Inelastic resonant K scattering near threshold

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Introduction

Besides normal x-ray fluorescence lines, a low-energy K satellite has long been observed and attributed to a "radiative Auger" effect. Here, photoelectron emission is followed not by a single Auger electron or fluorescence x-ray emission but by simultaneous emission of both an electron and a photon [1]. This curious "radiative-Auger" effect is quite strong and challenges our understanding of x-ray excitation and decay processes. Despite its identification almost 30 years ago, there has been little work to understand its nature or to quantify the strength of the transition. Using tunable synchrotron radiation, we have studied the relative strengths and energy spectra of secondary fluorescence compared to the radiative-Auger emission in Ge. By studying the radiative-Auger effect below, near, and above threshold, we have eliminated alternative explanations for the observed spectral dependence of the radiative-Auger line.

Methods and Materials

Measurements were made on the UNI-CAT beamline 33-ID at the Advanced Photon Source [2]. A schematic of the setup is given in Figure 1. A high-heat-load, double-crystal Si(111) monochromator was used to produce an ~2 eV bandpass monochromatic x-ray beam. This beam was focused onto a single crystal of Ge. The beam size at the sample was ~ 200 x 700 μm^2. The vertical beam size could be further reduced with a slit mounted just upstream of the sample.

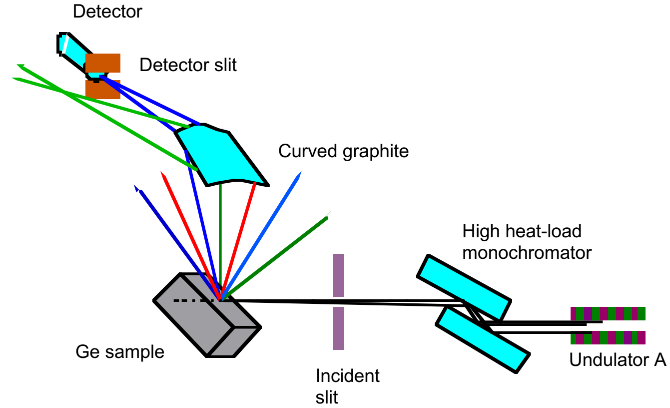

Figure 1: Curved graphite spectrometer with ~10–20 eV resolution.

The sample was mounted on a large, eight-circle Kappa diffractometer. The sample's crystallographic orientation and asymmetry could be adjusted to study the influence of absorption and crystal symmetry on the measured fluorescence.

The x-ray emission spectra were measured with a graphite energy-dispersive spectrometer [3]. This wavelength dispersive spectrometer utilizes a sagittally bent mosaic-graphite crystal that doubly focuses the fluorescence radiation onto a detector. For these measurements, the energy of the fluorescence was measured in a theta/two-theta geometry with a slit in front of the detector. The energy of the spectrometer was ~20 eV with a 0.2 mm slit at the detector and a 0.2 mm slit in front of the sample.

Results

The competing secondary fluorescence spectral shape and intensity were studied by tuning the incident x-ray energy to the Ge $K_{\alpha1}$ and $K_{\alpha2}$ energies. The two spectra were summed according to the ratio of $K_{\alpha1}$ to $K_{\alpha2}$. The resultant spectra is illustrated in Figure 2.

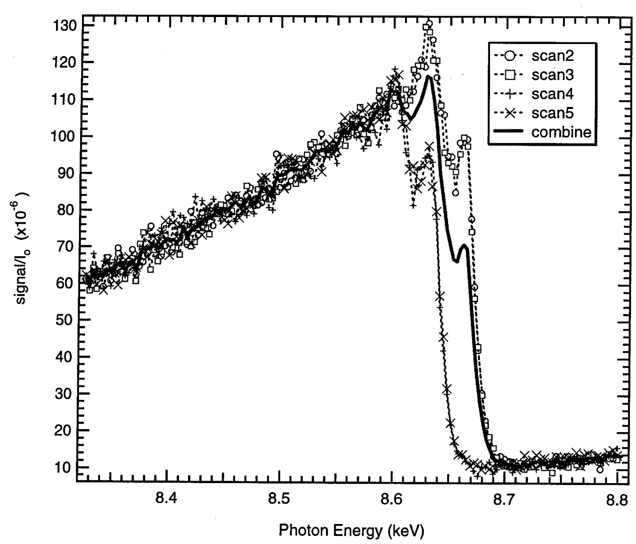

Figure 2: Resonant Raman scattering from Ge excited by incident photon energies of 9.886 keV (Ge $K_{\alpha1}$) and 9.855 keV (Ge $K_{\alpha2}$) and then combined to represent the spectrum for Ge fluorescent radiation. The leading-edge maxima are the L_{II} and LIII electrons involved in the shakeup.

Next, the x-ray energy was tuned near the K absorption edge of Ge and the spectra were again measured. Although there was still a small signal from the secondary induced resonant Raman scattering, this part of the spectra was dominated by the intense radiative-Auger channel. A simple estimate of the intensity of the secondary resonant Raman scattering can be made.

A ratio can be calculated for the expected resonant Raman scattering caused by the incident Ge K_α flux generated by the synchrotron spectrum to that internally generated by excitation at and above the Ge K threshold [4].

With the assumption that the incident flux from the synchrotron at 9.876 keV is the same as that at 11.103 keV, the ratio of the expected resonant Raman scattering at our typical scattering angle $2\theta = 20^o$ is:

I_{RRS} Ge K_α (Incident) / I_{RRS} (Secondary) = 5.6

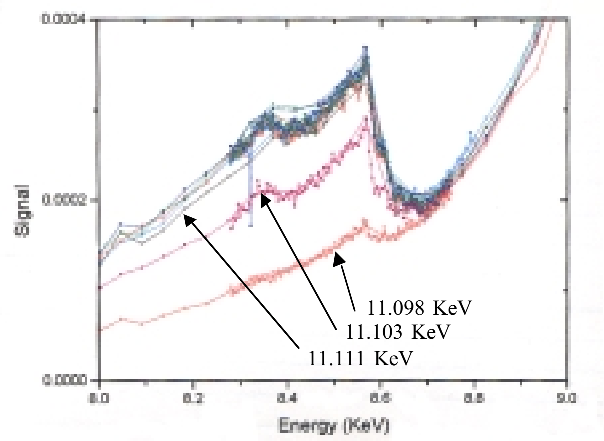

Figure 3: Resonant Raman scattering excited at the Ge K edge (11.103 keV) increases in intensity as the incident x-ray energy increases through the edge. The energy of the resonant Raman scattering is shifted lower by ~65 eV, which is less than the 110 eV expected from a Z+1 atom (which could be caused by post collision interactions) [5]. The resonant Raman scattering signal strength excited at threshold dwarfs that generated by Ge Ka in the sample, though it should be more intense by about a factor of 5.6.

Discussion

Calibration of the incident flux for the two photon energies will allow for a direct comparison of the resonant Raman scattering generated by external photons and those created at threshold. The effect of a core hole state on the emission of resonant Raman scattering allows studies of multiple electron interactions. The resolution of this experiment can be greatly enhanced through the use of spherical single-crystal optics. We have begun tests of a Ge analyzer with ~3.5 eV resolution at the 333 reflection. This analyzer will allow for much more accurate measurements of chemical shifts near threshold. As shown in Figure 4, the measured energy resolution is near its theoretical limit.

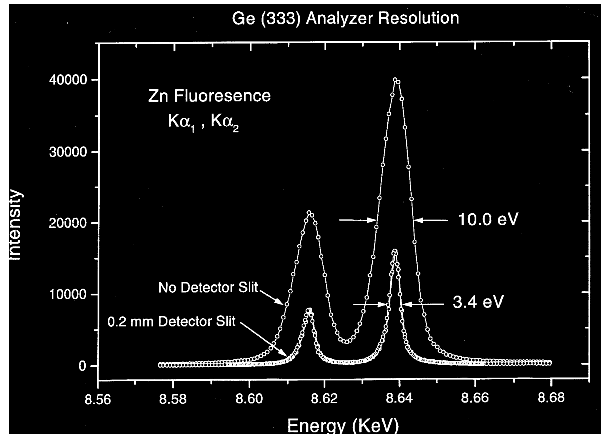

Figure 4: Energy resolution obtained with a spherical Ge analyzer and 333 reflection.

Acknowledgments

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References

[1] T. Aberg and J. Utrianen, *Phys. Rev. Lett.* **22**, 1346 (1969); Keski-Rahkonen and J. Utriainen, *J. Phy. B.* **7**, 55 (1974).

[2] P. Zschack, G.E. Ice, J.Z. Tischler, H. Hong, D. Robinson, P.R. Jemian, B.C. Larson, H. Chen, G.G. Long, and R.W. Broach, *Synchrotron Radiation Instrumentation: 11th U. S. National Conference, Stanford Linear Accelerator Center, Stanford, CA.. AIP* in print.

[3] G.E. Ice and C.J. Sparks, *Nucl. Instr. and Methods* **A291**, 110 (1990).

[4] C.J. Sparks, *Advances in X-ray Analysis Vol. 19*, (Kendel/Hunt Publishing, Dubergque, IA, 1976)19–52.

[5] G.S. Brown, M.H. Chen, B. Crasemann, and G.E. Ice. *Phys Rev Lett.* **54**, 1142 (1985).