Perturbing an electromagnetically induced transparency in a Λ system using a low-frequency driving field. II. Four-level system

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The effect a perturbing field has on an electromagnetically induced transparency within a three-level Λ system is presented. The perturbing field is applied resonant between one of the lower levels of the Λ system and a fourth level. The electromagnetically induced transparency feature is split and this is measured experimentally for both single and bichromatic driving fields. In the single-driving-field case a density matrix treatment is shown to be in reasonable agreement with experiment and in both single and bichromatic cases the structure in the spectrum can be explained using a dressed-state analysis.

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I. INTRODUCTION

In this work we are interested in how an electromagnetically induced transparency (EIT) is affected by the application of a perturbing electromagnetic field and specifically how an EIT associated with a three-level Λ system is affected by a field resonant with one or both of the lower levels. Where both the lower levels are involved the perturbing field will be resonant with the transition between the two lower levels and, hence, will be within the three-level system. This is the case presented in the preceding paper [1]. In the current paper we are interested in the case where the perturbing field is applied resonant with a transition between only one of the lower levels of the Λ system and an additional level. Thus, the study is that of a four-level system. Experimental observations associated with both the threeand four-level cases have been presented in an earlier paper but no theoretical modeling was presented [2]. The main motivation is to present a theoretical account of the observed effects using parameters determined from separate experiments.

The reason for the interest in electromagnetically induced transparency and modification of the transparency is associated with potential application of the phenomenon. Examples of applications have been given in the preceding paper [1] and with the ability to modify a transparency it has been suggested that there are opportunities for useful control over the EIT effect. For further discussion of this aspect of electromagnetically induced transparency the reader is referred to the preceding paper [1] and references therein.

The system we consider is a three-level Λ system where there are two lower hyperfine levels $|1\rangle$ and $|2\rangle$ and one upper level $|3\rangle$ (Fig. 1). The coupling field is applied near resonance with the $|2\rangle$ - $|3\rangle$ transition and a conventional EIT is observed when probing transition $|1\rangle$ - $|3\rangle$ [3]. In this paper the EIT is perturbed by applying a driving field between one of the ground-state levels, $|1\rangle$ or $|2\rangle$, and a fourth level $|4\rangle$. The two cases give different results but are similar for the modest perturbing fields that can be accessed experimentally and they will, therefore, only be presented for one case. The perturbing field is chosen to be resonant with the $|1\rangle$ - $|4\rangle$ transition. The measurements involve spin levels in a color center in diamond and details of the unperturbed EIT have been given in previous papers [1,4,5]. The spin system is the same as that used for earlier reports of EIT [4] and perturbed EIT [2] although the specific spin levels used are different. The perturbation gives rise to a splitting of the EIT feature and it is shown that a plausible account of the experimental traces can be obtained using a density matrix model of the fourlevel system. The splitting can be explained in terms of the dressed states of the driven system and this dressed state can also be used to explain additional measurements involving a bichromatic perturbing field.

There has been only one other experimental measurement of an equivalent situation to our knowledge, and this is where Chen *et al.* [6] have shown that a microwave field applied to a hyperfine transition splits an EIT associated with a cold Rb atom system. For other level configurations there are other studies of an EIT split by an additional electromagnetic field. These include our earlier work [5] and similar studies of Rb vapor [7]. There are other papers dealing with the perturbation of EIT [8] and treatments of driven fourlevel systems [9] but these latter publications do not include experimental observations.

II. EXPERIMENT

Experimental details have been given in the previous paper [1] and only a summary is given here to indicate the changes for this work. The experiments involve spin (S=1, I=1) transitions within the ground state of the nitrogenvacancy center in diamond and the relevant energy levels are given in Fig. 1(a). A coupling field is applied resonant with transition D and an EIT is observed when scanning through transition F (the spectrum is shown in Fig. 2 in Ref. [1]). In the present work the perturbing field is applied resonant with the hyperfine transition Y. The strength of the applied field can be determined by observed splitting of associated levels using the Mollow [10] and Autler-Townes [11] effects. For example, applying a field resonant with the Y transition causes a splitting of the X transition as a consequence of the Autler-Townes effect. This splitting gives a reliable measure



FIG. 1. (a) Energy levels of the ground state of the nitrogenvacancy color center in diamond used in the experiments. The ground state is an orbital singlet with S=1 and I=1 and a magnetic field is used to bring the spin levels $|M_s=-1\rangle$ and $|M_s=0\rangle$ close to an avoided crossing. In this situation the three upper hyperfine levels $|M_s = -1, M_I = \pm 1, 0\rangle$ are typically 40 MHz higher in energy than the lower hyperfine levels ($|M_s=0, M_I=\pm 1, 0\rangle$ for a magnetic field of 1000 G. The relative energies of the states are given and the arrows indicate the transitions involved in the study. (b) Energy levels, fields, and decay rates used in experiments and theoretical treatment. The four levels are the same as those given in (a) and designated by the simpler notation $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$. The experiments involve the application of three fields simultaneously. The coupling field and probe are applied in each case and they have Rabi frequencies of Ω_{coup} and Ω_{probe} , respectively. The perturbing field is applied resonant with the Y transition and has a Rabi frequency of Ω_{hvpY} .

of the Rabi frequency Ω_{hypY} of the field applied to the *Y* transition. Similar observations can be made to obtain the strength of the coupling field and probe field (using scaling of the field strength as necessary). The dephasing and population relaxation can also be obtained by separate spin-echo and spin-recovery measurements and the relevant parameters determined in this way are given in Fig. 1(b). Inhomogeneous broadening of the electron spin transitions (inhomogeneous 1 MHz compared with 60 kHz homogeneous) and allowance is made for this in the calculations. The inhomogeneous broadening of the hyperfine transition is small (inhomogeneous 3 kHz compared with 1 kHz homogeneous) and the transition is taken to be homogeneous with

a width of 3 kHz. Calculations show that this approximation is satisfactory.

III. FOUR-LEVEL CASE: DENSITY MATRIX CALCULATION

The situation studied involves the three levels $|1\rangle$, $|2\rangle$, and $|3\rangle$. The coupling field is resonant with the $|2\rangle - |3\rangle$ transition and the $|1\rangle$ - $|3\rangle$ transition is probed. The perturbing field is applied resonant with a transition to a fourth level $|4\rangle$. There are two ways such a perturbing field can be applied. The perturbing field can share a level with the probe field by being resonant with the $|1\rangle$ - $|4\rangle$ transition or it can share a level with the coupling field by being resonant with the $|2\rangle$ - $|4\rangle$ transition. These two cases give different results but the difference is only obvious at high perturbing fields and these intensities cannot be accessed in the present experiments. A contrast of the two cases has been presented in a separate theoretical paper where the details of the solution for the density matrix equations are given for both cases [12]. It is sufficient in the present work to restrict ourselves to considering one case with a perturbing field driving the $|1\rangle$ - $|4\rangle$ transition.

As discussed in the preceding paper for weak laser excitation the majority of the population remains in levels $|1\rangle$, $|2\rangle$, and $|4\rangle$. A driving field also involves $|3\rangle$ but the system can then be treated as a closed four-level system. The density matrix equation of motion under the rotating-wave approximation can be written as follows:

$$\begin{aligned} \rho_{11} &= i\chi_{hypY}(\rho_{41} - \rho_{14}) + i\chi_{probe}(\rho_{31} - \rho_{13}) + \gamma_{31}\rho_{33} \\ &+ \gamma(\rho_{44} - \rho_{11}) + \gamma(\rho_{22} - \rho_{11}), \\ \dot{\rho_{44}} &= -i\chi_{hypY}(\rho_{41} - \rho_{14}) + \gamma_{34}\rho_{33} - \gamma(\rho_{44} - \rho_{11}) \\ &+ \gamma(\rho_{22} - \rho_{44}), \end{aligned}$$
$$= i\chi_{coup}(\rho_{32} - \rho_{23}) + \gamma_{32}\rho_{33} - \gamma(\rho_{22} - \rho_{44}) - \gamma(\rho_{22} - \rho_{11}), \end{aligned}$$

$$\begin{split} \rho_{33} &= -i\chi_{probe}(\rho_{31} - \rho_{13}) + i\chi_{coup}(\rho_{32} - \rho_{23}) \\ &- (\gamma_{31} + \gamma_{32} + \gamma_{34})\rho_{33}, \\ \rho_{41} &= d_{41}\rho_{41} - i\chi_{hypY}(\rho_{44} - \rho_{11}) - i\chi_{probe}\rho_{43}, \\ \rho_{21} &= d_{21}\rho_{21} + i\chi_{coup}\rho_{31} - \chi_{hypY}\rho_{24} - i\chi_{probe}\rho_{23}, \\ \rho_{24} &= d_{24}\rho_{24} + i\chi_{coup}\rho_{34} - i\chi_{hypY}\rho_{21}, \\ \rho_{31} &= d_{31}\rho_{31} - i\chi_{probe}(\rho_{33} - \rho_{11}) - i\chi_{coup}\rho_{21} - i\chi_{hypY}\rho_{34}, \\ \rho_{34} &= d_{34}\rho_{34} + i\chi_{probe}\rho_{14} + i\chi_{coup}\rho_{21} - i\chi_{hypY}\rho_{31}, \\ \vdots \end{split}$$

 $\rho_{32} = d_{32}\rho_{21} + i\chi_{probe}\rho_{12} - i\chi_{coup}(\rho_{33} - \rho_{22}), \qquad (1)$ where $d_{ij} = i\delta_{ij} - \Gamma_{ij}$ are complex detunings and the detunings

 δ_{ii} are given by

 ho_{22}

$$\delta_{41} = \omega_{hypY} - \omega_{41},$$

$$\delta_{31} = \omega_{probe} - \omega_{31},$$

$$\delta_{32} = \omega_{coup} - \omega_{32},$$

$$\delta_{21} = \delta_{31} - \delta_{41},$$

$$\delta_{34} = \delta_{31} - \delta_{41},$$

(2)

$$\delta_{24} = \delta_{21} - \delta_{22} - \delta_{41}, \tag{3}$$

Note that the Rabi frequencies are $\Omega_{probe} = 2\chi_{probe}$, $\Omega_{coup} = 2\chi_{coup}$, and $\Omega_{hypX} = 2\chi_{hypX}$ for the probe, coupling, and hyperfine perturbing fields, respectively.

To calculate the absorption (and dispersion) of the field with frequency ω_p probing in the region of the ω_{31} transition we derive a steady-state solution for ρ_{31} . Numerical solutions are obtained accurate to the first order of the Rabi frequency $2\chi_{probe}$ and details are given in a separate publication [12]. The absorption profile for the probe field is given by plotting Im($\rho_{31}^{(1)}$) as a function of probe detuning δ_{31} and is calculated for various cases relating to the experimental situation.

We first consider a homogeneously broadened situation with a nominal width of 1 MHz. For this purpose the dephasing rate of the transitions associated with the upper level is taken as 1 MHz ($\Gamma_{31}=\Gamma_{32}=\Gamma_{34}=1$ MHz). Other relaxation rates are retained at those for the experimental system as given in Fig. 1(b). The application of a coupling field gives rise to a standard EIT feature and this is illustrated in the lowest trace in Fig. 2(a) for a coupling field with a Rabi frequency of 60 kHz.

Introducing a perturbing field resonant with the hyperfine transition $|1\rangle$ - $|4\rangle$ gives a symmetric splitting of the EIT and the splitting is equal to the Rabi frequency of the hyperfine field, Ω_{hypY} . As the strength of the perturbing field is increased the EIT splitting is increased and this is illustrated in Fig. 2(a). For this geometry of fields and a sufficiently strong coupling field each of the split EIT features is 50% deep. Should the hyperfine field be detuned the splitting pattern becomes asymmetric, with one feature moving toward zero frequency and gaining strength and the other feature moving to higher frequencies and becoming weaker [upper two traces in Fig. 2(a)]. If the strength of the coupling field is increased it has the effect of increasing the width of the transparencies in a similar fashion to that of the parent EIT. Such broadening for various coupling field strengths is illustrated in Fig. 2(b).

The above calculation is for a homogeneously broadened system. To model the EIT and the splittings associated with an inhomogenously broadened system the above calculation is repeated using the experimentally determined homogeneous linewidth of 60 kHz and the weighted contributions added for the transition frequencies over the inhomogeneous linewidth. The results of such a calculation are illustrated using a coupling field with a Rabi frequency of 60 kHz and a hyperfine field with a Rabi frequencies is shown in the lower half of Fig. 3. Summing the contributions gives the



FIG. 2. Calculation of a homogeneous four-level system giving the EIT for a Λ system when perturbed by a field resonant with a transition between one of the lower levels and a fourth level. The decay parameters used are those given in Fig. 1 except the dephasing rates associated with the electron spin transitions are set at 1 MHz. (a) gives the absorption response of a weak probe for a fixed coupling field of Ω_{coup} =60 kHz and various strengths of the perturbing field as indicated. For the five lower traces the perturbing field is resonant whereas for the highest Rabi frequency of Ω_{hypY} = 400 kHz the upper two traces show the spectrum for a detuning of Δ_{hypY} =±200 kHz. (b) gives the response for a fixed perturbing field with a Rabi frequency of Ω_{hypY} =200 kHz and various strengths of coupling field.

absorption for an inhomogeneous system as indicated in the second to top trace in Fig. 3. The spectrum has similar characteristics to that of a homogeneously broadened system, as can be seen by comparing the inhomogeneously broadened spectrum with the upper trace, which corresponds to a 1 MHz homogeneously broadened system. The effect of including the inhomogeneous broadening is to give a marginal increase in the linewidths and a reduction in the depths of the EIT features.





FIG. 3. Calculation of EIT associated with an inhomogeneous system perturbed by a hyperfine driving field. The lower traces show the spectra for individual homogeneous lines with various resonance frequencies detuned by Δ_{coup} as indicated. The coupling field is then detuned with respect to the individual resonances as indicated. In all cases the perturbing field is on resonance with the $|1\rangle$ - $|4\rangle$ transition and has a Rabi frequency of Ω_{hypY} =210 kHz. The contributions are summed to give the inhomogeneous spectrum above. The top trace is included for comparison and gives the equivalent spectrum for a homogeneously broadened system.

IV. EXPERIMENT AND COMPARISON WITH DENSITY MATRIX CALCULATION

An experimental measurement of the EIT in the absence of any perturbing field is shown in the lower trace in Fig. 4(a). The coupling field has a Rabi frequency of 60 kHz, a value established from separate nutation measurements. The experimental parameters are, hence, known and a calculation of the EIT spectrum is shown in Fig. 4(b). When a perturbing field driving the $|1\rangle$ - $|4\rangle$ transition is introduced, the extra field can be seen to split the EIT into two components separated by the Rabi frequency of the perturbing field [see lower trace of Fig. 4(a)]. The magnitude of the two EIT features is reduced by close to a factor of 2. The changes are in good correspondence with that calculated [see lower trace of Fig.

FIG. 4. (a) Experimental measurement of EIT perturbed by hyperfine driving field. The coupling field drives the *F* transition with a Rabi frequency of Ω_{coup} =60 kHz and this gives the EIT in the probed transition *D*. In the upper trace a resonant driving field is applied to the *Y* transition with a Rabi frequency of Ω_{hypY} =210 kHz. (b) Calculation of the above situation.

4(b)]. Accepting that a simplified set of parameters are used and these are determined from independent measurements the overall agreement between experiment and calculation is satisfactory. Although not shown, the agreement is maintained when the strength and detuning of coupling field and/or perturbing field is varied.

V. DRESSED-STATE ANALYSIS

An analysis in terms of the dressed states [13] of the driven hyperfine transition is straightforward. The energy levels for the current situation are given in Fig. 5(a), including an indication of the dressed states for the field driving the $|1\rangle$ - $|4\rangle$ transition. It can be considered that the perturbing field splits the lower level of the probed transition to give rise to two probed transitions. The coupling field applied to the $|2\rangle$ - $|3\rangle$ transition will give 100% deep EIT in each of these probed transitions and as they overlap and have equal



FIG. 5. Energy levels and fields of four-level scheme (i). The associated dressed states are shown in (ii) and the doubly dressed states in (iii).

strength the effect is to have two 50% deep transparencies in the overall absorption. The separation of the two transparencies is due to the separation of the dressed-state levels and equal to the Rabi frequency of the perturbing field, Ω_{hvpY} .

VI. BICHROMATIC FIELD

In this section we consider the effect of a bichromatic perturbing field driving a transition to a fourth level. For brevity we restrict the consideration to special cases. The single perturbing field has been shown to split the EIT into two transparency features and this situation is illustrated again in trace (ii) in Fig. 6(a) for a perturbing field resonant with the hyperfine transition Y using a Rabi frequency of Ω_{hypY} =210 kHz. A second field with a Rabi frequency of $\Omega_{hypY}^{(2)}$ = 100 kHz is introduced detuned from the Y transition by 210 kHz. With this detuning the second perturbing field is resonant with a dressed-state transition and gives a further splitting (by half the Rabi frequency). The probe transition exhibits this splitting in the EIT structure. The second field has a Rabi frequency of 100 kHz and the two EIT features are each split by 50 kHz [trace (iii), Fig. 6(a)]. It can be deduced from the dressed states that the features in (iii) will be located at $\pm \Omega_{hypY}/2 \pm \Omega_{hypY}^{(2)}/4$.

Should an alternate detuning be utilized the splitting pattern will be different. This is illustrated in a second special case. The second perturbing field is detuned by half of the Rabi frequency of the first field, or 105 kHz. The second field is resonant for a two-photon transition between the onephoton dressed states and gives another simple splitting pattern. Each of the EIT features is split, although the split is smaller than previously. There is also a weak doublet with the same splitting positioned at the frequency of the unperturbed EIT feature. This pattern is shown in trace (iv) in Fig. 6(a).

VII. COMPARISON OF EIT WITH DIRECT PROBE OF DRIVEN HYPERFINE TRANSITION

The conventional approach of studying a driven two-level system is through a one-photon absorption using a weak probe field monitoring the transition from one of the driven levels to a third level and this what is shown in Fig. 6(b). The



FIG. 6. (a) Experimental measurements of (i) EIT in a Λ system, (ii) EIT when perturbed by driving a transition to a fourth level as in Fig. 4, and (iii), (iv) EIT spectrum when perturbed by a bichromatic field. The coupling field is the same for all traces and has a Rabi frequency of Ω_{coup} =60 kHz. For traces (ii)–(iv) a resonant field is applied to the Y transition with a Rabi frequency of Ω_{hypY} =210 kHz. For traces (iii) and (iv) a second field with a Rabi frequency of Ω_{hypY} =100 kHz is also applied to the Y transition. For (iii) the field is detuned by 210 kHz and for (iv) by 105 kHz. (b) The absorption associated with the hyperfine transition Y when probed directly. The perturbing fields are the same as in (a).

spectrum is frequently termed the Autler-Townes spectrum [11] and a study of the Autler-Townes spectrum for bichromatic fields as shown in Fig. 6(b) has been presented in an earlier publication [14]. It is very clear that there is a close correlation between the EIT traces (i)–(iv) in Fig. 6(a) with the equivalent traces (i)–(iv) in Fig. 6(b). This correlation demonstrates that the two-photon resonance condition provides an Autler-Townes-type probe of the driven system. In the preceding paper [1] treating EIT within a three-level Λ system there was an analogous correlation of the perturbed EIT spectrum and a Mollow spectrum. In that case the correlation was largely restricted to the energies of the feature. However, here the correlation is with energy, linewidth, and intensity. In the Mollow configuration the driving field quenches the absorption and it is this that results in the lack of intensity correlation. This does not occur in the Autler-Townes case as the absorption is not quenched when probing to a fourth level. It is, therefore, acceptable that there is a closer relationship of the strength of features probed using a one- or a two-photon probe. However, given that the transitions represent multiphoton events the agreement is remarkable and it would be interesting to investigate to what degree the correlation between the one- and two-photon probes can be verified theoretically.

VIII. CONCLUSION

The work in this paper presents an experimental investigation of an EIT perturbed by single and bichromatic driving fields. The experiments are compared with density matrix calculations and the agreement is good considering all the parameters are determined from separate measurements. It has also been shown that dressed-state analysis is useful in

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predicting the number and position of the EIT features. These are satisfactory for a single perturbing field but also enable predictions for more complex perturbing fields where full density calculations are laborious.

This is the third paper dealing with perturbing an EIT in a Λ system. The papers [1,5] have dealt with different situations and included perturbing fields resonant with various transitions both homogeneous and inhomogeneous. Clearly not all possible Λ cases have been covered but it is considered that the experimental measurements and the accompanying modeling will enable the behavior of EIT in other situations to be anticipated. These studies should, therefore, be of value for planning further fundamental studies and assessing use of perturbation techniques for EIT applications.

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