Hyperfine spectroscopy of highly-excited atomic states based on atomic coherence

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Abstract

The hyperfine structures of highly-excited atomic states can be determined by electromagnetically-induced transparency experiment based on atomic coherence. We report an experiment on the excited states $5D_{5/2}$ in $^{85}$Rb atoms in a vapor cell. A theoretical model is presented and is in good agreement with the experimental results.

1. Introduction

Recently, lasing without inversion (LWI) [1-4], electromagnetically-induced transparency (EIT) [5-7], and enhancement of dispersion with reduced absorption [8-10] in atomic systems have attracted many interests. The physical mechanism behind these effects is the atomic coherence, i.e., a well-defined phase relation between atomic states. In a three-level $\Lambda$-type system with the lower levels being a near-degenerate state pair (the transitions between the upper state and the two lower states are dipole allowed), coherent superposition states between the two lower states (atomic coherence) can be established when a coherent coupling field is tuned to be on resonance with the atomic transition between the upper state and one of the lower states and a weak probe field is tuned to the other transition [6]. In a three-level cascade atomic system, as another example, atomic coherence can be established between the ground state and the highly-excited state by applying a coherent coupling field to the upper transition and a weak probe field to the lower transition [7]. As a direct result of the atomic coherence, EIT for the weak probe field is achieved, as demonstrated by recent experiments in different atomic systems [5-7]. Applications of this atomic coherence effect in enhancing the efficiency of nonlinear optical processes were also demonstrated [11].

In real atomic systems, the atomic states related to the coupling field usually have many hyperfine levels which may contribute to the atomic coherence. When the FWHM linewidth of the coupling field or the power-broadened natural linewidth of the associated atomic transition is broader than the separations between the hyperfine components, one cannot resolve these components simply due to the fact that the hyperfine structures are completely concealed by these broad linewidths. The interesting case is that, when the linewidths of the coupling field and the power broadening are much smaller than the hyperfine separations, the hyperfine components will contribute to the atomic coherence separately. In such a case, one will be able to resolve the closely spaced hyperfine components as the probe frequency is scanned through the whole range of the hyperfine structures and the absorption

Fig. 1. Cascade atomic system (\(^{85}\text{Rb}\)). (i) Energy levels with hyperfine structures. \(A\) and \(A'\) are atomic detunings; \(f_2\) and \(f_3\) are atomic natural decay rates; (ii) dressed-state picture.

spectrum of the probe beam is recorded. Motivated by this basic idea, we proposed a new spectroscopic method with high resolution and used it to measure the hyperfine structures of highly-excited atomic states [12]. Compared with conventional Doppler-free high-resolution spectroscopic methods, such as two-photon absorption [13], this method has several advantages. For example, the coupling field can be quite weak (much weaker than required for two-photon absorption method). The hyperfine structures of the highly-excited states are determined through the frequency scanning of the probe field not the frequency scanning of the pumping field. These properties make this method quite different from other spectroscopic methods of measuring the hyperfine structures of highly-excited atomic states [14] and more effective.

In this communication, we report the hyperfine-structure measurement of the states \(5D_{5/2}\) of rubidium atoms (\(^{85}\text{Rb}\)) in the EIT experiment in a vapor cell at room temperature. A theoretical model with all these hyperfine levels is presented, which includes finite laser linewidths for both probe and coupling fields and the light-induced linewidth broadening. Quantitative comparisons between the theoretical calculations and experimental results are given.

2. Hyperfine spectroscopy in EIT experiment

The atomic system used in this experiment is a cascade system in rubidium (\(^{85}\text{Rb}\)) atoms, as shown in Fig. 1(i), with all the relevant hyperfine components. There are two associated transitions: the lower transition is between \(5S_{1/2}, F = 3\) (the ground state, \(|1\rangle\)) and \(5P_{3/2}, F' = 4\) (state \(|2\rangle\)) with a transition wavelength of 780.0 nm (\(D_2\) line); the upper transitions are between \(5P_{3/2}, F' = 4\) and the hyperfine levels of \(5D_{5/2}\) (state \(|3\rangle\)) with transition wavelengths around 775.8 nm. The state \(|2\rangle\) serves as a common state for the two transitions in this cascade system. The hyperfine level separation between \(F' = 3\) and \(F' = 4\) in \(5P_{3/2}\) is about 120 MHz and is irrelevant in this experiment, since it is much larger than the hyperfine separations in state \(5D_{5/2}\) (less than 10 MHz). A peculiar feature of this system is that these two transitions have close frequencies, which leads to a quasi-Doppler-free condition when the two laser beams with frequencies \(\omega_c\) (coupling field) and \(\omega_p\) (probe field) propagate in the opposite directions. This configuration enables us to lower the requirement for the coupling field intensity to establish an effective atomic coherence [7]. The Rabi frequency of the coupling field now only needs to exceed the square-root of the FWHMs (about 2.45 MHz) of the two transitions (\(|2\rangle \rightarrow |1\rangle\) and \(|3\rangle \rightarrow |1\rangle\)) for substantial absorption reduction, as will be discussed later, not the much broader Doppler-broadened linewidth (about a few hundreds MHz) as required in the early experiments [5]. Meanwhile, the lower power for the coupling field also makes the light-induced linewidth broadening for the upper transition far below the hyperfine separations in the state \(5D_{5/2}\) and makes it possible to resolve the related hyperfine structures in the experiment. As a result, we can use the output of a cw low power laser diode as the coupling field for this system, as demonstrated in our previous experiment [7].

EIT effect in this atomic system can be clearly explained in the dressed-state picture. When the coupling (or, pumping) field is applied to the transition between the state \(|2\rangle\) and state \(|3\rangle\), a pair of equivalent dressed-states \(|2d\rangle\) and \(|2d'\rangle\) (separated by twice of the Rabi frequency of the coupling field when on resonance) is produced, which has strong correlation, as shown in Fig. 1(ii). The atoms on the ground state \(|1\rangle\) will have two possible transition paths (\(|1\rangle \rightarrow |2d\rangle\) and \(|1\rangle \rightarrow |2d'\rangle\) as denoted by the dashed up-arrows) to get excited by absorbing photons from the probe field. A destructive interference between these two transition amplitudes leads to an absorption reduction (EIT) for the probe field [1].
The experimental scheme is depicted in Fig. 2. Both the probe and coupling lasers are single-mode frequency-stabilized tunable laser diodes. The linewidths of these cw laser diodes are measured by using the homodyne beating technique with: $\gamma_p \approx \gamma_c \approx 2\text{ MHz}$, $\gamma_p$ ($\gamma_c$) is the FWHM linewidth of the probe (coupling) light. These two laser beams are arranged to have orthogonal polarizations and to propagate in opposite directions to satisfy the quasi-Doppler-free condition in this cascade system. The natural transition linewidths of the state $5P_{3/2}$ and the highly-exited state $5D_{5/2}$ are about 6.0 MHz and 0.97 MHz, respectively. The Doppler FWHM linewidth at room temperature (21°C in this experiment) is about 520 MHz for the D_2 line. L_1 and L_2 are two 10 cm positive lenses for focusing both the coupling and probe beams onto the center of a 7.6 cm long rubidium vapor cell to give better spatial matching. An effective Rabi frequency of 5.0 MHz is obtained when an average power intensity for the focused Gaussian coupling beam is estimated. Two 0.5 mm diameter apertures A_P1 and A_P2 are used for better laser beam profiles. The probe beam is attenuated to be around 1 $\mu$W which leads to a power intensity far below the saturation intensity for the rubidium D_2 line (9.7 mW/cm^2). The absorption of the probe beam is measured by a photodiode.

When the coupling laser is blocked, the normal absorption spectrum for Doppler-broadened two-level atoms is recorded as the frequency of the probe beam is scanned through the D_2 line of $^{85}$Rb. With the coupling laser on and tuned to be on resonance with the upper transition, the absorption for the probe beam is modified by the induced atomic coherence. The result is shown in Fig. 3 with the heavy solid curve. As expected, the hyperfine structures of the highly-excited state appear on the absorption spectrum of the probe beam. This is actually a two-photon process, since the probe field is not directly related to the upper-excited state $5D_{5/2}$. In this case, all the three hyperfine components are observed ($F' = 4 \rightarrow F'' = 3, 4, 5$ are dipole-allowed). The measured separations between the hyperfine levels for $^{85}$Rb atoms are 9.6 ± 0.2 MHz ($F'' = 4, 5$) and 9.1 ± 0.2 MHz ($F'' = 3, 4$), which are in good agreement with that in literature by Doppler-free two-photon spectroscopy \cite{15}. In order to achieve a sufficiently large EIT effect, the Rabi frequency of the coupling field needs to be large enough. A stronger coupling field, however, is always accompanied by a larger light-induced linewidth broadening. If the power-broadened linewidth is larger than the hyperfine separations of the peculiar atomic state, the hyperfine components will be masked by this broadened linewidth, as mentioned earlier. The restrictions for the Rabi frequency of the coupling field (denoted by $\Omega_c$) to resolve the hyperfine structures in this EIT experiment are given by
where $\delta_{\text{min}}$ is the minimum separation of the hyperfine structures in the state $|3\rangle$. $\gamma_{m1} (m = 2, 3)$ represents the transverse relaxation rate for the transition from the excited level $|m\rangle$ to the ground state level $|1\rangle$, which also corresponds to the HWHM (one half of FWHM) of the corresponding transition line. In the above experiment, the Rabi frequency of the coupling field is about 5.0 MHz, which is sufficiently larger than $2\sqrt{\gamma_{21}\gamma_{31}} \approx 2.45$ MHz (with $\gamma_{21} \approx 3$ MHz and $\gamma_{31} \approx 0.49$ MHz). Consequently, an effective atomic coherence for the probe field is established. Meanwhile, the light-induced broadened FWHM for the two-photon transition $|3\rangle \rightarrow |1\rangle$ is about $4.6\gamma_{31} \approx 2.3$ MHz, which is much smaller than the related hyperfine separations in the state $5D_{5/2}$. So, the requirement given by Eq. (1) is satisfied in this experiment.

3. Theory

In this section, we present a theoretical analysis for the above cascade atomic system. In our previous work, a theoretical calculation about an EIT experiment in an ideal three-level cascade system has been given with the inhomogeneous Doppler-broadening and the linewidths of both coupling and probe fields included, which is in good agreement with the experimental results [7]. In the previous EIT experiments, only the achievable absorption reduction was emphasized and, therefore, a strong coupling field was used in both the experimental and the theoretical treatments. In the existence of a significant light-induced linewidth broadening, the hyperfine structures have very small effect on the atomic coherence and, therefore, can be ignored. In the current experimental conditions, however, these hyperfine levels are really important and critical. The light-induced broadened linewidth (HWHM) $\gamma_{31}^{\text{R}}$ of the two-photon transition for a Rabi frequency $\Omega_c$ can be expressed as [16]

$$\gamma_{31}^{\text{R}} \approx \gamma_{31} \sqrt{1 + \frac{\Omega_c^2}{4\gamma_{21}\gamma_{31}}}.$$ (2)

Here, we have assumed that the natural lifetime of the ground state $5S_{1/2}$ is infinite. The Rabi frequency used in the early EIT experiment was about 100 MHz [7], which could give a quite large power-broadened linewidth (FWHM): $2\gamma_{31}^{\text{R}} \approx 40.8$ MHz. This means that $2\gamma_{31}^{\text{R}}$ is much larger than the hyperfine separations of the state $5D_{5/2}$. As a result, the condition (1) was violated and no hyperfine structures of that state were observed in experiments. In other aspect, although the frequencies of the two transitions in this three-level cascade system are quite close (nearly Doppler free), there exists a residual two-photon Doppler broadening. The condition for neglecting this residual broadening is given as [7]

$$\Omega_c \ll \gamma_{31} \sqrt{\frac{\omega_p}{\omega_c - \omega_p}}.$$ (3)

This requires the Rabi frequency $\Omega_c \ll 6.8$ MHz for $\gamma_{31} \approx 0.49$ MHz. On the other hand, in order to get sufficiently large atomic coherence and to achieve measurable EIT effect, the Rabi frequency of the coupling field needs to be large enough. As a result, the condition given by Eq. (3) usually can not be satisfied very well in real experimental situations. For our experimental conditions, both the hyperfine structures of the atomic states and the residual two-photon Doppler broadening should be considered to get a general theoretical analysis for this inhomogeneously broadened system. However, it is very difficult to include both effects in the theory. We believe that, provided the Rabi frequency of the coupling field is smaller than the value of 6.8 MHz given by inequality (3), one can still use the quasi-Doppler free approximation by neglecting the residual two-photon Doppler broadening. As will be shown later, the theoretical results with such an approximation are still in agreement with the experimental data.

The absorption coefficient for a weak probe field tuned near resonance with the lower transition can be written as

$$\alpha(\Delta) = \frac{-4N\hbar\omega_p n_0 g_{21}}{\epsilon_0 E_p} \text{Im}(\rho_{21}(\Delta)),$$ (4)

where $N$ is the density of atoms; $n_0$ is the background index of refraction; $2\hbar g_{21}$ is the dipole-moment matrix element of a single atom for the transition between state $|1\rangle$ and state $|2\rangle$; $E_p$ is the complex amplitude of the electric field of the probe field (here, we take it to be real). Following the derivation procedure in Ref. [7], the steady-state dipole-moment of a single atom
for the lower transition modified by the coupling field applied to the upper transition, with all the relevant hyperfine components of the upper state \( |3\rangle \) \((F'' = 3, 4, 5, \) for \( F' = 4 \) of the state \( |2\rangle \)) being considered, can be obtained as follows

\[
\rho_{21}(\Delta) \approx -ig_{21}E_p \times \left[ \gamma_{21} - i\Delta + \frac{Q^2 A(\Delta)}{4(\mu_{c2} + \mu_{b2} + \mu_{a2})} \right]^{-1},
\]

where

\[
A(\Delta) \equiv \frac{\mu_{c2}}{\gamma_{c1} - i(\Delta - \Delta')} + \frac{\mu_{b2}}{\gamma_{b1} - i(\Delta - \Delta' - \delta_1)} + \frac{\mu_{a2}}{\gamma_{a1} - i(\Delta - \Delta' - \delta_1 - \delta_2)}.
\]

\( \Delta \equiv \omega_p - \omega_{21}, \Delta' \equiv \omega_{c2} - \omega_c, \delta_1 \) and \( \delta_2 \) are the separations of the three hyperfine components of the level \( |3\rangle \) \((F'' = 3, 4, 5, \) as shown in Fig. 1. \( \gamma_{n1} \approx \gamma_{c1} \equiv \gamma_3 \) with \( \gamma_{n1} \) \( (n = a, b, c) \) being the decay rate for each of the three hyperfine sublevels of the upper state \( |3\rangle \) to the ground state. \( \mu_{a2}, \mu_{b2}, \) and \( \mu_{c2} \) are coupling parameters related to the transition probabilities (or line strengths) of the three transitions from the state \( |2\rangle \) \((F' = 4) \) to the three hyperfine sublevels \( |3\rangle \) \((F'' = 3, 4, 5, \) denoted by the subscripts \( (a, b, c) \). These relative strengths can be determined using the standard Racah method in atomic spectroscopy [17], which gives: \( \mu_{a2} : \mu_{b2} : \mu_{c2} \approx 0.25 : 0.55 : 1 \) for \(^85\)Rb atoms. Here, we have used the approximations that \( \rho_{22} \approx \rho_{33} \approx 0 \) and \( \rho_{11} \approx 1 \). These conditions are satisfied in our experimental situation, i.e., the probe field is so weak (far below the saturation intensity of the D2 line) that nearly all the atoms stay at the ground state \( |1\rangle \) at the lowest order in probe intensity. From Eq. (5), one can readily find that the Rabi frequency of the coupling field should exceed 2\( \sqrt{\gamma_{21}\gamma_3} \) to achieve an effective atomic coherence between the highly-excited state \( |3\rangle \) and the ground state \( |1\rangle \), when all the frequency detunings are set to be zeros, which is the left part of the inequality (1). With the inhomogeneous Doppler-broadening included, the absorption coefficient \( \alpha(\Delta, v) \) for a single atom with a velocity \( v \) along the probe light direction can be derived from Eq. (5). The total absorption coefficient can be obtained with an integration over the whole thermal velocity distribution (which is assumed to be a Maxwellian) as

\[
\alpha(\Delta) \approx \frac{8\sqrt{\pi}\ln 2 h_2^2 N_0 \omega_p n_0 \omega_D}{c e_0 n_0} \times \text{Re}\left[ \exp\left(\Theta(\Delta)^2\right) \text{erfc}(\Theta(\Delta)) \right],
\]

with

\[
\Theta(\Delta) \equiv \frac{2\sqrt{2}}{\Delta \omega_D} \left[ \gamma_{21} - i\Delta + \frac{Q^2 A(\Delta)}{4(\mu_{c2} + \mu_{b2} + \mu_{a2})} \right],
\]

where \( N_0 \) is the density of atoms inside the Rb cell. \( A(\Delta) \) in the argument \( \Theta(\Delta) \) of the complementary error function is defined by Eq. (6). Also, the finite linewidths of the probe and coupling laser fields can be taken into account simply through the transformations [7]: \( \gamma_{21} \rightarrow \gamma_{21}^p + \gamma_c/2 \equiv \gamma_2 \) and \( \gamma_{31} \rightarrow \gamma_{31}^p + \gamma_c/2 + \gamma_c/2 \equiv \gamma_3 \), if the lineshapes of both laser fields are assumed to be Lorentzians with \( \gamma_p \) and \( \gamma_c \) as their FWHMs, respectively. The light-induced broadened linewidths \( \gamma_{21}^p \) and \( \gamma_{31}^p \) for the two transitions are also included in these transformations. The theoretical result is shown in Fig. 3 with the light solid curve, together with the experimental data. The parameters are: \( \Omega_c = 5.0 \text{ MHz}, \gamma_p = 2.0 \text{ MHz}, \gamma_c = 2.0 \text{ MHz}, \delta_1 = 9.1 \text{ MHz}, \delta_2 = 9.6 \text{ MHz}, \) and \( \Delta \omega_D = 520 \text{ MHz} \) is the FWHM Doppler width. It is evident that the above theoretical analysis is in good agreement with our experimental measurement, although there exists a small mismatch. This is mainly because that we neglected the residual longitudinal two-photon Doppler-broadening in the theoretical treatment. Also, the Rabi frequency of the coupling field used in the theoretical calculations has an uncertainty due to the fact that we can not accurately determine its intensity distribution inside the atomic cell and an average value was used. Besides, the spatial matching between the probe and coupling beams inside the vapor cell is not perfect in the experiment.

Since any kind of broadenings will strongly affect the spectroscopic resolution, it is necessary to find the influences of the linewidths of the two laser fields on this spectroscopic method. It is clear from the above discussions that the resolution will primarily depend on \( \gamma_p, \gamma_c, 2\gamma_{21}^p, \) and \( 2\gamma_{31}^p \). The fact is that if any one of these FWHM linewidths exceeds the hyperfine separations, the hyperfine structures will be hidden. For this cascade system, although there is no direct relation between the probe field and the highly excited state \( |3\rangle \), the linewidth of the probe field does affect the res-
solution through the two-photon process. This is also shown by the above transformation for $\gamma_{31}$. In Fig. 4, we plot the absorption spectra of $^{85}$Rb atoms with different linewidths for the probe and coupling fields, while other parameters are kept the same as that in Fig. 3. We find that as the laser linewidths take moderate values of $\gamma_p = \gamma_c = 6$ MHz, the hyperfine components can no longer be resolved because in this case, $2\gamma_3 \approx 14.3$ MHz $> \delta_{\text{min}}$ ($\approx 9.1$ MHz). On the other hand, since the resolution is related to the two-photon transition linewidth $\gamma_{31}$, which is modified by the coupling field to be $\gamma_{31}^B$, the Rabi frequency (proportional to the amplitude of the coupling field) also greatly affects the resolution. As a result, we conclude that the resolution of this spectroscopic method depends mainly on the light-induced broadened linewidth of the upper transition (the Rabi frequency of the coupling field) and the linewidths of the two laser fields.

4. Conclusions

In this experiment, a spectroscopic method to measure highly-excited hyperfine splittings based on atomic coherence effect is demonstrated in rubidium atoms. A basic theoretical calculation is also presented and is in good agreement with our measurement. Due to the arrangement of a quasi-Doppler-free configuration for our cascade system, this EIT experiment can be carried out with low power coupling field. Consequently, an effective atomic coherence can be generated, and, at the same time, only a small light-induced linewidth broadening is introduced. In this experiment, when an effective EIT effect is observed, the light-induced broadened linewidth for the upper transition is still much smaller than the relevant hyperfine separation (approximately, only a natural linewidth wider than the natural linewidth). This enables us to determine the hyperfine structures of the highly-excited state. Since this spectroscopic method is based on atomic coherence, the resolution strongly depends on the natural decay rates of the related atomic states and the linewidths of both the probe and coupling fields. Meanwhile, the light-induced linewidth broadenings of the related transitions are also important factors to affect the resolution, even when the Rabi frequency is rather small.

It should be noted that there is actually, in principle, no requirement for the minimum intensity of the coupling field to establish atomic coherence. As far as the intensity of the coupling field is non-zero, an atomic coherence will be produced although it will be rather small. When the coupling field is very weak, the EIT effect will be very small and the related hyperfine structures of the highly-excited atomic states may be obscured by a strong background (the Doppler-broadened D2 line). We let the coupling field to be relatively strong in the above experiment ($\Omega_c > 2\sqrt{\gamma_{21}\gamma_{31}}$ in Eq. (5) to establish a relatively strong effective atomic coherence). So, the normal method of measuring EIT can be used to carry out this spectroscopic measurement. Actually, this is not a limitation for this spectroscopic method. When the coupling field is relatively weak (i.e., $\Omega_c < 2\sqrt{\gamma_{21}\gamma_{31}}$), we can use more sensitive detection methods (for example, the lock-in detection technique) to measure the weak EIT effect and to extract the related hyperfine structures from the Doppler-broadened background. Besides, the light-induced linewidth broadenings will become rather small in this case and the resolution will be increased. Meanwhile, the signal-to-noise ratio can be greatly enhanced because the Doppler-broadened background will be rejected in this kind of
measurement. Therefore, the technique demonstrated in this experiment could be an effective spectroscopic method with high resolution.

For this special cascade atomic system, this method is proven to be simple and has high resolution since the two cascade transitions have very close frequencies. For many cases, this is not true. Still this experiment shows that one can use the atomic coherence to be an useful way for atomic spectroscopy. In other aspect, this method is particularly suitable for atomic beam or trapped atomic systems where nearly all the longitudinal Doppler broadening is removed (Doppler-free) and the above limitation for this method (two transitions with close frequencies) can be removed. Other three-level atomic configurations, such as A-type and V-type systems, also can be used. Therefore, this spectroscopic method, in principal, can be used for any highly-excited transitions in different atoms with high resolution.

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