Threshold electron-Auger electron coincidence spectroscopy of sulphur 2s hole decay in H$_2$S

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Introduction

Measurement of the Auger electron spectrum in coincidence with the photoelectron emerging from a given inner-shell hole presents the advantage that it is possible to select exclusively the Auger lines associated with this specific hole. It offers moreover the possibility to achieve higher resolution on accumulation of the Auger spectrum than the core-hole lifetime width that is imposed on conventional Auger spectroscopy. Here, the complete definition of the doubly charged final states, brought about by the coincidences, leads to this ‘sub-natural linewidth’ regime [1].

We have developed a dedicated experimental set-up based on the use of synchrotron radiation in which the photoelectrons are produced at threshold [2]. Detection of threshold photoelectrons takes advantage of the high energy resolution (few meV) and high (4π) collection efficiency that can be attained. We present here results on sulphur 2s Auger decay in H$_2$S using this threshold electron-Auger electron coincidence technique. The 2s hole has a very short lifetime resulting in a 1.8 eV broadening of the non-coincident Auger lines. Our coincidence technique enables us to achieve higher resolution than the lifetime width and to extract information on H$_2$S$^{++}$ formation associated with the decay of the sulphur 2s hole.

Experimental results

Figure 1 shows a two-dimensional plot of threshold electron-Auger electron coincidence yield from H$_2$S as a function of both the incident photon energy, $h\nu$ and Auger electron energy, $KE$. The energy resolution of the Auger electron analyser is set to around 0.7 eV (FWHM). Each H$_2$S$^{++}$ state has a definite ionization energy, $IE$, and one of the two electrons is restricted to have essentially zero kinetic energy, so that the formation of the state appears along a diagonal line defined as $KE = h\nu - IE$. Several diagonal stripes are exhibited on the plot, therefore, corresponding to H$_2$S$^{++}$ states. In analogy to the Ar 2s Coster-Kronig decay [3], and as has been assigned a priori, the orbital characters of the H$_2$S$^{++}$ states are attributable to (S2p)$^1$(v)$^1$. A conventional Auger spectrum shows much blurred structures in the corresponding kinetic energy range, though it was measured with a much higher resolution (0.1 eV) than the present one [4]. This fact demonstrates the power of the threshold electron-Auger electron coincidence technique for spectroscopic use on dication states. Coincidence yields for formation of the H$_2$S$^{++}$ states are enhanced above the sulphur 2s threshold (~236 eV), which implies that decay of the sulphur 2s core hole results in formation of H$_2$S$^{++}$ states. Formation of H$_2$S$^{++}$ states is seen also below the threshold. This results from direct double ionization to the H$_2$S$^{++}$ states, and autoionization from H$_2$S[(S2p)$^1$(v)$^1$+(nl)$^1$] lying near the H$_2$S$^{++}$ states and probably enhances the formation of the H$_2$S$^{++}$ states.

References

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