Resonant enhancement of photoemission leading to the Ne^+ $[2p^2]({}^1D)3p\ {}^2P$ state across the $[1s2p]({}^3P)3p^2\ {}^1P$ double-excitation resonance of Ne

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(Received 25 February 2008; published 16 May 2008)

We have measured the relative intensity of the photoemission channel leading to the Ne^+ $[2p^2]({}^1D)3p\ {}^2P$ final ionic state as a function of excitation photon energy, where square brackets indicate hole states, across the $[1s2p]({}^3P)3p^2\ {}^1P$ double-excitation resonance. Resonant enhancement is observed in the intensity's variation curve, we find that the enhancement is definitely seen at the double-excitation resonance, i.e., 902.4 eV. The result confirms the spectator-participator–type resonant Auger transition associates with the $[1s2p]({}^3P)3p^2$ doubly excited state.

DOI: 10.1103/PhysRevA.77.054702 PACS number(s): 32.80.Aa, 32.80.Fb, 32.80.Hd

With the advent of synchrotron radiation (SR) sources, a detailed investigation of single-photon multielectron excitation and/or ionization processes for isolated atom has become possible. Since such study can provide a basis for understanding the intra-atomic electron correlations, extensive studies had been performed especially for light elements, e.g., He atom, because it provides an archetypal example of three-body Coulomb problem [1–3]. On the other hand, in spite of the importance in studying the characteristics of the resonance states as well as the excitation and relaxation dynamics in the multiply excited heavier atoms, a detailed investigation of the resonant multiple excitation involving core-electrons had been hampered due to the lack of intense photon beam in the vuv to soft x-ray region. Prior to the appearance of the third generation light sources, resonant excitation of the ^1P states of Ne $[1s2p]3p^2$ [4], Ar $[1s3p]4p^2$ [5], Ar $[1s2p]4p^2$ [6], and Kr $[1s4p]5p^2$ [7] observed in photoabsorption spectra had been rare examples of resonant multiple excitation involving $1s$ electron of heavier elements. Nowadays, however, with the dramatic improvement in the performance of the modern high-brilliant SR facilities, we can apply the resonant Auger (RA) electron spectroscopy [8] in studying the multiply excited heavier atoms.

Recently, the resonantly photoexcited $[1s2p]npn'p$ and $[1s2s]nsn'p$ $(n,n' \geq 3)$ ^1P states of Ne atom, which are locating in the excitation-energy range between 900 and 940 eV, have been independently investigated by several groups [9–15]. Among these investigations, the RA electron spectroscopy was applied to the $[1s2p]3p^2$ two-particle–two-hole (hereafter referred as 2p-2h) state [10,13]. They identified the double-spectator type RA transitions, in which both of two excited electrons stay excited as spectators, by comparing the observed spectra with those theoretically calculated based on the Hartree-Fock approximation. They also studied the spectral evolution for the double-spectator–type RA transitions leading to the Ne^+ $[2p^3]3p^2$ ionic 2p-3h states from the $[1s2p]({}^3P)3p^2$ intermediate 2p-2h state.

In the present study, we will investigate the excitation and relaxation processes for the $[1s2p]({}^3P)3p^2$ multiply excited Ne atom and focus our interest on the spectator-participator–type RA transition [16,17], in which one of the excited electrons participates and the other one stays excited, leading to the Ne^+ $[2p^2]({}^1D)3p\ {}^2P$ state, since the spectator-participator–type RA transition for the doubly excited Ne atom is not experimentally confirmed yet. It should be noted here that the Ne^+ $[2p^2]({}^1D)3p\ {}^2P$ final ionic state can be also populated via the $2p \rightarrow 3p$ shakeup process accompanying $2p$ photoionization of the Ne atom [18,19]. So that the spectral evolution measurement is indispensable to distinguish the spectator-participator–type RA transition channel from the direct photoemission channel leading to the same Ne^+ $[2p^2]({}^1D)3p\ {}^2P$ state. We will extract the resonance energy and width of the doubly excited state through the measurement with constant-ionic-state (CIS) scan for the spectator-participator–type RA decay channel.

The measurements were carried out on the *b* branch of the soft x-ray undulator beamline BL17SU at SPring-8, an 8-GeV SR facility in Japan. This beamline is equipped with the highly stabilized and high-resolution varied-line-spacing-plane-grating monochromators [20,21], which can provide highly stabilized monochromatic soft x rays with resolving power of over 10000. Energy calibration of the incident photon beam was performed by recording the photoion yield spectra of Ne atom in the regions of the $[1s]np$ $(n=3,4,\ldots,\epsilon)$ as well as the $[1s2p]({}^3P)3p^2$ excited states. The accuracy of energy scale for the incident photon beam was estimated to be about ± 0.12 eV (± 0.03 eV) for absolute (relative) scale. In the present study, the photon band pass, at a photon energy of 867.12 eV, was set to be about 104 meV to achieve higher photon flux. In the estimation of photon band pass, we have adopted the value of natural width Γ to be 240 meV [22].

By using a hemispherical electron energy analyzer SES-2002 equipped with a gas-cell (VG Scienta), the measurements of RA emission spectra of Ne were performed in the excitation-energy region of the $[1s2p]({}^3P)3p^2$ 2p-2h state. The lens axis of the analyzer was in the horizontal direction, at right angles to the photon beam direction. The direction of the polarization vector of the incident photon beam, generated by the multipolarization-mode undulator [23–25], can

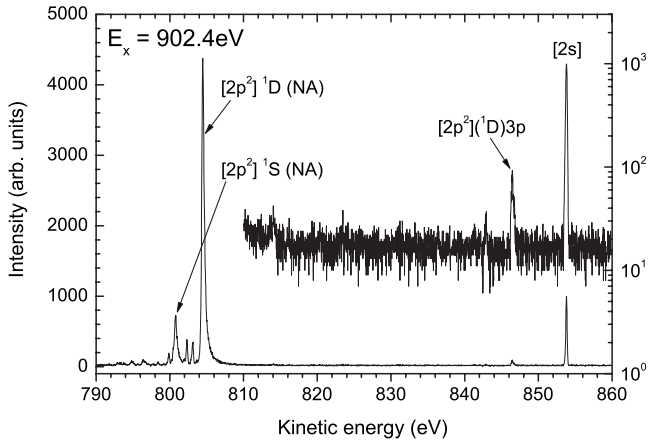

FIG. 1. Electron spectrum measured at an incident photon energy of 902.4 eV. In addition to the NA lines, several RA lines can be seen between 790 and 810 eV. At the higher kinetic energy, the objective photoemission lines are also seen.

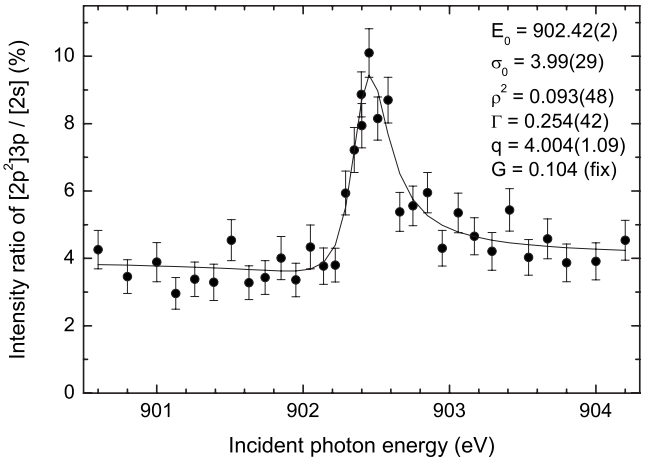

FIG. 2. Relative intensity's variation of the $[2p^2]3p$ state to the $[2s]$ state across the $[1s2p](^3P)3p^2$ double-excitation resonance. The solid line shows the result of Fano profile fitting determined by the least-squares method.

be changed by switching the operational mode of the undulator. Although the degree of linear polarization is not so high [26], we can perform the angle-resolved electron spectroscopy without rotating the electron analyzer. In the present study, however, we have set up the multipolarization-mode undulator to be a pseudohorizontal mode, in which the degree of linear polarization P_L is calculated to be about 0.86.

In the measurements, the energy resolution of the analyzer was set to be about 156 meV, whereas the Doppler broadening due to thermal motion of the sample in the gas-cell at room temperature is about 79 meV. The photon band pass was set to be about 104 meV. Thus the overall resolution for the measurements can be estimated to be about 203 meV, which is slightly smaller than the width of resonance 235 meV [10] of the Ne $[1s2p](^3P)3p^2$ 2p-2h state. The fine-energy step for the spectral evolution measurement was chosen to be about 50 meV in the vicinity of the double-excitation resonance. All of the electron spectra were normalized by accumulation time and gas pressure of Ne specimen. During the measurement, the photon flux was highly stabilized owing to the top-up-injection operation of SPring-8 [27].

Figure 1 shows a part of the electron spectrum of Ne taken at $h\nu$=902.4 eV. Several small peaks are clearly seen adjacent to the normal Auger (NA) lines, in which the NA lines are located at 800.7 and 804.4 eV. These small peaks are attributed to the double-spectator–type RA transitions leading to the $Ne^+ [2p^3](^2D)3p^2\ ^2P$ (~802.4 eV) and ^2F (~803.2 eV) states, and to the $Ne^+ [2p^3](^2P)3p^2\ ^2D$ (~799.9 eV) state, respectively [10,13]. Very weak structures are also seen below 799 eV. These might be due to the spectator-shakeup–type RA transitions [10]. Figure 1 also indicates some photoemission lines, which are coming from the Ne valence region, at higher kinetic energy region. The $[2s]$ main line is seen at 853.9 eV and the weak $[2p^2](^1D)3p\ ^2D$ satellite line can be recognized at 846.4 eV. Although we expected that we were able to observe a number of satellite lines in this region [18,19], we could not see them clearly because of the low signal-to-noise ratio. This is mainly due to the small photoionization cross sections of valence electrons in the present excitation-energy region. As we mentioned previously, this weak satellite line at 846.4 eV can be attributed to a mixture of the $2p \rightarrow 3p$ shakeup process accompanying $2p$ photoionization and the spectator-participator–type RA transition [10,15] leading to the same final ionic configuration. This implies that we may observe some interference effects if we perform the CIS-scan measurement for the $[2p^2]3p$ final ionic state, as Prince *et al.* have observed the resonant enhancement and the asymmetric shape of the partial ionization cross section for the $[1s]$ ionic state in this excitation region [12].

Figure 2 represents the energy dependence of the weak $[2p^2](^1D)3p$ satellite intensity relative to that of the $[2s]$ main line measured across the $[1s2p](^3P)3p^2$ 2p-2h state. We have obtained the intensities of these peaks by the least-squares fitting procedure and normalized the intensity of $[2p^2](^1D)3p$ line to that of $[2s]$ main line for a convenience in comparing the result with the previously reported value [19]. This normalization also cancels out the slight changes of sample-pressure in the gas-cell which gives rise to the fluctuation of the $[2p^2](^1D)3p$ intensity to be obtained. As is seen in Fig. 2, the intensity's variation curve shows the resonant enhancement of the $[2p^2](^1D)3p$ satellite line at the $[1s2p](^3P)3p^2$ double-excitation resonance. This resonant enhancement reveals the spectator-participator–type RA transition associating with the doubly excited intermediate state. As can be seen in Fig. 2, the intensity's variation curve also indicates a subtle asymmetric shape. Thus we applied the well-known parametrized equations describing a single resonance line interacting with a continuum [28] given by

$$\sigma = \sigma_0\{\rho^2(q+\epsilon)^2/(1+\epsilon^2) + 1 - \rho^2\}, \quad \epsilon = 2(E - E_0)/\Gamma,$$

where $\sigma_0 = \sigma_a + \sigma_b$ and $\rho^2 = \sigma_a/(\sigma_a + \sigma_b)$, σ_a represents the portion of the cross section corresponding to the transition to a continuum state which interacts with the resonance, E_0 the resonance energy, Γ the width of the resonance, q and ρ^2 are the so-called Fano parameters giving the shape of the reso-

nance and the strength of the continuum, respectively. The Fano profile was carefully determined by the least-squares fitting procedure using the above formulae. We have convoluted the narrow bandpass ~104 meV of the monochromator in the fitting procedure. The final fit to the experimental data yielded values of E_0=902.42(02) eV, Γ =0.254(42) eV, q=4.00(1.09), σ_0=3.99(29), and ρ^2 =0.093(48), respectively.

We found that the resonance energy was in good agreement with the previously reported values [10,13,14]. The width of the resonance was also confirmed to be in agreement with those in the previous paper within experimental uncertainty. According to the Fano profile fitting, the present result of the profile index q was positive and relatively large value, i.e., 4.00(1.09), but the q parameter deduced from the photoion yield spectroscopy was negative value, e.g., −2.86(5) [10]. According to the fact that the q parameter is related to the amplitudes for the "modified" discrete state and the unperturbed continuum state [29], we can understand why the sign of q parameter has changed. This is due to that the objective pair of the discrete and continuum states for the present CIS study differs from that for the previous photoion yield spectroscopic study.

As for the intensity ratio of $[2p^2]({}^1D)3p/[2s]$ for the nonresonant portion, we have estimated the corresponding ratio by applying the reported values of atomic subshell photoionization cross sections and asymmetry parameters [30] with adequate interpolation. The following formula was used for the estimation:

$$\frac{d\sigma}{d\theta} \propto \sigma\left\{1+\frac{\beta}{4}[3P_L\cos(2\theta)+1]\right\},$$

where σ is the photoionization cross section, P_L the degree of linear polarization of the radiation, θ the angle between the direction of emission and the polarization of the incident light, and β the asymmetry parameter. We employed the β for the $2s$ electron to be the energy-independent value 2 and the asymptotic shakeup probability for $[2p^2]({}^1D)3p$ state to be 0.075 [19]. By treating the present excitation-energy range as the sudden limit regime, we could calculate the intensity ratio to be 0.033 which is comparable to that of the present intensity ratio, i.e., ~0.036. Furthermore, we have roughly estimated the intensity of the transition leading to the $[2p^2]({}^1D)3p$ final ionic state relative to that of the double-spectator–type RA transition, e.g., the $[2p^3]({}^2D)3p^2({}^3P)\ {}^2F$ state at 803 eV. The result was deduced by the peak fitting to be about 16%. By simply applying the Fano profile fitting results, the ratio between the resonant and nonresonant terms can be considered as 2:1. Thus, at the $[1s2p]({}^3P)3p^2$ double-excitation resonance, the intensity of the spectator-participator–type RA transition leading to the $[2p^2]({}^1D)3p$ final ionic state is almost ten times smaller than that of the double-spectator–type RA transition leading to the $[2p^3]({}^2D)3p^2({}^3P)\ {}^2F$ state.

In summary, we have investigated the spectral evolution of the photoemission channel leading to the Ne^+ $[2p^2]({}^1D)3p\ {}^2P$ final ionic state in the region of the $[1s2p]({}^3P)3p^2$ double-excitation resonance. As in the case of Xe [16] and Kr [17], the present result confirms the spectator-participator–type RA transition associated with the $[1s2p]3p^2$ doubly excited intermediate state of Ne. The transition probability for the spectator-participator–type RA transition was estimated to be almost ten times smaller than that of the double-spectator–type RA transition.

This experiment was carried out with the approval of the RIKEN SPring-8 Center (Proposal No. 20070113). We are grateful to Dr. S. Shin and Dr. T. Ishikawa of RIKEN for their understanding and encouragement during the accomplishment of this experiment at BL17SU.

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