Laser-induced collisional energy transfer in Sr–Li system

Chen De-Ying(陈德应)[a)†], Lu Zhen-Zhong(鲁振中)[a)], Fan Rong-Wei(樊荣伟)[a)], Xia Yuan-Qin(夏元钦)[a)], Zhou Zhi-Gang(周志刚)[a)], and Ji Yi-Qin(季一勤)[b)]

[a)] *National Key Laboratory of Science and Technology on Tunable Laser, Harbin Institute of Technology, Harbin* 150001, *China*

[b)] *Tianjin Key Laboratory of Optical Thin Films, Tianjin Jinhang Institute of Technical Physics, Tianjin* 300192, *China*

(Received 12 December 2011; revised manuscript received 19 January 2012)

A four-state model considering the relative velocity distribution function for calculating the cross section of laser-induced collisional energy transfer in a Sr–Li system is presented and profiles of laser-induced collision cross section are obtained. The resulting spectra obtained from different intermediate states are strongly asymmetrical in an opposite asymmetry. Both of the two intermediate states have contributions to the final state, and none of the intermediate states should be neglected. The peak of the laser-induced collisional energy transfer (LICET) profile shifts toward the red and the FWHM becomes narrower obviously with laser field intensity increasing. A cross section of 1.2×10^{-12} cm^2 at a laser field intensity of 2.17×10^7 V/m is obtained, which indicates that this collision process can be an effective way to transfer energy selectively from a storage state to a target state. The existence of saturation for cross section with the increase of the laser intensity shows that the high-intensity redistribution of transition probabilities is an important feature of this process, which is not accounted for in a two-state treatment.

Keywords: laser-induced collisional energy transfer, collision cross section, Sr–Li system

PACS: 32.70.Jz, 34.50.–s **DOI:** 10.1088/1674-1056/21/8/083202

1. Introduction

A laser-induced collision is an optical process in which a laser field is used during the collision between two different particles to induce selective energy transfer from a level in one atomic or molecular species to another level in a different species. It cannot occur unless the two mechanisms, i.e., collision and radiative interaction, are both present; any of them singly cannot induce this interparticle transition.

The issue of laser-induced inelastic collisions was first addressed some 30 years ago. As plenty of the experimental studies have been carried out,[1–3] people also have proposed many relevant theoretical models on laser-induced collisional energy transfer (LICET).[4–20] The theoretical and experimental research on this issue have made great advances, but some questions remain unanswered. The theoretical results obtained provide only a modest insight into the physics of laser-assisted collisions. An important advance in the understanding of LICET was made by Bambini and Berman[7] who demonstrated that an intermediate state must be treated as real state which may be populated in the LICET process. And in subsequent theoretical work, Geltman[5] proposed that as the intensity increases, the increase of Rabi oscillation frequency in the B atomic transition will lead to strong-field shift in the LICET line shape.

The study of a laser-induced collision process that might broaden and deepen our general understanding of dynamic atomic processes has reached a certain maturity for laser-matter interactions and field-free collision. More specifically, it might pave the way for the control of ultrashort quantum processes, which are deemed to be important for a number of applications, such as laser-driven fusion, plasma heating, and the development of fast optical electronic devices. Energy transfer between different particles occurs more quickly and efficiently because of the participation of the laser field; on the other hand, the effect of interparticle collisions can implement transfers which are difficult for single photon excitation, thus can

†Corresponding author. E-mail: luhit@126.com

© 2012 Chinese Physical Society and IOP Publishing Ltd

obtain the expected highly excited state and consequently short wavelength laser. Therefore, the study of laser-induced collision processes is of great significance for the development of short wavelength laser sources. In addition, the ability to excite the special target level of the chosen particle permits potential applications in controlling the pathways of chemical reactions. The work by Seely[21] showed that LICET reactions were useful for the measurement of particle densities in a tokamak plasma. Hansen *et al.*[22] used a laser-induced charge exchange detection technique to observe a laser-induced high resolution absorption spectra of N_2^+ $A-X(4,\,0)$, which were difficult to obtain by laser-induced fluorescence (LIF).

In the present paper, the four-level LICET model with considering the relative velocity distribution function is proposed. The strong-field LICET process is discussed in detail for predicting the dependence of the line shape on the transfer laser parameter and further revealing the contribution of intermediate state in LICET process, which may have significance in unraveling the mechanism of LICET process.

2. Theory

Sr–Li system LICET process can be expressed by the following formula:

$$\begin{aligned}&\mathrm{Sr}(5\mathrm{p}\ {}^1\mathrm{P}_1^0)+\mathrm{Li}(2\mathrm{s}\ {}^2\mathrm{S})+\hbar\omega(669.8\ \mathrm{nm})\\&\longrightarrow \mathrm{Sr}(5\mathrm{s}^2\ {}^1\mathrm{S})+\mathrm{Li}(4\mathrm{d}\ {}^2\mathrm{D}).\end{aligned}$$

Figure 1 illustrates those energy levels relevant to the LICET process.

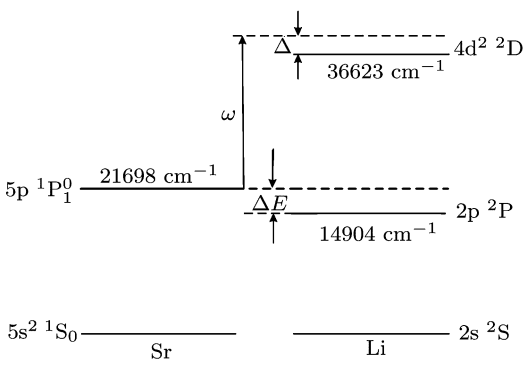


Fig. 1. LICET transition in Sr–Li system. The energy levels in the figure are given in cm^{-1}.

The states of Sr–Li compound system shown in Fig. 1 can be defined as

$$\left.\begin{aligned}|1\rangle &= |\mathrm{Sr}(5\mathrm{s}^2\ {}^1\mathrm{S})\rangle|\mathrm{Li}(2\mathrm{p}\ {}^2\mathrm{P})\rangle,\\|2\rangle &= |\mathrm{Sr}(5\mathrm{s}5\mathrm{p}\ {}^1\mathrm{P}_1^0)\rangle|\mathrm{Li}\,(2\mathrm{s}\ {}^2\mathrm{S})\rangle,\\|3\rangle &= |\mathrm{Sr}(5\mathrm{s}5\mathrm{p}\ {}^1\mathrm{P}_1^0)\rangle|\mathrm{Li}\,(2\mathrm{p}\ {}^2\mathrm{P})\rangle,\\|4\rangle &= |\mathrm{Sr}\,(5\mathrm{s}^2\ {}^1\mathrm{S})\rangle|\mathrm{Li}\,(4\mathrm{d}\ {}^2\mathrm{D})\rangle.\end{aligned}\right\}$$

The wave function can be expressed by the basis states as follows:

$$\Psi=\sum_{n=1}^{4}c_n\,(t)\,|n\rangle\exp\left(-\mathrm{i}E_nt/\hbar\right). \tag{1}$$

The trajectory is assumed for $R(t)=(b^2+v^2t^2)^{1/2}$, where b is an impact parameter and v is the relative velocity. And then the interaction Hamiltonian can be written as

$$H(t)=H_{\mathrm{r}}+V_{\mathrm{AB}}. \tag{2}$$

The radiative interaction H_{r} can be given by

$$H_{\mathrm{r}}=-\,\mathrm{e}\boldsymbol{E}_0\cdot(\boldsymbol{r}_{\mathrm{A}}+\boldsymbol{r}_{\mathrm{B}})\cos\omega t, \tag{3}$$

where the $\boldsymbol{E}_0(t)=\boldsymbol{E}_0\cos(\omega t)$ is the inducing laser field with frequency ω.

The interaction V_{AB} can be represented by the multipole expansion[23,24]

$$\begin{aligned}V_{\mathrm{AB}}=&\sum_{j=3}\sum_{l=1}^{j-2}\sum_{m=-1<}^{l<}(-1)^{j-l-1}\\&\times\left[\frac{(2j-2)!}{(2j-2l-2)!(2l)!}\right]^{1/2}\\&\times C(j-l-1,l,j-1;m,-m,0)\\&\times Q_l^m(\mathrm{A})Q_{j-l-1}^{-m}(\mathrm{B})R(t)^{-j}.\end{aligned}$$

Here, $l_<$ is the lesser of l and $j-1-l$, $Q_l^m(\mathrm{A})$ and $Q_{j-l-1}^{-m}(\mathrm{B})$ are multipole moment operators, and $C(j-l-1,\,l,\,j-1;\,m,\,-m,\,0)$ is the Clebsch–Gordan coefficient.[23]

By making formula (3) simultaneous with the Schrödinger equation and supposing that the laser field is sufficiently intense, the motion equation of formula (2) can be obtained as follows:

$$\left\{\begin{aligned}\mathrm{i}\dot{c}_1&=V_{12}c_2\,\mathrm{e}^{\mathrm{i}\omega_{12}t}+V_{13}\,\mathrm{e}^{\mathrm{i}(\omega_{13}+\omega)t}c_3\\&\quad+V_{14}\,\mathrm{e}^{\mathrm{i}(\omega_{14}+\omega)t}c_4,\\\mathrm{i}\dot{c}_2&=V_{21}c_1\,\mathrm{e}^{\mathrm{i}\omega_{21}t}+V_{23}\,\mathrm{e}^{\mathrm{i}(\omega_{23}+\omega)t}c_3,\\\mathrm{i}\dot{c}_3&=V_{31}\,\mathrm{e}^{\mathrm{i}(\omega_{31}-\omega)t}c_1+V_{32}\,\mathrm{e}^{\mathrm{i}(\omega_{32}-\omega)t}c_2\\&\quad+V_{34}c_4\,\mathrm{e}^{\mathrm{i}\omega_{34}t},\\\mathrm{i}\dot{c}_4&=V_{41}\,\mathrm{e}^{\mathrm{i}(\omega_{41}-\omega)t}c_1+V_{34}c_3\,\mathrm{e}^{\mathrm{i}\omega_{43}t},\end{aligned}\right. \tag{4}$$

where

$$V_{mn}=\langle m|H(t)|n\rangle,$$

and

$$\omega_{mn} = (E_m - E_n)/\hbar.$$

Through solving the coupled equations as a function of impact parameter, we can obtain the transition probabilities of the final state. And then, by considering the probability density function of relative velocity between two colliding atoms in three-dimensional space under thermal equilibrium condition, the laser-induced collision cross section at a given temperature can be obtained as

$$\sigma = \int_0^\infty \int_0^\infty |c_4(b, \upsilon, t = \infty)|^2 F(\upsilon) 2\pi b \mathrm{d}b \mathrm{d}\upsilon. \quad (5)$$

The probability density function $F(\upsilon)$ can be represented by[13]

$$F(\upsilon) \equiv 4\pi\upsilon^2 f(\upsilon) = 4\pi \left(\frac{M}{2\pi kT}\right)^{3/2} \upsilon^2 \, \mathrm{e}^{-M\upsilon^2/2kT}.$$

Here, M is the reduced mass of the two colliding atoms, T is the thermodynamic temperature, and k is the Boltzmann constant.

3. Results and discussion

When the laser field intensity is fixed to be 2.4×10^6 V/m, the relative speed between the two colliding atoms is 700 m/s, for the Sr–Li system the collisional cross section as a function of transfer laser detuning is shown in Fig. 2. Figure 2(a) shows the case where the collisional interaction takes place first, coupling the initial state to the intermediate state, followed by the laser interaction, i.e., the excitation channel includes the three states of $|1\rangle$, $|2\rangle$, and $|4\rangle$. While figure 2(b) shows the opposite order of the occurrence of these interactions, i.e., the laser interaction takes place first, followed by the collisional interaction, coupling the intermediate state to the final state, and the excitation channel includes the three states of $|2\rangle$, $|3\rangle$, and $|4\rangle$. Figure 2(c) shows the results from the four-state model. The resulting spectrum in Fig. 2(b) is strongly asymmetrical in an opposite asymmetry compared with that in Fig. 2(a). An understanding of this is based on the adiabatic molecular curves given in Fig. 3. Within a simple molecular picture of the process we note that the LICET process can be viewed as a resonant radiative transition between adiabatic molecular states as figure 4 shows. Molecular curves of $|1\rangle$ and $|2\rangle$ become strongly repelling energy curves by the coupling between $|1\rangle$ and $|2\rangle$. When $\omega > \omega_0$,

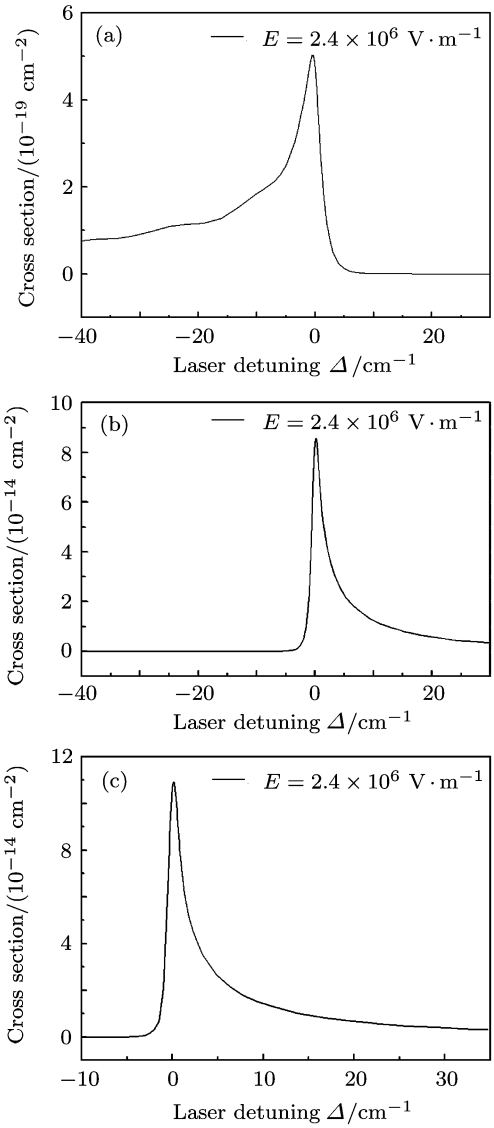


Fig. 2. LICET spectra for the Sr–Li system, where the excitation channel includes the three states of $|1\rangle$, $|2\rangle$ and $|4\rangle$ (a); $|2\rangle$, $|3\rangle$, and $|4\rangle$ (b); all of the four states (c).

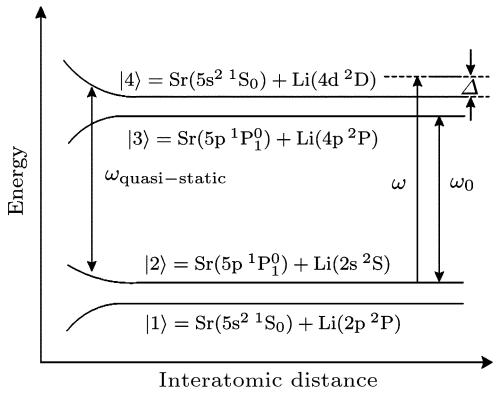


Fig. 3. Schematic adiabatic quasimolecular potential for the Sr–Li system.

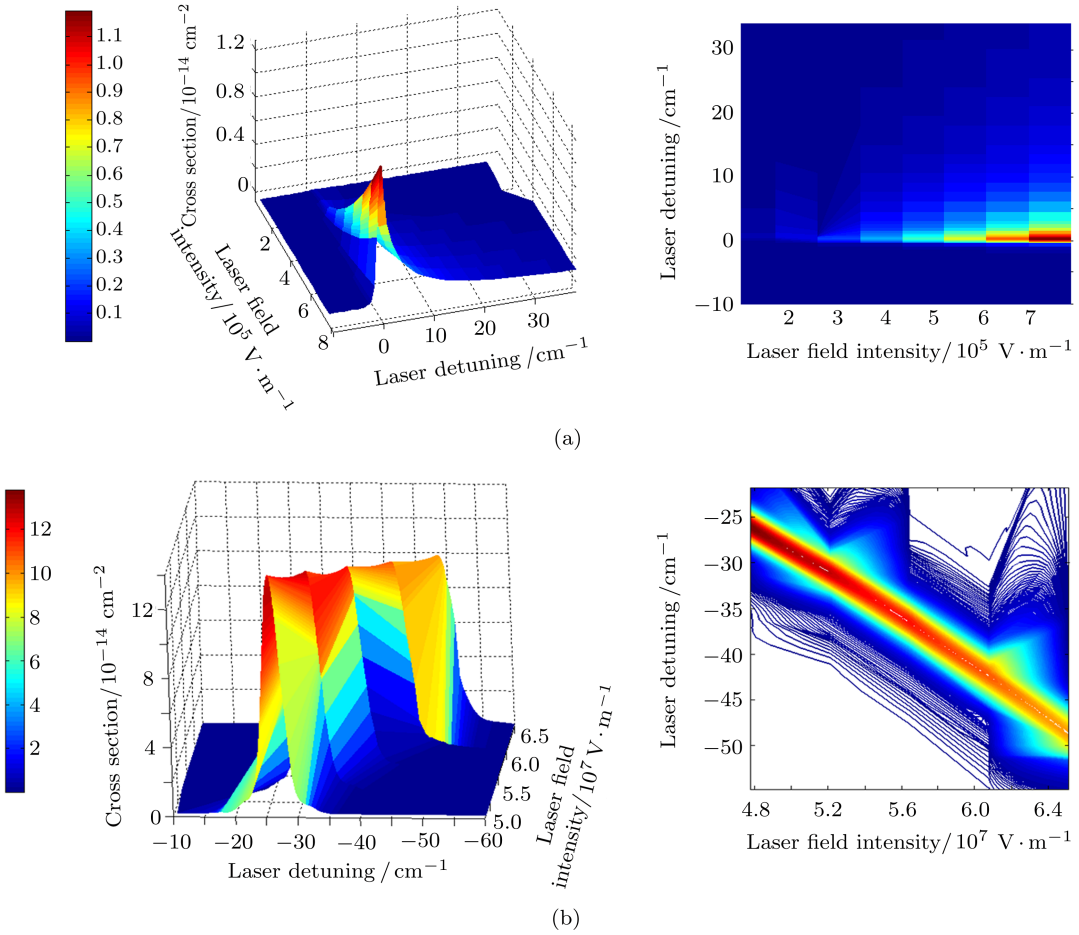


Fig. 4. (colour online) LICET spectrua for the Sr–Li system at various transfer laser intensities.

there are internuclear separations for which the transition frequency of the quasimolecule is resonant with the laser field, which leads to the slow reduction of the transition probability in the static wing as shown in Fig. 4(a). On the other hand, molecular curves of $|3\rangle$ and $|4\rangle$ become strongly repelling energy curves by the coupling between $|3\rangle$ and $|4\rangle$ states, thereby accounting for the universal asymmetric line shape shown in Fig. 2(b). It is also apparent from Fig. 2 that both of the intermediate states have contributions to the final state, so none of the intermediate states should be neglected.

To illustrate the influence of laser field on the LICET profile, we take more laser field intensities into consideration. A series of cross sections at various field intensities are shown in Fig. 4. In weak field as shown in Fig. 4(a), the results are characterized by a universal asymmetric line shape and the magnitude is proportional to the field intensity. The profile can be divided into two parts, i.e., the quasi-static wing and the anti-static wing.

As the field intensity increases, shown in Fig. 4(b), the resulting spectrum shifts in frequency toward the anti-static wing. An understanding of this is that as the laser intensity increases, the Rabi oscillation frequency in the Li(2s–2p) atomic transition increases and become dominant over the van der Waals quasimolecule mechanism. It tends to enhance the LICET cross section as ω approaches to ω_{32} which is $< \omega_{42}$, resulting in the bathochromic shift.[5] Figure 6 exhibits the shift of the peak cross section as a function of transfer laser intensity. When the laser field intensity is 8.25×10^7 V/m, the maximum occurs at $\Delta = -78.35$ cm^{-1}, making this configuration very promising for a quantitative experimental study.

The peak cross section as a function of transfer laser intensity for the Sr–Li system is given in Fig. 5. A cross section of 1.2×10^{-12} cm^2 at a laser field intensity of 2.17×10^7 V/m is obtained. The collision cross section rises with laser intensity increasing and tends to saturate. It has a significant deviation from the two-state treatment result that the LICET peak cross

section is monotonically increasing with the laser intensity increasing which does not conform to physical reality. The existence of saturation for cross section with the increase of the laser intensity shows that the high-intensity redistribution of transition probabilities is an important feature of this process which is not accounted for in a two-state treatment.[5]

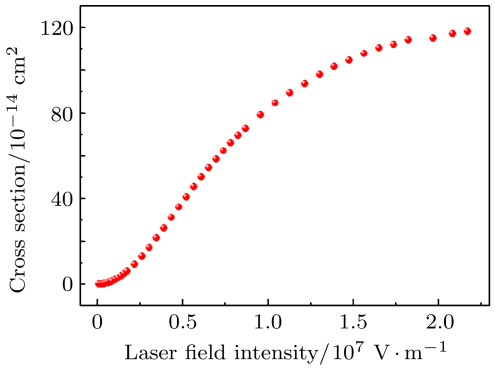


Fig. 5. Peak cross section as a function of transfer laser intensity for the Sr–Li system.

The previous theoretical work[5−12] was based on the assumption that the relative velocity was fixed. Nevertheless, all of the previous experimental investigations on LICET were made for mixed metal vapors, atoms that were collisional, should have many different speeds, which were distributed in a certain way, around an average value that had been used in the early theoretical calculation. Therefore, it is necessary to consider this distribution in the calculations. In the present calculation, the distribution function of relative velocity between two colliding atoms in three-dimensional space is obtained based on Maxwell velocity distribution function under thermal equilibrium condition. And the profiles of laser-induced collision cross section at different temperatures are obtained and shown in Fig. 7. It appears that the profiles of the spectra keep invariant at different temperatures. Since the average velocity decreases as temperature increases, it leads the Weisskopf radius to rise. As a result, the magnitude of the cross section increases with temperature decreasing.

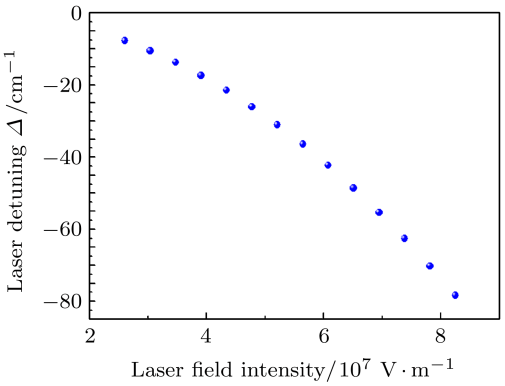


Fig. 6. Shift of the peak cross section as a function laser field intensity.

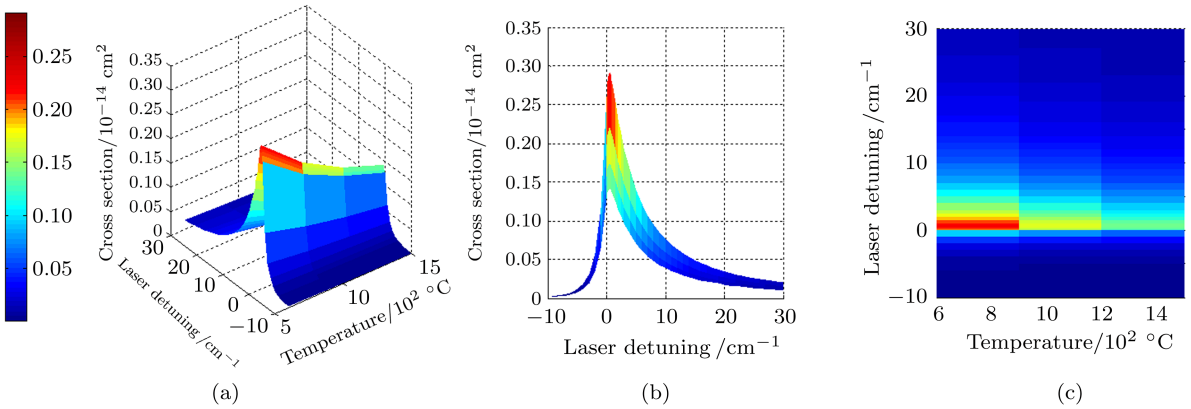


Fig. 7. (colour online) LICET spectra for the Sr–Li system at different temperatures.

4. Conclusion

In this paper, the laser-induced collisional energy transfer process in the Sr–Li system is studied. A four-state model considering the relative velocity distribution function for calculating the cross section is presented and profiles of laser-induced collision cross sections are obtained. The resulting spectra obtained

from different intermediate states are strongly asymmetrical in an opposite asymmetry, which is due to the opposite order of the occurrence of laser field and collision interaction. Both of the intermediate states have contributions to the final state, and none of the intermediate states should be neglected. The peak of the LICET profile shifts toward the red and the FWHM becomes narrower obviously with the laser field intensity increasing. When the laser field intensity is 8.25×10^7 V/m, the maximum occurs at $\Delta = -78.35$ cm^{-1}, making this configuration very promising for a quantitative experimental study. The collision cross section rises with laser intensity increasing, and becomes smaller as the system temperature increases. A cross section of 1.2×10^{-12} cm^2 at a laser field intensity of 2.17×10^7 V/m is obtained which indicates that this collision process can be an effective way to transfer energy selectively from a storage state to a target state. The existence of saturation for cross section with the increase of the laser intensity is in sharp contrast to the two-state results where cross section goes over to the purely increasing indicating that the high-intensity redistribution of transition probabilities is an important feature of this process which is not accounted for in a two-state treatment.

References

[1] Green W R, Lukasik J, Willison J R, Wright M D, Young J F and Hass S E 1979 *Phys. Rev.* A **15** 1333

[2] Debarre A 1983 *J. Phys.* B **16** 431

[3] Brechnignac C, Cahuzac P H and Toschek P E 1980 *Phys. Rev.* A **21** 1969

[4] Matera M, Mazzoni M, Buffa R, Cavalieri S and Arimondo E 1987 *Phys. Rev.* A **36** 1471

[5] Geltmen S 1992 *Phys. Rev.* A **45** 4792

[6] Harris S E 1977 *IEEE J. Quantum Electron.* **13** 972

[7] Bambini A and Berman P R 1987 *Phys. Rev.* A **35** 3753

[8] Agresti A, Berman P R, Bambini A and Stefanel A 1988 *Phys. Rev.* A **38** 2259

[9] Lu Z Z, Chen D Y, Fan R W and Xia Y Q 2011 *Chin. Phys.* B **20** 063202

[10] Chen D Y, Wang Q and Ma Z G 1996 *Acta Opt. Sin.* **16** 1653 (in Chinese)

[11] Chen D Y, Wang Q and Ma Z G 1997 *Sci. Chin.* Ser. A **27** 449 (in Chinese)

[12] Zhang H Y, Chen D Y, Lu Z Z, Fan R W and Xia Y Q 2008 *Acta Phys. Sin.* **57** 7600 (in Chinese)

[13] Zhang H Y, Chen D Y, Lu Z Z, Xia Y Q and Fan R W 2008 *Chin. J. Laser* **35** 77 (in Chinese)

[14] Bambini A, Matera M, Agresti A and Bianconi M 1990 *Phys. Rev.* A **42** 6629

[15] Lisitsa V S and Yakovlenko S I 1974 *Sov. Phys. JETP* **39** 759

[16] Li Y J, Feng H, Sun W G, Zeng Y Y, Wang X L, Li H D and Fan Q C 2011 *Acta Phys. Sin.* **60** 043401 (in Chinese)

[17] Yu Y J, Xu Q and Xu X W 2011 *Acta Phys. Sin.* **60** 123402 (in Chinese)

[18] Lu Z Z, Chen D Y, Fan R W and Xia Y Q 2011 *Sci. Chin.* G: *Phys. Mech. & Astron.* **54** 1

[19] Wang H Y, Zhao G Z and Wang X Q 2011 *Acta Phys. Sin.* **60** 043202 (in Chinese)

[20] Chen X and Mi X W 2011 *Acta Phys. Sin.* **60** 104204 (in Chinese)

[21] Seely J F 1981 *J. Chem. Phys.* **75** 3321

[22] Hansen J C, Kuo C H, Grieman F J and Moseley J T 1983 *J. Chem. Phys.* **79** 1111

[23] Hirschfelder J O and Meath W J 1967 *Adv. Chem. Phys.* **12** 3

[24] Rose M E 1958 *J. Math. Phys.* **37** 215