Transparency of a thin absorber in Moessbauer optics: effect of electron relaxation

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Abstract. A model of Moessbauer absorption taking into account the effects of the electron relaxation and the nuclear levels anticrossing simultaneously has been proposed. The dependence of the absorption deficit (transparency) on the relaxation and mixing parameters has been obtained using the stochastic theory of Moessbauer relaxation spectra. The role of quantum interference in occurrence of a partial transparency on Moessbauer transitions has been explored under these conditions.

1. Introduction

The problem of Moessbauer radiation transparency has took growing attention after the experiment [1] on the observation of the absorption deficit in FeCO₃ at 30.5 K under nuclear levels anticrossing conditions [2]. The theory of this phenomenon is discussed in references [3, 4, 5, 6] and recently a progress in understanding of possible mechanisms of the absorption deficit formation is achieved. As it was for the first time shown in [4], the electromagnetically induced transparency (EIT) [7] analog for Moessbauer radiation is possible if the crossing nuclear levels have different decay rates (see [8] also). In this report we consider this problem concerning to the case of thin absorber [5] in order to investigate conditions which lead to the realization of this mechanism. A stochastic relaxation model of the Moessbauer absorption [9] is used for this. One enables to determine the conditions leading to different relaxation widths of the crossing nuclear levels and establishes a quantitative relation between degree of the transparency and the electron relaxation parameters. It should be noted, this model provides an adequate explanation of the transparency observed earlier [1,4] and in some extent evolves a model proposed in [4].

2. The partial transparency at Moessbauer transitions due to electron relaxation

The main feature of the model used below is taking into account by the explicit way a fluctuation of the electron magnetic moment when the absorption cross section under a nuclear level anticrossing condition is calculated. The Hamiltonians of a nucleus in the ground (g) and excited (e) states have the form [9]:

\[ H^g = H^g f(t), \quad H^e = H^e f(t) + H^Q + H^Q⊥, \]

where \( H^g,e = Z_{e,g} \hat{I}_z^g,e \) and \( H^Q = Q[(\hat{I}_z^e)^2 - (\hat{I}_e^e + 1)/3] \) are Zeeman and coaxial to them quadrupole interactions, respectively. \( H^Q⊥ = Q^⊥[(\hat{I}_z^e)^2 + (\hat{I}_e^e)^2] \) is quadrupole interaction.
leading to a nuclear level mixing, where $Q^\perp = Q\eta/6$ and $\eta$ is the asymmetry parameter. The crossing condition has the form $Z_0 = Q$. The stochastic function $f(t)$ in Eq. (1) reflects stepwise changes in the hyperfine magnetic field due to the fluctuations of the electron spin. The function $f(t)$ in the simplest case has two values $a = \pm 1$ (two-level relaxation). The random process is characterized by the matrix of the stochastic transitions $W_{ab}$, where $W_{++} = \nu_1$ is the probability of the transition from the state characterized by the value $f(t) = +1$ to the state corresponding to $f(t) = -1$. It should be noted, that $W_{++} = -W_{+-} = -\nu_1$, and $W_{--} = \nu_2 \neq \nu_1$ in the general case. The cases $\nu_2 = \nu_1$ and $\nu_2 \neq \nu_1$ are called symmetric and asymmetric relaxation processes, respectively. According to Blume [9] the absorption cross section for gamma photons is given by the known expression:

$$\sigma(p) \sim \text{Re} \sum_{MmM'm',a} p_a \langle m | k_1 \sigma | M \rangle U_{MmM'm',a}^{ab} \langle M' | k_1 \sigma | m' \rangle. \quad (2)$$

Here, $\langle M' | k_1 \sigma | m' \rangle$ is the matrix element of the absorption of a photon with the wave vector $k_1$ and polarization $\sigma$ (the transition of the nucleus from the ground state $I_g, m'$ to the excited state $I_e, M'$). $p = -i(\omega_\gamma - \omega_0) + \Gamma/2$, where $\omega_\gamma$ is the energy of the photon and $\omega_0$ and $\Gamma$ are the energy and width of the Moessbauer level, respectively. $p_a = p(a)$ is the probability that the stochastic function $f(t)$ takes the value $a = \pm 1$. The condition $p(+)|W_{++} = p(-)|W_{--}$ is assumed. $U_{MmM'm',a}^{ab} = \{pE' - W - i L_1\}^{-1}$ is the matrix of the Laplace transform of the evolution superoperator. This matrix has the dimension $(2I_g + 1)(2I_e + 1)$, where a factor of 2 corresponds to the dimension of the stochastic space ($\pm 1$). The matrices $W$, $E$, and $L_1$ have the form:

$$W = \begin{pmatrix} -W_{ab}E' & W_{ab}E' \\ W_{ba}E' & -W_{ba}E' \end{pmatrix}, \quad E = \begin{pmatrix} E' & 0 \\ 0 & E' \end{pmatrix}, \quad L_1 = \begin{pmatrix} L^Q + L & 0 \\ 0 & L^Q - L \end{pmatrix}. \quad (3)$$

where $E'$ is the identity matrix and $L$ and $L^Q$ are the Liouville operator matrices; all of these matrices have the dimension $(2I_g + 1)(2I_e + 1)$. For the conditions of anticrossing described above:

$$L_{MmM'm'} = H_{MM'}^{\perp} \delta_{mm'} - H_{m'm}^b \delta_{MM'}, \quad L^Q_{MmM'm'} = \left(H_{MM'}^Q + H_{MM'}^{\perp} \right) \delta_{mm'}. \quad (4)$$

**Figure 1.** a) The Zeeman structure of Moessbauer transitions under levels crossing condition ($FeCO_3$). b) The absorption deficit $\Delta \sigma(p, \Omega)$ at crossing point, $\omega_\gamma = \omega_{cr}$; the dependence on $\Omega$ the same for polarizations $\sigma^\perp$ and for unpolarized photons. The longitudinal geometry and the values of $\nu_1, \nu_2$ as for Fig. 2 are used.
If $H_{Q}^{\pm}$ is neglected, the matrix $[pE - W - iL_{1}]$, which should be inversed, consists of second-order block matrices $Mm$. The inversion of matrix $Mm$ enables one to obtain the effect of the stepwise change in the hyperfine field on the line shape of the Mössbauer transition $Mm$. A small perturbation $H_{Q}^{\pm}$ becomes significant when two transitions, $Mm$ and $M'm'$, have the same energy $(L + L')_{Mm}Mm = (L + L')_{M'm'M'm'}$. Now we should to inverse a fourth-order matrix composed of second-order matrices $Mm$ and $(M \pm 2)m$ joined with each other by the perturbation $\Omega$: $L_{MmM'm'}^{Q} = H_{Mm'}^{Q} \delta_{m'm'} = 2\sqrt{3}Q^{\perp} \delta_{Mm'2} = \Omega \delta_{Mm'2}$. The expression (2) becomes as sum of the terms represented by a fourth-order matrices $(M'M')$: 

$$\sigma(p, \Omega) \sim \text{Re} \sum_{(M'M'); M, M' \in (M'M'), m, \sigma, ab} p(a) \langle m|k_{1}\sigma|M'\rangle U_{m'M'mM'}^{ab} \langle M|k_{1}\sigma|m\rangle. \tag{5}$$

Using expression (5) it was shown [5] the outer lines of Zeeman sextet are broader than the inner lines $\Gamma_{1} > \Gamma_{3}$ (see transitions presented in fig.1) for the asymmetric electron relaxation ($\nu_{1} \neq \nu_{2}$). The expression (5) enables us to introduce an absorption deficit factor $\Delta\sigma(p, \Omega) = (\sigma(p, 0) - \sigma(p, \Omega))/\sigma(p, 0)$, which defines the degree of the transparency at the crossing point. It should be noted, the $\Delta\sigma$ factor depends on mixing parameter and on relaxation parameters $\nu_{1}, \nu_{2}$ (or the values $\Gamma_{1}, \Gamma_{3}$ corresponding to them). The dependence of $\Delta\sigma(p, \Omega)$ versus $\Omega$ at the crossing point, $\omega_{cr} = \omega_{cr}$, is shown in Fig. 1b. Let us discuss possible mechanisms leading to the absorption deficit.

If the widths of crossing levels are equal, $\Gamma_{1} = \Gamma_{3}$ (it means $\nu_{1} = \nu_{2}$), the deficit $\Delta\sigma(p, \Omega)$ is a consequence of Rabi splitting (the repulsion) of nuclear levels [2]. One can be called as Rabi deficit. A different result we have if $\Gamma_{1} \neq \Gamma_{3}$. Now the decrease in the absorption cross section cannot be attributed only to Rabi effect, the explicit mechanism of deficit depends essentially on the mixing parameter value $\Omega$. While $\Omega < \Omega_{sp} = (\Gamma_{1} - \Gamma_{3})/4$ the nuclear levels anticrossing leads to two mixed states with the same energy. The decay constants of these states vary from the values $\Gamma_{1}$ and $\Gamma_{3}$ to the common value $(\Gamma_{1} + \Gamma_{3})/2$ as $\Omega$ increases from 0 to $\Omega_{sp}$. On the contrary, if $\Omega > \Omega_{sp}$, the mixed states have the same decay constants but their energies are different: $\omega_{cr} \pm \sqrt{\Omega^{2} - (\Gamma_{1} - \Gamma_{3})^{2}}/16$. There is no Rabi splitting in first case, so the interference of the quantum amplitudes (QI) is responsible for the deficit in this case. If $\Omega > \Omega_{sp}$ two mechanisms (the Rabi and QI ones) contribute to the total absorption deficit. The last one is proportional to $\Gamma_{1} - \Gamma_{3}$ and it vanishes when $\Gamma_{1} = \Gamma_{3}$. The figure 2 illustrates aforesaid for the longitudinal geometry, $k \parallel H_{z}$ (Fig. 1a). In this case the transitions 1 and 3 have the pure polarizations $\sigma^{-}$ and $\sigma^{+}$. The spectra $\sigma(p, \Omega)$ and $\Delta\sigma(p, 0)$ are calculated (thin and thick solid curves respectively) using (5) for $\Omega < \Omega_{sp}$ and $\Omega > \Omega_{sp}$. Let us note, in cases 2 a) and 2 b) the $\sigma(p, \Omega)$ (thin curve) consists of two interference parts (dotted lines). In both cases however the entire result of interference is destructive one. It results in decreasing of the absorption cross section in the vicinity $\omega_{cr} = \omega_{cr}$. Let us now pay attention to the cases 2 c) and 2 d). Now the thin curves are a sum of four terms. There are two Rabi contributions (dashed lines) and two interference terms (dotted lines) in each case. It is easy to see, the interference contribution is constructive in case 2 c) and destructive in case 2 d). In other words for the transition to the level with a larger width (absorption of the $\sigma^{-}$ polarization photon) QI leads to the additional decrease of $\sigma(p, \Omega)$, or increase of the Rabi deficit. That is similar to phenomenon of EIT discussed firstly in [7]. For $\sigma^{+}$ photon the effect is opposite, now the interference contribution causes the decreasing of available value of Rabi deficit.

Separate contributions of two mechanisms to the absorption cross section $\sigma(p, \Omega)$ (presented by dashed and dotted lines in a Fig. 2) have been calculated by us using the method of the optical theorem. For this purpose the scattering amplitudes were constructed on the basis of nuclear spin states having different decay rates and mixed in anticrossing regime. Such calculations enable
Figure 2. Cross section $\sigma(p, \Omega)$ in the vicinity $\omega \simeq \omega_{cr}$. The values of $\nu_1$, $\nu_2$ used correspond to $\Gamma_1 = 6.49\Gamma$, $\Gamma_3 = 1.16\Gamma$. Thick lines represent $\sigma(p, 0)$, thin lines - $\sigma(p, \Omega)$. The interference contributions to $\sigma(p, \Omega)$ are indicated by dotted lines, Rabi splitting terms - by dashed lines. The mixing parameters $\Omega = \Gamma < \Omega_{sp} (a,b)$ and $\Omega = 2\Gamma > \Omega_{sp} (c,d)$ are used. Longitudinal scheme of experiment is supposed, a) and c) correspond to polarization $\sigma^-$, b) and d) - to $\sigma^+$. us to obtain simple analytical expressions for $\sigma(p, \Omega)$ in the case of longitudinal geometry. The mixing of levels having different decay rates was discussed earlier in [10] and partly in [4].

3. Concluding remarks
The asymmetric relaxation of the electron spin essentially changes the Moessbauer sublevels anticrossing mechanism. It leads also to the absorption deficit (partial transparency) of the Moessbauer spectrum at the crossing point due QI mechanism. Both of these are a result of the difference between the relaxation widths of the crossing nuclear levels. The absorption deficit model proposed recently [5] describes adequately the results of the experiments [1,4]. It serves also as a tool of the quantitative relation between the partial transparency on the Moessbauer transitions and the dynamical (relaxation) parameters of the system. In weak relaxation limit this model yields a results which are very close to a results obtained by methods [4,7] using an empirical widths $\Gamma_1, \Gamma_3$ for Moessbauer sublevels.

The same effect of partial transparency due QI is expected if the mixing of Moessbauer sublevels with different relaxation widths is induced by strong (RF and optical) electromagnetic field.

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