Exploring the limits of single emitter detection in fluorescence and extinction

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We present an experimental comparison and a theoretical analysis of the signal-to-noise ratios in fluorescence and extinction spectroscopy of a single emitter. We show that extinction measurements can be advantageous if the emitter is weakly excited. Furthermore, we discuss the potential of this method for the detection and spectroscopy of weakly emitting systems such as rare earth ions.

The progress of nanoscience and technology in the past two decades has been accompanied by a growing interest in the optical study of single nano-objects [1]. A major thrust in this research area came from cryogenic spectroscopy [2, 3] as well as room temperature detection [4] and microscopy [5, 6] of dye molecules. Although a fluorescent atom suspended in vacuum can be seen even by the naked eye, achieving a high signal-to-noise ratio (SNR) in the detection of single molecules is a nontrivial task in the condensed phase. In particular, the background light and noise associated with the fluorescence or scattering from the environment can easily dominate the small signal of a single emitter. Furthermore, the dark counts and noise of photodetectors put a limit on the lowest signals that one might hope to detect.

As shown in Fig. 1a, the level scheme of a fluorescent molecule consists of vibrational manifolds in the electronic ground (g) and excited (e) states. For an appropriate combination of an emitter and its surrounding matrix, the linewidth of the so-called zero-phonon line (ZPL) of the 0-0 transition between the vibrational ground states of g and e can become lifetime limited at cryogenic temperatures, thus enhancing the emitter's absorption cross section σ [7]. A very successful method for detecting a single molecule with a narrow 0-0 ZPL has been fluorescence excitation spectroscopy [3] where the red-shifted incoherent fluorescence of the molecule at wavelength λ_{\text{red}} is separated from the light at the laser wavelength λ_{\text{las}} by using high quality spectral filters. The SNR of this technique is determined on the one hand by the detector noise, which can be as low as 20-100 counts per second (cps) for very good avalanche photodiode single photon counters. On the other hand, saturation limits the maximum attainable signal to typical values of $10^5 - 10^6$ cps.

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on the detector for a good dye molecule. Thus, fluorescence excitation spectroscopy can enjoy a
very healthy SNR when applied to strongly fluorescent systems. Detection of very weak emitters,
however, remains a challenge. In particular, fluorescence detection of single rare earth ions has
been hampered owing to their long lifetimes and therefore ultra weak fluorescence.

FIG. 1: a) The level scheme of a dye molecule. b) The schematics of our experimental setup. A laser
beam is focused onto the sample using an aspheric lens (AL) and a hemispherical solid-immersion lens
(SIL). A second aspherical lens is used to collect the transmitted laser beam as well as the forward emitted
fluorescence of the molecule. LP: long pass filter, SP: short pass filter, PD: photodetector.

An alternative approach to the detection of single solid-state emitters is to go back to the
first method that was applied in single molecule spectroscopy [2], namely to detect the extinction
of the laser light caused by a single molecule in its path. This method was successfully revived
by Plakhotnik and Palm in 2001 [8] where the coherent scattering of the excitation light was
interfered with the residual reflections from the interfaces in the setup. Closely related efforts
followed on quantum dots, especially with the aim of acquiring access to the linewidth of the
main optical transition in these systems [9, 10]. Recently, we have extended this approach to
detect the extinction of a laser beam by a single molecule in transmission without the need for any
noise suppression technique [11, 12, 13]. In this paper, we compare the conventional fluorescence
excitation technique with extinction measurements in terms of the SNR and discuss the potential
of the latter for detecting emitters with very weak optical transitions.

The experimental arrangement of our discussion is depicted in Fig. 1b, and its details are
described in Refs. [11, 13]. Briefly, the excitation laser light was focused onto the sample consisting
of DBATT molecules embedded in a n-tetradecane matrix inside a cryostat. For this we used an
aspheric lens with a numerical aperture of 0.68 and a cubic zirconia hemispherical solid immersion
lens (SIL). After interaction with the sample, a second aspheric lens collimates the beam and directs
it to an avalanche photodiode (PD). Two different filter sets are used to either reject $\lambda_{\text{las}}$ and detect
$\lambda_{\text{red}}$ or vice versa. The former arrangement delivers a fluorescence excitation spectrum while the
latter allows a direct resonant measurement. Fig. 2 shows examples of fluorescence and extinction
spectra recorded from the same single molecule and on the same detector at three different incident powers. In this article we adopt the unit of counts per second (cps) for power. When the detected laser power reads \(10^6\) cps on PD (corresponding to an excitation regime well below saturation) both extinction (a) and fluorescence (b) yield comparable SNR of \(\approx 100\ \sqrt{\text{Hz}}\). For a detected laser power of \(3.2 \times 10^4\) cps, the fluorescence of the molecule is hardly above the detector dark count rate of 100 cps. However, the extinction is still easily observable at 10% visibility. Even at an ultra-low illumination level of 2000 cps the extinction signal succeeds in detecting the molecule whereas the fluorescence peak is fully buried under the detector noise.

\[
P = \frac{\varepsilon_0 c}{2 \sqrt{\omega}} \int_{\Omega} \left( \langle \hat{E}_{\text{las}}^- \cdot \hat{E}_{\text{las}}^+ \rangle + \langle \hat{E}_{\text{m}}^- \cdot \hat{E}_{\text{m}}^+ \rangle + 2 \Re \langle \hat{E}_{\text{las}}^- \cdot \hat{E}_{\text{m}}^+ \rangle \right) d\Omega
\]

\[
= P_{\text{las}} + P_{\Omega} - P_{\text{ext}}
\]

Assuming a perfect transmission channel and detector, the power on PD in the absence of any spectral filter is given in cps by

\[
P = \frac{\varepsilon_0 c}{2 \sqrt{\omega}} \int_{\Omega} \left( \langle \hat{E}_{\text{las}}^- \cdot \hat{E}_{\text{las}}^+ \rangle + \langle \hat{E}_{\text{m}}^- \cdot \hat{E}_{\text{m}}^+ \rangle + 2 \Re \langle \hat{E}_{\text{las}}^- \cdot \hat{E}_{\text{m}}^+ \rangle \right) d\Omega
\]

\[
= P_{\text{las}} + P_{\Omega} - P_{\text{ext}}
\]

where \(E_{\text{las}}\) and \(E_{\text{m}}\) represent the electric fields associated with the laser and the molecular emission at the detector, respectively, and \(\omega\) is the frequency of the emitted photon. \(\Omega\) denotes the solid angle of light collection and is assumed to cover all the transmitted laser light. The molecular emission \(P_{\Omega}\) consists of a part that originates from the 0-0 ZPL transition and is resonant with the laser light and a red-shifted component which results from molecular and lattice vibronic transitions.
The electric field associated with the coherent part of the resonance fluorescence \[13, 14\] gives rise to a nonzero third term \( P_{\text{ext}} \) of Eq. (1), signifying the interference between the molecular emission and the laser beam. This component, which is known as the “extinction” term \[15\] is equivalent to a homodyne signal where the excitation laser beam acts as the local oscillator \[16, 17\].

It is helpful for the following discussion to write the terms of Eq. (1) in an explicit manner:

\[
\begin{align*}
    P_{\text{las}} &= \frac{\varepsilon_0 c^2}{2\hbar\omega} \int_{\Omega} \langle \hat{E}_{\text{las}}^- \cdot \hat{E}_{\text{las}}^+ \rangle \, d\Omega \\
    P_m^{4\pi} &= \frac{\varepsilon_0 c^2}{2\hbar\omega} \int_{4\pi} \langle \hat{E}_m^- \cdot \hat{E}_m^+ \rangle \, d\Omega = \Gamma_1 \rho_{22} = \frac{\Gamma_1}{2} \frac{S}{1 + S} \\
    P_{\Omega}^\Omega &= \zeta P_m^{4\pi} \\
    P_m^{\text{res}} &= \alpha P_m^\Omega \\
    P_m^{\text{red}} &= (1 - \alpha) P_m^\Omega \\
    P_{\text{ext}} &= -\frac{\varepsilon_0 c^2}{2\hbar\omega} \int_{\Omega} 2\Re \langle \hat{E}_{\text{las}}^- \cdot \hat{E}_m^+ \rangle \, d\Omega .
\end{align*}
\]

(2)

The quantity \( P_m^{4\pi} \) gives the total power emitted by the molecule into the \( 4\pi \) solid angle. \( \rho_{22} \) is the population of the excited state, and the on-resonance saturation parameter \( S \) reads \[14\]

\[
S = \frac{\nu^2}{\Gamma_1 \Gamma_2},
\]

(3)

where \( \nu \) is the Rabi frequency defined by \( \hbar\nu = d_{\text{ZPL}} \cdot E_{\text{las}}(O) \). The transition dipole moment \( d_{\text{ZPL}} \) and the incident electric field \( E_{\text{las}}(O) \) at position of the molecule are assumed to be parallel for simplicity. The factor \( \alpha \) describes the ratio of the power emitted on the 0-0 ZPL to the total excited state emission. Thus, \( d_{\text{ZPL}} = \sqrt{\alpha} d_{\text{eg}} \) where \( d_{\text{eg}} \) denotes the dipole moment associated with the total spontaneous emission rate of the excited state given by \( \Gamma_1 = d_{\text{eg}}^2 \omega^3/(3\pi \varepsilon_0 \hbar c^3) \). \( \Gamma_2 \) represents the transverse decay rate which equals \( \Gamma_1/2 \) in the absence of any dephasing. The parameter \( \zeta \) signifies the fraction of the total emitted molecular power to that collected into the detection solid angle \( \Omega \). We note that in addition, one might have to account for total internal reflection and waveguiding in the substrate which influence the angular distribution of the laser light and the molecular emission \[11\]. Finally, the quantities \( P_m^{\text{res}} \) and \( P_m^{\text{red}} \) represent the portions of the molecular emission into the solid angle \( \Omega \) that are resonant with the excitation laser and red shifted from it, respectively.

It is now instructive to separate the properties of the laser beam from the spectroscopic features of the emitter. Using the definitions of \( \Gamma_1 \) and \( \nu \), one can rearrange the saturation parameter in Eq. (3) to read

\[
S = \frac{\alpha}{\Gamma_2} K P_{\text{las}}
\]

(4)
where $K$ is a unitless geometrical factor that relates $|E_{\text{las}}(O)|^2$ to the laser power $P_{\text{las}}$. More precisely, $K$ denotes the ratio of the total power scattered by a weakly excited two-level system and the incident power. It depends on the spatial mode of the laser beam and the focusing optics. The reader is referred to Ref. [18] for details.

The expressions in Eq. (2) provide us with the red shifted fluorescence $P_{\text{m}}^{\text{red}}$. The noise on this signal is given by the fluctuations in the detector dark counts $P_{\text{drk}}$ if we assume that the excitation light is completely rejected by the filters. Thus, the SNR for a fluorescence excitation measurement becomes

$$\text{SNR}_{\text{red}} = \frac{\mu P_{\text{m}}^{\text{red}}}{N_{\text{red}}} = \frac{\mu \zeta (1 - \alpha) \Gamma_1}{2 \sqrt{P_{\text{drk}}}} \frac{S}{1 + S},$$

where we have introduced $\mu$ to account for losses (e.g. cryostat windows, filters, etc.) and the detector efficiency. The SNR maximum is given by $\text{SNR}_{\text{red}}^{\text{max}} = \mu \zeta (1 - \alpha) \Gamma_1 / (2 \sqrt{P_{\text{drk}}})$ and occurs in the fully saturated regime.

Considering that the solid angle $\Omega$ collects all the incident laser light, a simple energy balance argument implies that $P_{\text{ext}}$ in Eq. (1) must correspond to the total power $P_{\text{m}}^{4\pi}$ emitted by the molecule. Now we insert a spectral filter to select only the part of the transmitted light that is resonant with the laser light. Denoting the size of the dip in the power that is detected in this case by $P_{\text{dip}}^{\text{res}}$, Eq. (1) and Eqs. (2) yield,

$$P_{\text{dip}}^{\text{res}} = P_{\text{ext}} - P_{\text{m}}^{\text{res}} = P_{\text{m}}^{4\pi} - P_{\text{m}}^{\text{res}} = (1 - \alpha \zeta) \frac{\Gamma_1}{2} \frac{S}{1 + S}.$$  

The noise on a resonant extinction measurement is composed of the shot noise $\sqrt{P_{\text{las}}}$ of the laser power, the laser intensity fluctuations $\kappa P_{\text{las}}$ where $\kappa$ is a proportionality constant, and $\sqrt{P_{\text{drk}}}$. Since these contributions are statistically independent, the total noise can be written as $N_{\text{res}} = \sqrt{\mu P_{\text{las}} + (\mu \kappa P_{\text{las}})^2 + P_{\text{drk}}}$, where again, $\mu$ accounts for losses and the detection efficiency. Assuming that intensity fluctuations have been mastered at a sufficient level and that $\mu P_{\text{las}} \gg P_{\text{drk}}$, one finds $N_{\text{res}} \simeq \sqrt{\mu P_{\text{las}}}$. Thus, the signal-to-noise ratio for an extinction measurement becomes

$$\text{SNR}_{\text{res}} = \frac{\mu P_{\text{dip}}^{\text{res}}}{N_{\text{res}}} \simeq (1 - \zeta \alpha) \frac{\Gamma_1}{2} \sqrt{\frac{\mu \alpha K}{\Gamma_2}} \frac{\sqrt{S}}{1 + S}.$$  

Fig. 3 presents $\text{SNR}_{\text{res}}$ as a function of the detected laser power $\mu P_{\text{las}}$ and of the saturation parameter $S$. In each case, $S$ was directly derived from the power broadened linewidth of the fluorescence excitation spectrum. Our system could perform at the shot-noise limit down to the sub Hertz bandwidth over the whole power range presented here. The green theoretical fit curve is obtained using Eq. (7). With the parameters that have been independently determined for our setup ($\mu = 0.2$, $\zeta = 0.8$).
$\kappa=0.5, \alpha = 0.2, \Gamma_2 = \Gamma_1/2, \Gamma_1/2\pi = 17 \text{ MHz}$), an excellent agreement with the measured data is achieved. The deterioration of $\text{SNR}_{\text{res}}$ under very strong excitation is clearly visible and stems from the fact that for a quantum emitter, $P_{\text{dip}}^{\text{res}}$ saturates at high incident powers. The maximal attainable SNR in a shot-noise limited resonant detection then becomes $\text{SNR}_{\text{res}} = \sqrt{\Gamma_1^2 \alpha \kappa \mu/(16\Gamma_2)}$ and occurs at $S=1$ if we assume $\zeta\alpha \ll 1$.

![Graph showing signal-to-noise ratio as a function of saturation parameter and detected power.](image)

**FIG. 3:** The signal-to-noise ratio of a transmission measurement at the laser frequency as a function of the the laser power on the detector and of the saturation parameter.

To compare the SNR of fluorescence and extinction measurements directly, we have recorded spectra such as those shown in Fig. 2 for low excitation powers corresponding to $S = 6 \times 10^{-6}$ to $10^{-2}$ as shown in Fig. 4. The circles and the triangles display the SNR for the extinction and fluorescence detections, respectively. To determine the experimental SNR, we first fitted the spectra with Lorentzian functions. Then we extracted the on-resonant signal and divided it by the off-resonant rms noise. The data were recorded by adding 100 scans with 10 ms integration time, corresponding to a total acquisition time of 1 second per frequency pixel. This procedure helped to correct for possible laser drifts and spectral diffusion of the molecule. The fitted green and red theoretical curves depend on $\sqrt{S/(1+S)}$ and $S/(1+S)$, respectively (see Eqs. (5) and (7)) and show a very good agreement with the experimental data. We conclude that in case of a weakly excited system, an extinction measurement can be superior to fluorescence detection in terms of SNR. We point out in passing that both $\text{SNR}_{\text{red}}$ and $\text{SNR}_{\text{res}}$ scale as the square root of the integration time and thus, the comparison between the fluorescence and extinction methods holds for fast and slow measurements alike.

We remark that in the existing literature, the root-mean-square (rms) fluctuation of the signal
FIG. 4: The signal-to-noise ratios of the resonant transmission (green) and fluorescence (red) signals as a function of the excitation power in the weak excitation regime. Symbols display the experimental data and the lines denote theoretically expected behavior.

An exciting question that arises is whether extinction detection opens doors for studying weakly fluorescent nano-objects. Conventional single molecule detection has been successful for molecules that have fluorescence lifetimes of a few nanoseconds, corresponding to $\Gamma_1/2\pi \sim 10 - 100$ MHz. Such a high photon flux provides a good SNR even considering realistic collection plus detection efficiency of a few percent and $P_{\text{dark}} = 100$ cps. However, for weakly emitting systems such as rare earth ions with lifetimes of the order of milliseconds, $\text{SNR}_{\text{red}}$ becomes comparable or smaller than unity. Fig. 5 displays the expected $\text{SNR}_{\text{res}}$ as a function of the detected laser power for various radiative decay rates $\Gamma_1$. Here we have assumed a laboratory value of $\mathcal{K} = 0.5$, $\alpha = \mu = 1$, $\Gamma_2 = \Gamma_1/2$, and $P_{\text{dark}} = 20$ cps, but extension of the results to other situations is straightforward by following Eq. (7). These plots indicate that single emitters with spontaneous emission times as long as a millisecond should be detectable using extinction spectroscopy even when realistic detection parameters (e.g. $\mathcal{K} = 0.5, \alpha = 0.5, \mu = 0.2$) are considered. In addition, we emphasize that extinction measurements have the great added value that they provide direct access to the

itself has been often included as a noise source in fluorescence but ignored in extinction measurements [19, 20]. In our comparison of the two methods, we have consistently chosen to define the signal as the response of the system of interest, namely a single emitter, and the noise as all fluctuations stemming from other sources. Therefore, we do not include the signal rms noise in our analysis of SNR. This strategy is particularly convenient for the evaluation of SNR from single spectra.
coherent interaction of the incident light and the emitter.

![Graph showing SNR vs Detected laser power for different decay rates.](image)

FIG. 5: The SNR for a resonant transmission detection of emitters with different radiative decay rates. Here we have assumed $\alpha = \mu = 1$, $P_{\text{dark}}=20$ cps, and $K = 0.5$. $\Gamma_1$ is given in units of rad/sec.

Although the basic concepts discussed in this paper have been known in signal processing and electrical engineering [16], their direct experimental investigations at the single emitter level have been made possible through advances in cryogenic spectroscopy [8, 13, 21]. Inspired by this progress, very recently we have also succeeded in extinction detection of a single solid-state quantum emitter at room temperature [22] despite the fact that the extinction cross section is reduced by 5-6 orders of magnitude due to severe broadening of the transition ($\Gamma_2 \gg \Gamma_1$). Another interesting application of extinction or homodyne detection has been demonstrated almost independently for imaging small metallic and dielectric nanoparticles [23, 24, 25, 26, 27, 28]. Conventional methods of nanoparticle detection such as dark-field [29] or total internal reflection [30] microscopy rely on the elimination of the incident light from the detection path and the detection of the power scattered by the particles. To this end, these techniques are analogous to fluorescence excitation spectroscopy where frequency spectra are replaced by spatial images, and spatial filtering substitutes spectral filtering for the discrimination of the incident laser light. However, in practice the two systems are limited in different ways because in the case of spatial imaging, the persistent source of ”noise” is the light scattered from residual optical roughness of the medium [26]. The equivalent of this problem usually does not arise in extinction detection of emitters because they are typically embedded in well-behaved matrices [1, 2] without any optical transitions in the spectral region of interest.

In conclusion, we have explored the signal-to-noise ratio in the spectroscopic detection of single emitters. We have provided expressions for evaluating the performance of both fluorescence and extinction measurements. In particular, we have demonstrated that extinction measurements can be superior to fluorescence detection in the weak excitation regime. Furthermore, we have shown
that even weakly fluorescent emitters should be detectable using coherent extinction spectroscopy. This prospect is especially interesting for the optical storage and read out of quantum information in new systems such as rare earth ions.

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