# Rydberg atom mediated non-destructive readout of rotational states of polar molecules and indirect molecular interactions

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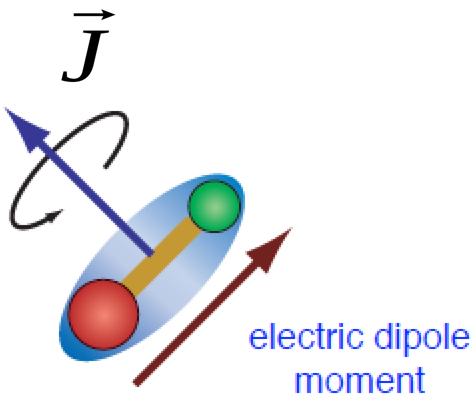
# Ultracold polar molecules: basic features



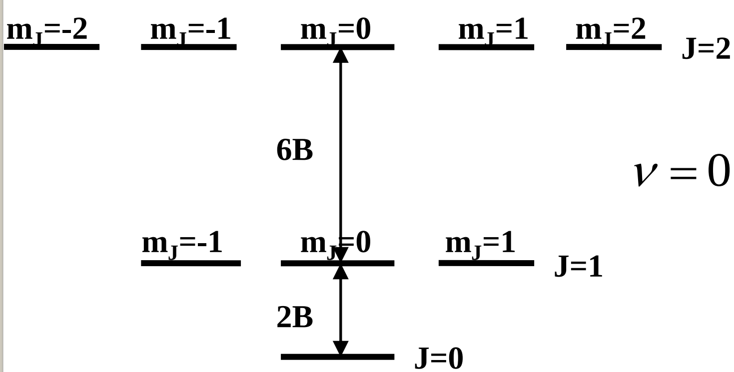

Rich level structure:

-electronic E~ $10^4$ cm$^{-1}$
-vibrational ~ 100 cm$^{-1}$
-rotational ~ 1-10 GHz
- hyperfine ~ kHz-MHz transitions

-Permanent electric dipole moment d~0.1-6 D

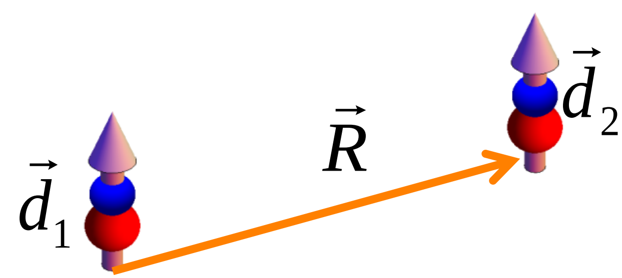
-State control by AC, DC electric and magnetic fields

-Long-range, anisotropic dipole-dipole interaction




$$V_{dd} = \frac{\vec{d}_1\vec{d}_2}{R^3} - \frac{3(\vec{d}_1\vec{R})(\vec{d}_2\vec{R})}{R^5}$$

$$\nu = 0$$




$$H_{rot}\,|J,m_J\rangle = B\vec{J}^2\,|J,m_J\rangle = BJ(J+1)\,|J,m_J\rangle$$

$|\langle J,m_J\,|\,\vec{d}\,|\,J\pm1,m_J'\rangle| \sim d$ - rotational transitions dipole moment of the order of permanent dipole moment

# Ultracold polar molecules: applications

-Quantum computation and quantum simulation

quantum magnetism: Ising, Heisenberg, XXZ models (A.V.Gorshkov, et. al. PRL **107**, 115301(2007))

topologically ordered states (N.Yao et. al., PRL **109**, 266804(2011))

highly entangled states, e.g. cluster state for MBQC (K.R.A. Hazzard et. al. PRA **90**, 063622(2014))

- novel quantum phases and phase transitions induced by dipole-dipole interactions

dipolar crystals (H.P. Buchler et al., PRL 98, 060404 (2007))

Hubbard model with long-ranged interactions (B. Capogrosso-Sansone et al. PRL 104, 125301 (2010))

- quantum chemistry at cold and ultracold temperatures (S. Ospelkaus, et al. Science 327, 853 (2010))
- high precision spectroscopy to measure fundamental constants, tests of Standard Model (J.J.Hudson et al. Nature 473, 493 (2011))

# Coherent dipolar spin-exchange with KRb molecules in 3D optical lattice (B.Yan et. al. Nature **501**, 521 (2013))

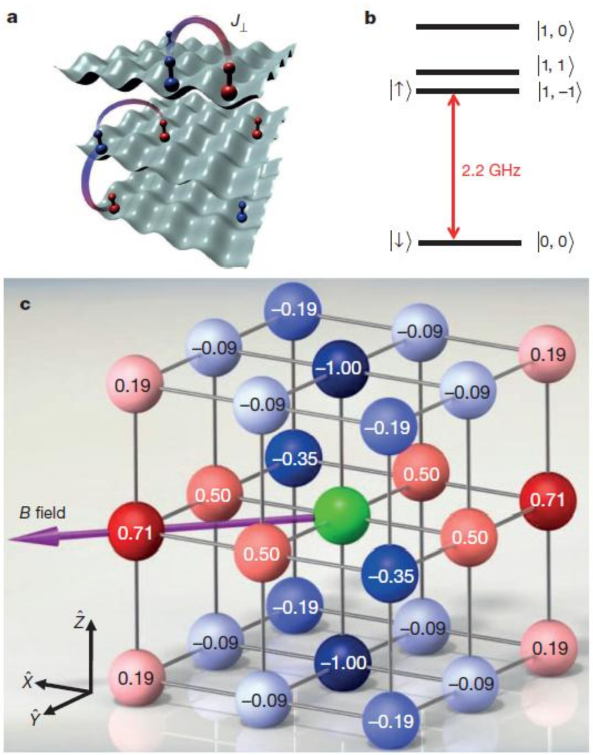



**Figure 1 | Dipolar interactions of polar molecules in a 3D lattice.** **a**, Polar molecules are loaded into a deep 3D optical lattice. Microwaves are used to address the transition between two rotational states (red and blue represent different rotational states). $J_\perp$ characterizes the spin-exchange interaction energy. **b**, Schematic energy diagram (not to scale) for the ground and first excited rotational states. The degeneracy of the excited rotational states is broken as a result of a weak coupling of the nuclear and rotational degrees of freedom. We use $|0,0\rangle$ and $|1,-1\rangle$ as our two spin states. **c**, The interaction energy between any two molecules depends on their relative position in the lattice. The numbers shown give the geometrical factor $-V_{dd}(r_i - r_j)$ for the dipolar interaction of each site relative to the central site (green), under the specific quantization axis ($B$ field). Negative values (blue) correspond to attractive interactions, and positive values (red) correspond to repulsive interactions.

- Rotational states $|\downarrow\rangle = |J=0, m_J=0\rangle$, $|\uparrow\rangle = |J=1, m_J=-1\rangle$ can form a spin-1/2 system
- In the absence of DC electric field interaction Hamiltonian is:

$$\hat{H} = \frac{J_\perp}{2}\sum_{i>j} V_{dd}(\vec{r}_i - \vec{r}_j)\left(\hat{S}_i^+\hat{S}_j^- + \hat{S}_i^-\hat{S}_j^+\right)$$

$$V_{dd} = \left(1 - 3\cos^2\Theta_{ij}\right)/|\vec{r}_i - \vec{r}_j|^3$$

$$J_\perp = d_{\uparrow\downarrow}{}^2/4\pi\varepsilon_0 a^3 \approx 48\ Hz$$

-strongest nearest neighbor interaction strength

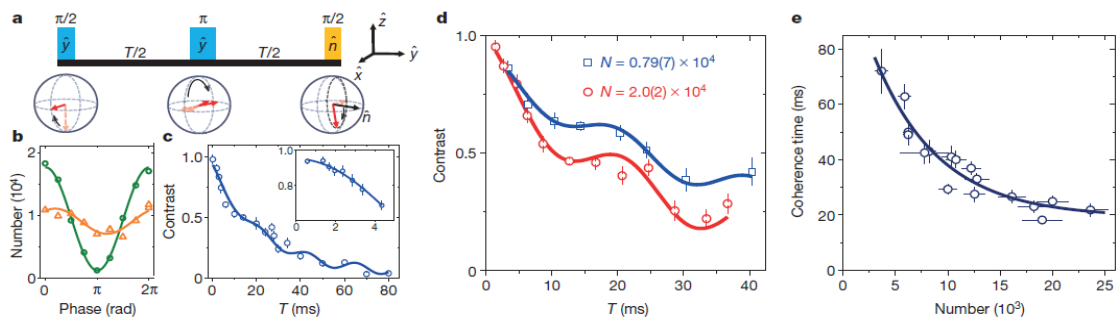
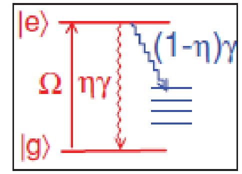



**Figure 2 | Coherent spin dynamics of polar molecules.** **a**, A $(\pi/2)_y$-pulse initializes the molecules in a coherent superposition of rotational states. A spin-echo pulse sequence is used to correct for effects arising from single-particle inhomogeneities across the sample, such as residual light shifts. **b**, The phase of the final $\pi/2$-pulse is scanned (corresponding to rotations around a variable axis, $\hat{n}$) to obtain a Ramsey fringe. Two fringes are shown, corresponding to the short (green circles) and intermediate timescales (orange triangles). **c**, The contrast of the Ramsey fringe is measured as a function of interrogation time. Because the molecules' spin states are initially all in phase, at very short times, $T < 2h/J_\perp$, the contrast decay curve should be quadratic[17], as shown in the inset. **d**, The contrast of the Ramsey fringe versus interrogation time is shown for two different filling factors, characterized by the initial molecule number. In addition to the density-dependent decay, we observe oscillations, which arise from spin-exchange interactions between neighbouring molecules. **e**, The spin coherence time decreases for increasing molecule number. The solid line shows a fit to $C + A/N$, where $C$ and $A$ are constants. Error bars, 1 s.d.

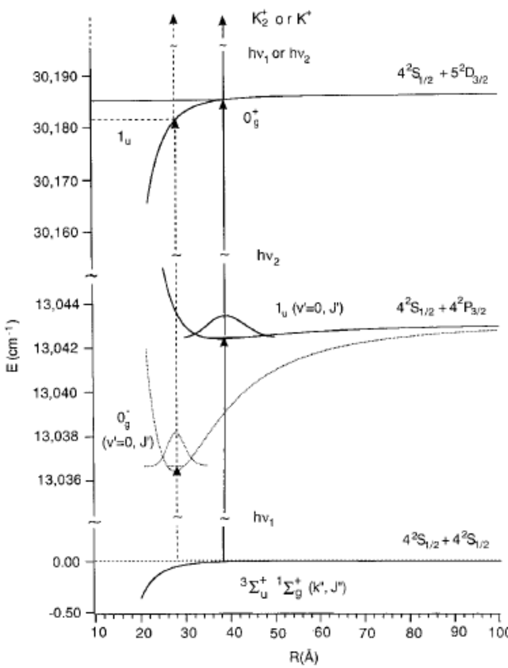
# Readout of molecular states

- molecular states have to be read out at the end of evolution

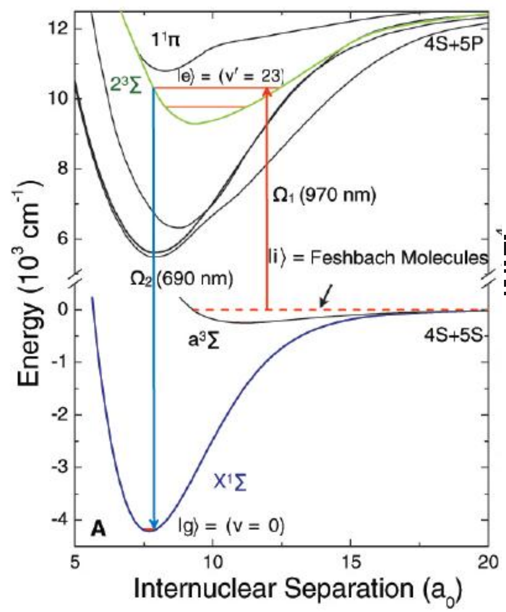
-lack of cycling transitions prevents state readout by fluorescence detection

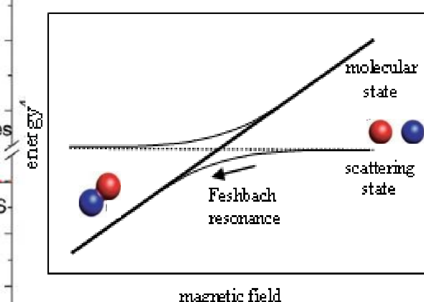



- molecular states are detected destructively via REMPI or Feshbach dissociation + detection of atomic fluorescence



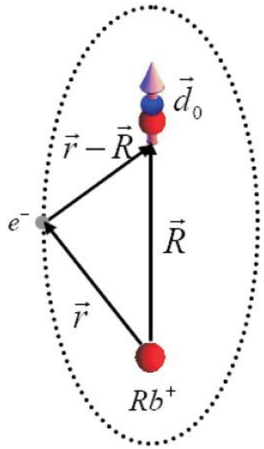

K$_2$ detection, W.C. Stwalley et. al., J. Mol. Spectr. **195**, 184 (1999)




KRb detection, K.-K. Ni et. al., Science, **322**, 231 (2008)

# Charge-dipole interaction of polar molecule and Rydberg atom




- molecular dipole interacts with electric field produced by Rydberg atom ionic core and outer electron

$$V_{ch-dip} = \frac{e\vec{d}_0\vec{R}}{R^3} - \frac{e\vec{d}_0(\vec{R}-\vec{r})}{|\vec{R}-\vec{r}|^3}$$

-at $d < d_c = 1.63\,D$ bound Rydberg atom-polar molecule states are predicted to form(polyatomic Rydberg molecules)
S.T. Rittenhouse, H.R. Sadeghpour, PRL **104**, 243002 (2010);
S.T.Rittenhouse, M. Mayle, P. Schmelcher, H.R. Sadeghpour, J. Phys. B **44**, 184005 (2011)

$$H = H_{Rydb} + H_{mol} + V_{ch-dip}$$

-system Hamiltonian is diagonalized in a basis of unperturbed atomic electronic and molecular rotational states $|nlm\rangle|Jm_J\rangle$ to obtain dressed atom-molecule states

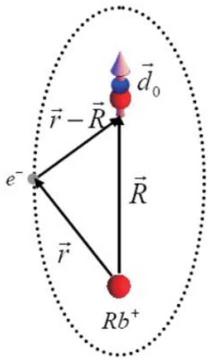
$$\langle J,m_J|\langle nlm|H_{Rydb}|n'm'l'\rangle|J',m_J'\rangle = -\frac{1}{2(n-\mu_l)^2}\delta_{nn'}\delta_{ll'}\delta_{mm'}\delta_{JJ'}\delta_{m_Jm_J}$$

$$\langle J,m_J|\langle nlm|H_{mol}|n'm'l'\rangle|J',m_J'\rangle = BJ(J+1)\delta_{nn'}\delta_{ll'}\delta_{mm'}\delta_{JJ'}\delta_{m_Jm_J}$$

$$\langle J,m_J|\langle nlm|V_{ch-dip}|n'm'l'\rangle|J',m_J'\rangle =$$

$$= e\langle J,m_J|\vec{d}|J',m_J'\rangle\left(\frac{\vec{R}}{R^3}\delta_{nn'}\delta_{ll'}\delta_{mm'} - \langle nlm|\frac{\vec{R}-\vec{r}}{|\vec{R}-\vec{r}|^3}|n'l'm'\rangle\right)$$

# Readout of molecular rotational states via interaction with Rydberg atom




- use probe qubit to measure spectroscopic qubit similar to quantum logic spectroscopy

-electric field of Rydberg ionic core and electron interacts with molecular dipole and changes energies of rotational states

$|\uparrow\rangle = |J=1, m_J=0\rangle$

$2B$

$|\downarrow\rangle = |J=0, m_J=0\rangle$

$$H = \begin{pmatrix} E_{Rydb} & V_{ch-dip} \\ V_{ch-dip} & E_{Rydb}+2B \end{pmatrix}$$

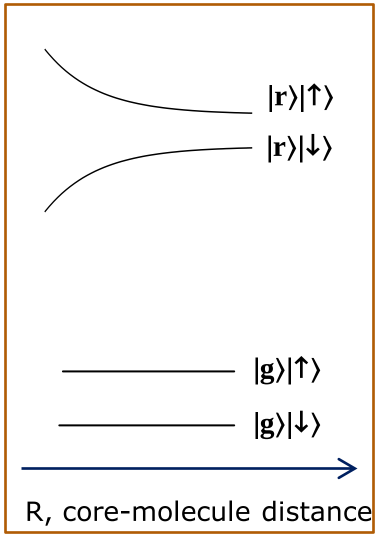
- energies of dressed atom-molecule states

$$E_\pm = E_{Rydb} + B \pm \sqrt{B^2 + V_{ch-dip}^2} \propto \begin{cases} E_{Rydb} + 2B + V_{ch-dip}^2 / 2B \\ E_{Rydb} - V_{ch-dip}^2 / 2B \end{cases}, \quad |V_{ch-dip}| << B$$

$$V_{ch-dip} = \langle ns | \langle J=0, m_J=0 | \hat{V}_{ch-dip} | J=1, m=0 \rangle | ns \rangle \sim 1/R^2$$

$$|-\rangle \approx |J=0, m_J=0\rangle$$

$$|+\rangle \approx |J=1, m_J=0\rangle$$




E.Kuznetsova, S.T. Rittenhouse, H.R. Sadeghpour, S.F. Yelin, Phys. Chem. Chem. Phys. **13**, 17115 (2011).

# Energies of dressed atom-molecule states for KRb+Rb(60s) and RbYb+Rb(60s)

$$\approx |60s\rangle\,|J=0, m_J=0\rangle, \quad |60s\rangle\,|J=1, m_J=0,\pm1\rangle$$

-basis states: Rb

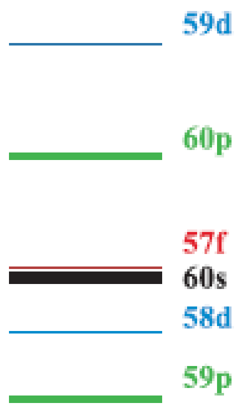
$$60s,\ 60p,\ 59p,\ 59d,\ 58d,\ 57f$$

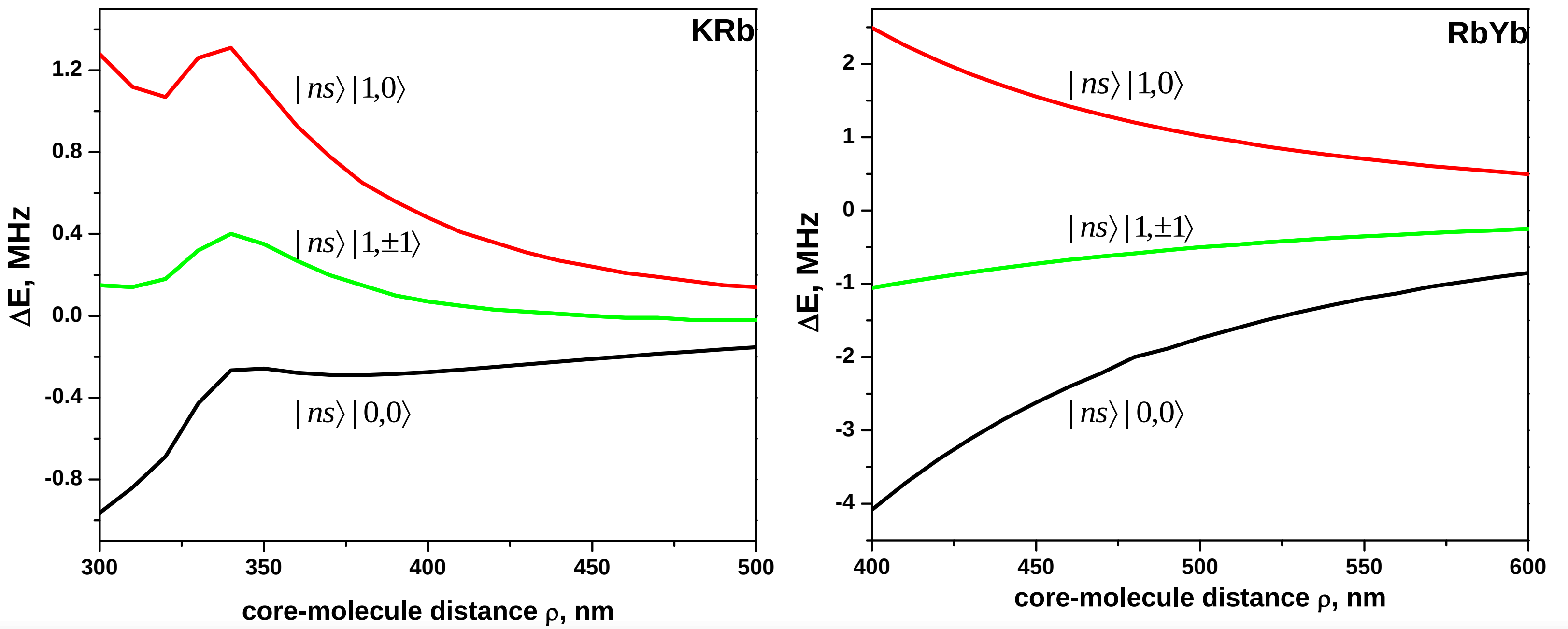
$$\mu_s = 3.13,\ \mu_p = 2.65,\ \mu_d = 1.34,\ \mu_f = 0.016$$

- rotational molecular states

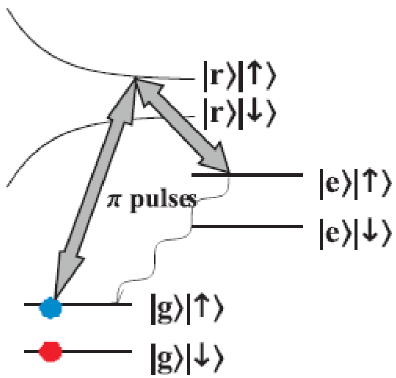
$$|J=0, m_J=0\rangle,\ |J=1, m_J=0,\pm1\rangle,\ |J=2, m_J=0,\pm1,\pm2\rangle$$

- KRb: $d = 0.566\ D, \quad B = 1114\ MHz$
- RbYb: $d = 1\ D, \quad B = 353\ MHz$



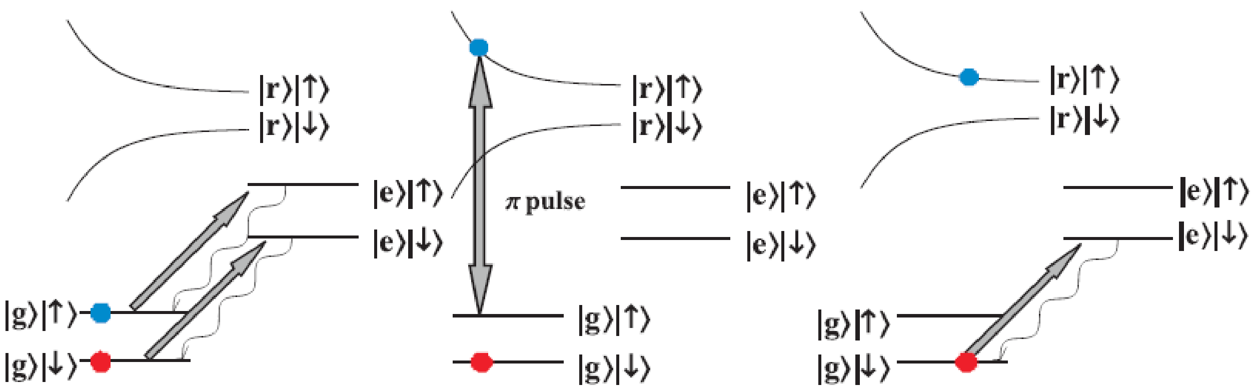



# Readout of rotational states via selective excitation to dressed atom-molecule states




$$(\alpha|\downarrow\rangle+\beta|\uparrow\rangle)|g\rangle \Rightarrow \alpha|\downarrow\rangle|g\rangle+\beta|\uparrow\rangle|r\rangle \Rightarrow \alpha|\downarrow\rangle|g\rangle+\beta|\uparrow\rangle|e\rangle$$

photon detection: $|\uparrow\rangle$ state
no photon: $|\downarrow\rangle$ state




1) Atom in $|g\rangle$ , full atomic fluorescence signal measured

2) System is selectively excited to Rydberg state $|g\rangle|\uparrow\rangle \rightarrow |r\rangle|\uparrow\rangle$: $(\alpha|\downarrow\rangle+\beta|\uparrow\rangle)|g\rangle \Rightarrow \alpha|\downarrow\rangle|g\rangle+\beta|\uparrow\rangle|r\rangle$

3) Population $|\alpha|^2$ in $|g\rangle$ measured via atomic fluorescence

4) Measurement is of QND type (T.C. Ralph et al., PRA 73, 012113 (2006))

# Readout of collective rotational molecular states via interaction with Rydberg superatom

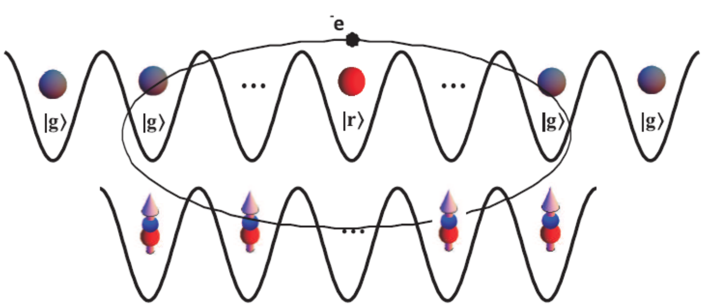
-molecular array interacts with atomic system in a single collective Rydberg excitation (superatom)

- energies of dressed states of molecules and superatom depend on collective molecular rotational states

$$|\downarrow\uparrow\uparrow\ldots\rangle$$

$$|\downarrow\rangle = |J=0, m_J=0\rangle,\ |\uparrow\rangle = |J=1, m_J=0,\pm1\rangle$$

- selective excitation of atom to Rydberg state and detection of atomic fluorescence will measure population of the selected collective rotational state




Model system:
-1D array of polar molecules (KRb or RbYb), each in superposition of $|\downarrow\rangle, |\uparrow\rangle$ states
- parallel 1D array of atoms in superatom state

$$|\Psi_{atom}\rangle = \frac{1}{\sqrt{N_a}} \sum_{j=1}^{N_a} e^{i\vec{k}\vec{r}_j} |g_1, g_2, \ldots r_j \ldots g_{N_a}\rangle$$

E. Kuznetsova, S.T. Rittenhouse, H.R. Sadeghpour, S.F. Yelin, PRA **94**, 032325 (2016).

- Arrays of $N = 3,5$ molecules and $N_a = N + 2$ atoms

- System Hamiltonian was diagonalized in a basis of atomic electronic and collective rotational molecular states

$$|\Psi_{atom}\rangle|\Psi_{mol}\rangle, \quad |\Psi_{mol}\rangle = |a_1, a_{2,} ... a_N\rangle$$

- Simplified basis: only $|r_j\rangle = |60s\rangle$ and $|a_i\rangle = |J=0, m_J=0\rangle, |J=1, m_J=0\rangle$ basis states were used

$$\langle\Psi_{atom}|\langle\Psi_{mol}|H_{atom}|\Psi_{mol}'\rangle|\Psi_{atom}\rangle = -\frac{1}{2(ns-\mu_s)^2}\Pi_{i=1}^{N}\delta_{a_i,a_i'}$$

$$\langle\Psi_{atom}|\langle\Psi_{mol}|H_{mol}|\Psi_{mol}'\rangle|\Psi_{atom}\rangle = \left(\sum_{i=1}^{N} BJ_i(J_i+1)\right)\Pi_{i=1}^{N}\delta_{a_i,a_i'}$$

$$\langle\Psi_{atom}|\langle\Psi_{mol}|V_{ch-dip,i}|\Psi_{mol}'\rangle|\Psi_{atom}\rangle = \left(\frac{1}{N_a}\sum_{j=1}^{N_a}\langle a_i|V_{ji}|a_i'\rangle\right)\Pi_{k=1,k\neq i}^{N}\delta_{a_k,a_k'}\delta_{J_i,J_i'\pm 1}$$

$$V_{ji} = \langle r_j|V_{ch-dip,i}|r_j\rangle = \langle r_j|\frac{e\vec{d}_i\vec{R}_{ji}}{R_{ji}^3} - \frac{e\vec{d}_i\left(\vec{R}_{ji}-\vec{r}\right)}{|\vec{R}_{ji}-\vec{r}|^3}|r_j\rangle$$

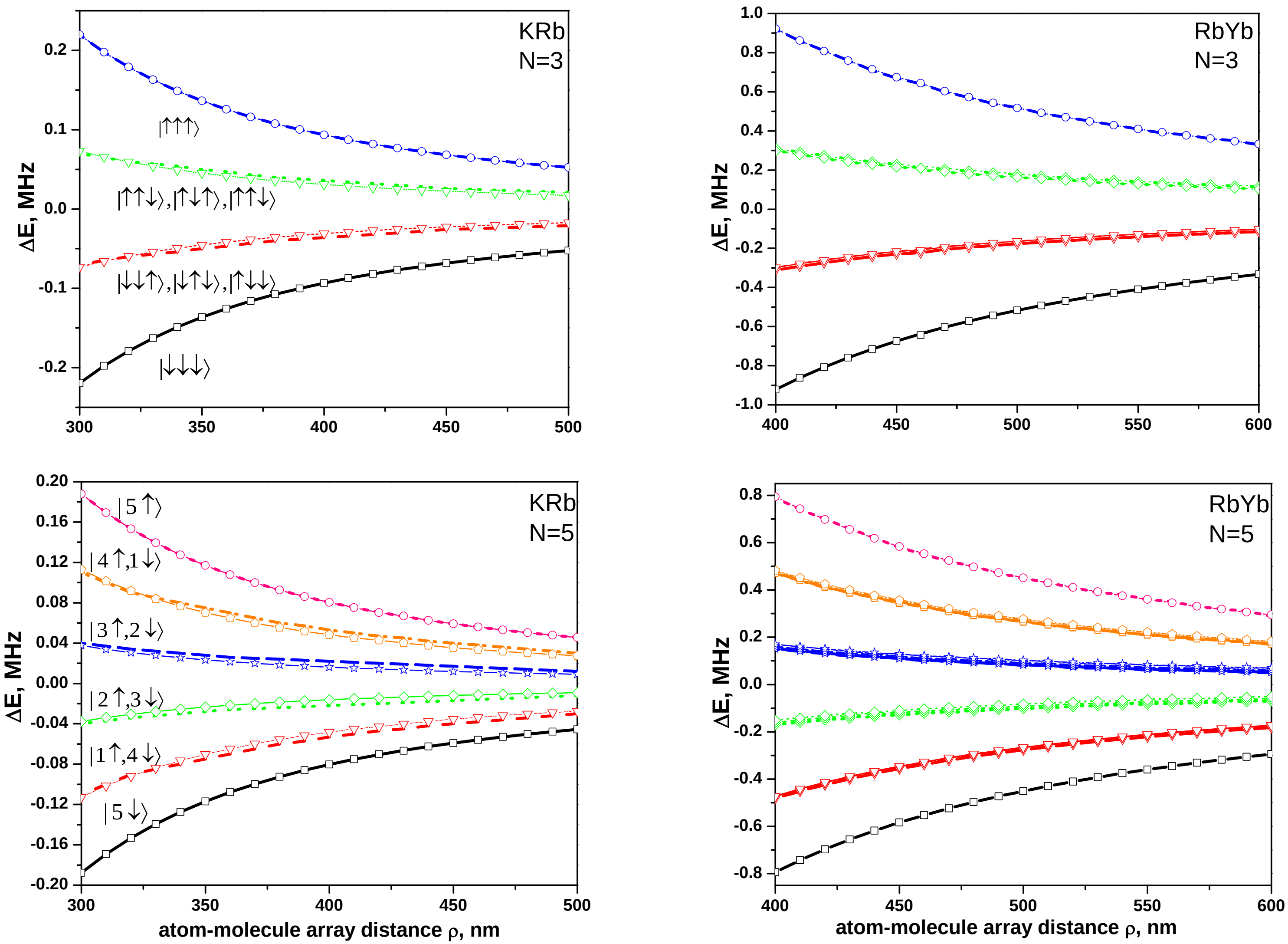
-interaction matrix element between $i^{th}$ molecule and $j^{th}$ atom

- For $|V_{ji}| << E_{rot}$ the energy shift of $(k\uparrow,(N-k)\downarrow)$ state

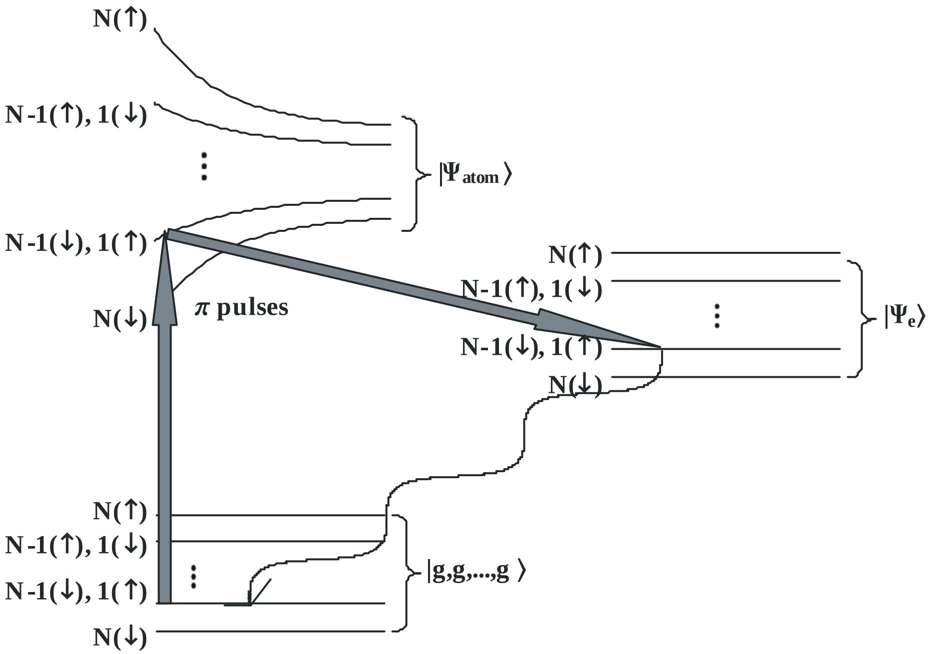
$$\Delta E_{k\uparrow,(N-k)\downarrow} \approx -\frac{(N-2k)}{N_a^2}\frac{|V_{j=i}|^2}{E_{rot}} \sim -\frac{1}{N}\frac{|V_{j=i}|^2}{E_{rot}}$$ for linear molecular array

$$\Delta E_{k\uparrow,(N-k)\downarrow} \approx -(N-2k)\frac{|V_{j=i}|^2}{E_{rot}}$$ for ring molecular array

# Energy shifts of dressed states of 1D linear N molecules array + 1D linear array of atoms in superatom state



# Readout of collective molecular rotational states




-atoms are excited to superatom state selectively only for

$$|\Psi_{molk\uparrow}\rangle = |k\uparrow,(N-k)\downarrow\rangle$$

- superatom is transferred to short-lived state

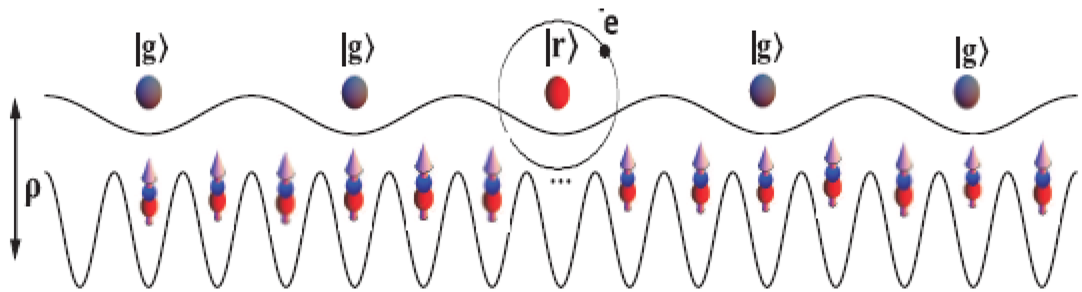
$$|\Psi_e\rangle = \frac{1}{N_a}\sum_{j=1}^{N_a} e^{i(\vec{k}-\vec{k}_e)\vec{r}_j}\,|g_1, g_2, \ldots e_j \ldots g_{N_a}\rangle \rightarrow |ggg\ldots\rangle$$

-fluorescence intensity gives population of

$$|\Psi_{molk\uparrow}\rangle$$

-measurement is of QND type
-if a photon is detected molecules are projected to $|\Psi_{molk\uparrow}\rangle$ - state preparation

# Indirect molecular interactions mediated by Rydberg atoms

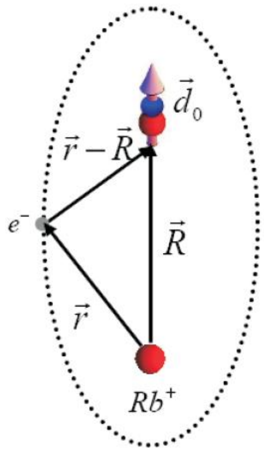



-polar molecules trapped in deep optical lattice (no tunneling)

- Rydberg superatom trapped in a shallow optical lattice/periodic trap array – strong tunneling

- Rydberg atom simultaneously interacts with all molecules: mediates molecular interaction

Direct polar molecule – Rydberg atom charge-dipole interaction

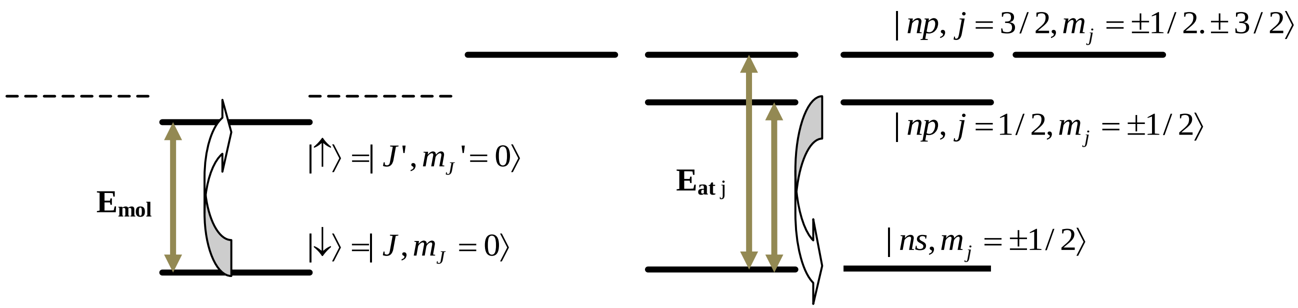



$$V_{ch-dip} = \frac{e\vec{d}_0\vec{R}}{R^3} - \frac{e\vec{d}_0(\vec{R}-\vec{r})}{|\vec{R}-\vec{r}|^3} \approx \frac{\vec{d}_0\vec{d}_{Rydb} - 3(\vec{d}_0,\vec{R})(\vec{d}_{Rydb},\vec{R})}{R^3}$$

Indirect molecular interaction

$$V_{mol} \sim \sum_f \frac{|V_{ch-dip,if}|^2}{E_i - E_f}$$

# Rydberg atom-single molecule




- molecular spin-1/2: $|\downarrow\rangle=|J=0,m_J=0\rangle$, $|\uparrow\rangle=|J=1,m_J=0\rangle$ rotational states

- $|J=0,m_J=0\rangle \leftrightarrow |J=1,m_J=0\rangle$ transition is near resonant with $np_j$ – ns Rydberg transition

- atom-molecule system can oscillate between near resonant $|ns,\uparrow\rangle \leftrightarrow |np_j,\downarrow\rangle$ states: atom-molecule Forster resonance

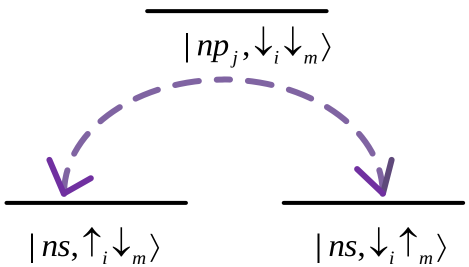
- if molecular states have induced dipole moments, transitions $|ns,\uparrow\rangle \leftrightarrow |np_j,\uparrow\rangle$, $|ns,\downarrow\rangle \leftrightarrow |np_j,\downarrow\rangle$ also possible

$$V_{ch-dip} = \sum_{m,i,f} |i\rangle_m V_{if}^m \langle f|_m$$

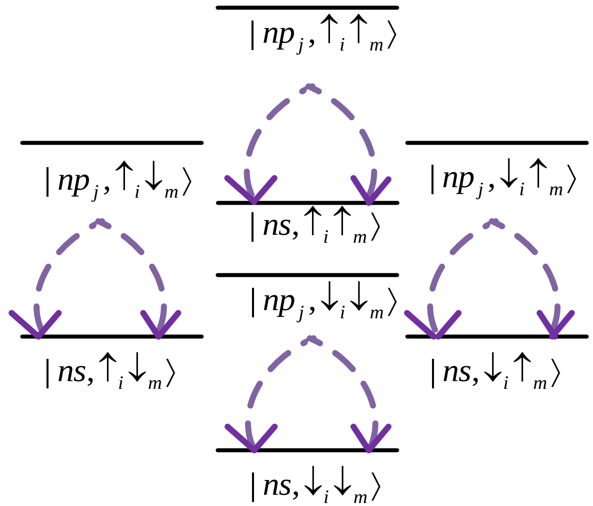
$$|i\rangle_m = |ns, m_j = 1/2, \vec{k}_0; \alpha_m\rangle, \quad |f\rangle_m = |np_{j'}, m_{j'}, \vec{k}; \beta_m\rangle, \quad \alpha_m, \beta_m = \uparrow, \downarrow$$

- $\vec{k}_0, \vec{k}$ denote quasimomenta of the initial and final atomic motional state, described by Bloch functions $\Psi_{\vec{k}}^{(n)} = u_{\vec{k}}^{(n)}(\vec{x}) e^{i\vec{k}\vec{x}}$

# Rydberg atom-two molecules




molecular spin-exchange interaction




molecular $s_z s_z$ interaction

- molecular spin-spin interaction: Schrieffer-Wolff transformation

$$H = H_{at} + H_{mol} + V_{\text{int}} = H_0 + V_{\text{int}}$$

$$\tilde{H} = e^S H e^{-S} = H + [S,H] + \frac{[S,[S,H]]}{2} + O(S^3)$$

$$[S,H_0] = -V_{\text{int}} \quad \Rightarrow$$

$$\tilde{H} = H_0 + \frac{[S,V_{\text{int}}]}{2} + O(V_{\text{int}}^3)$$

$$S \sim V_{\text{int}}$$

# Effective molecular spin-spin interaction

For an atom initially in $ns$ state the effective interaction is projected

$$V_{eff} = P_{ns}\frac{[S,V_{\text{int}}]}{2}P_{ns}, \quad P_{ns} = |ns, m_j, \vec{k}_0\rangle\langle ns, m_j, \vec{k}_0|$$

$$V_{eff} = |ns, m_j, \vec{k}_0\rangle\langle ns, m_j, \vec{k}_0| \left( \sum_{\substack{i,m,\\ \alpha,\beta,\gamma,\delta=\uparrow,\downarrow}} K^{im}_{\alpha\beta,\gamma\delta} |\alpha_i \beta_m\rangle\langle\gamma_i \delta_m| \right)$$

Assuming $|V_{\text{int}}| << E_{\uparrow} - E_{\downarrow}, E_{np} - E_{ns}$ and keeping only resonant terms

$$V_{eff} \approx |ns, m_j, \vec{k}_0\rangle\langle ns, m_j, \vec{k}_0| \left( \sum_{i,m} J^{zz}_{im} S^z_i S^z_m + \frac{J^{\perp}_{im}}{2}\left(S^+_i S^-_m + S^-_i S^+_m\right) \right) + |ns, m_j, \vec{k}_0\rangle\langle ns, m_j, \vec{k}_0| \sum_i b^z_i S^z_i$$

$$S^+_i = |\uparrow\rangle_i\langle\downarrow|, \quad S^-_i = \left(S^+_i\right)^+$$

$$S^z_i = \frac{1}{2}\left(|\uparrow\rangle_i\langle\uparrow| - |\downarrow\rangle_i\langle\downarrow|\right)$$

-XXZ interaction in the presence of magnetic field

# Calculation of interaction coefficients

$$J_{im}^{\perp} \approx -\sum_{\substack{j'=1/2,3/2\\ m_{j'}=\pm 1/2,\pm 3/2}} \sum_{\vec{k}} \frac{2V^{i}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\left(V^{m}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\right)^*}{E_{np_{j'}} - E_{ns} + \varepsilon^{np}_{kin}(\vec{k}) - \varepsilon^{ns}_{kin}(\vec{k}_0) - \left(E_{\uparrow} - E_{\downarrow}\right)}$$

$$J_{im}^{zz} \approx -\sum_{\substack{j'=1/2,3/2\\ m_{j'}=\pm 1/2,\pm 3/2}} \sum_{\vec{k}} \frac{\left(V^{i}_{ns,m_j,\vec{k}_0,\downarrow;np_{j'},m_{j'},\vec{k},\downarrow} - V^{i}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\uparrow}\right)\left(\left(V^{m}_{ns,m_j,\vec{k}_0,\downarrow;np_{j'},m_{j'},\vec{k},\downarrow}\right)^* - \left(V^{m}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\uparrow}\right)^*\right)}{E_{np_{j'}} - E_{ns} + \varepsilon^{np}_{kin}(\vec{k}) - \varepsilon^{ns}_{kin}(\vec{k}_0)}$$

$$-\sum_{\substack{j'=1/2,3/2\\ m_{j'}=\pm 1/2,\pm 3/2}} \sum_{\vec{k}} \frac{\left(V^{m}_{ns,m_j,\vec{k}_0,\downarrow;np_{j'},m_{j'},\vec{k},\downarrow} - V^{m}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\uparrow}\right)\left(\left(V^{i}_{ns,m_j,\vec{k}_0,\downarrow;np_{j'},m_{j'},\vec{k},\downarrow}\right)^* - \left(V^{i}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\uparrow}\right)^*\right)}{E_{np_{j'}} - E_{ns} + \varepsilon^{np}_{kin}(\vec{k}) - \varepsilon^{ns}_{kin}(\vec{k}_0)}$$

Interaction matrix elements, using the form of Bloch functions $|\vec{k}_0\rangle, |\vec{k}\rangle$

$$V^{m}_{ns,m_j,\vec{k}_0,\alpha;np_{j'},m_{j'},\vec{k},\beta} = \langle ns, m_j, \vec{k}_0 | \langle \alpha |_m V_{\text{int}} | \beta \rangle_m | np_{j'}, m_{j'}, \vec{k} \rangle = c^{m}_{ns,m_j,\vec{k}_0,\alpha;np_{j'},m_{j'},\vec{k},\beta} e^{i(\vec{k}-\vec{k}_0)\vec{x}_m}$$

# Calculation of interaction coefficients (cont'd)

$$J_{im}^{\perp} \approx -\sum_{\substack{j'=1/2,3/2 \\ m_{j'}=\pm 1/2,\pm 3/2}} \sum_{\vec{k}} \frac{2c^{i}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\left(c^{m}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\right)^{*} e^{i(\vec{k}-\vec{k}_0)(\vec{x}_i-\vec{x}_m)}}{E_{np_{j'}} - E_{ns} + \varepsilon^{np}_{kin}(\vec{k}) - \varepsilon^{ns}_{kin}(\vec{k}_0) - (E_{\uparrow} - E_{\downarrow})}$$

Assuming $c^{i}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}$ to weakly depend on k and $|\Delta E| = |E_{np_{j'}} - E_{ns} - (E_{\uparrow} - E_{\downarrow})| >> \varepsilon^{np}_{kin}, \varepsilon^{ns}_{kin}$

$$J_{im}^{\perp} \approx -\sum_{\substack{j'=1/2,3/2 \\ m_{j'}=\pm 1/2,\pm 3/2}} \frac{2c^{i}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\left(c^{m}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\right)^{*}}{\Delta E} \sum_{\vec{k}} e^{i(\vec{k}-\vec{k}_0)(\vec{x}_i-\vec{x}_m)}$$

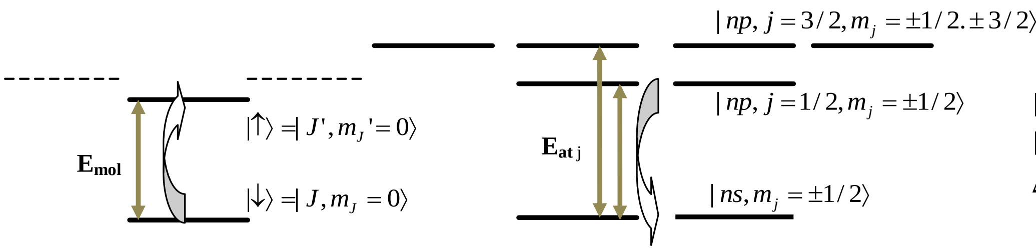
results in periodical dependence of interaction on spin-spin distance similar to RKKY interaction

E.g. in 1D case, assuming BEC initial atomic state with $\vec{k}_0 = 0$

$$J_{im}^{\perp} \sim \int_{-k_{BZ}}^{k_{BZ}} dk e^{ik(x_i - x_m)} = \frac{\sin(k_{BZ}(x_i - x_m))}{x_i - x_m}$$

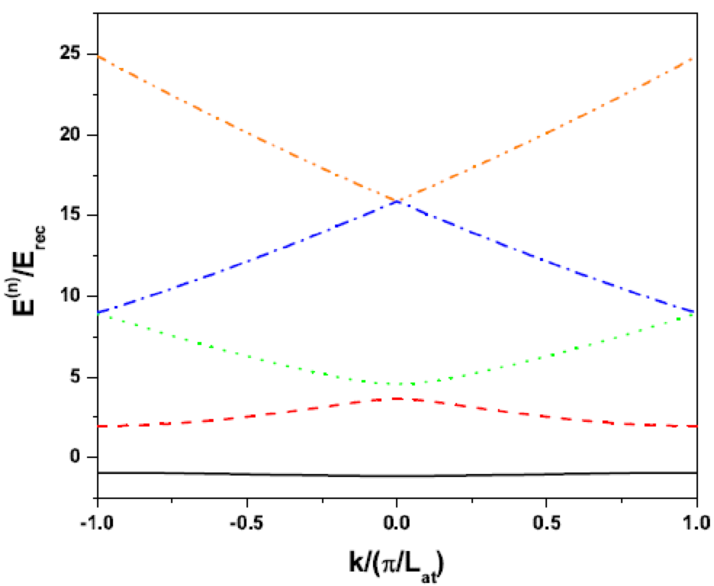
$k_{BZ} = \dfrac{\pi}{L^{latt}_{at}}$ - wavevectors are counted in the 1st Brillouin zone

# Numerical results for LiNa + Rb(n=55)




LiNa: $E_\uparrow - E_\downarrow = 0.38\ cm^{-1} = 22.784$ GHz
Rb: $E_{55p3/2} - E_{55s} = 22.759$ GHz
$\Delta E = E_{55p3/2} - E_{55s} - (E_\uparrow - E_\downarrow) = -25$ MHz

1D case
Atomic optical lattice: $V(x)=V_0\cos^2 K_{at}x$, $V_0=-6E_{rec}$, $K_{at}=\pi/L_{at}$, $L_{at}=1.5\ \mu m$
Molecular optical lattice: $L_{mol}=500$ nm, atomic-molecular lattice distance $\rho=400$ nm




Atomic Bloch energies and states

1D Schrodinger equation for Bloch wavefunction:

$$-\frac{\hbar^2}{2M_{at}}\frac{d^2\Psi_k^{(n)}}{dx^2} + V_0\cos^2(K_{at}x)\Psi_k^{(n)} = E^{(n)}\Psi_k^{(n)}$$

$$\Psi_k^{(n)} = u_k^{(n)} e^{ikx}, \quad k = \frac{2\pi Q}{L_{at}N_{at}}, \quad Q = -\frac{N_{at}}{2},\dots,\frac{N_{at}}{2}$$

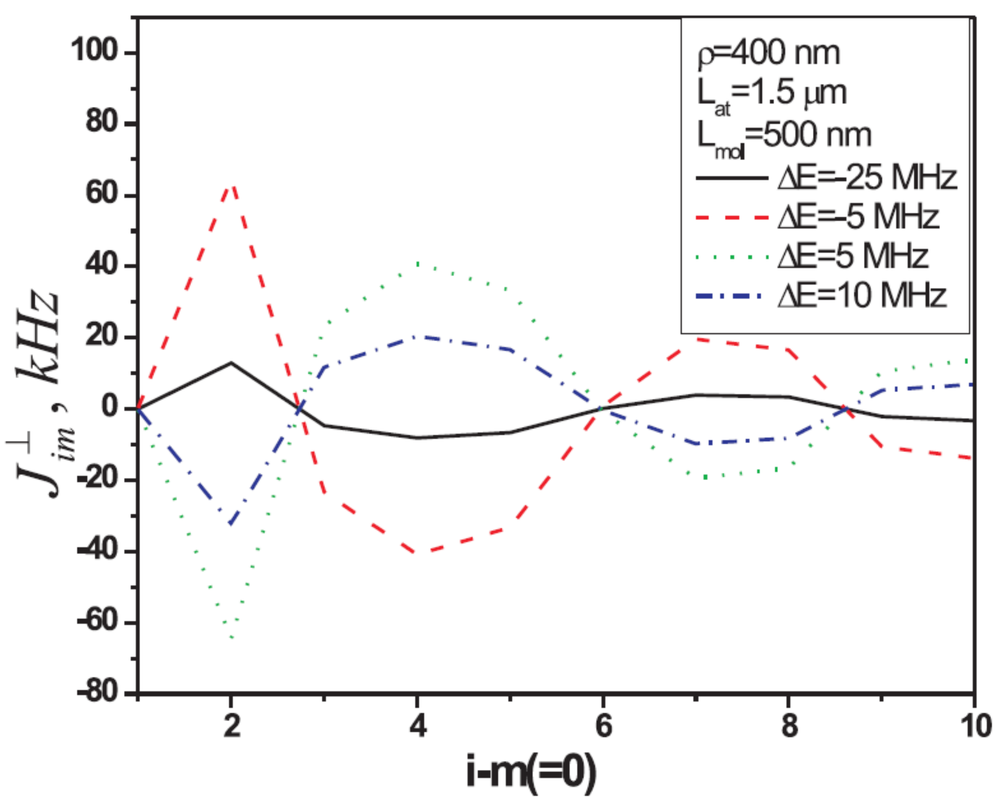
$$u_k^{(n)} = \sum_{s=-S_{\max}}^{S_{\max}} c_s^{(n)}(k) e^{2isK_{at}x}$$

$$\left(\frac{Q}{N_{at}}+s\right)^2 c_s^{(n)} + \frac{V_0}{16E_{rec}}\left(c_{s-1}^{(n)} + c_{s+1}^{(n)}\right) = \frac{E^{(n)} - V_0/2}{4E_{rec}} c_s^{(n)}$$

# Numerical results for LiNa + Rb(n=55)(cont'd)

$$J_{im}^{\perp} \approx -\sum_{\substack{j'=1/2,3/2\\ m_{j'}=\pm 1/2,\pm 3/2}} \sum_{\vec{k}} \frac{2V^{i}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\left(V^{m}_{ns,m_j,\vec{k}_0,\uparrow;np_{j'},m_{j'},\vec{k},\downarrow}\right)^{*}}{\Delta E + \varepsilon^{np}_{kin}(\vec{k}) - \varepsilon^{ns}_{kin}(\vec{k}_0)}$$

$$V^{m}_{ns,m_j,\vec{k}_0,\alpha;np_{j'},m_{j'},\vec{k},\beta} = \langle ns, m_j, \vec{k}_0 \,|\, \langle \alpha |_m \, V_{\text{int}} \,|\, \beta \rangle_m \,|\, np_{j'}, m_{j'}, \vec{k} \rangle = c^{m}_{ns,m_j,\vec{k}_0,\alpha;np_{j'},m_{j'},\vec{k},\beta} e^{i(\vec{k}-\vec{k}_0)\vec{x}_m}$$




-sum over 10 lowest Bloch bands

- periodic sign variation with interspin distance similar to RKKY interaction

- combined with random occupation of molecular lattice can lead to spin glass behaviour

- $J^{zz}_{im} \sim J^{\perp}_{im}\Delta E/(E_{np_j} - E_{ns}) \sim 5\cdot 10^{-4} J^{\perp}_{im}$ : highly anisotropic XXZ model

# Conclusions

- We analyzed a hybrid system of polar molecules and Rydberg atoms interacting via charge-dipole interaction. Energies of dressed atom-molecule states were calculated in a single molecule-single atom system, and a system of a molecular array+Rydberg superatom

- Energies of dressed states change differently for different molecular rotational states, which can be used for selective excitation of the atom to a Rydberg state depending on the rotational state

- Selective excitation of an atom interacting with polar molecule(s) to a Rydberg state, followed by detection of atomic fluorescence can be used for non-destructive readout of rotational states in the spirit of quantum logic spectroscopy

- Rydberg atom-polar molecule interaction can be used to realize indirect molecular spin interactions with sign periodically varying with interspin distance, similar to RKKY interaction between magnetic impurities mediated by conduction electrons

# Матричные элементы Гамильтониана заряд-дипольного взаимодействия

$$\left(V_{ch-dip}\right)_{nlmJm_J,n'l'm'J'm_J'} = \langle J,m_J\,|\,e\vec{d}\,|\,J',m_J'\rangle\frac{\vec{R}}{R^3}\delta_{n,n'}\delta_{l,l'}\delta_{m,m'} - \langle J,m_J\,|\,e\vec{d}\,|\,J',m_J'\rangle\langle nlm\,|\frac{\vec{R}-\vec{r}}{|\vec{R}-\vec{r}\,|^3}|\,n'l'm'\rangle$$

считаем для простоты $\vec{R} = \mathrm{R}\vec{\mathrm{e}}_z, \quad \vec{r} = \left(r\sin\theta\cos\varphi, r\sin\theta\sin\varphi, r\cos\theta\right)$

$$V_{ch-dip} = \frac{ed_z}{R^2} - \frac{ed_z(R-r\cos\theta)}{\left(R^2+r^2-2rR\cos\theta\right)^{3/2}} + \frac{ed_x r\sin\theta\cos\varphi + ed_y r\sin\theta\sin\varphi}{\left(R^2+r^2-2rR\cos\theta\right)^{3/2}}$$

1) $$\left(V_{ch-dip}^{core}\right)_{nlmJm_J,n'l'm'J'm_J'} = \frac{ed_z^{J,m_J;J',m_J'}}{R^2}\delta_{n.n'}\delta_{l,l'}\delta_{m,m'}\delta_{J\pm1,J'}\delta_{m_J,m_J'}$$

2) $$V_{ch-dip}^{I} = -\frac{ed_z(R-r\cos\theta)}{(R^2+r^2-2rR\cos\theta)^{3/2}} = ed_z\frac{d}{dR}\frac{1}{(R^2+r^2-2rR\cos\theta)^{1/2}} =$$

$$= ed_z\frac{d}{dR}\left\{\begin{array}{ll}\sum_{l''=0}^{\infty}\sqrt{\frac{4\pi}{2l''+1}}Y_{l''0}(\theta,\varphi)\frac{r^{l''}}{R^{l''+1}} & for\ r<R\\ \sum_{l''=0}^{\infty}\sqrt{\frac{4\pi}{2l''+1}}Y_{l''0}(\theta,\varphi)\frac{R^{l''}}{r^{l''+1}} & for\ r>R\end{array}\right. =$$

$$= ed_z\left\{\begin{array}{ll}-\sum_{l''=0}^{\infty}(l''+1)\sqrt{\frac{4\pi}{2l''+1}}Y_{l''0}(\theta,\varphi)\frac{r^{l''}}{R^{l''+2}} & for\ r<R\\ \sum_{l''=0}^{\infty}l''\sqrt{\frac{4\pi}{2l''+1}}Y_{l''0}(\theta,\varphi)\frac{R^{l''-1}}{r^{l''+1}} & for\ r>R\end{array}\right.$$

3) $$V_{ch-dip}^{II}=\frac{d_x r\sin\theta\cos\varphi+d_y r\sin\theta\sin\varphi}{(R^2+r^2-2rR\cos\theta)^{3/2}}=\frac{r}{2}\sin\theta\frac{(d_x-id_y)e^{i\varphi}+(d_x+id_y)e^{-i\varphi}}{(R^2+r^2-2rR\cos\theta)^{3/2}}=$$

$$=-\frac{1}{2R}\left((d_x-id_y)e^{i\varphi}+(d_x+id_y)e^{-i\varphi}\right)\frac{d}{d\theta}\frac{1}{(R^2+r^2-2rR\cos\theta)^{1/2}}$$

---

$$\frac{1}{\left(R^2+r^2-2rR\cos\theta\right)^{1/2}}=\begin{cases}\sum_{l''=0}^{\infty}P_{l''}(\cos\theta)\frac{r^{l''}}{R^{l''+1}} & for\ r<R\\ \sum_{l''=0}^{\infty}P_{l''}(\cos\theta)\frac{R^{l''}}{r^{l''+1}} & for\ r>R\end{cases}$$

$$\frac{d}{d\theta}P_{l''}(\cos\theta)e^{\pm i\varphi}=\begin{cases}P_{l''}^{1}(\cos\theta)e^{i\varphi}=\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}Y_{l''}^{1}(\theta,\varphi)\\ -l''(l''+1)P_{l''}^{-1}(\cos\theta)e^{-i\varphi}=-\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}Y_{l''}^{-1}(\theta,\varphi)\end{cases}$$

---

$$V_{ch-dip}^{II}=\frac{d_x r\sin\theta\cos\varphi+d_y r\sin\theta\sin\varphi}{(R^2+r^2-2rR\cos\theta)^{3/2}}=$$

$$=-\frac{d_x-id_y}{2R}\begin{cases}\sum_{l''=0}^{\infty}\frac{r^{l''}}{R^{l''+1}}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}Y_{l''}^{1}(\theta,\varphi) & for\ r<R\\ \sum_{l''=0}^{\infty}\frac{R^{l''}}{r^{l''+1}}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}Y_{l''}^{1}(\theta,\varphi) & for\ r>R\end{cases}+$$

$$+\frac{d_x+id_y}{2R}\begin{cases}\sum_{l''=0}^{\infty}\frac{r^{l''}}{R^{l''+1}}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}Y_{l''}^{-1}(\theta,\varphi) & for\ r<R\\ \sum_{l''=0}^{\infty}\frac{R^{l''}}{r^{l''+1}}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}Y_{l''}^{-1}(\theta,\varphi) & for\ r>R\end{cases}$$

$$\langle J,m_J|\langle nlm|V_{ch-dip}^{I}|n'l'm'\rangle|J',m_J'\rangle =$$

$$=\delta_{m,m'}\delta_{J',J\pm1}\delta_{m_J,m_J'}(-\sum_{l''=0}^{\infty}(l''+1)\sqrt{\frac{4\pi}{2l''+1}}\frac{1}{R^{l''+2}}\int_0^R r^{l''+2}R_{nl}(r)R_{n'l'}(r)dr\int_0^{\pi}Y_l^{m*}Y_{l''}^{0}Y_{l'}^{m}\sin\theta d\theta +$$

$$+\sum_{l''=0}^{\infty}l''\sqrt{\frac{4\pi}{2l''+1}}R^{l''-1}\int_0^R\frac{1}{r^{l''-1}}R_{nl}(r)R_{n'l'}(r)dr\int_0^{\pi}Y_l^{m*}Y_{l''}^{0}Y_{l'}^{m}\sin\theta d\theta)ed_z^{J,m_J;J',m_J'}$$

---

$$\langle J,m_J|\langle nlm|V_{ch-dip}^{II}|n'l'm'\rangle|J',m_J'\rangle =$$

$$=-(\sum_{l''=0}^{\infty}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}\frac{1}{R^{l''+1}}\int_0^R r^{l''+2}R_{nl}(r)R_{n'l'}(r)dr\int_0^{2\pi}d\varphi\int_0^{\pi}Y_l^{m*}Y_{l''}^{1}Y_{l'}^{m'}\sin\theta d\theta +$$

$$+\sum_{l''=0}^{\infty}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}R^{l''}\int_0^R\frac{1}{r^{l''-1}}R_{nl}(r)R_{n'l'}(r)dr\int_0^{2\pi}d\varphi\int_0^{\pi}Y_l^{m*}Y_{l''}^{1}Y_{l'}^{m'}\sin\theta d\theta)\frac{e\langle J,m_J|d_x-id_y|J',m_J'\rangle}{2R}+$$

$$+(\sum_{l''=0}^{\infty}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}\frac{1}{R^{l''+1}}\int_0^R r^{l''+2}R_{nl}(r)R_{n'l'}(r)dr\int_0^{2\pi}d\varphi\int_0^{\pi}Y_l^{m*}Y_{l''}^{-1}Y_{l'}^{m'}\sin\theta d\theta +$$

$$+\sum_{l''=0}^{\infty}\sqrt{\frac{4\pi l''(l''+1)}{2l''+1}}R^{l''}\int_0^R\frac{1}{r^{l''-1}}R_{nl}(r)R_{n'l'}(r)dr\int_0^{2\pi}d\varphi\int_0^{\pi}Y_l^{m*}Y_{l''}^{-1}Y_{l'}^{m'}\sin\theta d\theta)\frac{e\langle J,m_J|d_x+id_y|J',m_J'\rangle}{2R}$$

---

$$d_z^{0,0;1,0}=d/\sqrt{3},\ d_z^{1,0;2,0}=2d/\sqrt{15},\ d_z^{1,\pm1;2,\pm1}=d/\sqrt{5},\ d_{\pm}^{0,0;1,\pm1}=-d/\sqrt{3},$$

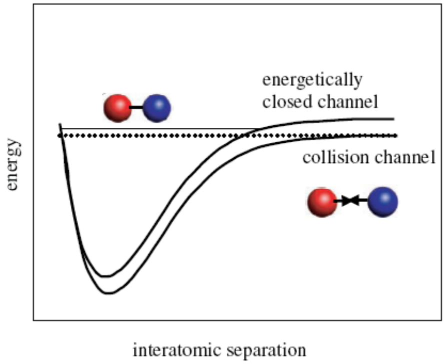
$$d_{\pm}^{1,0;2,\pm1}=-d/\sqrt{5},\ d_{\pm}^{1,\pm1;2,\pm2}=-d\sqrt{2}/\sqrt{5},\ d_{\pm}^{1,\pm1;2,0}=-d/\sqrt{15}$$

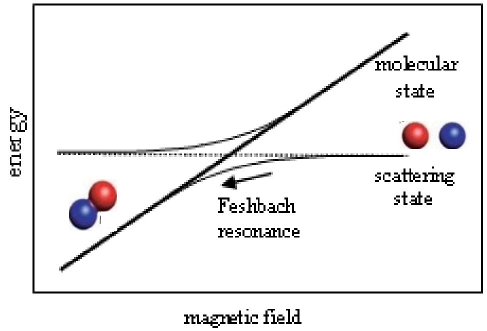
$$d_{\pm}^{J,m_J;J',m_J'}=\pm\langle J,m_J|d_x\mp id_y|J',m_J'\rangle/\sqrt{2}$$

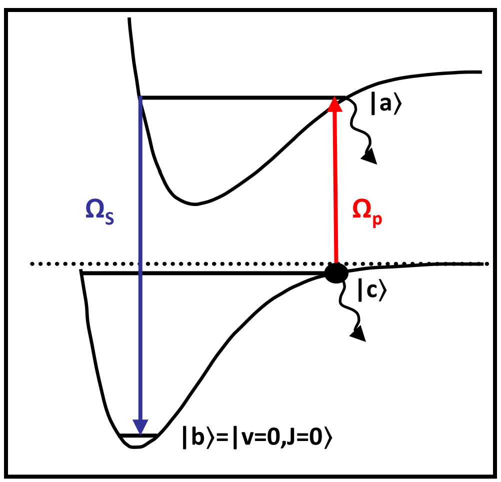
# Production of ultracold molecules

- direct laser cooling is not possible due to lack of closed transitions (vibrational transitions do not have strict selection rules)
- molecules are associated from ultracold atoms by

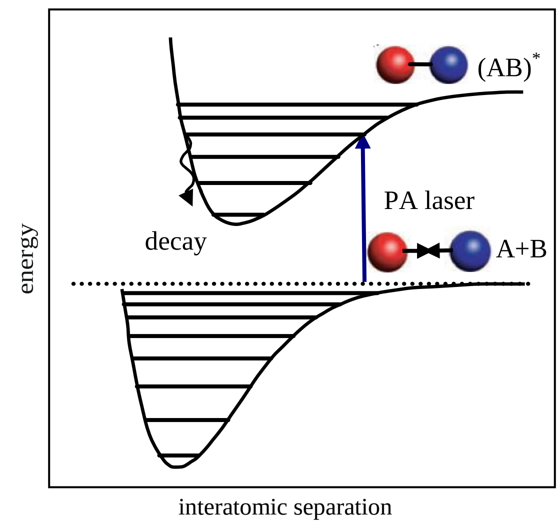
**Feshbach resonance+STIRAP** **Photoassociation**




D.Jin(JILA):KRb
W.Ketterle(MIT):NaLi
M.Zwierlein(MIT):NaK
H.C.Nagerl(Innsbruck):RbCs,KCs
R.Grimm(Innsbruck):RbSr
S.Cornish(Durham,UK):KCs




Jones, *et al.*, RMP **78**, 483 (2006)
D.DeMille(Yale): RbCs
M.Weidemuller(Heidelberg):LiCs
N.Bigelow(Rochester):NaCs

# Квантовая логическая спектроскопия

-основной спин/кубит не имеет состояний, позволяющих эффективное считывание, инициализацию, охлаждение
- используется взаимодействие основного кубита с пробным кубитом, имеющим удобные для управления оптическими полями состояния
- экспериментально продемонстрировано: ультрахолодные ионы (Кулоновское взаимодействие), N-V центры (сверхтонкое взаимодействие)
- предложения: ионы-полярные молекулы (заряд-дипольное взаимодействие), атомные смеси (Ридберговское взаимодействие)

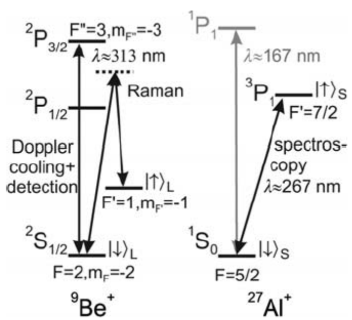



**Fig. 2.** Partial $^9Be^+$ and $^{27}Al^+$ energy level diagrams (not to scale). Shown are the relevant transitions for Doppler and Raman cooling on the $^9Be^+$ ion, the spectroscopy transition, and the difficult-to-reach Doppler cooling transition at 167 nm on the $^{27}Al^+$ ion.

Science **309**, 749 (2005)

состояние основного кубита когерентно переносится на пробный кубит используя общую колебательную моду ионов

$$\left(\alpha|\downarrow\rangle_S+\beta|\uparrow\rangle_S\right)|\downarrow\rangle_L|0\rangle_m\rightarrow\left(\alpha|\downarrow\rangle_S|0\rangle_m+\beta|\downarrow\rangle_S|1\rangle_m\right)|\downarrow\rangle_L\rightarrow$$
$$\rightarrow|\downarrow\rangle_S\left(\alpha|\downarrow\rangle_L|0\rangle_m+\beta|\uparrow\rangle_L|0\rangle_m\right)=|\downarrow\rangle_S|0\rangle_m\left(\alpha|\downarrow\rangle_L+\beta|\uparrow\rangle_L\right)$$

# Квантовые неразрушающие (QND) измерения

- измерения с использованием пробного кубита могут быть квантово неразрушающего QND типа

$\hat{H}(t)$ - Гамильтониан системы, включающий взаимодействие, необходимое для измерения

$\hat{O}_S$ - оператор измеряемой величины

$\left[\hat{H}(t), \hat{O}_S\right] = 0$ - условие QND измерения, процесс измерения не возмущает измеряемую величину

Критерии QND измерения для дискретных переменных (кубитов):

T.C.Ralph, et. al., PRA **73**, 012113 2006

$\{|\Psi_i\rangle, i = 1,...d\}$ - набор базисных состояний системы, d- размерность

$\hat{\rho}$ - матрица плотности системы до измерения

$p_i^{in} = \langle \Psi_i | \hat{\rho} | \Psi_i \rangle$ - распределение вероятностей состояний системы до измерения

$\hat{\rho}'$ - матрица плотности системы после измерения

$p_i^{out} = \langle \Psi_i | \hat{\rho}' | \Psi_i \rangle$ - распределение вероятностей состояний системы после измерения

$\hat{\rho}^m$ - матрица плотности измерителя после измерения

$p^m = \langle \Psi_i | \hat{\rho}_m | \Psi_i \rangle$ - распределение вероятностей состояний измерителя после измерения

1) Корреляция между результатами измерения и начальным состоянием

$$F_M = \left( \sum_i \sqrt{p_i^{in} p_i^m} \right)^2 = 1$$ - точность измерения

2) Измерение не должно возмущать измеряемую систему (для собственных состояний измеряемой величины)

$$F_{QND} = \left( \sum_i \sqrt{p_i^{in} p_i^{out}} \right)^2 = 1$$ - степень QND

3) Корреляция между состоянием измерителя и системы после измерения (измеритель в $|\Psi_i\rangle \Rightarrow \hat{\rho}' = |\Psi_i\rangle\langle\Psi_i|$)

$$F_{QSP} = \sum_i p_i^m p_{|i\rangle|i}^{out} = 1$$ - точность приготовления квантового состояния

$p_{|i\rangle|i}^{out}$ - вероятность системы после измерения быть в $|\Psi_i\rangle$ если измеритель в $|\Psi_i\rangle$

# QND измерение состояния основного кубита с помощью CNOT с участием пробного кубита

1) $\left(\alpha|0\rangle_S+\beta|1\rangle_S\right)|0\rangle_m$ - начальное состояние кубитов

2) $\alpha|0\rangle_S|0\rangle_m+\beta|1\rangle_S|1\rangle_m$ - CNOT на пробном кубите

3) измерение пробного кубита: $|0\rangle_m$ с вероятностью $|\alpha|^2$ - основной кубит проецируется в $|0\rangle_S$

$|1\rangle_m$ с вероятностью $|\beta|^2$ - основной кубит проецируется в $|1\rangle_S$

4) после измерения
$$\hat{\rho}'=|\alpha|^2|0\rangle_S\langle 0|+|\beta|^2|1\rangle_S\langle 1|$$
$$\hat{\rho}^m=|\alpha|^2|0\rangle_m\langle 0|+|\beta|^2|1\rangle_m\langle 1|$$

5) $p^{in}=p^{out}=p^m=\left\{|\alpha|^2,|\beta|^2\right\}$

$$F_M=\left(\sum_{i=1,2}\sqrt{p_i^{in}p_i^m}\right)^2=1,\quad F_{QND}=\left(\sum_{i=1,2}\sqrt{p_i^{in}p_i^{out}}\right)^2=1$$

$$F_{QSP}=\sum_{i=1,2}p_i^m p_{|i\rangle|i}^{out}=1$$