High-energy-resolution monochromator for ^{83}Kr nuclear resonant scattering

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We have built a high-energy-resolution monochromator for nuclear resonant scattering from the 9.4 keV nuclear transition of ^{83}Kr. The monochromator consists of two highly asymmetric silicon single crystals arranged in a dispersive geometry and produces an energy bandwidth of 2.3 meV. This monochromator has been successfully used for a study of nuclear forward scattering and nuclear resonant inelastic scattering from a ^{83}Kr sample that was solidified in a diamond anvil cell at 2.15 GPa and room temperature. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1445822]

I. INTRODUCTION

Nuclear resonant scattering (NRS) using synchrotron radiation (SR) has evolved greatly since it was first demonstrated by Gerdau *et al.* in 1985.[1] Up to now, most studies have concentrated on ^{57}Fe- and ^{119}Sn-containing materials and a few on ^{161}Dy, ^{151}Eu, ^{149}Sm.[2] Little attention has been paid to ^{83}Kr, one element with outstanding properties for NRS among the isotopes with a low-lying nuclear level. Johnson *et al.*[3] observed nuclear forward scattering (NFS) from a ^{83}Kr bulk film and a monolayer adsorbed on exfoliated graphite. Baron *et al.*[4] determined the wavelength of the nuclear resonance energy of ^{83}Kr by measuring the incoherent NRS from gaseous krypton. Recently research interest has been increasing on the rare-gas solids under high pressure and on the lattice dynamics of hydroquinone clathrates.[5,6] Such scientific interest motivated us to build a high-resolution monochromator (HRM) to study the lattice dynamics of ^{83}Kr-containing materials by using the unique nuclear resonant inelastic x-ray scattering (NRIXS).[7,8]

We have successfully built a HRM with an energy bandwidth of 2.3 meV at 9.4 keV radiation. In this article, we present the design and tests of the monochromator. Preliminary results of the phonon density-of-states measurement of a solid Kr sample under high pressure and at room temperature are also presented.

II. DESIGN OF THE MONOCHROMATOR

For NRIXS measurements, the resolution of the experiment is determined solely by the resolution of the HRM. A HRM with meV or sub-meV energy resolution is required to resolve structure in the density of states and to measure low-energy dynamical excitations. Our goal is to build a HRM with energy resolution around meV or even sub-meV and with good efficiency.

Toellner *et al.* obtained sub-meV energy resolution for both 14.4 keV (Ref. 9) (^{57}Fe resonance) and 23.9 keV (Ref. 10) (^{119}Sn resonance) x rays by adopting two high-order reflections from highly perfect silicon single crystals arranged in a dispersive (+,+) geometry. A HRM for 9.4 keV is more problematic, because the intrinsic Darwin width of silicon reflections becomes relatively large at lower photon energy. On one hand, it is easier to obtain wide angular acceptance of a HRM to match the emittance of the incoming x rays. On the other hand, it is difficult to get sub-meV-energy bandwidth at lower photon energies. For example, the Darwin width of the highest index reflection Si (7,3,3) at 9.4 keV is 16.3 μrad (Bragg angle $\theta_B = 83.5°$). Two symmetric Si (7,3,3) crystals in a (+,+) setup only produce an energy bandwidth of about 12.6 meV. At the 3ID undulator beamline of the Advanced Photon Source (APS), where this monochromator is designed to work, the vertical divergence of the x-ray beam from the undulator is about 15.8 μrad at 9.4 keV and the angular acceptance of a symmetric Si (7,3,3) already matches this source emittance. In order to narrow the energy bandwidth, the Si (7,3,3) crystals have to be cut asymmetrically.

For our HRM, we chose the asymmetry angle of $-83.0°$ and $+82.8°$ for the first and second crystal, corresponding to asymmetry factors of 0.04 and 18.5, respectively. Those Si (733) arranged in a (+,+) dispersive geometry are expected to produce an energy bandwidth of about 2.2 meV. Figure 1 shows the geometry of this monochromator. A highly perfect single crystal is needed to make such a high-resolution monochromator. The silicon single crystal was grown using the floating-zone method by TOPSIL Co. A high-resistivity crystal ingot (>50 kΩ cm) was chosen for our monochromator. The crystal surface was mechanically and chemically polished.

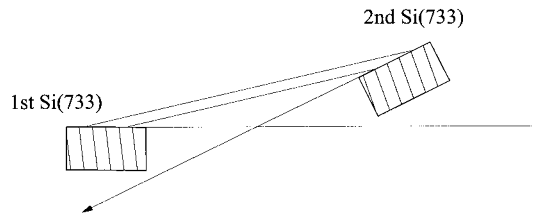

FIG. 1. Schematic of the high-energy-resolution monochromator for 9.4 keV x rays.

III. RESULTS AND DISCUSSION

The best way to characterize the performance of a HRM is to measure its response function with NFS, because the width of the nuclear resonance is negligible (on the order of several neV) compared with the meV width of the HRM. The nuclear resonance behaves like a delta function. The energy resolution of the HRM can be measured by recording the time-delayed NFS while tuning the HRM around the resonance.

In our experiment, a solid ^{83}Kr sample is used to characterize the monochromator. The sample was loaded cryogenically in a diamond anvil cell (DAC). We condensed commercial gaseous krypton (11.5% ^{83}Kr) by flowing the gas into a brass container sitting in a liquid nitrogen bath. The DAC was placed in the container; thus it was cooled by the liquid nitrogen through the brass wall. Most of the Kr liquified while the part close to the container solidified. When the DAC was filled with liquid Kr, the loading was applied to seal the sample in the gasket (made of Be) hole. After adequate pressure had been applied, the DAC was warmed to room temperature. According to the phase diagram, Kr remains in the solid state at room temperature when the pressure is above 0.85 GPa. The sample pressure was measured to be 2.15 GPa by the ruby fluorescence technique.

Characterization of the monochromator was done at the 3ID undulator beamline of the APS. The experimental setup is shown in Fig. 2. The monochromatic beam after the HRM transmitted through the solid Kr sample in the DAC. Along the beam, the sample thickness is about 60 μm. The NFS signal was collected by an avalanche photodiode detector (APD).

The result of the resolution measurement is shown in Fig. 3 along with a simulation using x-ray dynamical theory.[11] The energy resolution is determined by the full width at half maximum (FWHM) of the measured energy response function, which is 2.3 meV. The throughput of the

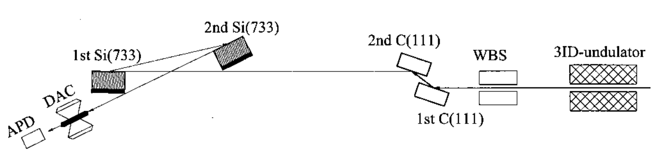

FIG. 2. Experimental setup to measure the energy response function of the monochromator. WBS: white beam slits; DAC: diamond anvil cell with solid Kr sample; APD: avalanche photodiode time-resolved detector.

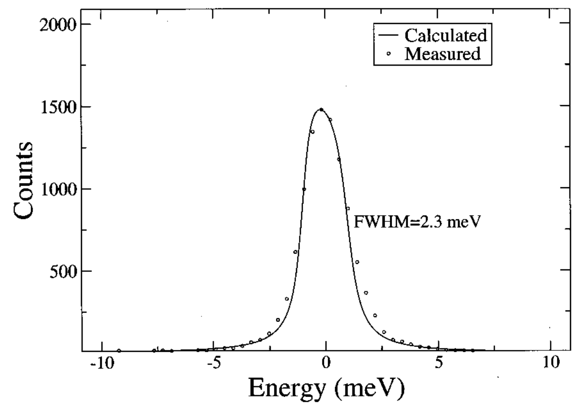

FIG. 3. Measured energy response function compared with a simulation. FWHM of the measured energy response function is about 2.3 meV.

monochromator is measured by placing a pin-photodiode before and after the HRM. The flux of the x-ray beam into the HRM was measured as 1.3×10^{13} photons per second per 100 mA storage ring current in an energy bandwidth of 0.57 eV, which corresponds to a spectral flux of 2.3×10^{13} phs/s/100 mA/eV. After the monochromator, we obtained 1.2×10^{10} phs/s/100 mA in the energy bandwidth of 2.3 meV, or a spectral flux of 5.2×10^{12} phs/s/100 mA/eV. The efficiency of this monochromator, the ratio of the outgoing and incoming spectral flux, is ~23% which is close to the theoretical estimation.

The monochromator was used to perform NRIXS on solid Kr under high pressure (2.19 GPa) and at room temperature. The HRM is tuned over an energy range of ±20 meV around the nuclear resonance of ^{83}Kr. Two APDs were placed close to the sample to detect the incoherent scattering signal. The photon counts are integrated over a time window of 20–150 ns after the synchrotron x-ray pulse. The total collecting time is 1600 s for each point. From the measured

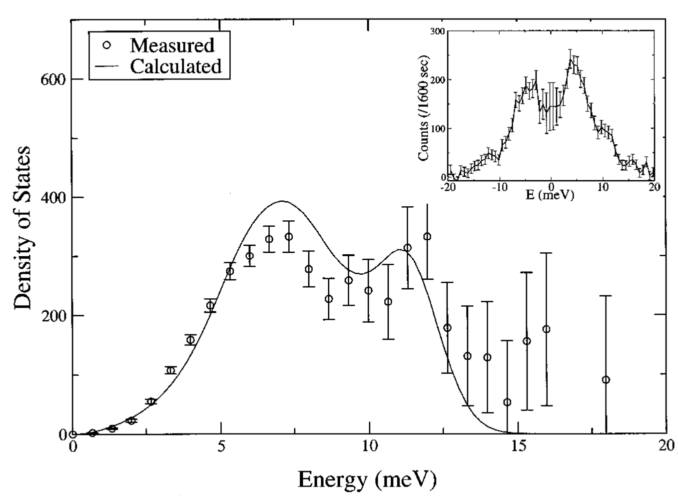

FIG. 4. Phonon DOS of solid Kr derived from the measured data of NRIXS (dots) and calculation by the density functional theory at a pressure of 2.9 GPa. The inset drawing shows the data after the elastic peak has been removed.

results, the photon density of states (DOS) of ^{83}Kr can be derived. Figure 4 shows the derived DOS of ^{83}Kr from the measured NRIXS data. The inset in Fig. 4 shows the normalized data after the elastic peak has removed. The derived phonon DOS of ^{83}Kr is in relatively good agreement with a calculation based on the density functional theory.[12] From such measurements, certain dynamical and thermodynamical quantities can be obtained. For Kr under these conditions, we derive a Lamb–Mossbauer factor of 0.47(9) and a mean kinetic energy per Kr atom of 36.9(15) meV. The detailed analysis and discussion of this spectrum compared with the theory will be presented elsewhere.

IV. CONCLUSION

We have designed and constructed a meV monochromator for ^{83}Kr nuclear resonance. The monochromator was characterized by NFS, and an energy resolution of 2.3 meV was achieved. The monochromator was successfully used to perform NRIXS on solid ^{83}Kr under high pressure and at room temperature.

ACKNOWLEDGMENT

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