Experimental evidence of strong-field effects in light-induced collisional energy-transfer processes in europium and strontium atoms

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Strong-field effects in the light-induced collisional energy-transfer spectral profile were investigated for the europium-strontium binary mixture. When the laser amplitude is increased, the slope in the quasistatic wing changes, the spectral peak reaches saturation, and the spectral line shape loses asymmetry. These features are in fair to good agreement with predictions of existing theoretical models. However, no appreciable shift of the spectral peak was observed at intensities ranging from 23 MW/cm^2 up to 3.75 GW/cm^2, in contrast with the predictions of those models.

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INTRODUCTION

The light-induced collisional energy transfer (LICET) between two atoms is a process described by the reaction

$$A^* + B + \hbar\omega \rightarrow A + B^{**} , \tag{1}$$

where the asterisks denote excited or double-excited states of atoms A and B, and ω is the laser frequency, nearly resonant with the interatomic transition of frequency $\omega_0 = [E(B^{**}) - E(A^*)]/\hbar$; the reaction can be seen as a radiative transition in the transient molecule formed by the atoms during the collision.

This process was first proposed theoretically by Gudzenko and Yakovlenko [1]; in successive investigations, Lisitsa and Yakovlenko [2] gave an expression for the radiative cross section dependent on laser detuning. The predicted asymmetric line profile was then observed by Falcone *et al.* [3] and by Cahuzac and Toschek [4], by recording the fluorescence from the excited state of an atom.

High-resolution experimental studies in the weak-field regime were made by Brechignac, Cahuzac, and Toschek [5] and Debarré [6]; at increasing detunings of the laser field a falloff of the LICET profile was found, with a constant slope which was different from the one predicted by theory. The discrepancy was explained in successive theoretical papers [7,8], where the original two-level model was considered inadequate because of the strong collisional interaction of the atoms. An effective three-level model was proposed, which included an intermediate level of the quasimolecule. The cross section $\sigma(\omega)$ in the quasistatic wing was found to fall off to zero according to a double-slope behavior:

$$\sigma(\omega) \propto (|\omega - \omega_0|)^{-3/2} |\omega - \omega_0|^{-1/2} , \tag{2}$$

where Δ is the frequency difference between the initial and intermediate levels. This law was confirmed by Matera *et al.* [9] for the europium-strontium colliding partners, demonstrating the validity of the approximation of the theoretical model in the weak-field regime.

Successively, Dorsh, Geltman, and Toschek [10] extended the experimental investigations to higher field amplitudes. They observed the spectral peak to follow closely the law predicted by the high-field theory; moreover, the far-wing behavior of the line shape was found to be consistent with the double-slope law (2).

To our knowledge, no other experiments in the high-field regime have been reported since then, even if many experiments on different pairs were done in the past in the weak-field regime (see, for example, the review articles by Berman and Robinson [11] and Weiner [12]).

Recently, in a paper by Bambini *et al.* [13], the evaluation of the LICET spectrum was extended to the high-field regime; this was made both in the core, using a three-level system, and in the quasistatic wing, integrating an effective two-level system. The authors showed that the double-slope behavior loses significance at growing field amplitudes. Instead, a single-slope shape

$$\sigma(\omega) = c|\omega - \omega_0|^{-k} \tag{3}$$

was found to be in better agreement with the LICET line shape in the wing; in Eq. (3), c is a proportionality constant. The values of the fitting k parameter were evaluated for the europium-strontium configuration, in correspondence to several values of the field amplitude. The line core of the spectrum was also evaluated for the same values of laser intensities, and the position of the LICET peak was found to be shifted from the interatomic transition frequency ω_0 by a quantity which was interpreted as a mixed collisional and Stark shift. The values of the shift were confirmed by Geltman [14] in a successive work.

We extended the LICET experimental studies to the high-field regime for the europium-strontium pair. In this paper we present our results and the comparisons made with theory.

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EXPERIMENTAL SETUP

The reaction studied is described by the equation

$$\mathrm{Eu}(6s6p)^8P_{9/2}+\mathrm{Sr}(5s^2)^1S_0+\hbar\omega \rightarrow \mathrm{Eu}(6s^2)^8S_{7/2}+\mathrm{Sr}(5p^2)^1D_2 . \quad (4)$$

The configuration of atomic energy levels of europium and strontium atoms is shown in Fig. 1.

In the experiment, two homemade dye lasers were used; they were pumped by the second and third harmonic, respectively, of a Nd:YAG laser (where Nd:YAG denotes neodymium-doped yttrium-aluminum-garnet) equipped with a Gaussian coupled resonator and optimized for a 10-Hz repetition rate. The oscillators' beams of both dye lasers were at grazing incidence in the Moja configuration, with a four-prism beam expander; two amplification stages with transverse pumping were used in cascade, delivering 5-nsec full-width-at-half-maximum pulses with a bandwidth of 0.2 cm^{-1}. As in previous experiments [15,16], the two beams were made to counterpropagate and entered the crossed heat pipe, where the binary mixture of the metallic vapors was prepared, with a relative angle of 8°. The transfer laser was delayed with respect to the pump laser by an optical delay line of 20 nsec.

The europium atoms were prepared in the initial, excited state $(6s6p)^8P_{9/2}$ by the pump dye laser (the "blue" laser, Coumarin 460 diluted in methanol); LICET transitions were induced by the second dye laser, the transfer laser, [the "red" laser, organic dye 4-(dicyanomethylene)-2-methyl-6(*p*-dimethyl-aminostyryl)-4H-pyran (DCM) diluted in dimethyl sulfoxide (DMSO) tuning of the emission frequency about the resonance ω_0 of the interatomic transition $\mathrm{Eu}((6s6p)^8P_{9/2}\leftrightarrow\mathrm{Sr}(5p^2)^1D_2)$ was made by a computer-controlled multistep scanning system, with a step width of 0.043 cm^{-1}. Data were collected every fifth scanning step, i.e., at a resolution of 0.215 cm^{-1}, comparable to the emission bandwidth of the dye lasers.

The transfer laser was optimized for an output energy of 40 mJ. Single-pulse energy of the transfer dye laser was measured directly at the entrance of the heat pipe after the delay line. In the full scanning range (50 cm^{-1}) the energy was found to be stabilized to within 8%. The beam intensity was attenuated by means of calibrated neutral filters, and its spatial distribution was measured by means of a digitizing system equipped with a charge-coupled detector (CCD) solid-state camera interfaced with a computer.

Lenses with focal lengths of 240 and 800 mm were used to focus the transfer beam inside the crossed heat pipe. The shorter-focal-length lens was used when the LICET antistatic and quasistatic wings were detected, whereas the other focal length was used near the peak of the spectral line shape.

The fluorescence of the $\mathrm{Sr}(5p^2)^1D_2\rightarrow\mathrm{Sr}(5s5p)^1P_1$ transition at $\lambda=658$ nm was collected in a direction perpendicular to both laser beams; the signal was filtered by a 0.85-m double monochromator and detected by a high-gain photomultiplier with an S-11 photocathode; synchronous integration was performed with a 50-nsec gate duration. Data acquisition was performed by computer, averaging over ten pulses.

In two series of experiments, the temperature was set at 690 and 760 °C, respectively, and was stabilized within a few tenths of a degree. 10 mbar of argon buffer gas was used. Notwithstanding the presence of the buffer gas, vapor deposition was found against the cell windows. The effect was greatly reduced when strontium was put in the heat pipe 2–3 cm away from the center, where europium was still kept.

The energy of the pump dye laser was kept low (from hundreds of microjoules to a few millijoules) and the collimated beam was sent to the oven axis, in order to minimize the influence of processes induced by the high intensity of the blue-laser radiation. The pump laser was also detuned to the red side of the europium absorption resonance. In this way the LICET spectra were not influenced by the two-photon signal in correspondence with the peak. The two-photon signal was greatly reduced by the optical delay of the transfer laser. The tilted-beam geometry was found to reduce further the two-photon signal, because of the smaller overlapping region of the two laser beams inside the oven.

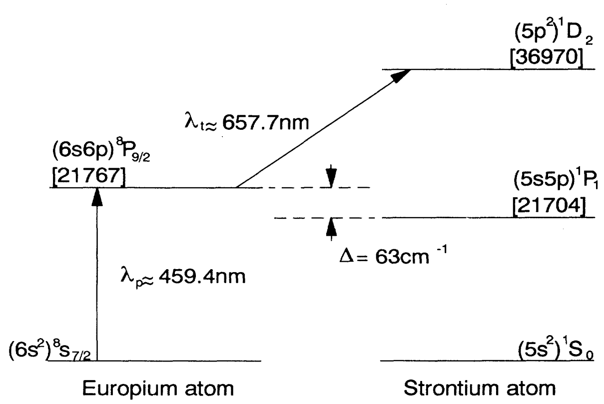

FIG. 1. Diagram of the energy levels relevant to the Eu-Sr LICET process. Level energies are given in cm^{-1}.

EXPERIMENTAL RESULTS

In the series of measurements reported in Fig. 2, the focusing lens of 240-mm focal length was used in conjunction with a working temperature of 690 °C. The transfer laser intensity was changed in the range from 50 $\mathrm{MW/cm}^2$ to 3.75 $\mathrm{GW/cm}^2$. From the laser intensity of 250 $\mathrm{MW/cm}^2$ and up the spectral peak was found in the frequency window next to the resonance and on the antistatic side, i.e., displaced by 0.215 cm^{-1} from resonance. Thus the shift was of the same order as the transfer laser bandwidth, but one order of magnitude smaller than the shift predicted by theoretical models.

In the antistatic wing, a background signal is present; at increasing laser intensities, it loses the aspect of a constant level signal. We made the hypothesis that the background signal is mainly due to the fluorescence following nonresonant excitations of the strontium D_2 state. This

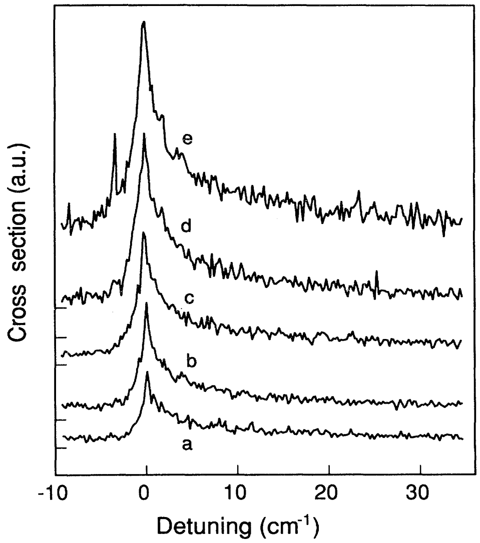

FIG. 2. Experimental line shapes obtained at 690 °C. The values of the transfer laser intensities I are (a) 50 MW/cm^2, (b) 125 MW/cm^2, (c) 250 MW/cm^2, (d) 750 MW/cm^2, (e) 3.75 GW/cm^2.

signal was assumed to be described by a Lorentzian function centered at the resonance fluorescence frequency ω_{32} of the strontium D_2-P_1 transition.

Analysis of the antistatic region

In the antistatic region the cross section is expected to decrease exponentially due to the absence of level crossing [13]. However, the experimental spectra did not manifest the shift predicted by the theory [13,14]. As predicted by Payne, Anderson, and Turner [17], the spatial and temporal distributions of the transfer laser pulse may reduce the shift and depress the cross section. In an attempt to describe the experimental behavior, we made a fit of this side of the spectrum by using the following function:

$$F(|\omega-\omega_p|)=Ae^{-l|\omega-\omega_p|}+B\frac{r^2}{r^2+|\omega-\omega_{32}|^2} \quad (5)$$

made up by the sum of exponential and Lorentzian functions; the latter function describes the background signal mentioned above. In general, we have found that it was possible to evaluate the exponential decay function from the peak of the LICET profile; hence, in (5), ω_p is the measured LICET peak frequency. However, in correspondence to the maximum value of the laser intensity (3.75 MW/cm^2), the antistatic region near the peak is not well reproduced by an exponential function up to the peak frequency. In this case, the fitting with (5) was made starting the evaluation from a frequency detuned 0.215 cm^{-1} from the LICET peak.

The function used to fit the data was considered to be accurate enough to describe the experimental behavior even if the Lorentzian parameters were obtained with large errors, due to the large detuning value $|\omega-\omega_{32}|$ at which the Lorentzian function was evaluated in the fit.

TABLE I. Values of l parameter with uncertainties (Δl) deduced from the fits of LICET experimental profiles with (5).

l	1.14	1.03	0.87	0.75	0.74
Δl	±0.04	±0.04	±0.03	±0.06	±0.06
I (MW/cm^2)	50	125	250	750	3750

The exponential decay rate l found by the fitting procedure decreases as the power density increases, while the parameter increases. The values for fit parameter l and the corresponding uncertainties Δl are reported in Table I as a function of transfer laser intensity I.

In Fig. 3 the fits for three cases only are reported after subtraction of the Lorentzian background and normalization. In Figs. 4 and 5 the numerical results of the fit are reported in conjunction with the experimental data for all the values of the transfer laser intensity.

Analysis of the quasistatic region

For each value of transfer laser intensity, the corresponding Lorentzian function found in the analysis of the antistatic region was also used as the background signal in the quasistatic part. After having subtracted the background signal from the experimental data, a best fit was done with the single-slope formula (3) using k and the proportionality constant c as fitting parameters. All the numerical results of the fits were obtained starting from a detuning of 5 cm^{-1} and are shown in Fig. 6 after normalization. From these fits, a slowly decreasing slope is obtained at increasing power density.

The numerical results of these fits are shown in Figs. 4 and 5, in which the background signal has been added again to the data. The values of the k parameter and of the relative experimental uncertainties Δk are reported in

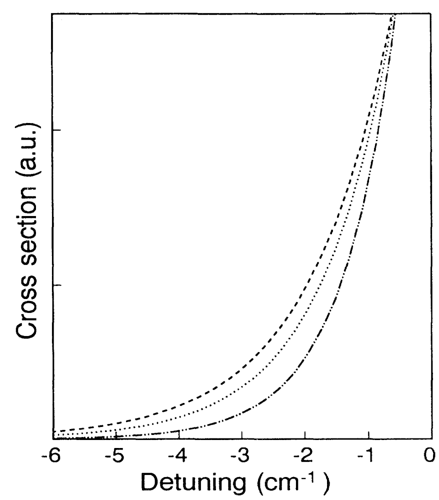

FIG. 3. Some of the fitting profiles obtained in the antistatic part shown after subtraction of the Lorentzian background contribution. The dot-dashed, dotted, and dashed fit profiles correspond to the values of transfer laser intensity (a), (c), and (e), respectively, as reported in Fig. 2. The values of l are (a) 1.14, (c) 0.87, (e) 0.74. All the curves are normalized to 0.43 cm^{-1}.

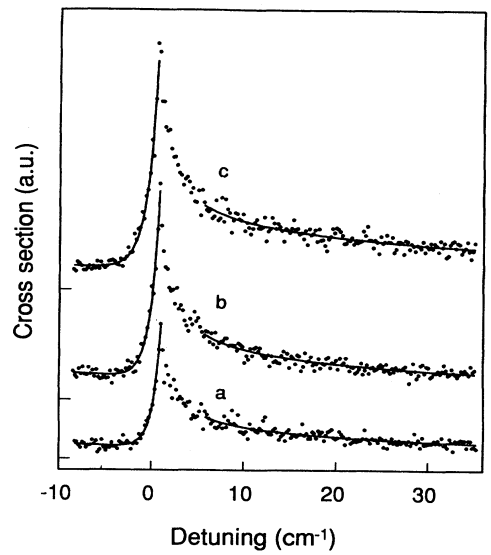

FIG. 4. Some of the experimental data shown in Fig. 2 with the corresponding fits. In the antistatic region (5) was used as a fitting function. In the quasistatic region the numerical results of the fits obtained with the single-slope law (3) were added to the signal background computed using the r and l values obtained in the analysis of the antistatic region. The transfer-laser intensities in cases (a)–(c) are the same as in the corresponding graphs of Fig. 2. The values of k are (a) 0.55, (b) 0.56, (c) 0.39.

Table II with the corresponding transfer laser intensities I.

At the high temperature of 760 °C, the entire LICET profile (Fig. 7) was also measured in correspondence with 514 MW/cm^2 and to 23 MW/ cm^2 using the 800-mm fo-

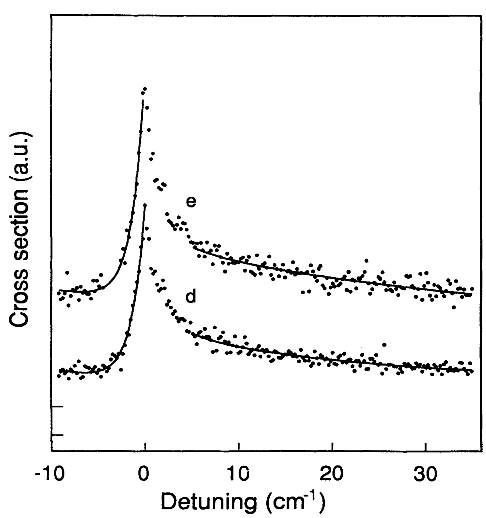

FIG. 5. Some of the experimental data shown in Fig. 2 with the corresponding fits. For case (e) formula (5) was used in the antistatic region analysis considering ω_p a frequency detuned 0.215 cm^{-1} from the effective peak. The transfer laser intensities in cases (d) and (e) are the same as in the corresponding graphs of Fig. 2. The values of k are (d) 0.38, (e) 0.24.

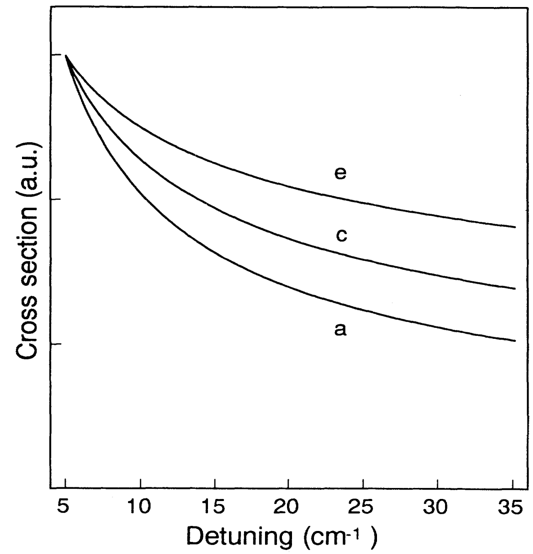

FIG. 6. Some of the single-slope fitting profiles obtained in the quasistatic wings after subtraction of the Lorentzian background to the experimental results. Normalization was made in correspondence with the detuning value of 5 cm^{-1}.

cal lens to focus the transfer laser inside the oven. From the fit of the quasistatic data obtained at the highest intensity, a k value of 0.4 ($\Delta k = \pm 0.02$) was obtained, consistent with the value obtained at the lower temperature, operating in the same range of the laser intensity. The value of k obtained at the lowest laser intensity was 0.52 ($\Delta k = \pm 0.03$), lower than expected from theory [13] (see also Table III).

Saturation behavior of the LICET peak

In order to get the behavior of the LICET peak versus the intensity of the laser field, we have operated at a temperature of 760 °C. Such a high temperature was chosen to enhance the signal even at lower laser intensities. This value was the one used in our previous LICET experiments in the low-field regime [16,18].

In Fig. 8 the peak values of fluorescence are plotted versus the transfer laser intensity. These plots refer to a series of measurements from 14.4 to 514 MW/cm^2, which were made by averaging over 30 shots. The Lorentzian background was subtracted from the data.

The power density at which saturation sets in is about 49 MW/cm^2, which corresponds to a Rabi frequency of $\sim$7.9 cm^{-1}. The straight lines reported in Fig. 8 result from best fits of experimental data. The full lines have slopes m (and corresponding experimental uncertainties Δm): $m = 0.96$ ($\Delta m = \pm 0.01$) and $m = 0.46$

TABLE II. Values of k parameter with uncertainties (Δk) deduced from the fit of LICET experimental profiles with (3).

k	0.55	0.56	0.39	0.38	0.24
Δk	±0.06	±0.04	±0.03	±0.03	±0.02
I (MW/cm^2)	50	125	250	750	3750

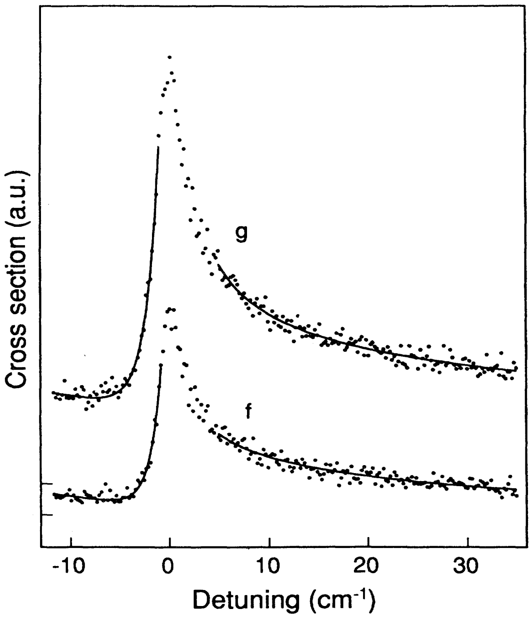

FIG. 7. Experimental profiles obtained at a working temperature of 760 °C in correspondence with two different laser intensities I: (f) $I = 13$ MW/cm^2, (g) $I = 514$ MW/cm^2. Experimental values in (f) are multiplied by a factor of 2. Best fits are shown in the antistatic and in the quasistatic wings. The slope values k are (f) 0.52, (g) 0.40. In the antistatic region formula (5) was used considering a frequency ω_p detuned in (f) 0.87 cm^{-1} and in (g) 1.08 cm^{-1}, from the effective peak.

($\Delta m = \pm 0.05$), respectively, beneath and beyond the saturation point. Over 120 MW/cm^2, the slope deviates from the $m = 0.46$ value, and the experimental data are shown fitted by a dashed line of slope $m = 0.26$ ($\Delta m = \pm 0.03$).

The peak values of the LICET profiles, as obtained in the other series of measurements performed at 690 °C (see Fig. 2) are also reported in Fig. 8 as triangles; these values were stripped of the Lorentzian background signal, too. Since in these measurements the lower atomic density produced lower signals, the peak values of this series of measurements were matched to the values in the previous series at 50 MW/cm^2. In the common range of intensities, the slopes were similar, showing a result which does not seem to be influenced by the temperature.

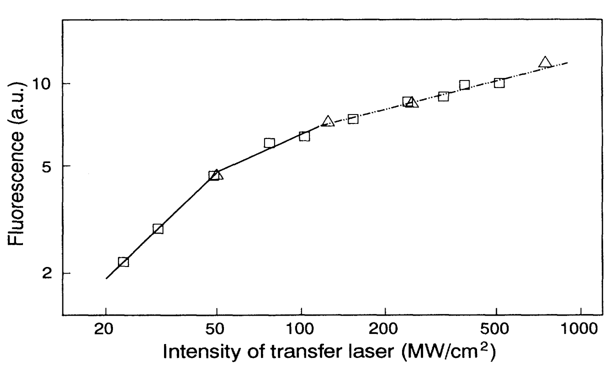

FIG. 8. Peak fluorescence vs transfer laser intensity. Open square symbols refer to the data collected averaging over 30 shots at a working temperature of 760 °C. The best fits of experimental data are shown (solid lines). The corresponding slope values are $m = 0.96$ and $m = 0.46$. The slope value $m = 0.26$ is obtained from the best fit beyond 120 MW/cm^2 (dashed line). Triangles refer to the peak values obtained averaging over 10 shots at a working temperature of 690 °C (Fig. 2).

THEORETICAL RESULTS

The theoretical model adopted in Ref. [13] for the analysis of the quasistatic wing was employed for evaluating the LICET profiles. The results were obtained by using a Monte Carlo simulation in which the proper statistical distributions of the relative speed and the impact parameter were taken into account [16,18]. The k values obtained by fitting the new curves with (3) in the same detuning range of the experimental reported profiles are given in Table III with the corresponding values of Rabi frequency and transfer laser intensities I. Also in Table III the l-parameter values obtained by fitting the theoretical curves in the antistatic region with an exponential function are reported, as are the values of the peak shift δ.

The same theoretical model [13] was used in another Monte Carlo simulation in which also the spatial and temporal distributions of laser pulse were taken into account. For this case, the computation of the quasistatic wing profile was made for a laser amplitude distribution with a maximum value of 137.5 MW/cm^2 (corresponding to a Rabi frequency of 13.25 cm^{-1}), in the same detuning range as in the previous evaluation. From the fit of the numerical results with(3), a value of 0.57 was obtained for k.

DISCUSSION AND CONCLUSION

In the graph of the LICET peak versus laser intensity, the experimental slopes 0.96 and 0.46 were found to be in fair to good agreement with the corresponding asymptotic theoretical values 1 and 0.5 [13]. Furthermore, saturation was found starting from a Rabi frequency consistent with the value reported by Bambini *et al.* [13]. At higher intensities (in our case, for intensities higher than 120 MW/cm^2), the measured slope's deviation from the expected saturation value of 0.5 was observed also by Dorsch, Geltman, and Toschek [10] in an experiment made on the Li-Sr pair. In his calculations, Geltman [14]

TABLE III. Theoretical values of l and k parameters obtained in correspondence with several Rabi frequencies and transfer laser intensities. The LICET peak shifts (δ) are also reported.

l	1.47	1.3	1.35	1.26
k	0.64	0.52	0.38	0.25
Rabi frequency (cm^{-1})	5.3	8	10.6	13.25
I (MW/cm^2)	22	50	88	137.5
δ (cm^{-1})	0.33	0.99	1.8	2.9

found a similar deviation, attributing the effect to the choice of a three-level model in the computation of the cross section in the core. The very existence of the fluorescence signal at larger detunings in the antistatic wing (see Fig. 3) was considered to be a high-intensity effect.

Also, the quasistatic slope values, deduced from the analysis of the experimental data, evidence a significant reduction of the k values versus laser intensities (see Table II). The slope value $k=0.55$ obtained in correspondence to a laser intensity of 50 MW/cm^2 may be considered consistent with the value found in the computation of the LICET spectrum relative to the same intensity value (see Table III).

However, at the intensity of 125 MW/cm^2, the experimental quasistatic slope ($k=0.56$) results are higher than the theoretical slope ($k=0.25$) obtained in correspondence with the transfer laser intensity of 137.5 MW/cm^2 and higher also than the theoretical slope ($k=0.38$) obtained in correspondence with 88 MW/cm^2. Furthermore, the experimental slope $k=0.38$ is obtained at an intensity five times higher than the value predicted by theory.

We thought that at intensities beyond the saturation onset of the peak, when curves computed at constant fields become more symmetric, the spatial and temporal distributions might influence the experimental behavior to the point of invalidating the comparison with the constant-field theoretical results even in the quasistatic wing. As expected, the slope value obtained when those distributions are taken into account is higher than the value obtained at constant field, owing to the influence of lower-intensity contributions. The value ($k=0.57$), obtained for an intensity distribution with a maximum value of 137 MW/cm^2 results in fair to good agreement with the experimental value ($k=0.56$) obtained in correspondence with 125 MW/cm^2. The predicted theoretical shift of the peak was not observed. Also, the l values found from the fits of the experimental profiles in the antistatic region (see Table I) are lower than the corresponding theoretical ones (see Table III), but again the experimental findings might have been influenced by the spatial and temporal distribution of the laser pulse. Computations are in progress to determine the line shape in the core with the spatial and temporal distribution also taken into account. From the results obtained up to now, a significant reduction of the shift was obtained, but the spectrum in the antistatic region is not well reproduced by an exponential decay function. Hence the disagreement with the theoretical results may indicate that the existing models are inadequate for the description of LICET processes under these conditions. For example, these models assume that an effective collision occurs at most for each atom in the mixture. But the strontium atom, left in its upper state after the *first* effective collision with the europium atom, may undergo *many* other long-range collisions with europium atoms, i.e., collisions that affect the core of the spectral profile. The effect of these collisions depends on the internal state of the newly encountered europium atoms. In particular, if the strontium atom collides with a europium atom in the ground state, then it may return to the ground state through an inverse LICET process stimulated by the field, thus lowering the fluorescence signal by which we measure the overall transition rate. The rate for such a process will depend on the particular experimental conditions in which the LICET profiles are measured. Also, other processes may affect the peak intensity. We suppose that, when the distributions of the laser field are introduced in new models that take into account more than one effective collision, the experimental features might be better reproduced. Otherwise, it will be necessary to take into account other more complicated schemes.

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