INTRODUCTION

Femtosecond electron diffraction (FED) is a technique used to study the ultrafast structural dynamics that occur during excited atomic states and phase transitions. Short pulses of high-energy electrons are scattered off an incident target through Bragg diffraction (Figure 1).

The wave functions of the scattered electrons overlap, resulting in either constructive (coherent) or destructive (incoherent) interference. This produces a diffraction pattern of intense rings (or "peaks") with intensity and spacing dependent upon the geometric atomic structure of the sample. By studying the electron diffraction pattern, it is possible to measure the structural characteristics of solid materials and how they change as a function of temperature and time.

Figure 1: Bragg’s Law for coherent (left) and incoherent (right) scattering from a crystal lattice.

In a crystal, atoms are arranged in a highly ordered and periodic lattice, and having constant interatomic spacing gives rise to well-defined diffraction peaks. In an amorphous solid, there is no global structure so the diffraction pattern is diffuse. However, there still exists an amount of localized structure resulting from densely packed shells (Figure 2)(1).

RESEARCH METHODS

In order to obtain femtosecond electron pulse generation, we used a frequency-tripled Ti:Sapphire pulse laser to back-illuminate a photocathode. By applying a magnetic field, electrons are ejected from the cathode, accelerated to an energy of 70keV, and focused towards a sample target by a magnetic solenoid lens. The diffracted electrons strike a phosphorescent screen and are photographed with a CCD camera. Our apparatus is shown in Figure 3. Further details can be found in [2].

RESULTS

In this research we studied the structural properties of Al film and amorphous ZrCu using femtosecond electron diffraction. We measured the location and intensity of the second diffraction peak (R(220)) of our aluminum sample (20nm standing film) for temperatures ranging from T = 139K to T = 300K (Figure 7). The coefficient of thermal expansion is the slope of the best-fit line through these points and was measured to be (2.25 ± 0.06) x 10^-5 m/K. Similarly, we measured the temperature dependence of the relative intensity of the second (R(220)) and first (R(111)) peaks to be (3.9 ± 0.4) x 10^-8 K^-1.

Various image processing techniques are first applied to the diffraction image to isolate a single diffraction ring (Figure 5). When a suitable ring has been found, a quasi-Newtonian optimization algorithm is used to minimize the square difference between the ring pixels and a circle centered at the estimated diffraction center.

Figure 2: Atoms in a crystalline solid (left) are closely packed in an ordered lattice arrangement, while in an amorphous solid (right), there is no global structure so the diffraction pattern is diffuse. However, there still exists an amount of localized structure resulting from densely packed shells.

For time-resolved applications, a new "pump" laser can be used to excite the sample a few picoseconds before the electron pulse. By changing the path-length difference of the pump laser, it is possible to observe energy transition relaxation time and lattice structure dynamics in real-time [2].

Custom software was developed this summer to aid in the analysis of the diffraction pattern images. Its purpose is to accurately determine the center location of the diffraction rings and perform a radial average about the center to obtain a radial intensity curve. A graphical user interface (GUI) was developed using Python’s Qt4 package to make the software easier to use for future researchers (Figure 4).

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This method was used to obtain the location and intensity of the second diffraction peak to within 0.01 pixels for crystal samples, and 0.04 pixels for amorphous samples. After the first diffraction image has been processed, the program can automatically run through multiple images with minimal user interaction. Using a 2.8GHz computer processor with 6GB of RAM, the algorithm takes 5-20 seconds per image to complete.

REFERENCES


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Using this method, we were able to precisely find the center coordinates of the diffraction patterns to within 0.01 pixels for crystal samples and 0.04 pixels for amorphous samples. After the first diffraction image has been processed, the program can automatically run through multiple images with minimal user interaction. Using a 2.8GHz computer processor with 6GB of RAM, the algorithm takes 5-20 seconds per image to complete.

A 2D radial intensity profile was then constructed for the diffraction images (Figure 6). Subtracting the background allows for precise measurement of diffraction peak position and intensity [3]. Diffraction peaks were found to be a convolution of Gaussian and Lorentzian distributions, and were fit with Voigt profiles [4].

In this research we studied the structural properties of Al film and amorphous ZrCu using femtosecond electron diffraction. We measured the location and intensity of the second diffraction peak (R(220)) of our aluminum sample (20nm standing film) for temperatures ranging from T = 139K to T = 300K (Figure 7). The coefficient of thermal expansion is the slope of the best-fit line through these points and was measured to be (2.25 ± 0.06) x 10^-5 m/K. Similarly, we measured the temperature dependence of the relative intensity of the second (R(220)) and first (R(111)) peaks to be (3.9 ± 0.4) x 10^-8 K^-1.

The radial intensity curve for amorphous ZrCu is shown in Figure 8a with scattering vector Q = 4π sin θ/λ, where λ is the wavelength of the incident electrons and θ is the scattering angle. We found that the background follows a Lorentzian profile (red curve), with the fit passing through the mean of the intensity oscillations caused by the shell structure. After subtracting the background and normalizing the intensity using [5]

$$\tilde{I}(Q) = \frac{I(Q) - \langle Q \rangle}{\langle Q \rangle}$$

we observed the reduced structure factor behavior (Figure 8b). We then applied a Fourier transform via numerical integration:

$$y(2) = \frac{2}{\lambda} \int_0^\lambda [I(Q) - \langle Q \rangle] \sin(2\pi Q) dq$$

going from momentum-space to real-space to obtain the total pair correlation function using [6]. We observed that the 1st shell of ZrCu is located at 5.4 angstroms, 2nd shell at 9.0 angstroms, and 3rd shell at 13.9 angstroms. The accepted literature value for ZrCu is 2.7 angstroms.

Future investigations will study how the shell structure of amorphous ZrCu changes with temperature and pump-probe excitation. We would also like to investigate the structural dynamics of PbSe quantum dots, a superconducting material, using the pump-probe method.

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