

Laser-induced collisional-energy transfer in thermal collisions of lithium and strontium

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Fluorescence of Li excited by laser light and simultaneous thermal Sr* collisions in the reaction $\text{Sr}(5p) + \text{Li}(2s) + \hbar\omega$ ($\lambda = 669.8 \text{ nm}$) $\rightarrow \text{Sr}(5s^2) + \text{Li}(4d)$ shows an asymmetric excitation spectrum of the energy transfer versus λ . The peak variation with light intensity demonstrates the strong-field regime and verifies current models. A change in the slope of the quasistatic wing agrees with the result of a recent three-level model.

I. INTRODUCTION

The transfer of energy of internal atomic excitation in the course of the collision of two thermal atoms is, in general, much less probable than an elastic collision. If, however, the energy defect between the actual state of one atom and a possible excited state of the other atom is matched by a light field of suitable frequency and field strength, that probability can assume dramatically large values. This "light-induced collisional-energy transfer" (LICET) was first proposed by Gudzenko and Yakovlenko¹ and detected by Harris *et al.* for the Ca-Sr collision pair.² Spectra of the transfer rate versus frequency of the inducing light, which could be fitted to calculated data, were demonstrated by Cahuzac and Toschek for Eu and Sr.³ Since then, a wealth of systems have been investigated, see, e.g., the review articles by Berman and Robinson and by Weiner.^{4,5} Some of the conspicuous features of the excitation spectra, in particular their nonsymmetric shape, have been explained in terms of conventional models for atomic collisions.

LICET is of fundamental interest since it allows one to study the collisional interaction of the atoms at the very time of the collision, as opposed to studies of the collision partners in the output channel.

We describe an experiment on LICET between lithium and strontium atoms, which has an unusually large excitation cross section due to the small frequency gap between the intermediate virtual level and a real excited level of the collisional system. We show that most, but not all, experimental results agree with predictions derived from current quasi-two-level models of the interaction. Of the few remaining features the most remarkable one, a change of slope in the quasistatic wing of the line profile, requires the application of a recent effective three-level model to include transient population of the near-resonant intermediate level.⁶

In Sec. II we briefly discuss the theoretical models used for comparison with LICET experiments. Section III characterizes the collision pair Li-Sr. We describe the Li-Sr experiment in Sec. IV and present the results in Sec. V. Data and models are critically assessed in the final section.

II. THEORETICAL MODEL

The theory of LICET has been discussed by a number of authors. For the purpose of understanding the present measurements we may consider the asymptotic quasi-molecular states which are designated by the asymptotic pairs of atomic states to which they go

$$\begin{aligned} |i\rangle &= \text{Li}(2s^2S)\text{Sr}(5p^1P), \\ |d\rangle &= \text{Li}(2p^2P)\text{Sr}(5p^1P), \\ |f\rangle &= \text{Li}(4d^2D)\text{Sr}(5s^2^1S). \end{aligned} \quad (1)$$

The basic theoretical approach is to consider this three-state system under the joint influence of the dipole-dipole interaction between the atoms and the applied laser field. The intermediate state $|d\rangle$ is important because its asymptotic energy lies only $\hbar\Delta\omega \approx 21 \text{ cm}^{-1}$ below the final state $|f\rangle$. The induced dipole-dipole interaction

$$H_d = \frac{1}{R^3}(\mathbf{r}_a \cdot \mathbf{r}_b - 3z_a z_b) \quad (2)$$

is seen to couple only $|d\rangle$ and $|f\rangle$ because it requires simultaneous dipole-allowed transitions in each of the atoms, while the laser interaction

$$H_r = (\mathbf{r}_a + \mathbf{r}_b) \cdot \mathbf{E}_0 \cos(\omega t) \quad (3)$$

couples $|i\rangle$ to $|d\rangle$ since only a single dipole transition is allowed, where \mathbf{E}_0 is the amplitude of the laser electric field.

A full numerical solution for the set of equations for the state amplitudes and the resulting LICET cross section, even under the assumption of straight-line classical paths, has not yet been carried out. There is a model in which the transition amplitude of $|d\rangle$ is first approximated by the known solution of the two-state ($|i\rangle$ and $|d\rangle$) laser-coupled problem, and then used to eliminate $|d\rangle$ from the three-state problem, leaving a two-state problem (in $|i\rangle$ and $|f\rangle$) to be solved. This resulting two-state problem was solved numerically (for a different physical case, Sr and Ca) by Harris and White,⁷ and by Yagisawa and Yagisawa⁸ using analytic methods. They find the result for the cross section at zero detuning in the

weak-field limit

$$\sigma = A \pi \left[\frac{\Delta C_6}{\hbar} \right]^{3/5} \frac{(E_0 \mu_{B_2})^2}{4 \hbar^2 \Delta \omega} v^{-8/5}, \quad (4)$$

where $\Delta C_6 = 4(\mu_{A_1} \mu_{B_1})^2 / 3 \hbar \Delta \omega$, and the two calculations lead to different values for the constant A . Harris and White find $A = 5.9$ and Yagisawa and Yagisawa find $A = 0.87$. For the present case μ_{A_1} is the atomic dipole matrix element for $\text{Sr}(5p - 5s)$, while μ_{B_1} and μ_{B_2} are those for $\text{Li}(2p - 4d)$ and $\text{Li}(2s - 2p)$, respectively. Furthermore, both treatments give the frequency dependence in the quasistatic wing of the line to be $(\delta\omega)^{-1/2}$. The quasistatic wing is that one for which there is a crossing of the dressed potential energy curves of the quasimolecule, i.e., for which $E_i(R) + \hbar\omega = E_f(R)$, where R is the separation of the colliding atoms, or for

$$\delta\omega = \omega - [E_f(\infty) - E_i(\infty)] > 0$$

in the present case since the difference of the van der Waals coefficients $\Delta C_6 = C_6^{(f)} - C_6^{(i)}$ is positive for states $|f\rangle$ and $|i\rangle$ in (1).

A simple way to understand this shape is on the basis of the quasistatic law for photon absorption by a pair of atoms. For an atomic pair at separation R , having effective dipole moment $\mu(R)$ and energy separation $\Delta E(R)$, the cross section for photon absorption is governed by⁹

$$\sigma(\delta\omega) \sim R^2 |\mu(R)|^2 \left/ \left| \frac{d\Delta E(R)}{dR} \right| \right., \quad (5)$$

where the energy resonance condition requires that the photon is absorbed at a given R such that $\hbar\omega = \Delta E(R)$. In the present LICET problem, $\mu(R) \sim 1/R^3$, $\Delta E = \Delta C_6/R^6$, which leads to $\sigma(\delta\omega) \sim (\delta\omega)^{-1/2}$. Gallagher and Holstein⁹ have treated the LICET problem in the spirit of line broadening theory, evaluating the collision-induced absorption for a system described by initial and final adiabatic potential curves and true molecular wave functions. They produced a general line shape function for the van der Waals quasimolecular potential which is seen to go smoothly into the above quasistatic form for large values of $\delta\omega$.

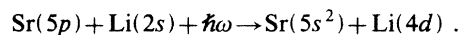
A strong signal model of LICET has been described by Payne, Anderson, and Turner.¹⁰ A recent treatment of this problem by Geltman¹¹ used first-order perturbation theory to evaluate the transition between the quasimolecular states $|i\rangle$ and $|f\rangle$ under the influence of the laser field. As is the case in first-order perturbation theory, there is no explicit appearance of the intermediate state $|d\rangle$, but it does make its presence felt in the evaluation of the van der Waals constants used in the adiabatic energies $E(R)$. In fact it is the smallness of $\hbar\Delta\omega = E_f(\infty) - E_d(\infty) = 21 \text{ cm}^{-1}$ which leads to the very large value $C_6^{(f)} = 1.30 \times 10^5 \text{ a.u.}$, which makes this particular cross section a large one [see Eq. (4), where $\mu_{B_2}^2 \mu_{A_1}^2 / \Delta\omega \sim C_6^{(f)}$]. The result of this treatment also gives a quasistatic wing dependence of $(\delta\omega)^{-1/2}$ in the low-intensity limit. This method allows the calculation of

LICET cross sections even with very intense laser fields when care is taken to ensure unitarity in a reasonable physical way. It is found that the very asymmetric shape for $\sigma(\delta\omega)$ in the low-intensity limit goes over into a symmetric shape of increasing peak cross section and decreasing FWHM as the intensity is increased, in agreement with the findings of Harris and White⁷ and of Payne, Anderson, and Turner.¹⁰

A recent paper by Bambini and Berman⁶ on the three-state system comes to the conclusion that the quasistatic wing behavior $(\delta\omega)^{-1/2}$ should be replaced by $(\delta\omega)^{-1/2}(\omega_{fd} + \delta\omega)^{-3/2}$. This model differs from the result of the other treatments in so far as the real intermediate level is not immediately eliminated such that it may contain transient population in the transfer process. It turns out that the Bambini-Berman shape is quite consistent with our present data, as will be shown later.

III. THE COLLISION PAIR

We study collisions of lithium ground-state atoms with strontium atoms in the $5p$ resonance state in the presence of light. The light frequency is such as to permit the excitation of the lithium atoms to the $4d$ level according to the reaction



Relevant parts of the level scheme are shown in Fig. 1. The system is characterized by a small energy defect of the resonant transfer light quantum from the resonance transition energy in lithium,

$$(\omega_0 - \omega_{2s-2p}) / 2\pi c = \Delta\tilde{\nu} = 21 \text{ cm}^{-1},$$

where $\omega = \omega_0 + 2\pi c \delta\tilde{\nu}$, and $2\pi c / \omega_0 = 669.82 \text{ nm}$.

The characteristic radial dipole moment matrix elements for the collision pair are

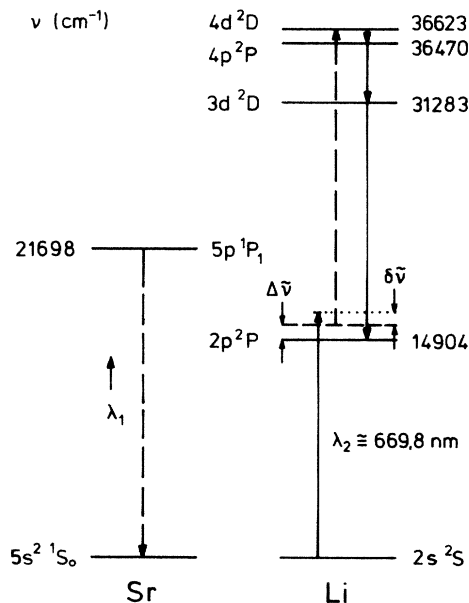


FIG. 1. Simplified level schemes of Sr and Li.

$$(5s \mid r \mid 5p)_{Sr} = 3.82 \text{ a.u.},$$

$$(2p \mid r \mid 4d)_{Li} = 1.70 \text{ a.u.},$$

$$(2s \mid r \mid 2p)_{Li} = 4.08 \text{ a.u.},$$

which are based on the best available oscillator strengths,¹² and $\bar{v} = 1760 \text{ ms}^{-1}$ for a relative velocity at 700°C. Using these in (4) gives the zero-detuning cross sections of

$$\sigma(\delta\omega=0) = 0.25 \times 10^{-13} \text{ cm}^2 \times I \text{ (in MW cm}^{-2}\text{)}$$

and

$$0.037 \times 10^{-13} \text{ cm}^2 \times I \text{ (in MW cm}^{-2}\text{)}$$

using $A = 5.9$ (Harris and White⁷) and 0.87 (Yagisawa and Yagisawa⁸), respectively. The calculation of Geltman using modified Bates-Damgaard radial matrix elements gave the weak-field result

$$\sigma(\delta\omega=0) = 2.23 \times 10^{-13} \text{ cm}^2 \times I \text{ (in MW cm}^{-2}\text{)}.$$

If the same best available oscillator strengths were used in that calculation the result would be changed to $1.80 \times 10^{-13} \text{ cm}^2 [I \text{ (MW cm}^{-2}\text{)}]$. These may be compared with the experimental estimate of Zhang, Nikolaus, and Toschek,¹³

$$\sigma(\delta\omega=0) \cong 2 \times 10^{-13} \text{ cm}^2 \times I \text{ (in MW cm}^{-2}\text{)}.$$

The critical light intensity marking equality of collisional and radiative coupling is 930 kW cm^{-2} for the present system, according to Ref. 1, and 700 kW cm^{-2} from Ref. 11. This level is easily achievable with pulsed dye lasers and permits one to study strong-field effects. The vapor pressure of Li and Sr varies over the temperature range of interest almost identically. Thus the relative concentration does not vary substantially, and the evaluation of the observed spectra is facilitated.

IV. EXPERIMENTAL

The experiment is an advanced version of a previous preliminary one,¹³ with better detection sensitivity, variable detection geometry, and improved data processing. A scheme of the experimental setup is shown in Fig. 2. The metal vapor was contained in a 20-cm long, temperature-controlled heat-pipe oven,¹⁴ either of conventional cylindrical geometry (internal diameter 2.8 cm), or of cross type (internal diameter 2.0 cm). Argon at 10 mbar was used as buffer gas. The observed fluorescence signals did not depend on the buffer gas pressure from 2 through 20 mbar. The temperature was set to values in the range 620–800°C, corresponding to the range of density between 10^{15} cm^{-3} and $2 \times 10^{16} \text{ cm}^{-3}$.

The vapor was irradiated by light pulses from two N_2 laser-pumped dye lasers. Dye laser 1, the "pump" laser, is of Littman type;¹⁵ its light excites Sr atoms to the $5p \ ^1P_1$ resonance level. Pulse width, peak power, and emission bandwidth of this laser are 3.4 ns, 5 kW, and $0.02 \text{ nm} \cong 0.9 \text{ cm}^{-1}$, respectively. Laser 2, the "transfer" laser, is of Hänisch type;¹⁶ its light mediates the energy transfer from Sr to Li. Its corresponding characteristics

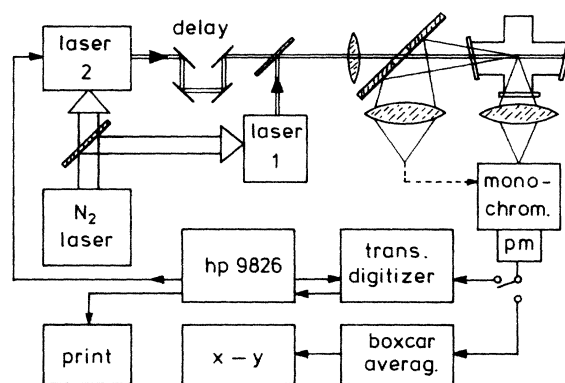


FIG. 2. Experimental setup.

are 3.8 ns, 20 kW, and $0.02 \text{ nm} \cong 0.6 \text{ cm}^{-1}$. The pulses of the second laser were delayed by a preselected time interval with respect to the pulses from the first laser. Having been coaxially combined, the two beams were focused, by a lens of 15 or 20 cm focal length into the heat pipe oven. With the transfer detection, fluorescence was observed from the center of the cross oven, but 5 cm off the beam focus. With axial detection, the fluorescence light picked up by a pierced mirror at 45° inclination is predominantly from the focal region. In both cases, the fluorescence light at 610 nm corresponding to the $Li(3d \ ^2D - 2p \ ^2P)$ line was narrow-band filtered by a 1-m Czerny-Turner monochromator and recorded by a low-noise photomultiplier. The electric signal was processed by a boxcar averager and x-y recorder (with the axial detection), or else by a fast transient digitizer (Tektronix 7912), controlled by a microcomputer (HP 9826) and a printer. In contrast with the boxcar averager, which is a pulse-sampling device, the transient digitizer records *individual* pulses. The repetition rate is limited to 12 pulses per s by the data transfer; actually it was 10 pulses per s.

The emission wavelength of the transfer laser was made to step across the LICET line. At one setting, 50 fluorescence pulses were integrated, corresponding to the effective integration time 5 s. The stepwidth was 0.25 cm^{-1} and the scan speed $2 \times 10^{-3} \text{ nm/s}$. Some of the recorded spectra have been numerically smoothed by averaging over neighboring data points.

The boxcar averager allowed recording at 20 Hz repetition rate, with the laser stepwidth 0.125 cm^{-1} at 1 Hz step frequency, corresponding to a scan speed $5 \times 10^{-3} \text{ nm/s}$. The actual length of the fluorescence pulses is estimated to be about 50 ns, taking into account the contributing steps in the decay cascade. In contrast, the signal output pulse of the photomultiplier showed a 50-ns long leading edge determined by the pm time constant and a 200-ns long trailing edge from the decay of fluorescence excited by spurious processes. We have checked that maximum contrast of the LICET signal versus background occurred in the leading edge. Thus the detection window was selected to start 3 ns past the peak of the transfer light pulse with 25 ns width.

The pump laser was tuned to resonance with the Sr

$5p\ ^1P_1$ level. Since it almost matches the $2p\ ^2P-4d\ ^2D$ transition of lithium—the energy defect is only 21 cm^{-1} —direct two-photon excitation of Li is comparable in probability with LICET. Thus a superposition of the two contributions to the total signal is observed (see Fig. 3). Note that the two-photon signal shifts in accord with the tuning of the pump laser. In order to separate the LICET signal, the pulses of the transfer laser 2 were time delayed by more than a pulse length against the pulses of pump laser 1 (Ref. 3). The delay time was measured with a photodiode of 5 GHz bandwidth. This delay is possible since the $5p$ level of Sr lives long enough, due to radiation trapping, to maintain collision-induced excitation upon the arrival of the transfer pulse. Even with pulse delay, a background signal appears. It is dominated by two components that are present with only one of the light pulses, 1 or 2, applied.

(1) With pump light only present, multistep collisional processes can lead to the excitation of Li $3d$ and the subsequent fluorescence. These processes form a spectrally constant background signal, which has been subtracted in the evaluation of the experimental data. This background increases as the square of the vapor density (see Fig. 4).

(2) With transfer light only present, another background contribution appears which increases toward the red end of the spectra. It results from off-resonant excita-

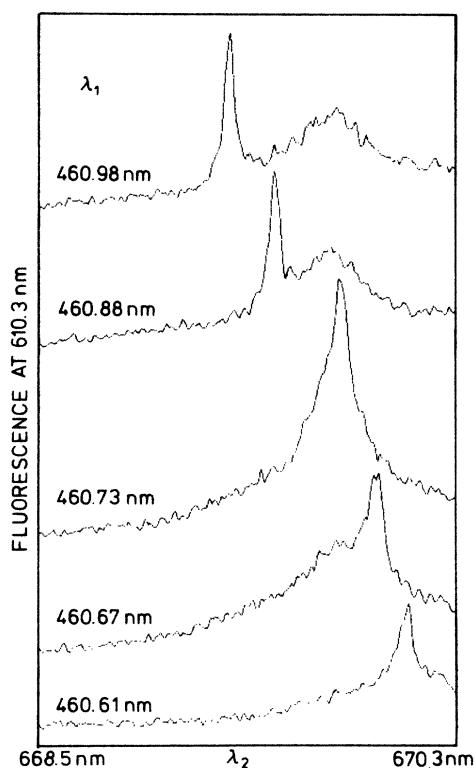


FIG. 3. Superposition of two-photon direct excitation (narrow resonance) and LICET excitation of Li $4d$ level with time overlap ($\tau=4\text{ ns}$) of pump and transfer light pulses. $T=700^\circ\text{C}$. The data are a weighted numerical average over five channels corresponding to 0.05 nm .

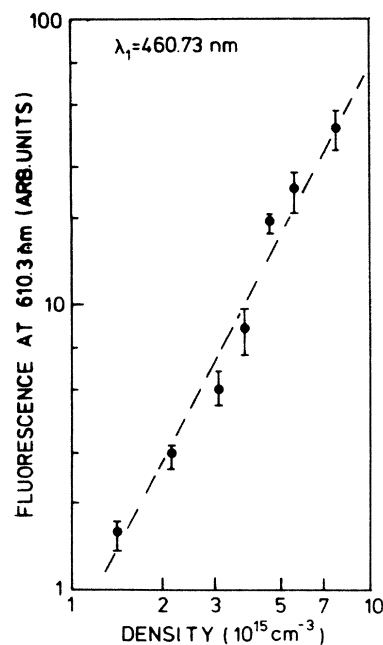


FIG. 4. Fluorescence from Li $3d$ atoms excited by collisions and with pump light only vs vapor density.

tion of Li $2p$ followed by Raman-like or collisionally aided excitation of the Li $3d$ level. This background component can be thoroughly fitted by a Lorentzian centered at the Li $2s-2p$ resonance at 670.78 nm . When using the transient digitizer, this component was recorded in addition to each individual spectrum and numerically subtracted.

With transverse detection, the 15-cm laser-focusing lens was placed 20 cm in front of the center of the oven. Thus at this region of observation, the average diameter of the illuminated cone is 1.6 mm , corresponding to the peak intensity 2.4 MW/cm^2 of the transfer pulse. Longitudinal collection of the fluorescence was combined by imaging the focal region inside the oven to the input slit of the monochromator. Accordingly, the effective intensity is much higher. The focal diameter of the laser beam was measured to be $200\ \mu\text{m}$, since higher order transverse laser modes are excited. We estimate the nominal maximum peak intensity as 120 MW/cm^2 . However, the distribution of the light-power density across the focus may be modified in this situation by nonlinear interaction with the vapor, leaving this estimate considerably more uncertain than the determination of the off-focus local intensity.

V. RESULTS

Spectra of the excitation of fluorescence at the Li $3d-2p$ line by the collisional transfer upon interaction with the scanned transfer light are shown in Fig. 5. Direct two-photon absorption is avoided by the 9-ns delay of the transfer light pulse, and only the LICET line shows up. The signal increases with vapor density, which is controlled by the temperature, but much less than the

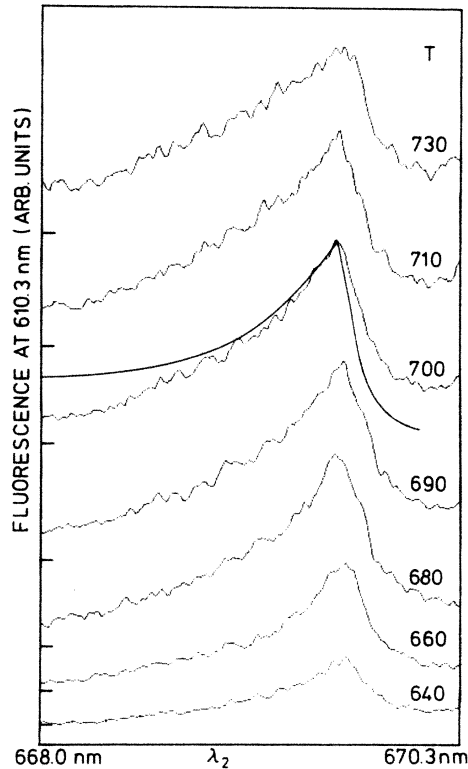


FIG. 5. LICET excitation of Li 4d monitored by fluorescence of Li 3d vs wavelength of transfer laser. Stepwidth 0.01 nm; 50 pulses per data point; delay $\tau=9$ ns. Background from transfer laser only subtracted. Pump laser $\lambda_1=460.73$ nm; $I_2=2.4$ MW/cm². The zero level is marked for each trace. Temperature in centigrade. Data average as in Fig. 3.

product of the densities. This variation is in qualitative agreement with light absorption before the interaction zone, although the effect is considerably larger than expected from the coefficients of absorption.

The spectra corresponding to the weak-field regime show the anticipated asymmetry with the quasistatic wing to the blue. The peak is located at the resonance wavelength for infinite separation of the Li and Sr atoms. The recordings match the calculated spectra, within 10%.

Corresponding excitation spectra, at various intensity values of the transfer light, are shown in Fig. 6 for transverse detection. The intensity range covers the weak-field regime and extends slightly into the strong-field regime. Note that neither the absolute linewidth nor the asymmetry of the line shape decreases with increasing intensity as would be inferred from the current theoretical models.^{10,11}

Observations at higher intensity values with axial fluorescence detection are shown in Fig. 7. The Lorentzian background from Li 2p excitation is fitted with the solid line. From the widths of the quasistatic wing (a) and antistatic wing (b), the asymmetry is determined as $(a-b)/(a+b)$. It is approximately constant, upon increasing intensity of the transfer light, for this observation as well as for transverse detection of fluorescence.

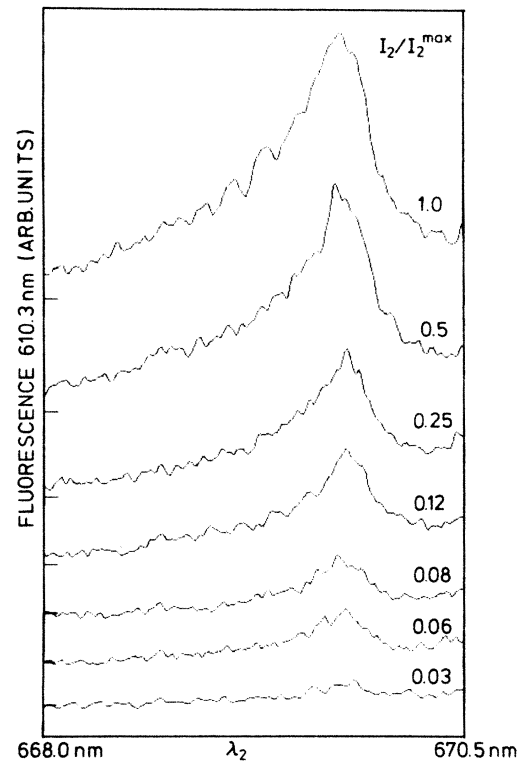


FIG. 6. LICET excitation of Li 4d monitored by fluorescence of Li 3d vs wavelength of transfer laser. $T=690^\circ\text{C}$; $I_2^{\text{max}}=2.4$ MW/cm². Other parameters as in Fig. 5.

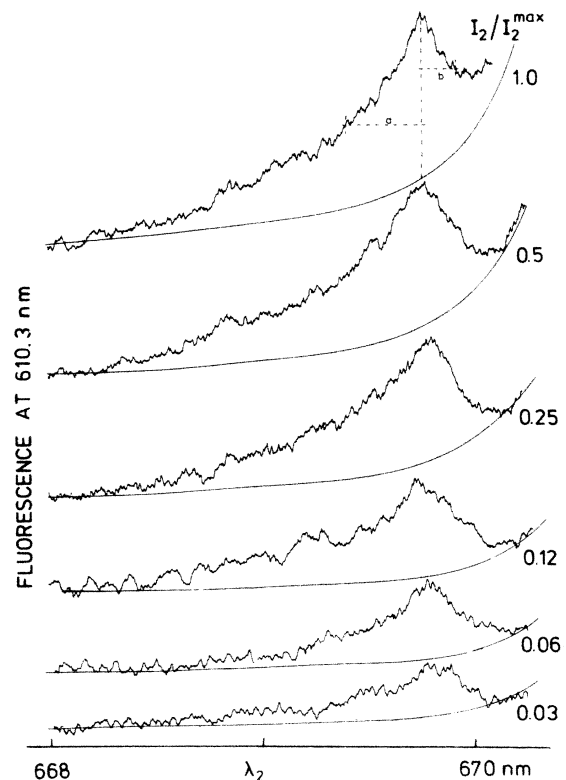


FIG. 7. Fluorescence of Li 3d recorded in axial direction with boxcar averager. $I_2^{\text{max}} \approx 120$ MW/cm². Background from stepwise excitation in the wing of the Li resonance line by pump laser.

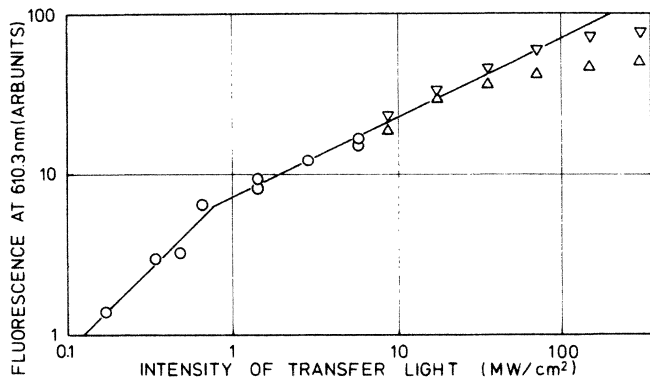


FIG. 8. Peak amplitude of LICET excitation vs transfer light intensity. The absolute scales of the experimental data are fitted. A tentative calibration of the focal power density showed about 2.5 times smaller values. The high-intensity data (axial detection) have been corrected for a flat (∇) or Lorentzian (Δ) background. Solid lines have slopes 1 and $\frac{1}{2}$.

The peak signal amplitude, as a function of transfer light intensity, is given in Fig. 8. Here, the absolute scale of the high-intensity data from axial detection has been fitted to the scale of the other data. Note the satisfactory agreement with the result of the theoretical model. The transition from a weak- to a strong-field regime is obvious, the two regimes characterized by slopes 1 and $\frac{1}{2}$, respectively.

Close inspection of excitation spectra as in Figs. 5–7 shows that the quasistatic wing falls off in proportion to

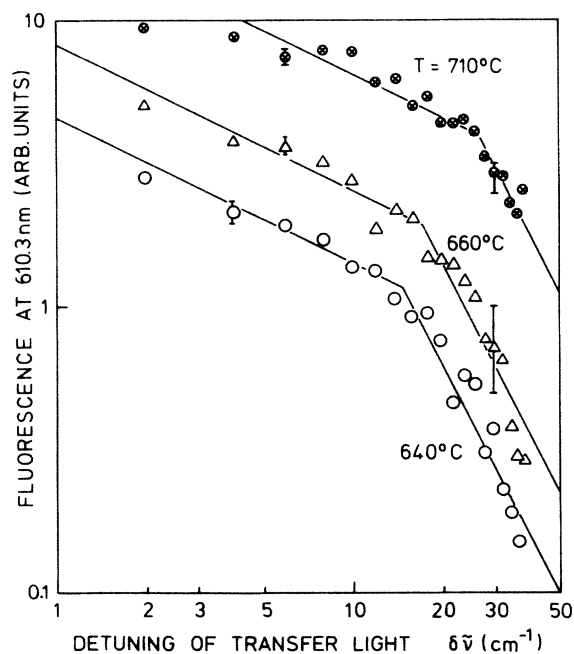


FIG. 9. Large-scale display of LICET-generated fluorescence at 610 nm in the quasistatic wing for three vapor-density values. Lines show slopes $-\frac{1}{2}$ and -2 . $\lambda_1 = 460.73$ nm; $I_2 = 2.4$ MW/cm².

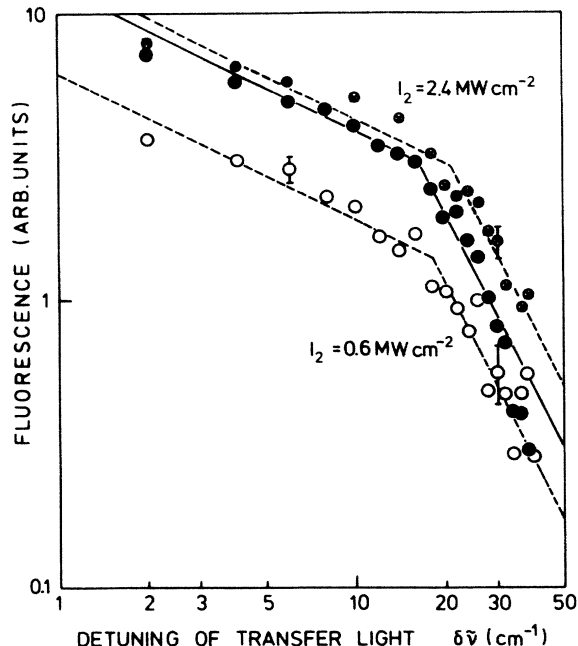


FIG. 10. Large-scale display of LICET-generated fluorescence in the quasistatic wing at 610.3 nm, $T = 690^\circ\text{C}$ (\times , \circ , ---), and at 460.2 nm, $T = 700^\circ\text{C}$ (\bullet , —). Lines show slopes $-\frac{1}{2}$ and -2 ; $\lambda_1 = 460.73$ nm.

$(\omega - \omega_0)^{-1/2}$, as it is expected for collisional dipole-dipole interaction.¹⁷ However, this anticipated behavior holds only up to a detuning of about 20 cm^{-1} . Beyond, the roll off is much more pronounced with the exponent being at least -2 . This is demonstrated in Figs. 9 and 10, where numerical data from digitizer recordings are displayed on a blown up scale. The conspicuous change of slopes moves to larger detuning values with increased vapor density and temperature (Fig. 9). It is also observed at widely differing intensity levels of the transfer light and with excitation spectra of the Li $4d - 2p$ fluorescence at 460.2 nm (Fig. 10).

VI. DISCUSSION AND CONCLUSIONS

We find that our present observations are in substantial agreement with the various theories of the LICET process, but there are still some issues which require further elucidation before full understanding can be obtained.

(a) The asymmetric line shape with the extended wing in the direction of positive detuning ($\delta\omega > 0$) is consistent with the calculated $\Delta C_6 > 0$.

(b) The maximum occurs at $\delta\omega = 0$ as expected, since the powers required for an observable Stark shift are much higher than those used in these measurements.

(c) The observations show the expected linearity of the maximum cross section as a function of transfer laser intensity at lower intensities, and the break to $I^{1/2}$ behavior. This is shown in Fig. 8, where the data are compared with the calculated result of Geltman.¹¹ In that theoretical work it is shown that the high-intensity behavior at any detuning $\delta\omega$ has the form

$$\sigma(\delta\omega) \sim I^{\nu(|\delta\omega|)},$$

where the exponent $\nu(|\delta\omega|)$ is a decreasing function of $|\delta\omega|$ starting at $\nu(0) = \frac{1}{2}$. Thus for any fixed observational resolution $(\Delta\omega)_{\text{exp}}$, the effective cross section at "zero" detuning will include some components for which $\nu < \frac{1}{2}$, and this dispersion would lead to an eventual downward departure from pure $I^{1/2}$ behavior, as is observed.

(d) The measured magnitude of the cross section at $\delta\omega = 0$ is $2 \times 10^{-13} \text{ cm}^2 \times I$ (in MW cm^{-2}) at the lower intensities, in good agreement with the calculated $2.23 \times 10^{-13} \text{ cm}^2 \times I$ (in MW cm^{-2}).

(e) The observations that linewidth and asymmetry increase with increasing intensity are somewhat inconsistent with the theoretical results of Payne, Anderson, and Turner¹⁰ and those of Geltman.¹¹ Those calculations show a uniform narrowing of the cross-section line shape as well as a trend toward symmetry as the laser intensity is increased.

(f) The departure from $(\delta\omega)^{-1/2}$ behavior at detunings of about 20 cm^{-1} as seen in the data of Figs. 9 and 10 are unexpected on the basis of quasistatic theory. If such a departure were attributed to departure of the difference

potential curves from pure van der Waals behavior, then such a departure would have to occur at $R \cong 34 \text{ a.u.} = 18 \text{ \AA}$. It is difficult to imagine the reason for such a departure at such large interatomic separations.

On the other hand, the new Bambini-Berman theory does predict the quasistatic wing dependence $(\delta\omega)^{-1/2} (\omega_{fd} + \delta\omega)^{-3/2}$, which would lead to a change from $(\delta\omega)^{-1/2}$ behavior to $(\delta\omega)^{-2}$ behavior for $\delta\omega > \omega_{fd}$. For the present pair of atoms and their states, $\omega_{fd} = 21 \text{ cm}^{-1}$ (as discussed earlier), and this is precisely where the observed change in slope occurs, and the slopes observed are $(\delta\omega)^{-1/2}$ and $(\delta\omega)^{-2}$ as well.

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