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Population of individual Ne $1s^{-1}np$ Rydberg states measured across the $1s$ ionization threshold and the evolution of shake-up into shake-down

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New high-resolution experimental results on Ne $1s$ near-threshold photoionization with subsequent decay into a $2p^4(^1D_2)n'p$ or $2p^4(^1D_2)\varepsilon' p$ state are presented. The population of Rydberg states up to $n'=12$ is determined as a function of excess energy. These data allow one to track in detail the transition from resonant Auger decay going along with shake-up of the spectator electron to shake-down and photoionization with recapture of the photoelectron. Results are in good agreement with a time-dependent quantum mechanical theory of Auger decay in the presence of a slow photoelectron.

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Photoionization of an atomic system near an inner shell threshold and the following relaxation is a very complex phenomenon which substantially involves many-electron correlations. It is especially interesting for light or medium-mass atoms where the dominant relaxation process is Auger electron emission. In this case the relaxation of the inner shell vacancy involves an interaction of at least three charged particles in the final state: photoelectron, Auger electron, and doubly charged ion—a Coulomb three-body problem with various interacting open channels that gets even more complicated by the multielectron structure of the ion. In some energy regions, simplified pictures (mechanisms) of the process are successfully used [1]. However, most interesting is the transition regime at the ionization threshold, where different mechanisms merge.

When the photon energy is slightly below threshold, core-excited resonances are formed in which a core electron is transferred to an unoccupied Rydberg orbit. These resonances usually decay by spectator resonant Auger electron emission [2]. As a rule the spectator electron is shaken-up to a higher orbital during the decay [3]. (If the excited electron is in the lowest possible state then spectator transitions are dominant.) The picture of a shake-modified resonant-Auger transition describes the experimental spectra of emitted electrons very well in this case (see [2], and references therein). When the photon energy is tuned just above threshold, normal Auger decay occurs, but since the energy of the photoelectron is small, strong post-collision interaction (PCI) effects distort both the Auger and photoelectron lines [4,5]. The energy of the excited (photo) electron is reduced and in some cases it can even be recaptured by the doubly charged ion so that only one electron (Auger) is emitted [6,7]. This shift of the photoelectron energy or recapture to a negative energy state may be seen as a shake-down due to PCI. The existing theories of PCI adequately describe the distortion of the photoelectron and Auger lines as well as the photoelectron recapture [8–11].

When the photon energy approaches threshold from either side, the two pictures blend. This happens in the so-called *transition regime* [1,3] when the excess energy E_{exc}, defined as the difference between the photon energy and the ionization potential ($E_{exc}=E_{ph}-IP$) is close to zero: $-\Gamma^{2/3}/2 \leqslant E_{exc} \leqslant 2\Gamma$ (Γ is the inner vacancy lifetime width in atomic units). In the transition regime shake-modified spectator emission with more shake-up than shake-down develops into PCI distorted emission with more shake-down than shake-up. Conceptually and theoretically the transition regime is well understood (see, for example, [1,10–13]). It is clear that all characteristics of the photoemission process evolve smoothly through the transition region without any drastic changes at the threshold [12,13]. On the other side, experimental studies of this regime are far from satisfactory. There were several investigations of the evolution from spectator to normal Auger lines across the threshold [14–16], including a recently published experiment by the authors [17]. However, the overall resolution of experiments was not good enough for a quantitative investigation of the transition from the shake-up to the shake-down regime. A detailed study of this transition will be reported here, which has become possible by virtue of further improvements in the data quality.

We present detailed high-resolution photoionization measurements in Ne which enabled us to follow the transition between the two regimes as the energy is tuned across the $1s$ ionization threshold. Results are obtained for Ne core level excitation or ionization followed by relaxation into the $2p^4(^1D_2)n'p,\varepsilon' p$ states, where $n'p$ designates a bound outer electron (resonant Auger decay or recapture) and $\varepsilon' p$ a free photoelectron. We have studied the radiationless decay spectra for these transitions by high-resolution electron spectroscopy in an excess energy interval of $[-0.6, 0.6]$ eV around the $1s$ ionization threshold of 870.17 eV [18]. Results for the

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same transition which cover a wider energy range, but in less detail, have been presented in [17].

The experimental setup has been described in our earlier publication [17]. Here, we only mention that the beamline BL27-SU at the Japanese synchrotron radiation facility SPring8 was used. This beamline is equipped with an endstation for gas phase electron spectroscopy that employs a large hemispherical analyzer (Scienta SES 2002). The polarization of the synchrotron radiation was linear and horizontal, and the analyzer axis was mounted horizontally in the plane perpendicular to the photon propagation axis. Electrons were thus probed at an angle of 0° with respect to the electric field vector of the synchrotron radiation. The photon energy resolution was 66 meV, better than in a previous series of measurements [17], and the total apparatus resolution amounted to 115 meV. The energy scale of the analyzer was corrected by placing the resonant Auger decay into the $2p^4(^1D_2)3p(^2P)$ final state, recorded on the maximum of the $1s \rightarrow 3p$ resonance, on the expected kinetic energy (final state energy 55.835 eV [19], photon energy 867.12(5) eV [20], see also [17,21]). Then, excess energies of the remaining spectra were derived from the measured kinetic energies of this line. The resulting scale error common to all excess energies can be estimated as 55 meV. No indications of fluctuations in the analyzer energy scale were observed within the limits set by the experimental resolution.

Experimental results are compared with two theoretical approaches. One is a numerical implementation of the semiclassical theory by Russek and Mehlhorn [22]. Basically, the Auger amplitude in this *ansatz* is written as a Fourier transform of the time-dependent decay probability, the time evolution of which is modified by a time and energy dependent phase factor, which takes the PCI into account. The energy exchange between the slow photoelectron and the fast Auger electron is $1/\rho$ with ρ the radius at which the Auger electron overtakes the photoelectron. Details of our implementation have been described in [17]. Although unforeseen in the original publication [22], we found our implementation of this approach usable in the extreme threshold region and even below threshold. The other approach is a fully quantum-mechanical theory based on a nonstationary description of photoionization accompanied by the Auger decay [23]. Its key point is the modeling of the slow electron with a time-dependent wave-packet, which evolves under the influence of the electromagnetic excitation pulse, the Hamiltonian in the intermediate state, and the (Auger) instability which projects it into the ionic final states. Physical observables are obtained by projecting the full final state wave-packet onto the time-independent eigenstates, the asymptotic population of which (recapture probability) we would like to obtain. Again we refer to our more extensive publications for details [17,23].

Experimental spectra are shown in Fig. 1. Contributions of resonant Auger decay or recapture, respectively, into final states up to $n'=10$ can be spectroscopically resolved. Final states with $n'=11$ and 12 can be discerned as shoulders on the high kinetic energy side of the normal Auger peak in most spectra where these states are populated. In order to compare the experimental spectra with our semiclassical calculations we employ a smooth continuation of the Auger part

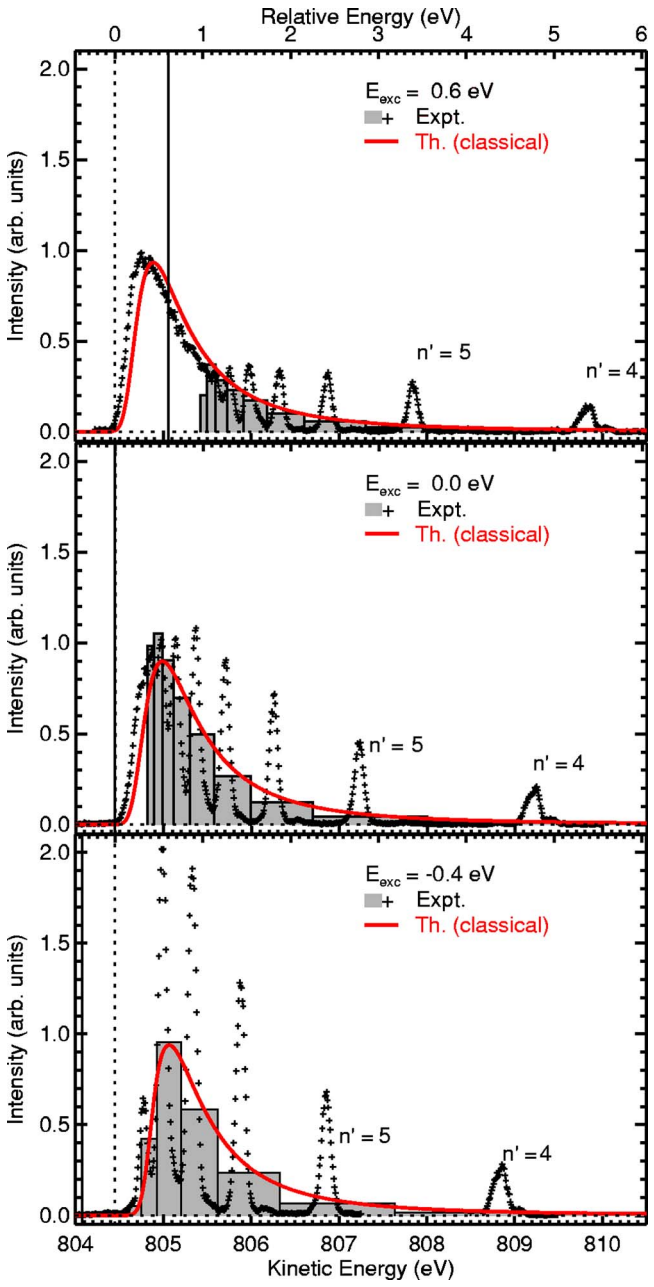

FIG. 1. (Color online) Electron spectra of Ne recorded with excitation energies near the Ne 1*s* threshold. The excess energy is indicated in the panels. Discrete lines due to the recapture of the photoelectron into a high Rydberg state dominate the spectrum even above the threshold. The gray shaded boxes are drawn as a histogram representation of the experimental spectrum, their areas are proportional to the experimental intensities of the $2p^4(^1D_2)n'p$ states with $n'=4-12$ (see text for details). The solid curve is derived from a semiclassical model of PCI-mediated Auger decay [17,22].

of the spectrum into the region below threshold. This problem has been discussed in the theory of atomic oscillator strength [24], and the gray boxes are drawn accordingly, taking into account the density of the final states. The area of the boxes represents the *experimental* intensity of the $n'=4-12$ states. For positive and negative excess energies, the gross behavior of the spectrum is in very good agreement

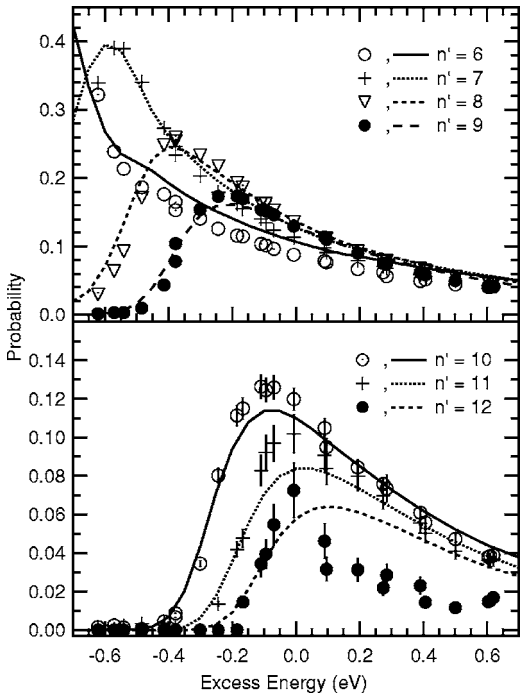

FIG. 2. Probability of populating $2p^4(^1D_2)n'p, n'=7-12$ states of Ne by resonant Auger decay, or—above threshold—photoionization followed by recapture of the photoelectron. Various symbols show the experimental probabilities for different n'. Experimental error bars in the upper panel are equal or smaller than the symbol size. The curves show the theoretical values derived from the time-dependent quantum-mechanical model [17,23].

with the semiclassically calculated spectra (solid curves). The quantum-mechanical calculations give very similar results (cf. Ref. [17]).

A more quantitative representation of our results is given in Fig. 2. Here the probability for the population of the experimentally resolvable final states is plotted. To arrive at this quantity, least-squares fit to all spectra were performed and the area of each resulting line was divided by the total area underneath the fit curve. The continuum part of the spectrum in the case of positive excess energy was modeled as well. Since the lifetime broadening of the Ne 1*s* vacancy state is 270 meV [20], the transition regime discussed above reaches from approximately −630 to 540 meV, and is completely covered by our measurements. The experimental probabilities are compared with the theoretical results (curves) calculated using the nonstationary quantum-mechanical approach [23]. For the lower values $n' \leqslant 9$ (upper panel) the agreement between theory and experiment is excellent. It is somewhat worse for larger $n'=11,12$ (see discussion below).

Certainly, the most intriguing finding is that the population of all n' final states exhibits a local maximum at an excess energy, which increases with n'. Qualitatively similar results may be deduced from the experimental data by Aksela *et al.* [15] for Kr(3*d*) and Xe(4*d*) excitations. Also Okada *et al.* [25] have observed such behavior for some Rydberg states across the L_3 threshold in Kr. As seen in Fig. 2, in the case of $n'=11$ and 12 the maximum is at the ionization threshold, that is these states are populated more

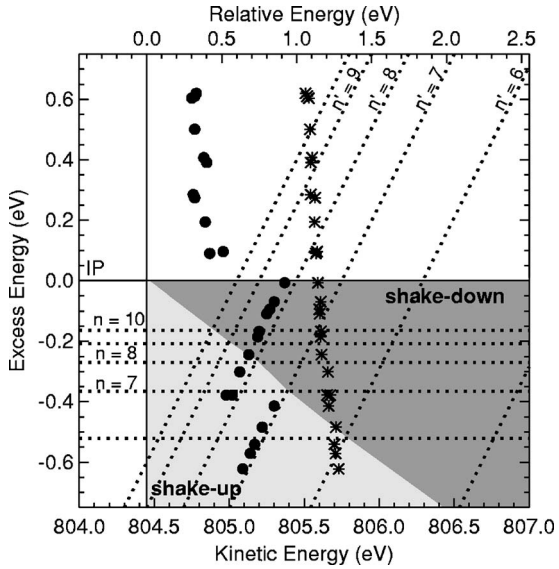

FIG. 3. Behavior of the center of gravity (asterisks) and the maximum of the spectrum (circles) as a function of photon energy. (See text for explanation.) Horizontal lines mark the excitation energy of some 1*snp* neutral excited states, and diagonal lines give kinetic energies of relaxation into some $2p^4(^1D_2)np'$ final states. IP designates the experimental Ne 1*s* ionization potential. Neutral excitation energies Ref. [26], final state energies Ref. [19].

effectively by shake-down or recapture than by shake-up at lower excitation energies. Therefore, for these final states the transition from shake-up to shake-down dominated behavior has already taken place. It is interesting to note that both the experimental data and theoretical results show that at every energy above the local maximum of the $n'=8$ state, this state has the maximum in population. The excess energy corresponding to the maximum of the $n'=8$ state is lower than the position of $1s^{-1}8p$ state in neutral Ne ($E_{exc}=-0.28$ eV). Therefore, for the $n=8$ excitation the most probable decay mode is a pure spectator transition in which the spectator electron does not change its principal quantum number. For higher resonances $n \geqslant 9$ the dominant transition is shake-down, for lower resonances $n \leqslant 7$ the shake-up dominates. This observation confirms the assumption made in our previous work [17], although as follows from our calculations, the exact position of the border between the shake-up and shake-down regions depends on the spectral width of the exciting radiation. We should note, however, that for increasing n' more and more final states overlap within their natural linewidths. This needs to be taken into account, and the behavior of the maximum of the spectrum should be considered, as we shall discuss below. The overlap of lines makes it difficult to single out the contribution of the $n'=12$ state in the experimental data, and might be responsible for the worse agreement to the theoretical data in this case.

The behavior of the maxima of the experimental spectra and the transition from shake-up to shake-down is depicted in Fig. 3. We see that the observed maximum of the spectrum at the lowest excess energies probed, which correspond to an excitation between $n=5$ and $n=6$ in the neutral atom, is at a final state with $n'=7$, that is, as expected, shake-up dominates. In the two-dimensional kinetic energy—excess energy

plane in Fig. 3, the half-plane pertaining to shake-up is shaded light gray. We see that the maxima of the spectra (shown by circles) reside in that half-plane until the excess energy corresponds to neutral excitations between $n=8$ and $n=9$, where the transition in the shake-down plane (dark-gray shading) occurs. Closer to threshold (but still at negative E_{exc}) the maxima remain close to $n'=8$. Above the $1s$ threshold, the maxima of the spectra are located in the continuous part of intensity (cf. Fig. 1). Note that the center of gravity of the spectra (asterisks in Fig. 3) smoothly evolves across the threshold in accordance with the above discussion, showing a slight increase of the Auger electron energy with decreasing photon energy due to PCI. (The center of gravity is calculated as $\Sigma_i a_i E_i / \Sigma_i a_i$, where E_i is the energy and a_i the intensity in data point i and the summation runs over the part of the spectrum pertaining to the final states discussed here.)

It is interesting to speculate how these findings would vary in other targets. The factors that enter into our classical model of near-threshold Auger emission are the excess energy E_{exc}, the lifetime Γ, and the radii of the orbitals emitting the photo- and the Auger electron [17,22]. These four quantities will determine the spectral behavior in the excess energy vs kinetic energy plane of Fig. 3. To determine the crossover from shake-up to shake-down, moreover it is necessary to know the difference in quantum defect from the neutral to the singly ionized state, which will determine the slope of the diagonal separating the two regimes from each other (border between the two gray shaded regions in the figure). Classically, it should thus be possible to transfer our findings to other systems with proper adjustments for the quantities mentioned above. Whether further changes in the emission behavior will be caused by the quantum nature of the system, or, e.g., by the pulse length, is a topic for future study [23].

In conclusion, from the high-resolution electron spectroscopy of the first relaxation step after Ne core level photoexcitation or ionization, we were able to observe for the first time precisely, where the transition from shake-up modified resonant Auger decay to PCI-mediated shake-down and/or recapture and normal Auger transitions occurs. We have found that the classical PCI theory is applicable even for energies below this limit, which suggests that already for sufficiently high Rydberg excitations a Coulomb interaction between the excited electron and the autoionization electron is mainly responsible for the spectral shape. We have tested a new time-dependent theory of PCI and found that it reproduces our results for the state resolved recapture probability excellently.

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