Analysis of optical differential transmission signals from co-propagating fields in a lambda system medium

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We analyze theoretically and experimentally how nonlinear differential-transmission spectroscopy of a lambda-system medium can provide quantitative understanding of the optical dipole moments and transition energies. We focus on the situation where two optical fields spatially overlap and co-propagate to a single detector. Nonlinear interactions give cross-modulation between a modulated and non-modulated laser field, yielding differential transmission signals. Our analysis shows how this approach can be used to enhance the visibility of relatively weak transitions, and how particular choices in the experimental design minimize systematic errors and the sensitivity to changes in laser field intensities. Experimentally, we demonstrate the relevance of our analysis with spectroscopy on the donor-bound exciton system of silicon donors in GaAs, where the transitions from the two bound-electron spin states to a bound-exciton state form a lambda system. Our approach is, however, of generic value for many spectroscopy experiments on solid-state systems in small cryogenic measurement volumes where in-situ frequency or polarization filtering of control and signal fields is often challenging.

I. INTRODUCTION

Laser spectroscopy is a main research tool to study electronic states of materials, across research fields ranging from atomic physics to cellular biology. In modern optics, nonlinear behavior of a medium is of primary interest, often studied with multiple laser fields which spatially overlap. In such cases, the presence of a strong control laser field makes it challenging to extract the signal from a weak probe field, in particular when further constraints enforce co-propagation of these fields. In practice one cannot always rely on spectral or polarization filtering (for example in cryogenic measurement volumes) and in these cases low-frequency intensity modulation combined with lock-in detection techniques provides a means to separate a signal from co-propagating fields. Such modulation techniques also improve the signal to noise ratio, as signals become less sensitive to external light and 1/f noise sources.

With this modulation technique applied to a nonlinear medium, the susceptibilities as seen by the control and probe fields become time dependent. This yields cross-modulation, where modulation of the applied intensity of one field influences the transmitted intensity of another field, giving rise to so-called differential transmission signals. Notably, for this paper the notion of differential transmission refers to this slow modulation of a susceptibility with continuous-wave lasers, effectively comparing two different steady-state conditions. The modulation timescale is orders of magnitude slower than the intrinsic electronic dynamics of the medium (that can be directly studied in pump-probe experiments with ultrafast pulsed lasers).

Low-frequency intensity modulation has already been applied in a variety of optical experiments with differential signals from transmission, reflection or luminescence. Textbooks on nonlinear spectroscopy qualitatively discuss the benefits of studying crosstalk signals from modulation techniques in saturation spectroscopy. Generically, the advantages lie in enhancing the visibility of weak transitions, and it was applied in experimental work on donor centers in semiconductors, quantum dots, and other atomic-like systems. However, a detailed analysis of the differential signals is mostly not presented, despite the fact that this is essential for optimal design of these experiments, interpretation of the results, and insight into systematic errors that may occur.

We present here an analysis of the signals from differential transmission spectroscopy (DTS) on a medium with three-level lambda systems, based on both model calculations and experimental results. We show how DTS can strongly enhance the spectroscopic visibility of weakly absorbing transitions, and how this strongly depends on the relative strength of the dipole moments. We also investigate how the method can provide quantitative results for the strength of the optical dipole moments (which are key ingredients if one also wishes to unravel the combined DTS signal from two co-propagating fields into the separate transmission components). Further, our analysis shows how in two-laser experiments (where one laser is fixed and one is used for spectroscopic scanning) choosing which laser is modulated makes a key difference for being robust against unintentional intensity variations in the applied fields. Similarly, we show how
the spectroscopic lines of a transition can show an apparent frequency shift. We show quantified results for this shift and describe the best approach for minimizing this systematic error.

The relevance of our analysis is demonstrated with spectroscopy experiments on donor-bound exciton centers in Si-doped gallium arsenide. While our experiments and part of our calculations thus focus on one particular material system and experimental arrangement, the analysis and methods that we present here are of generic value for spectroscopy experiments on media with multi-level systems.

The paper is organized as follows. Section III presents simulations of DTS on a three-level lambda-system medium. We introduce a model to calculate differential signals and describe it both at the macroscopic scale of the measurement setup and in terms of the microscopic electron dynamics. Subsequently, this model is used to analyze the benefits of differential transmission and how possible disadvantages can be minimized. Section III provides experimental results of DTS on silicon donor centers in gallium arsenide, demonstrating the effects described by our model.

II. SIMULATIONS

A. Model

We model a setup where two continuous-wave laser fields co-propagate through a medium, as shown in Fig. 1(a). One field, depicted by its angular frequency $\omega_{\text{mod}}$, is undergoing on-off modulation at frequency $f_{\text{mod}}$ by a chopper. The other field, with angular frequency $\omega_{\text{nmod}}$ is not modulated. The total transmitted power is converted into an electrical signal by a linearly behaving photodiode after the sample. A lock-in amplifier isolates the electrical component at frequency $f_{\text{mod}}$ from all other components.

The modulation is orders of magnitude slower than the electronic dynamics in the medium and is assumed to have a square on-off envelop. This leads to two steady-state situations with transmittance $T_{\omega_{\text{mod}}}$ for the modulated field and transmittances $T_{\omega_{\text{nmod}}}$ and $T_{\omega_{\text{off}_{\text{nmod}}}}$ for the non-modulated field. If the probed medium has nonlinear components in the susceptibility, i.e. the susceptibility depends on the presence of laser fields, $T_{\omega_{\text{mod}}}$ and $T_{\omega_{\text{off}_{\text{nmod}}}}$ are in general not equal. Consequently, the transmittance of the non-modulated field is time-dependent with frequency $f_{\text{mod}}$ in the form of amplitude modulation of its transmission. This transfer of amplitude modulation from one field to another via the susceptibility will be called cross-modulation, and the part of the transmission at frequency $f_{\text{mod}}$ for the non-modulated beam will be called differential transmission. The frequency component at $f_{\text{mod}}$ of the total transmitted power thus

![FIG. 1](image-url)
consists of two parts and is given by

\[ P_{f_{\text{mod}}} = \frac{P_{\text{mod}} T_{\text{mod}}^{\text{on}}}{\text{normal transmission}} + P_{\text{mod}} (T_{\text{mod}}^{\text{on}} - T_{\text{mod}}^{\text{off}}) \]  

\text{(1)}

where \( P_{\text{mod}} \) (\( P_{\text{mod}} \)) is the power of the modulated (non-modulated) field, and where \( P_{\text{mod}} \) and \( P_{\text{mod}} \) are incident on the medium while \( P_{f_{\text{mod}}} \) is measured after the medium. Figure 1(b) illustrates the transmitted power for the individual fields and the total transmitted power, for qualitatively different levels of cross-modulation. If cross-modulation is absent, the contribution of differential transmission in Eq. (1) is zero, and the lock-in signal consists purely of the normal transmission of the modulated field. For increasing levels of cross-modulation, the differential transmission contributes more to the lock-in signal. When the differential transmission term is larger then the normal transmission term, and of opposite sign (which is the case for lambda systems), the trace of the total transmitted power shows a 180° phase shift. This phase shift manifests itself in the lock-in signal either as a negative signal or a 180° phase shift, depending on lock-in operation settings (for examples see Fig. 5 and Fig. 1(c), respectively).

The medium of interest consists of lambda systems, as shown in Fig. 1(c). Two ground states \( |a\rangle \) and \( |b\rangle \) have optical transitions to common excited states \( |e\rangle \), \( |e'\rangle \), etc. There is no optical transition between \( |a\rangle \) and \( |b\rangle \). Relaxation parameters \( \Gamma_{ij} \) describe both spontaneous emission rates from the excited states to the ground states and thermalization of population in the ground states. Furthermore, all states except \( |a\rangle \) undergo pure dephasing \( \gamma_i \), etc. For the sake of simplicity, the simulations will be restricted to three-level lambda systems. The experimental results in Sec. III however, show multiple excited states.

Differential transmission spectroscopy is modeled with two laser fields coupled to the optical transitions with transition dipole moments \( \mu_{\text{mod}} \) and \( \mu_{\text{nonmod}} \), see Fig. 1(d). One field is held resonant with its transition frequency, while the other is scanned over the resonance by changing its detuning \( \Delta \). It is assumed that each laser couples only to one transition. The lasers drive transitions between the levels at Rabi frequencies \( \Omega_{\text{mod}} \) and \( \Omega_{\text{nonmod}} \). The population in \( |e\rangle \) can spontaneously decay to both ground states, with relaxation rates \( \Gamma_{ij} \). Hence, a field present at transition \( \omega_{ae} \) (\( \omega_{be} \)) will pump population to state \( |b\rangle \) (\( |a\rangle \)), increasing the absorption coefficient for the field at transition \( \omega_{be} \) (\( \omega_{ae} \)). In Eq. (1) this results in the contribution \( (T_{\text{mod}}^{\text{on}} - T_{\text{mod}}^{\text{off}}) \), which is always negative for lambda systems. For the present analysis, we neglect effects related to coherent population trapping that can take place in a narrow spectral window around two-photon resonance when two lasers drive a lambda system. The amount of population pumped from one low-energy state of the lambda system to the other depends on the ratio of relaxation coefficients \( \Gamma_{ae} \) and \( \Gamma_{eb} \). The relaxation coefficient of a transition is related to its electric dipole moments by

\[ \Gamma_{ij} = \frac{2n\omega_{ij}^3}{3e_{0}hc^3} \]  

\text{(2)}

where \( n \) is the bulk refractive index. The branching ratio \( \Gamma_{ea}/\Gamma_{eb} \) is given by

\[ \frac{\Gamma_{ea}}{\Gamma_{eb}} = \frac{\omega_{ea}^2}{\omega_{eb}^2} \approx \left( \frac{\mu_{\text{mod}}}{\mu_{\text{nonmod}}} \right)^2, \quad \text{for } \omega_{ae} \approx \omega_{be} \]  

\text{(3)}

The ratio \( \mu_{\text{mod}}/\mu_{\text{nonmod}} \), which we will call relative dipole moment, is the main parameter that determines the amount of cross-modulation and differential transmission (Fig. 2).

The transmittances are determined by the imaginary part of the susceptibility by

\[ T(\omega) = e^{-z \left[ \chi(\omega) \right]} \]  

\text{(4)}

where \( z \) is the sample thickness and \( c \) is the speed of light in vacuum. For each field \( \chi(\omega) \) is calculated by

\[ \chi(\omega) = \frac{2N\mu_{ij}^2 \sigma_{ij}(\omega)}{e_{0}hE(\omega)} \]  

\text{(5)}

where \( N \) is the number density of lambda systems in the material, \( E \) is the amplitude of the oscillating electric field, and \( \sigma_{ij} \) is the slowly oscillating part of the transition’s coherence. The coherence \( \sigma_{ij} \) is obtained from the steady-state solution of the master equation for the density operator

\[ \frac{d\rho}{dt} = -i [\hat{H}, \rho] + \hat{L}(\rho) \]  

\text{(6)}

where \( \hat{H} \) is the Hamiltonian of the laser-driven system in Fig. 1(c, d) (using a standard approach). The Lindblad operator \( \hat{L} \) includes all relaxation and decoherence rates. We solve the master equation numerically, applying the rotating wave approximation.

B. Results

Figure 2(a) shows DTS traces for three different values for the relative dipole moments. The values of the dipole moment varied in a manner that keeps the total relaxation rate \( \Gamma_{ea} + \Gamma_{eb} \) from the excited state the same for the three cases. The solid lines present results of Eq. (1) normalized to \( P_{\text{mod}} \), and represent the total signal as seen by the lock-in. The dashed lines present (on the same scale) the normal transmission of the modulated laser, such that the difference with the solid lines signifies the magnitude of the differential transmission part. The Rabi frequencies are \( \Omega_{\text{mod}} = 10 \text{ MHz} \) for the modulated laser and \( \Omega_{\text{nonmod}} = 0.5 \text{ GHz} \) for the non-modulated laser, for all three cases. A normalized measure of the transition visibility, which we will call dip depth, can be
FIG. 2. Simulation of normalized differential transmission (NDT) for three different relative dipole moments $\mu_{nmod}/\mu_{mod}$. a) NDT while scanning the modulated beam over the resonance, with $\Omega_{nmod} = 10$ MHz and $\Omega_{nmod} = 0.5$ GHz. The full lines show the total signal as seen by the lock-in, which consists of normal transmission of the modulated field and differential transmission of the non-modulated field due to cross-modulation. The dashed lines show the contribution of purely the transmission of the modulated field. b) Enhanced transition visibility as a function of Rabi frequency of the non-modulated laser, with $\Omega_{nmod} = 10$ MHz. The full lines show the dip depth of the total differential signal, while the dashed lines show only the normal transmission of the modulated field. Cross-modulation dramatically enhances the transition visibility for systems with a small relative dipole moment (top panel).

assigned to each scan as shown for the deepest scan. Figure 2(b) shows the dip depth as a function of $\Omega_{nmod}$, again comparing the total lock-in signal with the normal transmission part.

For lambda systems with a small relative dipole moment (in Fig. 2 shown for $\mu_{nmod}/\mu_{mod} = 0.1$), the visibility of the transition is strongly enhanced by cross-modulation. At high $\Omega_{nmod}$, the differential transmission is two orders of magnitude larger than the normal transmission (top panel Fig. 2(b)). For systems with a large relative dipole moment (shown for $\mu_{nmod}/\mu_{nmod} = 10$), differential transmission is negligible, since the amount of cross-modulation is low. The absorption of the modulated laser is sufficient to have a good transition visibility at both low and high $\Omega_{nmod}$ (bottom panel Fig. 2(b)). Here the presence of a strong non-modulated laser only slightly enhances the absorption of the modulated beam and hence the dip depth. In the intermediate case (shown for $\mu_{nmod}/\mu_{nmod} = 1$) DTS approximately doubles the dip depth (middle panel Fig. 2(b)). For a range of $\mu_{mod}/\mu_{nmod}$ values, strong cross-modulation can increase the dip depth to values greater than 1. This yields a negative DTS signal, as can be seen in Fig. 2(a) (solid trace for $\mu_{mod}/\mu_{nmod} = 1$) and in the top two panels of Fig. 2(b) at high $\Omega_{nmod}$. Whether this occurs, depends on both the relative and absolute dipole moments (for smaller absolute dipole moments the results are very similar to Fig. 2 with smaller dip depths).

Optical transitions in spectroscopy are observed as peaks or dips against a background signal. When this appears on a background signal with a slope or even additional structure (from other effects), it is harder to distinguish transitions and this also causes an offset for a peak or dip that should identify the transition frequency. In addition, as we analyze below here, one can have apparent shifts in the transition frequency when the intensity of the scanning laser is not constant while scanning over the resonance. For this latter effect, the causes can be in the experimental setup (for example changes in laser power during frequency scans) or fundamental to the sample (for example due to Fabry-Perot cavity effects inside the sample, for an experimental example see Fig. 3(a)).

We modeled the effect of intensity changes on the observed transition frequency, comparing the apparent shift when scanning the modulated or the non-modulated laser. We change the intensity of the scanning laser linearly with frequency. The unit of this intensity change is percentage per GHz scan range, with the intensity at the transition center defined as 100%. The point that appears lowest in a spectroscopic dip and the real transition frequency, $(\Delta I/I)/\Delta \omega$ is the percentage change in laser intensity per GHz scan range. Full (dashed) lines show the apparent shift while scanning the non-modulated (modulated) laser. Results for $\Omega_{nmod} = 10$ MHz and $\Omega_{nmod} = 0.5$ GHz.
a GHz can be obtained for realistic changes in intensity over scan range. The error is largest for small relative dipole moments. However, this error can be suppressed a factor of three by scanning the non-modulated laser. On the contrary, larger relative dipole moments show an enhanced spectroscopic error when the non-modulated laser is scanned. Scanning the non-modulated laser always results in a transition shift with the same sign as the intensity change (solid lines in the three panels). The sign of the error when scanning the modulated laser is less straightforward and depends on the competition between opposite shifts in the normal and differential transmission parts of Eq. (1).

To derive quantitative information on the relative and absolute strength of the optical transition dipole moments from such DTS data, a single DTS trace does not provide sufficient information. Instead, one needs to take data for a range of $\Omega_{\text{mod}}$ and $\Omega_{\text{nmod}}$ values, and fit the observed trends in dip depth to traces as in Fig. 2. If not known from independent measurements, the fitting should also yield parameters for the decay and dephasing parameters, and the optical Rabi frequencies. The non-linear behavior of the overall system allows for such a multi-parameter fitting analysis. Notably, this approach is thus also needed if one wishes to unravel the combined DTS signal from two co-propagating fields into the separate transmission components.

III. EXPERIMENTS ON SI:GAAS

A. Material and methods

We used DTS as a spectroscopic technique to find the optical transitions in lambda systems provided by silicon donors in epitaxial gallium arsenide. We studied epitaxially grown samples of 10 $\mu$m thickness and with a donor concentration of $\sim 3 \times 10^{13}$ cm$^{-3}$. The samples were studied at a temperature of 4.2 K, and in an applied magnetic field of 6.5 T. At such low donor concentrations and low temperature the donor centers are not ionized, thus providing an ensemble of localized electrons (so-called $D^0$ systems). These electrons have selective optical transitions from the two Zeeman-split spin $S = \frac{1}{2}$ levels of the $D^0$ ground state to a donor-bound exciton complex ($D^0 X$ system). The $D^0 X$ system consists of two electrons and one hole bound at the donor side, and it has a frequency window of tens of GHz a range of energy levels due to quantum confinement around the donor site and Zeeman shifts from the spin of the three particles. In our study the propagation of optical fields was in Voigt geometry. Two co-propagating laser fields were guided to the sample through a single mode fiber, and we had a detector directly behind the sample for recording DTS signals (for further details see Refs. 2 and 3). The results presented here were obtained with a scanning laser field of 50 mW cm$^{-2}$ on the sample, and a fixed laser of significantly higher intensity (presented in units of $I_0 = 0.5$ W cm$^{-2}$).

For applying the DTS technique and getting the results in Figs. 4 and 5, one laser was fixed at the optical transition in the spectrum from the $D^0$ spin-down state to the lowest level $|e\rangle$ of the $D^0$ complex (see Fig. 1(c.d)), while the other laser was scanned to probe the transitions from the $D^0$ spin-up state to the sequence of levels $|e\rangle, |e'\rangle, \ldots$ etc. The laser fields had linear polarizations and were for the examples presented here parallel to the magnetic field for the fixed laser, and orthogonal for the scanning laser. Strictly speaking, the two-laser driving thus only addressed a three-level lambda systems for probing level $|e\rangle$. However, for the subsequent levels $|e\rangle, \ldots$ the two-laser approach counter-acts optical pumping into one of the $D^0$ spin states, and for the investigation of how this yields cross-modulation signals it can for each transition be analyzed using our three-level modeling.

B. Results

Figure 4 shows DTS results, resonances with $D^0$-$D^0 X$ transitions appear as dips in transmission. In Fig. 4(a), the visibility of transitions is deteriorated by a strongly varying intensity background, caused by the Fabry-Perot effect inside the sample experienced by both lasers (it
is a chirped Fabry-Perot effect due to the very strong free-exciton absorption line at \( \sim 366900 \) GHz). Here the modulated laser was used for spectroscopic scanning. Conversely, Fig. 3(b) presents results from a nominally identical sample, from spectroscopic scanning the non-modulated laser. Notably, one can now clearly recognize at least six \( D^0-D^X \) resonance lines in the experimental data (the Fabry-Perot resonances are almost not visible anymore). Transitions now appear on a flat background (the shift in spectral position of the lines for Fig. 3(a, b) is here for a significant part due to a different amount of strain in the sample). The field of the scanning non-modulated laser only causes a strong contribution to the DTS signal when it scans over a transition, and the full signal is thereby much less sensitive to the Fabry-Perot modulation of the intensity inside the sample for this laser field. This technique hence provides clear high resolution spectroscopy data, free from the clutter with the underlying Fabry-Perot pattern. Figure 3(c) shows traces taken with the approach as for Fig. 3(a), where the non-modulated laser intensity is varied. Both a flip of the lock-in signal and a shift in the transition of hundreds of MHz can be observed. We ran simulations for the apparent transition shift with different \( \Omega_{\text{mod}} \), using changes in intensity as observed in the Fabry-Perot background, and found shifts very similar to these experimentally observed shifts.

Figure 5 presents experimental traces taken with the modulated laser resonant with one transition and the non-modulated laser scanning over another transition. The intensity of the non-modulated laser is varied. The dip depth increases with power, starting at a normalized value around 0.5 and saturating slightly above 1. This provides an example of how strong cross-modulation yields enhanced signals. We performed a fit of this sequence of traces with the theory from Eqs. (4)-(6) (yielding traces as in Fig. 2(a)). This yields equal dipole moments of \( \sim 2 \) emu for the two transitions involved.

IV. CONCLUSION

We have analyzed how low-frequency amplitude modulation in two-laser spectroscopy yields differential signals for lock-in detection with a single photo diode. We applied this to DTS studies of a medium with lambda systems. Cross-modulation provides a very useful tool for enhancing the visibility of transitions that are otherwise difficult to observe. We quantified this enhancement and showed how the resulting spectroscopy provides information about the underlying dipole moments. Choosing which laser to scan can improve signal disturbance from laser-field intensity changes, and minimize the spectroscopic error due to these effects. Experimental DTS results on Si:GaAs show the relevance and validity of our model. We envision that our modeling can be used for spectroscopy on a wider range of systems with multiple levels in the ground and excited state.

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