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LETTER TO THE EDITOR

The spin dependence of the excitation of metastable autoionising lithium ^{4}P states by electron impact

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Abstract. We measured the spin-dependent cross section asymmetry A for the formation of long-lived autoionising ^{4}P states of lithium by electron impact. The asymmetry is defined as

$$A=(\sigma^{\downarrow\uparrow}-\sigma^{\uparrow\uparrow})/(\sigma^{\downarrow\uparrow}+\sigma^{\uparrow\uparrow})$$

where $\sigma^{\downarrow\uparrow}$ and $\sigma^{\uparrow\uparrow}$ are the cross sections for antiparallel and parallel spins of the colliding electron and atom. The lifetime measurements of Feldman and Novick suggest that only the ^{6}Li(1s2s2p)^{4}P$_{5/2}$ substate contributes to our measured asymmetry. We obtain $A=-0.47$ with a statistical error of ±0.03 and a systematic error of $-0.05/+0.03$. This value is slightly lower than the theoretical expectation of $A(^4\mathrm{P}_{5/2})=-0.400$.

A deviation in this direction would occur if the lifetime of the ^{4}P$_{1/2}$ substate were not as short as has been measured in a strong magnetic field but rather were as long as that calculated for zero magnetic field.

In low-energy electron collisions with low-Z atoms the spin dependence of the cross sections is caused by electron exchange. The exchange interaction between the free electron and the valence electron of an alkali atom can be studied in crossed-beam experiments with spin-polarised electrons and spin-polarised atoms.

In such an experiment the polarisation asymmetry is determined from

$$A=(P_eP_a)^{-1}(N^{\downarrow\uparrow}-N^{\uparrow\uparrow})/(N^{\downarrow\uparrow}+N^{\uparrow\uparrow})$$

where $N^{\downarrow\uparrow}$ and $N^{\uparrow\uparrow}$ are the signal rates for antiparallel and parallel beam polarisations, respectively, P_e is the degree of polarisation of the electron beam and P_a that of the atomic beam. For completely polarised crossed beams ($P_a=P_e=1$), the signal ratio $N^{\downarrow\uparrow}/N^{\uparrow\uparrow}$ equals the cross section ratio $\sigma^{\downarrow\uparrow}/\sigma^{\uparrow\uparrow}$ and the formula above becomes that given in the abstract.

The free and valence electrons can either form a singlet (total spin $S=0$) or a triplet ($S=1$) collision complex. Since

$$\sigma^{\uparrow\uparrow}=\sigma_{\text{triplet}}(S=1,\ M_S=\pm1)$$

and

$$\sigma^{\downarrow\uparrow}=\tfrac{1}{2}\sigma_{\text{triplet}}(S=1,\ M_S=0)+\tfrac{1}{2}\sigma_{\text{singlet}}(S=0,\ M_S=0)$$

the asymmetry can be expressed as

$$A=\frac{1-r}{1+3r}\qquad\text{with}\qquad r=\frac{\sigma_{\text{triplet}}}{\sigma_{\text{singlet}}}.$$

Therefore, A can vary in two-electron collision complexes from +1 (pure singlet) to $-\frac{1}{3}$ (pure triplet). For example, in electron impact ionisation of H, Li and Na the asymmetry A_I of the ionisation cross sections, integrated over all scattering angles and

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partitions of energy, is almost +0.5 near threshold (but, however, only +0.25 for K), decreases with increasing energy and approaches zero at energies above 20 times the ionisation energy (Alguard *et al* 1977, Baum *et al* 1985); A_I never becomes negative.

However, the ionisation via electron impact excitation of an autoionising state can have a markedly different spin dependence. In a continuation of our experiment on the impact ionisation of spin-polarised ^{6}Li atoms by spin-polarised electrons (Baum *et al* 1985) we also investigated the autoionisation following the excitation of the state ^{6}Li(1s2s2p) $^4P_{5/2}$, a reaction channel which even at the maximum of its cross section contributes less than $\frac{1}{100}$ of the ions produced at those energies. To our knowledge this is the first polarisation experiment on the formation of autoionising states.

The metastable autoionising ^{4}P states of lithium were discovered by Feldman and Novick (1963, 1967). The lowest state (1s2s2p)^{4}P lies 57.3 eV above the (1s^{2}2s)^{2}S ground state. In the energy range above the first ionisation threshold at 5.39 eV (leading to the Li$^+$(1s^2)^{1}S continuum) and below the second ionisation threshold at 64.42 eV (leading to the Li$^+$(1s2s)^{3}S continuum) the ^{4}P states are metastable because they cannot decay by Coulomb interaction to the singlet ground state of the ion. The ^{4}P states with higher energy, e.g. the (1s2p^2)^{4}P state at 60.75 eV, will decay down to the lowest ^{4}P state (Feldman and Novick 1967). The fine-structure multiplet of this state exhibits 'differential metastability' by having different lifetimes for different J values. The substate with $J=\frac{5}{2}$ can only decay by means of spin-spin interaction into the (1s^2, εf)$^2F_{5/2}$ continuum; the substates with $J=\frac{3}{2}$ and $\frac{1}{2}$ can also decay to the (1s$^2\varepsilon$p) $^2P_{3/2}$, $^2P_{1/2}$ continua involving spin-spin and spin-orbit interactions, which lead to shorter lifetimes (Manson 1971). Levitt *et al* (1971) measured the lifetimes to be $T_{5/2}=5.8\pm1.2\ \mu$s, $T_{3/2}=0.46\pm0.10\ \mu$s and $T_{1/2}=0.14\pm0.07\ \mu$s. In the measurements reported here the metastable atoms travelled for about 9 μs between excitation and the detection region; therefore, we can assume that only atoms in the $^4P_{5/2}$ substate contribute to the ion signal.

There has been considerable scientific interest in these long-lived autoionising states for a variety of reasons. Metastable states offer the possibility of studying forbidden decay channels. Their small linewidth allows high-resolution measurements; their high excitation energy makes detection easy. Particularly the low-Z atoms provide interesting test cases because the theory is less complicated and selection rules are more rigorously obeyed than for heavy atoms. The (1s2s2p)$^4P_{5/2}$ state of ^{6}Li has also been discussed because of its usefulness in producing polarised nuclei (Levitt *et al* 1971). Furthermore, this state is of interest for the construction of XUV lasers (Harris 1980).

The ^{4}P states have been the subject of thorough investigations, both experimentally (Feldman and Novick 1967, Levitt *et al* 1971, Rassi *et al* 1977) and theoretically (Balashov *et al* 1967, Laughlin and Stewart 1968, Manson 1971). Our polarisation experiment provides a new way of investigating those states.

In order to excite a lithium atom from its (1s^{2}2s)$^2S_{1/2}$ ground state to a ^{4}P state (for example, to the lowest lying state (1s2s2p)^{4}P) by electron impact, core excitation and exchange with a core electron have to occur. The final state can be reached only if the excited core is in a triplet state ($S=1$). The spin of the valence electron then couples with the spin of the core to form quartet (or doublet) states. The evaluation outlined in table 1 leads to an asymmetry for quartet formation of $A(^4\mathrm{P})=-\frac{1}{3}$, the same value as for pure triplet scattering.

The collision time is on the order of 10^{-15} s. About 10^{-12} s after the inelastic collision the fine-structure (FS) coupling is strong enough to split the ^{4}P state into FS substates with $J=\frac{5}{2}$, $\frac{3}{2}$ and $\frac{1}{2}$ whereby the selection rule $\Delta M_L=0, \pm1$ is obeyed. If the Zeeman

Table 1. Evaluation of the asymmetry for quartet formation.

Initial spins of free and valence electron	Antiparallel		Parallel	
Initial spins, symbolic (1s1s)2s + ε	$(\downarrow\uparrow)\uparrow+\downarrow$		$(\downarrow\uparrow)\uparrow+\uparrow$	
Spin of free electron				
incoming	Down	Down	Up	Up
outgoing	Down	Up	Up	Down
Final spins, symbolic (1s2p)2s + ε	$(\downarrow\uparrow)\uparrow+\downarrow$	$(\downarrow\downarrow)\uparrow+\uparrow$	$(\downarrow\uparrow)\uparrow+\uparrow$	$(\uparrow\uparrow)\uparrow+\downarrow$
Probabilities for formation of core triplet	1/2	1	1/2	1
Probabilities for formation of atomic quartet	2/3	1/3	2/3	1
Product of probabilities	1/3	1/3	1/3	1
Sum of products	2/3		4/3	
Resulting asymmetry	−1/3			

levels with the three values of M_L become equally populated, the asymmetries for the different FS substates are all given by $A(^4\mathrm{P}) = -\frac{1}{3}$.

However, in our experiment the axis of quantisation was determined by a weak magnetic field orthogonal to the incoming electron beam, applied to stabilise the atomic beam polarisation. (If a polarised atomic beam goes through a zero-field region the polarisation is destroyed by Majorana transitions between adjacent levels of the Zeeman multiplet.) With this magnetic-field direction, the selection rule

$$\Delta M_L = \pm 1$$

applies for electron energies close to the threshold and the angular momentum algebra yields the following asymmetries: $A(^4\mathrm{P}_{5/2}) = -0.400$, $A(^4\mathrm{P}_{3/2}) = -0.067$, $A(^4\mathrm{P}_{1/2}) = -0.667$.

Our experiment was performed with 96% pure lithium-6 which has a nuclear spin of $I = 1$. The HFS coupling, which is established long after the FS coupling, then leads to substates $|F, J\rangle$; the asymmetry values for the excitation of these substates, however, depend only on the J value.

The apparatus used is similar to that used for measuring the dependence of the electron impact ionisation of alkali atoms on spin polarisation (Baum *et al* 1985). In a crossed-beam experiment polarised lithium atoms are ionised by polarised electrons. The interaction region is shown in figure 1. To measure the *ionisation asymmetry* A_I the produced ions are extracted by a small electric field toward the mesh (which is biased by $-2\,V$ with respect to the interaction region) and after traversing the mesh the ions are deflected into the channeltron detector. To study the *asymmetry* associated with the formation of *autoionising states*, however, the mesh is slightly positively biased (+5 V) to insure that all ions are repelled. Only the long-lived metastable atoms in the $^4\mathrm{P}_{5/2}$ state will travel through the mesh with the thermal velocities of the atomic beam (except for the small momentum transfer acquired in the excitation process). Some of them will autoionise beyond the mesh so that their ions can be pulled into the channel electron multiplied (CEM) and be registered.

The distance from the excitation region to the mesh is 1.8 cm. The lithium atoms travel with an average velocity of 2×10^5 cm s^{-1}. Thus they traverse the mesh and enter the detection region after about 9 μs, a time which is long enough for complete decay

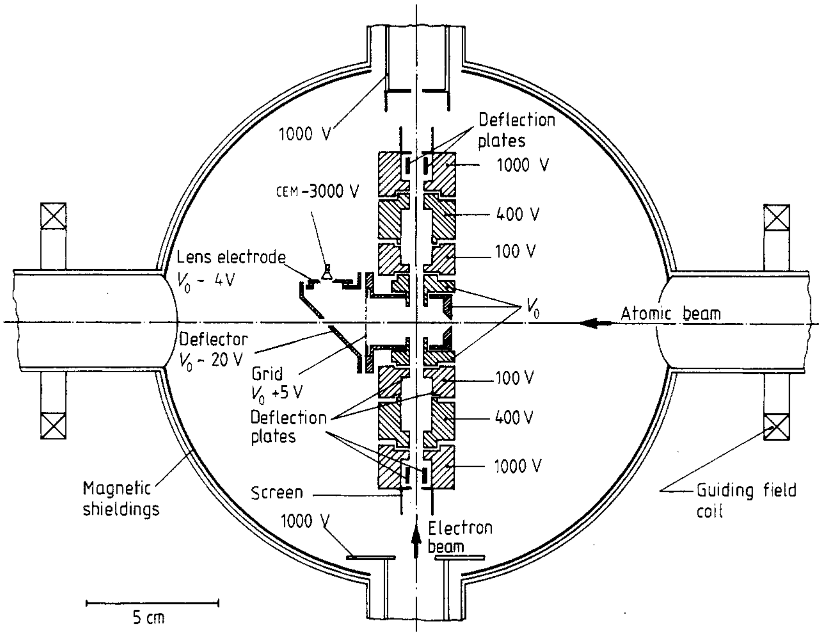

Figure 1. Cut through the interaction region where spin-polarised ^{6}Li atoms are excited or ionised by impact of spin-polarised electrons. The ions produced directly are repelled by the positively biased mesh. The metastable $^4P_{5/2}$ atoms travel with the ground-state atoms of the beam through the mesh, some of them autoionise between the mesh and the deflector and their ions are detected by the CEM.

of the atoms with $J=\frac{3}{2}$ and $\frac{1}{2}$ having measured lifetimes of 0.46 and 0.14 μs, respectively. Thus, it will only be atoms in the $J=\frac{5}{2}$ substate (lifetime 5.8 μs) which get into the detector region.

Figure 2 shows the ion yield as a function of incident electron energy, measured with a positively biased mesh. The excitation function shows peaks at 57.8, 60.5, 62.3 and 65-66 eV, features which were also present in the curve measured by Feldman and Novick (1967). The background signal (of about 390 counts) is thought to originate from beam electrons which are scattered through the mesh and then ionise ground-state lithium atoms as well as residual-gas atoms between mesh and deflector.

The result of our asymmetry measurements is given in figure 3. The error bars represent the statistical errors. In the data evaluation the background was assumed to have no asymmetry. The part of the background which comes from the atomic beam might at most have an asymmetry of $A=+0.15$ as we determined for direct ionisation at these energies (Baum *et al* 1985). This would lower the observed asymmetry further, a fact which is included in the systematic error quoted below, together with the 'scale uncertainty' due to the error in P_eP_a. The ion counting rate was about 100 s^{-1} for an incident electron current of 1 μA.

The four asymmetry values measured at different energies (figure 3) yield the average

$$A(^4\mathrm{P}) = -0.47 \pm 0.03 \text{ statistical error}$$
$$+0.03/-0.05 \text{ systematic error}$$

which lies below the theoretically expected value of -0.400. To calculate the expected A value we used the selection rule $\Delta M_L = \pm 1$ which is rigorously valid only *at threshold.*

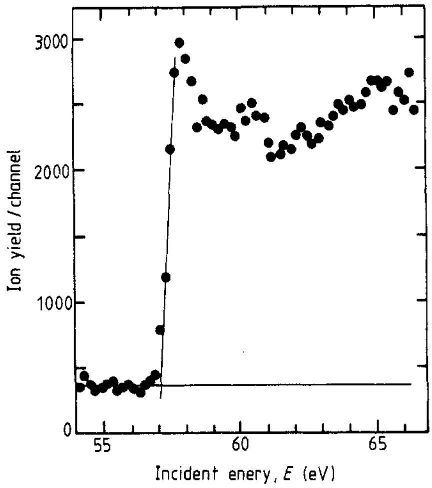

Figure 2. Ion yield plotted against electron energy. The energy width of the electron beam is 0.3 eV; the energy of the lowest-lying autoionising state was measured by Feldman and Novick (1967) as 57.3 ± 0.3 eV. This value was used by us to calibrate our energy scale.

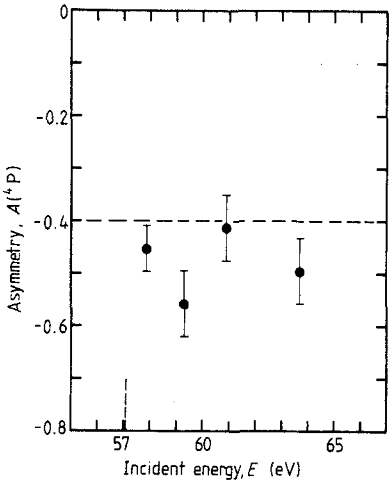

Figure 3. Asymmetry for the formation of autoionising atoms which decay between about 9 to 15 μs after formation measured at different energies. According to the measured lifetimes for the different fine-structure substates only $^4P_{5/2}$ atoms of the lowest ^4P state live long enough for detection in our experiment.

At higher energies, however, transitions with $\Delta M_L = 0$ are also expected and they should move the A values closer to $A = -\frac{1}{3}$, the result obtained for equal probability of the transitions with $\Delta M_L = 0$, $+1$ and -1. Therefore, the application of a no longer rigorously valid selection rule cannot account for a measured A value *below* -0.4.

The only theoretical A value which lies below -0.400 is $A(^4P_{1/2}) = -0.667$. For the $J = \frac{1}{2}$ substates Manson (1971) calculated very long albeit uncertain lifetimes (table 2, values marked by an asterisk). Since the doublet and quartet substates of the same J value autoionise to the same final state, their transition matrix elements add or subtract

Table 2. Estimates for $A(^4P)$, as measured in our experiment, based on the measured lifetimes of the states with $J=\frac{5}{2}$ and $\frac{3}{2}$ and the calculated lifetimes of the states with $J=\frac{1}{2}$ (lifetimes marked with an asterisk are theoretically uncertain).

State		Lifetime (μs)		Percentage of atoms surviving		Difference (atoms) ionising)	Statistical Weight	A_J	$A_{\rm av}$
F	J	Calc.	Measured	9 μs	15 μs				
7/2	5/2	5.88							
5/2	5/2	5.88	5.8 ± 1.2	34.1	16.7	17.4	9	−0.400	
3/2	5/2	5.05							
5/2	3/2	0.24							
3/2	3/2	0.25	0.46 ± 0.1	0.0	0.0	0.0	6	−0.067	
1/2	3/2	0.29							−0.428
3/2	1/2	100*	(0.14 ± 0.07)	94.0	90.1	3.9	2	−0.667	
1/2	1/2	29*		80.6	70.0	10.6	1	−0.667	

coherently, depending on the signs of the mixing coefficients. This then leads to anomalously small decay rates and correspondingly long lifetimes for the $^4P_{1/2}$ substates. The uncertainty comes from large cancellations in the computation.

Suppose for the sake of the argument that the atoms in the substates with $J=\frac{1}{2}$ would indeed live much longer than the lifetime measured by Levitt *et al.* (1971) In order to explain the measured average asymmetry value of $A(^4P)=-0.47$ in this way, one would have to assume that about one quarter of the detected signal ions (above the flat background, cf figure 2) come from atoms in the $^4P_{1/2}$ substates. The long calculated lifetimes lead to a somewhat smaller but non-negligible admixture of $J=\frac{1}{2}$ atoms (table 2) which shifts the A value to −0.43, a result which agrees with the measurement within the experimental errors.

The assumption of a lifetime which is longer than measured does not necessarily contradict the experimental results of Levitt *et al* (1971). These authors used a time-of-flight method for determining $T_{5/2}$ but inferred the values of $T_{3/2}$ and $T_{1/2}$ from the widths of anticrossing curves. Therefore, the lifetimes measured in this way are valid for rather high magnetic fields of 0.6–1T. As Manson (1971) pointed out 'a relatively small field would quench long-lived lower J states, so that even if our results are correct, it is unlikely that these states would be seen unless they were looked for in a field-free region'. In our experiment the magnetic field strength was only about 30 μT. Obviously our result for $A(^4P)$ does not prove the validity of the long lifetimes for the $^4P_{1/2}$ substates in a weak magnetic field. However, our speculative interpretation should stimulate further studies on these lifetimes.

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