Predicting and verifying transition strengths from weakly bound molecules

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We investigated transition strengths from ultracold weakly bound $^{41}$K$^{87}$Rb molecules produced via the photoassociation of laser-cooled atoms. An accurate potential energy curve of the excited state ($3\,^1\Sigma^+$) was constructed by carrying out direct potential fit analysis of rotational spectra obtained via depletion spectroscopy. Vibrational energies and rotational constants extracted from the depletion spectra of $\nu'$ = 41–50 levels were combined with the results of the previous spectroscopic study and they were used for modifying an ab initio potential. An accuracy of 0.14% in vibrational level spacing and 0.3% in rotational constants was sufficient to predict the large observed variation in transition strengths among the vibrational levels. Our results show that transition strengths from weakly bound molecules are a good measure of the accuracy of an excited state potential.

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I. INTRODUCTION

Ultracold molecular gases are prominent candidates for realizing novel distinctive applications in physics and chemistry, including precision measurements, quantum computation, ultracold chemistry, and novel quantum phases [1,2]. Thus far the production of ultracold molecules in the vibrational ground state has been dependent on the optical transfer of weakly bound molecules formed via either photoassociation [3] or magnetoassociation [4]. Among the previously proposed methods [5–10], stimulated Raman adiabatic passage (STIRAP) [11–13] of weakly bound molecules is the most efficient method for preparing a molecular sample in a single quantum state.

The transfer of an excited state is used as an intermediate state. It is important to select an excited state having large transition strengths from both weakly bound and deeply bound levels. In general, the strengths of molecular transitions from low-vibrational levels are readily predicted on the basis of the Franck-Condon factors (FCFs) calculated from a potential energy curve or molecular constants [14]. The small number of nodes in the radial wave function for a low vibrational level indicates a low sensitivity to deviations in the radial direction, thereby enabling us to explain the intensity distribution over vibrational levels from molecular constants. However, it is difficult to predict transition strengths from weakly bound molecular levels because weakly bound levels have a large number of nodes in their wave functions; hence the FCFs are quite sensitive to the wave functions of excited states. In this article we show that transition strengths from weakly bound levels can be predicted on the basis of an accurate potential energy curve of the excited state, constructed via direct potential fit (DPF) analysis [15] of both vibrational energies and rotational constants.

We focus on the ($3\,^1\Sigma^+$) state of K$\text{Rb}$ in the range 11 400–11 800 cm$^{-1}$ with respect to the ground atomic threshold 4$^3\Sigma^+$ which was proposed as a potential candidate for the STIRAP transfer of weakly bound molecules to the rovibrational ground state [16] (see Fig. 1). An Rydberg-Klein-Rees (RKR) potential curve of the ($3\,^1\Sigma^+$) state was reported from the potential minimum up to 10 400 cm$^{-1}$ [17]. An ionization spectrum for $^{39}$K$^{87}$Rb obtained by using a pulsed laser and a depletion spectrum near 11 700 cm$^{-1}$ were provided in Ref. [16]. Recently we have realized the STIRAP transfer of weakly bound molecules ($\nu'$ = 91, $J'' = 0$ of $X\,^1\Sigma^+$) to the rovibrational ground state ($\nu'' = 0$, $J'' = 0$ of $X\,^1\Sigma^+$), mediated by the $\nu'$ = 41 level of the ($3\,^1\Sigma^+$) state [19].

In this article we report on the spectroscopic studies on the ($3\,^1\Sigma^+$) state carried out in the range 11 400–11 800 cm$^{-1}$, which reveals 10 vibrational levels ($\nu'$ = 41–50). We find that the width of the observed spectra is highly dependent on the vibrational levels of the ($3\,^1\Sigma^+$) state. By introducing an analytical representation for power broadening, we extract the transition strengths for each vibrational level. The other electronic states that correlate with the ($3\,^1\Sigma^+$) state via spin-orbit interaction are far away; hence we assume that most of the perturbations from these electronic states are negligible. Thus we can analyze the experimentally obtained spectra on the basis of a single potential curve. In addition to the previous spectroscopic work near the bottom of the potential [17], vibrational energies and rotational constants extracted from the spectra are used to construct an accurate potential via DPF analysis. Using the modified potential, it is possible to explain the variation in transition strengths among the vibrational levels of the ($3\,^1\Sigma^+$) state in terms of the FCFs.

II. DEPLETION SPECTROSCOPY

Previously our experimental setup for the spectroscopy of ultracold $^{41}$K$^{87}$Rb molecules was described in detail [19]. We provide a brief summary here. We start with a dual-species magneto-optical trap (MOT) of $1 \times 10^8$ $^{41}$K atoms and $2 \times 10^8$ $^{87}$Rb atoms. A compressed MOT (C-MOT) procedure is

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intermediate state for REMPI ionization was the \((4)\Sigma^+\) state. The spectroscopy of molecules in the \(v'' = 91\) level of \(X^1\Sigma^+\). The intermediate state for REMPI ionization was the \((4)\Sigma^+\) state.

applied for 40 ms to compress and cool the \(^{41}\text{K}\) and \(^{87}\text{Rb}\) atoms. The typical densities and temperatures of \(^{41}\text{K}\) and \(^{87}\text{Rb}\) at the end of C-MOT are \(2 \times 10^{11} \text{ cm}^{-3}\) and 400 \(\mu\text{K}\) and \(4 \times 10^{11} \text{ cm}^{-3}\) and 100 \(\mu\text{K}\) for \(^{87}\text{Rb}\), respectively. A photoassociation (PA) laser (wave number 12 570.13 cm\(^{-1}\), intensity \(1 \times 10^3 \text{ W/cm}^2\)) is applied for 10 ms at the end of the C-MOT process. The produced molecules are detected using microchannel plates (MCP) after they are ionized via resonance enhanced multiphoton ionization (REMPI) using a pulsed dye laser (wave number 16 543 cm\(^{-1}\), intensity \(3 \times 10^6 \text{ W/cm}^2\)).

Depletion spectra are obtained by monitoring ion counts in the \(v'' = 91\) level of \(X^1\Sigma^+\), while a CW Ti:sapphire laser (Sirah Matisse TX, intensity 50 W/cm\(^2\), beam waist 350 \(\mu\text{m}\)) is continuously applied and scanned. In the present study we analyze spectra for the \(v'' = 91\), \(J'' = 2\) level, whose binding energy with respect to the atomic threshold \(F_{\text{K}} = 1 + F_8\text{Rb} = 1\) was measured as \(-12.454(1) \text{ cm}^{-1}\). The frequency of the Ti:sapphire laser is monitored using a Fabry-Perot cavity which is locked to the \(^{87}\text{Rb}\) \(D_2\) line. The cavity transmission signal is used to calibrate the variation in the scanning speed and to measure the relative frequency of the laser with respect to the cavity transmission peak with a precision of 3 MHz. The absolute frequency is measured using a commercial wave meter (Wavelength WS-7, accuracy 60 MHz). The accuracy of measurements for rotational constants is limited by the spectral width, which is of the order of 100 MHz, whereas that for vibrational energies is limited by both the spectral width and the wave meter.

Figure 2 shows a depletion spectrum for the \(v' = 41\) level of the \((3)\Sigma^+\) state, which was used as an intermediate state for the STIRAP transfer from the \(v'' = 91\), \(J'' = 0\) level of \(X^1\Sigma^+\) to the \(v'' = 0\), \(J'' = 0\) level [18]. We can extract transition strengths as well as rotational constants for both ground and excited states from the spectra. In the Appendix we show that an approximate representation of the full width at half maximum (FWHM) of a depletion spectrum is given by

\[2\Delta \sim 0.79\Omega\sqrt{\Gamma\tau},\]  

where \(\Omega\) is the Rabi frequency, \(\Gamma\) is the natural width of the excited state, and \(\tau\) is the interaction time. Roughly speaking, the width increases not only with the light intensity but also with the interaction time. In our case \(\tau\) was estimated as a few milliseconds on the basis of the temperature of the molecules and the beam diameter of the depletion laser. The natural width of the \((3)\Sigma^+\) state has not been precisely

![Image](https://via.placeholder.com/150)

**FIG. 1.** (Color) Relevant potential energy curves of KRb. Weakly bound molecules were produced by the photoassociation of laser-cooled \(^{41}\text{K}\) and \(^{87}\text{Rb}\) atoms. The \((3)\Sigma^+\) state was investigated via the spectroscopy of molecules in the \(v'' = 91\) level of \(X^1\Sigma^+\). The produced molecules are detected using microchannel plates (MCP) after they are ionized via resonance enhanced multiphoton ionization (REMPI) using a pulsed dye laser (wave number 16 543 cm\(^{-1}\), intensity \(3 \times 10^6 \text{ W/cm}^2\)).

![Image](https://via.placeholder.com/150)

**FIG. 2.** (Color online) Depletion spectrum of \(v' = 41\) level of \((3)\Sigma^+\) taken with molecules in \(v'' = 91\) level of \(X^1\Sigma^+\). The spectrum gives rotational constants of both ground and excited states. In addition, the width of the spectrum enables us to estimate the transition strength.

![Image](https://via.placeholder.com/150)

**FIG. 3.** (Color online) Comparison between calculated and observed rotational constants. Calculated rotational constants of five molecular states in this range are plotted against energy levels from the ground atomic threshold. The observed rotational constants are in good agreement with those obtained from the \(ab\ initio\) potential of the \((3)\Sigma^+\) state. There is a systematic deviation of \(-1\%\) between the observed values and \(ab\ initio\) values, which indicates the inaccuracy of the \(ab\ initio\) potential.
TABLE I. Parameters for EMO potential obtained via DPF analysis of the observed spectra. The units are cm⁻¹ for $V_{\text{min}}$ and $D_v$, Å for $R_v$, and Å⁻¹ for $\phi_i$.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
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<td>$V_{\text{min}}$</td>
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<td>$\phi_5$</td>
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<td>$D_v$</td>
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<td>$\phi_1$</td>
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<td>0.40684012</td>
<td>$\phi_{11}$</td>
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<tr>
<td>$\phi_3$</td>
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<td>$\phi_{12}$</td>
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<td>$\phi_4$</td>
<td>−0.47634930</td>
<td>$\phi_{13}$</td>
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</table>

known, but it was estimated as $2\pi \times 300$ kHz from an \textit{ab initio} calculation [20]. In the following discussion we derive the Rabi frequency from the observed spectrum by assuming $\tau = 2$ ms, $\Gamma = 2\pi \times 300$ kHz, and $\sqrt{\Gamma \tau} = 60$.

III. ANALYSIS

Although the \textit{ab initio} potential of Ref. [21] enabled us to identify the symmetry of the observed spectra without any ambiguity, it gave rotational constants that exceeded the experimental ones by approximately 1% (Fig. 3). There are two reasons for this deviation. First, the potential minimum exceeds the minimum of the RKR curve by 97 cm⁻¹. Second, the outer turning point is at a shorter internuclear separation than the RKR potential. We find that the variation in the observed transition strengths cannot be understood with the \textit{ab initio} potential. The observations cannot be attributed to the small change in the vibrational quantum number, which corresponds to the energy difference between the minima of the RKR and the \textit{ab initio} potentials. These facts indicate that the \textit{ab initio} potential is not sufficiently accurate. In order to obtain an accurate potential, we carried out a DPF analysis, whereby a potential is iteratively modified until its eigenvalues coincide with those determined from the experimental spectra [15]. With the aid of the phiFIT program code [22], we first construct an analytical extended Morse oscillator (EMO) potential of the (3) $^1\Sigma^+$ state on the basis of (1) the RKR curve, (2) a few points from the inner curve of the \textit{ab initio} potential, and (3) a few points around 12500 cm⁻¹ from the outer curve of the \textit{ab initio} potential. The potential curve of the ground state $X^1\Sigma^+$ is also required for the calculations. We use an EMO potential fitted to an accurate, experimentally determined potential [23]. Then the analytical potential of the (3) $^1\Sigma^+$ state is modified to reproduce our data by using the DPotFit program code [24]. A good convergence is achieved when we modify the potential significantly by manually moving the points from the \textit{ab initio} potential. This procedure is repeated until the eigenvalues of the potential are within 0.05 cm⁻¹ of the observed levels, that is, only 0.14% of the vibrational level spacing. The remaining deviations are presumably due to the incomplete analytical function used to represent the potential, although perturbations from other states can also be the origin of the deviations for some levels. Table I lists the final potential parameters. These parameters are used to represent the potential energy curve in the following form:

$$V(R) = V_{\text{min}} + D_v(1 - e^{-\phi(R)(R-R_v)})^2,$$

$$\phi(R) = \sum_{i=0}^{12} \phi_i y(R,R_v)^i,$$

$$y(R,R_v) = \frac{R^3 - R_v^3}{R^3 + R_v^3}.$$  

Figure 4 shows the RKR potential, \textit{ab initio} potential, and modified potential. With the new potential, rotational constants are reproduced within 0.3% of the observed values (Table II). We excluded the $\nu' = 44$ level from the analysis because the characteristics of this level are anomalous; a much larger transition strength, a much smaller rotational constant, and a much larger deviation in a vibrational energy were observed at this level. These features indicate that this level is coupled to the (2) $^1\Sigma^+$ state which was observed at $\sim 1 \text{ cm}^{-1}$ above the $\nu' = 44$ level.

TABLE II. Comparison between observed, \textit{ab initio}, and modified values for vibrational energies (E) and rotational constants (B). Vibrational energies are energy levels of the $J' = 1$ level. Error bars in vibrational energies are $2 \times 10^{-3} \text{ cm}^{-1}$, whereas those in rotational constants are $5 \times 10^{-3} \text{ cm}^{-1}$. Observed vibrational energies are reproduced within $5 \times 10^{-2} \text{ cm}^{-1}$, whereas observed rotational constants are reproduced within $7 \times 10^{-5} \text{ cm}^{-1}$. The vibrational numbering for \textit{ab initio} values is shifted by 2 because the potential minimum lies below that of the correct potential.

<table>
<thead>
<tr>
<th>$\nu'$</th>
<th>Obs.</th>
<th>\textit{ab initio}–Obs.</th>
<th>Mod.–Obs. (units of $10^{-3}$)</th>
<th>Obs.</th>
<th>\textit{ab initio}–Obs. (units of $10^{-5}$)</th>
<th>Mod.–Obs. (units of $10^{-5}$)</th>
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<td>2.5</td>
<td>2.067</td>
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<td>111466.644</td>
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<td>29.9</td>
<td>2.056</td>
<td>2.3</td>
<td>−1.7</td>
</tr>
<tr>
<td>43</td>
<td>111504.193</td>
<td>−1.8</td>
<td>42.6</td>
<td>2.05</td>
<td>2.4</td>
<td>−1.5</td>
</tr>
<tr>
<td>44</td>
<td>111541.484</td>
<td>−1.9</td>
<td>164.2</td>
<td>1.99</td>
<td>7.8</td>
<td>52.6</td>
</tr>
<tr>
<td>45</td>
<td>111578.919</td>
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<td>−11.4</td>
<td>2.038</td>
<td>2.4</td>
<td>−1.5</td>
</tr>
<tr>
<td>46</td>
<td>111616.036</td>
<td>−2.4</td>
<td>−28.0</td>
<td>2.037</td>
<td>1.8</td>
<td>−6.8</td>
</tr>
<tr>
<td>47</td>
<td>111652.982</td>
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<td>48</td>
<td>111689.751</td>
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<td>2.5</td>
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<td>2.005</td>
<td>2.5</td>
<td>−1.4</td>
</tr>
</tbody>
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The most important implication of this analysis is that the variation in the transition strengths with respect to the vibrational levels of the excited state can be accurately explained on the basis of the new potential. Figure 5 shows a plot of the Rabi frequencies derived from the observed spectra against those calculated from the corrected potential. For comparison, the same plots based on the ab initio potential are also shown. The calculated values are calibrated on the basis of our recent measurement of the transition dipole moment between (3)1 Σ+*, v′ = 41 and X1 Σ+, v″ = 91, 0.035(2)e a₀, determined via dark resonance spectroscopy of the rovibrational ground-state molecules. Weakly bound molecules have more than 90 nodes in their wave function; hence the FCFs are highly dependent on the wave function of the excited state. In other words, the FCFs can serve as a sensitive measure of the accuracy of wave functions. Our results show that accuracies of 0.14% in vibrational level spacings and 0.3% in rotational constants are sufficient to predict the FCFs from weakly bound levels; these values are justified by considering the typical size of nodes in the radial wave function. On one hand, the weakly bound level v″ = 91 in the ground state X1 Σ+ has an outer turning point of ~10 Å and an inner turning point of ~2 Å in the internuclear distance. Within these two points there are 91 nodes; hence each node has a typical size of ~0.1 Å. Therefore, the required accuracy for representing the wave function is ~10^-2 Å. On the other hand, in the present analysis an accuracy of ~0.3% in rotational constants or ~0.15% in internuclear distance is obtained for the (3)1 Σ+ state because the relation between the rotational constant B and the internuclear distance R is given by B ∝ R^-3. Assuming the typical size of molecules in the v′ = 41–50 levels of (3)1 Σ+ as 6 Å, we can derive the accuracy of the modified potential in the radial direction as ~10^-2 Å; this value is in good agreement with the required accuracy for representing the weakly bound level.

Now that we obtained an accurate potential curve as well as the absolute values of the transition dipole moment of the v′ = 41, (3)1 Σ+ level with the v″ = 0 and v″ = 91 levels of X1 Σ+, we can predict the transition dipole moment for each transition. Figure 6 shows our prediction for the transition dipole moments of the (3)1 Σ+ state with the least-bound state and for those with the lowest rovibrational level (v″ = 0). The v′ = 41 level used in Ref. [18] has favorable wave function overlaps with both the weakly bound and the lowest rovibrational levels; however other levels such as v′ = 38 and v′ = 39 can potentially serve as an intermediate state for the STIRAP transfer of weakly bound molecules to the rovibrational ground state. The potential presented can enable an accurate prediction for other isotopic combinations of KrB. Furthermore, the present method for achieving an accurate potential and verifying its accuracy can be extended to other molecular states that exhibit significant spin-orbit interaction by evaluating eigenvalues via coupled channel calculations including spin-orbit interaction.
calibrated by dark resonance spectroscopy for the transition accurate potential. The absolute values for weakly bound levels are described by a single potential. The simplicity of the \((3)^1 \Sigma^+\) state enabled us to modify the potential to reproduce our observations as well as to assign the spectra. We observed those calculated using an ab initio potential. By carrying out a DPF analysis we constructed an accurate potential and verifying its accuracy on the basis of rotational spectra for weakly bound molecules.

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**APPENDIX: ANALYTICAL EXPRESSION FOR THE LINE SHAPE OF A DEPLETION SPECTRUM**

We consider a general situation (as shown in Fig. 7) and we assume that radiative transitions occur much faster than the time variation of the population in each molecular level because of the photoassociative creation of molecules. We first consider the evolution of the population in the ground and excited states of a single molecule. The optical Bloch equation for this system can be written as

\[
\frac{ds}{dt} = -\left(\Gamma_2 - \Gamma_1\right) \frac{s - w}{2},
\]

\[
\frac{dw}{dt} = \left(\Gamma_2 + \Gamma_1\right) \frac{s - w}{2} - 2\Omega \text{Im}(\bar{\rho}_{eg}),
\]

\[
\frac{d\bar{\rho}_{eg}}{dt} = -\left(\frac{\Gamma_2}{2} - i\Delta\right)\bar{\rho}_{eg} + \frac{i}{2}w\Omega,
\]

where \(s = \rho_{ee} + \rho_{eg}, w = \rho_{eg} - \rho_{ee},\) and \(|e\rangle|g\rangle\) denote an excited state and a weakly bound level, respectively. \(\Delta\) and \(\Omega\) denote the detuning frequency and the Rabi frequency, respectively. The decay rates from the excited state to the initial weakly bound level and to other levels are given by \(\Gamma_1\) and \(\Gamma_2\), respectively. The photoassociation rate is denoted by \(A_{PA}\). The width is much larger than the Rabi frequency in the experiment; hence we can assume that the time evolution of \(s\) and \(w\) is much slower than that of \(\bar{\rho}_{eg}\). Thus, we can set \(d\bar{\rho}_{eg}/dt = 0\) and obtain the following expression for \(\bar{\rho}_{eg}\):

\[
\bar{\rho}_{eg} = \frac{i\Omega w}{\Gamma_2 - 2i\Delta}.
\]
Substituting Eq. (A2) into Eqs. (A1) we obtain alternative equations for $s$ and $w$ as

$$\frac{ds}{dt} = -\left(\Gamma_2 - \Gamma_1\right) \frac{s - w}{2},$$

$$\frac{dw}{dt} = \left(\Gamma_2 + \Gamma_1\right) \frac{s - w}{2} - \frac{2\Omega^2 \Gamma_2 w}{\Gamma_s^2 + 4\Delta^2}. \tag{A3}$$

Here we assume that the time taken by the mean value of the population ratio $w/s$ to attain a constant value $z$ after is greater than the typical time for radiative transitions. By using the relation $w = zs$, Eqs. (A3) give the following equations:

$$\frac{dz}{dt} = \left(\Gamma_2 + \Gamma_1\right) \frac{1-z}{2} - \frac{2\Omega^2 \Gamma_2 zs}{\Gamma_s^2 + 4\Delta^2}, \tag{A4}$$

Substituting the first equation in the second equation we obtain a time-independent equation for $z$:

$$(1-z)[\Gamma_2(1+z) + \Gamma_1(1-z)] = \frac{4\Omega^2 \Gamma_2}{\Gamma_s^2 + 4\Delta^2} z. \tag{A5}$$

This equation is readily solved, and it gives the following expression for $z$:

$$z = \frac{\sqrt{1 + k^2}\Gamma_s^2 + 2\Gamma_1\Gamma_2 - k(\Gamma_2 + \Gamma_1)}}{\frac{\Gamma_2 - \Gamma_1}{\Gamma_s^2 + 4\Delta^2}}, \tag{A6}$$

$$k = \frac{2\Omega^2}{\Gamma_s^2 + 4\Delta^2}.$$

This expression is used in the following discussion. Next, we derive rate equations for the population in the ground and excited molecular levels for the number of molecules:

$$\frac{dN_g}{dt} = A_{PA} - (\Gamma_2 - \Gamma_1)N_e,$$

$$N_e = \frac{1-z}{2}N,$$ \tag{A7}

$$N_g = \frac{1+z}{2}N,$$

where $N_g$ and $N_e$ denote the number of molecules in the ground and excited states, respectively, and $N = N_g + N_e$ is the total number of molecules. In these rate equations a typical time scale is of the order of 1 ms and it is governed by $A_{PA}$. This is much longer than the typical time scale for radiative transitions in most cases ($\ll 1 \mu$s). The time evolution of $N_g$ is given by

$$\frac{dN_g}{dt} = \frac{k+1}{2}A_{PA} - \frac{k-1}{2}(\Gamma_2 - \Gamma_1)N_g. \tag{A8}$$

Thus, the solution for $N_g$ is given by

$$N_g(t) = \frac{A_{PA}^2}{(\Gamma_2 - \Gamma_1)} \frac{1+z}{1-z} \left(1 - \exp\left[-\frac{1-z}{2}(\Gamma_2 - \Gamma_1)t\right]\right). \tag{A9}$$

This expression gives the line shape of a depletion spectrum for an interaction time $t$. Assuming $(\Gamma_2 - \Gamma_1)t \gg 1$, Eq. (A9) gives

$$N_g(t) \to \frac{A_{PA}}{\Gamma_2 - \Gamma_1}. \tag{A10}$$

on resonance ($\Delta = 0$ and $w \to 0$), whereas $N_g$ at an infinite detuning ($\Delta \to \infty$ and $w \to 1$) is given as

$$N_g(t) \to \frac{1}{2} A_{PA} t. \tag{A11}$$

$N_g$ at an infinite detuning is much larger than $N_g$ on resonance. Thus the width of the spectrum is determined by finding $w$ such that it satisfies

$$N_g(t) = \frac{1}{2} A_{PA} t. \tag{A12}$$

Substituting Eq. (A9) into Eq. (A12) and rewriting the equation with a new variable $x \equiv 1 - z (\ll 1)$, we obtain the following equation:

$$\frac{2}{x} \left[1 - \exp\left[-\frac{x}{2}(\Gamma_2 - \Gamma_1)t\right]\right] = \frac{\Gamma_2 - \Gamma_1}{2} t. \tag{A13}$$

An analytical solution of this equation is given by

$$x = \frac{4 + 2W(-2/e^2)}{(\Gamma_2 - \Gamma_1)t} \times 3.18724\ldots, \tag{A14}$$

where $W$ is the Lambert $W$ function. Thus the parameter $k$ in Eqs. (A6) is given by

$$k = \frac{3.18724(1-1.59362/\Gamma_2 t)}{(\Gamma_2 - \Gamma_1) t - 3.18724}. \tag{A15}$$

Assuming $(\Gamma_2 - \Gamma_1) t \gg 1$, we obtain the following expression for the FWHM:

$$2\Delta \approx 0.79\Omega\sqrt{(\Gamma_2 - \Gamma_1)t}. \tag{A16}$$

When decays from the excited state to the initial state are negligible $(\Gamma_2 \gg \Gamma_1)$, we obtain a simple relation (1). Expression (1) was experimentally confirmed by observing the relation between the width and the intensity (Fig. 8). It is difficult to evaluate the numerical factor $\sqrt{\Gamma} \tau$ precisely. Therefore the width of a depletion spectrum cannot serve as an accurate measure of the transition strength. However, expression (1) enables us to systematically compare transition strengths for different vibrational levels.

![FIG. 8. (Color online) Width of the depletion spectrum vs intensity. The widths of the depletion spectra for the $v' = 42$ level are plotted with respect to the intensity of the depletion laser. The errors indicate standard deviations in the fits to the spectra.](image-url)