Observation of Angle Dependent Postcollision Interaction in the Electron Impact Ionization of Xe $4d_{5/2}$

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The angular dependence of postcollision-interaction effects has been studied in the electron impact ionization of the Xe $4d_{5/2}$ orbital at 1000 eV incident energy. The Xe $N_5O_{23}O_{23}$ ($^1S_0$) Auger electrons have been measured in coincidence with ejected electrons of 30 eV at two different relative angles, namely, 170° and 25°. At the smallest relative angle, the coincidence spectrum displays a shift towards lower Auger energies and a shape that is consistent with the predictions of angular dependent postcollision-interaction models.

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Since its discovery in 1923 [1] and its interpretation as a radiationless process a few years later [2], the Auger effect has attracted extensive theoretical [3] and experimental [4] efforts. This is due to the finding that Auger electron spectroscopy is a powerful tool for chemical analysis and for the investigation of the valence structure of molecular systems, solids, and surfaces.

In the common wisdom, the Auger effect is assumed to be a two-step process: the creation of the inner hole and its subsequent decay; the latter being completely decoupled from the former. This assumption holds if (i) the probability of direct excitation of the doubly charged final state is negligible, i.e., interference effects such as the ones often observed in autoionizing processes can be excluded, and (ii) the lifetime of the intermediate state is long enough to prevent any final state interactions between the charged particles in the continuum.

In recent years it has been shown within the framework of the Bethe-Born theory [5] that the two-step assumption is violated in any charged particle impact ionization experiment, provided that the kinematics is not restricted by coincidence techniques [6,7]. The process responsible for this violation is the interaction, named the postcollision interaction (PCI), of the Auger electron and the other free charged particles produced in the primary ionization event. Close to the ionization threshold, PCI results in considerable energy shifts of the Auger peaks and distortion of the line shapes [8]. However, in an experiment where only the Auger electron is detected, the other two charged particles in the continuum can share the excess energy continuously. Thus even at high incident energy there is always a nonvanishing probability that a low energy particle is present in the final state. Both noncoincidence experiments [6] and calculations [7] have shown that even in the limit of high impact energies the line shapes are asymmetric, shifted with respect to the theoretical Auger energy E_A^0, and their width may exceed the decay width Γ_0 by up to 10%. On the contrary, in the case of inner shell photoionziation it has been experimentally shown [9] and theoretically proved [10–12] that the two-step assumption is valid for sufficiently high photon energy.

Up to now most of the experimental work has been devoted to studying the dependence of PCI effects on the excess energy above the excitation threshold [8]. Less attention has been paid to the PCI angular dependence [13–16] predicted by the recent formulations of the different theories [11,12,17]. This is due mainly to the time consuming coincidence experiments needed to investigate such an effect. In particular, no definite experimental proof of the anisotropic effects has been given in the case of electron impact ionization.

In this Letter, we present the observation of the PCI angular effects in the electron impact ionization of the Xe $4d_{5/2}$ orbital;

$$e_0(E_0 = 1000 \text{ eV}) + \text{Xe} \rightarrow \text{Xe}^+(4d_{5/2}^{-1}) + e_1(E_1 = 902.5 \text{ eV}) + e_2(E_2 = 30.5 \text{ eV})$$
$$\downarrow$$
$$\text{Xe}^{2+} + e_A[N_5O_{23}O_{23}(^1S_0)], \quad (1)$$

where the ejected electron e_2 is detected in coincidence with the $N_5O_{23}O_{23}$ ($^1S_0$) Auger electrons. The complete understanding of process (1) would be achieved only by an experiment in which all three final electrons are detected in coincidence. The feasibility of triple coincidence experiments has recently been provided [18], but the energy resolution and long accumulation time of these experiments hamper their application to the present study.

Although the nonisotropic PCI theories [11,12,17] predict that the main effect arises because of the interaction between the slow ejected and Auger electrons, in the previous investigations [13–15] the fast scattered and Auger electrons have been measured in coincidence. The PCI effects have then been averaged over all the directions of the ejected electrons weighted according to their angular distribution derived from the knowledge of the $(e, 2e)$ angular distributions of the primary ionizing process. In this experiment, we have chosen to detect in coincidence the slow ejected and Auger electrons, because the effect of the fast electron, mainly scattered in the forward direction [19], as the PCI inducer is expected to be negligible. The energy of the ejected electron has been chosen close to that of the $N_5O_{23}O_{23}$ ($^1S_0$) Auger electron in order to maximize the interaction time and therefore the energy and momentum exchange between the two emerging particles.

The apparatus used for the present measurements is an electron impact spectrometer specially designed for electron coincidence experiments. It contains an electron gun, two twin electrostatic analyzers, and an effusive gas source [20]. The electron gun was operated at an energy of 1000 eV and current of $\cong 2$ mA. The electron spectrometers are independently rotatable in the scattering plane from $-15°$ to $150°$ with a precision of $0.1°$. The angular acceptances of the analyzers were set to $\pm 0.5°$ and $\pm 2.5°$ for the slow ejected and Auger electron analyzers, respectively, while the energy resolution was $\Delta E_2 = \Delta E_A = 1.0$ eV. The energy resolution ΔE_c in the $(e, 2e)$ experiments depends on the incident beam resolution and the energy resolution in each outgoing channel, while in the $(e, e'$ Auger) experiments the resolution of the coincidence Auger spectrum is determined only by the resolution of the Auger electron analyzer. Therefore, in the present case, $\Delta E_c \cong 1.0$ eV. During the experiment the incident and ejected electron energies were held fixed, while the energy collected by the Auger electron analyzer was scanned across the region of the $N_5O_{23}O_{23}$ ($^1S_0$) Auger line. The coincidence electronics of the experiment has been tuned by measuring a He $(e, 2e)$ energy separation spectrum at $E_0 = 84.5$ eV with $E_1 = E_2 = 30$ eV and using the same operation mode of the spectrometer as in the $(e, e'$ Auger) measurements.

To test the PCI angular dependence, the $(e, e'$ Auger) spectrum has been measured at two relative angles of emission ϑ_{2A} of the ejected and Auger electrons, namely, $170°$ and $25°$. The coincidence count rate at the top of the $N_5O_{23}O_{23}$ ($^1S_0$) Auger peak was $\cong 0.03 \pm 0.005$ Hz, and the true-to-random ratio was never better than a few percent. Because of these values, the accumulation time was on average about 14 h per point. The noncoincidence Auger spectrum recorded simultaneously during the coincidence measurement allows one to check for any energy drift during the measurement itself. As a further check of the stability of the coincidence spectrometer, a scan over all the $N_{4-5}O_{23}O_{23}$ region was done for both the analyzers before and after each coincidence run.

In Fig. 1 the ejected-Auger coincidence spectra are shown together with the noncoincidence Auger line (full line in the figure), simultaneously measured during the coincidence measurement. The noncoincidence yield has been scaled to the coincidence one, after a subtraction of a constant background. The coincidence spectrum taken at $\vartheta_{2A} = 170°$ overlaps almost completely with the noncoincident one [Fig. 1(b)], while the one at $\vartheta_{2A} = 25°$ [Fig. 1(a)] is clearly shifted toward low electron energies. A shift in this direction is consistent with the prediction of nonisotropic PCI theories [11,12,17] when

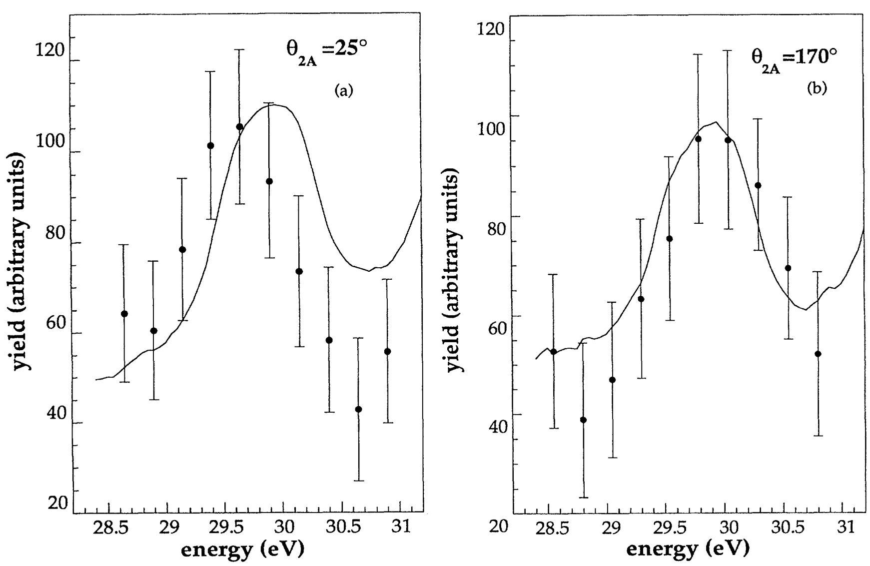

FIG. 1. Coincidence (dots) and noncoincidence (full line) spectra of the Xe $N_5O_{23}O_{23}$ ($^1S_0$) Auger line at 1000 eV incident energy, and (a) $\vartheta_{2A} = 25°$ ($\vartheta_2 = 15°$ and $\vartheta_A = 40°$) and (b) $170°$ ($\vartheta_2 = 260°$ and $\vartheta_A = 90°$).

the PCI inducer and the Auger electrons emerge in almost the same direction. To our knowledge, this is the first experimental observation of such an effect in an electron impact experiment. It is to be noted that the coincidence yield is superimposed on a continuum contribution. This contribution can be attributed to direct double ionization events, where the third electron remains undetected. From these measurements an upper rate limit of ≅0.01 Hz can be estimated for such a process.

The present results are compared with the predictions of the model developed by Kuchiev and Sheinerman [12] because this model, among all the angular dependent models of PCI [11,12,17], explicitly accounts for reactions with three electrons in the final state. In this model, the solution of the PCI problem is derived within the eikonal approach. The hypotheses of the derivation are *"(i) the potential energies of interaction between e_1 and e_2 with the ion and with the Auger electron are less than the kinetic energies of the electrons, and (ii) the electrons travel almost uniformly and rectilinearly at large distance"* [12]. The latter assumption implies that the relative angle does not change from the interaction region to the detector at infinity, i.e., the unbound electrons are assumed to travel along rectilinear trajectories. According to this model the cross section for process (1) is given by

$$\frac{d^8\sigma}{dE_2 dE_A d\Omega_1 d\Omega_2 d\Omega_A} = \frac{d^5\sigma}{dE_2 d\Omega_1 d\Omega_2} \times f(\Omega_A)\ell(E_A, \vartheta_{1A}, \vartheta_{2A}), \quad (2)$$

where $f(\Omega_A)$ is the angular distribution of the Auger electrons, and the PCI distorted line shape is given by

$$\ell(E_A, \vartheta_{1A}, \vartheta_{2A}) = \frac{\Gamma/2\pi}{(E_A^0 - E_A)^2 + \Gamma^2/4} k(E_A, \xi), \quad (3)$$

with

$$k(E_A, \xi) = \frac{\pi\xi}{\sinh(\pi\xi)} \exp\left[2\xi \tan^{-1}\frac{2(E_A^0 - E_A)}{\Gamma}\right] \quad (4)$$

and

$$\xi = -\frac{1}{K_1} + \frac{1}{|\vec{K}_1 - \vec{K}_A|} - \frac{1}{K_2} + \frac{1}{|\vec{K}_2 - \vec{K}_A|}. \quad (5)$$

E_A^0 in Eqs. (3) and (4) is the diagrammatic energy of the Auger electron, i.e., the energy difference between the one- and two-hole ion states. Because of hypothesis (ii), the time dependence of the momenta $\vec{K}_i$ is neglected and the constant asymptotic values are used in Eq. (5).

The eightfold cross section (2) can be determined only by an experiment in which all three unbound electrons are detected in coincidence. In our measurements, where only the ejected and Auger electrons are detected in coincidence, the sixfold cross section is measured:

$$\frac{d^6\sigma}{dE_2 d\Omega_2 d\Omega_A dE_A} = \int d\Omega_1 \frac{d^8\sigma}{dE_2 d\Omega_1 d\Omega_2 d\Omega_A dE_A}, \quad (6)$$

i.e., the cross section (2) integrated over all the directions of the fast scattered electron. The calculation of Eq. (6) implies knowledge of the angular distribution of the triple coincidence ejected-scattered-Auger-electron experiment, which at present is unknown. The triple coincidence angular distribution may be replaced by the product of the $(e,2e)$ cross section for the Xe $4d_{5/2}$ ionization, $d^5\sigma/d\Omega_1 d\Omega_2 dE_2$, and the angular distribution of the Xe $N_5O_{23}O_{23}$ ($^1S_0$) Auger electrons, $f(\Omega_A)$. Kuchiev and Sheinerman [12] suggested instead that the integration can be replaced by calculating Eq. (2) with a parameter ξ averaged over Ω_1. Recently, Kammerling, Krassig, and Schmidt [16], in their discussion of the different procedures used to account for angular effects in noncoincident inner shell photoionization, observed that this approximation can distort and shift the observed line shapes when the photoelectron and Auger electrons have almost the same energies. In the present case where the unobserved particle is ≅6 times faster than the observed ones and mainly scattered in the forward direction, we believe that (1) we can use the averaged ξ value given by

$$\bar{\xi} = -\frac{1}{K_2} + \frac{1}{|\vec{K}_2 - \vec{K}_A|}, \quad (7)$$

and (2) the change in the line shape due to this choice, if any, is not observable with our energy resolution.

In order to compare the PCI line shape with the experiment, the theoretical prediction has been convoluted with the energy and angular response functions of the spectrometer. The angular response function has been obtained by a computer simulation of the trajectories transmitted by the lens stack and the following dispersing element. The knowledge of this function is quite important at $\vartheta_{2A} = 25°$, because the theoretical PCI line shape convoluted with the energy response function can suffer a variation in the calculated shift as large as 80 meV when ϑ_{2A} varies from 20° to 30°. Experimental and calculated line shapes are compared in Figs. 2(a) and 2(b). A satisfactory agreement between theory and experiment is observed in both measured cases, although at $\vartheta_{2A} = 25°$ the experiment seems to indicate a shift slightly larger than the theory. This difference may be attributed to the theoretical assumption of rectilinear trajectories for the unbound electrons. Indeed, $(e,2e)$ studies [21] have shown that, due to their mutual interaction, free electrons of kinetic energy as high as 100 eV suffer a displacement of a few degrees from their original trajectory.

The angular dependence of PCI is due to the interaction among the unbound electrons. By changing the relative angle of emission of the PCI inducers and the Auger electrons, one observes an energy displacement of the Auger peaks, on either side of the diagrammatic energy. This effect shows the importance of treating on equal footing all the interactions [electron(s)-electron(s), electron(s)-ion interactions] in the final state of an ionization event induced by charged particle impact. This is a well-known fact in the study of the ionization mechanism of the outer shell by electron impact, but its relevance to PCI effects was neglected until the late 1980s, i.e., more than twenty

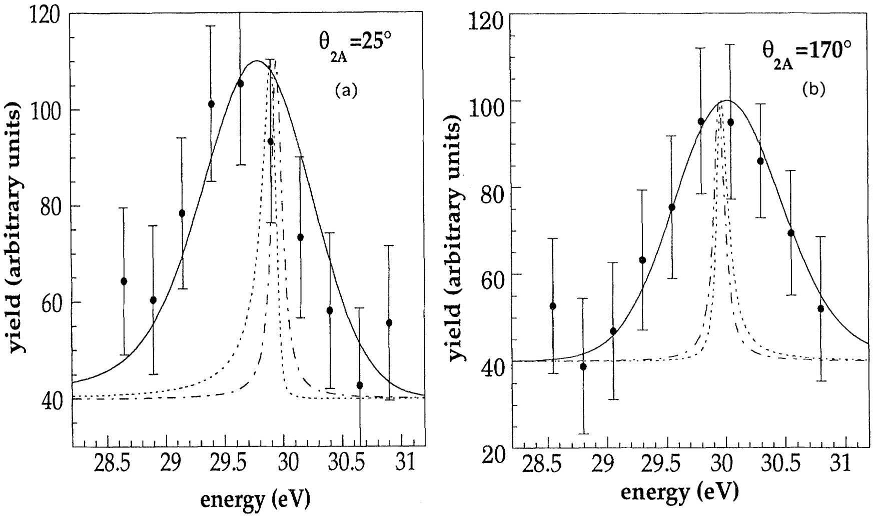

FIG. 2. Comparison between the experimental coincidence yields (dots) and the convoluted PCI line shapes (full line) of the Xe $N_5O_{23}O_{23}$ ($^1S_0$) Auger line at (a) $\vartheta_{2A} = 25°$ and (b) 170°. In the figures, the Lorentzian [dash-dotted line centered at 29.95 eV, i.e., at the diagrammatic energy of the $N_5O_{23}O_{23}$ ($^1S_0$) line] and unconvoluted PCI (dashed line) line shapes are also shown.

years after their first observation [22]. The present results and the previous experimental work [8] on the energy dependence of PCI effects show that, despite some crude assumptions, the actual PCI models give a satisfactory description of the shift and the change of the Auger line shapes. Therefore, in the future one can use PCI effects to study, at a finer level, fundamental quantities such as the atomic linewidths and the Auger transition energies. These data, of course, affect all of the branches of science where Auger line shapes are used to study "environmental" effects on the emitting atoms. For example, as pointed out by Sandner and Völkel [7], in the case of solids a comparison with the Auger lines from the free atoms may lead to wrong conclusions, when PCI effects are neglected in the analysis of the atomic data.

From a fundamental point of view, the PCI effects and the Auger resonant Raman effect [23,24] clearly show that the formation and decay of an inner hole cannot anymore be treated by two-step models, and that a unified picture of the process, such as the one proposed by Armen *et al.* [17], has to be adopted.

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