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Resonant double photoionization of lithium studied with medium energy resolution

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We have measured the relative photoionization cross section for the formation of Li^{2+} ions between 148 and 161 eV photon energy with higher photon-energy resolution than in previous Li^{2+} studies. This energy region is characterized by double and triple excitations that lead to strong enhancements in the Li^{2+} cross section. As a result, the double-to-single photoionization ratio shows a dramatic resonance structure not seen before. We have determined the resonance positions and widths using Fano-profile fits to the Li^{2+} data and compare them to previously published values and a calculated Li^{2+} cross-section curve.

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I. INTRODUCTION

The ejection of two electrons from an atom following absorption of a single photon is one of the fundamental few-body processes in atomic physics. This process is due to electron correlation that can conveniently be probed by measuring the double-to-single photoionization ratio. Double photoionization of He has been studied extensively since it is the simplest atom that exhibits electron correlations. Following He, atomic Li represents the next level of sophistication because now intershell correlation is possible and, in contrast to He, there is more than one process to create a doubly charged Li ion. Above the first double-ionization threshold at 81.03 eV doubly and triply excited states can decay and produce typically doubly charged ions by emitting two electrons sequentially. These excited states have an empty $1s$ shell and are, therefore, referred to as "hollow" Li. The first observation of a photon-induced triply excited state was made by Kiernan *et al.* [1] employing the photoabsorption technique. Besides the earlier ion-yield measurements by Huang *et al.* [2] using a low energy resolution of 0.5 eV, no higher resolution study of the doubly charged ion yield has been performed in the double-excitation region (148–161 eV) of Li. Recent measurements [3] concentrated on the higher-photon-energy region and have only one resonance included in their study. In another investigation [4] the total ion yield of Li was measured and the various doubly and triply excited states identified. Kiernan *et al.* [5] have investigated the same energy region with high energy resolution, but did not measure any doubly charged ions. Photoelectron measurements and *ab initio* *R*-matrix calculations have been carried out for the decay of triply excited states by Diehl *et al.* [6,7]. The doubly excited states of Li^{+} have also been investigated employing Auger spectroscopy [8,9] which allows them also to determine the energy positions of *triply* excited states.

Calculations of the energy positions of some of the triply excited states are presented in Refs. [4,5] and *ab initio* calculations were performed by Vo Ky *et al.* [10]. We will compare their assignments and energy positions with our Li^{2+} data. Calculations within the *R*-matrix model of the resonances near the hollow Li threshold were performed by Berrington and Nakazaki [11]. Other *R*-matrix calculations were performed for different decay channels into Li^{+} nl final states by Journel *et al.* [12] for triply excited states between 142 and 159 eV along with experimental data around the first resonance at 142.3 eV. An overview of previous investigations on hollow Li can be found, e.g., in Ref. [13].

II. EXPERIMENT

The experiment was performed at the Synchrotron Radiation Center (SRC) using the new VLS-PGM (U2) [14] undulator beam line. We have used a 10 μm exit slit which yields a resolution of approximately 110 meV at 153 eV as determined from scans across the Kr $3d \rightarrow 5p$ and Ar $2p \rightarrow 4s$ resonances. We have measured the double-to-single photoionization ratio of atomic Li employing the ion time-of-flight method. The monochromatized photon beam passed through a differential pumping stage and intersected the metal vapor emerging from an oven heated to 420 °C [15]. The crucible of the oven was electrically biased to prevent thermal electrons from reaching the interaction region because those electrons can ionize Li by impact. A pulsed electric field was applied across the interaction region in order to accelerate the ions into the drift tube of our time-of-flight spectrometer that detects the ions by a Z stack of microchannel plates. By measuring the ion's flight time we obtained a charge-state-resolved ion-yield spectrum. The experimental parameters were chosen so that the detection efficiencies for both charge states were the same. We have acquired ion time-of-flight spectra of about 10 min for each photon energy. We have repeated some of the spectra and found consistent results. Note that we have used a 0.5 μm thin carbon filter to suppress any unwanted scattered (nonmonochromatized) light which would result in too low of a double-to-single photoionization ratio.

III. RESULTS AND DISCUSSION

Our double-to-single photoionization ratio of Li is shown in Fig. 1 along with previously measured data [2,3]. The agreement with the data of Huang *et al.* [2] is very good in the regions without narrow resonances, but their data clearly suffer from a lower energy resolution. We find that the double-to-single photoionization ratio of Li is about 1% and

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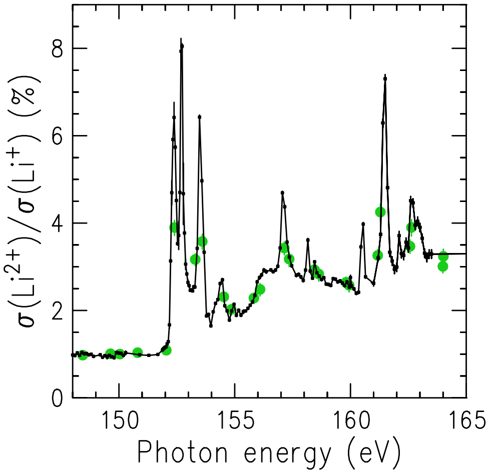

FIG. 1. (Color online) Double-to-single photoionization ratio of Li (data points connected by a solid line) and data points of Huang *et al.* [2] (gray filled circles). The statistical error bars of our data are smaller than the points representing the data. The data in the 161–164 eV region have been published previously [3].

remains essentially flat in the 140–152 eV photon energy range. Then, strong electron correlations enhance the ratio up to 8% on resonances while the nonresonant ratio increases only slowly to 2% at 160 eV. This pronounced resonance structure has not been seen before in the double-to-single photoionization ratio of Li because of the lower photon energy resolution used in the earlier experiment [2]. Even at higher photon energies the nonresonant ratio never exceeds 4.6% [3].

Now the following question arises: Are the resonances in the ratio due to resonances in the single- or double-ionization cross section? Using the well-known cross section of argon [16], we have determined the flux curve of the VLS-PGM beam line in the 140–161 eV range. Assuming a constant production of Li metal vapor, we can derive the relative double-photoionization cross section. Applying the known cross section for the production of Li^{+} ions [4] at 150 eV we can also calculate the partial cross section for Li^{2+}, which is shown in Fig. 2. This cross section is based on the absolute cross-section measurement by Mehlman *et al.* [17], which has an estimated error bar of 20%. As is known from previous measurements [4,5], even the two strongest resonances in the total (or Li^{+}) ion yield around 142 and 152 eV, respectively, exhibit an enhancement of the cross section of only a factor of 2, while the resonant enhancement at the other resonances in this energy region is much smaller. More pronounced, however, are the resonances in the Li^{2+} cross section considering the low nonresonant cross section. In this case, the cross section on resonance is enhanced by up to a factor of 15. Since the resonances in the Li^{+} cross section are weaker, the resonances in the ratio are mainly due to resonant enhancements in the Li^{2+} cross section.

In Fig. 3 we compare our experimental data and the data of Huang *et al.* [2] for the Li^{2+} cross section with the corresponding calculations within the *R*-matrix model by Berrington and Nakazaki [11]. The theoretical data are available only for the lower resonances and have been digitized from Fig. 1 in their paper (the curve may look more ragged than the actual calculation). As indicated in their paper, they expect a nonresonant double-to-single photoionization ratio of 2%. However, it is about a factor of 2 smaller [2,3]. After dividing their double-photoionization cross section by 2, not only the nonresonant cross section but also the lower resonances are approximately of the measured height. A correct comparison, however, requires a convolution of the theoretical curve with the experimental bandpass. This has not been done because the quality of the digitized theoretical data did not allow us to do it. We have energy shifted their theoretical data by 0.16 eV to match our *F* resonance. The *F* resonance has about the right height while the *G* resonance appears as too small and slightly shifted to lower energies (relative to the *F* resonance). The predicted resonance structure just below the *F* resonance is not visible in our Li^{2+} data; however, this Rydberg series of resonances have been seen for different single-ionization channels [7]. The *H* resonance appears at the right position but split into two resonances in their

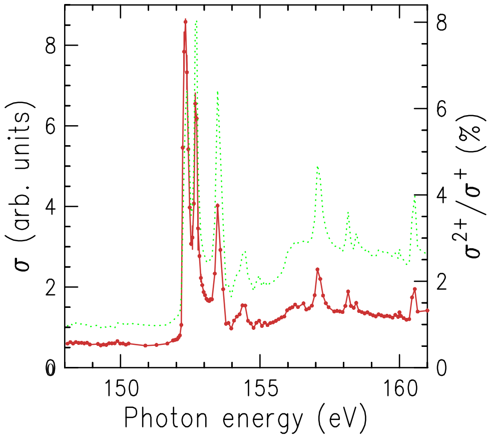

FIG. 2. (Color online) Relative photoionization cross section of Li^{2+} (solid curve). The statistical error bars of our data are smaller than the points representing the data. The double-to-single photoionization ratio (dotted curve) is shown for comparison with the corresponding scale on the right-hand side of the frame.

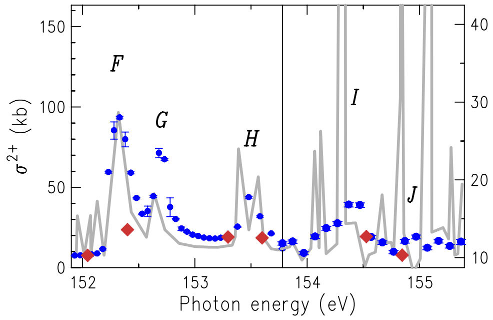

FIG. 3. (Color online) Our experimental (filled circles) and the calculated (solid line) Li^{2+} cross section data of Berrington and Nakazaki [11]. The diamonds represent the experimental data of Huang *et al.* [2]. The theoretical cross section was divided by 2.0 to match the experimental nonresonant cross section around 150 eV and shifted in energy by 0.16 eV to match the experimental *F* resonance. The resonances are labeled according to Azuma *et al.* [4]. Note the different cross-section scales for the left and right panels.

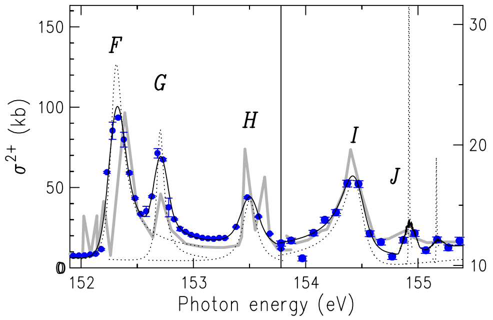

FIG. 4. (Color online) Fano-profile fit curve (solid line) to our Li^{2+} cross-section data. The individual, deconvoluted fit curves are shown as a dotted line. The resonances are labeled according to Azuma *et al.* [4]. The theoretical cross section [11] was divided by 2.0 to match the experimental nonresonant cross section around 150 eV and shifted in energy by 0.23 eV to match the experimental *G* resonance. The theoretical curve in the right panel was additionally smoothed and divided by 8 to match the experimental data. Note the different cross-section scales for the left and right panels.

theory. Also other measurements [4–6] did not resolve the *H* resonance possibly because the two components are too close to each other.

In the right panel of Fig. 3 we can see that theory predicts resonances (probably) converging toward the *I* resonance that is predicted to be rather narrow. The experimental data show a smooth increase of the Li^{2+} cross section due to the limited energy resolution. Noteworthy is the data point near 154 eV that shows the predicted dip in the cross section. The next higher resonance is hardly visible in the experimental cross section and the following *J* resonance is clearly visible but appears at a higher energy than the predicted resonance.

The resonance positions and widths were determined by a Fano-profile [18] fit to our double-ionization cross section data and the fit curves for the main resonances are shown in Fig. 4. This figure shows the total fit curve as well as the deconvoluted individual resonance fit curves. The deconvoluted curves mimic an ideal measurement with an infinitely high photon-energy resolution. Also included is the theoretical Li^{2+} cross section curve of Berrington and Nakazaki [11], which was energy shifted by 0.23 eV to match the *G* resonance and scaled down as described above. In the right panel of Fig. 4 we have smoothed the theoretical curve and scaled it down by a factor of 8 to get an approximate match to our data. The smoothing was performed in an attempt to mimic a theoretical curve convoluted with the experimental bandpass. With this scaling we get a reasonable agreement between the experimental and theoretical cross sections which means that the strength of these resonances is overestimated by theory. Since we aligned the theoretical curve to the *G* resonance, the *F* resonance seems to be predicted at too high of an energy. However, it is also possible that the Rydberg series of resonances on the lower energy side of the *F* resonance in the theoretical curve is affecting the experimental *F* resonance so that it appears at a lower energy than predicted.

Most of the resonances are slightly asymmetric and our monochromator bandpass is sufficiently narrow for the broader resonances while, e.g., the *J* and next higher resonance are too narrow to determine their width, even for the high-resolution measurements of Kiernan *et al.* [5]. As mentioned above the *H* resonance, predicted to consist of two resonances, has not been resolved in any experiment so far and the width of this resonance is in good agreement with the high-resolution experiment of Kiernan *et al.* [5]. The *I* resonance is predicted to consist of several resonances [11] that cannot be identified in the experiment and a fit of the overall shape of the experimental data was performed. However, the smoothed theoretical curve exhibits a shape that follows closely the experimental data.

The results of our Fano-profile fits are given in Table I. We compare our values with those obtained by Azuma *et al.* [4] from their total ion yield curve, by Kiernan *et al.* [5] from their Li^{+} ion yield curve, with the *ab initio* calculated energy positions by Vo Ky *et al.* [10], and with the experimental data of Diehl *et al.* [6,8].

The energy positions are in good agreement with Azuma *et al.* except for the resonances *C*, *H*, and *M*, with the latter resonance being weak in the Li^{+} channel and more prominent in the Li^{2+} channel. Our energy positions of the resonances *C* and *H* mentioned above are, however, in good agreement with the data of Kiernan *et al.* as well as the resonances *G*, *N*, and *O*. We found all resonances identified by Azuma *et al.* above the *F* resonance and some additional resonances, because they are more clearly visible in the double ionization cross section than in the total ion yield. There is a small discrepancy between our resonance energies and the energies of Kiernan *et al.* [5] for three resonances (*F*, *L*, and *M*). While the *L* and *M* resonances are rather weak and noisy in the Li^{+} spectrum of Kiernan *et al.*, we do not know the reason for the discrepancy in position for resonance *F*. Interestingly, their and our values for the width of this resonance are in good agreement. A possible problem may be the close proximity of the *F* and *G* resonance compared to their width and the fact that the *G* resonance is much smaller in the single-ionization channel while it is of similar size of the *F* resonance in the double-ionization channel.

The widths of the resonances are typically in the order of the monochromator bandpass and are all in good agreement with the high-resolution measurements of Kiernan *et al.* [5]. Some resonances, however, are so narrow (less than 10 meV) that even the high-resolution experiment of Kiernan *et al.* could not determine their width.

Turning to the assignments of the resonances given by Azuma *et al.* and Kiernan *et al.* with the latter being shown in Table I we find that the most prominent resonances in the Li^{2+} cross section (F,G,H,K) seem to have two of the three excited electrons in the same *nl* subshell $(2p)$, which results in a good overlap of the orbitals of those two electrons. This leads to a fast decay that manifests itself in a relatively broad resonance. The *G* resonance, which is rather weak in the Li^{+} cross section and has a $2s2p3s$ excited state according to Ref. [5], has strong admixtures of $2p^{2}5p$ and $2p^{2}4p$ [4] which can explain the strength of this resonance in the Li^{2+} cross section. Resonance *K*, which did not get an assignment in Ref. [5] but in Ref. [4], is mainly a mixture of a $2p^{2}3p$ and a $2p^{3}$ state. This would fit our expectation that two electrons in the same subshell lead to a particularly strong enhancement.

The assignment of the *C* resonance by Vo Ky *et al.* [10] is the same as the one given by Kiernan *et al.* However, its

TABLE I. Experimental resonance positions E_0 and widths Γ of resonances in the double-ionization cross section in eV. The resonances are labeled according to Azuma *et al.* [4] if possible. Included are the resonances in the Li^+ ion yield of Kiernan *et al.* [5] and the triply excited states detected by photoelectron spectrometry of Diehl *et al.* [6,8]. The energy positions of Vo Ky *et al.* [10] are *ab initio* calculations without any energy shift and are assigned to the experimentally found resonances according to their paper.

Resonance	Assignment Ref. [5]	This work		Ref. [5]		Ref. [4]	Ref. [10]	Ref. [6]	Ref. [8]
		E_0 (eV)	Γ (eV)	E_0	Γ (eV)	E_0 (eV)	E_0 (eV)	E_0 (eV)	E_0 (eV)
C	$2s2p3s$	149.90(7)		149.91(2)	0.08	149.79(3)	149.69	149.98(4)	
F	$2p^2 3p$	152.30(3)	0.128(3)	152.46(2)	0.13	152.32(3)	152.32	152.51(3)	
G	$2s2p3s$	152.70(3)	0.076(5)	152.75(5)		152.72(3)	152.57	152.90(5)	
H	$2p^2 4p$	153.50(3)	0.147(6)	153.54(5)	0.15	153.43(3)	153.35	153.66(5)	
I	$2s2p3d$	154.46(3)	0.215(21)	154.3(1)		154.43(3)			
J	$2s2p4s$	154.96(3)	<0.01	154.5(1)		155.0(1)			
		155.16(3)	<0.01						
K		157.08(3)	0.183(11)			157.0(1)			
		158.13(3)	0.067(15)						
		158.43(3)	<0.01						
		160.00(5)							
L	Unknown	160.55(3)	0.030(19)	160.71(5)	0.05	160.6(1)			160.65
M	$2s3s3p$	161.50(3)		161.66(5)	0.11	161.59(3)			161.52
N	Unknown	162.14(3)[a]		162.22(5)		162.2(1)			162.17
O	Unknown	162.65(3)[a]		162.62(5)		162.7(1)			162.47
		162.90(3)[a]							162.75

[a] This energy position was determined from the double-to-single photoionization ratio.

calculated energy position is at least 0.1–0.2 eV lower than the experimental energies, which is probably due to the difficulty to perform an *ab initio* calculation. This energy uncertainty may have resulted in giving the *G* resonance the assignment of the *F* resonance according to Kiernan *et al.*, because both resonances are closely spaced. Consequently, the *F* resonance has a different assignment in the paper by Vo Ky *et al.*, namely, $2s2p(^1P)3s$. This assignment does not meet our expectation of having two electrons in the same subshell and we believe that the assignments by Kiernan *et al.* are more reasonable. However, Vo Ky *et al.* and Kiernan *et al.* agree on the assignment of the well separated *H* resonance, although that "resonance" consists of two resonances according to calculations by Berrington and Nakazaki [11].

The energy positions of the triply excited states measured by Diehl *et al.* [6] employing Auger spectroscopy agree only for resonance *C* with our values. Interestingly, there are also slight discrepancies between their energy positions and those determined by others as shown in Table I.

The doubly excited states of Li^+ (i.e, not the triply excited states of neutral Li) found by Auger spectroscopy at fixed photon energies [8,9] can not immediately be related to the resonances in our data, because of the different pathways involved in creating a doubly charged ion. Possible pathways are schematically depicted in Fig. 5 and a more detailed energy level diagram is displayed in Fig. 1 of Ref. [9]. Because the differences between their and our experiments may not be obvious, the two different techniques are briefly described below.

Direct double photoionization (dot-dashed line) contributes to the nonresonant double-ionization cross section without producing an Auger line but will appear in an electron spectrum as a continuum.

The simultaneous emission of two Auger electrons (dashed line in Fig. 5) after resonant excitation will not appear as a line in an electron spectrum but rather as a continuum. However, it will be visible as a resonance in the doubly charged ion yield. A good example for it is the *A* resonance at 142.33 eV, which—for energy reasons—can

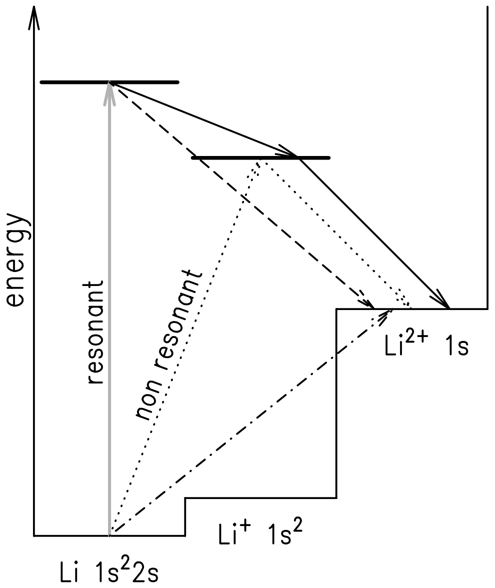

FIG. 5. Schematic energy diagram depicting different excitation and deexcitation pathways (see text for details).

decay only by the simultaneous emission of two Auger electrons [19] and does not produce any Auger line with a fixed kinetic energy.

On the other hand, satellite states (ionization with excitation) and double-shakeup states (ionization with double excitation) [8] above the double-ionization threshold can decay and produce Auger lines (dotted line) in an electron spectrum. This process is allowed at all energies above a certain photon energy, i.e., it does not require a resonant excitation, and thus contributes to the *nonresonant* double-ionization cross section and will not appear as a resonance in the doubly charged ion yield spectrum. This situation seems to be the case between 155.2 and 156.7 eV where we observe a smooth increase of the double-to-single photoionization ratio.

However, besides the nonresonant pathway (dotted line) there is (above 152 eV) a resonant pathway (solid line) via a triply excited state where two Auger electrons are emitted sequentially. The first Auger electron is emitted after the decay of the triply excited state into a doubly excited state of Li^{+}. The second Auger electron is then emitted, as in the case of a nonresonant excitation to Li^{+**}, leading to a doubly charged Li ion. The sum of the kinetic energies of both Auger electrons plus the energy of the final Li^{2+} state will correspond to the energy position of the triply excited state. However, it is not necessary to detect the first Auger line if one scans the photon energy while measuring the intensity of a particular Auger line. This has been done in the experiment of Ref. [8] where particular Auger lines show enhanced intensities whenever the doubly exited Li^{+} state is populated via the resonant pathway, i.e., the Auger electron emitted after nonresonant excitation to a doubly excited Li^{+} state appears at the same kinetic energy position as the second step Auger electron after resonant triple excitation.

The last column of Table I shows the energy positions of triply excited states by monitoring the Auger line intensity [8] that are published for the limited energy range of 160 to 163 eV in Table 2 of their paper. We find very good agreement between their and our energy positions for the *M* and *N* resonances and close agreement for the *L* resonance. However, there are some differences in energy for the *O* and higher resonance possibly due to the small intensity of the resonance.

IV. SUMMARY

In summary, we have measured the double-photoionization ion yield of Li between 148 and 161 eV photon energy and found good agreement with previous measurements [2,3] but also observed strong resonant enhancements of the double-to-single photoionization ratio not seen before due to the higher energy resolution used in this experiment. We have also derived the absolute double-photoionization cross section in that energy region. A comparison of our data with calculations by Berrington and Nakazaki [11] yields satisfactory agreement for the lower resonances except that theory seems to overestimate the double-photoionization cross section by a factor of 2. The higher resonances are much stronger in theory than in the experiment. However, they are also much narrower than the lower resonances so that the experiment may underestimate their strength due to limited energy resolution. Most of the resonances seen in the double-to-single photoionization ratio are due to enhancements in the double-photoionization cross section. These enhancements appear to be particularly strong when two of the three excited electrons are in the same subshell.

Employing Fano-profile fits to the resonances, we determined their energy positions and widths. Our energy positions of the resonances are overall in accord with the energies given by Azuma *et al.* [4] and Kiernan *et al.* [5]. We also find partial agreement with the resonance positions determined by Diehl *et al.* [6,8] using photoelectron spectroscopy. The resonance widths are in good agreement with the high-resolution measurement of Kiernan *et al.* [5]; however, only a few values are available for comparison.

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