

# Control of optical transitions with magnetic fields in weakly bound molecules: Supplemental material

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## MIXED QUANTIZATION IN AN OPTICAL LATTICE

To model tensor light shifts of the 1D optical lattice we may use an effective potential of the form

$$V = -(1/2) \langle T^2(\boldsymbol{\alpha}) \cdot T^2(\mathbf{E}, \mathbf{E}) \rangle, \quad (4)$$

where the brackets denote a time average and the spherical tensor notation follows Ref. [1]. Using the Wigner-Eckart theorem, this potential evaluates to the matrix

$$V_{\{|2,m\rangle\}_z} = \frac{\alpha_2 |E|^2}{16} \begin{pmatrix} 2 & 0 & \sqrt{6} & 0 & 0 \\ 0 & -1 & 0 & 3 & 0 \\ \sqrt{6} & 0 & -2 & 0 & \sqrt{6} \\ 0 & 3 & 0 & -1 & 0 \\ 0 & 0 & \sqrt{6} & 0 & 2 \end{pmatrix} \quad (5)$$

for a  $\{|J = 2, m = 2\rangle, \dots, |J = 2, m = -2\rangle\}$  ground-state basis quantized along the applied field  $\mathbf{B} = B\hat{z}$ , a lattice electric field  $\mathbf{E} = E \cos(\omega t)\hat{y}$  propagating along  $\hat{x}$ , and a tensor polarizability  $\alpha_2(\omega) = (2/\sqrt{105}) \langle 2 || T^2(\boldsymbol{\alpha}) || 2 \rangle$  that depends on the reduced matrix element of the polarizability operator  $\boldsymbol{\alpha} = \boldsymbol{\alpha}(\omega)$  for the particular  $J = 2$  ground state. Because  $\mathbf{E} \perp \mathbf{B}$ , the matrix (5) contains off-diagonal elements from virtual two-photon  $\sigma$  transitions that couple pairs of sublevels  $|2, m_1\rangle$  and  $|2, m_2\rangle$  with  $|m_1 - m_2| = 2$ .

The potential (4) does not uniquely specify the ground eigenstates because only one eigenenergy is unique. This is because tensor light shifts are independent of the sign of  $m$  for quantization parallel to  $\mathbf{E}$ . Therefore, another interaction is required to break degeneracy and obtain unique eigenstates. For ground-state  $^{88}\text{Sr}_2$ , a likely candidate is the rotational Zeeman interaction,  $-g_r \mu_N \mathbf{J} \cdot \mathbf{B}$  [1]. We do not resolve this weak interaction, which is on the order of a nuclear magneton,  $\mu_N$ , so we will proceed by taking the limit  $g_r \rightarrow 0$ . In the basis of matrix (5), the five unique eigenstates  $|\gamma\rangle = \sum_m |J, m\rangle \langle J, m | \gamma \rangle$  obtained from this limit are given by the following sets of  $\langle J, m | \gamma \rangle$

coefficients  $\{\langle 2, 2 | \gamma \rangle, \dots, \langle 2, -2 | \gamma \rangle\}$ :

$$\{0, 1, 0, 1, 0\}/\sqrt{2}, \quad (6)$$

$$\{0, 1, 0, -1, 0\}/\sqrt{2}, \quad (7)$$

$$\{\sqrt{3}, 0, \sqrt{2}, 0, \sqrt{3}\}/\sqrt{8}, \quad (8)$$

$$\{1, 0, -\sqrt{6}, 0, 1\}/\sqrt{8}, \quad (9)$$

$$\text{and } \{1, 0, 0, 0, -1\}/\sqrt{2}. \quad (10)$$

As observed experimentally, these eigenstates consist of mixtures of sublevels with even or odd  $m$ .

To highlight the mixed nature of this quantization, note that while off-diagonal elements of the tensor light shift (5) are responsible for mixing if the quantization axis is chosen to be along  $\hat{z}$ , the mixing would instead come from off-diagonal rotational Zeeman shifts if the quantization axis were chosen to be along  $\hat{y}$ , where the potential (4) evaluates to  $V_{\{|2,m\rangle\}_y} = -\alpha_2 |E|^2 [3m^2 - J(J+1)]/[4J(2J-1)]$ . In this case, the eigenstates would be proportional to the symmetric and antisymmetric combinations ( $|2, m\rangle \pm |2, -m\rangle$ ). Both cases are equivalent, since the choice of quantization axis is artificial.

## MEASUREMENT OF TRANSITION STRENGTHS

We measure the strength of a particular  $\text{Sr}_2$  transition using a procedure similar to that in Ref. [2]. Initially we treat the case without mixed quantization, and return to evaluate its effects in the following section. First, we apply a laser pulse of duration  $\tau$  and power  $P$  along the lattice axis to drive the transition between a ground state  $|\gamma\rangle$  with quantum numbers  $(v, J, m)$  and an excited state  $|\mu\rangle$  with  $(v', J', m')$ . Afterwards, we measure a signal  $S_{J,m}$  proportional to the number  $n_{J,m}$  of remaining ground-state molecules with  $(v, J, m)$  using a second laser pulse to drive a transition to an excited state that decays to Sr atoms [3]. Both laser pulses are linearly polarized along  $\mathbf{B}$  and drive  $\Delta m = m' - m = 0$  transitions for the spectroscopic quantization axis defined by  $\mathbf{B}$ .

We assume the first pulse drives an open transition with sufficiently low power  $P$  that the absorption process is linear and any emission back to the initial state is negligible. During the first pulse, the number of  $(v, J, m)$

molecules evolves as

$$\frac{d}{dt}n_{J,m}(t) = -\Gamma_{m,m'}(\delta)n_{J,m}(t), \quad (11)$$

where  $\Gamma_{m,m'}(\delta)$  is a stimulated absorption rate per molecule. The quantity we wish to determine is then

$$Q = Q(m, m') = \frac{1}{P} \int \Gamma_{m,m'}(\delta) d\delta = \frac{|\Omega_{\gamma\mu}(B)|^2}{4P}, \quad (12)$$

which is a measure of the strength of the  $(m, m')$  component of the transition under study, expressed in terms of the Rabi frequency for this component introduced before Eq. (1). This result follows in the rate-equation regime where the evolution (11) applies, because we may use Eq. (31) of Ch. 4 and Eqs. (23,32) of Ch. 5 in Ref. [4] to write the integrand as  $\Gamma_{m,m'}(\delta) = |\Omega_{\gamma\mu}(B)|^2 / \{\Delta\omega_{12}[1 + (4\pi\delta)^2/\Delta\omega_{12}^2]\}$ , where  $\Delta\omega_{12}$  is the transition linewidth. We have confirmed the result (12) for optical magnetic-dipole transitions to subradiant  $1_g$  excited states [2]. Note that the factor of 1/4 depends on the convention used to define  $P$ , on  $\Delta m$ , and on the laser polarization. For  $J = 0$ , additional relations to convert  $Q$  to an Einstein  $B$  coefficient and an absorption oscillator strength are available in Ref. [2].

Experimentally, we record the signal  $S_{J,m}(\delta) \propto n_{J,m}(\tau) = n_{J,m}(0) \exp[-\Gamma_{J,m}(\delta)\tau]$  as a function of the laser-frequency detuning  $\delta$  from resonance, which is roughly a Lorentzian absorption dip with a constant background. The quantity  $Q$  of Eq. (12) is then given by

$$Q = -\frac{1}{\tau P} \int \ln \left[ \frac{S_{J,m}(\delta)}{S_{J,m}(\infty)} \right] d\delta = \frac{A}{\tau P}, \quad (13)$$

where  $A$  is the area of a Lorentzian fitted to a plot of  $\ln[S_{J,m}(\delta)]$  versus  $\delta$ , and the shorthand  $S_{J,m}(\infty) \propto n_{J,m}(t=0)$  is the signal far from resonance. No adjustments are made to account for the angular momentum quantum numbers of the initial or final states of the transition. Care is taken to experimentally verify that  $Q$  is independent of  $P$  and  $\tau$ , as expected from Eq. (12) because  $|\Omega_{\gamma\mu}(B)|^2 \propto P$ . In particular, we use low powers  $P$  to avoid power broadening and use pulse times  $\tau$  that are short enough to avoid additional loss processes (e.g. collisions) but long enough to avoid transform-limited broadening.

### CORRECTION FOR MIXED QUANTIZATION

For  $J = 2$  ground states, we follow the same procedure to measure transition strengths as with  $J = 0$ . However, because of mixed quantization  $m$  is no longer a good quantum number. Instead, the ground eigenstates  $|\gamma\rangle$  are superpositions of  $|J = 2, m\rangle$  sublevels quantized along  $\mathbf{B}$ , as derived above. As a result, the quantity we measure is

no longer the  $Q$  of Eqs. (12–13), but instead a modified quantity  $Q_{\text{mix}}$ . To account for mixed quantization and obtain the values of  $Q$  we report, we multiply these values by a correction factor  $R(m_1, m_2)$ , as in

$$Q(m_1, m') = R(m_1, m_2) Q_{\text{mix}}(m_1, m', m_2), \quad (14)$$

which we derive as follows. Here, the two quantum numbers  $m_1$  and  $m_2$  are determined by the first and second laser pulses, respectively, and defined for quantization along  $\mathbf{B}$ . Because of mixed quantization, these two quantum numbers need not be equal.

Let us denote the number of molecules in the state  $|\gamma\rangle$  by  $M_\gamma(t)$ . For typical applied fields, the Zeeman interaction of the  $J'$  excited state ensures that  $m'$  is a good quantum number and that each excited-state sublevel is spectroscopically resolvable. Therefore, we can tune the frequency and polarization of the first laser to only allow absorption by the ground-state sublevel  $|J = 2, m_1\rangle$  quantized along  $\mathbf{B}$  (specifically,  $m_1 = m'$ ). During the first laser pulse, the evolution (11) is then replaced by

$$\frac{d}{dt}M_\gamma(t) = -\Gamma_{m_1,m'}(\delta)f_\gamma(m_1)M_\gamma(t), \quad (15)$$

where the mixing coefficients

$$f_\gamma(m) = |\langle J = 2, m | \gamma \rangle|^2 \quad (16)$$

may be computed using expressions (6–10). The number of molecules remaining afterwards is then

$$M_\gamma(\tau) = M_\gamma(0) \exp[-f_\gamma(m_1)\Gamma_{m_1,m'}(\delta)\tau]. \quad (17)$$

Likewise, we can tune the second laser to only detect molecules in the sublevel  $|J = 2, m_2\rangle$  quantized along  $\mathbf{B}$ . The signal we measure is then

$$S_{2,m_2}(\delta) \propto \sum_\gamma f_\gamma(m_2)M_\gamma(\tau). \quad (18)$$

Far off resonance we will denote this by  $S_{2,m_2}(\infty) \propto \sum_\gamma f_\gamma(m_2)M_\gamma(0)$ .

The initial numbers  $M_\gamma(0)$  depend on the molecule creation procedure, which consists of photoassociation to an excited state ( $J' = 1, m' = 0$ ) followed by spontaneous decay to ground states with  $J = 0$  and 2 (symmetry forbids ground states with odd  $J$ ) [3, 5]. For our experiment, they are

$$M_\gamma(0) = \sum_m f_\gamma(m) n_{2,m}(0), \quad (19)$$

where the initial numbers  $n_{2,m}(0)$  for the unmixed sublevels  $|J = 2, m\rangle$  are given by the branching ratios

$$n_{2,m}(0) = n_2(0) \sum_{q=-1}^1 \left( C_{2,m;1,q}^{1,0} \right)^2, \quad (20)$$

expressed here with Clebsch-Gordon coefficients [6]. Note that there would be no initial molecules with  $m = \pm 2$  without mixed quantization because of the  $\Delta m = 0, \pm 1$  selection rule, as in  $n_{2,\pm 2}(0) = 0$ . However, because of mixed quantization there will be molecules in states  $|\gamma\rangle$  composed of sublevels with  $m = \pm 2$ , as shown by (6–10).

Following Eq. (13), the quantity we measure is again

$$Q_{\text{mix}} = -\frac{1}{\tau P} \int \ln \left[ \frac{S_{2,m}(\delta)}{S_{2,m}(\infty)} \right] d\delta = \frac{A}{\tau P}. \quad (21)$$

Using (14–21), the correction factor is then

$$R(m_1, m_2) = -\frac{\tau \int \Gamma_{m_1, m'}(\delta) d\delta}{\int \ln [S_{2, m_2}(\delta)/S_{2, m_2}(\infty)] d\delta}. \quad (22)$$

For the mixed quantization considered here, the signs of the arguments do not matter:  $R(m_1, m_2) = R(|m_1|, |m_2|)$  because  $f_\gamma(-m) = f_\gamma(m)$ . Note that in the absence of mixed quantization we would be free to choose  $f_\gamma(m) = \delta_{\gamma, m}$ , which recovers  $R(m, m) = 1$ .

We measured the data for transitions involving ( $J = 2, m_1 = \pm 1$ ) using  $m_2 = 1$  or  $-1$ . For this case, the ratio

$$\left. \frac{S_{2,1}(\delta)}{S_{2,1}(\infty)} \right|_{m_1=\pm 1} = \exp \left[ -\frac{1}{2} \Gamma_{\pm 1, m'}(\delta) \tau \right], \quad (23)$$

using Eq. (16) with Eqs. (6–10) and (18–20). The correction factor (22) is then  $R(1, 1) = 2$ , which we denoted previously by  $R(m_1) = R(\pm 1)$ .

For transitions involving ( $J = 2, m_1 = 0$ ) we used  $m_2 = 0$ . In this case the ratio is bi-exponential, but we can approximate it as

$$\begin{aligned} \left. \frac{S_{2,0}(\delta)}{S_{2,0}(\infty)} \right|_{m_1=0} &= \frac{1}{4} \exp \left[ -\frac{1}{4} \Gamma_{0, m'}(\delta) \tau \right] \\ &+ \frac{3}{4} \exp \left[ -\frac{3}{4} \Gamma_{0, m'}(\delta) \tau \right] \approx \exp \left[ -\frac{3}{4} \Gamma_{0, m'}(\delta) \tau \right]. \end{aligned} \quad (24)$$

This correction factor (22) is then  $R(0, 0) \approx 4/3$ , which we denoted previously by  $R(0)$ .

Finally, for transitions involving ( $J = 2, m_1 = \pm 2$ ) we used  $m_2 = 0$ . Again, we can approximate the ratio

$$\begin{aligned} \left. \frac{S_{2,0}(\delta)}{S_{2,0}(\infty)} \right|_{|m_1|=2} &= \frac{1}{4} \exp \left[ -\frac{3}{8} \Gamma_{\pm 2, m'}(\delta) \tau \right] \\ &+ \frac{3}{4} \exp \left[ -\frac{1}{8} \Gamma_{\pm 2, m'}(\delta) \tau \right] \approx \exp \left[ -\frac{1}{8} \Gamma_{\pm 2, m'}(\delta) \tau \right]. \end{aligned} \quad (25)$$

This correction factor (22) is then  $R(2, 0) \approx 8$ , which we denoted previously by  $R(\pm 2)$ .

## CALCULATION OF TRANSITION STRENGTHS

The theoretical values for transition strengths are obtained by calculating the square of the transition dipole

matrix element between a ground state  $|\gamma\rangle$  and an excited state  $|\mu(B)\rangle$  for an electric field linearly polarized along the magnetic field,  $|\langle \gamma | d_Z | \mu(B) \rangle|^2$ . Here,  $d_Z$  is the space-fixed transition dipole moment, which is expressed in terms of the molecule-fixed  $d_i$  components by means of Wigner rotational matrices. We assume that the starting level  $|\gamma\rangle$  belongs to the electronic ground state  $X^1\Sigma_g^+$ , has well defined  $J$ ,  $m$ , and  $\Omega$  quantum numbers ( $\Omega$  is the projection of the total electronic angular momentum along the internuclear axis), and do not include any corrections for the coupling of  $m$  sublevels from mixed quantization by the optical lattice. The target level  $|\mu(B)\rangle$  belongs to the  $0_u^+/1_u$  potentials near the  $^1S_0 + ^3P_1$  asymptote, and is a superposition of states with different  $J'$  and  $|\Omega'|$ , as in Eq. (1), such that only  $m$  remains a good quantum number in the presence of the magnetic field. The correct form of  $|\mu(B)\rangle$  as a function of the field strength  $B$  is obtained by solving the coupled-channel equations with the Zeeman Hamiltonian (which couples states with  $\Delta J' = 0, \pm 1$ ) and nonadiabatic Coriolis coupling (which couples  $1_u$  and  $0_u^+$  electronic states of the same  $J'$ ) included. The calculations include couplings between states from  $J' = 1$  up to  $J' = 6$ , which results in nine coupled channels in total (each even  $J'$  contributes a single  $1_u$  channel, and each odd  $J'$  contributes two ( $1_u$  and  $0_u^+$ ) channels). We assumed that the molecule-fixed electronic transition dipole moment between the  $X^1\Sigma_g^+$  and  $0_u^+/1_u$  states is independent of the internuclear separation  $R$  and equal to its asymptotic atomic limit,

$$\langle X^1\Sigma_g^+ | d_i | 1_u/0_u^+ \rangle \approx \sqrt{2} \langle ^1S_0 | d_i | ^3P_1 \rangle, \quad (26)$$

which is a good approximation for weakly bound molecules [2]. Just as with the experimental results, the reported values are normalized to the average zero-field strength of the chosen allowed transition in Fig. 2(c).

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