Measurement of the argon-gas-induced broadening and line shifting of the barium Rydberg level 6s24d $^1D_2$ by two-photon resonant nondegenerate four-wave mixing*

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We apply two-photon resonant nondegenerate four-wave mixing with a resonant intermediate state to the observation of the broadening and shifting of the barium Rydberg level 6s24d $^1D_2$ by collision with argon. The collision broadening and shifting cross sections are measured. This technique is purely optical, and can investigate the pressure dependence of the transverse relaxation rate Γ_{21} between the Rydberg state and an intermediate state, as well as the transverse relaxation rate Γ_{20} between the Rydberg state and the ground state.

Keywords: four-wave mixing, collision-induced broadening, Rydberg state

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Collision-induced broadening and shifting of atomic transitions by various buffer gas perturbers have been extensively studied both experimentally and theoretically for a number of years. The research findings in this respect have provided valuable ideas for numerous areas including gaseous electronics, atomic physics, and astrophysics.[1−3] Especially, the research of broadening and shifting of absorption lines involving alkali-earth-metal atoms and alkali-metal atoms in Rydberg states have been of interest because the theoretical descriptions of collisions can be simplified by the large separations between Rydberg electrons and ionic cores.[4−7] In recent years, due to the unprecedented progress in laser cooling and trapping, the collision-induced broadening of Rydberg atomic spectral lines perturbed by buffer gas have attracted renewed interest because Rydberg atoms, having a large dipole moment and long lifetime, can interact with each other coherently over a relatively long time, which makes them a potential candidate for quantum information processing and trapping.[8] It is difficult to directly detect the fluorescence from Rydberg states because the long radiative lifetime implies a corresponding decrease of the fluorescent intensity. Modern experimental techniques employ lasers to populate specific Rydberg states and then detect the electrons or ions after field-ionization, photoionization or collisional-ionization.

Nonlinear spectroscopy based on the wave mixing is a valid tool to acquire dynamic information concerning the structure of matter because the nonlinear susceptibility shows resonance at the natural frequency of the medium. Lately, various resonance-enhanced nondegenerate wave-mixing techniques have been developed by our group. For instance, a new type of two-photon resonant nondegenerate four-wave mixing (NFWM) in a dressed atomic system has been demonstrated, which provides a spectroscopic technique for measuring not only the dephasing rate and resonant frequency but also the transition dipole moment between two highly excited atomic states.[9,10] We have reported on a generalize scheme for high-resolution resonance-enhanced $2n$-wave mixing in $(n+1)$-level systems with phase-conjugate geometry.[11] Due to the n-photon resonant excitation involved in this technique, we can study highly excited states with high sensitivity by purely optical means. Moreover, we have employed two-photon resonant nondegenerate

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four-wave mixing (NFWM) with a resonant intermediate state for the measurement of the Rydberg series of Ba atoms.[12]

In this paper we two-photon resonant NFWM to investigate the broadening and shifting of the barium Rydberg state 6s24d^{1}D$_2$ perturbed by argon gas. We can study Rydberg states with high sensitivity by purely optical means, which is unlike the collision ionization exploitation, photo ionization or field ionization used to detect Rydberg atoms in most experiments. Instead of detecting electrons or ions, here the signal to be detected is a coherent light. The experiment is performed in barium vapour. The technique can achieve Doppler-free resolution of narrow spectral structures of Rydberg levels when the incident lasers have narrow bandwidths.[9]

We consider two-photon resonant NFWM in a $|0\rangle$–$|1\rangle$–$|2\rangle$ cascade three-level system (Figs. 1(a) and 1(b)), where states between $|0\rangle$ and $|1\rangle$ and between $|1\rangle$ and $|2\rangle$ are coupled by dipolar transitions with dipole moment matrix elements μ_1 and μ_2 and resonant frequencies Ω_1 and Ω_2, respectively. As shown in Fig. 1(a), beam 1 with frequency ω_1 almost propagates along the opposite direction of beam 2, beams 2 and 2′ have the same frequency ω_2 and a small angle exists between them. Assuming that $\omega_1 \approx \Omega_1$ and $\omega_2 \approx \Omega_2$ so that ω_1 drives the transition from $|0\rangle$ to $|1\rangle$ while ω_2 drives the transition from $|1\rangle$ to $|2\rangle$, the simultaneous interactions of atoms with beams 1 and 2 will induce atomic coherence between $|0\rangle$ and $|2\rangle$ through two-photon excitation. This coherence is probed by beam 2′, and as a result an NFWM signal of frequency ω_1 is generated in the direction almost opposite to the direction of beam 2′.

The experiment was performed in barium vapor, where the ground state 6s^2 ^{1}S$_0$ ($|0\rangle$), the intermediate state 6s6p ^{1}P$_1$ ($|1\rangle$), and the Rydberg state 6s24d ^{1}D$_2$ ($|2\rangle$) form a three-level system. The barium vapour was contained in a stainless steel heat pipe oven with argon as the foreign gas. The oven temperature was around 933.15 K. Two dye lasers L1 and L2 were pumped by the second or third harmonics of a Quanta-Ray Nd:YAG laser, and had a linewidth of 0.007 nm and pulse width of 5 ns. All the incident beams were linearly polarized in the same direction. They were focused into spots with approximately 2.0-mm diameter. Laser L1 was tuned to 553.5 nm, the wavelength of the 6s^2 ^{1}S$_0$–6s6p ^{1}P$_1$ transition. The laser output from L2 was split into beams 2 and 2′, which, after passing through a beam splitter, intersected in the oven at a small angle of 0.5° to drive the transition from 6s6p ^{1}P$_1$ to a Rydberg 6s24d ^{1}D$_2$ level. The NFWM signal had the same polarization as the incident beams, and was directed by a photodiode and then fed into a signal averager for data averaging. A computer was used for data processing and controlling the stepping motor to vary the wavelength of L2.

We kept ω_1 fixed and scanned ω_2 in the vicinity of the 6s6p ^{1}P$_1$–6s24d ^{1}D$_2$ resonance line of the Ba atom at different argon pressures. To avoid strong absorption at the resonant frequency of transition from 6s^2 ^{1}S$_0$ to 6s6p ^{1}P$_1$, here the wavelength of L1 was detuned from the exact resonance by 0.48 nm. We studied the collisional broadening of the two-photon transition. Figure 2 presents the NFWM spectra of the 6s^2 ^{1}S$_0$–6s24d ^{1}D$_2$ transition at different argon pressures. Besides the broadening, a line-centre shift for 6s^2 ^{1}S$_0$–6s24d ^{1}D$_2$ transition has been observed. Since the line shapes of the spectra are dominated by the collisional broadening at high buffer gas pressure we can neglect the Doppler effect, then the NFWM signal intensity becomes[9]

$$I_{\rm e} \propto \frac{1}{(\Gamma_{10}^2 + \Delta_1^2)^2[\Gamma_{20}^2 + (\Delta_1 + \Delta_2)^2]}, \qquad (1)$$

where $\Delta_1 = \Omega_1 - \omega_1$ and $\Delta_2 = \Omega_2 - \omega_2$ are the detunings from the resonances, Γ_{n0} is the transverse relaxation rate between states $|n\rangle$ and $|0\rangle$, Γ_{20} is the half

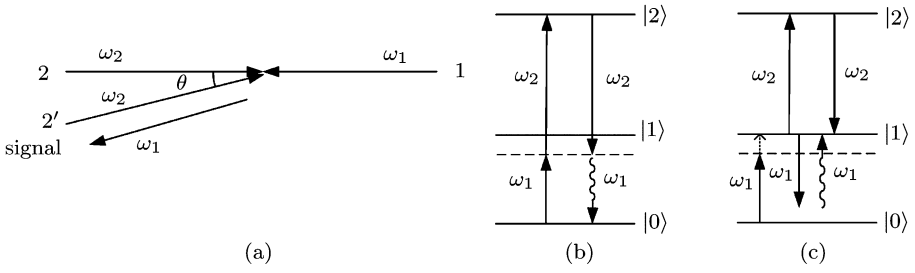


Fig. 1. (a) Schematic diagram of two-photon resonant NFWM, (b) energy-level diagram for two-photon resonant NFWM, (c) energy-level diagram for collisional redistribution.

width at half maximum (FWHM) of the line and can be expressed as $\Gamma_{20} = \gamma_2/2+\gamma_{20}^{\rm col}$. Here γ_2 and $\gamma_{20}^{\rm col}$ are the terms due to the spontaneous emission and collision broadening, respectively. The solid curves are the theoretical curves. In Fig. 2, as the pressure of Ar further increased, the violet shoulder began to swell conspicuously, indicating that a satellite line induced by the collisional redistribution was in the process of development.

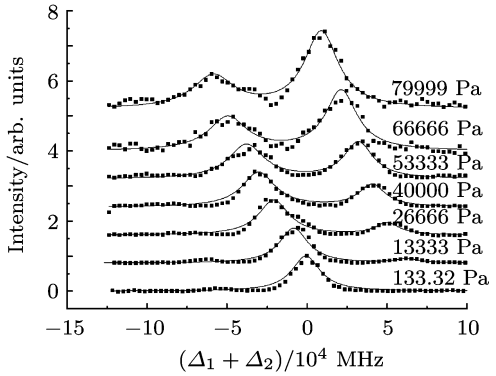


Fig. 2. NFWM spectra of the $6s^2\ {}^1S_0$–$6s24d\ {}^1D_2$ transition at different argon pressures.

Next, we study the satellite line induced by the collisional redistribution. The $6s6p\ {}^1P_1$ level was populated in the presence of collisions since the relevant wavelength of L1 was tuned near the 553.5-nm resonance line. As shown in Fig. 1(c), there is a corresponding nondegenerate four-wave mixing signal at frequency ω_1 generated from beams 2, 2′, and 1, via the perturbation chain $\rho_{11}^{(0)} \to \rho_{21}^{(1)} \to \rho_{11}^{(2)} \to \rho_{01}^{(3)}$. The NFWM signal intensity is given by

$$I_{\rm s} \propto \frac{1}{(\Delta_1^2+\Gamma_{10}^2)[\Delta_2^2+\Gamma_{21}^2]}\left|\rho_{11}\right|^2. \tag{2}$$

Here ρ_{11} is the diagonal density matrix element. As we scan Δ_2 the NFWM spectrum linewidth (FWHM) equals $2\Gamma_{21}$. When interactions between the Ba atom and pertuber in the gaseous system are due to binary collisions, ρ_{11} is proportional to the buffer gas pressure, i.e.

$$\rho_{11} \propto p. \tag{3}$$

As the buffer gas pressure increases, the intensity of the NFWM increases accordingly, and then forms the satellite line of the two-photon resonant NFWM spectrum originating from the energy level $|0\rangle$. The total signal can be represented with a coherent superposition of the signals from the two-photon resonant NFWM and the satellite NFWM. In Fig. 2 the signal intensity of the two-photon resonant NFWM signal is normalized to 1. Thus, with Eqs. (1)–(3), we can obtain the total signal intensity as

$$I = \frac{\Gamma_{20}^2}{[(\Delta_1+\Delta_2)^2+\Gamma_{20}^2]} + A(\Delta_1^2+\Gamma_{10}^2)\Gamma_{20}^2\frac{p^2}{\Delta_2^2+\Gamma_{21}^2}. \tag{4}$$

Here A is the proportionality factor, and together with the transverse relaxation rates Γ_{20} and Γ_{21} can be obtained through fitting the experimental data.

We plot the line-broadening and line-shifting rates against the pressure of the buffer gas. Figure 3(a) presents the pressure dependences of the transverse relaxation rates Γ_{20} and Γ_{21}. Figures 3(b) and 3(c) show the pressure dependences of the line-centre frequency shift $\delta\nu_{\rm e}$ of two-photon resonant NFWM spectrum and the line-centre frequency shift $\delta\nu_{\rm s}$ of the satellite line, respectively. The solid curves represent the theoretical results where Γ_{20}, Γ_{21}, $\delta\nu_{\rm e}$, and $\delta\nu_{\rm s}$ are assumed to be linear with respect to pressure p, i.e. $\Gamma_{20} = \Gamma_{20}^{(0)} + \eta_{\rm re}p$, $\Gamma_{21} = \Gamma_{21}^{(0)} + \eta_{\rm rs}p$, $\delta\nu_{\rm e} = \eta_{\rm se}p$, and $\delta\nu_{\rm s} = \eta_{\rm ss}p$. Here, $\eta_{\rm re}$ and $\eta_{\rm se}$ are the pressure-broadening and line-shifting rate coefficients of the two-photon resonant NFWM, $\eta_{\rm rs}$ and $\eta_{\rm ss}$ are the pressure-broadening and line-shifting rate coefficients of the satellite NFWM, respectively. For the data in Fig. 3, we obtain $\eta_{\rm re} = 0.0686$ MHz/Pa, $\eta_{\rm rs} = 0.0684$ MHz/Pa, $\eta_{\rm se} = -0.767$ MHz/Pa, and $\eta_{\rm ss} = -0.744$ MHz/Pa. The negative line-centre frequency shift indicates that the transition frequency shifts toward lower energies as pressure increases. That is, the collision between Ar atoms and Ba atoms during their transition shifts the energy levels to reduce the separation between the states.

For removing the temperature dependences inherent in the rate coefficients, the pressure-broadening and line-shifting rate coefficients are converted into cross sections using

$$\sigma_{\rm r} = \sqrt{\frac{\pi^3\mu k_{\rm B}T}{8}}\eta_{\rm r}$$

and

$$\sigma_{\rm s} = \sqrt{\frac{\pi^3\mu k_{\rm B}T}{2}}\eta_{\rm s},$$

respectively.[3] Here, $\sigma_{\rm r}$ is the collision-induced broadening cross section, $\sigma_{\rm s}$ is the line-shifting cross section, $k_{\rm B}$ is the Boltzmann constant, μ is the reduced mass of the barium–argon pair, and T is the temperature. We obtain $\sigma_{\rm re} = 3.47\times10^{-14}$ cm^{-2}, $\sigma_{\rm rs} = 3.46\times10^{-14}$ cm^2, $\sigma_{\rm se} = -7.76\times10^{-13}$ cm^{-2}, and $\sigma_{\rm ss} = -7.53\times10^{-13}$ cm^{-2}. Here, $\sigma_{\rm re}$ and $\sigma_{\rm se}$

are the pressure-broadening and line-shifting cross sections of the two-photon resonant NFWM, $\sigma_{\rm rs}$ and $\sigma_{\rm ss}$ are the pressure-broadening and line-shifting cross sections of the satellite line, respectively. These pressure-broadening and line-shifting results provide the means to extract a set of fundamental information about the interactions between Ba and Ar atoms.

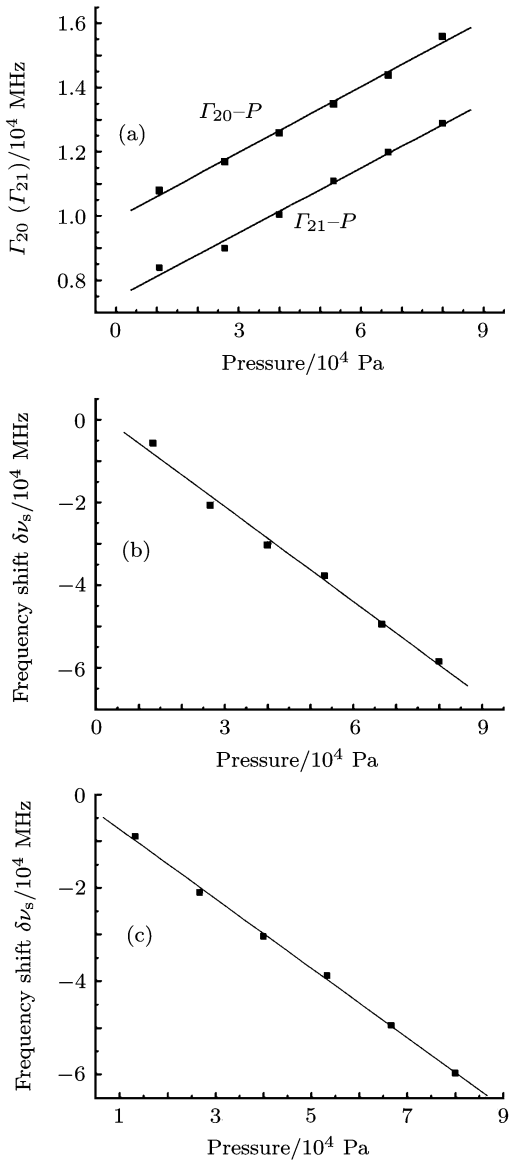


Fig. 3. Plots of Γ_{20}(a), Γ_{21} (a), $\delta\nu_{\rm e}$ (b) and $\delta\nu_{\rm r}$ (c) for the Rydberg state 6s24d $^1{\rm D}_2$ of Ba, each as a function of Ar pressure. Linear fits are superimposed on the data.

Traditionally, the experimental method of broadening and line-shifting of alkali-metal and alkali-earth-metal Rydberg levels is based on Doppler-free, two-photon spectroscopy with counterpropagating laser beams and the detection of the electrons or ions after field-ionization, photoionization or collisional-ionization.[4] Compared with these methods, our technique has the following features. First, our method is a coherence technique, where atomic coherences ρ_{20} and ρ_{21} are induced by a two-photon resonant NFWM process and by a satellite NFWM process, respectively. Due to the parametric nature of this process, the signal is highly directional coherent light, leading to a large signal for detection. Second, the phase matching of NFWM can be achieved for a very wide frequency range from many hundreds to thousands of $\rm cm^{-1}$. Most importantly, our method investigates the pressure dependences of both the transverse relaxation rates Γ_{20} and Γ_{21}, therefore, the contributions of collision to both the coherence between Rydberg state $|2\rangle$ and ground state $|0\rangle$ and the coherence between Rydberg state $|2\rangle$ and middle state $|1\rangle$ can be studied.

In conclusion, we have utilized two-photon resonant NFWM for studying the broadening and shift of the barium Rydberg state 6s24d $^1{\rm D}_2$ due to collisions with argon. The collision broadening and shift cross section have been measured. This method is purely optical, and has the advantages that the phase matching condition is not critical and the generation of the NFWM signal is efficient.

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