

## Angular Correlation between Photoelectrons and Auger Electrons from *K*-Shell Ionization of Neon

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We have used cold target recoil ion momentum spectroscopy to study the continuum correlation between the photoelectron of core-photoionized neon and the subsequent Auger electron. We observe a strong angular correlation between the two electrons. Classical trajectory Monte Carlo calculations agree quite well with the photoelectron energy distribution that is shifted due to the potential change associated with Auger decay. However, a striking discrepancy results in the distribution of the relative angle between Auger and photoelectron. The classical model predicts a shift in photoelectron flux away from the Auger emission direction, and the data strikingly reveal that the flux is lost rather than diverted, indicating that the two-step interpretation of photoionization followed by Auger emission is insufficient to fully describe the core-photoionization process.

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For a multielectron atom such as neon, core-level photoionization followed promptly by Auger decay is an example of the correlated three-body problem. In particular, just above the  $1s^2$  core-photoionization threshold, two interesting phenomena can occur: (1) the three-body post-collision interaction (PCI) between the photoelectron, residual ion, and subsequent Auger electron [1]; and (2) the possible recapture of the photoelectron [2,3]. Until now, these processes have been well described within the sudden approximation, where the loss in energy (in atomic units) of the photoelectron is simply the abrupt change in potential energy that occurs when the core-ionized  $\text{Ne}^{*+}$  ion decays to a  $\text{Ne}^{2+}$  ion via emission of a very fast Auger electron. Within that framework, the energy loss of the photoelectron is given by  $1/r$  (atomic units are used throughout), where  $r$  is the distance traveled from the ion before Auger decay occurs. If this energy loss is less than the original continuum energy, then the electron simply remains in the continuum with reduced kinetic energy (process 1). In this case, all three bodies can exchange momentum and energy. If, however, the energy loss is greater than the photoelectron's initial energy, the electron can be recaptured into a Rydberg state orbiting the  $\text{Ne}^{2+}$  core, which can in turn decay with the reemission of an electron with a discrete energy.

PCI effects in photoionization have been studied extensively, both theoretically and experimentally, by numerous groups [4–9] (and references therein). In the specific case of neon, the work by Ueda and collaborators [6–8] demonstrated that the escape probability and energy shifts in the electron spectra are fully consistent with the two-step,

sudden-approximation model. More recently, Penent *et al.* [9] used a magnetic bottle time-of-flight technique developed by Eland *et al.* [10] to measure the energies of the two emitted electrons in coincidence, showing the correlation between the two electrons in energy space. The angular correlation, however, between the Auger and photoelectron has only been studied theoretically for the case of equal or nearly equal continuum energies [11,12].

An experimental work on the double photoionization of xenon by Scherer [13] and collaborators demonstrated clearly how PCI plays a role in the interaction of the two continuum (photo and Auger) electrons. Their results were consistent with quantum calculations, which included PCI. However, in that experiment, the electrons had similar energies (17 and 30 eV) and so should be expected to interact strongly in a way that requires quantum mechanical description. In light of this of pioneering work, one fundamental question is yet to be explored: What happens when an energetic Auger electron ( $\sim 800$  eV) and low-energy photoelectron ( $\sim 1$  eV) are emitted from the atom in the *same* direction?

In this Letter, we present unexpected results for the case where both photo and Auger electron are emitted in the same direction. We have simultaneously measured both the photoelectron (directly) and the Auger electron (indirectly) following core photoionization of neon at 1.36 eV above threshold. At this energy, the majority (75%) of the photoelectrons escape, and the remainder are recaptured. By measuring both electrons and the residual neon ion in coincidence with  $4\pi$  solid angle collection efficiency, we have measured the final momentum of one electron in a

coordinate system defined by the direction of emission of the other.

Existing quantum theories of PCI have been derived when all of the particles are weakly interacting or when one of the pairs is strongly interacting (for example, see Refs. [14]). In our experiment, two pairs strongly interact: the Auger electron with the photoelectron and the photoelectron with the  $\text{Ne}^{2+}$  ion. Thus, none of the previous calculations of PCI are a valid starting point for the interpretation of our results [15]. Therefore, we compare our results to a Classical Trajectory Monte Carlo (CTMC) calculation that includes the continuum interaction between the electrons and the interaction of both escaping electron with the residual ion on equal footing, including PCI effects.

We performed our measurements at beam line 11.0.2 at the Advanced Light Source at Lawrence Berkeley National Laboratory using COLd Target Recoil Ion Momentum Spectroscopy (COLTRIMS) [16–18]. A beam of linearly polarized photons 1.36 eV above the neon  $K$  edge (871.6 eV) was crossed with a supersonically cooled jet of neon that was precooled to a temperature of 100 K. Low-energy electrons and ions were extracted from the overlapping volume of approximately  $0.1 \times 0.1 \times 1 \text{ mm}^3$  by electric and magnetic fields to position sensitive micro-channel plate detectors with delay-line anodes [19]. By measuring their positions of impact on the detectors and their times of flight, the momenta of the recoiling neon ion and the photoelectron were determined for each ionization event. Fields were chosen so that electrons up to 2.5 eV and all ions were collected with  $4\pi$  solid angle. The precooled of the neon target in combination with the cooling due to expansion from the jet nozzle and skimming of the atomic beam combined with 3-dimensional electrostatic focusing in the ion arm of the spectrometer allow for a ion momentum resolution (FWHM) of at least 0.3 a.u. in each direction. The resolution of the low-energy electron measurement is 0.02 a.u., which corresponds to 170 meV at the photoelectron energy. The electron spectrometer was calibrated using the sharp autoionization lines arising from the recapture process. The photon energy is then calibrated by measuring the photoelectron energy for the case where the excited ion decays via photon emission and no PCI takes place.

We observed four final charge states of neon:  $\text{Ne}^+$  (1.93%)  $\text{Ne}^{2+}$  (92.1%)  $\text{Ne}^{3+}$  (5.71%) and  $\text{Ne}^{4+}$  (0.28%), which is consistent with a previous measurement by Kanngießner *et al.* [20]. The radiative decay of the core vacancy leads to a singly charged ion, whereas the much more likely Auger decay primarily leads to the doubly charged ion. The remaining charge states are only accessible via a double-Auger process. In the case of Auger decay, the  $\text{Ne}^{2+}$  momentum is due to the small kick (0.3 a.u.) it receives from the photoelectron and the big kick it receives from the Auger electron (7.7 a.u.).

The full vector momentum of the Auger electron was determined by momentum conservation with the photoelectron and residual  $\text{Ne}^{2+}$  ion. The measured Auger energies and branching ratios were consistent with that of Kadar and collaborators [21] where the 804.5 eV  $^1D$  state has the largest branching ratio of 0.61. The Auger energy resolution was determined to be 30 eV. This large value relative to traditional spectroscopic measurements is due to the indirect momentum measurement via the recoiling  $\text{Ne}^{2+}$  ion. The momentum resolution of the Auger electron measurement is determined primarily by the resolution of the ion measurement and leads to an Auger angular resolution of  $2.2^\circ$ .

We have used a CTMC calculation to model the interaction between the photoelectron and Auger electron. Because the de Broglie wavelengths differ by a factor of 23, their interaction in the continuum should be well approximated classically. Each event in the calculation consisted of the emission from the origin of a 1.36 eV electron followed by the emission of an 800 eV Auger electron in a random direction after a random time delay with an exponential distribution determined by the Auger lifetime. For the calculations, we assume no correlation between the original outgoing directions of the two electrons. Once both electrons are launched, we solve the classical time dependent equation of motion to determine the final momenta of the two electrons, where photoelectrons with negative final energies are considered recaptured. Thus, the full electron-electron interaction in the field of the ion is incorporated in this classical calculation. The three components of each electron's momentum were stored in list mode, which allowed for the same sorting analysis of the CTMC data as was used in the analysis of the experimental data. This allowed for a realistic modeling of the experimental resolution.

Figure 1 shows the energy of the low-energy electrons in coincidence with the singly or doubly charged neon ion. The  $\text{Ne}^+$  charge state is produced following core photoionization through a radiative decay of the  $\text{Ne}(1s)$  hole. Because the ion charge state does not change, the photoelectron energy is simply given by the photon energy minus the  $\text{Ne}(1s)$  binding energy. The solid curve is a Voigt fit at 1.36 eV using the 270 meV natural line width in addition to the energy resolution of both the beam line monochromator and photoelectron measurement, which is fit at 300 meV. The  $\text{Ne}^{2+}$  charge state is produced, following core ionization, by the Auger decay of the  $\text{Ne}(1s)$  hole. The distribution in this case (open squares in Fig. 1) is shifted to lower energies by the post-collision interaction. On top of the  $\text{Ne}^{2+}$  distribution, one can see peaks or shoulders (indicated with vertical lines) that correspond to the recapture or reemission channel that comes with  $\text{Ne}^{2+}$  as well. The solid curve is produced by the CTMC calculation and agrees well with experiment. The recapture or reemission process is not included in the present CTMC model.

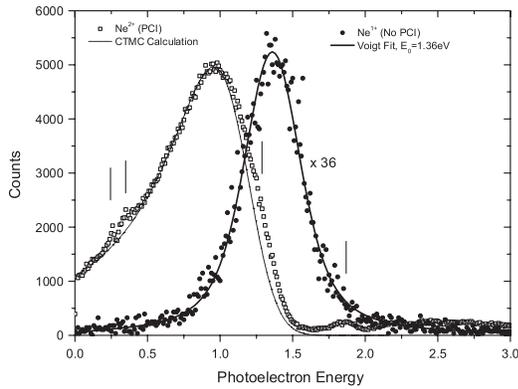


FIG. 1. Photoelectron energies. The closed circles (●) of the right hand peak are photoelectrons measured in coincidence with  $\text{Ne}^+$  ions and corresponding to direct photoionization with no PCI. The solid line is a Voigt fit. The open squares (□) of the left hand peak are photoelectrons measured in coincidence with  $\text{Ne}^{2+}$  ions and corresponding to photoionization followed by an Auger decay. The dashed line is the CTMC calculation described in the text. The short vertical markers indicate recapture-remission lines.

The momentum space images of the photoelectron in a frame defined with axes parallel and perpendicular to the momentum of the Auger electron are shown in Fig. 2. The upper panel corresponds to the experimental data, and the lower panel corresponds to the CTMC calculation. In both panels, the results are mirrored about the horizontal axis. Both distributions reveal a “C” shape that has an opening for angles near the direction of the Auger electron. In the experiment image, isotropic discrete rings can be seen that correspond to the recapture or reemission process for the autoionization of the  $\text{Ne}^+ 1s^2 2s^2 2p^4 np(^1D, ^1S)$  state (for a detailed discussion on the recapture or reemission process, see [6–8]). The general shape and size of the CTMC calculation and experimental result match quite well. However, there is a clear pileup of events in the CTMC case, where flux has been deflected by the outgoing Auger electron in the simulation. In the image from the experiment, however, the flux is missing at the opening of the “C.” It is this loss of electron flux that provides evidence for the incompleteness of a two-step model, where the total flux must be conserved.

This effect is more apparent in Fig. 3. Here, we show the data and CTMC calculation as a function of the cosine of the angle between the two electrons. The CTMC calculation is shown in three curves. The first is the model alone, and the other two correspond to including the  $1\times$  and  $2\times$  momentum resolution of the experiment in the initial conditions of the CTMC calculation. The calculated pileup of flux is not an effect of the classical treatment of the final state correlation. This redistribution of conserved flux is a consequence of the two-step assumption. The data show that the flux is not redistributed, but rather is suppressed. Because the wavelengths of the two electrons are so differ-

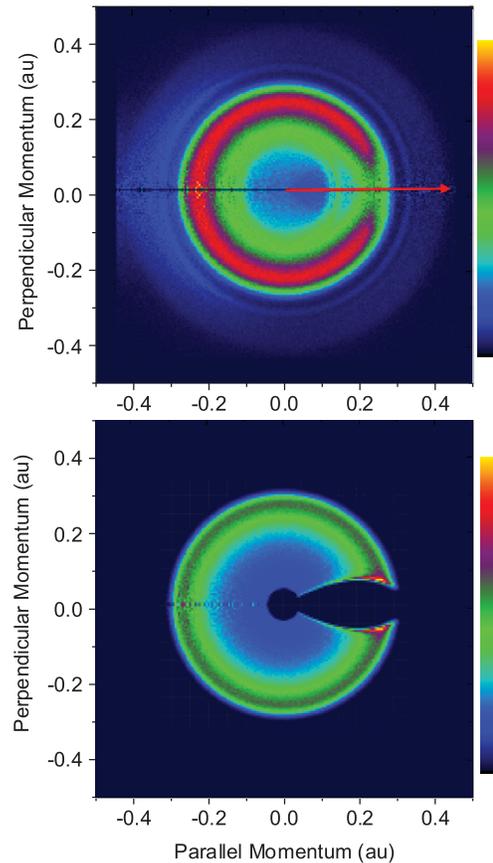


FIG. 2 (color). Momentum image of the photoelectron in a frame defined with axes parallel and perpendicular to the Auger emission direction (indicated by the red arrow). In both panels, the color scale is linear. The top panel shows the experimental result for low-energy electrons in coincidence with the Auger electron. The isotropic discrete rings correspond to the recapture or reemission process. The lower panel shows the CTMC result in the same format. The pileup along the edge of the opening of the “C” is not present in the experimental data.

ent, any calculation including only the continuum interaction will only change the angle of the outgoing flux and would be insufficient to explain the experimental results. We conclude from this that the initial state correlation must play a significant role in this process and that at this detailed level of investigation, the two-step model breaks down. Simply stated, for the case of coaxial emission, the initial emission of the photo and Auger electron pair is suppressed, and the total flux is slightly reduced.

In summary, we have used COLTRIMS and a CTMC model to explore the correlated final continuum state of a photoelectron and Auger electron pair. The ability to measure the momenta of the photoelectron and associated Auger electron into arbitrary directions has revealed an unexpected loss (as opposed to redistribution) of photoelectron flux for the subset of phase space where the two electrons are emitted in near parallel directions. This is a striking detail that is undetectable in noncoincident electron spectra.

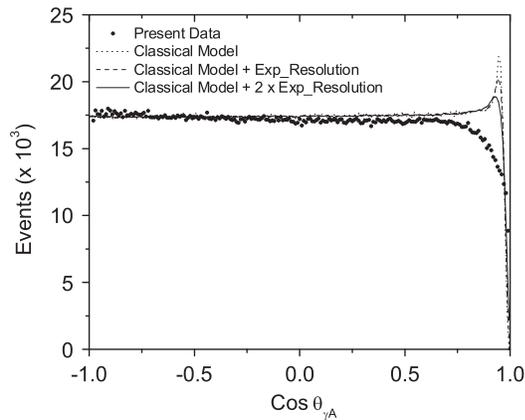


FIG. 3. Distribution in cosine of the angle between the Auger electron and photoelectron. Closed circles (●): experiment, lines: classical calculation with and without convolution with experimental resolution. The calculation clearly shows a redistribution of photoelectron flux, while it can be seen from the data the flux is actually lost.

Future measurements might include a system such as Argon to see what, if any, effect different initial angular momentum states  $\text{Ar}(2s, 2p)$  have on the final state correlation where both photo and Auger electrons emerge in the same direction. This would complement the work by Bolognesi and co-workers, where they measured the anisotropic angular distributions between the photo and Auger electrons for this system in rich detail for angles between the two electrons larger than  $45^\circ$  [22].

As far as we know, a complete quantum mechanical theoretical treatment of the system described here does not exist. However, we encourage the pursuit of detailed model calculations that include not just the continuum correlation of the photo and Auger electrons, but rather the full two-electron transition from initial to final state.

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