Emission spectrum of core-excited Li doublets

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Configuration-interaction calculations have been carried out for the core-excited $^2D^o$ bound states of neutral Li. The levels $1s2p\ ^3P\ 3d$, $1s2p\ ^3P\ 4d$, and $1s2p\ ^1P\ 3d$ are estimated to lie 524 235.5(3.0), 530 306.1(3.0), and 532 058.8(4.5) cm^{-1} above the ground state, respectively. Using additional theoretical and experimental data for core-excited Li levels and calculated oscillator strengths, we have developed a consistent scheme for the decay of Li** doublets which explains the main features of this spectrum. Two previously observed lines $\lambda = 2846$ and 3661 Å correspond to transitions between $^2D^o$ and ^2P bound states, in agreement with an earlier assignment. Three other lines $\lambda = 2640$, 3144, and 4590 Å result from bound states $^2D^o$ and ^2P decaying into ^2D and $^2P^o$ core-excited states which undergo rapid autoionization, in agreement with the prominent linewidths exhibited experimentally. Possible spin-forbidden electric-dipole transitions are discussed.

I. INTRODUCTION

During the last fifteen years much effort has been devoted to the elucidation of the spectrum of doubly excited Li.[1-7] This spectrum is profusely produced in the beam-foil source and most of the observed lines have been assigned to transitions between quartet states lying between 57.4128 and 66.6729 eV above the ground state.[8]

Because coulomb autoionization rates are associated with $\tau = 10^{-13}-10^{-15}$ sec,[9] photon emission can only be expected from states which live long enough to allow for electric-dipole radiative decay ($\tau = 10^{-7}-10^{-9}$sec). Such states lie below the continuum with the same LS symmetry and parity, and they are believed to autoionize slowly ($\tau = 10^{-5}$-10^{-7}sec for neutral systems)[9] on account of the small mixings of the adjacent continuum with states of different spin and orbital angular momentum.

Garcia and Mack[1] first noted that Li $1s2pnp\ ^2P$ and $1s2pnd\ ^2D^{\circ}$ states should be metastable against autoionization. Years later, Andersen *et al.*[10] rediscovered the Li Iλ3144 Å line[11] and pointed out that it should originate from doubly excited Li on account of the similarity of its excitation function with those from other lines known to belong to the Li quartet system. Contrary to the narrow-line transitions between nonautoionizing quartet states, the λ3144 Å line exhibited a large natural linewidth[12] proper of a transition from a bound state to an autoionizing state, and thus it became the first line to be unambiguously assigned to the Li** doublet system.[12]

The lines from core-excited Li doublets are expected to have very short mean lifetimes because the Li** doublets readily decay to the singly excited Li $1s^2nl\ ^2L$ states emitting photons in the 200 Å region. Berry *et al.*[4] used this knowledge to correctly assign several Li** lines to the doublet system.

In 1977 McIlrath and Lucatorto[13] used a high-power pulsed dye laser tuned to the $1s^22s \to 1s^22p$ resonance transition to produce large quantities of odd-parity $1s^22p$ Li atoms. These were subsequently excited by photons from a continuum probe pulse in the 100–300-Å region, and over twenty lines corresponding to transitions from the $1s^22p$ level to core-excited ^{2}SPD states were observed. Extensive calculations by Weiss[14] were used to unambiguously assign[13] ten of the observed lines. Higher series members were assigned[13] using quantum-defect comparisons and relative intensities. The ensuing calculations[15] showed that the lowest four core-excited ^2P states had been observed in the McIlrath-Lucatorto experiment.

Emission from Li $1s2p^2\ ^2P$ to $1s^22p$ was first observed by Buchet *et al*[16] in the beam-foil source at 207.5 Å. Recently, Willison *et al.*[17] studied the far uv emission spectrum of a microwave heated Li plasma, and observed transitions from the first four even-parity ^2P states which are believed to be good candidates for lasers in the 200 Å region.[18]

In the latest beam-foil study of the Li** spectrum,[7] unprecedented high-quality data were obtained and roughly one-half of the observed lines were left unclassified. We then decided to undertake the present study of the emission spectrum of Li** doublets, expecting to be able to account for many of the unclassified transitions. Since the ^2P levels have already been investigated[13-17] and the bound states ^2F, $^2G^0$, etc, either decay in the 200 Å region or in the infrarred, we restricted our study to the $^2D^o$ states which are expected to emit photons in the well-studied 2000–5500 Å range.[7] In Sec. II we present configuration interaction (CI) calculations for the first three core-excited $^2D^o$ states and calculate term values relative to the ground state. Transitions between Li** doublets are discussed in Sec. III together with absolute term values for the Li** doublet system. In Sec. IV we discuss the possible occurrence of spin-forbidden transitions in the Li** spectrum. The present study has been complemented with calculations on Li quartet states,[19,20] core-excited nonautoionizing Li^- states,[21] and recent experimental work.[22-24]

II. CALCULATIONS OF THE $^2D^o$ STATES

The nonrelativistic energies E_{nr} are calculated variationally by means of CI wave functions Ψ,[25]

$$\Psi = \sum_{Kp}^{\approx 200} \Phi_K^{(p)} a_{Kp} \tag{1}$$

based on (i) an energy optimized Slater-type orbital (STO) basis, (ii) orthogonal one-electron symmetry adapted spinorbitals, with equivalence of partner orbitals in degenerate representations, (iii) three electron LS eigenfunctions, and (iv) the consideration of STO and CI truncation energy errors ΔE_{STO} and ΔE_{CI}, respectively. Following the experience obtained in the companion papers,[8,20] relativistic, radiative, and mass polarization corrections are assumed to be equal to those in the corresponding well-defined Li^+ cores, which are $1s2p\,^3P$ in $^2D^o(1)$, $^2D^o(2)$, and $1s2p\,^1P$ in $^2D^o(3)$.

In Table I we give the STO parameters and corresponding truncation energy errors ΔE_{STO}. It may be seen that, although there is some change from one state to another, there is some sort of systematic trend in the build up of the STO set which may help considerably in the calculation of higher excited $^2D^o$ states. This has not been pursued, however, because the last two states, $^2D^o(2)$ and $^2D^o(3)$ already contributed with faint uv lines, due to their preferential decay to singly excited $1s^2nd$ states in the 200 Å region. The larger ΔE_{STO} error for the $^2D^o(3)$ state reflects the poor convergence of the CI expansion for a Li^+ $1s2p\,^1P$ core. In Table II we collect the data needed to compute the nonrelativistic energies E_{nr} as sums of rigorous energy upper bounds E_u and truncation energy errors:

$$E_{nr} = E_u + \Delta E_{CI} + \Delta E_{STO}. \tag{2}$$

We then use experimental and nonrelativistic energies for Li^+ $1s2p\,^{1,3}P$ states to deduce absolute term values as[8,20]

$$T = T(Li^+ \text{ core}) + E_{nr}(^2D^o) - E_{nr}(Li^+ \text{ core}). \tag{3}$$

III. TRANSITIONS BETWEEN Li** DOUBLETS

From theoretical data in Ref. 15 and the ionization potential of Li,[26] it follows that Li $1s2p^2\,^2P$ should be 496 943(10) cm^{-1} above the ground state. By combining this result with the absolute term values in Table II we find that the first three $^2D^o$ states decay to $1s2p^2\,^2P$ with λ(air) = 3663.0(1.7), 2996.4(1.2), and 2846.9(1.1) Å, respectively. These are close but not quite overlapping with the lines observed at λ = 3660.9,[4,10,22] 2994.1(0.3),[7] and 2846 (2) Å.[4] If instead of using the theoretical result above one places Li $1s2p^2\,^2P$ exactly 496 927.5 cm^{-1} above the ground state, the theoretical wavelengths become λ = 3660.9(0.4), 2995.0(0.3), and 2845.6 (0.3) Å exhibiting a more satisfactory agreement with experiment (we have assumed that the exper-

TABLE I. STO parameters for the $1s2pnd\,^2D^o$ states of Li and STO truncation energy errors, ΔE_{STO}, in μhartree.

l	State	STO's	ΔE_{STO} $^2D^o(1)$	$^2D^o(2)$	$^2D^o(3)$
	$^2D^o(1)$	$1s=3.00$; $3s=2.7$; $4s=4.8$; $3s=1.3$			
0	$^2D^o(2)$	$1s=3.00$; $3s=2.7$; $4s=5.1$; $3s=1.4$			
	$^2D^o(3)$	$1s=3.00$; $3s=3.1$; $4s=5.3$; $3s=1.75$			
	$^2D^o(1)$	$2p=1.01$; $2p=2.29$; $2p=0.54$; $3p=1.49$; $4p=3.55$; $5p=3.55$			
1	$^2D^o(2)$	$2p=0.98$; $2p=2.29$; $2p=0.72$; $3p=1.49$; $4p=3.10$; $5p=3.65$			
	$^2D^o(3)$	$2p=0.88$; $2p=2.25$; $2p=0.49$; $3p=1.49$; $4p=3.50$; $5p=4.00$			
	$^2D^o(1)$	$3d=0.378$; $3d=0.88$; $4d=0.32$; $3d=3.4$; $4d=2.18$; $5d=4.3$			
2	$^2D^o(2)$	$3d=0.357$; $3d=0.87$; $4d=0.258$; $5d=0.38$; $3d=3.4$; $4d=2.18$; $5d=4.3$			
	$^2D^o(3)$	$3d=0.360$; $3d=0.82$; $4d=0.210$; $5d=0.353$; $3d=2.5$; $4d=3.08$			
	$^2D^o(1)$	$4f=0.65$; $5f=1.3$; $4f=3.22$; $5f=3.5$			
3	$^2D^o(2)$	$4f=0.75$; $4f=0.32$; $4f=3.22$; $5f=3.5$			
	$^2D^o(3)$	$4f=0.63$; $5f=1.4$; $4f=3.22$; $5f=3.5$			
	$^2D^o(1)$	$5g=0.93$			
4	$^2D^o(2)$	$5g=1.0$			
	$^2D^o(3)$	$5g=0.97$; $5g=3.5$; $6g=4.0$			
		Total truncation energy error	69 ± 13	61 ± 13	157 ± 20

TABLE II. Nonrelativistic energies E_{nr} for the $1s2pnd\ ^2D^o$ states, in a.u.(Li) and estimates of their absolute term values T, in cm^{-1}. 1 a.u. (^{7}Li) = 219 457.48 cm^{-1}.

	$^2D^o(1)$	$^2D^o(2)$	$^2D^o(3)$
E_u, 200-term CI	−5.089 207	−5.061 562	−5.053 535
ΔE_{CI}	−0.000 015	−0.000 006	−0.000 006
ΔE_{STO}	−0.000 069(13)	−0.000 061(13)	−0.000 157(20)
E_{nr}, Eq. (2)	−5.089 291(13)	−5.061 629(13)	−5.053 668(20)
E_{nr}, Li^+ $1s2p\ ^3P$	−5.027 715 70(1) [a]		
E_{nr}, Li^+ $1s2p\ ^1P$			−4.993 351 08(1) [a]
T Li^+ $1s2p\ ^3P$, in cm^{-1}	537 748.6 [b]		
T Li^+ $1s2p\ ^1P$, in cm^{-1}			545 295.8 [c]
$\Delta E \simeq \Delta E_{nr}$	−0.061 575(13)	−0.033 913(13)	−0.060 317(20)
ΔE_{nr}, in cm^{-1}	−13 513(3)	−7 742(3)	−13 237(4.5)
$T \simeq T$ $Li^+ + E_{nr}$, Eq. (3)	524 235.5(3.0)	530 306.1(3.0)	532 058.8(4.5)

[a] Y. Accad *et al.*, Phys. Rev. A 4, 516 (1971).

[b] Reference 8.

[c] Obtained by adding the ionization potential of Li (Ref. 26) to the energy difference between Li^+ $1s^2$ and $1s2p\ ^1P$ [G. Herzberg and H. R. Moore, Can. J. Phys. 37, 1293 (1959)].

imental error in $\lambda = 3660.9$ is negligible). From an analysis of basis sets used in Ref. 15 it appears conceivable that truncation energy errors in the $1s2p\ ^1P$ core component of $1s2p^2\ ^2P$ were too small,[15] thus implying for Li $1s2p^2\ ^2P$ an energy lower than the one reported in Ref. 15. We therefore are led to assume that the correct value for this state is

$$T \text{ Li } 1s2p^2\ ^2P = 496\,927.5 \text{ cm}^{-1}. \tag{4}$$

In Table III we give wavelengths and f values for several transitions between ^2P, ^2D, and $^2P^o$, $^2D^o$ states. Branching ratios and transition probabilities associated to the $\lambda = 3661$ and 2846 Å lines are appreciable and therefore it may be concluded that indeed they correspond to the lines whose correct interpretation was first given by Berry *et al.*[4] It is surprising that the $\lambda = 2846$ Å line has not been detected in more recent work,[7,22] although it must be a very weak line on account of an unfavorable branching ratio. Future experimental work may possibly reveal that the weak line $\lambda = 2994.1(0.3)$ Å is blended with the $[1s2p\ ^3P\ 4d]^2D^o \to 1s2p^2\ ^2P$ line which we predict at $\lambda = 2995.0(0.3)$ Å.

An accurate theoretical estimate for the absolute term value of the autoionizing Li $1s2p^2\ ^2D$ state is difficult to obtain, and as we will see it is not worth the effort. From the absorption spectrum of McIlrath and Lucatorto[13] one finds T Li $1s2p^2$ $^2D = 492\ 482(50)$ cm^{-1}. Similar experimental work by Cantú *et al.*[27] yields T Li $1s2p^2\ ^2D = 492\ 413(23)$ cm^{-1}. We will assume that T is given exactly by

$$T \text{ Li } 1s2p^2\ ^2D = 492\ 443 \text{ cm}^{-1}. \tag{5}$$

It is then found that the first three $^2D^o$ states decay to $1s2p^2\ ^2D$ with $\lambda = 3144.5(0.3)$, 2640.3(0.2), and 2523.4(0.2) Å. The first two lines are in excellent agreement with the experimental lines $\lambda = 3144.5$ (0.2) and 2640.1(1.0) Å.[7] The transition probabilities in Table III explain why these lines are observed and they also explain why the $\lambda = 2523$ Å line is not observed, due to the preferential decay of $^2D^o(3)$ into $1s^23d$, $1s^24d$, $1s^25d$, and $1s2p^2\ ^2P$.

Although the even-parity ^2P states decay mostly to $1s^2np$ states in the 200 Å region,[15,17] some branching into the core-excited autoionizing $^2P^o$ states is expected. According to Ederer *et al.*[28] the lowest core-excited $^2P^o(1^*)$ resonance state is $[1s\ 2s2p\ ^3P]^2P^o$ lying 475 150(50) cm^{-1} above the ground state. Assuming that the term value of $^2P^o(1^*)$ is exactly 475 147 cm^{-1} we find the $^2P(1) \to {}^2P^o(1^*)$ transition at $\lambda = 4590.0$ Å in good agreement with the 10 ± 3 Å broadened $\lambda = 4590$ Å line assigned by Berry *et al.*[4] to the Li** doublet system. Furthermore, the transition $^2P(2) \to {}^2P^o(1^*)$ is at $\lambda = 2183$ Å coinciding with the $\lambda = 2183$ Å line which Berry *et al.*[4] assigned to the unlikely $1s12f \to 1s3d$ transition in Li II. Experimental information concerning the width of $\lambda = 2183$ Å will decide its classification.

One may also expect one-electron jumps from the odd-parity $^2D^o$ bound states into the core-excited autoionizing ^2D states. The $^2D^o(1) \to {}^2D(1^*)$ transition $\lambda = 3144.5$ Å has already been discussed. According to McIlrath and Lucatorto,[13] the $[1s2s\ ^3S\ nd]^2D$ states ($n = 3, 4$) lie 507 297(50) and 512 689 (50) cm^{-1} above the ground state, respectively. The corresponding one-electron jumps from $^2D^o(1)$ and $^2D^o(2)$ have $\lambda = 5902$ and 5675 Å, respectively, and they have not been observed. Analogously, $^2D^o(3)$ should also decay to $1s2s\ ^1S\ 3d$ [known to lie at 520 593(50) cm^{-1}][13] yielding a very weak line at $\lambda = 8719$ Å.

TABLE III. Wavelengths (in air), absorption f values, oscillator strengths, transition probabilities (in 10^8 sec^{-1}), and line strengths for transitions between some ^2P, ^2D, and $^2P^o$, $^2D^o$ states of neutral Li; A_{ki} and f values accurate to ten per cent.

Transition	Wavelength (Å, this work)	f	A_{ki}	$S(i,k)$
$1s2p^2\ ^2D \rightarrow [1s2p\ ^3P\ 3d]^2D^o$	3144.5(0.3)	0.154	1.03	16.0
$^2P \rightarrow$	3660.9(0.4)	0.128	0.37	9.32
$1s2p^2\ ^2D \rightarrow [1s2p\ ^3P\ 4d]^2D^o$	2640.3(0.2)	0.048	0.46	4.2
$^2P \rightarrow$	2995.0(0.3)	0.013	0.06	0.79
$1s2p^2\ ^2D \rightarrow [1s2p\ ^1P\ 3d]^2D^o$	2523.4(0.2)	0.022	0.24	1.8
$^2P \rightarrow$	2845.6(0.3)	0.31	1.53	17.5
$1s^23d \rightarrow [1s2p\ ^3P\ 3d]^2D^o$	202.860	0.0003	0.42	0.0018
$4d \rightarrow$	205.081	0.00005	0.08	0.0003
$5d \rightarrow$	206.126	0.00008	0.13	0.0006
$1s^23d \rightarrow [1s2p\ ^3P\ 4d]^2D^o$	200.392	0.0022	3.6	0.015
$4d \rightarrow$	202.559	0.00012	0.2	0.0008
$5d \rightarrow$	203.579	0.00014	0.2	0.0009
$1s^23d \rightarrow [1s2p\ ^1P\ 3d]^2D^o$	199.690	0.145	237.	0.97
$4d \rightarrow$	201.842	0.0010	1.6	0.007
$5d \rightarrow$	202.855	0.00072	1.1	0.005

The Li** emission spectrum has been studied in the 200-Å region.[15,17] Transitions from even-parity $^2P(n)$ states ($n=1,2,3,4$) into singly excited $1s^2mp$ ($m=2,3,4,5$) have been observed.[17] While $^2P(1)$ and $^2P(2)$ decay largely to $1s^22p$, it is found[17] that $^2P(3)$ and $^2P(4)$ decay mostly to $1s^23p$, in agreement with the theoretical predictions.[15] From the transition probabilities in Table III we see that prominent transitions from $^2D^o(1,2,3)$ to $1s^23d$ should occur at $\lambda=202.860$, 200.392, and 199.690 Å, respectively, but they have not observed so far.

In Table IV we compare theoretical and experimental wavelengths for some transitions between core-excited Li doublets. The prominent features of the emission spectrum of Li** doublets are illustrated in Fig. 1. The present scheme differs considerably from one given recently.[29] In the work of To *et al.*[29] the culprit turned out to be the medium intensity $\lambda=3490$ Å line which actually belongs to a transition between core-excited negative lithium quintet states.[21] The charge state of

TABLE IV. Comparison of theoretical and experimental wavelengths for some transitions between core-excited Li doublets.

Transition	Wavelength,[a] in Å	Wavelength (experimental)
$^2D^o(1) \rightarrow 1s2p^2\ ^2P$	3660.9(0.4)	3660.9[b]
$^2D^o(2) \rightarrow$	2995.0(0.3)	2994.1(0.3)?[b]
$^2D^o(3) \rightarrow$	2845.6(0.3)	2846(2)[c]
$^2D^o(1) \rightarrow 1s2p^2\ ^2D$	3144.5(0.3)	3144.5(0.2)[b]
$^2D^o(2) \rightarrow$	2640.3(0.2)	2640.1(1.0)[b]
$^2D^o(3) \rightarrow$	2523.4(0.2)	
$1s2p^2\ ^2P \rightarrow [1s2s2p\ ^3P]^2P^o$	4590	4590[c]
$[1s2p\ ^3P\ 3p]^2P \rightarrow$	2183	2183?[c]

[a] See discussion in text.
[b] References 7 and 22.
[c] Reference 4.

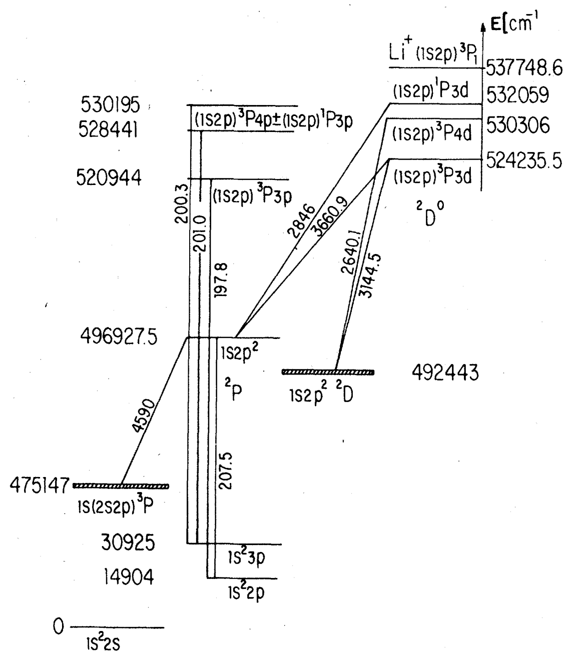

FIG. 1. Prominent features of the observed emission spectrum of Li** doublets.

TABLE V. Comparison of theoretical ($E1$) and experimental mean lifetimes, in nsec. Theoretical values larger than experiment suggest autoionization of the upper level.

State	$\tau(E1)$	τ_{expt}
$^2D^o(1)$	5.0 ± 0.5	1.5 ± 0.1 [a]
		(1.45 ± 0.1) [b]
$^2D^o(2)$	2.2 ± 0.2	0.51 ± 0.05 [a]
$^2D^o(3)$	0.040 ± 0.005	0.063 ± 0.05 [b]

[a] Reference 22.
[b] Reference 4.

species responsible for photon emission in the beam-foil source can now be easily determined by experiment.[23,24] In this regard, similar transitions between high-multiplicity bound states of negative ions are expected in other alkali, alkali-earths, chalcogens, and halogens.[30]

So far we have avoided any quantitative discussion about branching ratios. According to Mannervik,[31] the branching ratio of $^2D^o(1) \rightarrow 1s2p^2\,{}^2D$ to $^2D^o(1) \rightarrow 1s2p^2\,{}^2P$ is about 4. The results of our rather crude transition probability calculations (see Table III) yield a value of 2.8. Since $1s2p^2\,{}^2D$ is an autoionizing state, it is not entirely correct[32] to calculate branching ratios from transition probabilities which do not take into account all processes involved. It is nevertheless gratifying to verify that our simple calculation turns out to give the correct order of magnitude. With the above limitations in mind, we give in Table V mean lifetimes for $E1$ decay and compare them with experiment.[4,22] The larger theoretical estimates for the lifetime of $1s2p3d\,{}^2D^o(1)$ and $1s2p4d\,{}^2D^o(2)$ suggest autoionization of these levels. This is not surprising since Li $1s2p3d\,{}^4D^o$ is known to autoionize,[6] and Li $1s2p4d\,{}^4D^o$ possibly behaves likewise considering that its lifetime, $\tau = 1.11 \pm 0.05$ nsec.,[22] is too short for $E1$ decay in the uv-visible range of the spectrum. The experimental data for $^2D^o(3)$ is too uncertain to draw similar conclusions for this state.

The Li** doublets can also decay radiatively into their adjacent continuum.[33] Calculations[33] for the $1s2p^2\,{}^2P$ state show that radiative autoionization is negligible in this case.

IV. POSSIBLE SPIN-FORBIDDEN TRANSITIONS

If a quartet bound state mixes slightly with doublet states (through the relativistic interactions) it will participate in the faster decay modes of the companion doublets: (i) very rapid autoionization through doublets in the adjacent continuum, and (ii) spin-forbidden transitions through the small component of the bound-state doublets. (The opposite case, that one of a doublet slightly mixed with a quartet state is of less interest because of the larger transition probabilities for doublet decay.) The only intense transitions between Li** doublets in the beam-foil source originate from the decay of $^2D^o(1)$ to $1s2p^2\,{}^2D$ and to $1s2p^2\,{}^2P$ with $\lambda = 3144$ and 3661 Å, respectively. If the $^4D^o(1)$ state mixes with $^2D^o(1)$ considerably more than with its adjacent doublet continuum, very weak lines $\lambda = 3087$ and 3583 Å are expected. Since these lines have not been observed, the only significant modes of decay of Li $1s2p3d\,{}^4D^o(1)$ appear to be radiative decay in the quartet system and autoionization,[6] in comparable amounts. Spin-forbidden transitions are therefore ruled out in the Li** spectrum.

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