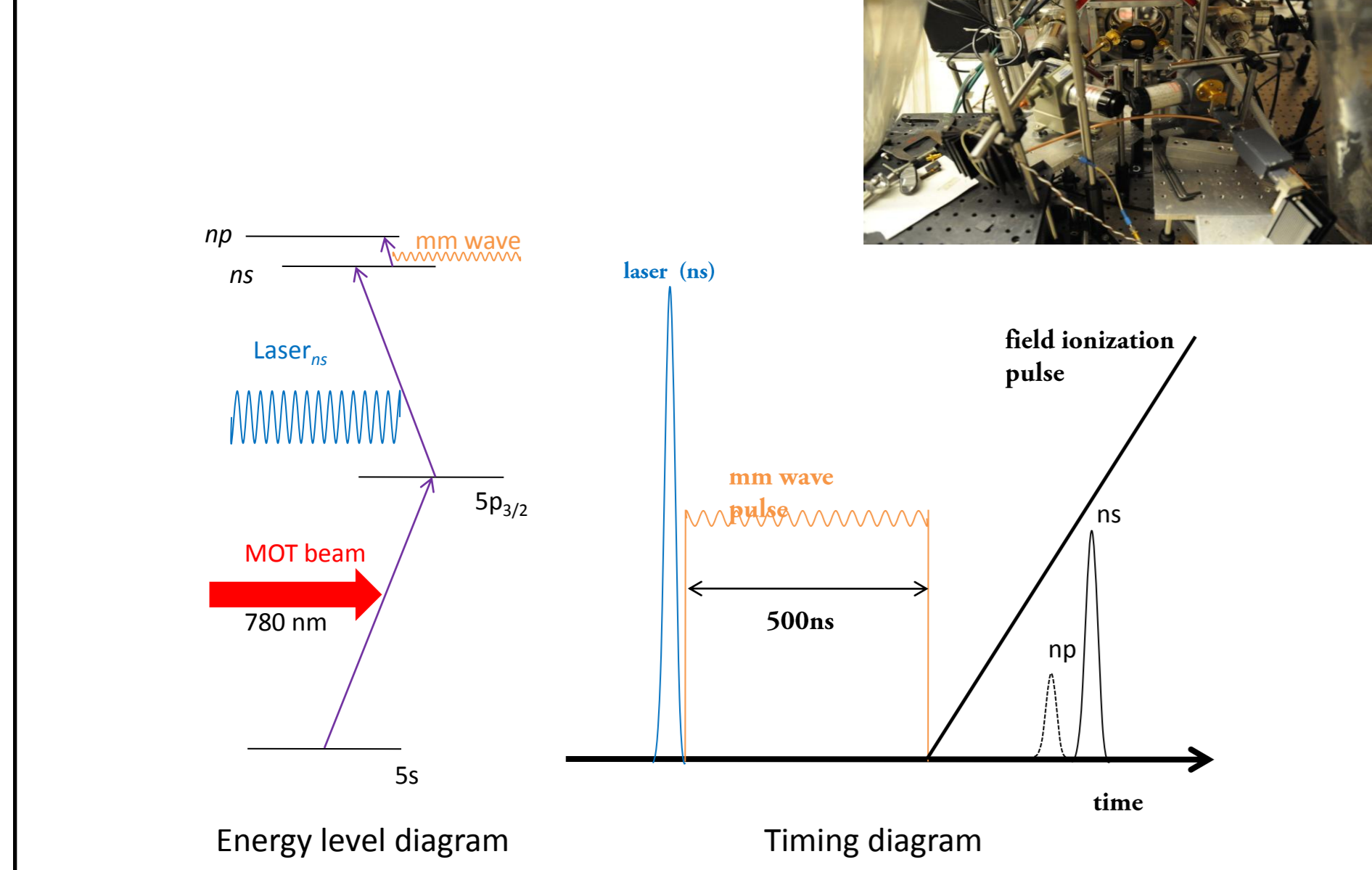


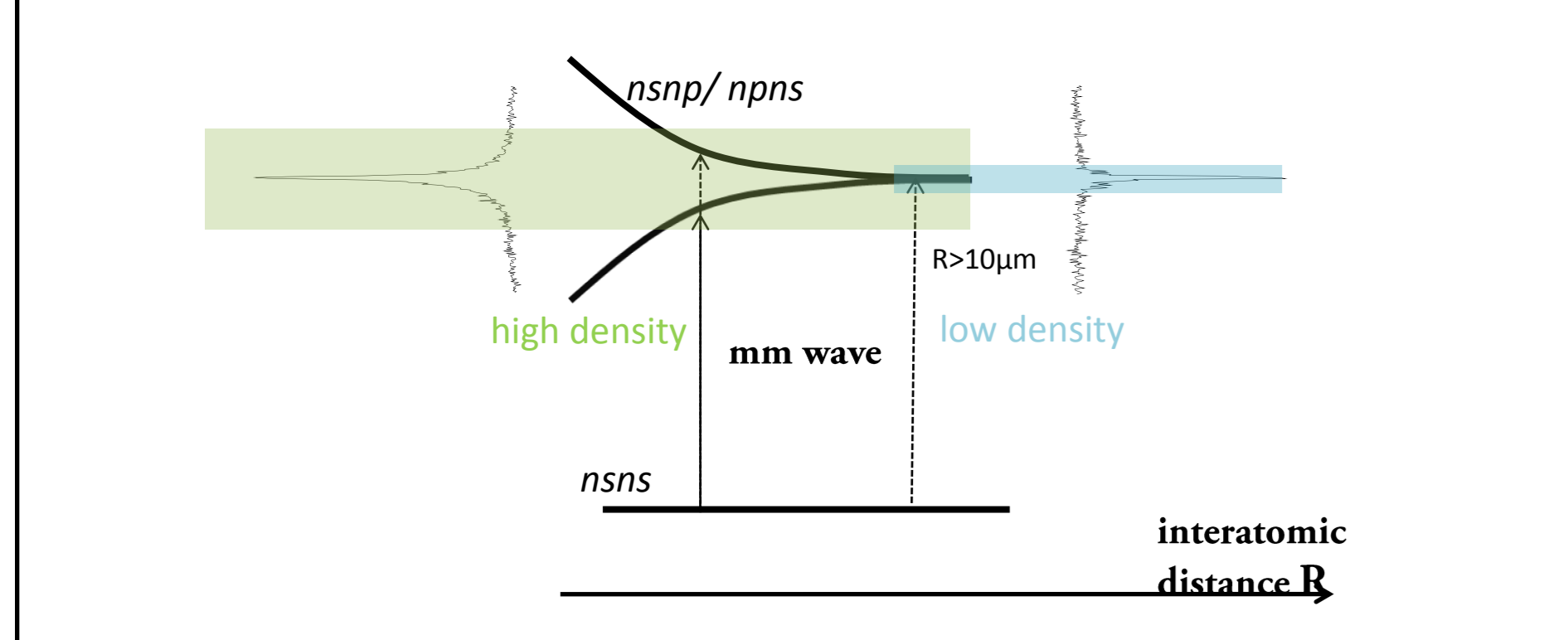
Introduction

The cold Rydberg atom is a fascinating system to study what happens between atoms and its applications. First of all, Rydberg atom is very sensitive to external fields due to the highly excited electron. Due to the fact that electron far away from the nucleus is not under the influence of the Coulombic potential, it is possible to investigate the detailed properties of atoms under moderate external fields. In practice, we succeeded to probe quite small electric field produced from a plasma in my recent work [1] using the sensitivity of Rydberg atoms to the fields. Furthermore, with the help of the magneto-optical trap (MOT), we readily manipulate the inter-atomic distance of almost stationary atoms. A MOT provides us with a 300μK, almost frozen atomic gas and the inter-atomic distance is controllable simply by adjusting the trapping laser power. The spatial control of atoms enables us to explore weak but fundamental forces such as van der Waals and dipole-dipole interactions. Especially, the dipole-dipole potential has been attracting a lot of interests. For instance, the dipole-dipole potential plays an important role in the early stage of evolution to a plasma from Rydberg atoms [2]. The atoms in the dipole-dipole attractive potential collide each other and this triggers the avalanche of the ionization. More interestingly, the dipole-dipole system can be applicable to a quantum gate through the dipole blockade [3]. With the first atom already excited, the excitation of the other one of an atom pair is suppressed due to the shifted energy level, which is caused by the dipole-dipole interaction. Accordingly, a close investigation of the fundamental properties of the dipole-dipole interaction is needed. In spite of the importance, not many works are performed so far. Van Vleck [4] demonstrated the interaction of the sp/ps system and Afrousheh et al.[5] reported the linear increase of the interaction strength in the density range up to 10⁷ cm⁻³. In this work, we extend the density range up to several times 10⁹ cm⁻³ and present a new model to describe the detailed properties including a spin-orbit coupling.

Experiments



How to probe the interaction? Line broadening technique



Depending on the density of the Rydberg gas, different widths of spectra are produced by the microwave resonance measurements.

Results

Theory
Two possible dipole-dipole pictures

The dipole-dipole interaction is given by

$$V_{dd} = \frac{\vec{\mu}_1 \cdot \vec{\mu}_2 - 3(\vec{\mu}_1 \cdot \vec{R})(\vec{\mu}_2 \cdot \vec{R})}{R^3} = \frac{\mu_1 \mu_2 (1 - 3 \cos^2 \theta)}{R^3}$$

$$= \frac{4\pi\rho\mu_{sp}^2 (1 - 3 \cos^2 \theta)}{3} = \frac{4\pi\rho n^4 (1 - 3 \cos^2 \theta)}{3}$$

where ρ is the atomic density and μ_i is the dipole moment of atom i .

According to above equation, the interaction is proportional to the atomic density in a certain n state, $V_{dd} \propto \rho$.

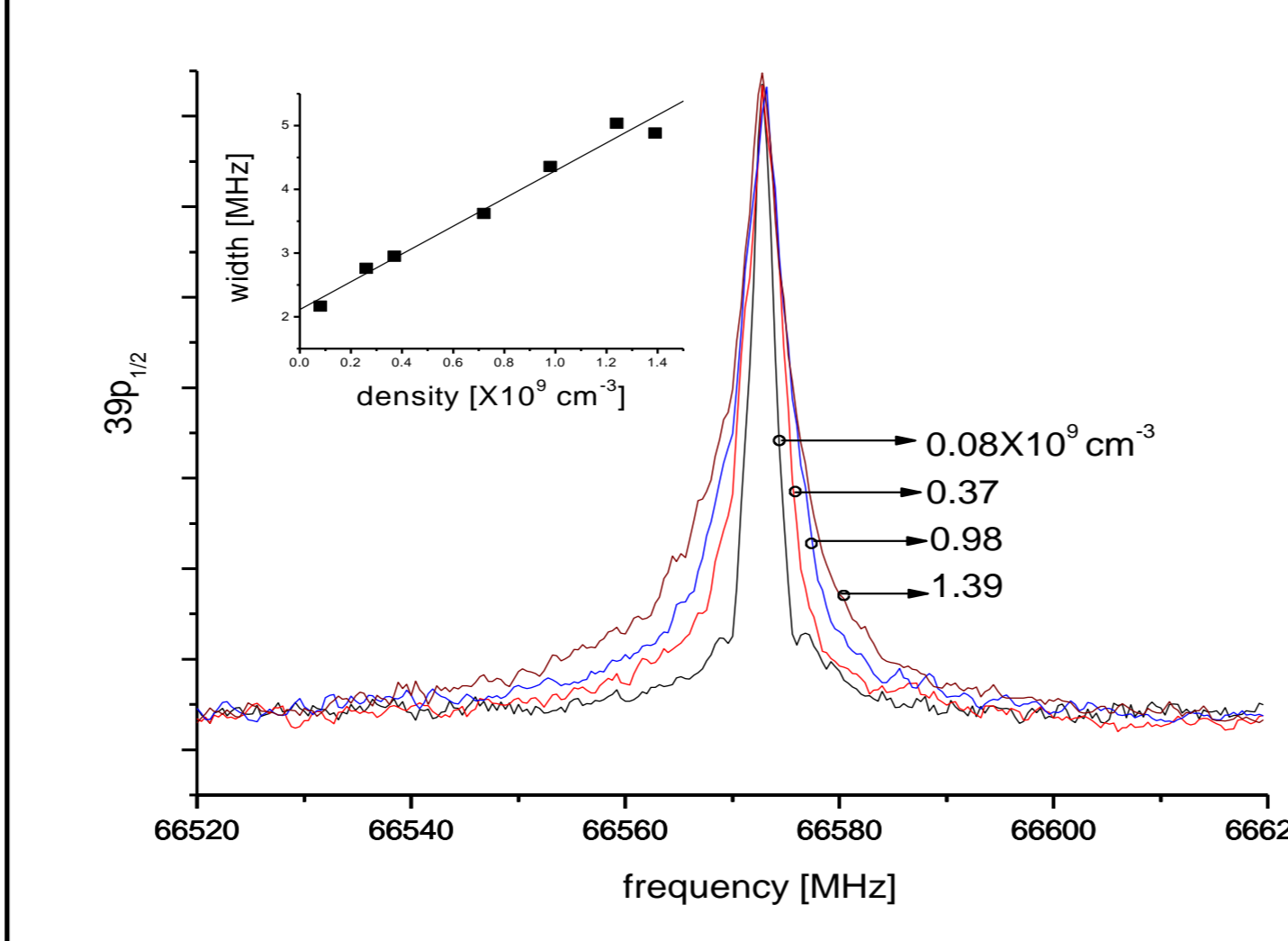


Fig. 1 The observation of line-broadenings of 39s-39p_{1/2} transition in various Rydberg atom densities. As the atomic density is increased, the broader spectrum is observed. The inset shows that the line-width, in other words, the dipole-dipole interaction increases linearly with the atomic density. The linewidth(FWHM) is extracted from a Lorentzian fit to the experimental data.

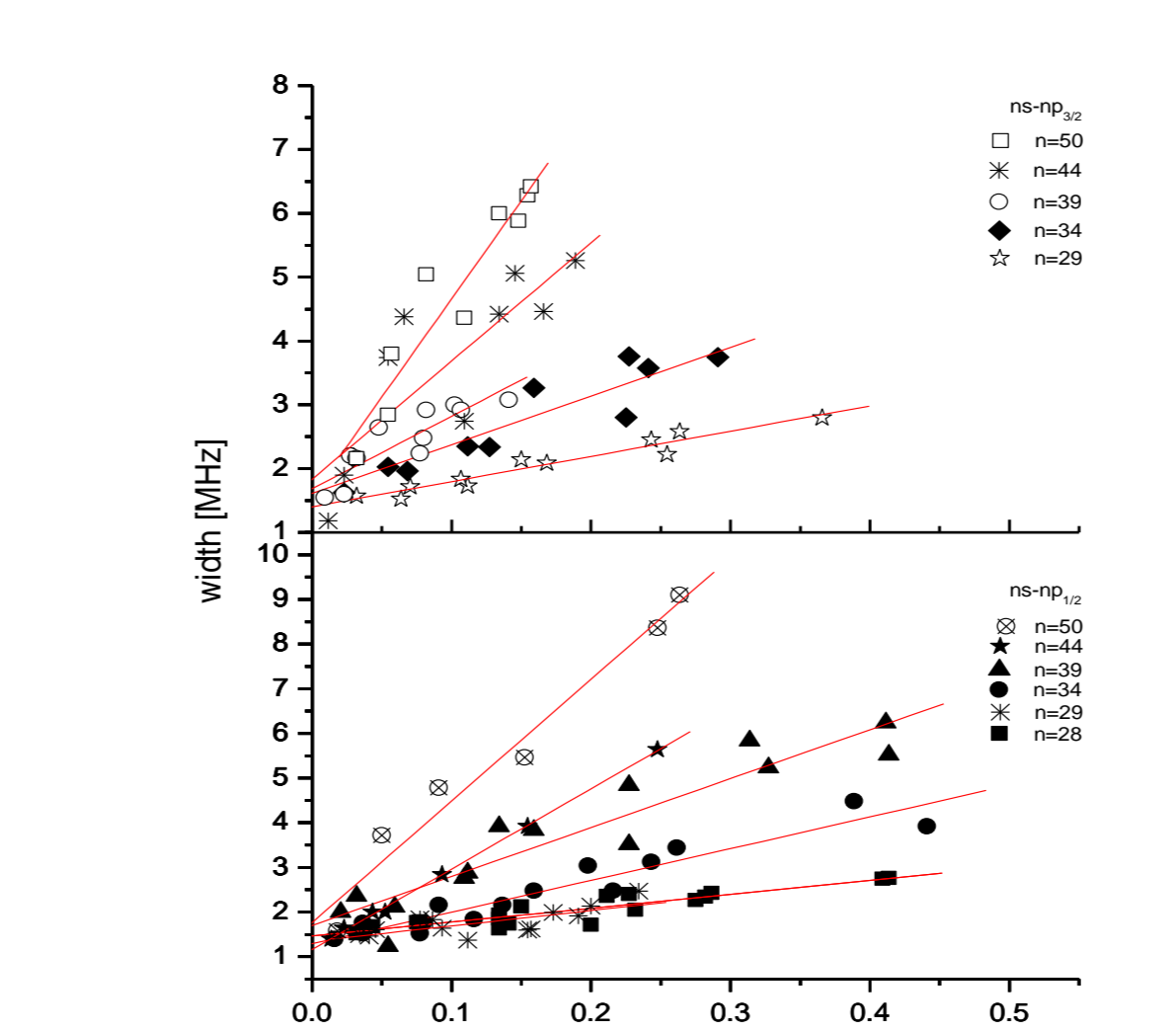


Fig. 2 ns- $np_{1/2}$ transition for $n=28, 29, 34, 39, 44$, and 50 . The broadening rate increases with increasing n due to the fact that the higher n states have the stronger dipole-dipole coupling.

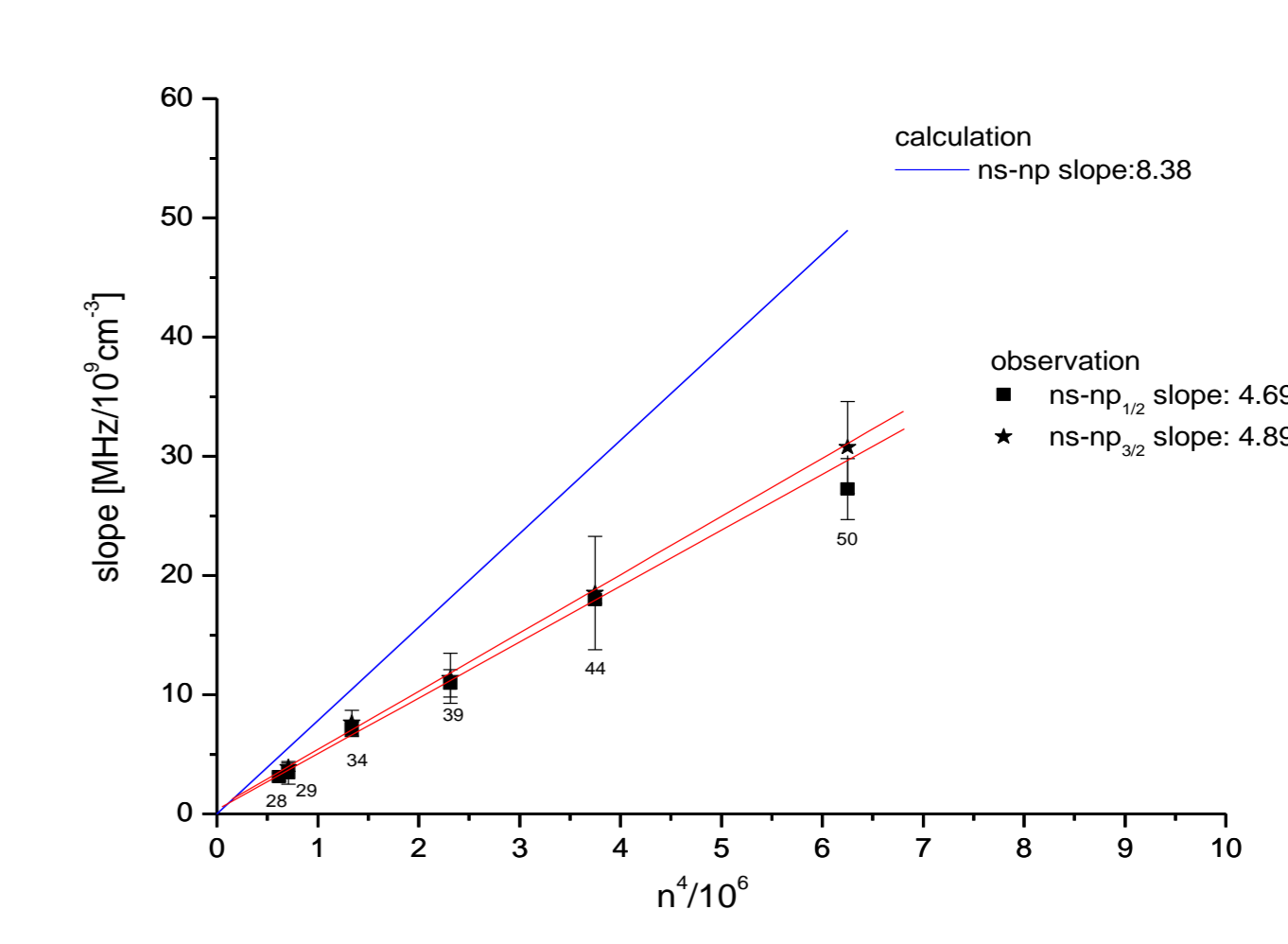


Fig. 3 n^4 scaling of the dipole-dipole interaction. Although the broadening rate shows n^4 scaling as expected, the measured slope does not agree with the theoretical value.

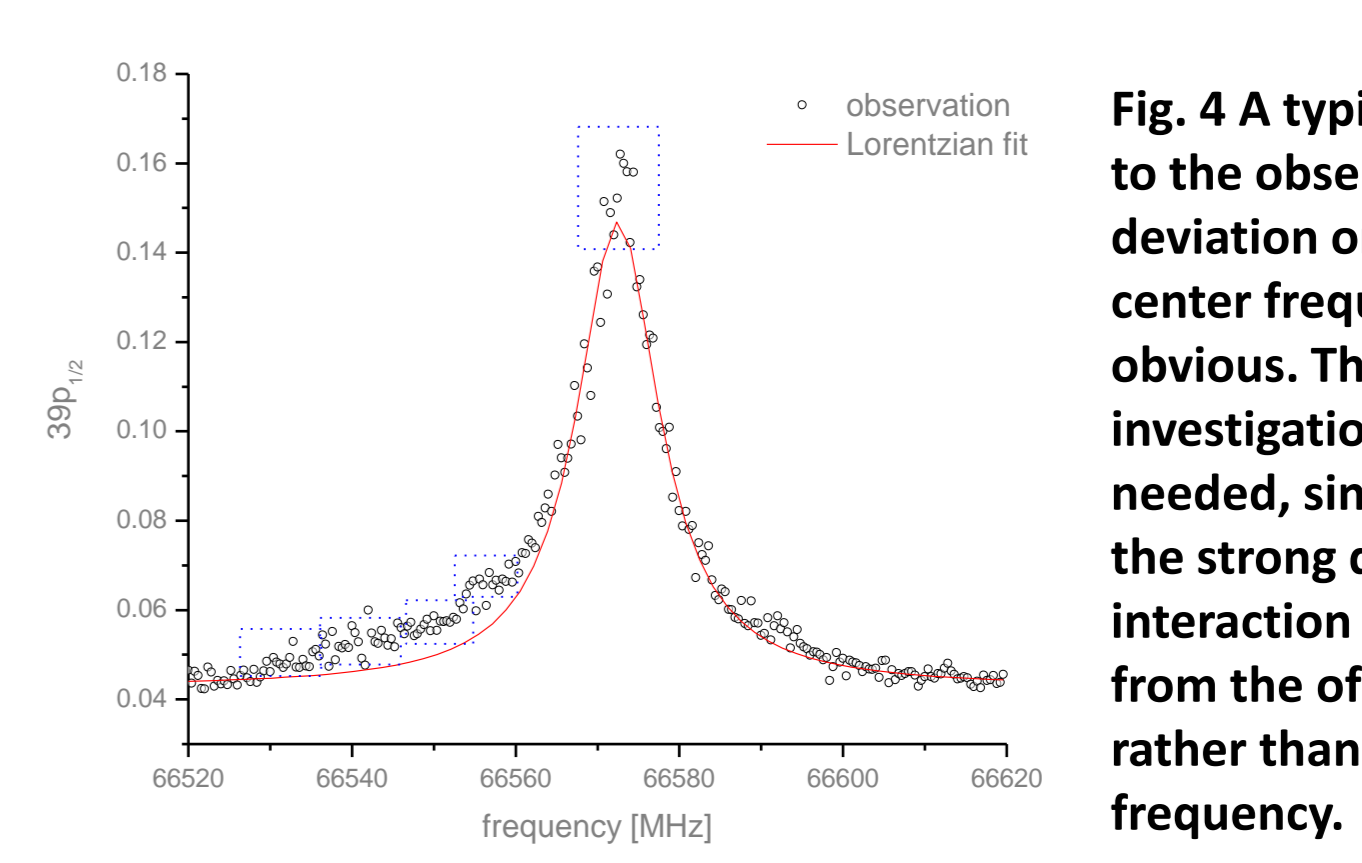


Fig. 4 A typical Lorentzian fit to the observation. The deviation on the wing and center frequency is quite obvious. The closer investigation on the wing is needed, since information of the strong dipole-dipole interaction can be extracted from the off-resonant region rather than at the atomic frequency.

New Model

Including spin, we rewrite V_{dd} and compute the transition probability. In the spinless case, only symmetric initial $nsns$ states are present, while spin creates three symmetric and one antisymmetric states

NOTE: All molecular states are direct products of two atomic states (ex. $nsnp$: atom1 in ns state and atom2 in np state) and symmetry is determined by the interchange of two atoms.

From the general form of the dipole-dipole interaction, we rewrite it in terms of Edmonds' C-tensor.

$$V_{dd} = \frac{\vec{\mu}_1 \cdot \vec{\mu}_2 - 3(\vec{\mu}_1 \cdot \vec{R})(\vec{\mu}_2 \cdot \vec{R})}{R^3} = \frac{x_1 x_2 + y_1 y_2 - 2z_1 z_2}{R^3}$$

where,

$$x + iy = -2\sqrt{\frac{2\pi}{3}} r Y_{11}$$

$$x - iy = 2\sqrt{\frac{2\pi}{3}} r Y_{1,-1}$$

$$z = 2\sqrt{\frac{\pi}{3}} r Y_{10}$$

$$C_k^q = \left(\frac{4\pi}{2k+1}\right)^{1/2} Y_{kq}(\theta, \phi)$$

The final form of the dipole-dipole interaction is,

$$V_{dd} = -\frac{\gamma_{nsnp}^2}{R^3} (C_1^1 C_{-1}^1 + C_{-1}^1 C_1^1 + 2C_0^1 C_0^1)$$

The spin-orbit coupled basis states of $nsnp$ state

M	basis
0	$(ns_{1/2} \uparrow np_{1/2} \uparrow, np_{1/2} \uparrow ns_{1/2} \uparrow, ns_{1/2} \downarrow np_{1/2} \downarrow, np_{1/2} \downarrow ns_{1/2} \downarrow)$
± 1	$(ns_{1/2} \uparrow np_{1/2} \downarrow, np_{1/2} \downarrow ns_{1/2} \uparrow, ns_{1/2} \downarrow np_{1/2} \uparrow, np_{1/2} \uparrow ns_{1/2} \downarrow)$

M	basis
0	$(ns_{1/2} \uparrow np_{3/2} \uparrow, np_{3/2} \uparrow ns_{1/2} \uparrow, ns_{1/2} \downarrow np_{3/2} \downarrow, np_{3/2} \downarrow ns_{1/2} \downarrow)$
± 1	$(ns_{1/2} \uparrow np_{3/2} \downarrow, np_{3/2} \downarrow ns_{1/2} \uparrow, ns_{1/2} \downarrow np_{3/2} \uparrow, np_{3/2} \uparrow ns_{1/2} \downarrow)$
± 2	$(ns_{3/2} \uparrow np_{3/2} \uparrow, np_{3/2} \uparrow ns_{3/2} \uparrow)$

Using the basis and V_{dd} , the angular matrix elements are computed. For example, the dipole-dipole matrix element between $ns_{1/2} np_{1/2}$ and $np_{1/2} ns_{1/2}$ is given by,

$$\langle ns_{1/2} \uparrow np_{1/2} \uparrow | V_{dd} | np_{1/2} \uparrow ns_{1/2} \uparrow \rangle = -\frac{\gamma_{nsnp}^2}{R^3} (s_{1/2} \uparrow | C_1^1 | p_{1/2} \uparrow) (p_{1/2} \uparrow | C_0^1 | s_{1/2} \uparrow)$$

The Hamiltonian matrix is divided into blocks according to the azimuthal angular momentum quantum number M. From now on, the radial matrix elements are omitted. For example, the Hamiltonian of $nsnp_{1/2}$ is given by,

$$H_{M=0}(nsnp_{1/2}) = \begin{pmatrix} 0 & 2/9 & 0 & 2/9 \\ 2/9 & 0 & 2/9 & 0 \\ 0 & 2/9 & 0 & 2/9 \\ 2/9 & 0 & 2/9 & 0 \end{pmatrix} \quad H_{M=\pm 1}(nsnp_{1/2}) = \begin{pmatrix} 0 & -2/9 \\ -2/9 & 0 \end{pmatrix}$$

Diagonalizing the Hamiltonian yields the energy eigenvalues and eigenvectors.

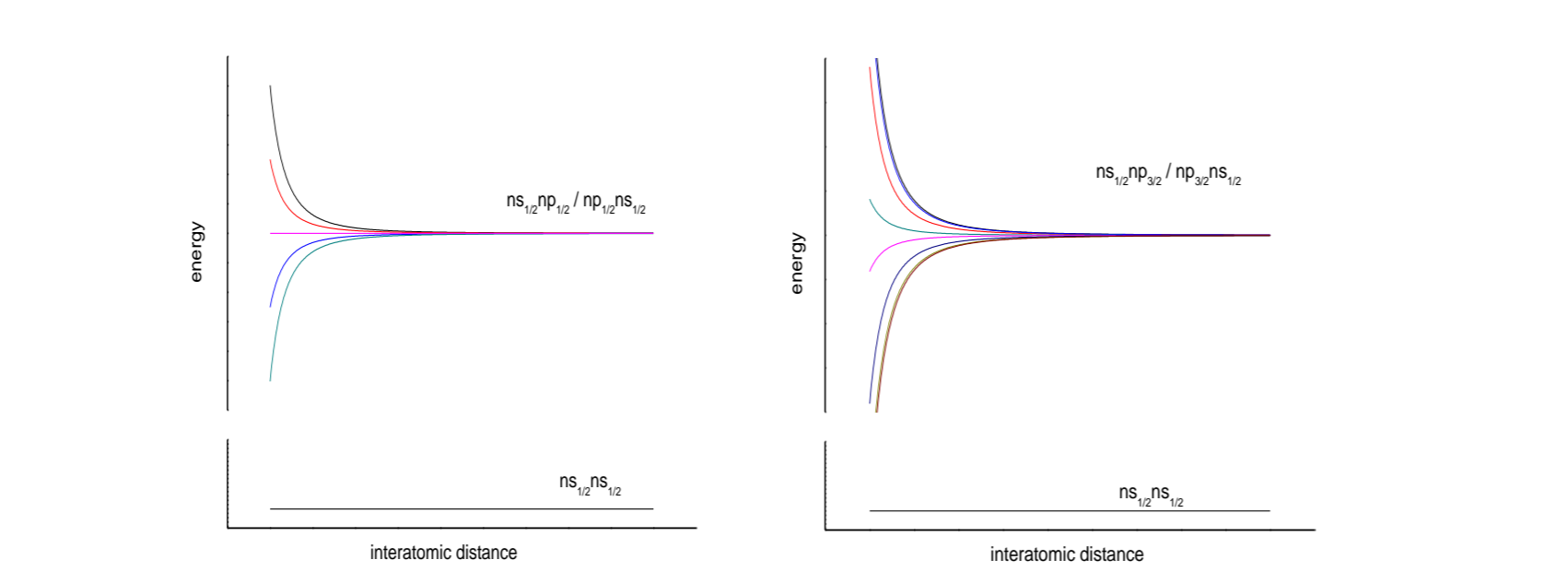


Fig. 5 The energy levels of $nsnp_{1/2}$ and $nsnp_{3/2}$ states. Interestingly, $nsnp_{1/2}$ includes two unshifted energy levels and $nsnp_{3/2}$ includes two slightly shifted level as well as strongly shifted ones. This can explain why a cusped line at the center frequency is observed.

The eigenvalues, eigenvectors, and relative transition probability are summarized in the below tables.

from M	to M	abs(E)	approx. transition matrix element
symmetric	0	0	2/9
	± 1	± 1	0.0718
	± 1	± 1	0.5162
	± 1	0	2/9
	± 1	0	1/9
antisymmetric	0	0	-1/9
	± 1	± 1	2/9
	± 1	± 1	0.0718
	± 1	± 1	-0.5162
	± 1	± 2	1/3
	± 1	± 2	2/9
antisymmetric	0	0	2/9
	± 1	± 1	-0.0718
	± 1	± 1	0.5162
	± 1	± 1	0.26

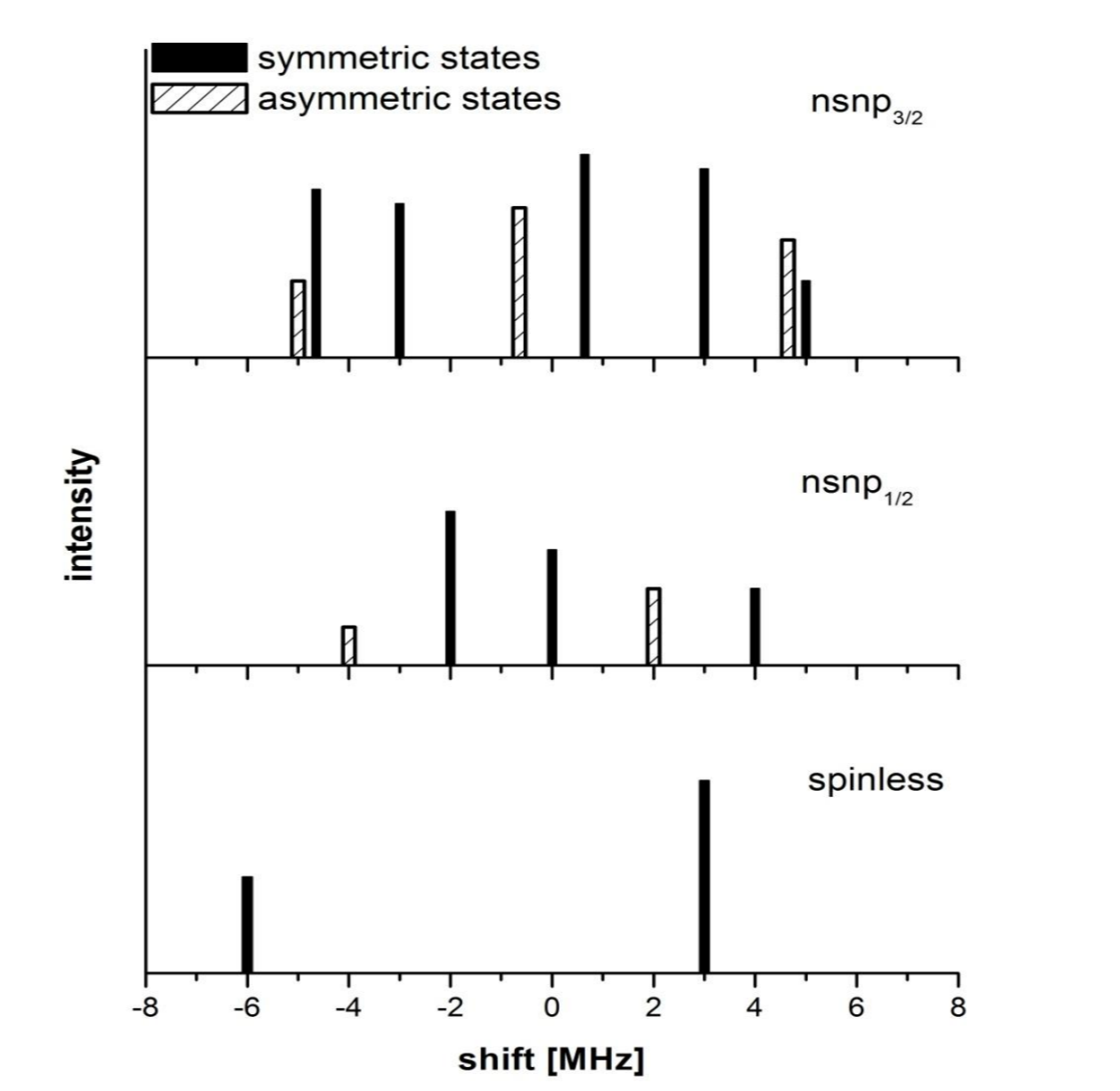


Fig. 6 The frequency shifts and the relative transition strength for with and without spin.

The density distribution of Rydberg atom in MOT

The density is given as a Gaussian form,

$$\rho(w, z) = \rho_0 e^{-\frac{w^2}{r_l^2} - \frac{z^2}{r_M^2}} = \frac{1}{r_l^2} \frac{1}{r_M^2} e^{-\frac{w^2}{r_l^2} - \frac{z^2}{r_M^2}}$$

where, ρ_0 the peak density, r_l the radius of 480nm laser, r_M is the MOT radius.

$$\frac{r_l^2 \ln(\rho_0/\rho)}{w^2} + \frac{z^2}{r_M^2 \ln(\rho_0/\rho)} = 1$$

From the above equation, the volume of Rydberg atom can be written as,

$$V(\rho) = \frac{4\pi}{3} r_l^2 r_M^2 (\ln(\rho_0/\rho))^{3/2}$$

and the total number of atom is given by

$$N = \rho_0 \pi^{3/2} r_l^2 r_M^2$$

Combining V and N gives the atom distribution as a function of the density

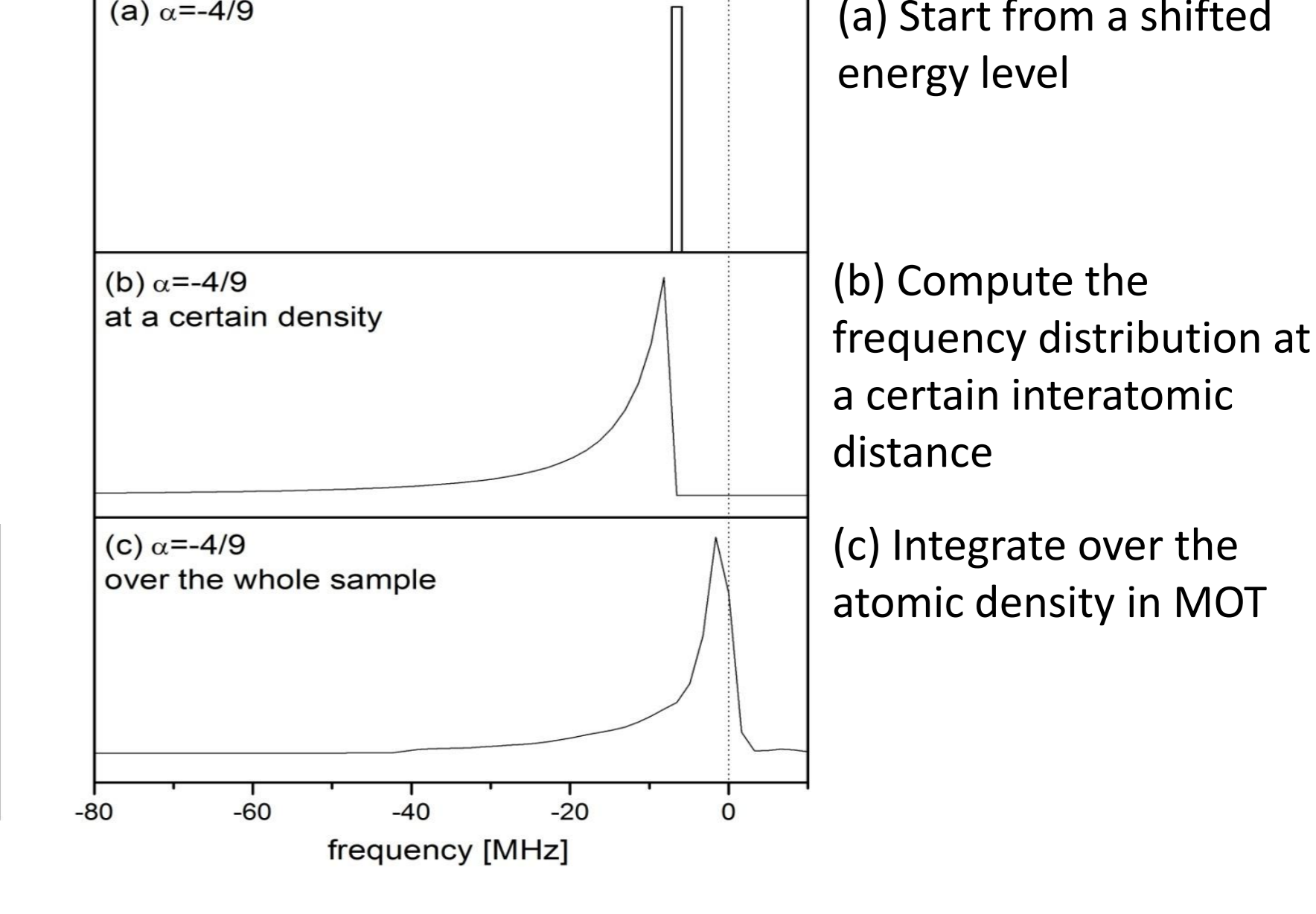
$$\frac{dN}{d\rho} = 2\pi^{3/2} r_l^2 r_M^2 (\ln(\rho_0/\rho))^{1/2}$$

!! Much more atoms are distributed in the low density region, which causes the cusped lineshape.

Lineshape Model

- The probability of R being between R and R+dR is $dP(R) = \frac{4\pi R^2 dR}{4\pi R_{av}^3} = 4\pi R^2 \rho(r) dr$ for $R < R_{av}$ where R_{av} is the average interatomic distance
- The frequency shifts due to the dipole-dipole interaction at R is $f(R) = \frac{\alpha}{R^3}$ where α is the eigenvalues.
- The probability of finding the frequency shift df between f and df, $\frac{dP}{dR} = \frac{dP(R)}{dR} \cdot \frac{dR}{df} = \left(-\frac{4\pi\alpha}{3} \rho(r)\right) \frac{1}{f^2}$
- Taking into account the whole Rydberg atom cloud, the frequency shifted lineshape is, $L_{\text{shift}}(f) = \sum_n \int_{r=0}^{\infty} \int_{\theta=0}^{\pi} \left(-\frac{4\pi\alpha}{3} \rho(r)\right) \frac{1}{f^2} 4\pi\rho(r)r^2 dr$
 L_{shift} is weighted according to the transition probability
- Convolution of L_{shift} and L_{Rabi}
 $L_{\text{conv}} = \text{convolution}(L_{\text{shift}}, L_{\text{Rabi}})$
where $L_{\text{Rabi}}(f) = \frac{b^2}{b^2 + f^2} \sin^2\left\{\frac{1}{2}\pi(b^2 + f^2)^{1/2}\right\}$ is the Rabi-oscillation line profile of two level system.

Fig. 7 The procedure of producing the lineshape model



Line fitting

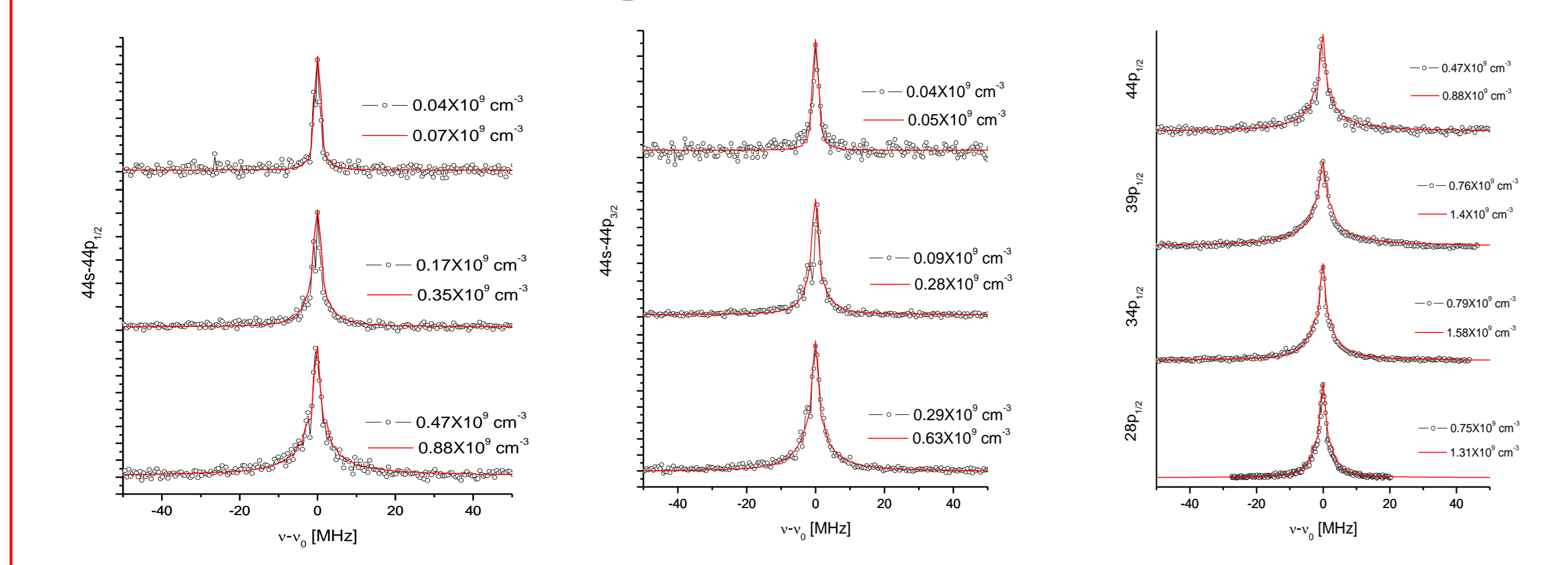


Fig. 8 The line fittings to the observations. The model agrees very well with the observed data including the cusped and side wings. The density discrepancy between the observation and the model possibly comes from the uncertainty of the density measurements, which is typically 100-200%.

Conclusion

- We have observed n^4 scaling of the dipole-dipole interaction.
- Dipole coupled energy levels(sp/ps) are calculated including the electron spin.
- The cusped lineshape is due to the reduced, even zero, dipole-dipole shifts due to the spin-orbit coupling and the density distribution of the trap.
- There is zero weighted average shift, but due to the asymmetry of the pattern of lines shown in Fig. 6, the lineshape is asymmetric.

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