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LETTER TO THE EDITOR

Interference effects in the branching ratio for the partial decay channels of the Ne ($1s^{-1}3p$) resonance

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Abstract. Strong variation in the branching ratio of partial decay channels across the resonance is observed for the $1s^{-1}3p$ resonance in Ne photoemission. It is attributed to the interference between the resonant and direct amplitudes of photoionization. The observed phenomenon is used to obtain the experimental ratio of the direct and resonant contributions to the resonance.

The study of core-excited resonant states of atoms and molecules is a comparatively new and quickly developing area within atomic and molecular spectroscopy (see, for example, Armen *et al* (2000) and references therein). In photoabsorption below a particular inner-shell ionization threshold, the electron is promoted from the inner shell to one of the unoccupied bound orbitals. The relaxation of such a highly excited state usually proceeds via electron emission, similar to the Auger process. The resonant Auger electron emission process is conventionally described within the framework of the two-step model in analogy with normal Auger decay. The two-step ansatz means that the excitation and the decay of the resonant state are considered as independent processes. In this way, a large body of experimental data has been successfully analysed; the energy positions and widths of the resonances have been obtained, together with the widths of the partial decay channels, the angular distributions of the resonant Auger electrons and their spin polarizations etc. In spite of the success of the two-step approach, or maybe because of this success, there have been many attempts to find indications of its breakdown. In principle, it is well understood that the resonant Auger process can be adequately described as a resonance in photoionization, i.e. as a one-step process. This is again in analogy with normal Auger decay after photoionization, which is in fact a resonance in double photoionization (Åberg and Howat 1982).

One of the indicators of the breakdown of the two-step model is the presence of interference, which is usually ignored in the two-step approach. It may be interference between the direct and resonant processes or interference between the two resonances. In fact, the core-excited resonance may be considered as a typical autoionizing state which is described by the Fano theory (Fano 1961, Fano and Cooper 1965) containing, naturally, the above mentioned interference effects. The first observation of interference between direct and resonant photoionization has been reported by Camilloni *et al* (1996) for the resonantly excited $2p_{3/2}^{-1}3d$ state in Ar. They observed a large variation of the branching ratio for the decay

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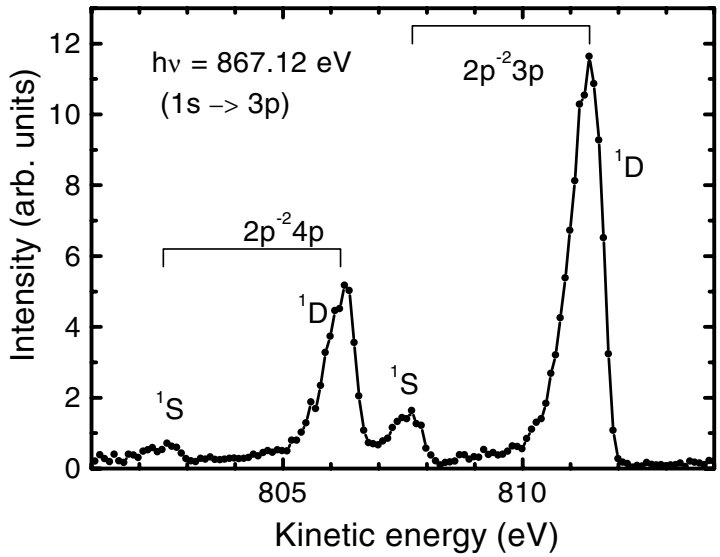

Figure 1. A part of the electron spectra of the resonant Auger transitions from the Ne $1s^{-1}3p$ state to the final Ne^+ ion states of the $2p^{-2}np$ configurations at a photon energy of 867.12 eV. The two peaks at kinetic energies of 811.4 and 807.7 eV are assigned to the ^1D and ^1S components of the $2p^{-2}3p$ configuration. The two peaks at kinetic energies of 806.2 and 802.5 eV are assigned to components of the $2p^{-2}4p$ configuration. The spectrum was recorded in the 4° direction relative to the linear polarization axis of the incident light.

into the spectator $3p^{-2}3d$ and the shake-up $3p^{-2}4d$ channels. Such variation is typical of the decay of autoionizing states; it comes about because of the different partial wave contributions in the resonant and direct processes (Starace 1977). Interference between two overlapping resonances has been observed by Rubensson *et al* (1996) in the excitation of Ne between the $1s^{-1}3p$ and $1s^{-1}4p$ resonances. In this letter we report our observation of the strong variation of the branching ratio for the decay of the $1s^{-1}3p$ state in Ne when the photon energy is scanned across the resonance. The effect is similar to that observed in Ar by Camilloni *et al* (1996). We interpret the result more consistently, taking into account both of the two interference effects described above, and show that it is possible to evaluate the relative strength of the direct transitions to the continuum.

The experimental setup and procedure are similar to those described in our previous paper (Yoshida *et al* 2000). Briefly, the experiment was performed at SPring-8 using a high-resolution plane grating monochromator (Ishiguro *et al* 1999) installed in the c-branch of the soft x-ray figure-8 undulator beamline 27SU. The photon band pass was set to ∼200 meV at a photon energy of ∼867 eV. The monochromatized photon beam was incident upon the focal point of a cylindrical mirror electron energy analyser (CMA); the energy resolution of the CMA was set to 0.5 eV. The spectra were recorded simultaneously in the directions 4° and 56° relative to the linear polarization axis of the incident light (see Yoshida *et al* 2000).

Figure 1 shows part of the spectra of the resonant Auger transitions to the final Ne^+ states of the $2p^{-2}3p$ and $2p^{-2}4p$ configurations at a photon energy of 867.12 eV (the top of the resonance). This spectrum was recorded in the 4° direction. In this figure we are particularly interested in the branching ratio to the $2p^{-2}(^1D_2)3p$ and $2p^{-2}(^1D_2)4p$ states; we measured this branching ratio R as a function of photon energy through the $1s^{-1}3p$ resonance. The measured ratio R for each photon energy was independent of the directions, 4° and 56°, of electron detection.

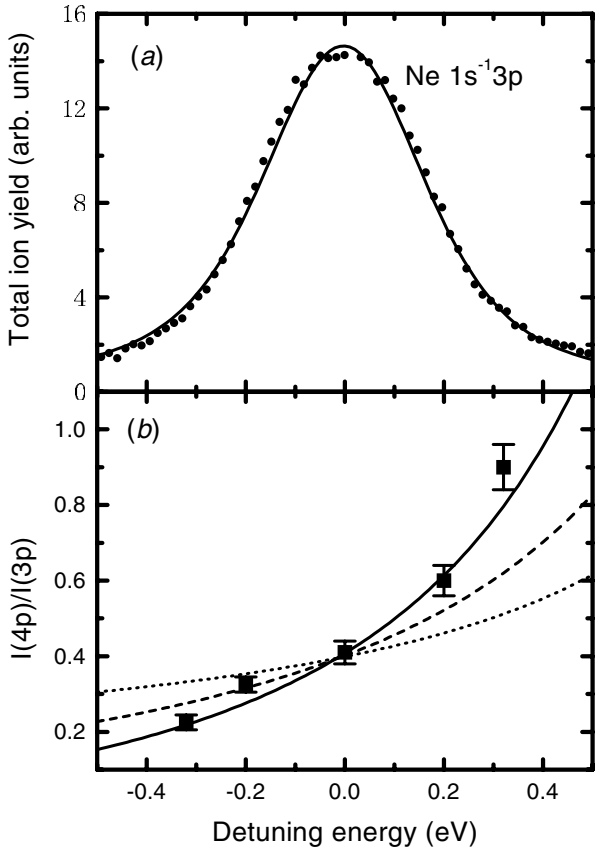

Figure 2. (a) Ion yield spectrum across the $1s^{-1}3p$ resonance of Ne. The solid curve is the summation of $|a_1(E)|^2$ and $|a_2(E)|^2$ calculated using a core hole lifetime-broadening (Γ) of 254 meV, $\eta = -0.035$ and a photon bandpass of 200 meV. The detuning energy is the energy difference between the photon energy and 867.12 eV, which corresponds to $E - E_1$. (b) Variation in the branching ratio to the $2p^{-2}(^1D_2)3p$ and $2p^{-2}(^1D_2)4p$ states. Solid curve, the best theoretical fit to the experimental data points ($\eta = -0.035$). Dashed curve, calculated with the value $\eta = -0.016$ estimated from the independent experiment. Dotted curve, calculated taking account of only the inter-resonance interference ($\eta = 0$).

The resulting branching ratio R is shown in figure 2(b) together with the theoretical curves calculated as described below, while figure 2(a) gives the photoabsorption spectrum of Ne in the region of interest.

The amplitude of photoionization with the photoelectron emitted in channel i can be written in the following form:

$$a_i(E) = \langle i \mid D \mid 0 \rangle + \sum_k \frac{\langle i \mid V \mid k \rangle \langle k \mid D \mid 0 \rangle}{E - E_k + i\Gamma_k/2} \,. \tag{1}$$

Here, the first term on the right-hand side of the equation is the amplitude of direct photoionization, the dipole matrix element describing the transition from the ground state $|0\rangle$ to the final channel $|i\rangle$. The second term describes the contribution of the resonances. E_k and Γ_k are the position and the width of the kth resonance, $\langle k \mid D \mid 0 \rangle$ is the dipole amplitude of the resonance excitation, while $\langle i \mid V \mid k \rangle$ is the amplitude of its decay to the ith channel. In our case, $k = 1$ corresponds to the resonance $1s^{-1}3p$ at a photon energy of 867.12 ± 0.05 eV and

$k = 2$ corresponds to the resonance $1s^{-1}4p$ at a photon energy of 868.69 ± 0.04 eV (Coreno *et al* 1999). We consider two direct channels which correspond to the two final ionic states: $2p^{-2}3p$ ($i = 1$) and $2p^{-2}4p$ ($i = 2$).

The amplitude (1) can be rewritten in a more convenient form using the normalization condition.

$$a_i(E) = f_i + \sum_{k=1,2} \gamma_{ki} D_k \frac{\sqrt{\Gamma_k/2\pi}}{E - E_k + i\Gamma_k/2} \,. \tag{2}$$

Here we have introduced a shorthand notation for the direct amplitude $f_i \equiv \langle i \mid D \mid 0 \rangle$ and for the resonant dipole amplitude $D_k \equiv \langle k \mid D \mid 0 \rangle$; we also introduced the relative partial decay width $|\gamma_{ki}|^2 = \Gamma_{ki}/\Gamma_k$ where Γ_{ki} is the partial decay width of the resonance k decaying into the channel i. If we ignore the contribution of the direct transitions and the interference, then in the region of each of the resonances the integral cross section summed over all channels is

$$\sum_i \int_{\Gamma_k} |a_i(E)|^2 \mathrm{d}E \approx |D_k|^2 \,. \tag{3}$$

We are interested in the energy dependence of the ratio $R = |a_2(E)|^2/|a_1(E)|^2$ in the region of the first resonance. In order to calculate it we make some additional approximations. We assume that both resonances have the same width, $\Gamma_1 = \Gamma_2 = \Gamma$. We ignore the contribution of the direct process, $|f_i|^2$, which is small in comparison with the resonant and interference terms. We assume also that the phase of the amplitudes f_i and γ_{ki} is the same since it is determined by the phase of the continuum wavefunction of the emitted electron (we ignore any additional phase correction due to channel coupling). The amplitudes D_k can be chosen to be real. With these assumptions the square of the channel amplitude can be written as

$$|a_i(E)|^2 = \frac{\Gamma}{2\pi} \sum_{k=1,2} \frac{|\gamma_{ki}|^2 D_k^2}{(E - E_k)^2 + \Gamma^2/4} + 2\sqrt{\frac{\Gamma}{2\pi}} \sum_{k=1,2} \frac{|f_i|\,|\gamma_{ki}| D_k (E - E_k)}{(E - E_k)^2 + \Gamma^2/4}$$
$$+ \frac{\Gamma |\gamma_{1i}|\,|\gamma_{2i}| D_1 D_2 [(E - E_1)(E - E_2) + \Gamma^2/4]}{\pi [(E - E_1)^2 + \Gamma^2/4][(E - E_2)^2 + \Gamma^2/4]} \,. \tag{4}$$

In expression (4), the first term represents the contributions from the two resonances and the second term gives the contribution from the interference between the direct and resonant amplitudes. This term was considered by Camilloni *et al* (1996) in the case of the Ar resonance. The final term gives the interference between the two resonances and was considered by Rubensson *et al* (1996). From equation (4) it can be seen that the ratio of the squared amplitudes R depends on the ratio of the dipole amplitudes D_2/D_1 and the ratio of the direct amplitudes $|f_2|/|f_1|$, as well as on the width parameters $|\gamma_{ki}|$ and Γ. Most essential is the ratio between the intensity of the direct and resonance transitions which may be characterized by the dimensionless parameter $\eta^2 = \Gamma |f_1|^2/D_1^2$ (for the first resonance). In general, the contribution from the direct process is much smaller than the resonant contribution. This is confirmed by the almost symmetric line-shape of the resonance. Therefore, it is very difficult to measure the relative intensity of the direct process. However, information on the direct amplitude can be obtained more readily from the interference effect because the interference term depends linearly on the amplitudes while the intensity term depends quadratically on the amplitudes.

In order to evaluate η, we derive all other parameters from the experimental data. According to expression (3), the ratio of the dipole amplitudes squared, $(D_2/D_1)^2$, can be estimated from the ratio of the photoabsorption cross sections of the two resonances. We obtained the ratio $1 : 0.37 \pm 0.02$ from our measured intensities of the $1s^{-1}3p$ and $1s^{-1}4p$ resonances, which is consistent with the data obtained by Rubensson *et al* (1996). The

measured ratio gives $|D_2/D_1| \approx \pm 0.61 \pm 0.03$; the choice of the sign will be considered later. The total width of the resonances was taken from the recent paper by Coreno *et al* (1999), from which $\Gamma = 0.254 \pm 0.02$ eV. The partial relative widths $|\gamma_{ki}|^2$ were taken from the results of our measurements of the intensity ratios of the resonant Auger emission to the $2p^{-2}3p$, $2p^{-2}4p$, and $2p^{-2}5p$ states, following excitation to the $1s^{-1}3p$ and $1s^{-1}4p$ levels. Based on our experiment we derived the following values: $|\gamma_{11}|^2 = 0.715 \pm 0.03$, $|\gamma_{12}|^2 = 0.285 \pm 0.03$, $|\gamma_{21}|^2 = 0.0 + 0.03$, and $|\gamma_{22}|^2 = 0.205 \pm 0.05$, in reasonable agreement with the calculations by Aksela *et al* (1989). The intensity ratio $|f_2|^2/|f_1|^2$ of the direct transitions to the two open channels $2p^{-2}4p$ and $2p^{-2}3p$ can be estimated from the relative intensity of the corresponding satellites in the 2s photoionization. Using experimental data published by Kikas *et al* (1996) and by Svensson *et al* (1988) and linearly interpolating them to our photon energy (∼867 eV), we obtain $|f_2|^2/|f_1|^2 \simeq 0.14 \pm 0.04$.

All the parameters necessary to describe the ratio $|a_2(E)|^2/|a_1(E)|^2$, as well as the total intensity $|a_1(E)|^2 + |a_2(E)|^2$ (i.e. the absorption spectrum), are fixed, except for the dimensionless parameter $\eta = \sqrt{\Gamma}|f_1|/D_1$ which characterizes the relative strength of the direct and resonance transitions. Considering η as a fitting parameter we have been able to obtain good agreement of the theoretical curve and experimental ratio with $\eta = -0.035(+0.004, -0.006)$ (see the solid curve in figure 2(*b*)). The indicated uncertainty includes the statistical error in the measurement of the ratio R as well as the uncertainties of all other parameters used in expression (4) and quoted above. It does not include the uncertainty due to the approximate character of expression (4) itself. The relative phases between the amplitudes f_1 and D_1 and between the amplitudes D_1 and D_2 are important for explaining the experimental observation. A good fit can be obtained only with negative values of η and D_2/D_1.

We also estimated η^2 indirectly through the relative intensities of the resonance Auger transition $1s^{-1}3p \rightarrow 2p^{-2}3p$ and the direct $2s^{-1}$ photoionization peak measured using the same experimental apparatus. With a photon bandwidth of 0.2 eV we have measured the ratio $I(2s^{-1})/I(1s^{-1}3p \rightarrow 2p^{-2}3p) \simeq 0.01 \pm 0.005$. Using this value and the extrapolated values of the relative intensities of the main $2s^{-1}$ and satellite lines from Kikas *et al* (1996) and Svensson *et al* (1988) we obtained an estimate for the $|\eta|$ parameter of 0.016 ± 0.008. This value is smaller than that obtained from fitting the interference effect to the experimental data. However, considering the rather simple approximations used, we conclude that our estimate is broadly in agreement with the value obtained from the branching ratio variations. The main source of the relatively large uncertainty in the estimate of $|\eta|$ is, in this case, the uncertainty of the measured ratio $I(2s^{-1})/I(1s^{-1}3p \rightarrow 2p^{-2}3p)$. We note, however, that calculations of the interference effect with the estimated value of $\eta = -0.016$ deviate considerably from the experimental points (see the dashed curve in figure 2(*b*)). We also note that the effect of the interference between the two resonances, though noticeable, is not as significant as the effect of the interference between the direct and resonant transitions. This is illustrated by the dotted curve in figure 2(*b*), calculated taking account of only the inter-resonance interference.

In conclusion, the relative strength of the direct and resonance processes has been obtained from the measured effect of the strong variation in the branching ratio across the resonance $1s^{-1}3p$ in Ne. The observed variation is interpreted as a manifestation of the interference between the direct and resonant transitions. It demonstrates the breakdown of the two-step model in the description of core excited resonances.

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