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Sub-natural linewidth of the probe absorption spectrum in a cold Cs atom–molecule system*

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We theoretically analysed the linewidth of the probe absorption spectrum in a cold Cs atom–molecule system. The tunnelling coupling between the two excited molecular states, especially for the Cs atom–molecule system, plays an important role in obtaining the sub-natural linewidth of the probe absorption spectrum. For example, when the tunnelling couple strength fulfils $\sigma_{12} = 10\gamma_{ab_1}$, the linewidth is only about 0.66 MHz. Moreover, since the linewidth of interest is dominated by the tunnelling coupling, the absorption peak becomes very narrow even in the case of large pump laser intensities.

Keywords: sub-natural linewidth, tunnelling coupling

PACC: 4250, 3270J, 3280

1. Introduction

The natural (even sub-natural) linewidth plays a crucial role not only in high-resolution spectroscopy and high-precision magnetometry,[1−4] slow light, and quantum information processing,[5−7] but also in laser cooling of atoms and ions, in which the natural linewidth decides the lowest attainable temperature (Doppler limit) and, when lasers are locked to atomic transitions, the natural linewidth determines the tightness of the lock.[8] Therefore, how to get a very narrow line is an interesting problem both for theorists and experimentalists. Since the discovery of the laser inducing line-narrowing effect in the couple Doppler broadened system,[9,10] a multi-level configuration with a coherent control laser field has been explored to obtain very narrow linewidths in the spontaneous emission spectrum or in the probe absorption spectrum.[11−17]

With the help of the control laser technique, by which the transitions from different ground states are driven, a sub-natural linewidth (two orders less than the Doppler broadening) has been observed in several systems, for example, room-temperature Rb vapour,[8] electromagnetically induced transparency (EIT)[18,19] and the tripod system[13] etc. When metastable 2S atomic hydrogen was trapped in a magnetic trap at a submillikelvin scale, a very narrow linewidth (3 kHz) was obtained in Ref.[20]. Usually, the narrow linewidth is limited to the natural line width of the excited levels. Therefore it is interesting to discover a new controllable parameter, compared to the strength and the detuning of laser field, in order to obtain a sub-natural linewidth. In the cold Cs atom–molecule system, there actually is an effective four-level structure and two laser fields (photoassociation (PA) laser and pump laser), which have been proposed in our recent work.[21] Based on extensive studies on the formation of cold Cs_2 molecules through PA,[22−26] it is known that there is a special double-well shape in the excited potential curve of $0_g^-(6S + 6P_{3/2})$, separated by a potential barrier at distance $R \approx 15a_0$. The PA process can be interpreted as follows: when a pair of cold Cs atoms absorbs a photon, an excited level of the outer well ($R > 15a_0$) is populated; tunnelling is suggested as an efficient mechanism for transferring the population to the inner well ($R < 15a_0$), where spontaneous emission may lead to formation of cold

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molecules in low vibrational levels of the electronic state $a^3\Sigma_u^+(6s,6s)$.[26] Therefore, four atom–molecule levels, which are composed of the atomic ground state $|a\rangle$, two molecular excited states $|b_{2,1}\rangle$ and the ground molecular state $|g\rangle$ (see Fig.1), are involved in this process. The tunnelling coupling between two excited molecular states is suggested as a new controllable parameter to obtain a sub-natural linewidth in the probe absorption spectrum.

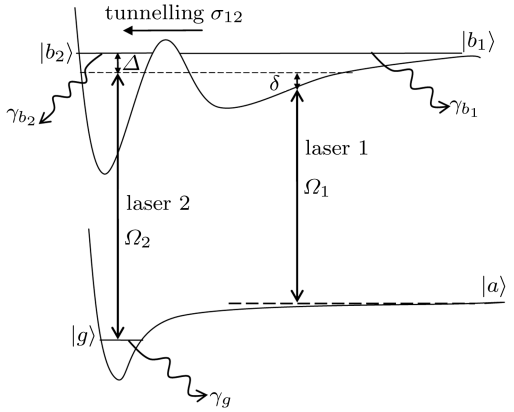


Fig.1. The level scheme. Δ and δ denote detunings. The inner and outer molecular states $|b_{1,2}\rangle$ spontaneously decay with a rate γ_{b_1,b_2} to levels outside this scheme. The molecular ground state $|g\rangle$ is contributed with a decay rate γ_g, which phenomenologically takes into account losses through inelastic collisions.

This paper is organized as follows. In Section 2, we describe our theoretical model and present the density matrix equation of motion. In Section 3, we analyse the linewidth of the probe absorption spectrum. Finally, a conclusion is given in Section 4.

2. Model and the density matrix equation

As shown in Fig.1, the free Cs atom (denoted by $|a\rangle$), the inner and outer excited molecular states of $0_g^-(6S+6P_{3/2})$ (denoted by $|b_{2,1}\rangle$) and the lower triplet state $a^3\Sigma_u^+$ ($|g\rangle$) construct an effective four-level system. The first laser field, called the probe laser in below, is a PA laser with Rabi frequency Ω_1, by which the excited molecules are formed in a precise vibrational level of the outer well in $0_g^-(6S+6P_{3/2})$ (denoted by $|b_1\rangle$) from the ultra-cold atoms (denoted by $|a\rangle$). The second laser field with Rabi frequency Ω_2, called the pump laser, provides a stimulated emission, coupling the inner excited molecular state $|b_2\rangle$ with the deeply bounded molecular state $|g\rangle$. The coupling between the outer and inner excited molecular states is realized by the tunnelling mechanism and the coupling strength is denoted by σ_{12}. From Ref.[26], we know that the tunnelling coupling strength depends on the energy difference between the two tunnelling levels lying in the inner and outer wells, which can be controlled by tuning and detuning the PA laser in the experiment. The dynamics of the system are governed by the following Hamiltonian (in the rotating wave approximation[27])

$$H = H_0 + H_I + H_t, \tag{1}$$

where

$$\begin{aligned} H_0 &= \hbar(\delta+\Delta)b_1^\dagger b_1 + \hbar(\delta+\Delta)b_2^\dagger b_2 + \hbar\delta g^\dagger g, \\ H_I &= -\frac{\hbar}{2}\left(\Omega_1 b_1^\dagger a a + \text{H.c.}\right) - \frac{\hbar}{2}\left(\Omega_2 g^\dagger b_2 + \text{H.c.}\right), \\ H_t &= \hbar\sigma_{12}\left(b_2^\dagger b_1 + b_1^\dagger b_2\right). \end{aligned} \tag{2}$$

The operators a and $a^\dagger$ are annihilation and creation operators for atoms in the state $|a\rangle$, similarly for $b_{1,2}$ ($b_{1,2}^\dagger$) and g ($g^\dagger$), annihilating (creating) molecules in states $|b_{1,2}\rangle$ and $|g\rangle$, respectively. In our model, $|b_1\rangle \longrightarrow |a\rangle$ and $|b_2\rangle \longrightarrow |g\rangle$ transitions are dipole allowed,[27] and the transition $|b_1\rangle \longrightarrow |b_2\rangle$ is realized by the tunnelling mechanism.

The atom–molecule coherence dynamics can be investigated by solving the following master equation

$$\frac{\partial}{\partial t}\hat{\rho}_{\mu\nu} = -\gamma_{\mu\nu}\hat{\rho}_{\mu\nu} - \frac{\mathrm{i}}{\hbar}\left[\hat{\rho}_{\mu\nu}, H\right], \tag{3}$$

where $\hat{\rho}_{\mu\nu}$ is the density matrix operator and describes the probability of being in the μ state for $\mu = \nu$ and the polarization for $\mu \neq \nu$. $\gamma_{\mu\nu}\,(\mu \neq \nu)$ is the transverse decay rate from state μ to ν $(\mu,\nu = a, b_1, b_2, g)$ and is defined as $\gamma_{\mu\nu} \equiv (\gamma_\mu + \gamma_\nu)/2$. In the absence of molecular decay, i.e., $\gamma_{b_1,b_2,g} = 0$, the total particle number is conserved and $\rho_{aa} + 2\left(\rho_{b_1b_1} + \rho_{b_2b_2} + \rho_{gg}\right) = 1$.

Substituting the Hamiltonian (1) into Eq.(3) and taking the mean values for the density matrix operator, $\rho_{\mu\nu} = \langle\hat{\rho}_{\mu\nu}\rangle$, we have found equations of motion for the density matrix elements ρ_{b_1a}, ρ_{b_2a} and ρ_{ga}

$$\begin{aligned} \frac{\partial}{\partial t}\rho_{b_1a} &= \left[-\mathrm{i}\left(\delta+\Delta\right) - \gamma_{ab_1}\right]\rho_{b_1a} - \mathrm{i}\sigma_{12}\rho_{b_2a} \\ &\quad -\mathrm{i}\frac{\Omega_1}{2}\left(4\rho_{b_1b_1}\rho_{aa} - \rho_{aa}^2\right), \\ \frac{\partial}{\partial t}\rho_{b_2a} &= \left[-\mathrm{i}\left(\delta+\Delta\right) - \gamma_{ab_2}\right]\rho_{b_2a} - 2\mathrm{i}\Omega_1\rho_{b_2b_1}\rho_{aa} \\ &\quad +\mathrm{i}\frac{\Omega_2}{2}\rho_{ga} - \mathrm{i}\sigma_{12}\rho_{b_1a}, \\ \frac{\partial}{\partial t}\rho_{ga} &= \left(-\mathrm{i}\delta - \gamma_{ag}\right)\rho_{ga} + \mathrm{i}\frac{\Omega_2}{2}\rho_{b_2a} \end{aligned}$$

$$-2\mathrm{i}\Omega_1\rho_{gb_1}\rho_{aa}. \tag{4}$$

Considering the weak probe field condition ($\Omega_1 = 0.01\gamma_{ab_1}$) and the long time limit, and starting with the case when all atoms are prepared in the atomic ground state $|a\rangle$, we arrive at the expression of the imaginary part of ρ_{b_1a} and follow procedures in Ref.[27], which is the experimental observable variable.

$$\begin{aligned}\mathrm{Im}(\rho_{b_1a}) = {} & \frac{\Omega_1}{a_1^2+a_2^2}\left[\Omega_2^2/4-\delta\left(\Delta+\delta\right)+\gamma_{ag}\gamma_{ab_2}\right]a_1 \\ & +\left[\delta\gamma_{ab_2}+\left(\Delta+\delta\right)\gamma_{ag}\right]a_2,\end{aligned} \tag{5}$$

where

$$\begin{aligned}a_1 = {} & \frac{1}{2}\Omega_2^2\gamma_{ab_1}-2\delta\left(\Delta+\delta\right)\left(\gamma_{ab_2}+\gamma_{ab_1}\right) \\ & +2\gamma_{ag}\left[\sigma_{12}^2-\left(\Delta+\delta\right)^2+\gamma_{ab_1}\gamma_{ab_2}\right],\end{aligned} \tag{6}$$

$$\begin{aligned}a_2 = {} & \left[\frac{1}{2}\Omega_2^2+2\gamma_{ag}\left(\gamma_{ab_2}+\gamma_{ab_1}\right)\right]\left(\Delta+\delta\right) \\ & +2\delta\left[\sigma_{12}^2-\left(\Delta+\delta\right)^2+\gamma_{ab_1}\gamma_{ab_2}\right].\end{aligned} \tag{7}$$

In the following section, we will prove that the linewidth of the absorption peak can be controlled by adjusting the tunnelling coupling strength, pump power and pump field detuning.

3. The linewidth of the probe absorption spectrum

The properties of the probe absorption spectrum are governed by Eq.(5). As shown in Ref.[21], when the tunnelling coupling strength increases, typical double dark resonance spectra emerge and it is interesting to note that a very narrow absorption spectral line is attained in the case of a large enough σ_{12} (see Fig.2(b1)). The linewidth is defined as the full width at half maximum for the absorption peak. For given system parameters Ω_1, Ω_2, Δ and all the decays of involved states, $\mathrm{Im}(\rho_{b_1a})$ is a function of δ (see Eq.(5)). Then by solving the equation $\mathrm{Im}(\rho_{b_1a}) = 50\%\mathrm{Im}(\rho_{b_1a})_{\max}$ on δ, one can find six roots. Here, we focus on the two nearest roots to δ_c, corresponding to the value of δ for the central absorption peak. The difference between these two roots (δ_1, δ_2, shown in Figs.2(a1) and 2(b1)) determines the linewidth of the central absorption peak. Figures 2(a1) and 2(b1) are the probe absorption spectra with different decay rates (γ_{ab_2}) and tunnelling coupling strengths. Note that the width of the central peak changes very little with increasing decay rate (γ_{ab_2}). Particularly, when tunnelling coupling is larger, the decay (γ_{ab_2}) has a little effect on the linewidth of the central peak (see Figs.2(b1) and 2(b2)). Hence, we will choose $\gamma_{ab_2} = 0$ to be simplified in the following calculation.

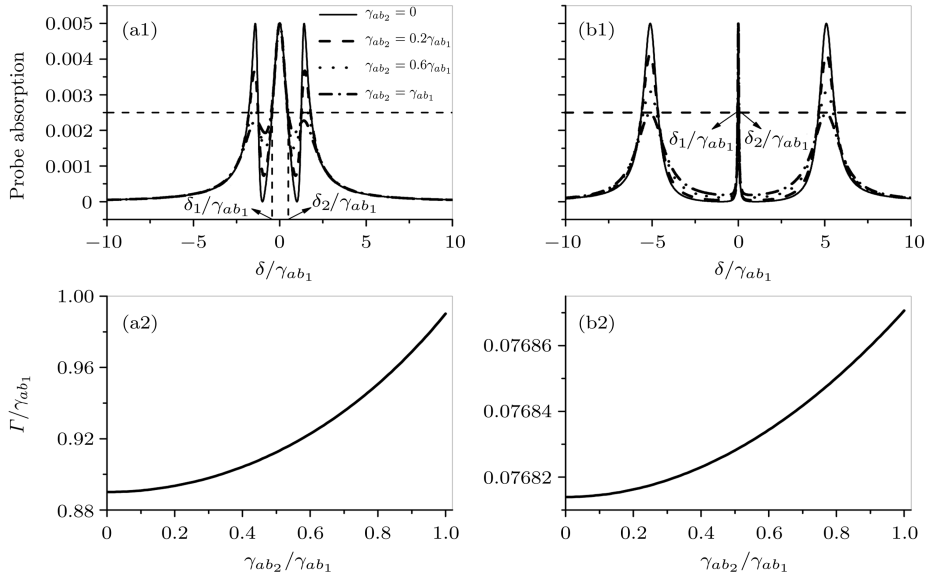


Fig.2. (a1) (b1) The probe absorption spectra for different decay and tunnelling coupling; (a2) (b2) the linewidth of the central absorption peak as a function of decay rate γ_{ab_2}, where (a1) (a2) $\sigma_{12} = \gamma_{ab_1}$; (b1) (b2) $\sigma_{12} = 5\gamma_{ab_1}$, with $\Omega_2 = 2\gamma_{ab_1}$, $\gamma_{ag} = 0$, $\Delta = 0$.

In Fig.3, we plot the linewidth of the central peak for the probe absorption spectrum versus the tunnelling coupling. As the tunnelling coupling increases, it can be found that the linewidth of the central absorption peak decreases rapidly and sub-natural linewidth ($\Gamma < \gamma_{ab_1} \approx 33.4$ MHz) is approached at $\sigma_{12} \gtrsim 0.8\gamma_{ab_1}$ (see the shaded part of Fig.3). When the tunnelling coupling fulfils $\sigma_{12} = 10\gamma_{ab_1}$, the linewidth is reduced to 0.66 MHz. As shown in Ref.[26], the characteristic tunnelling time related to the transfer of the population from the outer well to the inner well can be 400–450 ps, then a very narrow linewidth (around 12–15 kHz) may be achieved. Hence, in this cold Cs atom–molecule system, the tunnelling coupling of the two wells in excited molecular states of $0_g^-(6S + 6P_{3/2})$ plays a key role in obtaining the narrow spectral line.

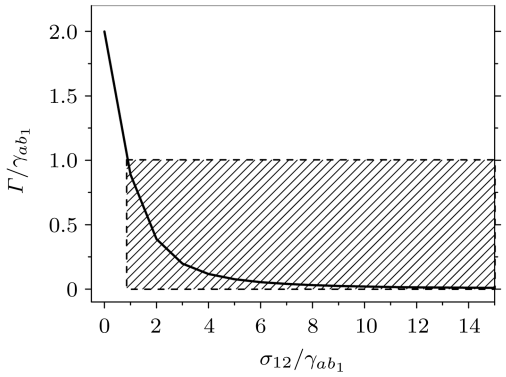


Fig.3. The linewidth of the central absorption peak as a function of tunnelling coupling; other parameter values are the same as in Fig.2.

In Fig.4, we present the linewidth as a function of the pump field Rabi frequency for different tunnelling coupling strengths. It is easy to see that a stronger pump power usually enlarges the linewidth for a given tunnelling coupling, as in Refs.[9], [10], [28] and [29], while the tunnelling coupling dramatically modifies the situation. For example, at larger Rabi frequencies of the pump field ($\Omega_2 = 8\gamma_{ab_1}$), the linewidth of the central peak can be suppressed from 62.56 MHz to 9.2 MHz with the tunnelling coupling increasing from $\sigma_{12} = \gamma_{ab_1}$ to $\sigma_{12} = 10\gamma_{ab_1}$. Therefore, in our model, even at larger pump laser intensities, sub-natural linewidth can be approached (see the shaded part of Fig.4). This is obviously different from the three-level system and other four-level systems. Considering the weak probe field condition, we remind the reader that our results will be authentic for Ω_2 larger than $0.5\gamma_{ab_1}$ (the black part is unauthentic in Fig.4).

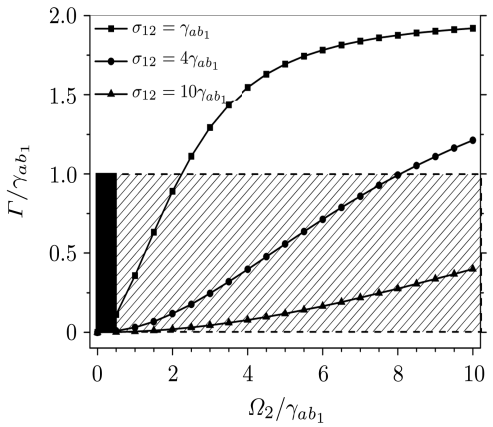


Fig.4. The linewidth of the central absorption peak as a function of the Rabi frequency of the pump laser for different tunnelling coupling; other parameter values are the same as in Fig.2.

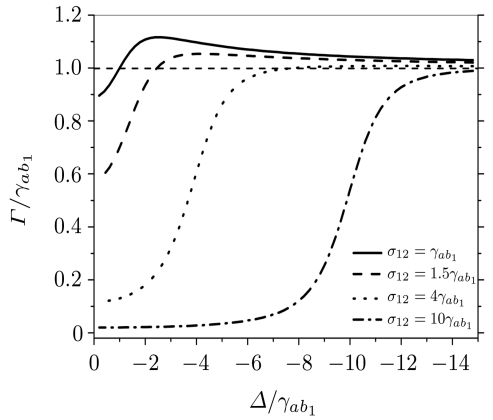


Fig.5. The linewidth of the central absorption peak as a function of pump detuning for different tunnelling coupling. Other parameter values are the same as in Fig.2.

For a given tunnelling coupling strength (σ_{12}), the linewidth of the central peak as a function of the pump field detuning is plotted in Fig.5. We can see that the linewidth is broadened with increasing pump field detuning for a given tunnelling coupling. This is different from the three-level system,[8,11,12] in which the linewidth becomes narrower with increasing strong-coupling laser detuning. Once again, the stronger the tunnelling coupling is, the narrower the linewidth of the central peak will become for a given pump field detuning.

4. Conclusions

In conclusion, we have demonstrated that it is possible to get a sub-natural linewidth for the probe absorption spectrum in a cold Cs atom–molecule system. It is interesting to see that the tunnelling coupling between two excited states dramatically modifies the conditions for obtaining the sub-natural linewidth in the central peak of the probe absorption spectrum. The nature of this controllable sub-natural linewidth system may be understood in terms of interference induced by the coherent interaction between two dark states,[14] which have been reported in our recent work.[21] The underlying physics is going to be explored in the future.

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