Oscillations in the Double-Photoionization Cross Section of Li near Threshold

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The threshold region of the double-photoionization cross section of lithium was investigated using monochromatized synchrotron radiation and ion time-of-flight spectrometry. While the overall energy dependence can be described by the Wannier power law, we found oscillations in the cross section which are in good agreement with a modulated threshold law as proposed by Temkin [Phys. Rev. Lett. **49**, 365 (1982)]. This behavior may be due to the unequal binding energies of the electrons involved in the double-photoionization process.

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The double-photoionization (DPI) process is an interesting and also challenging subject in physics because the breakup of a Coulomb system into three particles cannot be described analytically (see, e.g., [1–5]). In particular, the threshold region, where both electrons move slowly and have time to interact, has attracted the interest of theorists and experimentalists trying to find numerical models of this seemingly simple process. Two conceptually different theories were developed, namely, the Wannier theory [6] and the Coulomb-dipole theory [7]. Refinements of those theories followed along with experiments that tried to decide between the two different models. One of the differences between these two models is the energy dependence of the near-threshold cross section σ. The Wannier theory [6] predicts

$$\sigma = \sigma_0 E_{\mathrm{exc}}^{\alpha}, \tag{1}$$

where α is the Wannier exponent, σ_0 a proportionality constant, and $E_{\mathrm{exc}} = h\nu - E_0$ the excess energy, $h\nu$ the photon energy, and E_0 the threshold energy. However, no prediction for its range of validity is given. The picture behind this model is that both electrons are ''traveling'' together along the so-called Wannier ridge through the atom and leave the atom with the same kinetic energy; i.e., their distances (r_1, r_2) from the nucleus are approximately the same ($r_1/r_2 \approx 1$). After this classical approach by Wannier, a semiclassical [8] and quantum-mechanical treatment [9] followed confirming Wannier's original work. Although the exponent in Wannier's law is undisputed, its range of validity is still a subject of investigations and may be too small to be experimentally accessible as was predicted for electron-impact ionization (EII) [10,11]. In addition, α may also depend on the excess energy [11]. A more detailed introduction to Wannier's work can be found in Ref. [12].

In contrast to the Wannier power law, the Coulomb-dipole theory by Temkin [7] predicts an *oscillating* but nevertheless monotonically increasing cross section near threshold of the form

$$\sigma \propto E_{\mathrm{exc}}\{1 - C\sin[a\ln(E_{\mathrm{exc}}) + \mu]\}, \tag{2}$$

with E_{exc} the excess energy, and C, a, and μ suitable constants. This model was developed for EII of atoms and for double photodetachment of negative ions. It is based on the notion that the faster leaving electron is subject to a dipole potential formed by the residual ion and the slower electron. In this case, the interaction of the faster electron is governed by the dipole potential and not the Coulomb potential, because the system of remaining ion and slow electron is essentially neutral. Equation (2) was later refined [13] by taking into account that the dipole moment changes during the process:

$$\sigma \propto E_{\mathrm{exc}}[\ln(E_{\mathrm{exc}})]^{-2}\{1 - C\sin[a\ln(E_{\mathrm{exc}}) + \mu]\}. \tag{3}$$

However, the additional logarithmic term in Eq. (3) applies only *extremely* close to threshold [14].

Early experiments, such as EII of H [15] and He [16,17] confirmed, or in the case of DPI of He [18] were at least consistent with Wannier's threshold law, but the experimental investigations continued stimulated by the development of the Coulomb-dipole theory. First attempts to verify Temkin's model showed possible oscillations in the double-photodetachment cross section of H^- [19], He^- [20], and K^- [21]. These experiments proved to be inconclusive, but a later analysis [22] showed that the results of both experiments reveal the presence of structure and are not in agreement with Wannier's power law. Another experiment [23] involving spin asymmetry in EII of hydrogen revealed oscillations which, however, agreed only marginally with the Coulomb-dipole theory.

In the case of DPI, Eqs. (2) and (3) are not strictly valid because one expects that, in addition to a ''dipole'' potential, the outer (faster) electron sees an additional Coulomb potential from the charge of the residual ion shielded by one unit by the inner (slower) electron. Nevertheless, the dipole potential, formed by the doubly charged ion and the slower electron, may affect the DPI cross section and Eq. (2) may still apply. Very recently, a preliminary

formula for DPI, which takes the nonzero net charge of the dipole into account, has been derived by Temkin [24]:

$$\sigma \propto E_{\mathrm{exc}}\{1 + CE_{\mathrm{exc}}^{1/4} \sin[D/E_{\mathrm{exc}}^{1/2} + a \ln(E_{\mathrm{exc}}) + \mu]\}. \quad (4)$$

Here E_{exc} is the excess energy, and C, D, a, and μ are suitable constants whose values have not yet been calculated for any particular system.

Intensive investigations of the DPI cross section of He were undertaken by detecting one of the ejected photoelectrons (see, e.g., [25–29]). The measured Wannier exponent was in agreement with the theoretical value of 1.056 assuming a flat energy distribution between the ejected electrons. So far, the only DPI experiments that tested the Wannier power law on atoms by measuring ions was performed on He [30] and atomic oxygen [31]. They confirmed the theoretical Wannier exponent and determined the range of validity as 2 eV above threshold. While several experiments confirmed the Wannier law, it was tested only for a few selected targets. However, there is still no clear evidence for Temkin's threshold law [32].

Li is different from He in two main aspects: (a) The electrons involved have very different binding energies, namely, 5.4 and 64.4 eV. (b) The two ejected electrons can have two different spin couplings, namely, $^1P^o$ and $^3P^o$. Note that both spin couplings have the same DPI threshold of 81.03 eV [33]. The only Li double-to-single photoionization ratios available [34] were taken with larger photon-energy steps, lower photon-energy resolution, and with larger error bars.

The experiment was conducted in two separate runs at the Synchrotron Radiation Center (SRC). In both cases measurements were carried out at the plane grating monochromator undulator beam line [35]. Monochromatized photons entered the experimental chamber through a capillary and intersected the Li vapor emerging from a resistively heated oven. The temperature of the oven was typically 450 °C. The ions created were extracted by a pulsed electrical field across the interaction region, accelerated into a drift tube, and detected by a microchannel plate detector. By measuring the ions' flight time we obtained an ion-yield spectrum. The photon flux was measured with an XUV100 silicon photodiode which has a known quantum efficiency. Details of the setup can be found elsewhere [36].

During our first study of the Li DPI cross section near threshold, we found first indications for oscillations in the cross section but the error bars were too large to reach a decisive conclusion. Therefore, we repeated the experiment with a monochromator resolution of 40 meV. Our relative total cross section was normalized to an absolute cross section by using the value of Mehlman *et al.* [37] at 103.3 eV. We find very good agreement with their cross section data between 80 and 103 eV [38]. A smooth curve through our total cross section data was used to derive the Li^{2+} cross section. In order to test for fluctuations due to changing contact potentials and changes in the Li vapor production, we took a spectrum at 103 eV every few hours. Also, the spectra were taken at different photon energies in random order over the course of three weeks.

First we tried to apply the Wannier threshold law to our data. One data point taken below the DPI threshold indicates a small background in the cross section data. Since we are concerned with only a very small energy region of approximately 2 eV, we assume this contribution to be constant and changed Eq. (1) to

$$\sigma = \sigma_0(h\nu - E_0)^{\alpha} + B_0 \quad (5)$$

for the fit procedure. Here, $h\nu$ is the photon energy, E_0 the apparent DPI threshold, and B_0 the constant background. The resulting fit curve is displayed in Fig. 1 along with the residuum (difference between data and the fit curve). The residuum exhibits oscillations which cannot be explained by noise because they are larger than the corresponding error bars. In principle, however, there is the possibility that the background B_0 originates from second-order light. This would be a serious problem because of resonances in the DPI cross section in the corresponding energy region of 162 to 165 eV [34,39,40]. However, these resonances do not match the observed pattern of oscillations. Moreover, the second-order light contribution was measured to be ca. 0.7% at 80 eV so that the resonances due to second-order light would appear smaller than the error bars of our cross section data. A possible source of the observed background is EII by thermal electrons which originate from the oven and are accelerated by 100 V of the electric pulse across the interaction region. Because the oscillations cannot be explained by second-order light, we applied Eq. (2) with an additional constant background B_0 to our data:

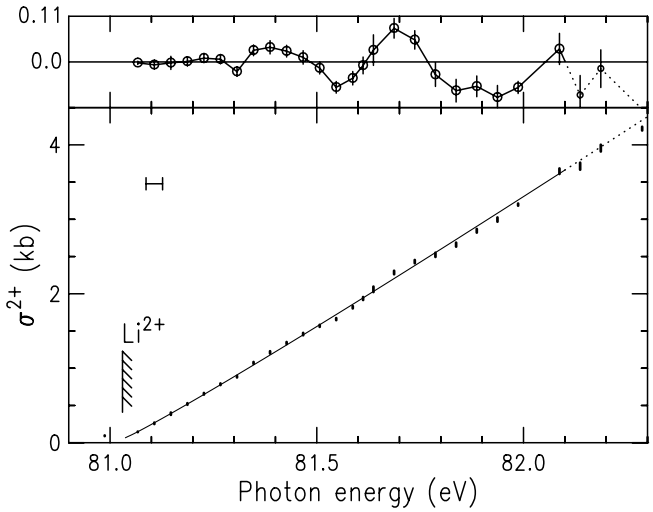

FIG. 1. DPI cross section of Li (points with error bars); fit curve according to Eq. (5): solid line; extrapolated fit curve: dotted line. The energy resolution is indicated in the upper right-hand corner. The upper panel shows the deviation of the data points from the fit curve.

$$\sigma = P(h\nu - E_0)\{1 - C\sin[a\ln(h\nu - E_0) + \mu]\} + B_0, \quad (6)$$

with parameters as described above and P a proportionality factor. We used the same values for E_0 and B_0 as in the previous fit procedure. The fit curve follows the energy dependence of our data fairly well for energies up to about 82.3 eV. As can be seen in Fig. 2, the maximal deviation of our data points from the fit curve is less than in Fig. 1. In order to compare the quality of the fit, we performed the fit with both the Wannier and Coulomb-dipole formula up to 81.8 eV. We obtain a χ^2 of 64.5 for the Wannier fit and 26.3 for the Temkin fit. Note that the period of oscillations get systematically larger as $h\nu$ increases in accord with Eq. (2). This is not what one would expect if the oscillations were an experimental artifact, particularly since the data was not recorded sequentially in time. This result shows that Temkin's dipole model [Eq. (2)] provides a reasonable description of our near-threshold cross section data.

As mentioned above, in case of DPI of a neutral atom, we have to take *both* the dipole potential and the Coulomb potential into account. Therefore, we used the following equation, based on Eq. (4) [24], as a fit model:

$$\sigma = P(h\nu - E_0)\{1 + C(h\nu - E_0)^{1/4} \times \sin[D/(h\nu - E_0)^{1/2} + a\ln(h\nu - E_0) + \mu]\} + B_0, \quad (7)$$

with parameters as described above.

As can be seen in Fig. 3, we achieve very good agreement between this preliminary formula derived by Temkin [24] and our data. For the same fit range as used before, i.e., up to 81.8 eV, we obtain a χ^2 of 21.7 which is a slight improvement over the previous fit where the dipole potential but not the Coulomb potential was taken into account. We used the same values for E_0 and B_0 as in the previous fit procedures, namely, $E_0 = 81.058$ eV and $B_0 = 9.42\times 10^{-2}$ kb. The resulting parameters for this fit are $P = 3.29 \times 10^{-3}$ kb, $C = 3.91 \times 10^{-2}$, $D = 280.8$, $a = -375.2$, and $\mu = 315.6$. The nonoscillatory part of Eq. (7), in contrast to Eq. (2), does not only have a linear energy dependence, $E_{\rm exc}$, but has an additional $E_{\rm exc}^{1.25}$ dependence which leads to a better description of the nonoscillatory energy dependence of the DPI cross section.

Assuming that we can apply the Wannier model [Eq. (5)] to our data, we performed least-squares fits for different energy regions with free parameters and then with fixed parameters except for α. The values obtained for α are shown in Fig. 4. Because of the oscillations in the cross section, α oscillates when using free parameters. A parabolic fit curve through these α values (gray curve in Fig. 4) guides the eye through the oscillations and indicates how the theoretical value of 1.056 may be approached near threshold. Using fixed parameters, α does not oscillate but remains slightly below the theoretical value before it reaches the theoretical value at energies below $\approx$81.35 eV. The proportionality factor σ_0 is 3.32(3) kb, which is about a factor of 3 larger than for He [30], demonstrating a much steeper rise of the DPI cross section for Li than for He.

In summary, we have carried out an extensive study of the DPI threshold region of Li. In contrast to He, where both electrons originate from the same shell, we have studied a system where one electron is tightly and the other one loosely bound to the nucleus. Unexpectedly, we have found oscillations in the DPI cross section that show the first indication of a dipole potential as proposed by Temkin. Although this threshold law was originally developed for EII of atoms and double photodetachment of negative ions, it describes the near-threshold DPI cross section fairly well, even better than the Wannier threshold law. A recently derived, preliminary formula for a

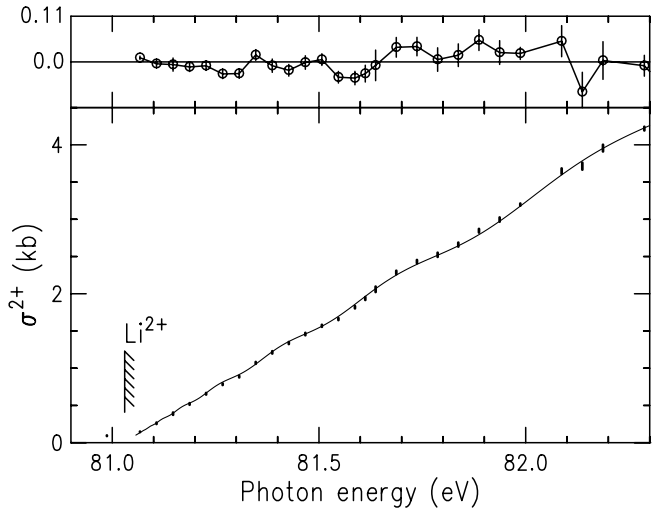

FIG. 2. DPI cross section of Li (points with error bars) and a fit curve according to Eq. (6). The upper panel shows the deviation of the data points from the fit curve.

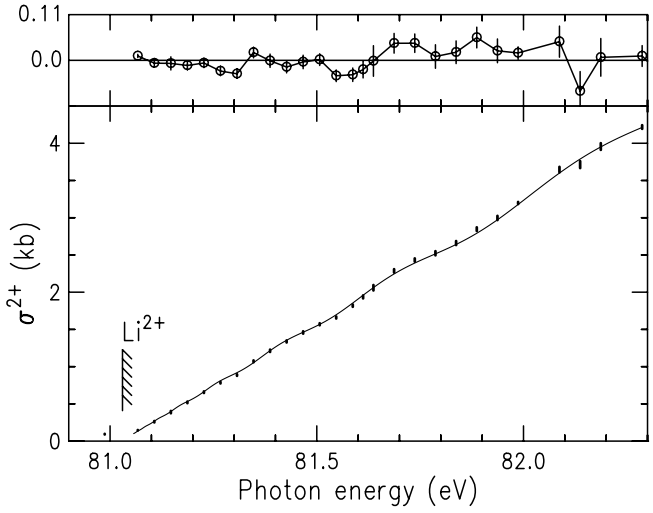

FIG. 3. DPI cross section of Li (points with error bars); fit curve according to Eq. (7). The upper panel shows the deviation of the data points from the fit curve.

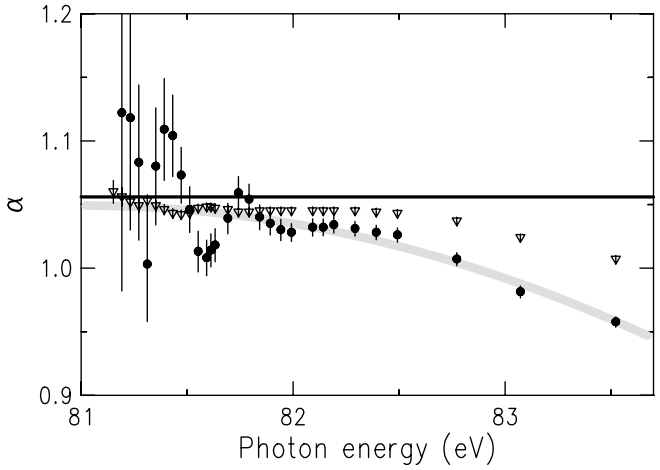

FIG. 4. The Wannier exponent α as a function of the upper limit of the fit range using Eq. (5) with free parameters (solid circles) and with fixed parameters except α (open triangles). The solid gray line is a parabolic fit curve to the filled circles. The horizontal line indicates the theoretical α value.

Coulomb-dipole model, which takes the net charge of this dipole into account, compares favorably with our data. A possible reason why we observe oscillations in the DPI cross section of Li is the large difference between the binding energies of the two ejected electrons. This energy difference could result in an unequal energy sharing even near threshold; i.e., one electron remains closer to the nucleus than the other.

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