Unexpected Behavior of the Near-Threshold Double-Photoionization Cross Section of Beryllium

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The threshold region of the double-photoionization cross section of atomic beryllium was investigated using monochromatized synchrotron radiation. The photon energy dependence of the double-photoionization cross section can be described by the Wannier power law up to 1.7 eV above threshold. However, we unexpectedly find oscillations in the cross section, which are in excellent agreement with a modulated threshold law based on the Coulomb-dipole theory [A. Temkin, Phys. Rev. Lett. **49**, 365 (1982)]. This new finding casts some doubts on the general applicability of the Wannier power law.

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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]). 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 models of this seemingly simple process.

In a recent investigation on near-threshold behavior of the Li ($1s^2 2s$) DPI cross section [2], oscillations were found in contrast to former DPI experiments on He [3] and atomic oxygen [4]. The oscillations in the Li^{2+} cross section were explained by the very different binding energies of the emitted $1s$ and $2s$ electrons. Obviously, the valence DPI process in Be resembles the one in He with two s electrons of the same binding energy in the valence shell rather than the one in Li with electrons emitted from different shells. Therefore, one expects a threshold behavior similar to the one for He. On the other hand, and in contrast to He, Be has still two electrons after the DPI takes place. Also, the $2s$ electrons have a much lower binding energy (9.323 eV) than the $1s$ electrons in He (24.587 eV), resulting in a quite low DPI threshold of 27.535 eV compared to 79.003 eV [5] for He.

Wannier has predicted that the DPI cross section σ rises *at* threshold according to a power law [6]:

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

Here, α is the "Wannier" exponent, σ_0 a constant, $E_{\mathrm{exc}} = h\nu - E_0$ the excess energy, $h\nu$ the photon energy, and E_0 the threshold energy. This theory makes the assumption that the initial conditions of the orbits are randomly distributed; i.e., the reaction zone can be ignored. It also assumes that both electrons have the same distance to the nucleus on their way out. Following Wannier's classical approach to the three-body Coulomb problem, other theorists using different approaches confirmed [7] but also extended Wannier's theory [8–10]. However, no prediction for its range of validity was given. While the exponent α in Wannier's law is undisputed, its range of validity is still a subject of investigation and may be too small to be experimentally accessible as was predicted for electron-impact ionization (EII) [11,12]. In addition, α may also depend on the excess energy [12]. An introduction to Wannier's threshold theory can be found in, e.g., Ref. [13].

Early EII experiments using H [14] and He [15] confirmed Wannier's threshold law. Later experiments on He, detecting one electron of the DPI process, were also in accord with a power law [16]. Except for the case of Li, the only two DPI experiments detecting the residual ion were performed on He [3] and atomic oxygen [4] and agreed with Wannier's power law.

Despite the seeming success of the Wannier threshold law, a conceptually different threshold law, namely, the Coulomb-dipole (CD) theory, was developed by Temkin for EII of atoms and for double photodetachment of negative ions [17]. This theory predicts, in contrast to the Wannier law, an *oscillating* but nevertheless monotonically increasing cross section near threshold. 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.

Similarly in the case of DPI, the slower electron forms together with the ion a "dipole" (but with a positive charge) so that the faster electron moves in a Coulomb *and* a dipole field. This dipole field, as any other attractive dipole-type interaction, causes a characteristic oscillatory-like behavior [10]. The final state wave function is thought of as the product of a Coulomb wave and a dipole wave. The dipole interaction is most pronounced in the (semiasymptotic) shorter range where the dipole wave function has a momentum-dependent cosine term. Thus, the oscillations are a phase effect coming from the dipole

wave of the outer electron and are *not* an interference effect [17,18].

Earlier 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 indeed reveal the presence of structure and are not in agreement with Wannier's power law. The conclusion, at that point, was that there is still no clear evidence for Temkin's threshold law [23]. In this Letter we report about the unexpected observation of oscillations in the DPI cross section of Be.

The experiment was carried out at the 4-m normal incidence monochromator of the Synchrotron Radiation Center (SRC). Photons with an energy resolution of about 30 meV at 28 eV intersected the Be vapor emerging from a resistively heated oven. Details of the experimental setup can be found in Ref. [24]. An ion-yield scan across the Ar $3s \rightarrow np$ resonances between 26 and 29 eV served as a photon energy calibration.

From the measured double-to-single photoionization ratio, we derived the DPI cross section by using the theoretical single-ionization data of Yeh and Lindau [25]. The energy dependence of their data agrees very well with our relative cross section data in the region of interest [26]. In order to test for fluctuations due to changing contact potentials and changes in the Be vapor production, we took a reference spectrum at certain photon energies every few hours. Also, the spectra were taken at different photon energies in random order to minimize the effect of systematical changes.

At first, we applied the Wannier threshold law [Eq. (1)] with an additional background B_0 to our data:

$$\sigma = \sigma_0(h\nu - E_0)^\alpha + B_0. \qquad (2)$$

Here, $h\nu$ is the corrected photon energy, and E_0 is the DPI threshold. B_0 is a constant background of 0.11 kb that takes into account a small contribution of second-order light. Note that there are no resonances in the DPI cross section below 115 eV. Another possible source of the observed background is EII by thermal electrons which originate from the oven and are accelerated by an electric pulse of 50 V across the interaction region. However, the thermal electrons were greatly suppressed by a small bias voltage applied to the crucible.

Both the fit curve as well as the theoretical curve with a Wannier exponent of 1.056, displayed in Fig. 1, agree well with our data below 1.7 eV. The fit parameter σ_0, using different fit ranges with all fit parameters free, turned out to be virtually independent of the fit range. After fixing σ_0 to an average value of 2.6 kb, we determined an average value for the background B_0. With σ_0 and B_0 fixed, we repeated the fit and determined the exponent α as a function of the fit range as displayed in Fig. 2. It is apparent from this figure that α drops down above ca. 1.7 eV, and the error bars do not overlap with the theoretical value for the exponent anymore. It is worthwhile to note that this drop around 1.7 eV is quite insensitive to the choice of fit parameters.

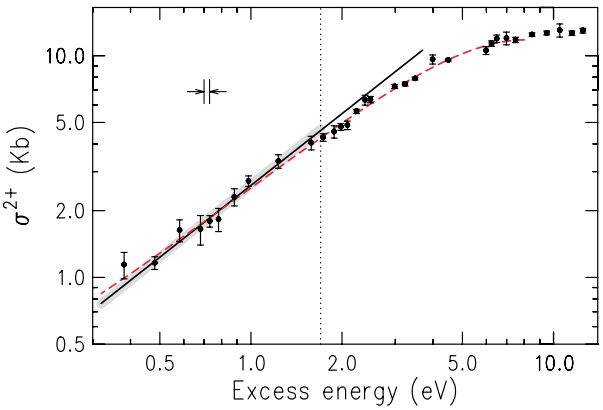

FIG. 1 (color online). Black solid line: DPI cross section of Be (points with error bars) and fit curve according to Eq. (2); gray line: theoretical curve ($\alpha = 1.056$); dashed line: fit curve according to fourth-order Wannier theory [9]. The energy resolution of 30 meV is indicated in the upper left-hand corner. The dotted line indicates the range of validity for the Wannier power law.

We also applied the fourth-order Wannier law [9] to our data and find good agreement for up to 8 eV. This fit curve, shown in Fig. 1, also provides a smooth curve through our data. The difference between that fit curve and our data exhibits an interesting oscillation, as shown in Fig. 3. This oscillation, in addition to the modulation in α (cf. Figure 2), prompted us to apply the CD theory to our data.

The original CD theory predicts oscillations in the cross section σ near threshold due to the dipole interaction of the form

$$\sigma \propto E_{\mathrm{exc}}\{1 - C\sin[a\ln(E_{\mathrm{exc}}) + \mu]\}, \qquad (3)$$

with E_{exc} the excess energy, and C, a, and μ are suitable constants.

However, this formula is only correct for EII and double photodetachment, i.e., pure dipole interaction in

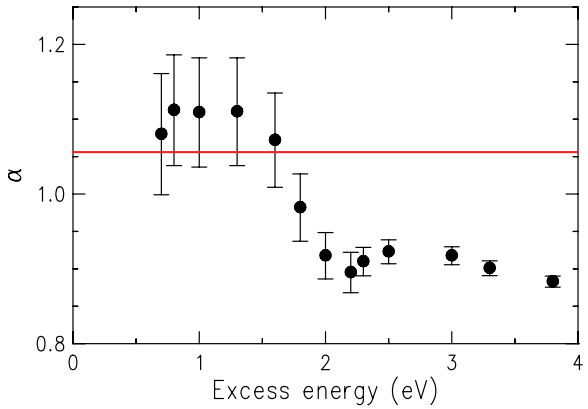

FIG. 2 (color online). The Wannier exponent α as a function of the upper limit of the fit range using Eq. (2) with fixed parameters except α. The horizontal line indicates $\alpha = 1.056$.

the ionization process. In our case of DPI, Eq. (3) is 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. A formula for DPI, which takes the nonzero net charge of the dipole into account, has been recently derived by Temkin [18]:

$$\sigma \propto E_{\mathrm{exc}}[1 + CE_{\mathrm{exc}}^{1/4} \times M(E_{\mathrm{exc}})] \tag{4}$$

with a suggested form of the modulation factor

$$M(E) \approx \sin[D \ln(E_{\mathrm{exc}})^2 + \mu]. \tag{5}$$

Here, E_{exc} is the excess energy, and C, D, and μ are suitable constants. The amplitude C is related to the overlap matrix element between the outer electrons of the neutral target and the Coulomb-dipole part of the two-electron final state. The constant D, which determines the "wavelength" of the oscillations, is related to the effective dipole moment. Note that Eq. (4) is slightly different from the one used in Ref. [2] due to the progress made in generating a correct formula. In practice, both curves appear to be very similar. We used the modulation factor $M(E)$ with its corresponding amplitude and an additional constant offset σ_0 to fit the difference $\Delta\sigma$ shown in Fig. 3; i.e., we used the formula

$$\Delta\sigma = CE_{\mathrm{exc}}^{5/4} \times M(E_{\mathrm{exc}}) + \sigma_0. \tag{6}$$

We subtracted the fourth-order Wannier curve (see Fig. 1) from our data because it provides a smooth curve through the data to higher energies than Eq. (4) does. As a result, our data points oscillate around zero with only a very small offset σ_0. The resulting fit curve using Eq. (6) represents the oscillations over a wide energy range very well, as can be seen in Fig. 3. Note that the period of oscillations becomes systematically larger as the excess energy increases in accord with Eq. (6). This is not what one would expect if the oscillations were an experimental artifact.

Using Eq. (4), we obtain an excellent fit to the Be^{2+} cross section data, as can be seen in Fig. 4. From various fits performed for different energy ranges, we conclude that the CD theory is applicable for energies up to 3.5 eV. At higher energies the fit curve is systematically too high.

The corresponding normalized χ^2 values for the Wannier and CD theories are listed for different fit ranges in Table I. It demonstrates that the CD theory is a better model to describe the near-threshold DPI cross section over an energy range of a few eV than the Wannier theory, although the fourth-order Wannier theory [9] describes the overall energy dependence without the oscillations quite well. However, these oscillations point to a new effect that has not been taken into account in the fourth-order Wannier theory.

The observation of oscillations is completely unexpected for Be. In the case of Li, the hypothesis for an oscillating cross section was based on the different binding energies of the two ejected electrons [2]. This is obviously not the case here. However, Li as well as Be have at least one electron with a relatively low binding energy as compared to He and O. These loosely bound electrons are little affected by the nucleus but rather by electron-electron interactions. Therefore, we expect that oscillations in the near-threshold DPI cross section should exist for all atoms with at least one loosely bound electron, namely, for all alkaline and alkaline-earth metals and for several other atoms. Comparing Be with Li, we find that the oscillations have an about 2 times larger amplitude for Be than for Li with a similar wavelength. The reason why the amplitude is clearly larger for Be may be due to the fact that Be has *two* quite loosely bound electrons in contrast to Li, which has only one.

In summary, we have measured the relative DPI cross section of Be in the threshold region. Surprisingly and against all expectations, we observed oscillations in the DPI cross section. This effect was observed for Li [2] and

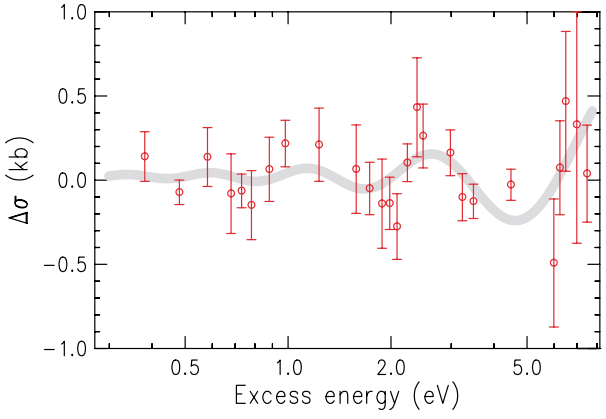

FIG. 3 (color online). Difference between our DPI cross section data and the fourth-order Wannier curve shown in Fig. 1 (points with error bars). The gray fit curve corresponds to the modulation term of the CD theory [Eq. (6)].

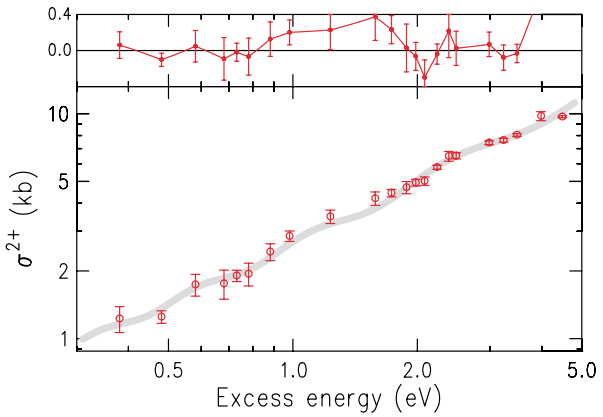

FIG. 4 (color online). DPI cross section of Be (points with error bars); fit curve according to Eq. (4). The upper panel shows the deviation of the data points from the fit curve.

TABLE I. Normalized χ^2 values for different upper fit limits (E_{max}) and models. W1: Wannier's power law with all parameters free; W2: Wannier's power law with only α free; F: fourth-order Wannier theory [9]; CD: CD formula for DPI.

E_{max} (eV)	W1	W2	F	CD
1.2	0.40	0.43	0.41	0.49
1.7	0.46	0.49	0.43	0.41
2.5	0.77	1.17	0.86	0.39
3.5	1.48	1.65	0.84	0.71

was ascribed to the fact that the electrons escaping from the Li ion have very different binding energies. However, our new data show that it is indeed a more general effect which may be observable for many atoms. The oscillations can be well modeled by the CD theory, which describes the threshold DPI cross section better than the Wannier threshold law. An approximate formula for a CD theory, which takes the net charge of the dipole into account [18], compares very favorably with our data. The Wannier power law gives a reasonable fit up to 1.7 eV above threshold, whereas the CD theory is applicable over 3.5 eV. From that we conclude that the Wannier threshold law may be applicable only very close to threshold, as suggested in Ref. [12], while for slightly higher energies an oscillatory structure emerges depending on the binding energies of the electrons involved. Further investigations of other few-body Coulomb systems are called for.

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