RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 48, NUMBER 3 SEPTEMBER 1993

Probing electron correlations in double photoionization of He at intermediate energies

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(Received 15 April 1993)

The ratio of double-to-single ionization of He has been measured between 280 and 1210 eV to investigate its behavior in this partially unexplored region. These measurements, compared with the most recent theories of Pan and Kelly (private communication) and of Hino [Phys. Rev. A **47**, 4845 (1993)], show the importance of including not only ground-state but also final-state correlations, in contrast to the high-energy behavior discussed by Dalgarno and Sadeghpour [Phys. Rev. A **46**, R3591 (1992)] where consideration of final-state correlations proves inessential. Our intermediate-energy results also appear to indicate the importance of including higher-order effects in the theory.

PACS number(s): 32.80.Fb

Accurate theoretical predictions of the energy dependence of the double photoionization of He are a fundamental problem in atomic physics that requires correspondingly accurate solutions of the Coulomb three-body problem. Since in photonionization, one photon can interact directly with only one electron, double photoionization can proceed only by electron-electron interactions. Although very recently there has been a great deal of progress experimentally and theoretically, both near and far above threshold, a number of unanswered questions remain at intermediate energies. Principal questions that arise are: (1) How does the interplay of electron correlations, in both the initial state and final states, affect the behavior of the ratio of double-to-single ionization in the intermediate energy range? (2) What is the relative importance of the basis set, and explicit consideration of higher-order correlation effects?

From the early 1980s until the present, the threshold region has been extensively investigated. In a many-body perturbation theory (MBPT) approach, the importance of choosing an appropriate basis set, ground- and final-state correlations, and of considering appropriate higher-order diagrams was shown by Carter and Kelly [1]. They found the velocity form of their MBPT calculations to be the most accurate, correctly describing the available data [2–6]. More recently, in the high-energy limit, measurements of the ratio of double-to-single photoionization at several photon energies, from 2 to 12 keV have been reported [7], consistent with an asymptotic value of 1.66%. Even though good agreement with this value of the ratio was obtained by older shake calculations [8,9], a new MBPT calculation [10] showed that this agreement may be fortuitous. With a procedure that uses the *acceleration form* of the dipole operator [11], it was only last year, after much debate over different and in part conflicting theories [8–12], that an understanding of the relative importance of the different processes in double photoionization in the asymptotic limit was achieved. In particular, in this limit only ground-state correlation need be considered when using the acceleration gauge (cf. Dalgarno and Sadeghpour [11]). From about 2 keV up, new theoretical studies of asymptotic behavior considering also the impact of Compton scattering by Andersson and Burgdörfer and by Samson, Greene, and Bartlett [13] have been made. In contrast to the threshold and high-energy regime, for intermediate energies only few reliable theoretical values exist. Very recent theoretical calculations [14,15] have investigated this range where the available data [16,17] to test these differing theories are also extremely scarce, in fact previously nonexistent from 560 eV to 2 keV. The only recent data up 560 eV were obtained by Bartlett *et al.* These data were scaled by them by a factor of 1.3 [17] in order to produce agreement with well-known threshold data.

With the threshold and asymptotic limit results better established than before, further progress in understanding requires answering a leading question: What are the dominant correlation effects in the energy gap between threshold and below about 1500 eV, which force the ratio to undergo a significant decrease from about 5% to 2% before settling slowly into the measured [7] asymptotic limit of about 1.5(2)%? Therefore, measurements in this intermediate-energy range are of critical importance because they test the capability of the *ab initio* calculations to describe the transition between the low- and high-energy regime in an adequate way. We present such benchmark data here.

In this Rapid Communication, we report on measurements between 280 and 1210 eV that test the most recent theories of Pan and Kelly [15] and of Hino [14] in order to understand the dominant electron correlation effects in this almost unexplored energy region.

The present measurements have been performed at the Berliner Elektronenspeicherring-Gesellschaft für Synchrotronstrahlung m.b.H (BESSY). Monochromatic light from the high-energy toroidal grating monochromator beamline (HE-TGM-1) operated by the Fritz-Haber-Institute [18] was tuned to several photon energies, $h\nu$,

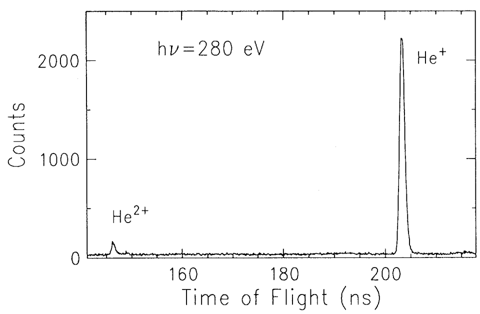

FIG. 1. Helium time-of-flight spectrum after photoionization with 280-eV photons.

from 280 to 1210 eV. Since the technique used is similar to the one used previously [7], discussion here is brief. He ions produced in the interaction region were analyzed by a 4.5-cm-long time-of-flight spectrometer adapted to enable the measurement of both He^{+} and He^{2+} within the 208-ns spacing of electron bunches in the ring. Both He^{+} and He^{2+} peaks recorded at $h\nu=280$ eV are shown in Fig. 1.

Several experimental effects can result in an inaccurate determination of the ratio of double-to-single ionization of helium [7]. In the present experiment, special emphasis has been put on suppressing possible higher-order and stray light contributions to the ion signal caused by the monochromator [19,4]. Both of these effects produce spurious He^{+}, and therefore contribute to giving a lower ratio than one would measure if the source were "clean" of these two effects. In order to take into account the light effects, we followed previous authors [19,4,17] and took two sets of measurements: (1) stray light effects were suppressed by using a succession of filters to absorb unwanted low-energy photons and by making the measurements just below the edge of each filter used. For example, at 280 eV, taking measurements with and without a carbon filter resulted in increasing the He^{+} signal by a large amount and therefore lowering the ratio of $\sigma(He^{2+})/\sigma(He^{+})$ by a factor of 8. (2) Higher-order light effects were taken into account, by taking measurements just above the edge of the appropriate filter. With this arrangement, and at the $h\nu$ selected, one would expect no creation of ions. If then an ion signal is recorded, it is attributed to higher-order light effects. For example, at 280 eV, we observed a small amount of He^{+}, which lowered the ratio by 4%. All the data were corrected accordingly. The filters used were C (thickness ~ 266 $\mu g/cm^2$), Ti (thickness ~ 200 $\mu g/cm^2$), and Cu (thickness ~ 318 $\mu g/cm^2$). At these thicknesses, the filters should have about 60% transmission [20]. We found that the Cu filter was very effective throughout the energy range of interest [20] and so it was used for the high-energy points. Photon energies were calibrated against the Ar $2p_{3/2}$-4s resonance at 2.44.4 eV, the π^* resonance in N_2 at 401.1 eV, the Xe M_V edge at 676.4 eV, the Ne K edge at 870.2 eV, the Xe M_{III} edge at 940.6 eV, and the Xe M_{II} edge at 1002.1 eV.

Additional experimental effects discussed previously [21] can also result in an incorrect measurement of He-

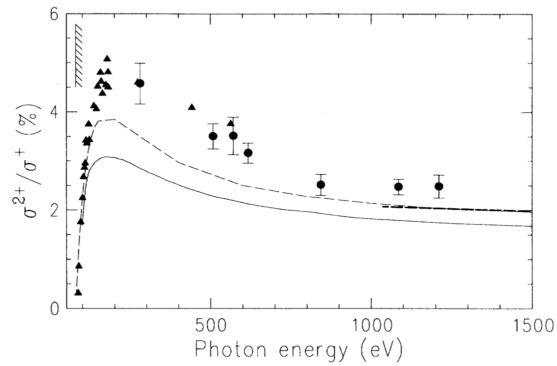

FIG. 2. Ratio of double-to-single ionization as a function of photon energy, comparing our data (full circles) and the data of Bartlett *et al.* (triangles) [17] with two theories by Hino, using MBPT and a correlated double continuum wave function (long and short dashed lines, respectively) [14], and by Ishihara, Hino, and McGuire (solid line) [10] (see text for details).

ion production. We tested for unequal detection efficiency by accelerating them to 4.05, 3.95, and 3.85 keV per charge, with no detectable change in the ratio. We also tested for target pressure dependence, and the results were consistent with pressure independence from 1.8×10^{-5} torr, to 7.4×10^{-6} torr, to 3.7×10^{-6} torr. Uncertainties in the data reflect both the statistical error in determining the areas of He^{+} and He^{2+} peaks as well as estimates of remaining stray light and higher-order light effects (the error bars shown are obtained by adding a 6% error of their values to each of the statistical errors).

Our present ratios of double-to-single ionization obtained at several photon energies are compared in Fig. 2 with the scaled data of Bartlett *et al.* [17]. As can be seen from Fig. 2 our data agree quite well with Bartlett *et al.*'s recent measurements.

We have compared our data with two calculations of Hino [14] and one of Ishihara, Hino, and McGuire [10] in Fig. 2. We then compare the data with an alternate reduced set of recent theories in Fig. 3. In Fig. 2 the solid

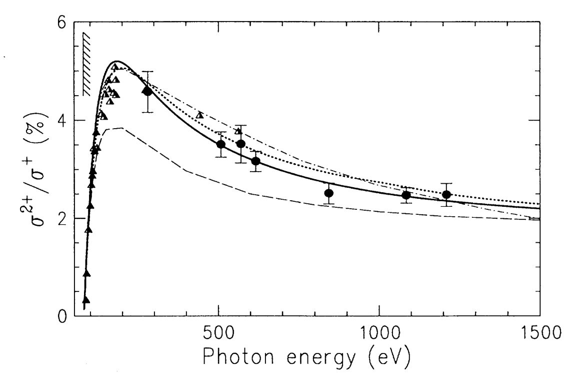

FIG. 3. Ratio of double-to-single ionization as a function of photon energy comparing the present data (full circles) and data of Bartlett *et al.* (triangles) [17] with theories by Pan and Kelly (solid line and short dashed line) [15], Hino (long dashed line) [14], and Samson, Bartlett, and He (chain-dashed line) [23] (see text for details).

curve is Ishihara, Hino, and McGuire's calculation [10] in the lowest-order many-body perturbation theory, using the length form of the electric dipole operator. In their terminology, ground-state correlation (GSC) and final-state correlation (FCS), which consists of shakeoff (SO) and TS1 (the inelastic scattering of the photoelectron on its way out of the interaction region with the remaining electron), were considered. This calculation [10], which models well the asymptotic behavior, and with which they find good agreement with the experimental high-energy-limit value of 1.5(2)% [7], lies lower than the data in the intermediate-energy range. The long dashed curve is a more recent calculation of Hino [14], this time using the acceleration form of the dipole operator, but otherwise it is similar to the previous calculation [10]. Despite the substantial change in the predicted ratios, the new MBPT calculation lies lower than our data up to about 800 eV with, however, better agreement above that energy. Hino [14] and Hino *et al.* [22] verified the gauge dependence of MPBT shown recently by Dalgarno and Sadeghpour [11] and concluded that, in the acceleration form, TS1 and GSC interfere constructively, while in the length form they interfere destructively. This could explain why the solid curve in their previous length form calculation shown in Fig. 2 is much lower than the data. According to Hino *et al.* [22], unlike in the asymptotic limit, TS1 is the dominant effect below 1 keV even in the acceleration gauge, which would imply that in our energy range of interest, in their terminology, both ground-state as well as final-state correlations (TS1 and SO) are important effects. Hino [14] obtained the short dashed curve using the acceleration form with an accurate ground-state wave function and a correlated double continuum wave function for the final state. This calculation models the high-energy limit quite well. Unfortunately it does not extend below 1000 eV and therefore cannot be compared with all our data points.

Figure 3 compares our data with the recent MBPT calculation of Pan and Kelly [15] in the length (short dashed line) and velocity (solid line) forms, along with the MBPT calculations of Hino [14] (long dashed line) and of a semiempirical calculation of Samson, Bartlett, and He [23] (chain-dashed line). Our data show excellent agreement with both the length and the velocity forms of the calculation of Pan and Kelly, and especially, with the velocity form. Pan has recently completed the calculation started by Pan and Kelly extending the threshold energy calculation of Carter and Kelly [1] up to 14 keV [15]. Throughout their calculation they included both ground-state and final-state correlations. But more importantly, as in the previous calculations [1], they show that while lowest-order results show reasonable agreement with experiment, they found that to achieve better agreement, certain higher-order correlation effects are significant. The agreement between the length and the velocity forms is very good over this energy range, apart from having the velocity form slightly dominate at the region near maximum. They find that separate total FSC and GSC diagrams, while individually large and of nearly the same magnitude, are of opposite sign and therefore interfere strongly, with FSC contribution being the larger of the two. They find, as in their previous work, that the length curve appears to be more sensitive to the higher-order corrections.

If indeed the main difference between Hino's MBPT calculation [14] and Pan and Kelly's MBPT calculation [15] is the inclusion of higher-order effects (both in GSC and FSC) and the use of a different basis set (important since the choice of a pertinent basis set enables the implicit inclusion of higher-order effects), we are led to conclude that, at intermediate energies (below 1500 eV), unlike the high-energy case, these higher-order effects are very important for a good description of the present data. We also note that the acceleration form, which is less sensitive to higher-order effects, as used by Hino [14], is not sufficient to describe the present data well.

Samson, Bartlett, and He [23] used a semiempirical model which conjectured that there should be a proportionality between producing a doubly charged ion by photon impact on a neutral atom and electron impact on a singly charged ion. Electron-impact data of Peart, Walton, and Dolder [24] were used to obtain their curve. The chain curve shows their calculation, which is in very good agreement with their data [17]. Compared to our data we also see good agreement, although it lacks the curvature of the MBPT calculation of Pan and Kelly. Overall good agreement is obtained in the present energy region, but their model appears to break down above 1300 eV, since their curve decreases rapidly compared with the measured ratios at high energies [21,7]. This difference may indicate the greater importance of shake-off effects at high photon energies compared to scattering effects.

In summary, we have shown by comparing our measurements with the most recent calculations of Hino and Hino *et al.* [14,22] and of Pan and Kelly [15] that at intermediate energies (below 1500 eV) both ground-state and final-state correlations appear to be important, and that inclusion of higher-order effects as well as a judicious choice of the basis set used seems to be essential to producing the excellent agreement observed.

Nora Berrah and Scott B. Whitfield are indebted to the Alexander von Humboldt Foundation for support. F. Heiser is indebted to the Deutscher Akademischer Austauschdienst for support. We would like to thank John Greene from Argonne National Laboratory for his expert help with the filters. We are grateful to C. Pan and K. Hino for supplying their calculations prior to publication. This work was supported in part by the U.S. Department of Energy, Office of Basic Energy Science, Division of Chemical Science under Contract No. DE-FG02-92ER14299, by BMFT, by WMU, by NIST, and by NSF.

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