K-Shell Photodetachment of Li^-: Experiment and Theory

J. D. Bozek[1], A.A. Wills[2], G. Turri[1,2], G. Akerman[1], B. Rude[1], H.-L. Zhou[3], S. T. Manson[3], N.D. Gibson[4], C.W. Walter[4], L. VoKy[5], A. Hibbert[6], R.A. Phaneuf[7], S.M. Ferguson[2] and N. Berrah[2]

[1]Lawrence Berkeley National Laboratory, Advanced Light Source, Berkeley, Ca 94720
[2]Western Michigan University, Physics department, Kalamazoo, MI 49008
[3]Department of Physics and Astronomy, Georgia state University, Atlanta, GA 30309
[4]Department of Physics and Astronomy, Denison University, Granville, Ohio 43023
[5]DAMAP, UMR 8588 du CNRS, Observatoire de Paris 92195, Meudon Cedex, France
[6]Queen's University of Belfast, Belfast, BT7 1NN, United Kingdom
[7] University of Nevada, Reno, NV89557

Introduction

Investigation of the dynamics in negative ions provides valuable insights into the general problem of the correlated motion of electrons in many-particle systems, such as heavy atoms, molecules, clusters and solids. Photoexcitation and photodetachment processes of negative ions stand out as an extremely sensitive probe and theoretical test bed for the important effect of electron-electron interactions because of the weak coupling between the photons and the target electrons. In addition, negative ions present a severe theoretical challenge since the independent electron model is inadequate for even a qualitative description of their properties. Finally, studies of the properties of negative ions are needed since their production and destruction strongly affects systems such as dilute plasmas appearing in the outer atmospheres of stars. It has long been thought that electron correlations mainly involve the outer shell electrons of a negative ion, and only outer shell correlations were considered in theoretical calculations. However, new theoretical [1] works including core-valence and core-core effects has recently led to a better agreement with experiments. The present work shows how all the four electrons of Li^- are strongly affected by correlations. Li^-, ground state $1s^2 2s^2$ (^1S), is one of the simplest negative ions. However, its extended nuclear core, as compared to H^-, has a profound effect on the resonance structure. The lifting of the degeneracy of different l states with the same quantum number n opens up new decay channels. Outer-shell structures in the photodetachment cross section of Li^- have been extensively investigated experimentally [2,3] and they were well explained by theoretical calculations [4,5]. However, up until a very recent calculation [6] no published work in inner-shell photodetachment of negative ions was available other than the inner-shell theoretical work in He^- [7,8]. With the advent of 3rd generation synchrotron light sources with higher flux, brightness and resolution, it is now possible to investigate experimentally inner-shell processes in tenuous negative ion targets.

Experiment and theory

In this work, we report dramatic structure measured and calculated for the K-shell photodetachment of Li^-. This process leads to a core-excited state of Li which decays predominantly to the Li^+ ion. The measurements were performed at ALS Beamline10.0.1, used in tandem with the photon-ion experimental apparatus [9]. Li^- ions were produced using a cesium sputtering source (SNICS II) [10]. The Li^- ion beam was accelerated to 11 KeV, and a flux of 20-150 nA reached the interaction region. The ions were merged collinearly with the counterpropagating photon beam in a 30 cm long energy-tagged interaction region, producing neutral Li atoms and Li^+ ions. Li^+ ions were detected as a function of photon energy, using a photon resolution of 75 meV. The resulting signal was normalized to the primary Li^- ion beam and the incident photon flux. In the case of negative ions, merged experiments are a serious challenge since the signal is very easily swamped by background noise due to stripping of negative ions with the residual gas (even though the background pressure in the interaction region was ~ 10^{-10} Torr) or with apertures in the ion beamline. In order to correct for the background ionization, the photon beam was chopped at 1 Hz and the photodetachment signal, corresponding to a relative cross section, was determined by subtracting the light-off signal from the light-on signal. The statistical error in the data was decreased by summing multiple sweeps of the photon energy of interest. The photon energies were calibrated separately using known resonance positions for neutral gasses and corrected for the Doppler shift, which amounts to about 108 meV for 60 eV photons. Calculations of the photodetachment of Li^- were performed using the R-matrix methodology which was enhanced to handle negative ions [11] and inner shells [12]. The discrete state input was generated with CIV3 [13]. A total of 29 target states were included in the close-coupling expansion: five $1s^2nl$ states, $n \leq 3$ and $l \leq 2$, and 24 1s2l3l', l=2,3, l'=0,1,2 core-excited states of Li. The cross section for Li^+ production for the Li^- photodetachment was obtained by summing all the cross sections for all of the channels leading to core-excited Li, since Li with a 1s-vacancy decays via Auger process virtually 100% of the time [14].

Results

The measured relative intensity was normalized to the magnitude of the calculated cross section at 62 eV. The calculated spectrum required only a small shift, +0.2 eV, to align to experimental data. It is interesting to note that this energy discrepancy is the same as that found in the case of photoionization of neutral Li [15]. The experimental and theoretical data are reported in Fig.1, where the neutral Li thresholds are reported.

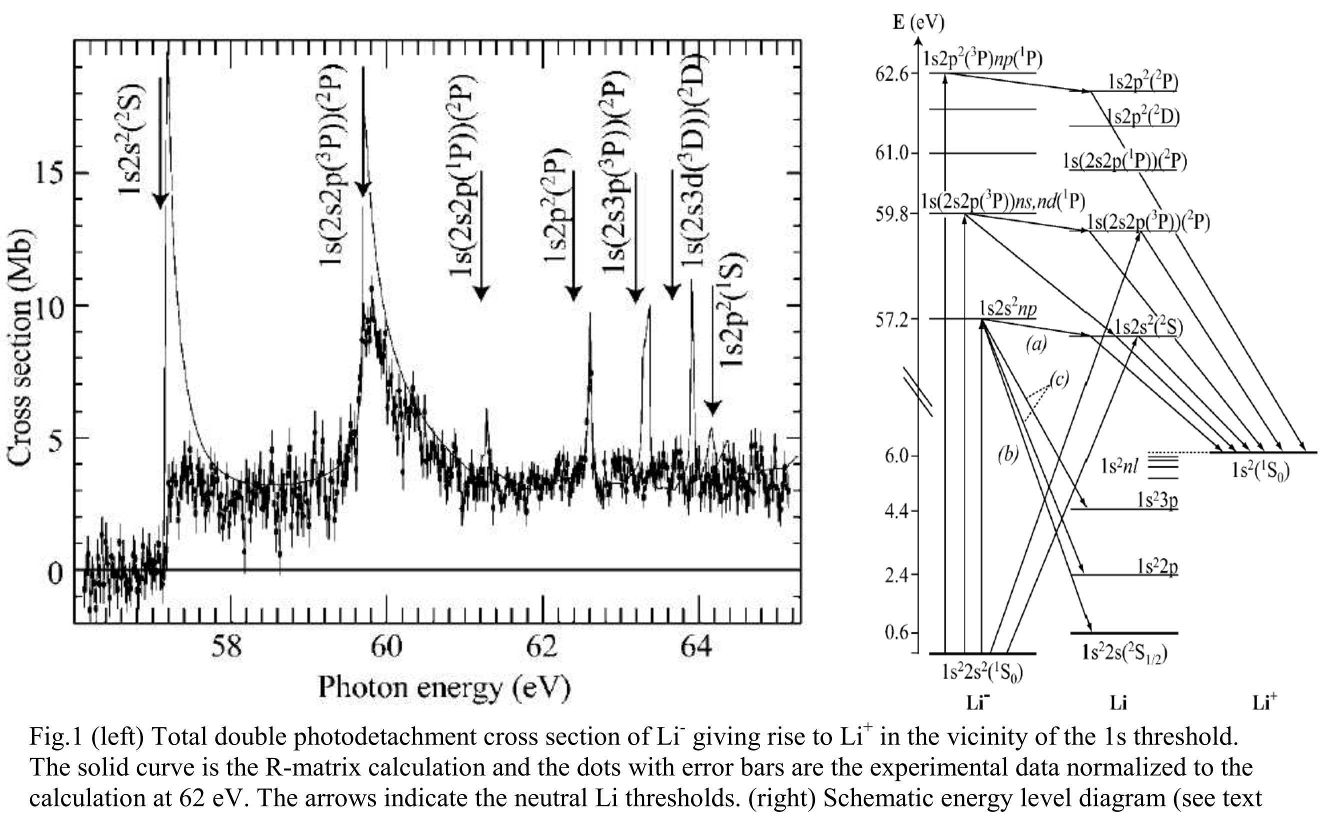

Fig.1 (left) Total double photodetachment cross section of Li^- giving rise to Li^+ in the vicinity of the 1s threshold. The solid curve is the R-matrix calculation and the dots with error bars are the experimental data normalized to the calculation at 62 eV. The arrows indicate the neutral Li thresholds. (right) Schematic energy level diagram (see text for details).

The experimental data clearly show three structures: first a step above the $1s2s^2\ ^2S$ threshold around 57.2 eV; second a shape resonance well defined by its sharp rise and decay tail above the second threshold $1s(2s2p\ ^3P)\ ^2P$; third a narrow resonance above the $1s2p^2\ ^2P$ threshold around 62.6 eV. Agreement between theory and experiment is quite good for the latter two structures, whereas the measured spectrum does not show the theoretically predicted shape resonance structure in the region above the first 1s detachment threshold. Li^+ can be produced in this energy region by autodetachment of the $1s2s^2np$ Li^- shape resonance to the core-excited $1s2s^2\ ^2S$ state of Li, which subsequently undergoes Auger decay to the ground state of Li^+, as labeled by process (a) in the energy level diagram of Fig, 1.

A similar process leads to the observed resonance structure above the second 1s threshold at 59.65 eV. In order for the $1s2s^2np$ Li^- states to produce Li^+, these states must be at higher energy than the first core-excited state of Li. As illustrated in the energy level diagram of Fig.1, this is the case for the $1s2s^2np$ excitations, which are found to be slightly above the $1s2s^2\ ^2S$ threshold for Li. Note that this situation is quite different from the case of neutral atoms, in the sense that there is not an infinite Rydberg series converging to each threshold. The lack of signal in the positive ion channel suggests that the $1s^2np$, $n>3$ states, omitted in the calculations, may play a significant role, since our results [16] are corroborated by an independent work [17].

It is surprising to find that the experiment was able to resolve and measure the predicted narrow structure above the $1s2p^2\ ^2P$ threshold around 62.6 eV, but not the predicted structures above the $1s(2s3p\ ^3P)^2P$ or $1s(2s3d^3D)^2D$ threshold above 63 eV or the weaker structure above the $1s(2s2p^1P)^2P$ around 61 eV. In the case of this last threshold, it may be that the signal to noise ratio is not sufficient to allow the observation of this weak structure. However the structure at 62.6 eV is expected to be stronger and narrower than the one at 63.2 eV, so at the moment we have no explanation for the discrepancy between experiment and theory in this region.

Conclusion

The first comparison between an experiment and theoretical K-shell study of the photodetachment of Li^- reveals dramatic structure, qualitatively and quantitatively unlike the same process in atomic Li or Li^+ ion. The calculations are able to predict the structures decaying to Li^+ in some cases, whereas the decay cross section is overestimated in the case of $1s(2s2p^3P)^2P$ and $1s2s^2\ ^2S$ thresholds. It is evident to us that although much of the essential physics of the inner-shell photodetachment problem is embodied in the calculation, there is still more to be understood, even in this simplest multishell negative ion.

Funding

DOE, Office of Science, BES, the NFS and NASA

Publication

N. Berrah *et al.*, K-Shell Photodetachment of Li^-: Experiment and Theory, Phys. Rev. Lett. 87, 253002 (2001).

References

[1] T. Andersen *et al.*, J. Phys. Chem. Ref. Data 28, 1511 (1999) and references therein

[2] G. Haeffler *et al.*, Phys. Rev. A 63, 053409 (2001) and references therein

[3] U. Berzinsh *et al.*, Phys. Rev. Lett. 74, 4795 (1995) and references therein

[4] C. Pan *et al.*, J. Phys. B 27, L137 (1994) and references therein

[5] E. Lindroth, Phys. Rev. A 52, 2737 (1995) and references therein

[6] H.-L. Zhou *et al.*, Phys. Rev. Lett. 87, 023001 (2001)

[7] D-S Kim *et al.*, J. Phys. B 30, L1 (1997)

[8] J. Xi and C.F. Fisher, Phys Rev. A 59, 307 (1999)

[9] A.M. Covington *et al.*, Phys. Rev. Lett. 87, 243002 (2001)

[10] R.D. Rathmell and G.A. Norton, Nucl. Instrum. Methods Phys. Res., Sect. B21, 270 (1987)

[11] H.-L. Zhou *et al.*, Phys. Rev. A 64, 012714 (2001)

[12] H.-L. Zhou *et al.*, Phys. Rev. A 59, 462 (1999)

[13] A. Hibbert, Comput. Phys. Commun. 9, 141 (1975)

[14] W. Bambynek *et al.*, Rev. Mod. Phys. 44, 716 (1972)

[15] S. Diehl *et al.*, Phys. Rev. Lett. 84, 1677 (2000)

[16] N. Berrah *et al.*, Phys. Rev. Lett. 87, 253002 (2001)

[17] B.H. Kjeldsen *et al.*, J. Phys. B 34, L353 (2001)

Principal investigator: Nora Berrah, Physics Department, Western Michigan University, Kalamazoo, MI 49008.
Telephone: (1) 616-387-4955. Fax: (1) 616-387-4939. Email: berrah@wmich.edu.