Observation of high-lying weak autoionizing resonances of Ne, Na and Mg atoms by charge-separated photoion-yield method

K Kato, T Osawa, S Obara1, Y Tohyama, T Nagata, Y Azuma1 and F Koike2
Department of Science and Technology, Meisei University, Hino, Tokyo 191-8506 Japan
1Photon Factory, Institute for Material Structure Science, KEK, Tsukuba, Ibaraki 305-0801 Japan
2School of Medicine, Kitasato University, Sagamihara, Kanagawa 228-8555 Japan

Abstract Doubly-excited 2s–12p–13pnp autoionizing resonance series of Ne atom as well as autoionizing resonances in the 2s region of Na and Mg atoms have been studied with monochromatized synchrotron radiation. Use of charge-separated photoion-yield method allowed us to detect these weak resonances in a clarified way in the yield curves of doubly-charged ions. The observed resonance states have been interpreted with the help of MCDF calculations, and the decay processes of these resonance states are discussed briefly.

1. Introduction
Autoionizing resonances in photoabsorption of atoms have been a subject of extensive experimental and theoretical studies. Resonances in rare-gas atoms, which show typical Beutler-Fano profiles [1], have been precisely studied and have often been used for calibrating the photon energies. It is quite natural that the spectrum of inner-shell resonance-excitations or outer-shell resonant double excitation is superimposed on a strong valence ionization continuum. As the inner shell concerned becomes deeper, the resonance states in the spectrum are superimposed on two or more continua and result in not only singly charged ions but also multiply charged ions with respective probabilities. Excitation of such high-lying resonance states is generally quite weak, and we may face a difficulty in discriminating the excitations from the spectra of photoabsorption, total photoions, or photoelectrons, especially for metallic vapors which need high oven temperatures. In this respect, the charge-separated photoion-yield method as used in this study is often advantageous because weaker resonances are observed in a clarified way in the double and even multiple ionization channels whose background levels are relatively low. In addition, from this kind of measurement, we may obtain information about the decay processes of observed resonance states, as well as the resonance energies and the associated configuration interaction comparing experiment with theoretical calculations.

We have carried out an experiment of charge-separated photoion spectroscopy for Ne, Na, and Mg atoms in the region of 2s ionization and have observed weak high-lying autoionizing resonances. In this report, we present the experimental spectra, describe the results of assignment and profile analysis of the resonance features, and, for the Ne and Na cases, mention briefly the decay processes of the resonance states. The details of the assignment based on furthermore precise calculations will be given and discussed in a forthcoming paper. As far as we know, the resonance series observed in this study for Ne and Mg atoms have not been reported previously.

2. Experiment
The present experiment was carried out with synchrotron radiation at the beam line BL-3B in Photon
Factory, KEK. The details of the experimental apparatus and procedure have been described in previous papers [2, 3]. Briefly, a time-of-flight mass spectrometer was used to separate ions with different charges. The monochromatized synchrotron radiation passes through the interaction region into which the target atoms are fed from a gas nozzle or an electron bombardment oven. Photoions resulting from the photon-atom interaction were pushed into a time-of-flight tube with the aid of periodic positive pulses and detected by a microchannel plate (MCP) and a subsequent electronics in counting mode. The intensities (number of counts) of singly and doubly charged photoions were recorded simultaneously with a change in the photon energy and stored in a personal computer. The photoion-yield spectra obtained were normalized to the incident photon flux monitored with the drain current at a post-focusing mirror. In the present study, the detection efficiency of doubly charged ions is larger by 5%~15% than that of singly charged ions, depending on the mass of the atom [4]. However, the present photoion-yield spectra have not been corrected for this difference because it is not essential for the discussion that follows. The energy scale was calibrated using the absorption edge and the doubly excited autoionization series of He atom [5]. The accuracy of the energy scale is estimated to be within 0.15 eV. The energy resolutions are 3 meV for Ne, 25 meV for Na, and 40 meV for Mg.

The operating temperatures of the metal oven were approximately 550 K for Na and 700 K for Mg. The vapor pressure corresponding to these temperatures is of the order of 10⁻¹ Pa, and the target pressure at the interaction section is estimated to be of the order of 10⁻³ Pa. The background pressure was less than 2 × 10⁻⁵ Pa.

3. Results and Discussion

Figure 1 shows the resonance series of Ne atom lying in the 84-90 eV region. With regard to the Ne result, we can refer to the photoabsorption measurement by Codling et al [6]. They observed resonance features at 73.8 eV and 84.9 eV. The feature at 84.9 eV corresponds probably to the first member of the resonance series observed at 85.37 eV in this study. The assignment that they suggested for this feature is 2s2p⁵(1P)3p²(1P). If this is the case, the present series can be assigned as 1s²2s2p⁵(1P)3pnp(1P).

In order to confirm this assignment, we have performed MCDF calculation using the GRASP92 code [7]. As a first step, we have made minimal-base calculation. The series limit of the Ne 1s²2s2p⁵(1P)3pnp(1P) resonances is Ne⁺ 1s²2s2p⁵(1P)3p ⁵P, which is a shake-up state associated with 2s ionization. The energies of the Ne⁺ 1s²2s2p 3p ⁵P configuration states are calculated to be 76.26 eV for the 1s²2s2p³P state and 88.35 eV for the 1s²2s2p³P state. The calculation of the 1s²2s2p³P(1P)3pnp(1P) members of the series converging to the latter limit is summarized in Table I. The calculation is in substantial agreement with the measurement, as is shown in figure 1. Table 1 includes the profile parameters $E_r$, $q$ and $\Gamma$ determined using

$$\sigma(E) = \sigma_a \frac{(E - E_r)^2}{1 + \frac{E_r}{\Gamma}} + \sigma_b$$

in which the energy resolution of the spectrometer was taken into account [8]. Here the reduced energy $\epsilon = (E - E_r)/(\Gamma/2)$ describes the deviation of the incident photon energy $E$ from the resonance energy $E_r$ scaled by $(\Gamma/2)$, where $\Gamma$ is the resonance width. The quantity $q$ is called the profile index. The quantities $\sigma_a$ and $\sigma_b$ are the interacting and non-interacting continuum cross sections [1].

The 1s²2s2p³P(3P)3pnp(1P) resonance states are considered to decay as follows. The 2s⁻¹ state of Ne⁺ (48.48 eV) lies below the threshold of Ne²⁺ formation (62.52 eV), and hence the 1s²2s2p³P(3P)3pnp(1P) resonance states autoionizing to the Ne²⁺(2s⁻¹) state end up with formation of Ne⁺ ion. However, the Ne⁺ (2s2p³s) shake-up state is estimated to lie around 72 eV. Consequently, these resonance states have non-negligible probabilities of decaying to Ne²⁺ ion either by two steps via the Ne⁺(2s2p³s) state, or directly with emission of two electrons (double autoionization). From figure 1, we find that the first member of the series, the 1s²2s2p³P(3P) state, appears as a window type resonance in the single ionization channel. This indicates that the excitation probability of this state is very small compared to the 2p ionization. We also find that there are still several weaker high-lying resonances below the resonance series under consideration.
Figure 1. The doubly-excited 2s–1p–1(P)3np autoionizing resonance series of Ne atom observed in the photoion-yield spectra. The vertical bars indicate the position of the series members calculated by GRASP92 code.

Table 1  Ne 1s22s2p5(1P)3pnp 1P autoionizing resonance series and Fano’s profile parameters

| Configuration               | Energy (eV) | E_r (eV) | Profile index, q | Width (eV) |
|-----------------------------|-------------|----------|------------------|------------|
| Ground state                | 0           | 0        |                  |            |
| 1s22p(1P)3p2               | 85.49       | 85.37 (1)| 2.5 (3)          | 0.17 (1)   |
| 1s22p(1P)3p4p              | 87.13       | 87.3 (2) | 3.4 (9)          | 0.17 (3)   |
| 1s22p(1P)3p5p              | 87.80       | —        | —                | —          |
| 1s22p(1P)3p6p              | 88.04       | —        | —                | —          |
| 2p(1P)3p limit             | 88.35       | —        | —                | —          |

The result on Na atom is shown in figure 2. These autoionizing features in Na have been previously observed in the photoabsorption spectra by LaVilla et al [9]. The resonance peaks below the 2s limit have been assigned as the members of the 2s’np autoionizing resonance series. The features above the limit have been assigned as 2s–1snl’np doubly-excited states, and the strongest feature at 72.6 eV has been assigned as 2s2p(6S)3p4s(3P),2P. It is found that the 2s–1p resonance state decays to Na2+ ion with a probability of about 2 % possibly by double autoionization. Also, it is found that roughly a half of Na atoms excited to this 2s2p(6S)3p4s(3P),2P state at 72.6 eV decay to Na+ ions and the other half decay to Na2+ ion states via the autoionization to the 2s–1 continuum.

Figure 3 shows the new result on Mg atom. We have assigned the observed features as due to 2s–1np resonances. This is based on the following consideration. Simple calculations using GRASP92 code gave values around 75 eV as the limits of Mg2+(2s22p53s) formation and approximately 124 eV as the lowest limit of Mg+(2s22p43s24p) formation. Consequently neither Mg 2s22p53sn1 resonance series nor the Mg 2s22p3s4pln series, even if they exist with appreciable intensities, lie in the present region of observation. The 2s–1np resonance energies obtained preliminarily using minimal-base

Figure 2. Photoion-yield spectra obtained for Na atom. The energy levels indicated by vertical bars refer to the paper of La Villa et al [9].

Figure 3. Photoion-yield spectra obtained for Mg. The energy levels indicated by vertical bars correspond to the present calculation, but the energy positions are shifted downwards by 2.2 eV.
Table II. The 2s–1np autoionizing resonances of Na atom.

| Configuration          | Energy (eV) | Profile parameters a) | Width (eV) |
|------------------------|-------------|-----------------------|------------|
| Ground State           | 0           | Energy $E_r$ (eV)     | Profile index, $q$ | Width (eV) |
| 2s2p$^3$3s(3S)3p $^2$P| 67.0        | 66.4 ($66.41\pm0.03$) | $-2.8$ ($-2.6$)   | 0.15 (0.20) |
| 2s2p$^3$3s(3S)4p $^2$P| 70.7        | $69.3$                | $-2.0$      | 0.11       |
| 2s–1 limits            | 71.8        | $72.6$ ($72.67$)      | $-3.1$      | 0.11 (0.14) |
| 2s2p$^3$3p4s(3P) $^2$P| 72.6        | $72.6$ ($72.67$)      | $-3.1$      | 0.11 (0.14) |

a) The values in parentheses are from the measurement of LaVilla et al [9]. Their $E_r$ values were determined at the absorption maxima.
b) The values in these lines are those determined from the Na$^{2+}$ spectrum. Other parameters were determined from the Na$^+$ spectrum.

calculation were higher by about 2.2 eV than the observation, suggesting the existence of important configurations. The energy positions indicated in figure 3 are due to the present calculation, but shifted by 2.2 eV to the lower energy side. To improve the calculation, more precise multi-configuration calculations are now in progress by taking into account the influence from 2p-doubly excited configurations. What we can say at the present stage is that the improvement of the calculation on the Mg 2s–1np resonances is unexpectedly difficult task and needs careful consideration of calculated radial wavefunctions and energies of the subshells concerned.

In the case of photoabsorption in the 2s region of Mg atom, double ionization has been found to dominate, and, as far as the observation and assignment of the resonance states are concerned, the present charge-separated spectroscopy is not always advantageous over standard photoabsorption spectroscopy. However, as has been mentioned in §1, the present method provides information about decay processes after the resonance excitation. The detailed discussion on the decay processes should be postponed until the improved calculations are over.

In summary, we have measured the charge-separated photoion-yield spectra on Ne, Na and Mg atoms in the regions of the high-lying autoionizing resonances associated with 2s excitation and analyzed them through the MCDF calculation and the fitting of their profiles. More precise multi-configuration analysis is required to obtain detailed information about the present high-lying resonance states and their decay processes.

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