{1}{2} \begin{pmatrix}
-5 & 0 & 0 & 0 & 0 & 0 \\
0 & -3 & 0 & 0 & 0 & 0 \\
0 & 0 & -1 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & 3 & 0 \\
0 & 0 & 0 & 0 & 0 & 5
\end{pmatrix}$$  \hspace{1cm} (6.9)
The splitting of the Zeeman components shows different behaviour as a function of the intensity of the magnetic field in three field regions. In low fields, where the Zeeman splitting is small compared to the splitting due to the crystal field, the splitting is nearly proportional to $B$, and independent of the magnitude of the crystal field. In the $\Gamma_6$ and $\Gamma_7$ doublets, this splitting is isotropic (independent of the direction of the field with respect to the crystal axes); in the $\Gamma_8$ quartet the splitting is, however, anisotropic.

In fields where the Zeeman splitting is comparable to that of the crystal field, there occurs a mixing of states with equal values of $\mu$ belonging to different crystal-field sublevels; as a consequence the splitting depends on the magnitude of the crystal field, deviates considerably from proportionality to $B$, and is always anisotropic. In still higher fields, where the Zeeman splitting is larger than the crystal field splitting, the Zeeman splitting becomes again proportional to $B$ and isotropic, and approaches that of the undisturbed atom.

### 6.4 Selection rules and consequences

Transitions between these Zeeman levels are subject to selection rules with the general form $\Delta \mu = 0, +/-1$. For electric dipole transitions with $\Delta \mu = 0$ the emitted light will be linearly polarized perpendicular to the magnetic field. For transitions with $\Delta \mu = +/-1$ circularly polarized light will be emitted parallel or antiparallel to the magnetic field. Such transitions have been observed in the present experiment. For the magnetic dipole transition then holds $\Delta \mu = 0$, and since the electric dipole intrashell transitions are already forbidden, magnetic dipole transitions can be of the same intensity or maybe even stronger than the electric dipole transitions [6.9].

From this analysis it follows that for a magnetic field in a non-high-symmetry direction no transitions will be forbidden by the selection rules. This can lead to doubling of the number of lines compared to the $<100>$ direction. For a $<111>$ direction again some selection rules become effective. The general rule is, the higher the symmetry, the lower the number of allowed transitions. The dependence of the sublevels on the direction of the magnetic field is given in figure 6.4. It can be observed that $\Gamma_7$ and $\Gamma_6$ show hardly any directional dependencies but $\Gamma_8$ shows a contraction at the $<111>$ direction. Subsequently it should be investigated what to expect for the different transitions of the tetrahedral Yb defect in a magnetic field parallel to the $<100>$ direction.

The values for $\mu$ are given in figure 6.3. One should take into account that $\Delta \mu$ has modulus 4, so therefore $\Delta \mu = 3$ is equivalent to $\Delta \mu = -1$. 

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The theoretical angular dependence of the energy levels of model 4 upon rotating a magnetic field of 5.3 tesla in a (110) plane of the crystal.

The calculated levels versus field are given in figure 6.5, for the lines of the lowest level of the excited state to the ground states: the "cold" lines. As expected the levels are seen to bend away from each other.

The following transitions can be expected in the ytterbium system:

- The $\Gamma_7 \rightarrow \Gamma_7$ transition has four possible transitions: one with $\Delta\mu = +1$, one with $\Delta\mu = -1$ and two with $\Delta\mu = 0$, the allowed transitions are the inner ones.
- The $\Gamma_7 \rightarrow \Gamma_6$ transition has also two allowed transitions $\Delta\mu = +/-1$ but they are the outer ones and the other two have $\Delta\mu = 2$.
- The $\Gamma_7 \rightarrow \Gamma_8$ transition and the $\Gamma_8 \rightarrow \Gamma_7$ transition have eight transitions: two $\Delta\mu = 0$, two $\Delta\mu = 2$ and four allowed transitions, the allowed transitions are the inner ones.
- The $\Gamma_8 \rightarrow \Gamma_6$ transition has the same kind of transitions as $\Gamma_8 \rightarrow \Gamma_7$ only the allowed ones are the outer ones.
- The $\Gamma_8 \rightarrow \Gamma_8$ transition has four $\Delta\mu = 2$, four $\Delta\mu = 0$ and eight allowed transitions and the allowed ones are the inner ones.
Figure 6.5 The calculated energy levels versus the strength of the magnetic field. For model 1 the lines arise for #3 from a $\Gamma_7$ to $\Gamma_7$ transition, for #4 from a $\Gamma_7$ to $\Gamma_8$ transition and for #8 from a $\Gamma_7$ to $\Gamma_6$ transition. The forbidden transitions are given with the dashed lines.
6.5 Experimental results and discussion

We have measured the Zeeman effect at lines #3, #4 and #8 in magnetic fields up to 16 T, at temperatures of 30 K, 4 K and some also at 1.8 K. In one specimen the field was directed along a <100> direction, in the other along an arbitrary crystallographic direction; always only the light emerging parallel to the field was observed. In order to find the polarization state, a λ/4 polarizing filter was inserted in the light beam and the signal was optimized. The other polarization state was detected by reversing the direction of the field (this being easier than changing the filter); arbitrarily the first measured direction, giving the strongest signal, will be referred to as positive.

Unfortunately the Zeeman-splitted lines could, due to experimental difficulties, only be detected when the sample was irradiated with a very high intensity of laser light. It was observed that, after having performed measurements for some time, the spectrum of the returned light changed considerably, see figure 6.6; we will call the original condition state I, and the new condition state II.

Figure 6.6 The Zeeman spectra of the two different states that could be observed in the InP:Yb samples at a magnetic field of 12 T immediately (I) and after a few hours of laser illumination (II).
6.5.1 State I

It was observed that some lines in one Zeeman multiplet increase in intensity at the cost of the other lines, if the magnetic field is increased above about 4 T, see figure 6.7 and 6.8. This can be understood, since in that case the Zeeman splitting of the lowest sublevel of the excited state, from which all these transitions originate, is no longer small compared to $kT$. As the lifetime of this excited state is long enough to establish some thermal equilibrium, the occupation of the lowest Zeeman level will increase at the cost of the other Zeeman level(s).

![The Zeeman spectrum of line #3 to #8 at, from bottom to top, 8, 10, 12, 14 and 16 tesla in the positive polarization direction in state I of the off-axis sample at 4.2 K.](image)

Figure 6.7 The Zeeman spectrum of line #3 to #8 at, from bottom to top, 8, 10, 12, 14 and 16 tesla in the positive polarization direction in state I of the off-axis sample at 4.2 K.
It was seen that in every case exactly half of the multiplet lines at the low-energy side increase in intensity (by about the same amount), while the other half decreases, see figure 6.8. This can only be explained if the lowest sublevel of the excited state splits into a Zeeman doublet; it should therefore be the $\Gamma_7$ level, in agreement with our earlier conclusion.

Due to the low intensity of line #3, the Zeeman splitting of this line could not be observed; at low field this line was invisible, whereas at a higher field only one Zeeman component became visible in positive polarization, see figure 6.7. The slope of the field dependence agrees well with the theoretical expectations, as can be seen comparing figure 6.5 with figure 6.9.

Line #4 was seen to split into 6 Zeeman components if the field is in an off-axis direction; when it is applied in a $<100>$ direction 6 components remain visible, not coinciding with the original ones, indicating a strong anisotropy of the splitting.

![Figure 6.8](image)

**Figure 6.8** The intensity versus the magnetic field of the positive (full line) and the negative (dashed line) components of lines #3, #4 and #8.
Figure 6.9 The positions of the lines versus the magnetic field for line #3, #4 and #8 in state I. The <100> oriented sample is indicated by a cross and the off-axis sample with a diamond.

Details of the magnetic field splitting of line #4 are depicted in figures 6.9 and 6.10. In the off-axis oriented material again one strong line was seen in the positive polarisation and the other ones were of equal intensity up to 8 T. The positively polarised line increases in intensity by a factor of about two, in going from 0 to 16 T, the negatively polarised line
decreases by a factor ten. The anisotropy, intensity dependence and number of lines all indicate that line #4 must be $\Gamma_7 \Rightarrow \Gamma_8$ transition which has 8 lines, all other possible transitions from a $\Gamma_7$ level show less lines.

Line #8 splits into 4 Zeeman components with an off-axis field, with the field in a $<100>$ direction the both innermost components become no longer visible, whereas the both outermost ones remain at the same position. This is exactly the behaviour expected for a $\Gamma_7 \Rightarrow \Gamma_6$ transition (in a $\Gamma_7 \Rightarrow \Gamma_7$ transition the both outermost components should disappear with a $<100>$ field), see figure 6.5. Details of the splitting of the photoluminescence line

Figure 6.10 The different spectra in the positive (+) and negative (-) polarization direction are given at 10 and 16 tesla.
#8 in the magnetic field are given figure 6.9 and 6.10. The total splitting width is about 4 nm in a field of 16 T, so also the magnitude of the experimentally found Zeeman splitting agrees with the theoretical expectations for this transition, see figure 6.9, further supporting such an assignment. Additional features observed in this region probably originate from the lines #5 – #7. From the Zeeman splitting of the photoluminescence lines it can therefore be decided that model 1 of table 5.2 is the only possibility for the energy level assignment of Yb in InP.

This energy level assignment is in agreement with all the available data mentioned before, except for the earlier Zeeman measurements of Aszodi et al. [6.3]; they observed the Zeeman splitting of line #3 into 8 components showing a large anisotropy, which is incompatible with a $\Gamma_7 \Rightarrow \Gamma_7$ transition. We also observe a splitting into 8 components, but only in state II, see figure 6.11.

![Diagram](image)

Figure 6.11 *The new Zeeman splittings in the "line #3 region". Eight lines can be observed alternating two in the positive polarization direction and two in the negative at 16 tesla.*

### 6.5.2 State II

The spectrum in the line #3 region changes dramatically as the specimen transforms from state I to state II, see figure 6.6. In state II eight lines in the off-axis oriented crystal and
four lines in the <100> oriented crystal are observed, yielding a splitting with a total width of about 4 nm in a field of 16 T. Several features of the Zeeman spectrum of line #3 are illustrated in the figures 6.11 through 6.13. In all cases the intensity of all lines increases tremendously when the field is increased above 10 T. One line, observed in positive polarisation at the low-energy side, becomes dominant and is four times stronger than the other ones, which are of similar intensity. In the off-axis-oriented specimen strong circular polarisation of the light was detected, where adjacent lines were polarised in opposite sense. For lines #4 and #8 the changes are not so striking, see figure 6.14. Both states of the off-axis oriented material show slightly different spectra for line #4, but at a sufficiently high field always six lines appear.

Fig 6.12 The Zeeman lines in the line #3 region for magnetic field of 0, 4, 8, 12, and 16 tesla. Decreasing the field from 16 T to 0 T for some time at 4.2 K a small "rest" is observed.
The only difference between both states of the crystal for line #8 is that in state I the line at highest energy increases its intensity with increasing field, which does not happen in state II. The origin of the transition from state I to state II, and of the eight lines seen in state II in the "line #3 region", are still unclear. A $\Gamma_8 \Rightarrow \Gamma_7$ or $\Gamma_7 \Rightarrow \Gamma_8$ transition is expected to have four lines in the <100> direction and eight in the random direction. The lines in the <100> direction oriented material should correspond to the inner components and the other lines should appear at the outer side. In contrast to that, two lines are observed at the inner side and two lines at the outer side.

Besides, if it were a $\Gamma_8 \Rightarrow \Gamma_7$ or $\Gamma_7 \Rightarrow \Gamma_8$ transition there would be no reason that all the lines gain intensity, as experimentally found.

A $\Gamma_8 \Rightarrow \Gamma_8$ transition, the hot line #2', should have 8 and 16 lines in the two materials. It is possible that not all the lines are observed because of transition probabilities or lower occupation of the higher levels.

Fig 6.13 Line #3, #4 and #8 in state II at 14 T. The negative polarization direction is three times amplified.
This assignment agrees with the fact that the lines can only be observed in the higher magnetic field where the lowest sublevels of the highest excited state have sufficiently lowered their energy to become populated.

A $\Gamma_8 \Rightarrow \Gamma_8$ transition, however, cannot explain the positions of the lines. Also no lines were observed of the other hot line, #2. The fact that the lines of the <100> direction oriented and the off-axis oriented material coincide seems to indicate that there is no, anisotropic, $\Gamma_8$ involved.

The fact that both the positively and the negatively polarised lines grow with the increasing magnetic field indicates that not the same $\Gamma_7$ level is involved as for line #4 and #8. It is possible that the origin of these lines cannot be found in this energy level diagram, and that a different defect is involved.

The fact that the Zeeman lines can be found in the two samples with the same intensities relative to line #4 indicates that there can be a connection with the yet unexplained lines #5 – #7 and #9, which can always be found in the photoluminescence spectrum of every InP:Yb sample with the same relative intensities as lines #3, #4, and #8. Such a defect would have to be of a high symmetry, since one line is split into eight at least one $\Gamma_8$ level should be

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Figure 6.14 The positions of the Zeeman splittings of line "#3" versus the magnetic field in state II. The triangles indicated the position of the lines in the off-axis material, the crosses in the <100> directed material.
involved, and is therefore not related with line #1, the trigonally distorted Yb$^{3+}$In – X$_p$ centre [6.3].

6.6 Conclusion

From the Zeeman splitting of lines #3, #4 and #8 in state I it can be concluded that model no.1 is the correct energy level diagram. This assignment is in agreement with literature and the measurements described in chapter 5. Upon application of a high magnetic field and under laser radiation a transformation of two states has been observed. The nature of this reversible transformation and the nature of state II remain unclear at this moment.

References


