New possibilities of photon echo: determination of ground and excited states $g$-factors applying a weak magnetic field pulse

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Abstract. We have measured the magnetic parameters of a paramagnetic ion in the ground and excited states by controlling the relative phases of excited dipoles with pulse of a weak magnetic field. Er$^{3+}$ in two crystals LuLiF$_4$ and YLiF$_4$ has been used as a paramagnetic ion. The second matrix was used for control. Optical transition is $^{4}I_{15/2} \rightarrow ^{4}F_{9/2}$.

1. Introduction

A weak perturbation pulse can lead to a significant change of the relative phases of radiators and so to the amplitude of the resulting dipole moment. It can be manifest in many cases, in which coherent effects are important (photon echo, the propagation of light, and so forth). In the optical systems, in which the Zeeman Effect is present, a pulse magnetic field can be used as a perturbation. The Zeeman Effect is the change of the period of light emitted by a substance when the substance was acted upon by magnetic force [1]. Pieter Zeeman observed only the broadening of optical lines in a magnetic field but not splitting. The frequency change of the emitted light should exceed the inhomogeneous line width, in order to observe the splitting of the optical line. A large magnetic field is needed to use this. Photon echo is the method which overcomes the difficulties associated with inhomogeneous line width and allows to observe the splitting of optical lines in a weak magnetic field.

2. Photon echo

Photon echo is coherent radiation of a system in the form of the short pulse, caused by restoration of the phase of separate radiators after the change of a sign on relative frequency of radiators. For two-pulse photon echo we have

$$\phi_\alpha(t) = \delta_\alpha(t_2 - t_1) + \pi - \delta_\alpha(t - t_2) = \{t = t_\text{echo} = 2t_2 - t_1\} = \pi,$$

$$P(t) \propto \sum_\alpha e^{i\phi_\alpha(t)} g\left(\delta_\alpha = \{t = t_\text{echo} = 2t_2 - t_1\} = -P(0)\right),$$

$$I(t) = |P(t)|^2 = \{t = t_\text{echo} = 2t_2 - t_1\} = I(0), \quad \delta_\alpha = (\omega_\alpha - \omega).$$

Here $t_1$ and $t_2$ are the times of switching on laser pulses, $\omega$ is the laser frequency, $\omega_\alpha$ is frequency of transition for $\alpha$-ion, $P$ is the dipole moment of a system, $I$ is echo intensity.
3. Zeeman splitting of optical line by magnetic field

Transition frequencies of two groups of paramagnetic ions \( \text{Er}^{3+} \) with spin projection in ground state \( \pm 1/2 \) are shifted by magnetic field on the different values (see figure 1)

\[
\Delta E(\frac{1}{2}I_{1/2}) = \beta H \langle \mu = \pm 3/2 | L_z + 2S_z | \mu = \pm 3/2 \rangle = \]

\[
= g(\frac{1}{2}I_{1/2}) \beta m, \ m = \pm 1/2
\]

\[
\Delta \omega = Zm, \ \text{(rad/s)}
\]

\[
Z = [g(\frac{4}{2}F_{1/2}) \pm g(\frac{1}{2}I_{1/2})] \beta H / \hbar,
\]

where \( g(\frac{4}{2}F_{1/2}) \) and \( g(\frac{1}{2}I_{1/2}) \) are the g-factors of excited and ground states of erbium ion, \( \beta \) is the Bohr magneton, \( \hbar \) is the Planck constant, \( \mu \) is crystal quantum number, \( L_z \) and \( S_z \) are projections on crystal axis \( C \) of total orbital and spin moments accordingly for \( \text{Er}^{3+} \). You can see that \( \pi \)- and \( \sigma \)-Zeeman splitting is proportional accordingly to the difference and to the sum of the excited and ground states g-factors.

![Energy levels of \( \text{Er}^{3+} \) in YLiF\(_4\) and LuLiF\(_4\) (H\(\parallel\)C). Also the transition and the transition wavelength are shown (the arrows indicate the transitions for \( \pi \) and \( \sigma \) polarizations of the laser). C is crystal axes, H is magnetic field and E is laser electrical field. E\(\parallel\)C is \( \pi \) polarization, E\(\perp\)C is \( \sigma \) polarization. \( \mu \) is crystal quantum number, \( \pm 1/2 \) indicate the \( \text{Er}^{3+} \) ion states whose spin projection on axis C are \( \pm 1/2 \).](image)

4. Photon echo and a pulse of magnetic field

If the magnetic field pulse switches on after the second laser pulse then the relative phase \( \phi \) of two groups of radiators is not equal to zero:

\[
\phi_\alpha(t) = \delta_\alpha(t - t_1) + \pi - \delta_\alpha(t - t_2) - \int_{t_0}^{t} dt' \Delta \omega(t') = \phi_\alpha^0(t) - m_\alpha \varphi(t),
\]

\[
\varphi(t) = \int_{t_0}^{t} Z(t'), \ m_\alpha = \pm 1/2,
\]

and the amplitude of the total dipole moment and the echo intensity are significantly changed:
\[
P(t) \propto \sum_{\alpha} e^{i\delta_\alpha(t)} g(\delta_\alpha) = \sum_{\alpha} e^{i\delta_\alpha(t)} e^{-im_\alpha \varphi(t)} g(\delta_\alpha) = P_0(t) \cos(\varphi(t)/2),
\]
(4)

\[
I(t) = |P(t)|^2 = I_0(t)(1 + \cos(\varphi(t))/2,
\]
where \(t_0\) is the time of the magnetic pulse switch on.

5. Magnetic pulse does not overlap in time with echo-pulse

The relative phase \(\varphi\) is not dependent on time in this case. The echo intensity oscillates versus relative phase as you can see from (4) and figure 2. Zeeman splitting \(Z=10\) MHz was observed [2] in \(YLiF_4:Er^{3+}\) and \(LuLiF_4:Er^{3+}\), which is much less than the inhomogeneous line width \(=600\) MHz.

![Figure 2](image)

**Figure 2.** Time sequence of laser, echo and magnetic pulses, phase’s evolution (left) and relative echo intensity versus magnetic pulse amplitude for sigma- and pi-polarizations in \(LuLiF_4:Er^{3+}\).

6. Magnetic pulse overlaps in time with echo-pulse

The phase \(\varphi\) depends on time. The echo time form is changed [3]. If the relative phase \(\varphi < \pi\) then the echo pulse is “compressed” and the time of echo “decreases”. It is shown in figure 3 (above): schematically on the left and experimental results on the right. If the relative phase \(\varphi\) is larger than \(3\pi\) then the echo time form is modulated. Figure 3 (below) shows the case of \(\varphi>5\pi\): schematically on the left and experimental results on the right. The modulation period \(T\) is determined from the relation

\[
\varphi(t + T(t)) - \varphi(t) = 2\pi.
\]
(5)

It generally depends on time. If echo \(I_0\) and Zeeman splitting \(Z\) are constant in the time interval \([t, t+T(t)]\) then (see (3))

\[
\varphi(t + T(t)) - \varphi(t) = 2\pi = \int_{t}^{t+T(t)} Z(t') dt' \approx Z(t)T(t).
\]
(6)

In our experiment, as you can see in figure 3 (below), the duration of the flat part on top of the magnetic pulse (25 ns) becomes approximately equal to the echo pulse duration (28 ns). Therefore, \(Z\)
is constant in (6). From the waveforms of echo signals observed at various times $t_0$ of the beginning of action of the magnetic pulse, similar to those shown in figure 3, the time intervals between the nearest minima in the echo signals were measured and the average value $T$ of the modulation period was determined. The next equation has been used for the determination of g-factors:

$$T|g(^4F_{9/2}) \pm g(^4I_{15/2})|\beta H / h = 2\pi .$$ (7)

Here $H$ is the magnetic pulse amplitude in the region of the flat part on top of the pulse.

![Figure 3](image.png)

**Figure 3.** Magnetic pulse overlaps in time with echo-pulse. The case $\phi < \pi$ is shown above and $\phi > 5\pi$ is shown below. It illustrates the waveforms of magnetic pulse and photon echo for $\pi$-polarizations of laser pulses when the magnetic pulse is on and off.

The dependence of the average period $T$ versus $H$ has been built. Table 1 shows these dependences for different polarizations of the laser pulses in the samples YLiF$_4$:Er$^{3+}$ and LuLiF$_4$:Er$^{3+}$. Also the values of the sum and differences of g-factors, determined using (7), are shown.
Here $g'_g = g(^4I_{9/2})$ and $g = g(^4I_{15/2})$ are g-factors of the excited and ground states of Er$^{3+}$.

Determining from this table the average value of the sum and the difference of the g-factors we find

$$g = 3.06, \quad g' = 9.6 \quad \text{for} \quad \text{LuLiF}_4: \text{Er}^{3+}$$

$$g = 3.12, \quad g' = 9.75 \quad \text{for} \quad \text{YLiF}_4: \text{Er}^{3+}$$

The ground state g-factor values are in good agreement with those measured by EPR method: $g({\text{LuLiF}}_4: \text{Er}^{3+}) = 3.09$ [4], $g({\text{YLiF}}_4: \text{Er}^{3+}) = 3.14$ [5]. The excited state g-factor value for $\text{YLiF}_4: \text{Er}^{3+}$ is in good agreement with those measured by optical methods: $g'({\text{YLiF}}_4: \text{Er}^{3+}) = 9.84 \pm 0.25$ [6], $g'({\text{YLiF}}_4: \text{Er}^{3+}) = 9.61$ [7].

7. Conclusion

In our study we measured the Zeeman splitting of the ground and excited states and determined magnetic parameters of the paramagnetic ion with an accuracy comparable to the EPR using the known values of the magnetic pulse amplitude. If the magnetic parameters are known, it is possible to determine the amplitude of the MP, using the relation (7). For example, if the magnetic field is created by the pulse change of the dipole-dipole interaction of the ion with the environment. This can be used to determine the distance to the centers of the environment by measuring the waveform of the echo modulation periods, not only in the optical range, but also in the EPR and NMR bands, using relations similar to (7).

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