Photoabsorption cross section and ion-yield spectra of helium double-excitation resonances

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The photoabsorption cross section and ion-yield spectra of He have been measured in the region of the double-excitation resonances below the $N=2$ threshold. For resonances decaying predominantly by autoionization [the $2,0_n$ series, low values of n; notation of Herrick and Sinanoğlu, Phys. Rev. A **11**, 97 (1975)], the two kinds of spectra yield the same information. For resonances where substantial decay by fluorescence occurs ($2,1_n$ and $2,-1_n$ series), the ion-yield spectrum differs significantly from the cross section. The ratios of peak intensity of one series to the other are clearly different and the Fano line shape parameter (q) of the $2,1_n$ series also changes as a function of n, in qualitative agreement with the trends expected when the fluorescence channel is taken into account. These direct measurements confirm the influence of fluorescence on the cross section and agree with previous indirect measurements.

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INTRODUCTION

The continuous development through the 1990s of soft x-ray synchrotron light sources has meant that the ever higher intensity and resolution has provided many opportunities for experimenters. Helium is the simplest noble gas and these improvements led first to the discovery of previously unobserved but theoretically predicted double-excitation resonances [1,2], then later to the observation of unexpected triplet and dipole forbidden states [3,4]. The detection of states via the fluorescence decay and metastable atom signals [5–7] led to a paradigm shift in our understanding of the decay of the higher excited states. The importance of spin-orbit coupling was recognized [7,8], and it became clear that fluorescence is not simply a minor correction to autoionization, as for deeper core levels, but rather it is the main decay channel for many states. Thus it was concluded that the ion yield may not approximate the cross section well for these resonances [6]. Berkowitz [9] has pointed out that above about 20 eV, and above the ionization potential in any case, for most atoms and molecules the quantum photoyield is close to 1, that is, one ion-electron pair is created for each photon absorbed, and so the ion yield is generally proportional to the photoabsorption cross section. This generalization needs to be refined in the narrow ranges near resonances, and in the present case just below the double-excitation thresholds. Since these resonances are very sharp, it is only with newer high resolution light sources that such investigations are possible.

So far conclusions concerning the cross section have been drawn from separate measurements of the ion yield and the fluorescence decay channel. The goal of the present work was to investigate whether measurement of the cross section is influenced by the fluorescence channel and, if so, how it differs from an ion-yield spectrum for the doubly excited states below the $N=2$ threshold. Absolute cross sections have been measured frequently in the continuum region before [[10], and references therein], but it is rare to measure the cross section on sharp resonances. Measurements with laboratory line sources have good resolution, but since the sources are not continuous in energy, spectral profiles cannot be determined. Electron scattering techniques have been applied to measure absolute cross sections but their low resolution (of the order of 1 eV) means they are not adapted to sharp resonances. For He in particular we are not aware of systematic cross section measurements in the double-excitation region, and the present work fills this gap.

EXPERIMENTAL DETAILS

The experiments were carried out on the gas phase photoemission beamline Elettra [11] with the resolution set to approximately 1.1 meV (2 meV for broader resonances). At the highest resolution the transmission function is not a simple Gaussian but consists of a main peak and two sidebands due to diffraction at the entrance slit; each sideband had an intensity of 5–8 % of the main peak. This transmission function was determined for the isolated $2,-1_3$ resonance and used in quantitative fitting to extract line shape parameters. The fits were carried out by convoluting this transmission function with the Fano profile:

$$I=\sigma_a\frac{(q+\varepsilon)^2}{1+\varepsilon^2}+\sigma_b, \quad (1)$$

where $\varepsilon=(E-E_r)/(\Gamma/2)$, and E is the photon energy, E_r the resonance energy, Γ the lifetime of the state, σ_a the component of the continuum cross section that interacts with the discrete state, σ_b the component that does not interact, and q the Fano parameter.

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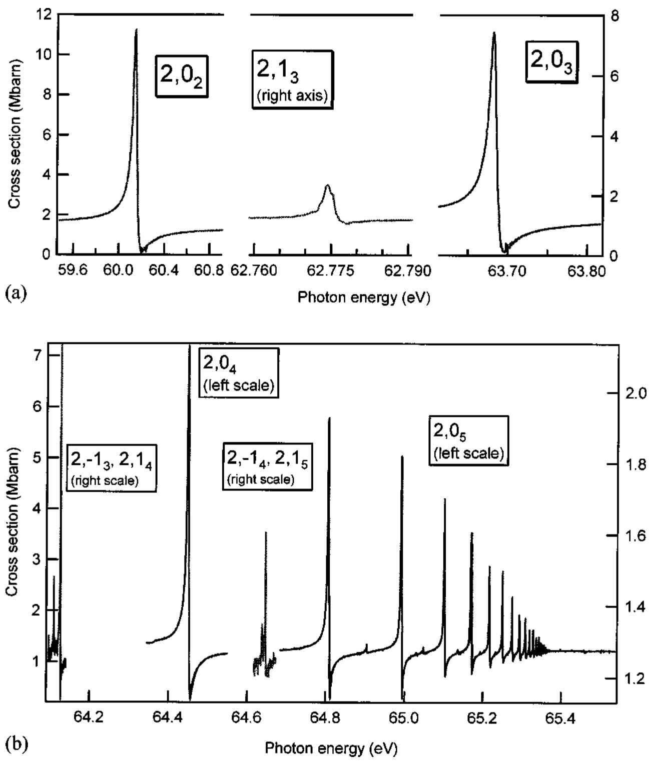

FIG. 1. Overview of the resonances below the $N=2$ threshold. (a) Lower resonances. (b) Higher resonances.

TABLE I. Line shape parameters.

State	Fano parameter: Cross section q'	Fano parameter: Ion yield q	Fano parameter: Ref. [2]	Lifetime (meV): Cross section	Lifetime (meV): Ion yield	Lifetime (meV): Ref. [2]
$2,0_2$	-2.75		-2.75	38		37
$2,0_3$	-2.5		-2.5	9.5		10
$2,0_4$	-2.7	-2.6	-2.4	4.4	3.3	4.0
$2,1_3$	-3.6	-3.6	-3.5	0.9	0.3	0.5
$2,1_4$	-3.9	-3.2	-3.2	<0.6	<0.7	0.3
$2,1_5$	-4.1	-3.0 ± 0.2	-3.2	<0.8	<0.3	<0.1

The spectra were measured with a double-ionization chamber [12] similar to that described by Samson *et al.* [13] at typical pressures of 0.7 mbar for the cross section and three orders of magnitude lower for the ion yield. This instrument measures the absolute cross section provided the ion yield is nonzero, and has the advantage that the effects of stray light, higher order light, and saturation effects are reduced with respect to single-ion-chamber methods. The ion-yield spectra were measured in the same cell by reducing the pressure so that the ion yield was in the linear region, and measuring the ion current from one cell. An aluminum filter was used to reduce the second order content of the radiation. The energies were calibrated to the values of Domke *et al.* [2].

We use the labels of Herrick and Sinanoğlu [14] and Lin [15], namely, $2,0_n$ for the strongest series ($n+$ in the original notation of Madden and Codling [16]), $2,1_n$ for the next strongest (originally $n-$), and $2,-1_n$ for the third strongest (originally $2pnd$).

RESULTS AND DISCUSSION

Figure 1 shows the photoabsorption cross sections of a set of states in the $2,0_n$ series, and Table I lists the parameters extracted. For the $2,0_n$ resonances, the values of the Fano parameter q and the lifetimes are in good agreement with the results of Domke *et al.* [2]. To obtain best fits, a small background offset is necessary [σ_b in Eq. 1 is nonzero]. There are no other continua present and decay by fluorescence is negligible and so σ_b should be identically zero (this is the case of a single bound channel and single continuum in the language of multichannel quantum defect theory). This background of 0.16 Mb therefore represents the signal due to residual experimental offsets (stray light, etc.) and finite resolution.

For the $2,1_3$ state, Fig. 1(a), the fits give a value of $q=-3.6$ for both ion yield and cross section and lifetime widths of 0.9 and 0.3 meV, respectively. The measured value of the lifetime is limited by the resolution and so is in poor agreement with theory (0.105 meV) while the q value is similar to that of Domke *et al.* (-3.5), Table I, and lower than their theoretical value (-4.25). We conclude that a reasonably accurate value of q can be extracted, but since the resolution is significantly larger than the natural width, only an upper bound for the lifetime can be extracted.

Figure 2 shows a comparison of the ion-yield spectra and absolute cross sections of the $2,-1_n$ and nearby $2,1_{n+1}$ resonances for $n=3$, 4, and 5. The relative peak heights of the states depend on the measurement technique, with the first state of each pair being markedly more intense when the absolute cross section is measured rather than the ion yield. This is because the $2,-1_n$ states decay mostly by fluorescence and this channel is not detected in an ion-yield spectrum. The absolute measurement gives a better approximation to the relative cross sections, although still not quantitative.

For the $2,1_n$ series, fluorescence begins to play a role at high values of n, although the autoionization transition rate is dominant for the states studied here [17]. In this case we have three channels: fluorescence, autoionization, and the continuum channel. In general terms we therefore expect in Eq. (1) that the background cross section σ_b will be nonzero. A second observation is that in the cross section measurements the q parameter increases with the quantum number n, but this is not so in the ion-yield measurements. Previous calculations [2] indicated that q should decrease or remain constant.

Robicheaux *et al.* [18] have discussed in detail the closely related case of photorecombination, the inverse process of photoabsorption, for the situation in which fluorescence and autoionization are competing channels. They found that the Fano factor q is proportional to $q'A/(A+R)$, where A and R are the autoionization and radiative decay probabilities, respectively, and q' is the Fano q parameter in the absence of radiative coupling. We suggest that this is also the case for

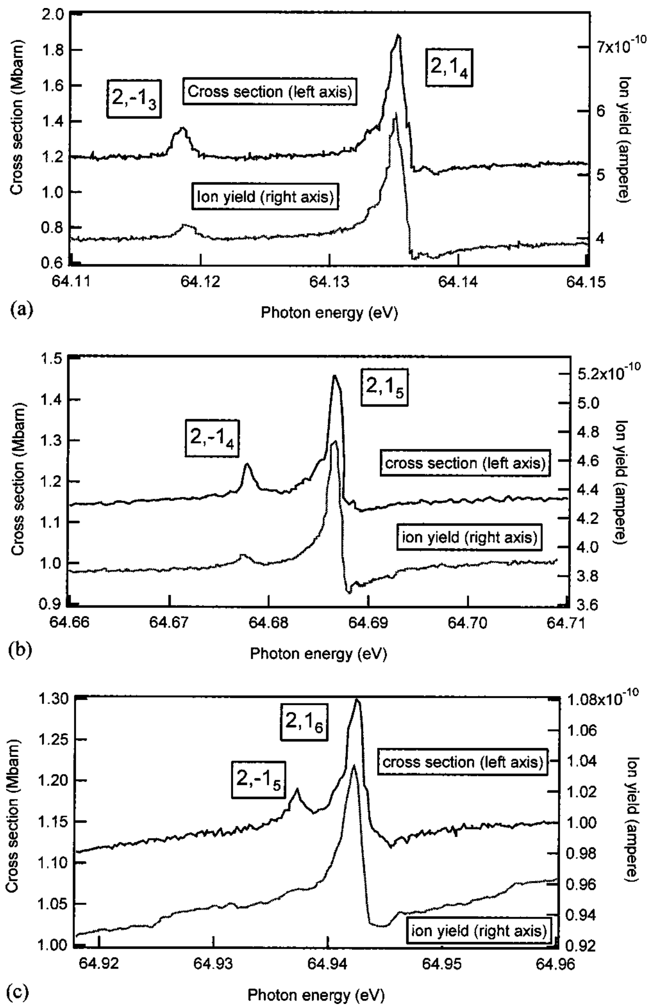

FIG. 2. (a) Comparison of ion yield and cross section for the $2,-1_3$ and $2,1_4$ resonances. (b) Comparison of cross section and ion yield for the $2,-1_4$ and $2,1_5$ resonances. (c) Comparison of cross section and ion yield for the $2,-1_5$ and $2,1_6$ resonances.

the present case of photoabsorption, $q=q'A/(A+R)$, where q is the line shape parameter observed by ion yield, and q' is the parameter observed for the absolute cross section. We can use the values of radiative and autoionization decay rates calculated by Liu *et al.* [17] to estimate the change in q, Table II. This qualitatively explains the observed trend in which the value of q' increases with quantum number n, and therefore with the relative fluorescence rate.

For autoionizing resonances in the absence of fluorescence and interacting with only a single continuum, the cross section must go to zero at $\varepsilon=-q$. Robicheaux *et al.* [18] have noted that in the presence of the fluorescence channel this is no longer true.

TABLE II. Experimental and calculated ratios of the Fano index measured with cross section to the Fano index measured with ion yield.

State	Ratio [(cross section)/(ion yield)]	
	Expt.	Theor.
$2,1_3$	1.0	1.03
$2,1_4$	1.22	1.09
$2,1_5$	1.36 ± 0.1	1.19

TABLE III. Comparison of ratio of peak heights of absorption resonances.

States	Yan *et al.* [19]	Schulz *et al.* [1]	Ratio (cross section)	Ratio (ion yield)
$2,-1_3/2,1_4$	0.34	0.05 ± 0.01	0.21	0.09
$2,-1_4/2,1_5$	0.39	0.13 ± 0.03	0.22	0.06
$2,-1_5/2,1_6$		0.11 ± 0.03	0.2	<0.02

The effective value q' obtained in the presence of fluorescence is different from the value of q related to the autoionization part of the resonance, and it is easy to understand the direction of change of q: the Lorentzian line shape increases the positive part of the cross section, increasing the modulus of q, i.e., if q is negative, q' is more negative, and if q is positive, q' is more positive.

Yan *et al.* [19] have calculated the photoabsorption cross section using R-matrix theory and convoluted their results with a spectral resolution function of 1 meV width, very close to the current value, to obtain ratios of the peak to peak heights of the $2,-1_n$ and $2,1_n$ states. Their results and ours are shown in Table III, together with values estimated roughly from Ref. [1] by enlarging the published figures. Clearly, the cross section gives better agreement with Yan *et al.*; the residual difference of a factor of 2 is likely to be due to the fact that the resolution is poorer than the intrinsic linewidths and so a simple convolution is only a rather approximate way of simulating the spectra. The ratio of our peak heights for ion yield is different from that of Schulz *et al.*, although their resolution was reported to be only slightly better than ours. The reason for this discrepancy is not known but may imply that our resolution is a little worse than estimated. Nevertheless, the cross section ratio is consistently higher than their ion-yield ratio.

CONCLUSIONS

To date it has been inferred indirectly that in measuring or calculating the line profiles of the helium double-excitation states, it is necessary to take into account the fluorescence decay channel. We have shown this directly, namely, that fluorescence influences the value of the cross section determined in an experiment, the line shape, and in particular the value of q. This is so even when the resolution is well below the lifetime broadening. Thus a correct understanding of the cross section under resonant conditions requires consideration of all decay channels.

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