Rydberg level shift due to the electric field generated by Rydberg-atom collision-induced-ionization in cesium atomic ensemble

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We demonstrate the phenomena of Rydberg level shift due to the electric field generated by Rydberg-atom collision-induced-ionization in cesium atomic ensemble. For $50S_{1/2}$ Rydberg state of cesium atoms, we measured the energy shift of hundreds MHz via electromagnetically-induced-transparency spectroscopy. We used a semi-classical model to explain the physical origin and found good agreement with numerical simulation. These energy shifts are important in self-calibrating measurements of microwave frequency standard. Moreover, in contrast to resonant excitation case, the no-broadening spectroscopy with high signal-to-noise ratio would be useful in high precision measurements.

Keywords: Rydberg atoms; ionization of Rydberg atoms; Rydberg electromagnetically-induced transparency; energy shift; cesium ensemble;
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I. INTRODUCTION

Recent progress in technical control offers a possibility for studying the interaction between pure quantum mechanical system and their coupling to reservoirs, which can be extend as a unique tool for quantum sensing [1]. The alkali metal atoms are convenient as regards the technical requirements needed for sensors. Neutral atoms in chip-scale atomic vapor have been utilized as a device for many fields, including atomic clocks [2], atomic magnetometers [3] and radio-frequency (RF) field [4]. Atom-based electromagnetic field sensor technologies hold great promise for realizing capabilities. The most advanced experimental demonstrations include high dynamic field ranges exceeding 120 dB, high-intensity radio-frequency field up to 10 kV/m [4, 5], broadband RF detection of THz [6], and high sensitivity of $\mu$Vcm$^{-1}$Hz$^{-1/2}$ [4, 7]. Recent advances have experimentally achieved a sensitivity of 55 nV cm$^{-1}$/Hz$^{1/2}$ which approaches the quantum projection noise limited [8], while going beyond is possible by using squeezed state of light or Schroedinger-cat states [9].

Recent advances have afforded new capabilities in RF or microwave (MW) sensing. The wavelength of typical high frequency microwave has a wavelength range of millimeter or centimeter. In the chip-scale atomic vapor, the atomic signal is limited by atomic density. Be different to the characters of atomic spin, the Rydberg atoms with high principal quantum number have exaggerated electric characters including dipole-dipole or Van der Waals interactions. Using buffer-gas- filled cell to improve the atomic coherence is not feasible, where collision

will destroy the electric dipole coherence. The high density can be obtained by ultraviolet light desorption effect or heating the atomic cell. Under high density atomic gas, the sensing accuracy and sensitivity are limited by the spectrum broadening, including Doppler broadening, collision broadening and interaction dephasing induced by Rydberg atoms collisions. The dephasing observed in the experiments has been explained by averaging effect resulting from Van den Waals interaction of Rydberg-Rydberg interaction or coupling in Rydberg atoms and glass inner surface. In this paper, we measure Rydberg state level shift using Rydberg electromagnetically-induced-transparency (EIT). The equivalent electric field is caused by Rydberg atoms collisions induced ionization in cesium atomic ensemble.

II. SETUP AND METHODS

The experimental setup (Fig. 1) consists of an external-cavity diode laser (ECDL) with a wavelength of 852 nm and used as a probe laser with a typical linewidth of ~90 kHz. The optical power of the 1018-nm ECDL laser was amplified to ~5 W by the fiber amplifier, and the output beam was frequency-doubled with a single-pass PPLN crystal to produce a 509-nm laser. The two beams were overlapped in cesium atomic vapor with a counter-propagating configuration. The spherical cesium cell has radius of ~1.2 centimeters to match Rayleigh length of the focused beams (~40μm waist radius). The 852-nm probe laser was stabilized to Cs $6S_{1/2}(F = 4) \rightarrow 6P_{3/2}(F' = 5)$ hyperfine transitions via saturated absorption spectroscopy (SAS). Wavelength of the probe laser and coupling laser were calibrated by a wavelength meter (HighFinesse WS-7, wavelength deviation sensitivity is about 10 MHz). The wavelength meter was calibrated through cesium atoms transition line. The
ladder-type Rydberg EIT signal is observed by scanning the frequency of 509-nm coupling laser while locking the 852 nm probe laser. The background-free EIT spectrum with its high signal-to-noise ratios is beneficial for the low-noise detected system, which decreases the intensity noise and phase noise.

FIG. 1. (a) Schematic diagram of the experimental setup. An external-cavity diode laser (ECDL) with a wavelength of 852 nm was used as probe laser. The laser power of a 1018-nm ECDL was amplified to ∼ 5 W by fiber amplifier, and frequency was doubled with a periodically poled lithium niobate (PPLN) crystal to produce a λc=509 nm laser. Then, the 852-nm and 509-nm laser beams were overlapped in cesium atomic vapor with a counter-propagating configuration. SAS setup is used as the frequency reference of hyperfine structure Cs D2 transition. (b) Energy level schematic of the cascade-type EIT of Cs atom with Rydberg state. The 852-nm probe laser resonates with Cs 6S1/2(F = 4) → 6P3/2(F′ = 5) → 50S1/2 cascade transition. The EIT signal were obtained under different two-photon resonance conditions. We fixed 852-nm laser frequency and scanned the 509-nm laser frequency to get the EIT signal at each fixed probe laser detuning. The EIT signal of maximum intensity is located at the position of the probe laser frequency detuning of about 260 MHz. We consider that Rydberg level shift due to the electric field generated by Rydberg-atom collision-induced-ionization in cesium atomic ensemble, so the EIT signal of maximum intensity appears at a position of 260 MHz, detailed explanation can be found in the fourth part. The linewidth of EIT signal is about 10 MHz, which is measured by a radio-frequency modulation spectroscopy. Fig. 2(b) shows the intensity of 50S1/2 Rydberg state EIT spectra for different cell temperatures. In order to clearly show the overall profile of the signal at different temperatures, there is no corresponding EIT peak at each point in the picture. The EIT signal of maximum intensity varies with the cell temperature parameters. For the case of temperature of 100°C, the probe laser detuning of maximum EIT signals intensity is about 440 MHz. Under the condition of high temperature, the Rydberg EIT signal for 6S1/2(F = 4) → 6P3/2(F′ = 5) → 50S1/2 cascade transitions will be very weak. Moreover, the observed linewidths of maximum EIT signals have a few

III. MEASUREMENT OF ENERGY SHIFT

Fig. 2 (a) shows the dependence of Rydberg EIT signal on probe detuning at a cell temperature of 40°C, each small peak in the figure represents an EIT signal. The zero detuning of probe laser shows Cs 6S1/2(F = 4) → 6P3/2(F′ = 5) → 50S1/2 cascade transition. The EIT signal were obtained under different two-photon resonance conditions. We fixed 852-nm laser frequency and scanned the 509-nm laser frequency to get the EIT signal at each fixed probe laser detuning. The EIT signal of maximum intensity is located at the position of the probe laser frequency detuning of about 260 MHz. We consider that Rydberg level shift due to the electric field generated by Rydberg-atom collision-induced-ionization in cesium atomic ensemble, so the EIT signal of maximum intensity appears at a position of 260 MHz, detailed explanation can be found in the fourth part. The linewidth of EIT signal is about 10 MHz, which is measured by a radio-frequency modulation spectroscopy. Fig. 2(b) shows the intensity of 50S1/2 Rydberg state EIT spectra for different cell temperatures. In order to clearly show the overall profile of the signal at different temperatures, there is no corresponding EIT peak at each point in the picture. The EIT signal of maximum intensity varies with the cell temperature parameters. For the case of temperature of 100°C, the probe laser detuning of maximum EIT signals intensity is about 440 MHz. Under the condition of high temperature, the Rydberg EIT signal for 6S1/2(F = 4) → 6P3/2(F′ = 5) → 50S1/2 cascade transitions will be very weak. Moreover, the observed linewidths of maximum EIT signals have a few
times larger than the resonance linewidth for different cell temperatures. There is no obvious linewidth broadening means that the electric field induced shift is not a dissipative process. For the above experimental phenomena, we first considered the interaction between Rydberg atoms. The theoretical results were shown in Fig. 3(a), where interaction strength is smaller than a few tens of MHz, and it is impossible to cause the energy level shift of several hundred MHz, therefore, energy level shift caused by the interaction between atoms is excluded.

![Figure 3](image)

**FIG. 3.** (a) Rydberg-Rydberg interaction energies of Cs Rydberg atoms for 50S₁/₂ and 50D Rydberg states. (b) Stark map for Cs 50S₁/₂ and 48D Rydberg states with electric fields. The red dots indicates the Stark shift of Cs 50S₁/₂ (m_j = ±1/2) Rydberg state, the blue solid line indicate Stark shift of Cs 48D₃/₂ (m_j = ±1/2, m_s = ±3/2) Rydberg state, and the green solid line indicate Stark shift of Cs 48D₅/₂ (m_j = ±1/2, m_s = ±3/2, m_s = ±5/2) Rydberg state.

### IV. ANALYSIS AND DISCUSSIONS

The most advanced experiments demonstrate that collisions in Rydberg atomic ensemble will produce positive ions and electrons. We expand this idea and use a semi-classical model to explain the physical origin of Rydberg level shift. In a thermal Rydberg atomic ensemble, the collisions between Rydberg atoms and between Rydberg atoms and ground-state atoms will induce charged particles (positive ions and electrons), which mediates the avalanche ionization [10-12]. For Cs 50S₁/₂ Rydberg state, the theoretical results show that Rydberg level shift crossover induced by Stark effect is close to 20 GHz, which corresponds to the electric field intensity of 20V/cm, as shown in Fig.3(b). Under the condition of weak electric field, Rydberg level shift will be dominated by Stark shift. The dependence of Rydberg level shift on weak electric field can be measured by Rydberg EIT spectroscopy. For the case of temperature of 100°C, the electric field caused by Rydberg atom collision is 5V/cm. There is no crossover between energy levels under this condition, therefore, hypothesis of self-ionization of energy levels is verified. In the theoretical model, we assume the electron density and Rydberg ion density to be approximately the same, and both parameters are given by N_i. The dependence of electron density and Rydberg atomic density can be obtained by solving the following rate equations,

\[ \dot{N}_{\text{Ryd}} = -N_{\text{Ryd}} N_g \sigma_g \pi - N_{\text{Ryd}} N_{\text{geo}} \sigma_\text{geo} \pi + R_{\text{pump}} \]  

\[ \dot{N}_i = N_{\text{Ryd}} N_g \sigma_g \pi + N_{\text{Ryd}} N_{\text{Ryd}} \sigma_\text{geo} \pi - N_i \Gamma_d \]

N_{\text{Ryd}} and N_{\text{ion}} are Rydberg atom density and induced ions density, respectively. \( \sigma_g \) is cross-section for Rydberg-ground state collisions, \( \sigma_\text{geo} \) describes the geometric cross-section of the Rydberg state, and \( \pi \) is the mean relative velocity of Rydberg atom collides with ground-state atom. \( \pi \) is the mean relative velocity of the collisions between Rydberg atoms. The value of \( \sigma_g \) is 0.06\( \sigma_{\text{geo}} \) [13], with \( \sigma_\text{geo} = \pi (a^* N^4) \) being the geometric cross-section of the Rydberg state with effective principal quantum number \( n^* \). The \( R_{\text{pump}} = \Omega^2_{\pi}/(\Omega^2_{\pi} + \Omega^2_{\text{geo}}) \cdot N_g \sigma_g \pi \rho_{\text{ion}} \) gives the losses in the charged particle number. \( \Gamma_d \) is ions diffuse rate, the most obvious contribution is that \( \Gamma_i \) describes the motional decay parameters of electrons leaving the interaction volume due to thermal motion, which vary slightly with temperature, \( \Gamma_i \) is \( \sim 0.25 \) MHz. The second term in \( \Gamma_d \) is represented by \( 2 \Gamma_r \) and describes recombination of ions with electrons into neutral particles. We refer to related literature [14], the expected value for \( \sigma_r \) can be estimated in semi-classical treatment with Kramers formula, which gives a value of \( 1 \times 10^{-5} \sigma_{\text{geo}} \). Here, \( m_e \) is the electron mass, \( \rho_{\text{ion}} \) is the ion population, we take \( \rho_{\text{ion}} \approx 1 \) in our model.

The electric field is \( \varepsilon = 2.603 \cdot |e|/(4\pi\varepsilon_0) \cdot N_{\text{ion}}^{2/3} \), and the corresponding energy shift is \( \Delta = - (1/2) a \varepsilon^2 \). The dependence of Rydberg level shift on the ion density can be given as following.

\[ \Delta = - \frac{1}{2} \alpha \left( \frac{2.603 \cdot |e|}{4\pi\varepsilon_0} \right) \cdot N_{\text{ion}}^{2/3} \]  

The blue detuned Rydberg level shift versus \( N_{\text{ion}} \) density and \( N_g \) density for Cs 50S₁/₂ Rydberg state are shown in Fig. 4. \( \Omega_p/2\pi = 5.0 \) MHz, \( \Omega_e/2\pi = 36.0 \) MHz. Theoretical simulation takes the absolute value of formula (3). For the red detuning for Rydberg state 50S₁/₂, there is also energy shift and maximum intensity of Rydberg EIT signal. For the case of Cs 6P₃/₂(F’ = 5) → 50S₁/₂ Rydberg transition, the red detuning of 500 MHz is corresponding to the blue detuning of Cs 6P₃/₂(F’ = 3) → 50S₁/₂ Rydberg transition. We conclude that the observed phenomenon is arising from the velocity-selective hyperfine splitting of intermediate states. For Rydberg EIT spectroscopy, it is difficult to eliminate the influence of hyperfine energy states. In general, the EIT signal of maximum intensity is located at the position where the probe laser frequency detuning of about 260 MHz in Fig.2 (a), which is due to the energy level shift 260MHz caused by the equivalent electric field.
FIG. 4. The blue detuned Rydberg level shift versus $N_{\text{ion}}$ density and $N_g$ density for Cs 50S$_{1/2}$ Rydberg state. The solid circles with error bar are Rydberg level shift measured date, and the solid line is the theoretical simulation.

Moreover, we measure the dependence of maximum intensity of Rydberg EIT signal on pumping laser power. As shown in Fig. 5(a), the intensity of EIT signal increase with the coupling laser power, but the peak shift is at almost of order MHz. In fact, the shift is possible due to the excitation probability increasing of Rydberg state that caused by optical pumping; The increasing of coupling beams Rabi frequency will suppress the factor of Rydberg in superposition dark state $|D\rangle = \alpha|g\rangle + \beta|r\rangle$. The $\beta$ describe the proportion of the Rydberg component in the dark state, which is defined by a factor $\Omega_p/\sqrt{(\Omega_p^2 + \Omega_c^2)}$; The Rydberg interaction will mitigate the EIT signal. All these effects result in transmission decreasing at resonance position and poor shift of the maximum EIT signal.

Under the condition of strong probe beam, the Rydberg blockade effect is much stronger than optical pumping in the condition of high atomic density [15-17]. Thus, the Rydberg interaction inhibits the ensemble occupation of Rydberg state and thus effectively suppress avalanche ionization. As shown in Fig. 5(b), we measured the dependence of probe laser detuning that can obtain the maximum EIT signal on probe Rabi frequency. From the picture, increasing the probe beams Rabi frequency cleanly decreases probe laser detuning, that is, decreases the energy shift, the nonlinearity shift compression is also observed. The transmission depends on the power of the probe laser is due to the excitation suppression of higher probability population of Rydberg state [18]. The three-level EIT system has atoms in a superposition where almost only one atom can be excited to the Rydberg state and thereby contributes to the EIT dark state, while the other atoms in the blockade volume acts as two-level system which absorb and scatter the probe laser. Under condition of hot atomic ensemble, the blockade effect is not strong enough to enable an efficient cooperative absorption or suppression of EIT signal. Even so, the excitation blockade effect still can be observed.

FIG. 5. (a). Rydberg EIT signal’s intensity versus the probe detuning at different Rabi frequency of the 509-nm coupling beams. The 852-nm probe beam’s Rabi frequency is $\sim 5.60$ MHz. Temperature of Cs vapor cell is $\sim 25^\circ$C. (b) The position of maximum intensity EIT signal varies with the Rabi frequency of probe laser. The 509-nm coupling beam’s Rabi frequency is $\sim 34.50$ MHz. Temperature of Cs vapor cell is $\sim 25^\circ$C.

V. CONCLUSION

In conclusion, we explored the non-broadening Rydberg level shift by Rydberg EIT spectroscopy combined with RF bandwidth modulation. We use a semi-classical model to explain the physical origin and find good agreement with numerical simulation. The ion density caused by Rydberg atoms collisions is modulated by controlling the ground state atomic density. Moreover, the nonlinearity shift compression is also observed, which arises from the cooperative blockaded interaction in Rydberg ensemble. Our results have direct consequences in Rydberg quantum sensing. This energy level shift due to the electric field generated by Rydberg atoms collisions induced ionization in cesium atomic ensemble may be important in quantum sensing device for microwave frequency standard.

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