Interference effects in the Ne double photoionization studied by photoelectron–Auger-electron coincidence experiments

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The resonant double photoionization of Ne has been studied via an electron-electron coincidence experiment at a photon energy of 92.21 eV. At this energy, the kinetic energy of the photoelectrons matches exactly the energy of the Auger electrons. The overall experimental energy resolution, narrower than the natural linewidth of the intermediate state, has allowed us to observe angular- and energy-dependent interference effects due to the indistinguishability of the two electrons.

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The formation of a doubly charged ion by absorption of a single photon may proceed either via a direct process with the simultaneous emission of two photoelectrons, or via an indirect process with the sequential emission of a photoelectron and an Auger electron. The latter process is due to the formation of an intermediate excited state of the singly charged ion that autoionize (Auger decay) to the double-ionization continuum and must be considered as a resonance embedded in the double-ionization continuum [1]. This process can be described in the frame of a two-step model, i.e., photo and autoionization are treated as incoherent successive processes. This approximation relies on the fact that (i) the two free electrons can be distinguished by their kinetic energies, (ii) the contribution of the direct double ionization process is negligible, and (iii) the intermediate state has a lifetime long enough to prevent any final state interactions in the continuum. This picture might be incorrect if the energy of the incident photon is such to produce photoelectrons with a kinetic energy close to the energy of the Auger electrons. In such a case, the conventional two-step formulation does not hold anymore, and must be replaced by a one-step model where the two outgoing electrons are not specified as photoelectron or Auger electron. In this case it has been demonstrated both experimentally [2] and theoretically [3] that strong interference effects due to electron exchange appear in the probability distribution of electron pairs.

These interference effects can be observed only in experiments where the two electrons in the final state are measured in coincidence after energy and angular selection, in other words by measuring the triple differential cross section (TDCS). Moreover, to ensure the indistinguishability of both electrons, it is vital to work with an energy resolution comparable to or better than the natural linewidth of the intermediate state. Such experiments have been pioneered by Schwarzkopft and Schmidt [4] as far as the study of the energy distributions is concerned, and by Schaphorst *et al.* [5] in the case of angular distributions. More recently, Viefhaus *et al.* [2], exploiting the high resolution of the BW3 beamline at Hasylab and the high efficiency of the time-of-flight technique, have observed a clear interference effect in a photoelectron-Auger electron coincidence experiment performed for Xe $4d$ photoionization. In such equal energy sharing experiments, the observed TDCS angular and energy distributions can be also distorted by post-collisional interaction (PCI) at small mutual angle between the outgoing electrons [6]. These PCI effects, first observed in the photoionization of the Xe 4d [7], have been recently investigated by Scherer *et al.* [8] in an experiment where the two electrons were detected at a mutual angle of $20° \pm 4°$.

Our aim, in the present study, is to investigate interference in the double photoionization process by taking full advantage of the high resolution of the Photoemission Gas Phase beamline at Elettra combined with the high efficiency of the multicoincidence apparatus [9]. We present here a report on the results obtained for the double photoionization of neon with an overall energy resolution better than the lifetime width of the intermediate state. In particular, we have chosen to investigate the following process

$$\begin{aligned} h\nu + \mathrm{Ne} \rightarrow \mathrm{Ne}^{+}[2s2p^5(^3P^o)3p(^2S^e)] + e_{ph}(E_{ph}, l=1) \\ \rightarrow \mathrm{Ne}^{2+}[2s^2 2p^4(^1D^e)] \\ + e_{Auger}(E_{Auger} = 13.24 \text{ eV}, l=2) \end{aligned} \tag{1}$$

at different photon energies close to the one where $E_{ph} = E_{Auger}$. The $\mathrm{Ne}^{+}[2s2p^5(^3P^o)3p(^2S^e)]$ state can decay non-radiatively to the $\mathrm{Ne}^{2+}[2s^2 2p^4(^1D^e)]$ and

$[2s^2 2p^4(^1S^e)]$ continua. However, the measurements [5,10] have shown, in agreement with the calculations [11], that the decay to the $^1D^e$ continuum largely dominates.

The experimental setup allows us, at present, to simultaneously collect in coincidence up to fourteen pairs of electrons, with the mutual θ_{12} angle varying from 60° to 180°. Thus, while scanning the kinetic energy of the two electrons, both energy and angular coincidence distributions can be simultaneously measured. The beamline as well as the multicoincidence set-up have been previously described [9,12], thus only a brief summary is given here. The radiation source is a 4.5-m undulator (12.5 cm period) [13]. The radiation is deflected to the monochromator by a prefocusing mirror that focuses the beam at the entrance slit of the monochromator in the vertical plane, and at the exit slit in the horizontal one. The variable angle spherical grating monochromator [14] consists of two optical elements: a plane mirror and a spherical grating. The multicoincidence end station houses two independently-rotatable arrays of electrostatic electron energy analyzers located in the plane perpendicular to the photon beam direction and containing the polarization vector. The efficiency of the different detectors in the array has been calibrated by measuring the Ne$(2p)$ photolines at about 29 eV photon energy, which are characterized by an almost isotropic angular distribution [15,16]. The spectrometers are composed of two four-element lenses that focus the photoelectrons from the target region onto the entrance slits of the hemispherical deflector. Their pass energies were 2.1 eV leading to an energy resolution in noncoincidence measurements of about 80 meV. This was lower than the width of the Auger line (155±25 meV), hence complied with the condition stated by Vegh and Macek [3] in order to observe interference effects in coincidence experiments.

To investigate the process given by formula 1 with $E_{ph}=E_{Auger}=13.24$ eV, the photon energy has been set to $h\nu=92.21$ eV. In Fig. 1(a), we present the mixed photoelectron/Auger spectrum measured at this photon energy and for $\theta=30°$ with respect to the photon polarization axis. The experimental peak at 13.24 eV is well represented by a Voigt profile with a width of the Lorentzian contribution equal to 155±25 meV. This shape does not change with the detection angle and reveals that the indirect process dominate.

The procedure used in measuring coincidence spectra is the one suggested by Sheinerman and Schmidt [6], i.e., the kinetic energies E_1 and E_2 of the two electrons were varied at fixed photon energy according to the relationship:

$$h\nu - V_{\text{ion}}[\text{Ne}^{2+}(^1D^e)] = E_1 + E_2, \tag{2}$$

where V_{ion} is the ionization potential. The results presented here were obtained for the constant value of $E_1+E_2=26.48$ eV with E_i $(i=1,2)$ varying from 12.7 to 13.5 eV. Figures 1(b) and 1(c) show the coincidence energy distributions measured for $\theta_{12}=180°$ ($\theta_1=30°$ and $\theta_2=210°$) and 60° ($\theta_1=30°$ and $\theta_2=330°$) respectively. Despite the statistical uncertainty, the data display a quite different shape at the two different mutual angles and both of them do not resemble to the noncoincidence spectrum. The spectrum

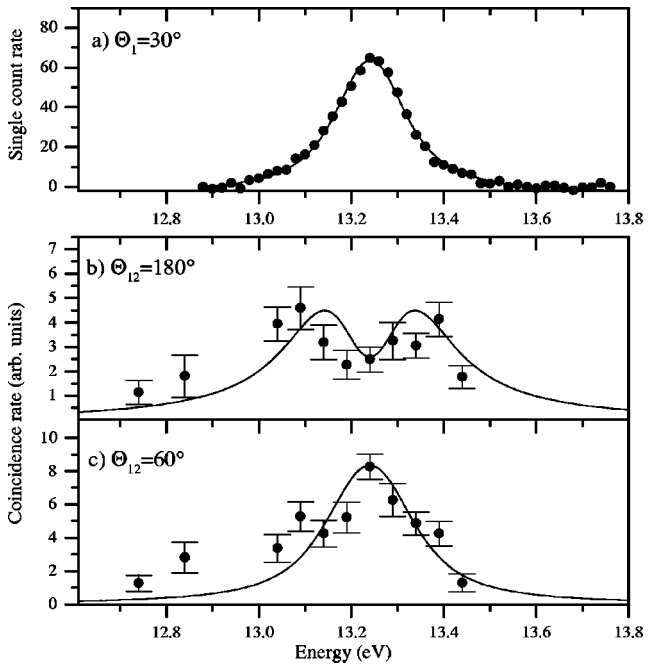

FIG. 1. Mixed photoelectron/Auger electron non-coincidence spectrum (a) at $h\nu=92.21$ eV and $\theta=30°$ with respect to the direction of the photon polarization. Photoelectron/Auger coincidence spectrum at the same photon energy for $\theta_{12}=180°$ (b) and for 60° (c). The full lines are a fit with a Voigt lineshape to the experiments (a) and the general formula by Vegh and Macek (Ref. [3]) convoluted with the apparatus function in (b,c).

taken at $\theta_{12}=180°$ clearly shows a minimum when $E_{ph}=E_{Auger}=13.24$ eV. According to the model of Vegh and Macek [3], this behavior can be qualitatively understood as the signature of the interference effect, which depends on the total spin and parity of the electron pair. In the LS coupling scheme, for the $^1D^e$ final state of Ne^{2+}, only the three $(^1P^o, ^1D^o, ^1F^o)$ electron pair configurations are allowed. They are of unfavoured type and therefore lead to a node for antiparallel emission. In a previous angle-resolved photoelectron-Auger electron coincidence experiments on neon [5] made with a photon energy resolution of 250 meV and detector energy resolutions of about 80 meV, a similar minimum was observed in the TDCS angular distribution.

The lineshapes reported in Figs. 1(b) and 1(c) can be compared with the general formula given by Vegh and Macek [3] for the one-step model. In our case, for the linearly polarized radiation produced by the undulator, the resonant amplitude reduces to the following form [5]:

$$T^m = \frac{a(E_1,E_2)Y_{10}(\Omega_2)Y_{2m}(\Omega_1)}{E_1 - E_{Auger} + i\Gamma/2} + \frac{a(E_2,E_1)Y_{10}(\Omega_1)Y_{2m}(\Omega_2)}{E_2 - E_{Auger} + i\Gamma/2}, \tag{3}$$

where the function a represents the slowly varying part of the amplitude and is assumed to be constant near the resonance [5]. Ω_1 and Ω_2 indicate the angle of emission of the two electrons. The Y_{10} and Y_{2m} spherical harmonics are as-

sociated with the ϵp and ϵd partial waves of the photoelectron and Auger electron, respectively. The TDCS can easily be calculated as an incoherent summation of the amplitudes T^m over the unobserved magnetic quantum number m of the electron associated with the ϵd partial wave. The results of the theoretical model have been folded with a 100 meV-wide Gaussian function describing the overall energy contribution of the coincidence spectrometer. In the comparison between theoretical prediction and experiment, the former has been rescaled to the data to give the best visual fit at $\theta_{12}=180°$. Then the same scaling factor has been used also for all the other measurements at the different angles.

As illustrated in Figs. 1(b) and 1(c), the theoretical predictions give a reasonable description of the experiments. In particular, for $E_1=E_2=E_{Auger}=13.24$ eV, the two denominators in formula (3) are equal, and one obtains for antiparallel emission a destructive interference leading to a theoretical zero value in the TDCS. On the contrary, for parallel emission, the two terms of the formula (3) interfere constructively and therefore explain the maximum observed at $E_1=E_2=E_{Auger}=13.24$ eV when the θ_{12} angle is decreasing. In the model used for the comparison with the measurements the contribution from the direct double photoioinization process and PCI effects have been neglected. The former approximation seems to be reasonable because the experimental observations of Schaphorst *et al.* [5] have shown that in resonance the coincidence count rate increases of about one order of magnitude with respect of the one off-resonance. PCI effects do produce appreciable effects only at quite small mutual angles [6,8]. At the mutual angles of the measurements reported in Fig. 1 the PCI effects, if any, result in a change of the coincidence line shape undetectable with the present quality of the data.

In summary, a clear interference effect, energy- and angular-dependent, has been observed in the study of the indirect double photoionization of Ne. This result has been achieved via photoelectron-Auger electron coincidence measurements, when the photon energy is tuned at the resonance value of 92.21 eV, where the photoelectron and Auger electron become indistinguishable. To investigate more carefully PCI effects, the complete study at all measurable angles and at different photon energies nearby the resonance value is in progress.

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