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# Comparison of the double- to single-photoionization ratio of Li with He

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We present precise double- to single-photoionization ratios and partial cross sections of lithium in the energy range from 81 to 110 eV. We compare our data with those of ground-state and metastable helium and find a remarkable similarity in the photon energy dependence of the double- to single-photoionization ratio of ground-state He when the energy axis is scaled in units of the energy difference between the double- and single-ionization threshold. We also present a simple explanation for this scaling behavior.

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## I. INTRODUCTION

The double-photoionization (DPI) process of helium was investigated thoroughly in the past [1,2]. Recent progress has led to a satisfactory agreement between experimental and theoretical data. Following helium, lithium as well as beryllium are only slightly more complicated systems but are already challenging targets for theorists and experimentalists. Therefore, DPI data of these targets are sparse compared to the amount of data available for He. For example, only recently experimental double- to single-photoionization ratios became available for Li [3,4] and Be [5]. In contrast to He, electrons from two different shells ($1s^{-1}2s^{-1}$) are ejected above the lowest DPI threshold of Li. We have measured precise double- to single-photoionization ratios as well as the total and DPI cross section of Li from threshold (81.03 eV) to 110 eV. Previous measurements [3] did not report the partial cross sections of Li below 100 eV and the error bars for the DPI ratios were larger compared to the results presented here. A recently performed experiment concentrated on the DPI threshold region (below 83 eV) only [4]. In the energy range below 140 eV only the simultaneous ejection of the $1s$ and $2s$ electrons contribute to the DPI cross section and not autoionization processes. Therefore, these data are well suited for a comparison with corresponding data for ground-state and metastable He.

## II. EXPERIMENT

The experiment was conducted at the Synchrotron Radiation Center. The measurements were carried out at the plane grating monochromator (PGM) undulator beamline [6]. Details of the setup can be found elsewhere [7]. Briefly, monochromatized photons enter the experimental chamber through a capillary and intersect Li vapor emerging from a resistively heated oven. The temperature of the oven is typically 450 °C, which corresponds to a Li vapor pressure of approximately $1\times10^{-3}$ mbar inside the crucible [8] whereas the background pressure in the vacuum chamber is in the low $10^{-8}$-mbar range during the experiment. At this temperature, the pressure of Li-dimers is a factor of 300 lower that of Li atoms [9]. The ions created are extracted by a pulsed electrical field across the interaction region, accelerated into a drift tube, and detected by a Z-stack MCP detector. By measuring the ions' flight time we obtain an ion-yield spectrum. In order to avoid dead-time problems with the electronics, we shifted the $^7\mathrm{Li}^+$ photoion peak outside the range of the time-to-amplitude converter and used the $^6\mathrm{Li}^+$ photoion peak to derive the DPI ratio.

## III. RESULTS AND DISCUSSION

The resulting $\mathrm{Li}^{2+}$:$\mathrm{Li}^+$ DPI ratio is shown in Fig. 1. The statistical error bars are at most the size of the plot symbols. Our ratios agree well with the data of Ref. [3] at higher photon energies; however, their data are systematically higher at lower photon energies, which becomes most obvious for their DPI ratios below threshold. Some of our spectra

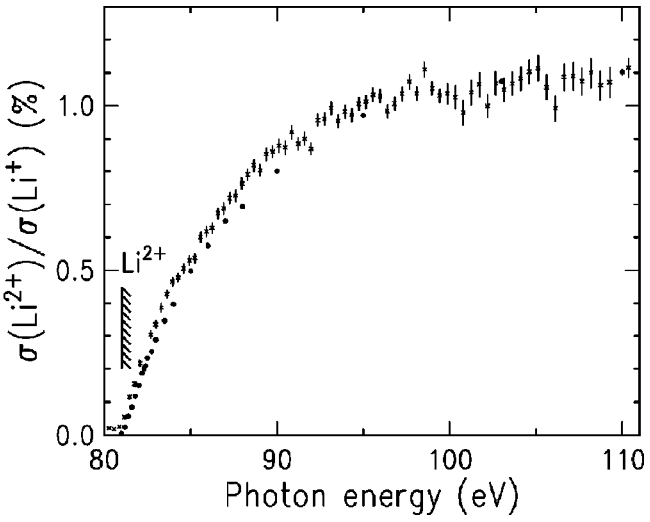

FIG. 1. Double- to single-photoionization ratio of Li (filled circles) together with data of Ref. [3] (crosses). Note that our statistical error bars are smaller than the symbol size.

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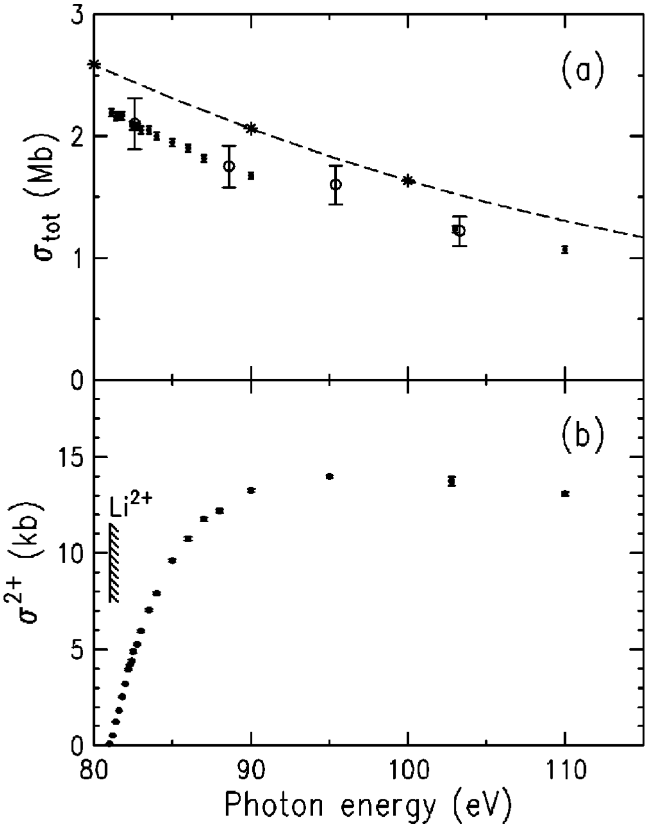

FIG. 2. (a) Total photoionization cross section of Li. Filled circles: this work; open circles: [10]; dashed line: [11]; asterisks: [12]. (b) Double photoionization cross section of Li.

were taken over a short period of time of a few hours, where the production of Li vapor can be regarded as constant, so that with the help of a known photon-flux curve the relative total cross section of Li could be determined. The photon flux was measured with an XUV100 silicon photodiode that has a known quantum efficiency. Our relative total cross section was normalized to an absolute cross section by using the value of Mehlman *et al.* [10] at 103.3 eV and is shown in Fig. 2(a). We find very good agreement with their cross-section data between 80 eV and 103 eV. A smooth curve through our total cross-section data was used to derive the Li$^{2+}$ cross section shown in Fig. 2(b).

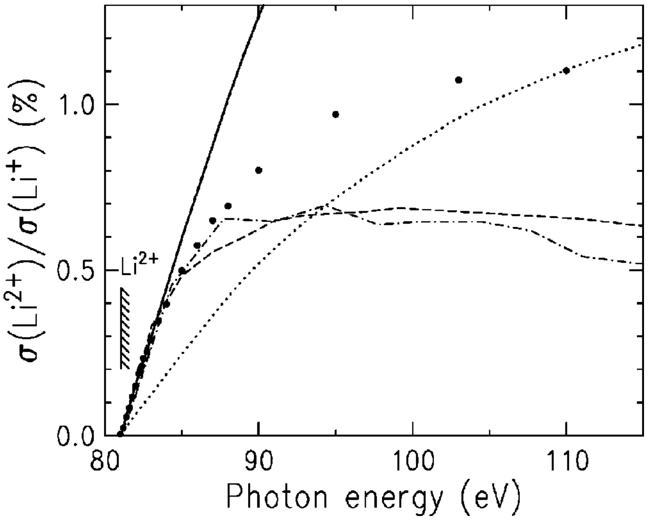

FIG. 3. Double- to single-photoionization ratio of Li (filled circles) and He (solid line) [13] as a function of excess energy. The He data (dotted line) [13] are scaled arbitrarily showing a different energy dependence than our Li data. Calculated ratios for metastable He$(1s2s)$ $^1S$ of Ref. [14] (dashed line) and of Ref. [15] (dotted-dashed line) scaled to fit the Li ratios at the lower energies.

In Fig. 3 we compare our Li double- to single-photoionization ratio with the one for He. We have chosen the He data of Ref. [13] because it is the most comprehensive set of data that is in overall agreement with the data of other authors (see references in Ref. [13]). While there is agreement for the first few eV, the He ratio rises to larger values at higher energies. The same He data divided by 2.5 (dotted line in Fig. 3) also show that the energy dependence is qualitatively different for Li compared to He and not just the same energy dependence on a different scale. Recent calculations for metastable He$(1s2s)$ $^1S$ [14,15] are also shown in Fig. 3 and are multiplied by 0.22 to match the Li ratio at lower energies. Metastable He$(1s2s)$ resembles Li if one assumes that the second $1s$ electron in Li is only screening one Coulomb charge, i.e., one regards it as an inert core. However, as Fig. 3 shows, this approximation may not be valid. The overall higher ratio for metastable He could be explained by the different binding energies of the two electrons in He and Li and, therefore, the different strength of interaction between them. Moreover, the additional $1s$ electron in Li manifest itself in a different energy dependence. While it is possible to align the ratios for Li and metastable He for the first 5 eV, there is no agreement at higher energies.

In general, different mechanisms contribute to the DPI process [16,17] namely, the shake-off (SO) mechanism, the two-step 1 (TS1) mechanism, and the ground-state correlation (GSC), which exhibit different energy dependences. Thus, the additional $1s$ electron in Li does not only affect the overall strength of interaction (as measured by the ratio) but also affects the contribution of the different mechanisms to the total double- to single-photoionization ratio.

As proposed in Ref. [5], we have plotted in Fig. 4 our DPI ratio on an energy scale in units of the difference between the DPI threshold and the single-ionization threshold in order to compare our results with those of He [13]. In the case of He, the energy unit is unambiguously determined since He has only one single- and one double-ionization threshold. In the case of Li, there is only one DPI threshold (81.03 eV [18]) in the region of interest, but three single-ionization thresholds.

The $2s$ photoionization contributes only marginally to the total photoionization cross section, and the corresponding $2s$ threshold is not considered here. The main contribution comes from the $1s$ photoionization, which can leave the Li$^+$ ion in either a $^1S$ or $^3S$ state. In Fig. 4 we have plotted our Li data using both the binding energy of the $^1S$ state (triangles connected with a dotted line) and the $^3S$ state (solid circles) for calculating the energy units $\Delta E$. As can be seen, the resulting data using the binding energy of the $1s$ $^3S$ (64.41 eV) agrees remarkably well with the corresponding He data after multiplying the He DPI ratio by 0.295 to fit the Li data. However, the same Li DPI ratios on a different en-

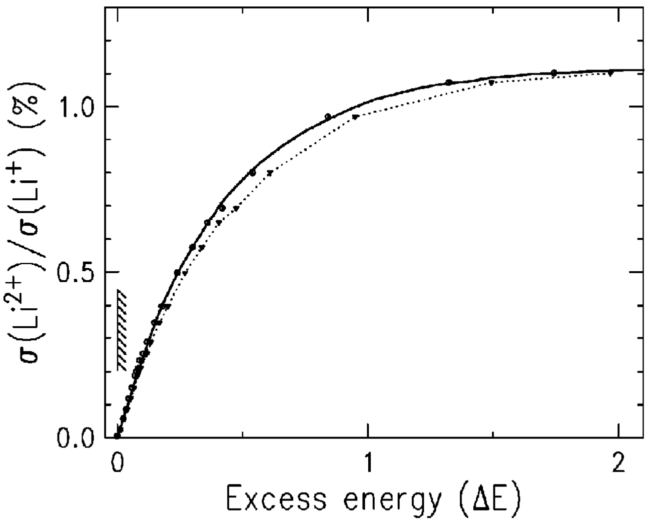

FIG. 4. Double- to single-photoionization ratio of Li (filled circles and triangles) and He (solid line) [13] as a function of excess energy. The triangles are connected by a dotted line for easier identification. The DPI ratio of He was multiplied to fit the Li data. See text for details.

ergy scale using the $1s\ {}^1S$ binding energy (66.31 eV) do not agree nearly as well as when using the $1s\ {}^3S$ binding energy. The deviation from the He data, particularly around an excess energy of 0.5 $\Delta E$ units, is clearly outside the error bars.

According to Samson [19], it is reasonable to assume that the DPI process not too far above threshold can be thought of as a two-step process: a photon is absorbed by one electron which, on its way out, collides with another electron, which may also leave the atom. This picture is also called TS1 [16] or half-collision model [20] and is one of the possible DPI mechanisms as mentioned above.

The possible spin orientations of the electrons in Li “during” the DPI process are shown in Fig. 5. As can be seen in Fig. 5(b), the $\mathrm{Li}^{2+}$ $1s^{-1}2s^{-1}\ {}^1S$ state can be created only after ionization of the $1s\ {}^3S$ single-ionization state without a “spin flip.” The obviously better agreement achieved when using the $1s\ {}^3S$ binding energy can then be explained by two facts. (a) The $1s\ {}^3S$ has a larger cross section than the $1s\ {}^1S$ [21,22]. (b) As calculations for metastable He($1s2s$) show [14,15], the ${}^1S$ state has a DPI probability three times higher than the ${}^3S$ state. Hence, we expect that in the case of Li, we are also more likely to end up in the ${}^1S$ DPI state than in the ${}^3S$ DPI state. Therefore, it is reasonable to expect that using the binding energy of the $1s\ {}^3S$ electron—which we think of as the precursor to the $1s^{-1}2s^{-1}\ {}^1S$ DPI state via the TS1 mechanism—yields a better agreement with the scaled He data. This agreement, on the other hand, is also an indication that the two escaping electrons are more likely to have an antiparallel spin [Fig. 5(b)] than a parallel spin [Fig. 5(a)].

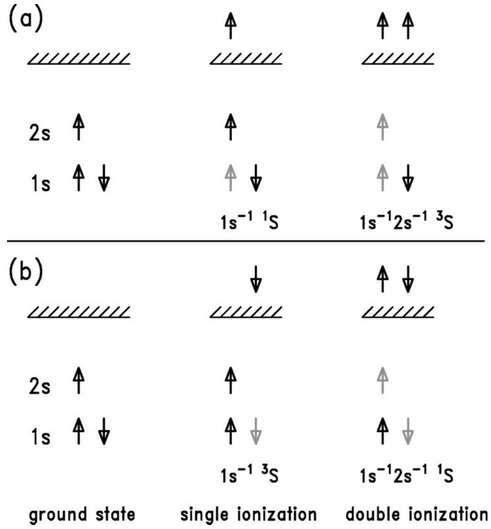

FIG. 5. Schematic diagram of possible spin orientations after single and double photoionization of ground-state Li. The black and gray arrows represent the spins of electrons and holes, respectively.

Although our simple model could be successfully applied to the $1s^{-1}2s^{-1}$ DPI of Li, the choice of energy units $\Delta E$ as the difference between double- and single-ionization threshold is not obvious. The reason why we scale the double- to single-photoionization ratios in units of $\Delta E$ is based on the similarity between the DPI process and electron-impact single ionization of a positive ion [19] for energies not too far above the DPI threshold. Furthermore, it is known that the single-ionization cross section by electron impact can be described by an empirical formula where the energy scales in units of the single-ionization potential [23,24]. In the case of a $\mathrm{Li}^{+}(1s^2)$ ion, the single-ionization potential is the energy $\epsilon$ necessary to remove one $1s$ electron. Since the final charge state is $\mathrm{Li}^{2+}$, this $\epsilon$ is the same energy as the double-ionization potential of neutral Li (81.0 eV) minus the ionization energy of the $2s$ electron (5.4 eV), because it is already removed in the case of a $\mathrm{Li}^{+}(1s^2)$ ion. In other words, $\epsilon$ and $\Delta E$ have the same values.

The energy dependence of the double- to total-photoionization ratio of an atom $A$ can be modeled by the appropriately scaled electron-impact single-ionization cross section of the corresponding singly charged ion $A^{+}$ [19] as long as the TS1 mechanism dominates the other mechanisms (SO and GSC):

$$\frac{\sigma_{\gamma}^{2+}(A)}{\sigma_{\gamma}^{tot}(A)} = F\sigma_{e}^{+}(A^{+}). \tag{1}$$

Here, $\sigma_{\gamma}^{2+}(A)$ is the DPI cross section of an atom $A$, $\sigma_{\gamma}^{tot}(A)$, the total photoionization cross section of an atom $A$, $\sigma_{e}^{+}(A^{+})$ the electron-impact single-ionization cross section of the ion $A^{+}$, and $F$ a suitable constant. Rewriting the equations yields

$$\sigma_{\gamma}^{2+}(A) = F\sigma_{\gamma}^{tot}(A)\sigma_{e}^{+}(A^{+}). \tag{2}$$

This equation makes it apparent that the cross section for DPI is given by the product of the total-photoionization cross section of the atom and the electron-impact single-ionization cross section of the “intermediate” ion. Hence, the DPI process can be written as

$$\gamma + \mathrm{Li} \rightarrow e^{-} + \mathrm{Li}^{+} \rightarrow \mathrm{Li}^{2+} + 2e^{-}. \tag{3}$$

Combining the scaling law for electron-impact ionization [23,24] with the similarity of DPI and electron-impact ionization [19], it is not surprising that the different DPI ratios scale with the energy $\Delta E$.

Note that at higher photon energies the proportionality of electron-impact ionization and DPI must fail because the TS1 mechanism is not the dominant process anymore and because of the different ratios for DPI and electron-impact ionization in the high-energy limit. While the electron-impact cross section will go to zero, the DPI ratio will reach a nonzero value. This also limits the applicability of the model to an energy region where the TS1 mechanism is the dominant process. As mentioned above, although metastable He resembles Li in some respect, the energy dependence of the double- to single-photoionization ratio is clearly different probably due to mechanisms other than the TS1 mechanism contributing to the total DPI process. Consequently, our model fails in the case of metastable He. On the other hand, however, this failure can also be seen as an indication that the SO and/or GSC mechanisms are more important for metastable He than for ground-state He near threshold.

## IV. CONCLUSION

To summarize, we have measured precise double- to single-photoionization ratios of Li in the photon energy range from 81 to 110 eV. We have also presented the total- and double-photoionization cross section in the same energy range. The energy dependence of the Li double- to single-photoionization ratio is different from the one for ground-state He and metastable He$(1s2s)$. Using a simple scaling model, based on the TS1 mechanism, to compare the photon energy dependence of our DPI ratios with the ones for ground-state He [13], we find excellent agreement confirming the previously proposed model [5].

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