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ION-INDUCED X-RAY STUDIES WITH A HIGH LUMINOSITY VON HAMOS

CRYSTAL SPECTROMETER *

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A high-resolution, high-efficiency, von Hamos geometry, Bragg crystal X-ray spectrometer has been developed and mounted on a beamline at the Holifield Heavy Ion Research Facility at the Oak Ridge National Laboratory. Measurements have been made of K and L X-rays emitted from a variety of targets and projectiles. Instrument performance characteristics are reported here along with spectra from fast projectile ions and very low intensity target emission — areas of measurement for which this spectrometer is especially suitable.

1. Introduction

The Nuclear Science Applications Group at the Oak Ridge National Laboratory has been studying for some time the effects of chemical environment on X-rays induced by heavy-ion impact [1-3]. To advance the study of these effects as well as for use in other research areas involving X-ray emission from a variety of projectile ions and targets, we have constructed a high-resolution, high-efficiency, position sensitive detector based, curved-crystal X-ray spectrometer and installed it on a beamline at the Holifield Heavy Ion Research Facility. A brief description of the spectrometer design was given previously [2]. Final instrument performance characteristics are presented below, along with data from two specific areas (other than the measurement of chemical effects) where the device is also especially effective.

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2. Spectrometer

First proposed by Gouy [4] and developed by von Hamos [5], the design shown in fig. 1 offers substantial gain in collected X-ray intensity over conventional flat crystal spectrometers. The von Hamos geometry differs greatly from the horizontal-focusing, curved-crystal geometries of Johann [6] and Johansson [7] which concentrate X-rays in the dispersive plane. For a localized source, the latter geometries normally require re-positioning of the crystal and detector in order to simultaneously collect data over a substantial range of energies. Vertical focusing instruments concentrate X-rays in the non-dispersive plane and permit, at one positioning of components, data collection over an energy bandwidth limited primarily by the crystal and detector lengths. As shown in fig. 1, a detected X-ray position (D) corresponds geometrically to a particular Bragg angle θ_B , and hence to a specific lab energy through the modified Bragg relation

$$E(\text{keV}) = 12.398 \, n / [2d(\text{\AA}) \sin \theta_B] , \quad (1)$$

where n is the diffraction order and $2d$ is the crystal lattice spacing. An extended crystal (C) with radius of curvature ρ subtends a range of angles for a given source point (S). In our application, the source of X-rays is a rectangular ion-target or electron-target interaction region, defined either by an X-ray limiting slit or by the beam spot on a solid target. A position-sensitive detector is located at the focus, on the crystal's axis of revolution. The source-to-crystal and crystal-to-detector distances are equal. Ideally, the detector axis should coincide with the crystal axis; however, in gas detectors position resolution is seriously degraded for X-rays having off-normal incidence, so that a proportional counter (like the one used here) must be oriented perpendicular to the incoming X-rays. Several position-sensitive,

vertical-focusing spectrometers have been constructed and used very successfully, especially in measurements for plasma diagnostics [8-10]. Previous instruments of this type have been limited, however, to relatively narrow bandwidth ($\Delta E/E$) and to specific operating energy regions. The need for a more versatile spectrometer to study a wide range of X-ray energies led us to develop the instrument shown in fig. 2 with the following performance design goals: total energy range using a few Bragg crystals = 1 to 10 keV; energy resolution = 1 eV fwhm at 2.3 keV and 5 eV fwhm at 10 keV; and detected energy bandwidth = 10% of the central energy. An additional aim was maximization of total efficiency.

The total instrumental energy resolution is determined by all the components which contribute to the smearing of Bragg angles for X-rays detected at a single position at the detector and by the spatial resolution of the detection system. An ion-induced X-ray source width narrower than a few hundred μm is difficult to achieve without substantial loss of beam intensity. This extended source size produces a major component of the overall resolution loss. The Bragg angle smearing due to finite source size can be reduced by increasing the crystal radius. However, a large radius leads to a significant reduction in collection efficiency which can only be compensated by increasing the size of the crystal. Unfortunately, large crystals result in increased imaging distortion and are difficult to make and mount.

To quantify these effects, imaging distortion was studied for a number of particular designs with ray-tracing programs written especially for this project [11]. The tradeoff between resolution and collection efficiency led us to choose a radius $\rho = 25.4$ cm such that the width of the distorted image (at the detector) of a 250 μm wide (Δx) by 1 cm high (Δz) source would result in a calculated total instrument energy resolution of ≤ 1 eV fwhw at 2.5 keV.

The crystal size was chosen as 10 cm x 10 cm, to satisfy the requirement of high collection efficiency and to fully illuminate the detector at all accessible Bragg angles. The thin crystals are permanently mounted on aluminum blocks machined to a precise concave cylindrical surface (Interactive Radiation, Inc., Northvale, NJ). The detector is a gas-flow, position-sensitive proportional counter (PSPC) with an active length of 10 cm and a spatial resolution of $\approx 150 \mu\text{m}$ for 2 keV X-rays using P10 (90% Ar, 10% CH_4) gas at one atmosphere pressure (TEC, Knoxville, TN).

As shown in fig. 2, the crystal and detector are mounted on pre-aligned, parallel dove-tail tracks in a stainless steel ultra-high vacuum chamber. Positions of both the crystal and detector are remotely controlled to allow operation for central Bragg angles between 30 and 70 degrees. A micro-computer system has been developed to automatically control three stepping motors which position the crystal and detector.

With a few commonly used crystals [ADP(101), Quartz(10T1), LiF(200), and Si(100)] nearly the full 1-10 keV energy region can be covered. Within a detected bandwidth the X-ray energy as a function of channel number is weakly non-linear. Energy calibration is normally achieved using several electron excited X-ray lines occurring in the chosen bandwidth.

The luminosity or "speed" of the spectrometer can be expressed in several ways. For estimates of count rates in a single line, the total efficiency for detection of X-rays within a resolution width is probably most useful. This can be defined as the ratio of the number of X-rays detected within a resolution width (I_d) to the total number emitted (I_{tot}). Assuming isotropic emission this ratio is given by

$$I_d/I_{\text{tot}} = R_{\text{avg}} (\Delta\Omega/4\pi) D_{\text{eff}} . \quad (2)$$

In eq. (2), R_{avg} is the crystal reflectivity averaged over the crystal rocking angle; $\Delta\Omega$ is the product of the crystal height (in radians from the source) and the opening angle corresponding to the angular total system resolution; and D_{eff} is the detector efficiency including window absorption. For a 10 cm high quartz($10\bar{1}1$) crystal, 250 μm wide source, 150 μm detector resolution, and at 2.3 keV, where the detection efficiency is estimated to be 35%, the calculated efficiency per resolution width is about 1×10^{-5} . The 10 cm long PSPC simultaneously intercepts over 300 resolution widths.

The spectrometer efficiency can vary significantly (even within a bandwidth) if the crystal reflectivity or detector efficiency changes with position due to imperfections. With our instrument the portions of the crystal and detector being used to detect a particular energy X-ray can be varied in a controlled way. The relative collection efficiency can be measured as a function of position along the crystal by monitoring the detected X-ray count rate for a line source while scanning the crystal along its mounting track, i.e., parallel to the x-axis in fig. 1. Characterization data acquired in this manner for a quartz($10\bar{1}1$) crystal using electron-excited sulfur K_{α} X-rays is displayed in fig. 3. The intensity variations shown there will result in distortions in X-ray spectra measured using this crystal. The relative efficiency of the detector along its active length for X-rays diffracted and focused by a given crystal is measured in a similar manner. Spectral data can be at least partially corrected with the aid of these two efficiency curves.

3. Sample data

The advantages of high resolution and "speed" make this instrument uniquely suited for a variety of ion-atom collisions studies. Two specific areas in which we have already begun measurements will be discussed below.

Intensities and energies of X-rays from few-electron heavy ions are of fundamental interest. Preparation of these ions normally involves acceleration to high energies and subsequent stripping in thin targets to achieve the required high charge states. Observation in the laboratory of X-rays emitted from the fast ions results in unavoidable Doppler shifting and broadening. Measurements are often made on X-rays emitted in a direction normal to the beam velocity to minimize the shifts. The spectrometer described here sacrifices energy determination somewhat but achieves low Doppler broadening and high efficiency by collecting projectile X-rays with large but nearly constant Doppler shifts. The geometric relationship between the Doppler and Bragg angles (θ_D and θ_B) in the lab frame is shown in fig. 1. When the crystal axis is oriented exactly normal to the ion beam velocity vector $\vec{\beta}c$ (c is the velocity of light),

$$\cos \theta_D = \sin \theta_B \cos \Psi . \quad (3)$$

The relativistic Doppler relation between projectile and laboratory frame X-ray energies (E_p and E_L , respectively) [12] is

$$E_L = E_p / [\gamma (1 - \beta \cos \theta_D)] , \quad (4)$$

where $\gamma = 1/\sqrt{1-\beta^2}$. When combined with eqs. (3) and (1), the lab frame X-ray energy is given by

$$E_L = (E_p/\gamma) \pm C_B \beta \cos \psi ; \quad C_B \equiv 12.398 \text{ n}/2d(\text{\AA}) . \quad (5)$$

The sign of the second term depends on the direction of $\vec{\beta}$, being negative for the case shown in fig. 1. For a 10 cm high crystal ψ varies from 0° at the center to $\psi_{\max} = 11^\circ$ at the crystal edge. This leads to a broadening and shifting of line energies such that the Doppler induced width δE_D and centroid energy $\langle E_L \rangle$ are given by

$$\delta E_D = C_B \beta (1 - \cos \psi_{\max}) , \quad (6)$$

and

$$\langle E_L \rangle = (E_p/\gamma) \pm [C_B \beta (1 - \cos \psi_{\max})/2] . \quad (7)$$

The measured energy and resolution are only weakly dependent on $\psi_{\max}$ and the broadening is therefore relatively small even for large area (i.e., fast) crystals. As an example, a spectrum of silicon X-rays emitted from 2.6 MeV/u Si ions passing through a 5 $\mu\text{g}/\text{cm}^2$ carbon foil is shown in fig. 4. Silicon Lyman α_1 and α_2 separated by 1.8 eV are clearly resolved there. Several lines from doubly-excited He-like Si are also identified according to tabulated energies [13]. By limiting the crystal height to 7 cm ($\psi_{\max} = 7.8^\circ$), the overall energy resolution including Doppler broadening (~ 1 eV) has been held to < 1.5 eV at 2.14 keV while the relatively high efficiency of the system has been retained. If the same height crystal were oriented in a configuration to collect X-rays emitted about a normal to the beam, the resulting Doppler spread would amount to > 40 eV.

The high "speed" of this spectrometer permits high-resolution studies of very-low-probability phenomena. For example, several low-energy structures exist in ion-induced spectra which are usually ignored because they occur at $\sim 1/100$ of the intensity of the characteristic lines and the higher energy satellites [14,15]. These structures include edges and peaks from multielectron transitions such as those occurring in Radiative Auger Emission (RAE) and Radiative Electron Rearrangement (RER) [16-18]. In RAE a free electron and an X-ray are simultaneously emitted. In RER a 2s electron fills the initial K vacancy with simultaneous filling of an initial 2p vacancy by the remaining 2s electron. Increasing the number of initial L (2p) vacancies shifts the X-ray

energies upward (as in the case of the $K L^n$ satellites) so that a separate peak occurs for each initial $K L^n$ vacancy state. A spectrum taken with the current instrument of aluminum RAE and RER structures is shown in fig. 5.

4. Summary

The attributes of this spectrometer that make it excellent for ion-atom collisions studies are: (1) High resolution so that individual atomic transitions can be measured, (2) High efficiency which allows low-probability events to be studied in detail, (3) Simultaneous collection of X-rays over a wide bandwidth so that time-dependent effects like radiation damage can be studied, (4) Ease of operation such that minimum time is spent on instrument adjustment during data acquisition, (5) Versatility of operation such that a wide range of energy regions are accessible and in a variety of configurations so that target and projectile X-rays can be measured taking advantage of Doppler shifting yet minimizing Doppler spreading, and (6) Compatibility with accelerator ultra-high vacuum environment.

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FIGURE CAPTIONS

- FIG. 1. Schematic diagram of a vertical-focusing, Bragg crystal spectrometer in the von Hamos geometry. S is an X-ray source point; C is the cylindrically-curved crystal of radius ρ ; and D is the detection point at focus.
- FIG. 2. Diagram of the ORNL von Hamos spectrometer. The labelled components are: (a) Target cell containing targets, X-ray limiting slits, and simple tungsten filament electron source, (b) Curved crystal mounted on a linear track, (c) Position-sensitive proportional counter mounted on a rotational adjustment carriage and linear track, and (d) ultra-high vacuum chamber.
- FIG. 3. Relative reflection/collection efficiency for sulfur K_{α} X-rays as a function of position along the length of the quartz($10\bar{1}1$) crystal ($2d = 6.687 \text{ \AA}$).
- FIG. 4. Silicon K X-rays from 2.6 MeV/u $\text{Si}^{12,13+}$ ions excited in passing through a $5 \mu\text{g}/\text{cm}^2$ carbon foil. The energies are in the laboratory frame, shifted upward from the projectile frame according to eq. 7 with the positive sign. Silicon Lyman-alpha transitions (α_1 and α_2 are separated by 1.8 eV) as well as several doubly-excited He-like Si lines are labelled.
- FIG. 5. Low energy satellites of Al K X-rays produced by 2 MeV/u sulfur ions on aluminum metal. Peaks are due to K L^n Radiative Electron Rearrangement (RER) and Radiative Auger Emission (RAE) as labelled and described in the text.

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Fig.3

Fig.4

Fig. 5

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