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Inner-shell photoionization in weak and strong radiation fields

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Abstract

The X-ray beams presently produced at synchrotron-radiation facilities interact weakly with matter, and the observation of double photoionization is due to electron–electron interactions. The intensities of future X-ray free-electron lasers are expected to produce double photoionization by absorption of two photons. The example of double K-shell photoionization of neon is discussed in the one- and two-photon cases. We also describe an experiment in which X rays photoionize the K shell of krypton in the presence of a strong AC field imposed by an optical laser.

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Keywords: X-ray; Photoionization; Synchrotron-radiation; Free-electron laser

1. Introduction

The intense, tunable X-ray beams at synchrotron-radiation facilities enable detailed studies of X-ray/atom interactions by measurements that are high resolution, angle resolved, or detect emitted electrons, X rays, and ions in coincidence. These measurements are sensitive to the accuracy of theoretical treatments of higher-order effects such as electron–electron and nondipole interactions. Our group has conducted several experiments on X-ray/atom interactions involving X-ray scattering, multiple ionization, vacancy-decay processes, and photoelectron angular distributions. Overviews of this work are given in Southworth et al. (2000); Dunford et al. (2003). While the high flux and brightness of third-generation synchrotron light sources have enabled experiments with unprecedented detail, the peak power and coherence properties of existing X-ray sources do not approach those of optical lasers. Single atoms interact with single X-rays, and the interactions can be described perturbatively, i.e., in the weak-field regime. In contrast, ultrafast optical lasers produce strong AC fields that interact with matter nonlinearly, so coherent multiphoton processes dominate (DiMauro and Agostini, 1995). However, accelerator-based, X-ray free-electron lasers (XFELs) are presently being designed and constructed to produce coherent, ultrafast (~ 100 fs), high peak-power ($\sim 10^{16}$ W/cm^2) X-ray pulses (Materlik and Tschentscher, 2001; Arthur et al., 2002). The X-ray pulses from XFELs are expected to approach the onset of strong-field interactions with atoms, e.g. by multiphoton ionization (Kornberg et al., 2002).

Understanding the physics of X-ray processes in the weak-field regime is the starting point for treating strong-field phenomena and designing initial experiments for XFELs. Here, we discuss double K-shell photoionization of the neon atom in both the weak- and strong-field regimes. The weak-field experiment has been performed at Argonne's Advanced Photon Source (APS) (Southworth et al., 2003), while the strong-field experiment is planned for Stanford's Linac Coherent Light Source (LCLS) (Freeman et al., 2000). Single and double K-shell excitation/ionization cross sections of neon by two photons have also been recently calculated (Novikov and Hopersky, 2000, 2002). We also describe

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the development of an experiment at the APS designed to study X-ray photoionization of the krypton K shell in the presence of a strong AC field imposed by an ultrafast optical laser. This experiment is expected to demonstrate strong-field phenomena involving inner-shell electrons that are relevant to future experiments using XFELs.

2. Double K-shell photoionization of neon

Since single photons interact with single electrons in the weak-field regime, the excitation or ionization of two or more electrons is attributed to electron–electron interactions (Starace, 1982; Amusia, 1990). Double photoionization of helium has been extensively studied to understand electron correlation and develop theoretical models in the simplest case of a two-electron system (McGuire et al., 1995; Sadeghpour, 1996). Three first-order interactions have been identified. First, the relative positions of the two 1s electrons in the ground state are correlated radially and angularly (‘‘ground-state correlation’’) (Schmidt, 1997). Second, when one of the electrons is photoejected, the other is no longer screened from the nuclear charge; this sudden change in the effective potential can result in the second electron also being ejected (‘‘shake off’’) (Åberg, 1970). Third, the photoexcited electron can collide with the second electron and both are ejected (‘‘two-step-one’’ or ‘‘knockout’’) (Hino et al., 1993). Intuitively, it can be seen that shake off and knockout are interdependent and vary with photon energy. In rigorous theoretical treatments such as the many-body perturbation theory (Hino et al., 1993), the three interactions are treated as amplitudes that are added coherently and are not considered as independent mechanisms. However, it has recently been argued that ground-state correlation plus shakeoff can essentially be separated from knockout (Schneider and Rost, 2003).

The same electron–electron interactions are expected to govern double K-shell photoionization in higher-Z atoms. K-shell ionization energies are large compared with those of outer-shell electrons, and, to a first approximation, the K electrons can be treated as non-interacting with outer electrons. The production of double-K vacancies can be observed by recording either KK-KLL hypersatellites in Auger-electron spectra or KK-KL hypersatellites in X-ray fluorescence spectra. Hypersatellites are shifted up in energy from the diagram lines associated with single K vacancies (Chen, 1991). X-ray fluorescence methods are favored for higher-Z atoms, because their fluorescence yields are large. We used X-ray/X-ray coincidence spectroscopy to isolate hypersatellites from diagram lines following double-K photoionization of molybdenum (Kanter et al., 1999) and silver (Kanter et al., 2004). Auger-electron spectroscopy was used to study double-K photoionization of the lower-Z atom neon (Southworth et al., 2003), and we briefly describe that work here.

The single-K ionization energy of neon is 870.21 eV (Schmidt, 1997) and the double-K ionization energy is 1863 eV (Pelicon et al., 2000). A gas jet of neon was photoionized by a 5000-eV X-ray beam at the APS, and the Auger-electron spectrum was recorded with a cylindrical-mirror analyzer. The spectrum plotted in Fig. 1 is dominated by the K-$L_{2,3}L_{2,3}$ ^1S and ^1D diagram lines near 805 eV, but the KK-$KL_{2,3}L_{2,3}$ ^2D hypersatellite line at 870.5 eV is also observed on an expanded scale. The position and width of this peak agree well with multiconfiguration Dirac–Fock calculations (Chen, 1991). The structure between 830–860 eV is complicated by contributions from several multivacancy states, apparently including L-shell ionization in addition to double K-shell ionization. The intensity ratio of the KK-$KL_{2,3}L_{2,3}$ ^2D hypersatellite to the K-$L_{2,3}L_{2,3}$ ^1D diagram line was combined with experimental and theoretical determinations of branching ratios from single- and double-K vacancies to determine the ratio of double-to-single K-shell photoionization cross sections to be 0.32(4)%. This ratio is ≈4 times larger than the ratio calculated for He-like neon in the high-energy limit (Forrey et al., 1995; Krivec et al., 2001). The knockout interaction vanishes at high excess energies, and double ionization results from ground-state correlation and shake off (Sadeghpour, 1996; Schneider and Rost, 2003). Therefore, our measurement indicates that a strong knockout interaction is involved at 5000 eV.

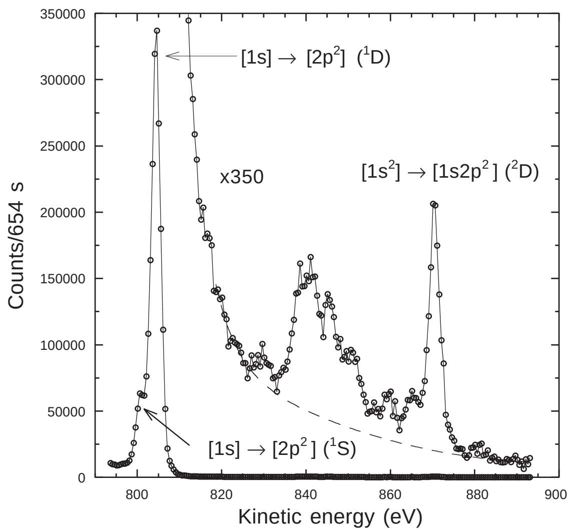

Fig. 1. Auger electron spectrum of Ne excited by absorption of 5000-eV X rays. The K-$L_{2,3}L_{2,3}$ ^1S and ^1D diagram lines and the KK-$KL_{2,3}L_{2,3}$ ^2D hypersatellite line are indicated. The structure between 830 and 860 eV is discussed in the text. The dashed curve beneath the peaks in the 820 and 890 eV region is the estimated background and tail of the diagram peaks.

Double-K photoionization with a single photon is a sensitive probe of electron correlation, but the effect is relatively weak. Even for He, the maximum of the double-to-single photoionization ratio is 3.76% (Samson et al., 1998). In comparison, it is interesting to estimate the probability of producing double-K photoionization in Ne by two-photon absorption at the LCLS as outlined by Freeman et al. (2000). In the energy range ~850–1000 eV, the unfocused LCLS beam (10 μm × 30 μm) will produce 2×10^{13} photons/pulse in a 0.42% bandwidth and 233 fs (FWHM) pulse width. This gives an instantaneous intensity of 2.9×10^{31} photons/s cm^2 that is many orders of magnitude larger than the instantaneous intensity $\approx 1.6 \times 10^{20}$ photons/s cm^2 we estimate for an APS undulator beam at the same bandwidth. While two-photon absorption is negligible at the APS, it becomes significant at the LCLS even for inner-shell photoionization for which vacancies decay on the femtosecond time scale. The probability for absorbing a second X ray within the Auger-decay lifetime of the first K vacancy can be estimated. Using the K-shell photoionization cross section of Ne near threshold (3×10^{-19} cm^2) and the K-vacancy lifetime (2.4 fs), the probability of double-K photoionization by two-photon absorption is 2.5% in the unfocused LCLS beam. If the LCLS beam can be focused as expected to 100 nm × 100 nm, the intensity increases to 4.3×10^{35} photons/s cm^2, and the calculated probability of double-K photoionization by two photons is 100%. Multiphoton processes in the X-ray regime are thus expected to become significant or dominant in LCLS experiments. This consideration clearly has implications for experiments on molecules and materials at the LCLS, because the weak-field models of X rays interacting with matter become inadequate. Understanding the physics of atoms in strong X-ray fields will be a necessary foundation for designing and understanding experiments on more complex samples. It should also be noted that two-photon excitation/ionization cross-sections of one or both K-shell electrons of Ne have been calculated by Novikov and Hopersky (2000, 2002) in anticipation of LCLS experiments. These calculations predict strong features in the two-photon cross sections due to two-electron excitations of K and L electrons into discrete and continuum states and consequent relaxation processes.

3. X-ray photoionization in a strong optical laser field

Pump-probe experiments using X rays and optical lasers are being performed at synchrotron-radiation facilities (Reis et al., 2001; Chen et al., 2001) and are expected to be widely used at XFELs to study atomic motion in molecules and materials on the sub-picosecond time scale. With the advent of laser-based sub-femtosecond pump–probe techniques, time-resolved studies of atomic inner-shell electronic motion has recently been demonstrated (Drescher et al., 2002). It is important to understand how the physics of X-ray processes are affected by the presence of intense optical laser radiation. A commonly used technique in synchrotron-radiation experiments is to tune the X-ray energy through an inner-shell absorption edge, so that electrons are excited from deeply bound states to Rydberg states and continuum (free electron) states. If the X-ray absorption process occurs in the presence of an intense laser field, the excited electrons "wiggle" due to stimulated scattering of laser photons (Freeman and Bucksbaum, 1991). The classical wiggle motion of a free electron in a laser field produces a time-averaged kinetic energy ("ponderomotive energy") $U_{\mathrm{p}} = e^2E^2/4m\omega^2$, where e and m are the electron charge and mass, and E is the peak electric-field strength produced by the laser of frequency ω. For an 800 nm laser at an intensity of 10^{14} W/cm^2, the ponderomotive energy is ≈ 6 eV. Since the wiggle energies of deeply bound states are small compared with those of free electrons, the presence of the laser field effectively increases the ionization energy by the free-electron wiggle energy U_{p}. That is, the absorbed X ray must provide the field-free ionization energy plus the ponderomotive energy in order to eject a photoelectron into the surrounding laser field. This effect has been observed at low energy using two lasers to study the photodetachment threshold of Cl^- (Davidson et al., 1993). The ponderomotive shift has not been studied at inner-shell thresholds.

We are developing an experiment to measure the shift of the krypton K-edge (nominal 14 327 eV ionization energy) due to the ponderomotive potential imposed by an 800 nm laser at intensities up to 10^{14} W/cm^2. We make use of the ultrafast laser/X-ray facility developed by the University of Michigan at the APS (Reis et al., 2001; Adams et al., 2002). A diagram of the experimental setup is shown in Fig. 2. To achieve the necessary laser intensity and to match the spatial overlap between the laser and X-ray beams, both beams must be focused to spot sizes of ≈ 10 μm^2. It is critical to develop diagnostic tools with which to measure the focal spot sizes and positions of the beams and to overlap them spatially and temporally. Our first experiments have

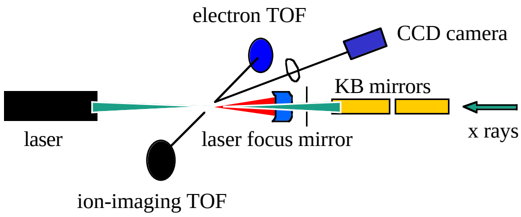

Fig. 2. Diagram of instrumentation for studying X-ray photoionization in the presence of an intense laser field. See text for description.

been devoted to developing the necessary instrumentation and methods. The X-ray beam was focused with Kirkpatrick–Baez (KB) mirrors that are capable of focusing APS beams to the required size with sufficient intensity for spectroscopic experiments (Eng et al., 1998). The focused X rays passed through a beryllium window into the vacuum chamber and through a hole in the spherical mirror used to focus the laser beam. A BGO crystal was inserted in the focal plane to produce fluorescence from the X-ray and laser spots that could be viewed with an optical CCD camera. This system allowed the two focal spots to be positioned within 100 μm in the plane perpendicular to the beam directions. By scanning the edge of the BGO crystal with submicron steps, the position of the focused X-ray beam could be accurately located, and an upper limit on its size was determined to be $\approx 8\ \mu m \times 12\ \mu m$ at a flux of 3×10^{11} X rays/s. The X-ray flux can be increased by coating the KB mirrors with platinum and using them at higher incident angles or by using longer mirrors. Locating the focused laser spot by scanning the BGO crystal did not work reproducibly, perhaps due to radiation damage. In future experiments, metal cross hairs will be used to estimate spot sizes and overlap the two beams. Laser photons scattered from the cross hairs will be detected by the CCD camera, and the X-ray beam will be detected by photoemission from the cross hairs or by shadowing the beam from a downstream ion chamber.

The focused laser and X-ray beams passed through an effusive krypton gas jet between the push- and pull-grids of an ion time-of-flight (TOF) spectrometer. We also attempted to use an electron TOF spectrometer, but the collection solid angle was too small to provide a useful signal from the spatially and temporally overlapped beams. With 4π collection, the ion TOF provides adequate count rate. The ion detector includes a position-sensitive anode to image the ion source region. Since the intensity of the laser beam decreases upstream and downstream of the focus, the ion position will provide information on the intensity dependence of the ion yields. In a separate measurement without X rays, the laser intensity can be determined from ion charge-state ratios (Maeda et al., 2000). To make use of the ion TOF method and to overlap the laser and X-ray pulses in time, the APS storage ring was operated in a special mode with a single bunch of stored electrons isolated before and after by 1.59 μs gaps from 56 closely spaced bunches. These gaps allowed the ion TOF spectrum from the single bunch to be isolated from ions produced by the multi-bunches. The single bunch contains $\approx 5\%$ of the total X-ray flux and produced an ion count rate of $10\ s^{-1}$ at a chamber pressure of 4×10^{-7} Torr. The count rate can be increased by operating at higher pressure. However, while the singlet X-ray pulses are produced at a rate of 272 kHz, the laser operates at 1 kHz, so only a small fraction of the X-ray pulses are overlapped by laser pulses. The laser oscillator is synchronized to the radio-frequency clock of the storage ring, and a fast photodiode is used to detect the laser and X-ray pulses and establish temporal overlap. The ions are collected in event mode and tagged with the presence or not of a laser pulse. In this way, ion TOF spectra with "laser on" can be compared with "laser off." The temporal width of the laser pulse was ≈ 25 ps compared with ≈ 87 ps for the X-ray pulses. In future experiments, it is desirable to reduce the X-ray pulse width or stretch the laser pulse to fully overlap them in time.

In Fig. 3 is plotted an ion TOF spectrum produced by photoionization of krypton by the X-ray singlet at 14 453 eV. The charge states are well resolved, and the substructure on the peaks is due to the natural isotopic abundances of krypton. The wide range in charge states results from alternative vacancy-cascade pathways involving the emission of X-ray fluorescence and Auger electrons (Kochur et al., 1995). The charge-state distribution is expected to vary as the absorbed X-ray energy is scanned across threshold, due to the interplay of bound- and continuum-state excitations and post-collision interactions (Armen et al., 1996). For example, the charge-state yields can be modified by the "recapture" of threshold photoelectrons into Rydberg states. In separate experiments at the APS, we measured Kr ion charge-state yields in coincidence with X-ray fluorescence as the incident X-ray energy was scanned across the K-shell threshold (Armen et al., 2003, 2004). We observed resonant enhancements or depletions of the charge-state yields near threshold that were modeled by accounting for both the threshold photoexcitation and the cascade behavior of the spectator electrons. It will be

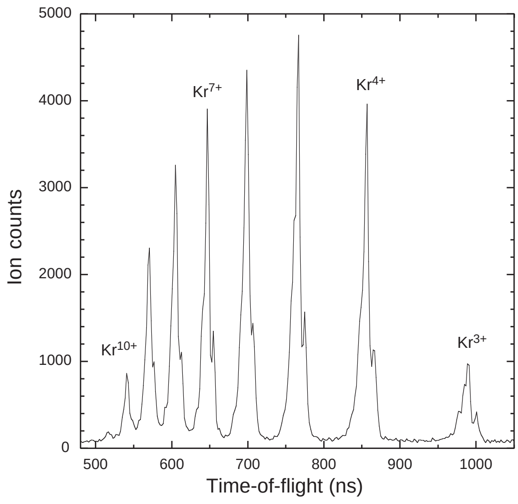

Fig. 3. Ion time-of-flight spectrum of krypton produced by photoionization at 14 453 eV.

interesting to study how inner-shell photoexcitation and decay dynamics are affected by the presence of a strong laser field. We expect that the ion charge-state yields will be modified due to effects on the photoelectron, Auger-electron, and Rydberg states of the ponderomotive potential and free–free or bound–free transitions in the laser field.

4. Summary

The intense, tunable X-ray beams produced at present synchrotron-radiation facilities enable detailed studies of X-ray/atom interactions in the weak-field regime. As an example, electron–electron interactions were investigated in the double K-shell photoionization of neon. X-ray free-electron lasers are being developed that will produce orders-of-magnitude higher peak intensities and introduce strong-field effects such as two-photon, two-electron inner-shell photoionization. By combining a synchrotron X-ray beam with an ultrafast optical laser, inner-shell photoionization in a strong-field environment is beginning to be studied.

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