Homogeneous linewidth of the $^4I_{11/2}$–$^4I_{15/2}$ optical transition of erbium in $LiNbO_3{:}Er^{3+}$

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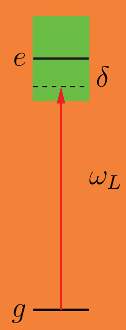
Abstract

We work out a simple, pulsed pump-probe measurement scheme to measure the homogeneous linewidth of an atomic transition in an inhomogeneously broadened spectral line in a solid state environment. We apply the theory to the $^4I_{11/2}-{}^4I_{15/2}$ optical transition of erbium in $LiNbO_3{:}Er^{3+}$ crystal. Beside obtaining the homogeneous linewidth, we have estimated the population relaxation time as well.

Introduction



A dopant atom in a crystal expiriences incoherent spectral line broadening due to AC Stark-shift induced by the surrounding atoms and crystal defects.

To design coherent control experiments in doped solids, we have to know the population relaxation T_1, and the diplole relaxation T_2 times of the involved transitions of the dopant atoms. In many cases $T_2 \ll T_1$ (depends on temperature, external magnetic field).

Model calculation

The Bloch vectors associated with the density matrices of a set of two-level systems are defined as

$$
\begin{aligned}
U_\delta(t) &= \varrho_{eg}(\delta;t) + \varrho_{ge}(\delta;t)\,, &\text{(1a)}\\
V_\delta(t) &= i[\varrho_{eg}(\delta;t) - \varrho_{ge}(\delta;t)]\,, &\text{(1b)}\\
W_\delta(t) &= \varrho_{ee}(\delta;t) - \varrho_{gg}(\delta;t)\,. &\text{(1c)}
\end{aligned}
$$

where the index δ denotes the frequency difference between the transition frequency ω_{eg} of the particular atom and some reference frequency ω, $\delta = \omega_{eg} - \omega$.

Our method consists of three subsequent steps:

Writing: If the atoms interact with a monochromatic, nearly resonant field, the dynamics is described by the Bloch equations

$$
\begin{aligned}
\dot U_\delta &= -\delta V_\delta - \frac{1}{T_2}U_\delta\,, &\text{(2a)}\\
\dot V_\delta &= \Omega_{\mathrm{w}} W_\delta + \delta U_\delta - \frac{1}{T_2}V_\delta\,, &\text{(2b)}\\
\dot W_\delta &= -\Omega_{\mathrm{w}} V_\delta - \frac{1}{T_1}(W_\delta + 1)\,, &\text{(2c)}
\end{aligned}
$$

where $\omega = \omega_{\mathrm{w}}$. The Rabi frequency Ω_{w} is given by $\Omega_{\mathrm{w}} = -d_{eg}E_{\mathrm{w}}/\hbar$, where d_{eg} denotes the dipole moment of the atomic transition, E_{w} describes the pump (write $\equiv$ w) field strength. After a long enough time, the atoms reach a steady state

$$
\begin{aligned}
U_\delta(0) &= \frac{\Omega_{\mathrm{w}}\delta}{\delta^2 + \frac{1}{T_2^2} + \frac{T_1}{T_2}\Omega_{\mathrm{w}}^2}\,, &\text{(3a)}\\
V_\delta(0) &= -\frac{\Omega_{\mathrm{w}}/T_2}{\delta^2 + \frac{1}{T_2^2} + \frac{T_1}{T_2}\Omega_{\mathrm{w}}^2}\,, &\text{(3b)}\\
W_\delta(0) &= -\frac{\delta^2 + 1/T_2^2}{\delta^2 + \frac{1}{T_2^2} + \frac{T_1}{T_2}\Omega_{\mathrm{w}}^2}\,. &\text{(3c)}
\end{aligned}
$$

Delay: All fields are switched off for a time t_d. The system undergoes free evolution. For $T_2 \ll t_d$ we have $U_\delta(t_d) = 0$, $V_\delta(t_d) = 0$, and $W_\delta(t_d) = (1 + W_\delta(0))\exp(-t_d/T_1) - 1$.

Read out: A short read out pulse probes the atoms. It is assumed that the length of the probe pulse τ satisfies the relation $T_2 \ll \tau \ll T_1$. Then the population relaxation does not take place, but the dipole relaxation does. As a result $W_\delta(t_d + \tau) \approx W_\delta(t_d)$ and

$$
\begin{aligned}
U_\delta(t_d+\tau) &= -\frac{(\Delta-\delta)\Omega_p}{(\Delta-\delta)^2 + \frac{1}{T_2^2}}W_\delta(t_d)\,, &\text{(4a)}\\
V_\delta(t_d+\tau) &= \frac{\Omega_p/T_2}{(\Delta-\delta)^2 + \frac{1}{T_2^2}}W_\delta(t_d)\,, &\text{(4b)}
\end{aligned}
$$

where Δ is the detuning of the probe pulse from the frequency of the pump (write) pulse.

Susceptibility The susceptibility of the ensemble of the atoms is obtained from $U_\delta(t_d+\tau)$ and $V_\delta(t_d+\tau)$ by summing up for all subset of atoms with transition frequency difference δ. The sum should be weighted by the inhomogeneous line profile function $g(\delta)$

$$
\chi^+(\Delta) = -\mathcal{N}\frac{|d_{eg}|^2}{\varepsilon_0\hbar}\int g(\delta)\frac{(\Delta-\delta)+i/T_2}{(\Delta-\delta)^2+\frac{1}{T_2^2}}W_\delta(t_d)\,d\delta\,, \tag{5}
$$

where $\mathcal{N}$ is the density of the two-state atoms participating in the process. The integral can be performed via application of the Fourier and inverse Fourier transforms

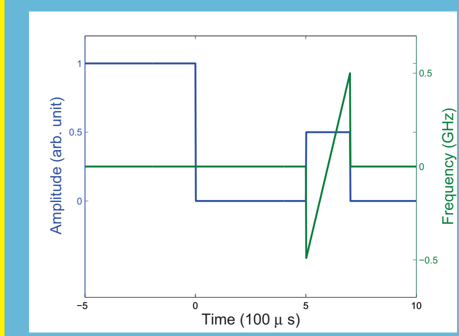
$$
\chi^+(\Delta) = \frac{\mathcal{N}|d_{eg}|^2}{\varepsilon_0\hbar}\pi g(0)\left[\frac{\frac{T_1}{T_2}\Omega_{\mathrm{w}}^2\Delta}{\Gamma\left((\frac{1}{T_2}+\Gamma)^2+\Delta^2\right)} + i\left(1 - \frac{\frac{T_1}{T_2}\Omega_{\mathrm{w}}^2(\frac{1}{T_2}+\Gamma)}{\Gamma\left((\frac{1}{T_2}+\Gamma)^2+\Delta^2\right)}e^{-t_d/T_1}\right)\right]\,, \tag{6}
$$

where $\Gamma^2 = \frac{1}{T_2^2} + \frac{T_1}{T_2}\Omega_{\mathrm{w}}^2$. The imaginary part of the susceptibility in Eq. (6) describes a *spectral hole*.

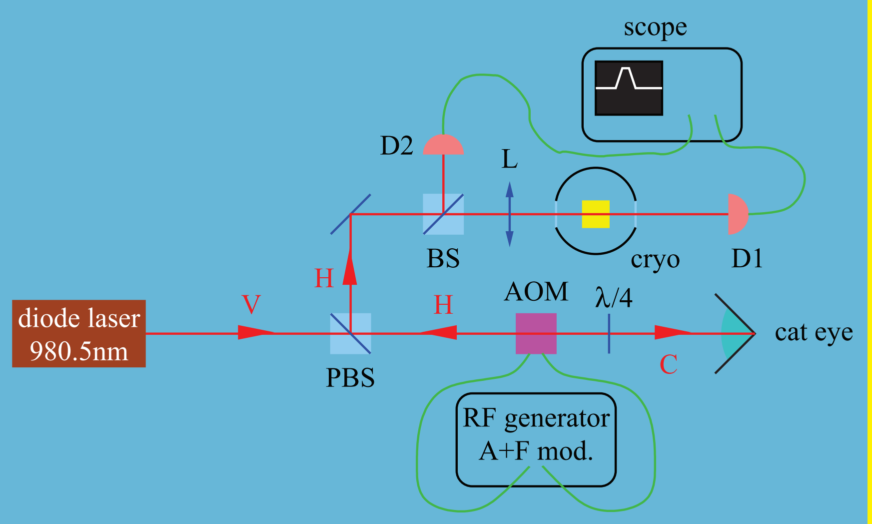
Here the quantity $(T_1/T_2)\Omega_{\mathrm{w}}^2$ corresponds to power broadening. For a low intensity pump (write) pulse the result of the convolution can be expanded, in lowest order of Ω_{w}^2 one finds

$$
\mathrm{Im}(\chi^+(\Delta)) = \frac{\mathcal{N}|d_{eg}|^2}{\varepsilon_0\hbar}\pi g(0)\left(1 - \frac{2\frac{T_1}{T_2}\Omega_{\mathrm{w}}^2}{\Delta^2+\frac{4}{T_2^2}}e^{-t_d/T_1}\right)\,. \tag{7}
$$

Experiment:

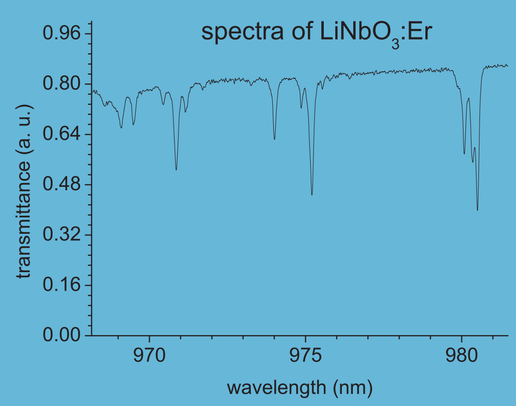


Pulse sequence.

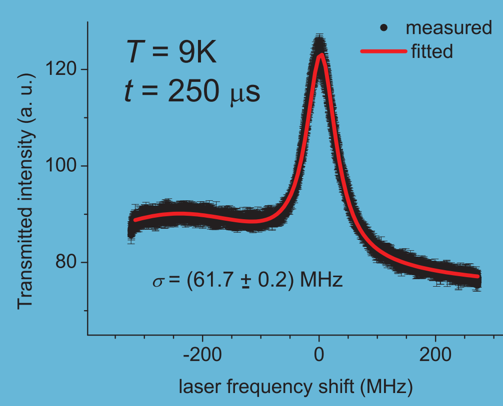


Experimental setup.

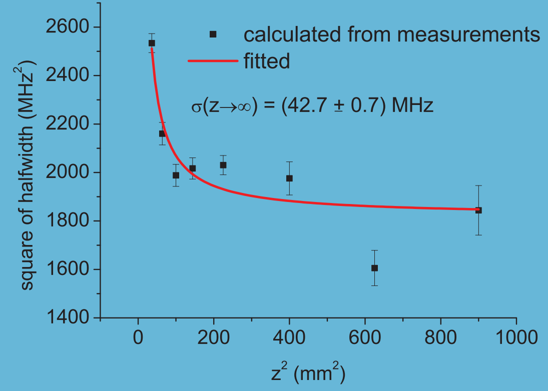
Results:



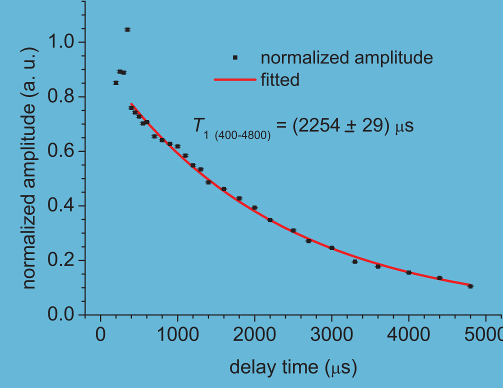
A section of the absorption spectrum of $LiNbO_3{:}Er^{3+}$.



A typical scan.

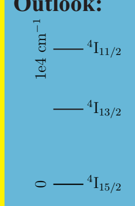


Z-scan measurement to obtain $T_2 = 4/\sigma \approx 90$ns.



T_1 measurement.

Outlook:



The erbium ions are not true two-level systems. The form of the exponential decay in t_d changes in a more realystic three-level model. In a three-level model the shape of the absorption line does not change in the weak field limit.

Future plans:

1. repeat the measurement with AOM aplitude control instead of Z-scan
2. photon-echo measuremet
3. CW pump-probe measurements
4. demonstration of coherent pulse propagation effects
5. ...

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