Line positions of a $\frac{3}{2} - \frac{1}{2}$ transition

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Contents

1 Abstract

The information carried by the positions of the absorption lines of an ^{57}Fe Mossbauer spectrum with combined electric and magnetic hyperfine interaction was a matter of concern of several theoretical papers [1], [2],[3],[4] (see also [5],[6],[7]).A Fortran routine which visualize the manifold of solutions of the ambiguity problem were written in a Diploma work in Erlangen [8].

2 Energy level scheme

The Energy level scheme of a $\frac{3}{2}-\frac{1}{2}$ transition is schown in Fig. 1 for ^{57}Fe. The 2 ground states are eigenstates of the I_z operator (z-axis choosen parallel to the magnetic field) with eigenvalues $\pm\mu_{1/2}H$ and eigenstates $|I = 1/2, \mp 1/2\rangle$ for $\mu_{1/2} = \mu_g > 0$, respectively, while the 4 exited states are of mixed type (linear combination of $|I = 3/2, m\rangle$ states) in the general case of combined electric and magnetic interaction. The eigenvalues $E_1, ..., E_4$ are obtained from the secular equation of a 4x4 Hermitian matrix. The 8 transitions are assigned as $L_1, ...L_8$ in Fig.1. In case of pure magnetic interaction the eigenstates are $|I = 3/2, +3/2\rangle, ..., |I = 3/2, -3/2\rangle$ fromE_1 to E_4 and the transitions $L_7 : -1/2 \rightarrow +3/2$ and $L_8 : +1/2 \rightarrow -3/2$ become forbidden.
The transition energies $E_r + L_i$ are the sum of the energy of the γ-quanta of the source E_r and the energy L_i provided by the Doppler velocity. If the difference in energy shifts by the electron densities at the nuclei of source and absorber, the isomer shift or

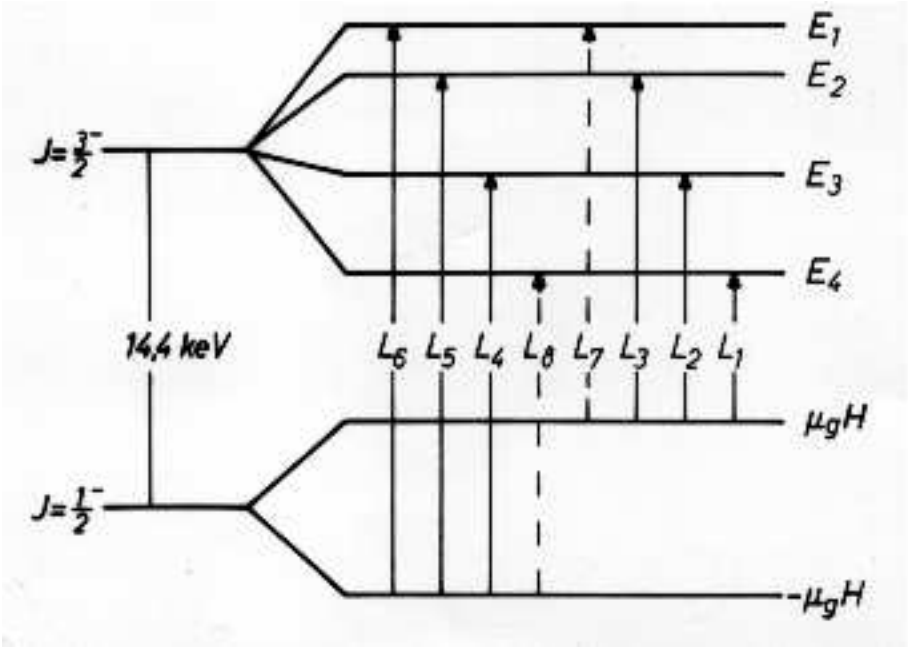

Figure 1: The $\frac{1}{2}-\frac{3}{2}$ 14.4keV transition of ^{57}Fe from the ground state to the exited state of an absorption experiment. The 8 transitions are assigned as $L_1, ...L_8$. The broken lines of L_7 and L_8 are forbidden transitions in case of pure magnetic interaction. The figure is taken from [8]

chemical shifts, is denoted by Is, the line positions are readily read from left to right hand side of Fig. 1.

$$\begin{aligned} L_6 &= Is + \mu_g H + E_1 \qquad L_7 = Is - \mu_g H + E_1 \\ L_5 &= Is + \mu_g H + E_2 \qquad L_3 = Is - \mu_g H + E_2 \\ L_4 &= Is + \mu_g H + E_3 \qquad L_2 = Is - \mu_g H + E_3 \\ L_8 &= Is + \mu_g H + E_4 \qquad L_1 = Is - \mu_g H + E_4 \end{aligned} \tag{1}$$

The center of gravity of the hyperfine energies E_i of nuclear Hamiltonians (see Eq. 5,6) of magnetic and electric quadrupole interactions is zero: $\sum_i E_i = 0$. This fact provides easy access to the isomer shift Is from the line postions L_i.

$$\begin{aligned} Is &= \frac{1}{8}\sum_{i=1}^{8} E_i \qquad and\ also \\ &= \frac{1}{4}\left(L_1 + L_2 + L_5 + L_6\right) \end{aligned} \tag{2}$$

The positions of the absorption lines of the Mossbauer spectrum are shown in Fig.2. $L = L_1$ and $R = L_6$ are always the outer lines in the spectrum. If the magnetic field interaction is large compared to the quadrupole interaction lines L_2 and L_5 are also readily identified and with them the isomer shift Is by Eq. 2.
The 8 line positions L_i dependent on 5 quantities ($\sum_i E_i = 0$ factored in) are related

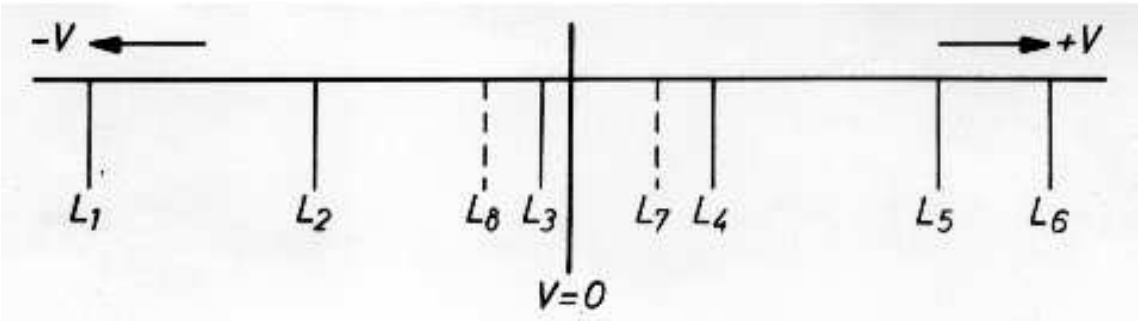

Figure 2: The $\frac{1}{2}-\frac{3}{2}$ 14.4keV transition of ^{57}Fe from the ground state to the exited state of an absorption experiment. The 8 line positions assigned as $L_1, ...L_8$ belong to the 8 transitions shown in Fig.1. The broken lines of L_7 and L_8 are forbidden transitions in case of pure magnetic interaction. The figure is taken from [8]

to each other as follows:

$$\begin{array}{llll} L_1 - Is = l & L_2 - Is = x & L_6 - Is = r & \\ L_3 - Is = -(r + l + x + 2\mu_g H) & L_4 - Is = x + 2\mu_g H & & \\ L_5 - Is = -(r + l + x) & L_7 - Is = r - 2\mu_g H & L_8 - Is = l + 2\mu_g H & \end{array} \tag{3}$$

These equations are used in the program **lines_alpl** in the directory effi/helproutines/ lineLR/lpos_LRIsDq, where alpl stands for the plot program **xalpl**, which plots a one parameter manifold of line positions together with an experimental spectrum in order to identify all 8 resonance lines.

3 Interrelations of the line positions

The interrelation of the line positions according to Eq. 3 are still a 2 parameter manifold if the positions of the outer lines are taken to be fixed. The solutions of the Hamiltonian are used to reduce the manifold of line positions, which are determined by 5 parameters, which are with respect to the principal axes system (PAS) of the electric field gradient (EFG) 3 parameters for the magnetic field $\mathbf{H}(\theta, \phi)$ and 2 for the EFG, $Deq = eQV_{zz}/4$ and $\eta = (V_{xx} - V_{yy})/V_{zz}$. The Hamiltonian H of the nuclear spin states with spin I reads

$$H_I = -\boldsymbol{\mu}_I \mathbf{H} + \frac{eQ_I V_{zz}}{4I(2I-1)} \left[3I_z^2 - I(I+1) + \frac{\eta}{2} \left(I_+^2 + I_-^2 \right) \right] \tag{4}$$

with the magnetic moment $\boldsymbol{\mu}_I = \mu_I \mathbf{I}/I$ and the quadrupole moment Q_I.
The magnetic field $\mathbf{H}$ defined with respect to the PAS of the EFG is given by $\mathbf{H}(\theta, \phi) = H\,(sin\theta cos\phi \mathbf{e}_x + sin\theta sin\phi \mathbf{e}_y + cos\theta \mathbf{e}_z)$. The spin Hamiltonian of the I=1/2 ground state is the 2x2 matrix:

$$H_{1/2}/(-\mu_{1/2}) = \begin{array}{c} \\ \langle +\frac{1}{2}| \\ \langle -\frac{1}{2}| \end{array} \begin{array}{c} \begin{array}{cc} |+\frac{1}{2}\rangle & |-\frac{1}{2}\rangle \end{array} \\ \begin{pmatrix} cos\theta & sin\theta e^{-i\phi} \\ sin\theta e^{i\phi} & -cos\theta \end{pmatrix} \end{array} \tag{5}$$

with eigenvalues $\pm\mu_{1/2} = \pm\mu_g(= 0.090604\mu_k)$ nuclear magnetons. The I=3/2 exited state is the 4x4 matrix:

$$H_{3/2}/m = \begin{array}{c} \\ \langle+\frac{3}{2}| \\ \langle+\frac{1}{2}| \\ \langle-\frac{1}{2}| \\ \langle-\frac{3}{2}| \end{array} \begin{array}{c} \begin{array}{cccc} |+\frac{3}{2}\rangle & |+\frac{1}{2}\rangle & |-\frac{1}{2}\rangle & |-\frac{3}{2}\rangle \end{array} \\ \begin{pmatrix} q+3cos\theta & \sqrt{3}sin\theta e^{-i\phi} & \frac{q\eta}{\sqrt{3}} & 0 \\ \sqrt{3}sin\theta e^{i\phi} & -q+cos\theta & 2sin\theta e^{-i\phi} & \frac{q\eta}{\sqrt{3}} \\ \frac{q\eta}{\sqrt{3}} & 2sin\theta e^{i\phi} & -q-cos\theta & \sqrt{3}sin\theta e^{-i\phi} \\ 0 & \frac{q\eta}{\sqrt{3}} & \sqrt{3}sin\theta e^{-i\phi} & q-3cos\theta \end{pmatrix} \end{array} \tag{6}$$

with $m = -\mu_e H/3$ and $q = eQV_{zz}/4/m$ (the magnetic moment $\mu_e = \mu_{3/2} = -0.15531\mu_k$ nuclear magnetons, the Quadrupole moment $Q = Q_{3/2} = 0.18 \cdot 10^{-24} cm^2$).

The characteristic polynomial $x^4 + A_3x^3 + A_2x^2 + A_1x + A_0 = 0$ for the eigenvalues $E = x \cdot m$ has 3 nonzero coefficients:

$$\begin{aligned} A_0 &= 9 - 2q^2(5-\eta^2) + q^4\left(1+\frac{\eta^2}{3}\right)^2 + 4q^2 sin^2\theta\left(3 + 2\eta cos2\phi - \eta^2\right) \\ A_1 &= 8q\left(sin^2\theta(3 - \eta cos2\phi) - 2\right) \\ A_2 &= -10 - 2q^2\left(1+\frac{\eta^2}{3}\right) \\ A_3 &= 0 \end{aligned} \tag{7}$$

The coefficients A_i depend on 4 parameters q, η, θ, and ϕ. Next it is shown, that the A_i can be determined from the line positions.

Viéte's formulas express the coefficients by the roots of the polynomial

$$\begin{aligned} A_0 &= x_1x_2x_3x_4 \qquad A_1 = -\sum_{\substack{i,j,k=1 \\ i<j<k}}^{4} x_ix_jx_k \\ A_2 &= \sum_{\substack{i,j=1 \\ i<j<}}^{4} x_ix_j \qquad A_3 = -\sum_{i=1}^{4} x_i \end{aligned} \tag{8}$$

which can be simplified by $A_3 = 0$ to

$$A_0 = E_1E_2E_3E_4/m^4 \qquad A_1 = -\frac{1}{3}\sum_{i=1}^{4}(E_i/m)^3 \qquad A_2 = -\frac{1}{3}\sum_{i=1}^{4}(E_i/m)^2 \tag{9}$$

Th eigenvalues E_i are connected to the line positions as directly read from Fig.1:

$$\begin{aligned} E_1 - E_2 &= L_6 - L_5 \\ E_2 - E_3 &= L_5 - L_4 = L_3 - L_2 \\ E_3 - E_4 &= L_2 - L_1 \\ 2\mu_g H &= L_5 - L_3 = L_4 - L_2 \end{aligned} \tag{10}$$

Using $A_3 = \sum_i E_i = 0$ and the abbreviations

$$\begin{aligned} c_1 &= L_6 - L_5 \qquad c_2 = L_2 - L_1 \\ d &= \frac{1}{2}\left((L_6 - L_1) - (L_4 - L_3)\right) \end{aligned} \tag{11}$$

the energies of the $J = 3/2$-state are expressed by line positions L_i as

$$\begin{aligned} E_1 &= (d + c_1)/2 \qquad E_2 = (d - c_1)/2 \\ E_3 &= (-d + c_1)/2 \qquad E_4 = (-d - c_1)/2 \end{aligned} \tag{12}$$

by which using Eq. 8 the coefficients A_i are calculated.

4 Range of line positions

If the isomer shift Is is known and the quadrupole interaction by the quarupole splitting ΔEq, which is the seperation of the two resonance lines with the magnetic interaction switched off, there is a one parameter manifold left for the line positions in between line numbers L_1 and L_6. In the program **lines_alpl** the position $x = L_2 - Is$ is taken as a parameter. Using Eqs. 9,12 the coefficient A_2 is expressed by the line positions as written by Eq. 3.

$$\sum_i E_i^2 = (r + l)^2 + (l + x)^2 + (r + x)^2 + 4(\mu_g H)^2 + 4(l + x)(\mu_g H) \tag{13}$$

On the other hand is the coefficient A_2 from Eq. 7 proportional to $\sum_i E_i^2$ such that

$$\sum_i E_i^2 = 20m^2 + 4q^2m^2\left(1 + \frac{\eta^2}{3}\right) \tag{14}$$

The second term of the sum is just the square $(\Delta Eq)^2$,of the quadrupole splitting. The last two equations can be resolved for $\mu_g H$

$$\begin{aligned} \mu_g H &= \frac{1}{2\kappa}\left((l + x) \pm \sqrt{(l + x)^2 + \kappa\alpha}\right) \\ \kappa &= \frac{5}{9}\left(\frac{\mu_e}{\mu_g}\right)^2 - 1 \\ \alpha &= (r + l)^2 + (l + x)^2 + (r + x)^2 - (\Delta Eq)^2 \end{aligned} \tag{15}$$

From the fact that $L_2 \leq L_5$ (see Fig. 1) the relation $x \leq -(l + r)/2$ is derived by Eq. 3 and in the same way from $L_8 \geq L_1$ the value of $\mu_g H$ of Eq. 15 always to be ≥ 0. These two lower bounds determine the sign of the square root in the equation for $\mu_g H$ to be the positive one since $\kappa = 1.2648376 > 0$ and $(x + l)$ comes out to be $\leq (l - r)/2 < 0$. The lower limit of x, $x \geq l$ and $x \geq -(2r + l)$, stems from $L_1 \leq L_2$ and $L_5 \leq L_6$, respectively.

Fig. 3 shows the manifold of line positions compatible with L_1 and L_6 and Eq. 15. The positions were taken from a simulated spectrum with $\mathbf{H}_i(\theta = 75, \phi = 25)$, $H_i = 330\,kG$, $\eta = 0.9$, $Deq = -4.0\,mm/s$, and $Is = -0.5\,mm/s$. This manifold is restricted to the

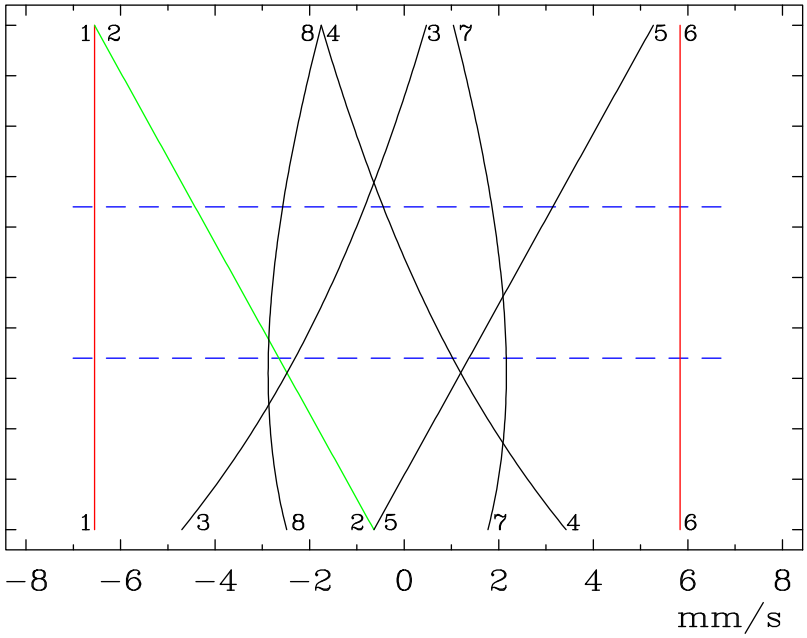

Figure 3: The one parameter ($L_2 - Is = x$) manifold based on Eq. 3 and 15 is shown. The values in [mm/s] of the outer line positions (red lines) are $L_1 = -6.55$, $L_6 = 5.83$ and an isomer shift $IS = -0.50$ and $Deq = -4.0$, that is a quadrupole splitting $\Delta Eq = |Deq|$ with a negativ V_{zz}. The dashed horizontal lines mark the region which in addition is compatible with $0 \leq sin^2\theta \leq 1$ and $-1 \leq cos\phi \leq 1$ restricting L_2 to $-4.5\,mm/s \leq L_2 \leq -2.7\,mm/s$.

region between the dashed horizontal lines of Fig. 3 by the conditions $0 \leq sin^2\theta \leq 1$ and $-1 \leq cos\phi \leq 1$ dirived from A_0 and A_1 of Eq. 7.
For each x the values A_0 and A_1 are known by Eq. 12 and 9 and the value of $m = -\mu_e H/3$ is known with $\mu_g H$ from above. With the quantities F and G

$$\begin{aligned} F(\eta) &= \frac{1+\eta^2/3}{m^2}\left(\frac{A_0 - 9m^4}{(Deq)^2} - \frac{1}{16}(Deq)^2\right) + (5-\eta^2)/2 \\ G(\eta) &= \frac{A_1}{4Deq}\sqrt{1+\eta^2/3} \end{aligned} \tag{16}$$

the equations are solved for $sin^2\theta$ and $cos\phi$.

$$\begin{aligned} sin^2\theta &= \left(F(\eta) + 2(G(\eta)+2)\right)/(9-\eta^2) \\ cos\phi &= \frac{1}{\eta}\left(3 - (G(\eta)+2)/sin^2\theta\right) \end{aligned} \tag{17}$$

Fig. 4 shows 4 different simulated spectra with the same line positions L_1 and L_6, isomeric shift $Is = -0.5\,mm/s$ and quadrupole splitting $\Delta Eq = |Deq|$ with a negative field gradient V_{zz} defining the input parameter $Deq = -4\,mm/s$. The 4 simulated 'experimental' spectra have different line positions of $x = L_2 - Is$ from top to bottom: $L2 = -4.23$, $L2 = -4.47$, $L2 = -3.37$ and $L2 = -2.70$ $[mm/s]$.
For each of the line positions defined by the values of L_2 there exists a manifold of solutions compatible with Eq. 7. The program **hyperfield** in the directory "EFFI/helproutines/lineLR/hyper_field" prints the manifold of solutions.

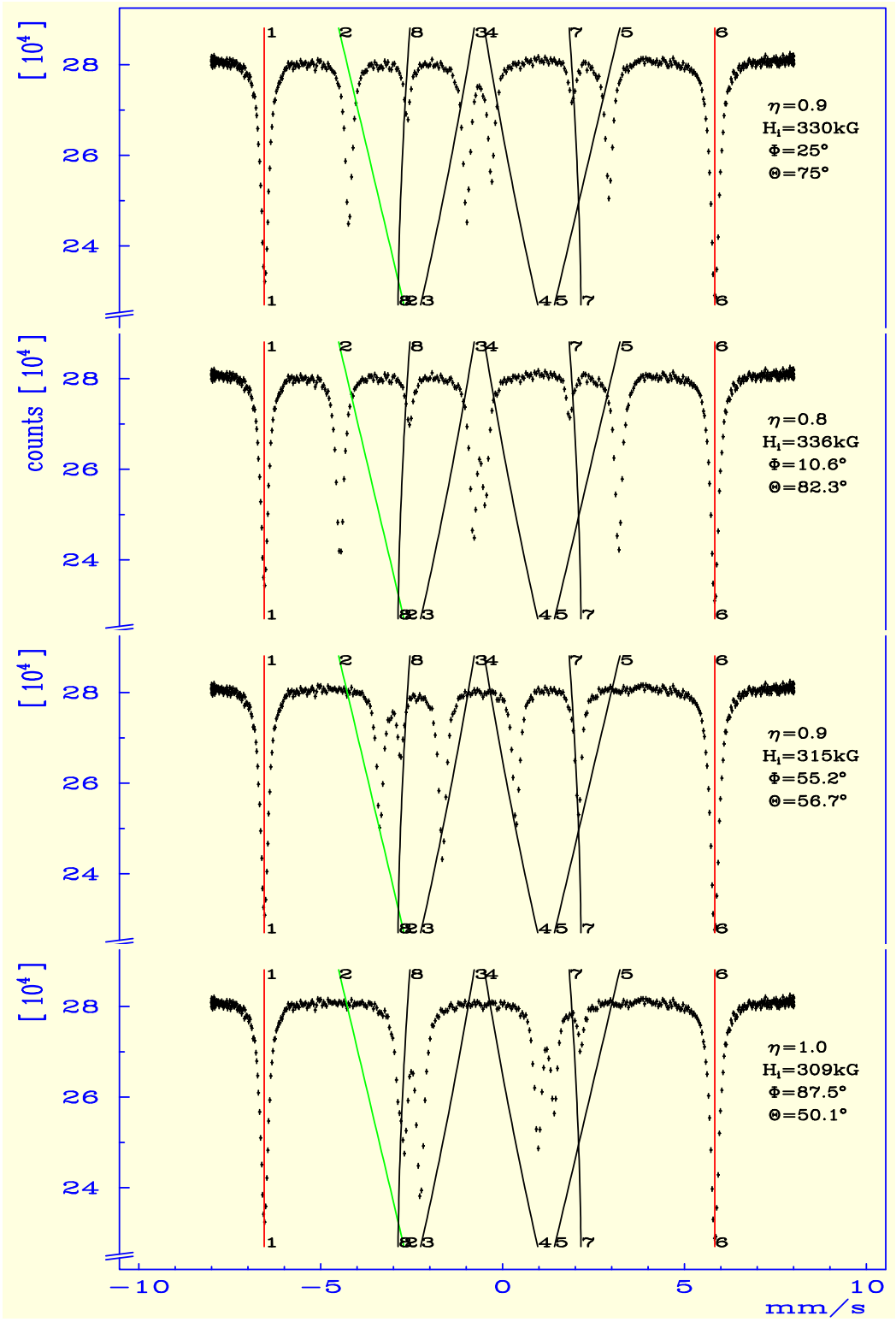

Figure 4: Four 'Experimental' absorption spectra of the $\frac{1}{2} - \frac{3}{2}$ 14.4keV transition of ^{57}Fe has been simulated with the program **effi**. All have the same line positions L_1 and L_6 and isomeric shift $Is = -0.5\,mm/s$ and quadrupole splitting $\Delta Eq = |Deq|$ with $Deq = -4\,mm/s$. As a consequence the manifold of 8 line positions $1, ..., 8$ are the same for all simulated spectra. Their different parameters of the hyperfine fields (η, H_i, ϕ and θ) are written at the right border.

5 Program hyperfine fields

The program **hyperfield** has three entries where two of them define 4 line positions and one is eqivalent to the program **lines_alpl** with L_1, L_6, Is and Deq as input parameters. The first two cases require L_1, L_2,L_5 and L_6 or eqivalently using Eq. 2 L_1, L_2, L_6 and Is.
If L_2 is given, two parameters η and Deq are tested for compatibility, so that the printout covers the manifold of solutions of two parameters. For the 4 spectra of Fig. 4 the solution with $Deq = -4\,mm/s$ and an η-value close to 0.9 (stepwidth was 0.1) has been selected.
If Deq is fixed for each L_2 the parameters η are tested.

References

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