# Laser-induced quadrupole–quadrupole collisional energy transfer in Xe–Kr

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By considering the relative velocity distribution function and multipole expansion interaction Hamiltonian, a three-state model for calculating the cross section of laser-induced quadrupole–quadrupole collisional energy transfer is presented. Calculated results in Xe–Kr system show that in the present system, the laser-induced collision process occurs for ∼4 ps, which is much shorter than the dipole–dipole laser-induced collisional energy transfer (LICET) process. The spectrum of laser-induced quadrupole–quadrupole collisional energy transfer in Xe–Kr system has wider tunable range in an order of magnitude than the dipole–dipole LICET spectra. The peak cross section decreases and moves to the quasi-static wing with increasing temperature and the full width at half peak of the profile becomes larger as the system temperature increases.

**Keywords:** laser-induced collisional energy transfer, collisional cross section, Xe–Kr system

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## 1. Introduction

For many years, laser-induced inelastic collision has been of considerable interest,[1−23] because the cross section of interparticle inelastic collisions can be greatly increased by several orders by the joint action of the collisional and radiative interactions. So the laser-induced collisional energy transfer (LICET) can realize the rapid and efficient energy transfer between particles; it is also a possible way to make the short-wave laser. The theoretical and experimental researches on this issue both have profound significance.

As is described by many works,[1,2,9] laser-induced collisional energy transfer (LICET) is an optical phenomenon that a pair of colliding atoms transfer to the final states by the joint action of the collisional and radiative interactions. An LICET process between two atoms can be expressed by the reaction

$$A^*(\mathrm{i}) + B(\mathrm{i}') + \hbar\omega \rightarrow A(\mathrm{f}) + B^{**}(\mathrm{f}'). \qquad (1)$$

As plenty of the experimental studies have been made including Sr–Ca,[1] Na–Ca,[2] Eu–Sr,[3] Li–Sr,[4] Rb–K,[5] Li–Sr,[6] people also have proposed many theoretical models on LICET.[7−17]

The previous experimental and theoretical researches on LICET were mainly concentrated on the dipole–dipole process and just few dipole–quadrupole LICET experimental researches[18,19] were made. As to the research on the higher multipole (higher than dipole–quadrupole) interaction process, no such researches were made. Actually, based on our theoretical results, we can clearly see that laser-induced multipole interaction collision cross section also can be large.

In this paper, a three-state model considering the relative velocity distribution function for calculating the cross section of laser-induced quadrupole–quadrupole collisional energy transfer in Xe–Kr system is presented. The factor of system temperature is discussed to illustrate the features of the quadrupole–quadrupole LICET process. It has been interesting to observe that the peak position and the spectrum profile depend strongly on system temperature, while in the previous dipole–dipole LICET systems the temperature has no obvious effect on the line shape.[24]

## 2. Theory

Equation (1) shows the LICET process between two different atoms. Taking independent atomic states of atoms $A$ and $B$ as basis states, the three

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states of the compound system can be expressed as

$$\begin{cases} |1\rangle = |A\rangle|B^*\rangle, \\ |2\rangle = |A^*\rangle|B\rangle, \\ |3\rangle = |A\rangle|B^{**}\rangle, \end{cases} \tag{2}$$

where $|A\rangle$ and $|A^*\rangle$ denote the ground and excited states of atom $A$, $|B\rangle$, $|B^*\rangle$, and $|B^{**}\rangle$ denote the ground, excited, and high-excited states of atom $B$ respectively.

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

$$\Psi = \sum_{n=1}^{3} c_n(t)|n\rangle \exp(-\mathrm{i}E_n t/\hbar). \tag{3}$$

We assume the coordinate system as shown in Fig. 1 where atom $B$ is fixed and atom $A$ moves in a straight line with velocity $v$ along the $z$ axis. The linear 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. Assuming $\boldsymbol{E}(t) = E_0\boldsymbol{a}_y \cos(\omega t)$ is the laser field with frequency $\omega$, then the interaction Hamiltonian can be written as

$$H(t) = -ey_A E_0 \cos\omega t - ey_B E_0 \cos\omega t + V_{AB}. \tag{4}$$

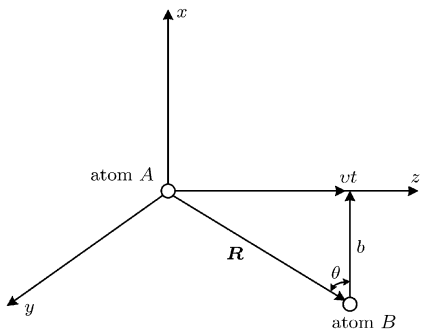



**Fig. 1.** Coordinate system formed by two colliding atoms.

The interaction $V_{AB}$ can be represented by the multipole expansion[21]

$$V_{AB} = \sum_{j=3} V_j R(t)^{-j}, \tag{5}$$

where

$$V_j = \sum_{l=1}^{j-2} \sum_{m=-1<}^{l<} F(j,l,m) Q_l^m(A) Q_{i-l-1}^{-m}(B).$$

Here $l_<$ is the lesser of $l$ and $j-1-l$, $Q_l^m(A)$ and $Q_{j-l-1}^{-m}(B)$ are multipole moment operators and $F(j,l,m)$ is given by[25,26]

$$\begin{aligned} F(j,l,m) &= (-1)^{j-l-1}\left[\frac{(2j-2)!}{(2j-2l-2)!(2l)!}\right]^{1/2} C(j-l-1,l,j-1;m,-m,0) \\ &= \frac{(-1)^{j-l-1}(j-1)!}{[(l-m)!(l+m)!(j-1-l-m)!(j-1-l+m)!]^{1/2}}, \end{aligned}$$

where $C(j-l-1,l,j-1;m,-m,0)$ is Clebsch–Gordan coefficient.

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

$$\begin{cases} \mathrm{i}\dot{c}_1 = V_{12}\,\mathrm{e}^{\mathrm{i}\omega_{12}t}c_2 + V_{13}\,\mathrm{e}^{\mathrm{i}(\omega_{13}+\omega)t}c_3, \\ \mathrm{i}\dot{c}_2 = V_{21}\,\mathrm{e}^{\mathrm{i}\omega_{21}t}c_1, \\ \mathrm{i}\dot{c}_3 = V_{31}\,\mathrm{e}^{\mathrm{i}(\omega_{31}-\omega)t}c_1, \end{cases} \tag{6}$$

where

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

and

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

Using unitary transformation, equation (6) can be simplified as

$$\begin{cases} \mathrm{i}\dot{a}_1 = -\omega_{21}a_1 + V_{12}a_2 + V_{13}a_3, \\ \mathrm{i}\dot{a}_2 = V_{21}a_1, \\ \mathrm{i}\dot{a}_3 = V_{31}a_1 + (\omega_{32}-\omega)a_3. \end{cases} \tag{7}$$

Formula (7) is a differential equation set with complex coefficients. Through solving the coupled equations as a function of impact parameter, we can obtain the probabilities of 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 by

$$\sigma = \int_0^\infty \int_0^\infty |a_3(b,v,t=\infty)|^2 \cdot F(v) \cdot 2\pi b \cdot \mathrm{d}b\,\mathrm{d}v. \tag{8}$$

The probability density function $F(\upsilon)$ can be represented by[24]

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

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

## 3. Application to Xe–Kr system

Xe–Kr system LICET process can be expressed by the following formula and figure 2 illustrates those energy levels relevant to the LICET process.

$$\mathrm{Xe}(4\mathrm{f}\ ^2[5/2]_2) + \mathrm{Kr}(4\mathrm{p}^6\ ^1\mathrm{S}_0) + \hbar\omega(538.753\ \mathrm{nm})$$
$$\rightarrow \mathrm{Xe}(5\mathrm{p}^6\ ^1\mathrm{S}_0) + \mathrm{Kr}(7\mathrm{d}\ ^2[7/2]_3).$$

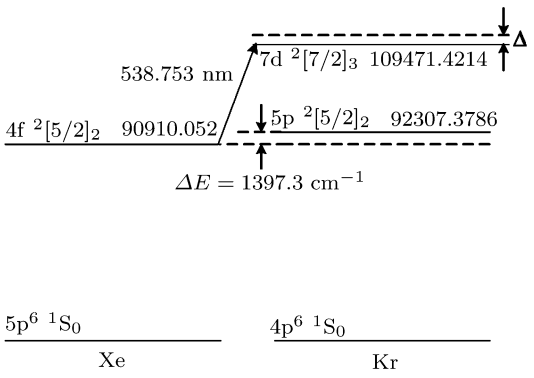



**Fig. 2.** The LICET transition in Sr–Ca system. The energy levels in the figure are given in $\mathrm{cm}^{-1}$.

The states of Xe–Kr compound system shown in Fig. 2 can be defined as

$$\begin{cases} |1\rangle = |\mathrm{Xe}(5\mathrm{p}^6\ ^1\mathrm{S}_0)\rangle|\mathrm{Kr}(4\mathrm{p}^6\ ^1\mathrm{S}_0)\rangle, \\ |2\rangle = |\mathrm{Xe}(4\mathrm{f}\ ^2[5/2]_2)\rangle|\mathrm{Kr}(5\mathrm{p}\ 2[5/2]_2)\rangle, \\ |3\rangle = |\mathrm{Xe}(5\mathrm{p}^6\ ^1\mathrm{S}_0)\rangle|\mathrm{Kr}(7\mathrm{d}\ ^2[7/2]_3)\rangle, \end{cases}$$

where the energy interval between state $|1\rangle$ and state $|2\rangle$ is $\omega_{21} = -1397.327\ \mathrm{cm}^{-1}$.

When the relative speed is 600 m/s, the laser detuning is 0 $\mathrm{cm}^{-1}$ and the impact parameters $b$ are for the two atoms equal to 0.2 nm and 0.25 nm respectively, the collisional transition probability $|a_3(t)|^2$ as a function of time $t$ in this Xe–Kr system is shown in Fig. 3. When $|t| \geq 2$ ps, $|a_3(t)|^2$ approaches to a definite value, which indicates that the laser-induced collision process occurs for $\sim 4$ ps, which is much shorter than the dipole–dipole LICET process.

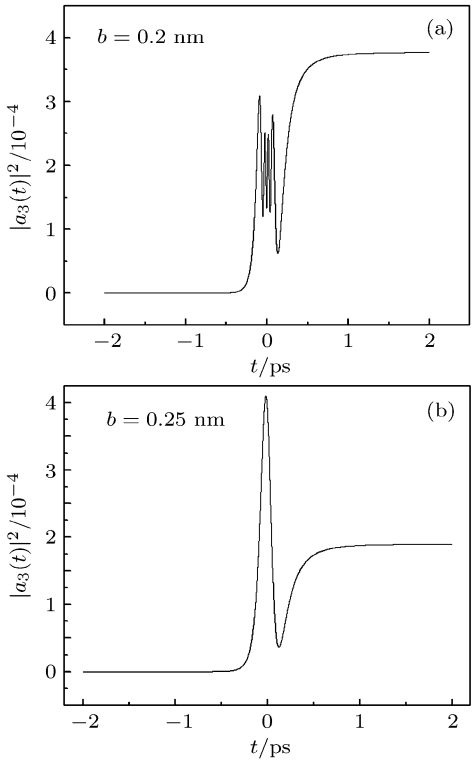



**Fig. 3.** The time dependence of laser-induced collision transition probability $|a_3(t)|^2$ for the Xe–Kr system: (a) $b = 0.2$ nm and (b) $b = 0.25$ nm.

Figure 4 shows the collisional transition probability as a function of impact parameter $b$. It appears

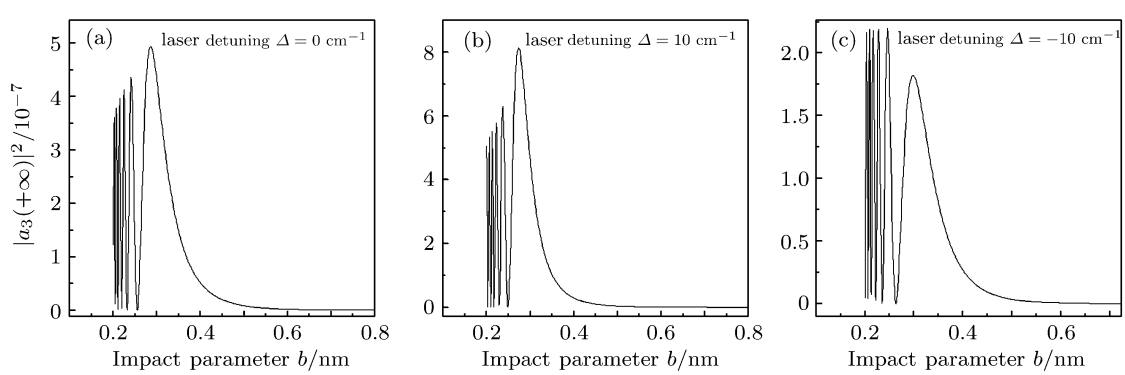



**Fig. 4.** Laser-induced collision transition probability $|a_3(+\infty)|^2$ as a function of impact parameter for the Xe–Kr system: (a) $\Delta = 0\ \mathrm{cm}^{-1}$, (b) $\Delta = 10\ \mathrm{cm}^{-1}$, and (c) $\Delta = -10\ \mathrm{cm}^{-1}$.

that $|a_3(+\infty)|^2$ oscillates as $b$ changes. The oscillation can be understood by the Landau–Zener theory of the effective curve crossing section. To the quasimolecule system formed in the collisional process, the argument of the exponent in the equation of motion for the probability amplitudes equals zero at some interatomic distance and the transition probability is only large for impact parameter less than that equal to interatomic distance. At a fixed impact parameter $a_3(t)$ accumulates during each of the two times that the crossing distances are near. And the adding and subtracting of the phased contributions at each of the two crossings lead to the oscillation of $|a_3(+\infty)|^2$.[9] It is also found that in the detuning case more of the contribution to the total collision cross section occurs at small impact parameter than in the strict resonance case. When laser detuning $\Delta > 0$, the maximum amplitude increases with increasing impact parameter and when $\Delta < 0$, amplitude maximum decreases as the impact parameter increases.

Figure 5 shows the LICET spectra for the Xe–Kr system at different temperatures. The profile of the excitation spectrum, markedly asymmetric, can be divided into two parts, the quasi-static and the anti-static wings. It can be seen that the variation in the LICET profile with laser detuning has a certain tunable range. The cross section $\sigma$ at fixed laser detuning becomes smaller as the system temperature increases.

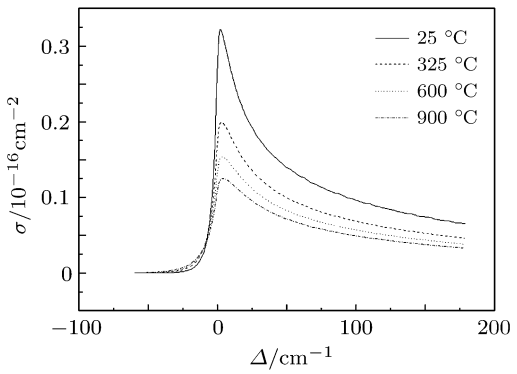



**Fig. 5.** LICET spectra for Sr–Ca system at various transfer laser intensities in weak field.

In contrast, our resulting spectrum has wider tunable range of an order of magnitude than the dipole–dipole LICET spectra.[9,15,16,26] When the laser field intensity is $1.0\times10^8$ V/m and the system temperature is 900 °C, the full width at half maximum (FWHM) is about 70 $\mathrm{cm}^{-1}$.

Furthermore, it is shown in Fig. 5 that the peak of the cross section profiles is not found when the transfer laser is strictly resonant (i.e. $\Delta = 0$ $\mathrm{cm}^{-1}$), but it is found when the transfer laser detunes from $\Delta = 0$ $\mathrm{cm}^{-1}$ in the quasi-static wing. And such a shift depends on the system temperature as shown below.

The significant variation of the spectrum profile with temperature $T$ is evident from Fig. 5. The peak position, FWHM and the profile of anti-static wing depend on temperature. It is found that in the dipole–dipole LICET system, the system temperature has no obvious effect on line shape.[20] It is interesting to observe that in the present system as the temperature is increased, the resulting spectrum is shifted in frequency towards the quasi-static wing and the full width at half peak of the profile becomes larger. Moreover, from Fig. 5 we can see that the anti-static wings become steeper as the temperature decreases and system temperature has the effect of increasing the transition probability in the quasi-static wing.

The shift of the profile of the peak and the peak cross section as functions of the system temperature $T$ are shown in Figs. 6 and 7. The peak cross section decreases rapidly with increasing temperature, while the shift of the line peak $\Delta_{\mathrm{peak}}$ increases as the system temperature increases.

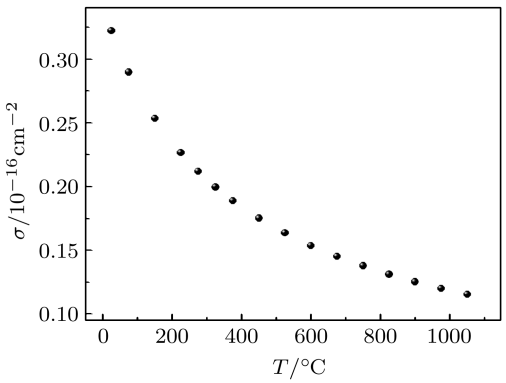



**Fig. 6.** The peak cross section as a function of transfer system temperature for the Xe–Kr system.

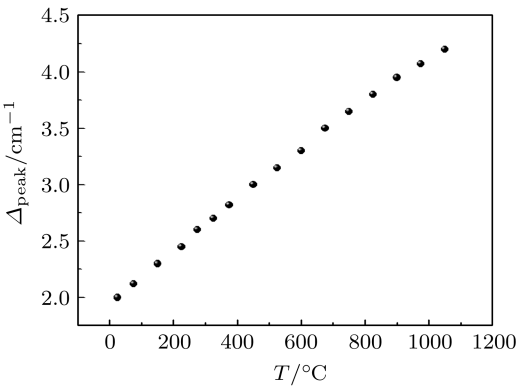



**Fig. 7.** Shift of the peak cross section as a function of system temperature for the Xe–Kr system in a strong field.

## 4. Conclusion

In this paper, in view of the multipole expansion interaction Hamiltonian, a three-state model for calculating the cross section of laser-induced quadrupole–quadrupole collisional energy transfer in Xe–Kr system is presented. 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 is obtained and the spectral profiles of LICET are calculated through immediate integrations. The results show that in the present system, the laser-induced collision process occurs for $\sim 4$ ps, which is much shorter than dipole–dipole LICET process. The spectra of laser-induced quadrupole–quadrupole collisional energy transfer in the Xe–Kr system have wider tunable range than the dipole–dipole LICET spectra. It is interesting to observe that the peak position and the spectrum profile depend on system temperature. As the system temperature increases, the resulting spectrum is shifted in frequency towards the quasi-static wing and the full width at half peak of the profile becomes larger, while in the previous dipole–dipole LICET systems the profile has no obvious effect on line shape at different system temperatures.

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