Autoionizing states in Li I observed in optical-emission spectra

Henrik Cederquist and Sven Mannervik
Research Institute of Physics, S-104 05 Stockholm, Sweden
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A beam-foil study of the lithium spectrum above 5000 Å has been undertaken. Four new observations of core-excited doublet transitions are reported. All of these transitions have autoionizing states as the final state. The autoionization width of the $(1s\,2s\,{}^3S)3d\,{}^2D$ has been measured, yielding a value of 0.89±0.04 meV. A comparison with calculations of the term energies and the width is presented.

I. INTRODUCTION

Core-excited doublet states in lithium, which all lie high above the ionization limit, have been studied by many different techniques. The first experimental information was obtained by Ederer *et al.*[1] in a photoabsorption experiment where core-excited $^2P^0$ states were excited from the ground state of Li I. This series is the only one accessible optically from the ground state. McIlrath and Lucatorto[2] developed the photoabsorption experiment further by preparing the lithium atoms in the first excited state by tuning a dye laser to the resonance transition $1s^22s$-$1s^22p$. From this excited state a number of even-parity states of ^2S, ^2P, and ^2D symmetries could be excited with use of a continuum background source.

The core-excited states have also been studied by the electron spectra from atomic collisions,[3–6] having Li either in the ion beam or as a target. The advantage here is that states which are not optically accessible can be studied. Low-energy-electron impact on lithium vapor has also been used in the study of the electron spectrum.[7] In the most favorable cases these experiments determine the energies of the levels with an accuracy of 10 meV, but in other cases errors of 50 or 100 meV are given. In the photoabsorption measurements of McIlrath and Lucatorto,[2] an accuracy is given which corresponds to an uncertainty in energy of 50 cm^{-1}, or 6 meV.

Core-excited configurations in Li I forming quartets have no optical link to the singly excited system. It was, however, found early that the optical beam-foil spectra exhibited many lines belonging to the quartet system of neutral lithium.[8,9] Thus, the beam-foil technique has been extensively used to study multiply-excited nonautoionizing states. The term energies of the Li I quartets have recently been established by Mannervik and Cederquist[10] at an accuracy of 1 meV or better on an absolute scale. While the quartets are prohibited from Coulombic autoionization by spin conservation, the core-excited doublets are not, since they are embedded in a doublet continuum. In order to autoionize, the parity of the state must, however, be conserved. This restriction will make also several doublets bound (in the sense that Coulombic autoionization is forbidden). For example, the $1s\,2p^2\,{}^2P$ state must go to a ^2P continuum in order to conserve L and S, but since the $1s^2\epsilon p\,{}^2P^0$ continuum has odd parity such an autoionization process is forbidden.

The fact that some doublet states are bound means that they can be observed in optical-emission spectra as transitions either within the core-excited system or down to the singly excited system. The latter type of transition was first observed by Buchet *et al.*[11] Some identifications of core-excited doublet intrasystem lines were also discussed by Berry *et al.*[12] Emission lines from the core-excited to the singly excited states have recently been observed from a microwave-heated plasma.[13] Berry *et al.*[14] first pointed out that since the core-excited doublet system both contains bound and autoionizing levels it should be possible to observe emission lines from transitions from bound to autoionizing levels. Such transitions should be observed as broadened spectral lines from which the lifetime of the autoionizing level could be determined from the linewidth. We have recently[15–18] utilized this idea to make accurate determinations of autoionization widths.

Since the energy is inversely proportional to the wavelength it is obvious that observations within the core-excited system where the transitions have wavelengths greater than 2000 Å will give the opportunity of much more accurate determinations of relative term energies as well as widths compared to studies of transitions between the core-excited and the singly excited system. In this context it should be pointed out that the identification of the intrasystem lines is crucial. Without correct identifications, measurements of linewidths are not meaningful. Since the transition wavelength will change drastically for uncertainties of, say, 0.1 eV in the term energies and since many transitions end in autoionizing levels, which limits the possibility of closed-loop considerations, the construction of the term system has been a difficult task. Very accurate calculations by Bunge[19] and by Jáuregui and Bunge,[20] together with our recent experimental results,[15,21,22] have made it possible to firmly establish six transitions within the system.[15,20]

II. EXPERIMENTAL

The experiment was performed at the 400-kV heavy-ion accelerator at the Research Institute of Physics, Stockholm, utilizing the beam-foil technique. The equipment has been described in detail earlier[22] as well as the method for scanning over the line profile.[18] It might still be ap-

propriate to repeat here that the scanning of the line profile is normalized to the beam intensity by a monitor unit, which records the total light yield at a fixed distance after the foil. This monitor signal is also connected to a gate unit which requires that the signal does not vary outside certain preset limits (typically $\pm 20\%$). The gate circuit is used to prevent the contribution from the detector background from varying over the line profile. The normalization unit and the gate circuit are essential for a correct experimental description of the line profile. In order to obtain good statistics we sweep over a spectral region several times. Thereby we obtain a more proper averaging over the line profile. The spectra recorded in different scanning directions are stored separately to avoid effects from mechanical backlash in the scanning system.

A Li^+ beam of about 10 μA at energies between 100 and 200 keV was passed through the foil. The residual gas pressure in the beam-foil chamber was 2×10^{-6} Torr. The photons were analyzed by a Heath EUE 700 monochromator equipped with a 600-groves/mm grating blazed at 7500 Å and counted by a Hamamatsu infrared photomultiplier, R 943-02. The photomultiplier was cooled to $-30\,^{\circ}$C by a Hamamatsu refrigerator C659, using coolant water and a Peltier element. At this temperature the detector background was typically 4 counts/s. In order to avoid second-order lines in the wavelength region above 5000 Å studied here, a long-pass filter with a cutoff wavelength of 4750 Å (50% transmission) was used. Data were stored in the memory of a BM 96 Intertechnique multichannel analyzer.

For the lifetime measurements it is essential to know the beam energy after the passage of the foil. This was measured in a 90° 0.5-m-radius electrostatic energy analyzer.

III. DATA ANALYSIS

The recorded spectra were analyzed by a peak-fitting program in which the lines are approximated with a Gaussian line profile. The program gives also the opportunity of folding the Gaussian with a Lorentzian profile. This is used for the analysis of broadened lines. A detailed description of the program has been given recently[18] and will not be repeated here. The lifetime curves were analyzed by the fitting program DISCRETE.[23]

IV. RESULTS AND DISCUSSION

The core-excited quartets of Li I are built on the $1s\,2s\,{}^3S$ and $1s\,2p\,{}^3P$ parent states. Thus, several satellites are observed[10] close to the $1s\,2s\,{}^3S-1s\,2p\,{}^3P$ transition at 5485 Å in Li II, which forms the series limit for Rydberg transitions $1s\,2snl\,{}^4L-1s\,2pnl\,{}^4L$. The core-excited doublets can be built both on singlet ($1s\,2s\,{}^1S, 1s\,2p\,{}^1P$) and triplet ($1s2s\,{}^3S, 1s\,2p\,{}^3P$) parent states. Thus, there are, in principle, two different series limits: $1s\,2s\,{}^1S-1s\,2p\,{}^1P$ at 9581 Å and $1s\,2s\,{}^3S-1s\,2p\,{}^3P$ at 5485 Å. Since the triplets have lower energies than the singlets, the triplets will, however, be most important for lower members of the doublet series. These arguments indicate that lines of the following type:

$$(1s\,2s\,{}^3S)np\,{}^2P^o-(1s\,2p\,{}^3P)np\,{}^2P\ ,$$

$$(1s\,2s\,{}^3S)nd\,{}^2D-(1s\,2p\,{}^3P)nd\,{}^2D^o\ ,$$

and

$$(1s\,2s\,{}^3S)nf\,{}^2F^o-(1s\,2p\,{}^3P)nf\,{}^2F$$

should be expected not far from the 5485-Å Li II transition.

In an earlier work[15] we were able to experimentally establish six transitions within the core-excited doublet system. For that work the accurate calculations of Jáuregui and Bunge[20] were very useful. They claimed that their calculation had an accuracy 1 order of magnitude better than what was obtained experimentally in the excellent work of McIlrath and Lucatorto.[2] Our work[15] could in most cases confirm the result of Jáuregui and Bunge.[20] It might be worth noting here that since we are observing lines within the core-excited system which have wavelengths greater than 2000 Å, we have the opportunity to determine energy differences between levels much more accurately than, e.g., McIlrath and Lucatorto, who observed transitions from the singly excited to the doubly excited system, which appear at wavelengths around 200 Å. A wavelength uncertainty as large as 0.5 Å at 5000 Å would still require an accuracy of better than 1 mÅ at 200 Å in order to determine an energy difference with the same accuracy. Another advantage of studying lines within the system is that for transitions having an autoionizing level as the final state, a typical width of a couple of meV will give a broadening of a couple of angstroms of the spectral line, which is possible to measure accurately.[15] To make a similar determination in Auger spectra or photoabsorption spectra an energy resolution $\Delta E/E$ of 10^{-4} or better is required.

Since we have determined the width of the lowest state in the series of $^2P^o$ and ^2D, we found it interesting to try to measure the widths of higher members of the series. The widths are, however, expected to decrease as the principal quantum number n for the outer electron increases. According to Bhatia[24] and $^2D(2)$ state [$(1s\,2s\,{}^3S)3d\,{}^2D$] should have the largest width and consequently be least difficult to measure.

The transition $(1s\,2s\,{}^3S)3d\,{}^2D-(1s\,2p\,{}^3P)3d\,{}^2D^o$ [$^2D(2)$-$^2D^o(1)$, in abbreviated form] have not been observed earlier. Combining the theoretical values of Jáuregui and Bunge[20] and of Chung,[25] a wavelength of 5935 Å is obtained. For the $^2D^o$ states the values of Jáuregui and Bunge are the only ones available. Wakid *et al.*[26] have calculated energies for the even ^2D states. They give two alternative values for each level depending on if they relate their energy to the ground state or to the Hartree-Fock energy of the excited threshold. Using these values instead of Chung's[25] energy, we obtain a transition wavelength of 5921 or 5949 Å. For the $^2D^o$ states, which are not accessible optically from the ground state nor from the first excited state $1s^2\,2p\,{}^2P^o$, no experimental values on an absolute scale have been published. If the experimental term energy of the $^2D(2)$ state given by McIlrath and Lucatorto[2] is combined with the $^2D^o(1)$ energy of Jáuregui and Bunge,[20] a transition wavelength of 5907 ± 18 Å is obtained.

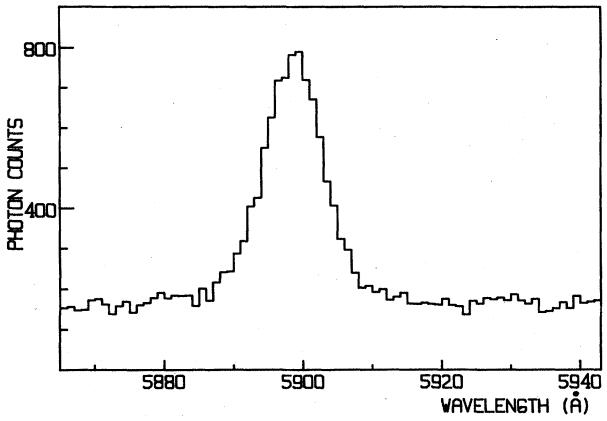

FIG. 1. Part of the beam-foil spectrum of Li recorded at 130 keV. The slit width was 150 μm.

In the present experiment we did find a line at 5900.3±0.2 Å (Fig. 1). This line was about 25% wider than other lines nearby. We also measured the lifetime of the upper level of the transition and found it to be 1.5±0.1 ns. This is in perfect agreement with the earlier measurements[22] of the lifetime of the $^2D^o(1)$ level using the decay branches at 3661 Å (1.4±0.1 ns) and 3144 Å (1.5±0.1 ns) (see Fig. 2). We conclude that the 5900-Å line should be identified as the $(1s2s\,^3S)3d\,^2D$–$(1s2p\,^3P)3d\,^2D^o$ transition.

Recording the line very close to the foil we found the line to be blended by a line having a wavelength 8–10 Å shorter than the stronger Li transition. By using different beam projectiles we could show that this line originated from sputtered carbon (a C II line). Contribution from this line could, however, be avoided by increasing the distance between the foil and the observation point.

The natural width of the $^2D(2)$ level was determined from 13 independent measurements (one of these is shown in Fig. 1). Ten of these were recorded at a slit width of 150 μm and three at 100 μm. Instrumental linewidths of about 8 and 6 Å, respectively, were obtained for these slit widths (the monochromator having a dispersion $D \approx 1/40$ mm Å^{-1}). A mean value of 2.5±0.1 Å was obtained for the Lorentzian width. In principle, it would be possible to increase the ratio between the Lorentzian and the instrumental width by using more narrow slits. As discussed in an earlier paper[18] we have, however, found that it is essential to obtain good statistics. In particular, it is necessary to have a signal level which is much higher than the detector background in order to obtain significant Lorentzian wings in the profile. The experimental response function (approximated by a Gaussian profile) was determined from the Li I $2p\,^2P^o$–$3d\,^2D$ transition at 6103 Å. All runs gave Lorentzian width consistent with each other, indicating that no occasional errors were present.

The Lorentzian width of the 5900-Å line corresponds to an autoionization width of 0.89±0.04 meV (7.2±0.3 cm^{-1}) for the $(1s2s\,^3S)3d\,^2D$ state. The observed width is, in principle, the sum of the natural width of the upper and the lower level. The upper level has, however, a lifetime of 1.5 ns corresponding to a width of only 0.4 μeV. The contribution from the radiative decay is negligible also for the lower state as can be seen from the oscillator

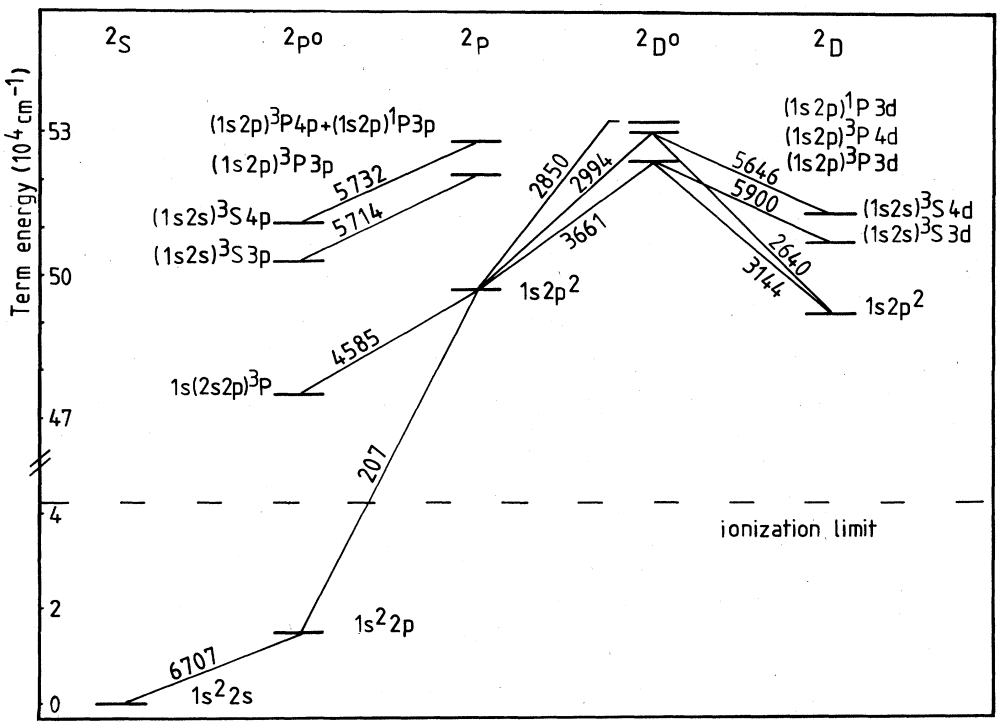

FIG. 2. The term system of the core-excited doublets of Li I. Wavelengths are given in Å.

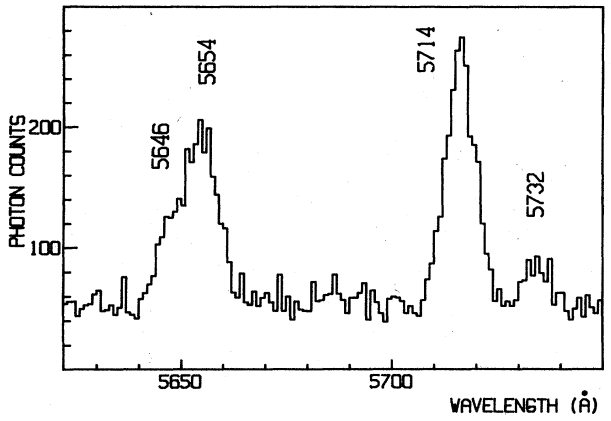

FIG. 3. Section of the beam-foil spectrum of Li recorded at 165 keV with use of 150-μm slits. The line at 5654 Å is the $4s\,^3S-6p\,^3P^o$ Li II transition. The other three lines which can be observed all belong to the core-excited system of Li I doublets.

strength calculated by Chung.[25] One more source of broadening to consider is the fine structure, which cannot be resolved spectroscopically in the present experiment. Chung[27] has calculated the fine structure of the $(1s2p\,^3P)3d\,^2D^o$ and the $(1s2s\,^3S)3d\,^2D$ state to be 0.3 cm^{-1} and 0.1 cm^{-1}, respectively, in calculations similar to those performed for Be II.[28] Thus, the fine structure will not give any significant contribution to the observed linewidth. The error given for the width reflects the statistical uncertainty (one standard deviation).

In Table I we compare our experimental width with the calculations available. It turns out that the experimental value falls in between the calculated values of Bhatia[24] and of Chung.[27] For completeness we also include the experimental widths of the $^2P^o(1)$ and $^2D(1)$ states which have been published earlier.[15]

The transition $(1s2s\,^3S)3p\,^2P^o$–$(1s2p\,^3P)3p\,^2P$ [or $^2P^o(3)$-$^2P(2)$, in abbreviated form] will appear at 5756 Å according to the calculations of Bunge[19] and Chung.[29] If we instead combine the energy for the $^2P(2)$ state of Bunge with the $^2P^o(3)$ energy of Wakid *et al.*,[26] we obtain 5698 or 5724 Å, depending on whichever value is used of the two values that they have presented. A combination of the experimental values of McIlrath and Lucatorto[2] [$^2P(2)$] and of Cantù *et al.*[30] gives a transition wavelength of 5714±18 Å. In Fig. 3 we show a spectrum recorded in the region 5600–5750 Å. Here a prominent line is observed at 5713.8±0.5 Å. We obtain a lifetime of 0.30±0.05 ns for the upper state of the transition. From the transition probabilities calculated by Bunge[19] we obtain a theoretical lifetime of 0.28±0.03 ns for the $^2P(2)$ level, thus in very good agreement with our experimental value. The lifetime for the $^2P(1)$ which can be derived from Bunge's calculations is 47±5 ps. This is in perfect agreement with the recent experimental value of Berry *et al.*,[31] 49±5 ps, which indicates that Bunge's values are reliable. In the deduction of the lifetime values above, we have not included the transition probabilities for decay within the core-excited system, since these are not available. They should, however, be small enough to be covered by the errors quoted.

In this context it might be worth mentioning that while the ^2P lifetimes seem to be well determined only by considering the radiative decay, this is not the case for the $^2D^o$ states[20] for which spin-induced autoionization seems to be a somewhat more important decay branch than the radiative decay. Similar effects have been observed for the Li I quartets.[17,22] As has been stated earlier,[17] these observations call for calculations of rates for spin-induced autoionization.

In the spectrum (Fig. 3) we also observe a weak line at 5731.8±0.6 Å. We suggest that this line should be identified as the transition between the $[(1s2p\,^3P)4p + (1s2p\,^1P)3p]^2P$ and the $(1s2s\,^3S)4p\,^2P^o$ states [$^2P^o(4)$-$^2P(3)$]. This is consistent with the experimental values of McIlrath and Lucatorto[2] and Cantù *et al.*,[30] from which a wavelength of 5749±20 Å can be derived. The lifetime of the upper state is found to be very short, but the weak signal does not permit a proper lifetime measurement.

In Fig. 3 still one more new line can be observed. This is not completely resolved from the Li II $4s\,^3S-6p\,^3P^o$ transition at 5654 Å. Lifetime measurements for the transition at 5646.4 Å yield a lifetime of 0.56±0.04 ns. This is in good agreement with the lifetime for the $^2D^o(2)$ level

TABLE I. Autoionization widths (in meV) in Li I.

State	Experimental	Calculations				
		P[a]	SS[b]	B[c]	NA[d]	C[e]
$1s(2s2p\,^3P)^2P^o$	2.6 ±0.1[f]	507	3.07	7	3.4	3.76
$1s2p^2\,^2D$	10.5 ±0.3[f]	23	123	5	10.0	10.9
$(1s2s\,^3S)3d\,^2D$	0.89±0.04[g]			0.75		1.11

[a]Propin, Ref. 35.
[b]Safranova and Senashenko, Ref. 36.
[c]Bhatia, Ref. 24.
[d]Nicolaides and Aspromallis, Ref. 37.
[e]Chung, Ref. 27.
[f]Cederquist and Mannervik, Ref. 15.
[g]This work.

TABLE II. Wavelengths of transitions within the core-excited doublet system of Li I.

Identification	Wavelength (Å) This work	Comparison wavelength[a] C-B	W_1-B	W_2-B	Ca-M	M-B
$(1s\,2s\ ^3S)3p\ ^2P^o$–$(1s\,2p\ ^3P)3p\ ^2P$	5713.8±0.5	5756	5698	5724	5714±18	
$(1s\,2s\ ^3S)4p\ ^2P^o$–$[(1s\,2p\ ^3P)4p + (1s\,2p\ ^1P)3p]^2P$	5731.8±0.6	5757	5760	5787	5749±20	
$(1s\,2s\ ^3S)3d\ ^2D$–$(1s\,2p\ ^3P)3d\ ^2D^o$	5900.3±0.2	5935	5921	5949		5907±18
$(1s\,2s\ ^3S)4d\ ^2D$–$(1s\,2p\ ^3P)4d\ ^2D^o$	5646.4±0.6	5678	5947	5976		5672±16

[a]Wavelengths are derived by combining results of different authors, C-B meaning Chung's values combined with those of Bunge, etc. C, Chung (Refs. 25 and 29); B, Bunge (Ref. 19) and Jáuregui and Bunge (Ref. 20); W_1, W_2, Wakid *et al.* (Ref. 26)—they give two alternative energies; Ca, Cantù *et al.* (Ref. 30); M, McIlrath and Lucatorto (Ref. 2).

which has been determined earlier[22] by the 2640-Å transition (0.51±0.05 ns). The lifetime of the $6p\ ^3P^o$ state in Li II is 10.6 ns.[32] Thus we identify the 5646-Å line as the $^2D(3)$-$^2D^o(2)$ $[(1s\,2s\ ^3S)4d\ ^2D$–$(1s\,2p\ ^3P)4d\ ^2D^o]$ transition. This is not completely consistent with the value obtained when the term energies of Bunge[19] $[^2D^o(2)]$ and McIlrath and Lucatorto[2] $[^2D(3)]$ are combined: 5672±16 Å. The $(1s\,2s\ ^3S)4f\ ^2F^o$–$(1s\,2p\ ^3P)4f\ ^2F$ transition should also be expected at a wavelength close to the wavelengths discussed here. There are, however, neither experimental or theoretical results available for the ^2F states. The wavelengths of the new observations are collected in Table II.

The fairly strong $^2P^o(3)$-$^2P(2)$ transition at 5714 Å would, in principle, give the opportunity of measuring the autoionization width of the $^2P^o(3)$ level. We were, however, not able to observe a significant broadening of the line. A small width is in agreement with the calculations of Bhatia,[24] who gives a width of 0.021 meV. This is the only theoretical result available.

V. SUMMARY

We have observed four new transitions in the core-excited system of Li I doublets. Together with our earlier results,[15] these transitions make it possible to construct a term system of 12 levels. The relative energies are determined with an accuracy better than 1 meV. It would be desirable that calculations of the ^2F and $^2F^o$ states could be performed. Recently, Agentoft *et al.*[33] have found that isoelectronic core-excited ^2F states in Be II are involved in strong transitions.

We have also reported the first experimental determination of the autoionization width of the $(1s\,2s\ ^3S)3d\ ^2D$ state. The theoretical results available are in fairly good agreement (±20%) with our results. To the best of our knowledge Manson[34] is the only one who has considered spin-induced autoionization of Li I. This was done for the lowest quartet state for which this is the only decay mode possible. As has been pointed out in this article and also earlier,[17,20] spin-induced autoionization seems to be an important decay mode also for more highly excited states. For core-excited $^2D^o$ states[20] as well as for ^4P and $^4D^o$ states it seems to be even stronger than the radiative decay mode. It would be very interesting if such calculations could be performed.

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