Theoretical studies of photoassociation in ultracold metastable helium

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Abstract. Line shifts and line shapes for photoassociation of spin-polarised metastable helium to long-range vibrational states in the $J = 1, 0^+$ potential dissociating to the $2s^2S_1 + 2p^3P_0$ limit are studied using a nonperturbative multichannel calculation valid for arbitrary laser intensities. Asymptotically-free dressed states of the laser plus matter system are used to obtain the S-matrix elements required to generate the photoassociation profiles. Issues associated with the very shallow nature of the potentials that support the excited states are investigated.

1. Introduction
Photoassociation (PA) experiments in which two interacting ultracold atoms (usually ground state alkali atoms or metastable rare gas atoms) are resonantly excited by a laser to a molecular bound state provide a powerful technique to study the dynamics of ultracold collisions [1]. Since the colliding atoms are so cold, the energy of the initial scattering state is well determined and the resonant laser energies corresponding to transitions to various bound states produce a very high resolution spectrum ($< 1\,\text{MHz}$). At this level of precision, energy level shifts induced by the PA laser can be significant [2] and an understanding of the dependence of the energy level shifts upon the laser intensity, polarization and frequency is crucial.

Line profiles for PA in metastable helium (see figure 1) have been observed experimentally by Kim et al [3]. A short laser pulse was applied to the magnetically trapped gas, the trapping potential turned off and ballistic expansion used to measure the PA-induced temperature profile of the gas for various laser frequencies. Lorentzian fits to the profiles yielded resonance shifts that varied linearly with laser intensity up to 6 W/cm$^2$. Difficulties in accurately determining the laser intensity incident upon the gaseous cloud led to large error bars.

Previous theoretical investigations of PA have either relied upon perturbative techniques [4, 2] or have assumed that the laser interaction vanishes at large interatomic separation [5]. To properly treat the PA process, one must use laser-dressed states [6] that asymptotically decouple, thus allowing the S-matrix to be correctly defined. This will be shown to be especially important for the weakly bound levels in the present study.

2. Theory
The eigenstate $|\Psi_j\rangle$ of two atoms colliding in the presence of a radiation field satisfies

$$[\hat{H}_{\text{mol}} + \hat{H}_{\text{int}} + \hat{H}_{\text{rad}}]|\Psi_j\rangle = E|\Psi_j\rangle$$

(1)
where $\hat{H}_{\text{mol}}$ is the total molecular Hamiltonian, $\hat{H}_{\text{rad}}$ is the Hamiltonian for a free laser field of angular frequency $\omega$, intensity $I$ and polarization $\epsilon$, and $\hat{H}_{\text{int}} = - (\sqrt{I/2c_0}) \mathbf{d} \cdot \epsilon$ describes the interaction between the laser and the molecular dipole moment $\mathbf{d}$. Using the expansion $|\Psi_j\rangle = \sum_i R^{-1} G_{ij}(R) |i\rangle$ in terms of a basis $\{|i\rangle\}$, yields the close-coupled equations

$$-\frac{\hbar^2}{2m} \frac{d^2}{dR^2} G_{ij}(R) + \sum_l W_{il}(R) G_{lj}(R) = 0$$

(2)

where $W_{il}(R) = \langle i | \hat{H} - E | l \rangle$ and $\hat{H}$ contains all the terms of $\hat{H}_{\text{mol}}$ except the radial derivative term.

In choosing the basis, we note that molecular interactions are most naturally described in the molecular frame whereas selection rules for the laser interaction refer to the space-fixed frame. Hence we use the basis $|i\rangle \equiv |\gamma, j_1 j_2, j\Omega_j, J m_J \rangle \otimes |n, \omega, \epsilon\rangle$, where $\Omega_j$ is the projection of the total electronic angular momentum $j$ upon the molecular axis and $m_J$ labels the projections of the total angular momentum $J$ in the space-fixed frame. In this basis, $W_{il}$ for the states coupled by $\sigma^-$ polarised light has the asymptotic $R \to \infty$ form,

$$
\begin{pmatrix}
-E_K & 0 & 0 & 0 & 0 & 0 & \hbar \Omega^{0}_{2j+1} \\
0 & -E_K & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -E_K & 0 & 0 & \hbar \Omega^{-1}_{5} & \hbar \Omega^{0}_{3} \\
0 & 0 & 0 & -E_K & 0 & 0 & \hbar \Omega^{0}_{5} \\
0 & 0 & 0 & 0 & -E_K & 0 & \hbar \Omega^{0}_{3} \\
\hbar \Omega^{-1}_{5} & 0 & \hbar \Omega^{-1}_{5} & \hbar \Omega^{0}_{3} & \hbar \Omega^{1}_{5} & 0 & -\Delta E - i \Gamma_j
\end{pmatrix}
$$

(3)

where $\hbar \Omega^{0}_{2j+1} \equiv \langle i | \hat{H}_{\text{int}} | l \rangle$ are the Rabi couplings and $\Gamma$ is the spontaneous decay width of the excited state.
Figure 2. Spontaneous emission cross section profiles as a function of laser detuning \( \Delta E \) averaged over degenerate entrance channels for laser intensities of (a) 64 mW/cm\(^2\) and (b) 3.2 W/cm\(^2\).

In this basis the system is not asymptotically uncoupled; in fact the shallow nature of the potential wells (small \( \Delta E \)), the ultracold temperatures (small \( E_K \)) and high laser intensities (large \( \hbar \Omega \)) can cause the off-diagonal terms to be of the same order of magnitude as the diagonal terms. To properly define the S-matrix, we must transform to a laser-dressed set of basis states \( |\beta\rangle = \sum_i U_{\beta i} |i\rangle \) that diagonalise the dressed potential matrix \( W_{\beta\beta'} \). Although Napolitano [6] was able to analytically form dressed state combinations for the sodium system investigated, the large number of couplings in \( W_{\text{il}} \) for our system forces us to perform a numerical diagonalisation.

All dressed states potentially include the undressed excited state, resulting in complex energies \( E_\beta = -W_{\beta\beta'}^D (R = \infty) \). However, for the current problem, we find only two combinations are formed with the excited state: one a closed channel and the other a ‘pseudo-open’ channel. The remaining dressed states are degenerate open channels (real \( E_\beta \)) that span a subspace with an arbitrariness that varies with laser frequency. The channel designated as the ‘pseudo-open’ channel has Re(\( E_\beta \)) \( \gg \) Im(\( E_\beta \)), which suggests open-channel behaviour, however the small imaginary component introduces exponential behaviour characteristic of closed channels. In a rigorous treatment, one would be forced to treat this as a closed-channel but for numerical reasons this is not a feasible approach. Napolitano [6] circumvented this problem by allowing a complex total energy, which removed the asymptotic imaginary component but has the side effect of introducing small complex perturbations to the excited states. Although reasonable for the sodium system, the method will introduce additional asymptotic imaginary components in the degenerate open states for the metastable helium system. Instead, we prefer to slightly redefine the scattering matrix, by matching the asymptotic functions to a finite interatomic distance \( R = R_{\text{max}} \) instead of \( R \to \infty \). Although the particular choice of \( R_{\text{max}} \) will modify the cross-sections, we do not expect it to modify the profile shape as long as \( R_{\text{max}} \) is outside the interaction region.

To extract the S matrix elements, we solve the radial equation (2) in the dressed state basis and, for the open and pseudo-open channel scattering states, match to the asymptotic form

\[
G_{\beta\beta'}(R) = A_{\beta\beta'} h^-_\beta(R) + B_{\beta\beta'} h^+_\beta(R)
\]

where \( h^\pm_\beta \sim 1/\sqrt{2|k_\beta|} \exp(\pm ik_\beta R) \) and \( k_\beta = \sqrt{2\mu \text{Re}(E_\beta)}/\hbar \). The S-matrix is then \( S = -BA^{-1} \) and the cross section for spontaneous photon emission from a particular entrance channel \( \beta \) is obtained from the loss of unitarity of the S-matrix:

\[
\sigma_\beta^{\text{photon}} = \frac{\pi}{k^2_\beta} [1 - \sum_{\beta'} |S_{\beta\beta'}|^2].
\]
3. Results and discussion

Two sample PA spectra for the $v = 0 J = 1, 0^+$ vibrational level are shown in figure 2. These spectra exhibit two features not present in previously calculated PA profiles: (i) a non-zero background variation in $g_{\text{photon}}$ and (ii) the appearance of interference behaviour at larger laser intensities. The non-zero background appears to arise from the shallow nature of the $0^+_u$ potential well as artificially deepening the well by two orders of magnitude completely removes the background. The interference feature can be matched perfectly by a Fano profile [7] of the form

$$A_{\text{bg}} - A_{\text{res}} \frac{(\epsilon + q)^2}{1 + \epsilon^2}, \quad \epsilon = \frac{E - E_{\text{res}}}{\Gamma_{\text{res}}/2}$$

where $E_{\text{res}}$ and $\Gamma_{\text{res}}$ are the position and width respectively of the resonance. The experimental measurements [3] do not show such interference and are fitted by standard Lorentzian profiles. A detailed comparison between actual experimental profiles and theoretical predictions needs further investigation.

The dependence of the resonance position and width on laser intensity is shown in figures 3 and 4 respectively. Our calculations indicate that the line shift has a linear dependence and the line width a quadratic dependence. Perturbative calculations of energy level shifts [2] predict a linear dependence and our result of 6.446 MHz / W.cm$^{-2}$ agrees closely with our perturbative result of 6.439 MHz / W.cm$^{-2}$ obtained using the F-operator technique [8] adapted to the laser-matter interaction. Details of this perturbative calculation, together with a more comprehensive account of the nonperturbative calculations presented here, will be the subject of a forthcoming paper.

References

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