# Overcoming inhomogeneous broadening from ensembles of quantum emitters in noisy environments with external control fields

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## ABSTRACT

In an ensembles of nominally identical quantum emitters, each emitter can have its emission frequency shifted randomly by its specific environment so that the emission spectrum of the overall system is inhomogeneously broadened over a large frequency range. This can make the system hard to probe and to utilize for a variety of applications. We show that it is possible, with realistic external control field protocols, to refocus the emission spectrum of the ensemble onto a lineshape with a well-defined central peak that has the linewidth of an individual isolated non-noisy emitter.

**Keywords:** Ensemble, quantum emitters, spectral control, inhomogeneous broadening

# **1. INTRODUCTION**

A quantum emitter in its minimal representation can be described as a two-level system with ground state  $|q\rangle$ and excited state  $|e\rangle$  with respective energy levels  $E_g$  and  $E_e$ . The states of such a quantum system in the solid state can be modified by fluctuations in its environment (e.g. charge, spin or strain). These dynamic processes can be essentially random and thus lead to the emission frequency  $\omega = (E_e - E_q)\hbar$  drifting randomly away from a target frequency  $\omega_0$ .<sup>1</sup> This behavior, spectral diffusion or spectral wandering, can have far-reaching effects on properties of an emitter and its usage in various applications, including as a qubit in quantum information processing operations. Hence, being able to mitigate it is of significant value. For the purpose of quantum information processing, the two-level system that makes up the quantum bit should ideally be well isolated from its environment in order to protect it from decoherence however some parasitic coupling typically persists for solid state systems. Several quantum emitters have shown great promise to be used as quantum bits. This includes quantum dots, and color centers such as nitrogen-vacancy (NV) Centers in diamond. These solid state emitters are all subject, to various degrees, to spectral diffusion and to inhomogeneous environments.

We have previously demonstrated that it is possible to suppress spectral diffusion with an externally applied pulse sequence<sup>2-4</sup> and that this can in turn enhance photon indistinguishability between quantum emitters in different environments.<sup>5</sup> Furthermore, the control protocols are effective even when the pulses are broadly imperfect.<sup>6</sup>

In an ensemble of quantum emitters, fluctuations in the environments and spectral diffusion are often manifested in spectroscopy experiments. Here, several emission peaks corresponding to different emitters, each wandering spectrally along its own path, may only partially overlap; Resulting in an emission spectrum for the ensemble that is made of a broad ill-defined single feature.<sup>1,7–9</sup> This can make it difficult to resolve spectral features of the system or to use ensembles of quantum emitters in applications such quantum sensing with ensembles of NV centers  $^{10-13}$  .

In this paper, we consider an ensemble of two-level systems in a dynamic environment in the dilute limit so that photon-mediated dipole-dipole interaction can be neglected. As a result of the fluctuating environment, emitters within the ensemble have their specific emission frequencies distributed according to a random distribution, which we consider here to be Gaussian over broad range of frequencies with a given standard deviation

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around a given average value. The emission spectrum of the ensemble is then spread over a frequency range that can be orders of magnitude greater than that of an individual isolated emitter. We place the ensemble under the influence of a periodic sequence of finite-width pulses an evaluate its emission spectrum. We show that the emission spectrum of the ensemble that has a broad Gaussian lineshape in the absence of the control protocol is refocused by the control protocol onto a lineshape with a central peak at the pulse carrier frequency. The central peak of this refocused spectrum has the linewidth of an individual isolated non-noisy emitter and is flanked by satellite peaks at frequencies determined by the inter-pulse delay. These procedures can facilitate the use of ensembles of quantum emitters in various applications such as optimal interfaces with inhomogeneously broadened ensembles of quantum emitters in quantum information processing.



Figure 1. Schematic representation of inhomogeneous broadening in the emission spectrum of an ensemble of two-level systems. (a) Due to variations in the environments, emitter *i* with  $i = 1, 2, 3, \cdots$  has an emission frequency that is detuned by  $\Delta_i$  from a target frequency  $\omega_0$ . (b) Each individual emitter has an emission spectrum with a Lorentzian lineshape centered around its emission frequency. (c) As a result of the random distribution of detunings across the ensemble, the overall emission spectrum of the ensemble is broadened over a wide frequency range with ill-defined features.

## 2. MODEL AND SOLUTION

We consider an ensemble of two-level systems (e.g. NV centers) under the influence of a control protocol made of equidistant finite-time pulses. The two-level systems have their respective emission frequencies ( $\omega_i$ 's) spread over a range such that the detunings, with respect to a target frequency  $\omega_0$ ,  $\Delta_i = \omega_i - \omega_0$ , follow a Gaussian distribution defined by

$$P(\Delta_i) = N(0, \Delta_i)$$
 with  $N(0, \Delta_i) = \frac{1}{\Delta_0 \sqrt{2\pi}} e^{-\frac{\Delta_i^2}{2\Delta_0^2}}$ 

i.e. average value 0 and  $stdev = \Delta_0$ .

We aim to determine the emission profile of this ensemble when it is placed under the influence of a control protocol consisting of periodically applied optical pulses. Namely, we periodically apply a field of amplitude  $\Omega_x$  and frequency  $\omega_0$  for a time  $t_p = \pi/\Omega_x$ : The system undergoes free evolution for time  $\tau - t_p$  followed by evolution in the presence of the pulse field for time  $t_p$  and this process is repeated periodically with a free evolution for time  $\tau - t_p$  followed by the application of the field for time  $t_p$ . An ideal  $\pi$  pulse has the effect of swapping the populations of the excited and ground states as well as the coherences of the two-level system.

We assume the system to be in the dilute limit, where direct dipole-dipole interactions and photon-mediated dipole-dipole interactions can be neglected, so that the emission spectrum of the ensemble can be obtained by adding up contributions from individual emitters. To obtain the emission spectrum of each emitter, we use the master equation and iteratively evolve the density matrix numerically on a discretized time axis.<sup>4,14</sup>

We assume that the emitter is initially in its excited state. The width at half maximum of the Lorentzian lineshape for the emission spectrum of an isolated emitter without control protocol is  $\Gamma = 2.0$ . This sets our unit of energy to be  $\Gamma/2$  and our time unit to be 2/Gamma. The numerical solution of the master equation allows us to obtain the emission spectrum for each two-level system. The emission of the ensemble of two-level systems is calculated by averaging, over all two-level systems with the appropriate Gaussian weights, the emission at each frequency.



Figure 2. Emission spectrum for an ensemble of two-level systems with detunings in a Gaussian distribution  $N(0, \Delta_0 = 10)$  (left) and  $N(0, \Delta_0 = 15)$  (right). In red (dashed line) the emission without control and in blue (solid line) the emission under the control protocol with  $\Omega_x = 50$ , and  $\tau = 0.2$  after 8 pulses.

### 3. RESULTS

The results are presented in Figures 2, and 3. The left panel of Figure 2 shows the emission spectrum of an ensemble with  $\Delta_0 = 10$ , under the influence of a pulse sequence of period  $\tau = 0.2$  with  $\Omega_x = 50$  after 8 pulses. The right panel shows the emission spectrum of an ensemble with  $\Delta_0 = 15$ , under the influence of a pulse

sequence of period  $\tau = 0.2$  with  $\Omega_x = 50$  after 8 pulses. The red line corresponds to the spectrum in the absence of the control protocol while the blue line corresponds to the emission spectrum of the ensemble under the influence of the pulse sequence. Note that the amplitude of the spectrum without control has been rescaled to that of the pulse-controlled spectrum. The controlled emission spectrum features a central peak at the pulse carrier frequency  $\omega_0$  and satellite peaks at multiples of  $\pm \pi/\tau$ . Both the central peak and the satellite peaks have the width of the isolated system emission peak. This central peak is much narrower than the inhomogeneously broadened linewidth. The overall lineshape is established after 2 pulses and the amplitudes of the peaks increase with time.

Figure 3 shows the emission spectrum of an ensemble with  $\Delta_0 = 10$ , under the influence of a pulse sequence of period  $\tau = 0.4$  with  $\Omega_x = 50$  after 8 pulses. As the interpulse delay increases, the amount of spectral weight in the central peak depends on the Rabbi frequency of the driving field and less spectral weight is refocused to the pulse carrier frequency for slower pulses.



Figure 3. Emission spectrum for an ensemble of two-level systems with detunings in a Gaussian distribution  $N(0, \Delta_0 = 10)$ . In red (dashed line) the emission without control and in blue (solid line) the emission under the control protocol with  $\Omega_x = 50$  and  $\tau = 0.4$  after 8 pulses.

### 4. CONCLUSION

We have calculated the emission spectrum of an ensemble of two-level systems, with respective emission frequencies spread across a random Gaussian distribution, when the ensemble is placed under a periodic sequence of finite-width optical  $\pi$  pulses. We show that the broad Gaussian lineshape in the absence of the control protocol is refocused by the control protocol onto a lineshape with a central peak at the pulse carrier frequency. The central peak of this refocused spectrum has the linewidth of an individual isolated non-noisy emitter and is flanked by satellite peaks at frequencies determined by the inter-pulse delay. These procedures can facilitate optimal interfaces with inhomogeneously broadened ensembles of quantum emitters.

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