Far-wing study of laser-induced collisional energy transfer

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An experimental study of laser-induced collisional energy transfer in a mixture of europium and strontium vapors is reported. The profile of the excitation spectrum in the static wing has been measured with high accuracy and compared with available calculations, demonstrating that simple two-level models are inadequate to describe the transfer process. A comparison with the predictions of a three-level model assuming dipole-dipole interaction, recently developed by Bambini and Berman, shows for the first time agreement between theory and experiment.

The development of high-power tunable laser sources has prompted in the last decade the study of atomic transitions induced by the simultaneous action of a laser field and a collision. The processes, predicted by Yakovlenko *et al.*,[1] can be described as a reaction of the general form

$$A_i + B_i + \hbar\omega \rightarrow A_f + B_f \ , \tag{1}$$

where $A_{i,f}$ and $B_{i,f}$ are the internal states of the colliding atoms and ω is the laser frequency, close to the difference between the transition frequencies of the atoms. Determination of quasimolecular transient states of the colliding atoms, efficient excitation of high-lying states inaccessible by direct photon absorption, and laser switching of chemical reactions represent the main objectives of the study of laser-induced collisions.

The laser-induced collisional energy transfer (LICET), also designated as radiatively assisted inelastic collision (RAIC), involves an energy transfer from one atom to another with the simultaneous absorption of a photon. Since the first experimental demonstration by Harris and co-workers in a strontium-calcium mixture,[2] radiative collisions have been studied in various binary atomic systems, such as Eu-Sr,[3–5] Rb-K,[6] Na-Ca,[7] and Li-Sr.[8] Among the laser-induced processes, charge transfer,[9] pair absorption,[10] and Penning or associative ionization[11] have been investigated.

The cross section of a LICET process, which is energetically forbidden in the absence of laser field, is maximum when the laser frequency ω is resonant with the frequency ω_0 of the interatomic transition. The line profile, as a function of the laser detuning from line center $\Delta\omega = \omega - \omega_0$, is characterized by an asymmetric shape, showing a gradual falloff on one wing and an abrupt (exponential) falloff on the other. By analogy with the line-broadening theory, the LICET line profile can be divided into an impact and a static region, corresponding to small and large values of $\Delta\omega$, respectively. In the static wing, where the main contributions to the cross section arise from short-range collisions, the profile is strongly affected by the interatomic potentials. In this region, for $|\Delta\omega| \gg 1/\tau_c$, where τ_c is a typical collision time, the process can be described as an instantaneous transition between adiabatic quasimolecular states (quasistatic approximation). From the quasistatic approximation, assuming a dipole-dipole interaction, the LICET cross section in the static wing has been predicted to follow the asymptotic law $|\Delta\omega|^{-1/2}$.[12,13] An asymptotic behavior very close to the $-\frac{1}{2}$ power law is also provided by a numerical resolution of the time-dependent Schrödinger equation for the compound system (atoms+field), under the usual two-level approximation.[14]

High-resolution measurements of the LICET spectrum, performed by Brechignac *et al.* on the Eu-Sr system[4] and by Débarre on the Na-Ca system,[7] have shown that the detuning dependence of the cross section in the static wing can be fitted by a power law $|\Delta\omega|^{\alpha}$ with $\alpha = -0.85$ and $\alpha = -0.8$, respectively. The discrepancy with the theoretical predictions was ascribed to the influence of short-range interactions, including a breakdown of the linear trajectory hypothesis, or to a role played by near-resonant atomic levels.

Recently, a new analysis of the LICET process performed by Bambini and Berman for the Eu-Sr system,[15] has shown that the usual two-level approximation is inadequate to describe accurately the line profile in the far wing. In fact, when the laser detuning $\Delta\omega$ is comparable to the energy defect Δ of the interatomic transition, the initial and intermediate states of the compound system (see Fig. 1) are mixed by the collisional interaction and a three-level treatment is required to describe the transfer process. A similar effect was observed by Niemax in a study of the static wing absorption of the 459.5-nm Eu line collisionally broadened by Sr atoms.[16] Assuming a

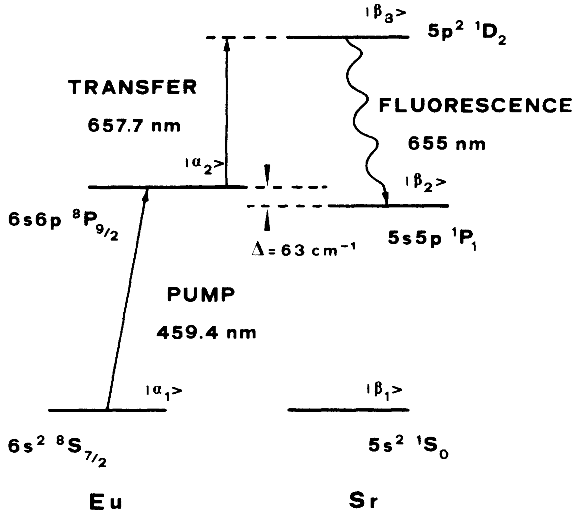

FIG. 1. Energy level diagram for the Eu-Sr laser-induced process. The compound states relevant to the transition are $|\alpha_2\rangle|\beta_1\rangle$ (initial state), $|\alpha_1\rangle|\beta_2\rangle$ (intermediate state), and $|\alpha_1\rangle|\beta_3\rangle$ (final state).

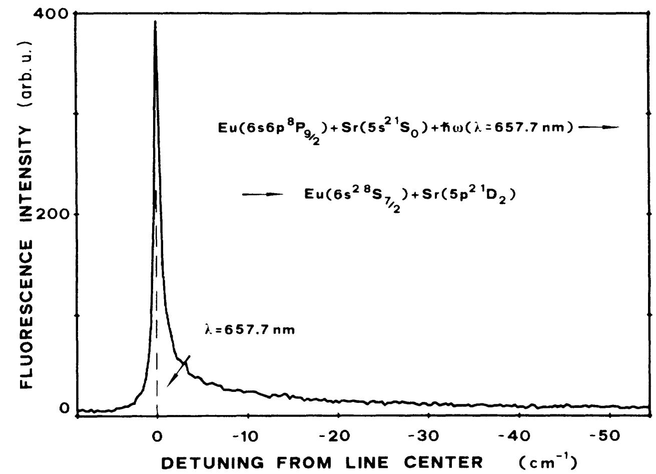

FIG. 2. Spectral profile of the Eu-Sr LICET process versus detuning of the transfer laser $\Delta\omega=\omega-\omega_0$. The spectrum peaks at the wavelength $\lambda=657.7$ nm, corresponding to the interatomic transition for separated atoms. For experimental conditions see text.

dipole-dipole interaction, the three-level model provides, at first order in the laser field amplitude, the following asymptotic law for the cross section in the static wing:

$$\sigma \propto |\Delta\omega|^{-1/2}(\Delta+|\Delta\omega|)^{-3/2}, \qquad (2)$$

where $\Delta=63$ cm^{-1} for the Eu-Sr system. This law reduces to the $-1/2$ power law predicted by the quasistatic approximation when $|\Delta\omega| \ll \Delta$ and approaches a -2 power law when $|\Delta\omega| \gg \Delta$.

In this paper a new high-resolution measurement of the far-wing profile for the Eu-Sr system is reported. The experimental setup, described in detail in a previous paper,[17] employed two dye lasers, pumped by the same nitrogen laser, providing 10–50 μJ, 2-nsec pulses with a bandwidth $\simeq 0.1$ cm^{-1}.[18]

The output of one laser (pump laser) was used to populate the Eu($6s6p\ ^8P_{9/2}$) state, while the output of the other laser, delayed by $T \simeq 16$ nsec in order to avoid the direct two-photon excitation of Sr, was used to induce the transfer process. The laser beams were focused into a heat-pipe oven providing the binary vapor mixture, with Eu number densities near 10^{16} atoms/cm^3 and Sr number densities near 10^{17} atoms/cm^3. The population of the final state was monitored by measuring the fluorescence from the Sr($5p^2\ ^1D_2$) level at $\lambda=655$ nm. Data acquisition, noise subtraction, monitoring of the energy of the laser pulses, and overall system control were performed by a computer.

A measurement of the LICET spectral profile is shown in Fig. 2. The line-core shape was found to be dependent on the intensity of the transfer laser, proving that the stimulated emission on the Sr($5p^2\,^1D_2$)$\rightarrow$Sr($5s5p\ ^1P_1$) transition played an important role in the final-state decay. However, the comparison between the line shapes measured at different laser intensities showed that, at the atomic densities of our experiment, stimulated emission effects were negligible for detunings larger than 5 cm^{-1}. The fluorescence signal measured on the antistatic (blue) side of the resonance revealed the presence of a background process, dependent on the pump laser alone. This background signal was probably due to a pump-laser excitation of high-lying europium levels followed by collisional transfer to strontium populating the Sr($5p^2\ ^1D_2$) state.

The far-wing profile measured in the detuning interval 6.3–53.2 cm^{-1} is shown in Fig. 3. The fine-tuning system, with a resolution better than 0.1 cm^{-1}, allowed the determination of the detuning from line center with an accuracy of ± 0.3 cm^{-1}. The average energy of the transfer laser, also presented in Fig. 3, remained constant during

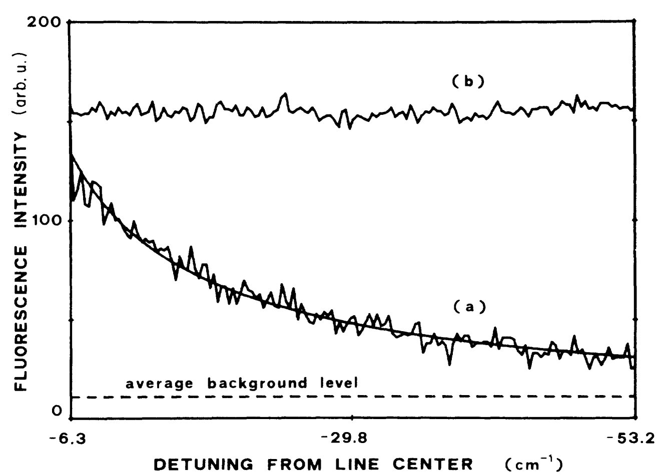

FIG. 3. (a) Line profile in the static wing measured in the detuning interval 6.3–53.2 cm^{-1}. Experimental points are averages over 16 laser pulses. The best fit of Eq. (2) is reported on the data. (b) Transfer laser energy measured during the frequency scanning. The average background level is represented as a dashed line.

the frequency scanning to within 1%.

In order to compare our results with the predictions of the quasistatic theory, the measured far-wing profile was fitted by a power law $|\Delta\omega|^{\alpha}$. The best straight line fitting the log/log plot of the recorded data was found to have a slope $\alpha=-0.79\pm0.05$. This value, consistent with the results of the previous high-resolution investigations, confirmed the discrepancy with the quasistatic law $|\Delta\omega|^{-1/2}$ predicted for a dipole-dipole interaction. The uncertainty in the α value arises from a combination of the statistical errors with the systematic errors mainly due to the inaccuracy of the detuning determination. When the fit was performed separately in the detuning intervals 6.3–29.8 cm^{-1} and 29.8–53.2 cm^{-1}, the slopes $\alpha=-0.72\pm0.05$ and $\alpha=-1.14\pm0.10$ were respectively obtained, proving that the measured profile can be more accurately described by a law with a variable slope.[17]

The analysis of our data showed that the accuracy of the measurement was adequate to the requirements for a quantitative test of theoretical models, making a comparison with the predictions of the three-level theory significant. Therefore, the measured profile in the overall detuning range was fitted with the theoretical law (2). From a two-parameter fit (including a normalization constant) an energy defect $\Delta=67\pm6$ cm^{-1} was estimated, to be compared to the value $\Delta=63$ cm^{-1} for the Eu-Sr interatomic transition. The corresponding line shape, shown in Fig. 3 as a continuous line through the experimental points, provides a good fit to the data with a small standard deviation. Since the detuning range explored in the experiment was comparable to the Eu-Sr energy defect, the asymptotic slope $\alpha=-2$, predicted by Eq. (2), could not be checked.

The results reported here provide an experimental evidence that the LICET cross section does not follow a simple power law in the static wing, since a significant slope change has been determined in a detuning interval $\simeq 50$ cm^{-1}. Moreover, the experimental results, consistent with the results of previous studies, are in agreement with the predictions of a theoretical model based on a three-level approximation. The deeper insight gained into the dynamics of radiative collisions stimulates further investigation of other types of dipole-dipole laser-induced collisions.

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