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Predominance of Coster-Kronig Preceded Auger Decay in Ru Determined by Auger–Photoelectron Coincidence Spectroscopy

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Introduction: As one moves to the left in the Periodic Table, Coster-Kronig (CK) preceded Auger transitions play an increasingly important role in the decay of core hole the open shell 3d and 4d transition metals. This behavior has been inferred by the difference between $3d_{3/2}$ and $3d_{5/2}$ core level line widths of the 4d transition metals and by the evolution in the shape of the $M_{45}VV$ Auger spectrum as one moves to the left through the series.[1] However, owing to the strong overlap of the M_4VV and M_5VV Auger lines, and large backgrounds upon which they ride, direct observation of the Auger emission associated with the CK preceded channel, or its relative probability, has not been possible. We have used Auger-photoelectron coincidence spectroscopy (APECS) and found that Ru $3d_{3/2}$ core holes are about four times as likely to decay via a CK-preceded transition compared to the direct channel. In contrast, for a near noble metal such as Pd, the direct channel is about 10 times more likely than CK preceded decays.

Methods and Materials: Using APECS, we have measured the line shape of the Ru M_4VV and M_5VV Auger transitions in coincidence with Ru $3d_{3/2}$ and $3d_{5/2}$ core photoelectrons, respectively, from a clean, well-ordered Ru(0001) surface. The surface was prepared by repeated sputtering and annealing followed by high temperature anneals in oxygen to remove residual carbon contamination. Two electron energy analyzers were used to perform the measurements. One remained fixed on the core level of interest while the other scanned the energy region of the Ru Auger transitions.

Results: Figures 1(a) and 1(b) below show the M_5VV and M_4VV Auger spectra obtained in coincidence with their respective core levels. The singles spectrum is shown as the dashed line. If we neglect matrix element effects, one would expect that the M_4VV spectrum has a similar line shape as the M_5VV spectrum but is shifted to higher kinetic energy by an amount equal to the difference in core level binding energies (~4.5 eV in this case). In contrast, we find that the two spectra are almost identical with essentially no shift. This is demonstrated in Fig. 1(c) where the two coincidence spectra (rescaled to the same peak height) are plotted on the same graph. The Figure shows that the M_4VV line has a small amount of excess emission at the high-energy edge as compared to the M_5VV line. The difference spectrum shown in Fig. 1(c) gives the intensity for direct M_4VV decay. The remainder of the spectral weight is associated with the CK-preceded transition. We find that the emission associated with the direct decay accounts for approximately 20% of the total M_4VV spectral weight indicating that the core hole has a vastly higher probability of decaying via the CK-preceded channel.

Conclusions: The intrinsic line shapes of the Ru M_4VV and M_5VV Auger spectra are remarkably similar, indicating that the overwhelming majority of Ru $3d_{3/2}$ core holes decay via a Coster-Kronig preceded transition. We also illustrate how APECS can be used to determine branching ratios in core hole decay paths.

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References: [1] N. Martensson and R. Nyholm, Phys. Rev. B 24, 7121 (1981).

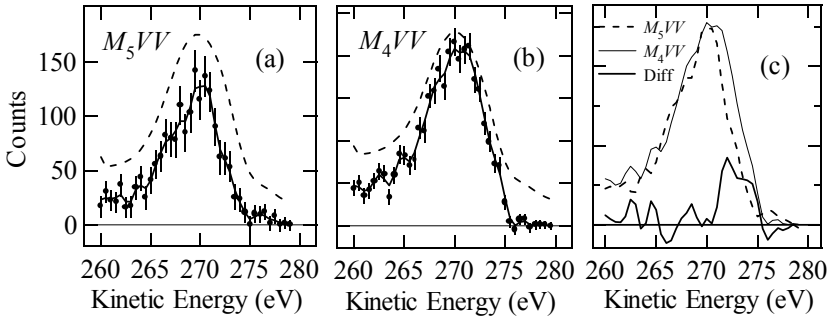

Fig. 1: Coincidence spectra of the (a) Ru M_4VV and (b) Ru M_5VV Auger transitions. (c) A direct comparison of the two.