Dielectric function spectra at 40 K and critical-point energies for $CuIn_{0.7}Ga_{0.3}Se_2$

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We report ellipsometrically determined dielectric function ε spectra for $CuIn_{0.7}Ga_{0.3}Se_2$ thin film at 40 and 300 K. The data exhibit numerous spectral features associated with interband critical points (CPs) in the spectral range from 0.74 to 6.43 eV. The second-energy-derivatives of ε further reveal a total of twelve above-bandgap CP features, whose energies are obtained accurately by a standard lineshape analysis. The ε spectra determined by ellipsometry show a good agreement with the results of full-potential linearized augmented plane wave calculations. Probable electronic origins of the CP features observed are discussed.
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$CuIn_{1-x}Ga_xSe_2$ is so far the most promising thin-film photovoltaic (PV) material and its best cell efficiency has been recorded as high as 20.3%.[1] The production capacity of $CuIn_{1-x}Ga_xSe_2$ solar cells was 426 MW in 2010 and is now approaching gigawatt-scale.[2] For design and optimization of high-efficiency PV device structures, accurate knowledge of the optical response for absorber materials is of great importance.[3] In the course of exploring $CuIn_{1-x}Ga_xSe_2$ for PV device applications, its optical properties were well characterized.[4,5]

Spectroscopic ellipsometry (SE)[6] is known to be a highly suitable method of determining a material's optical functions such as complex dielectric function $\varepsilon = \varepsilon_1 + i\varepsilon_2$ and complex refractive index $N = n + ik$ over a wide spectral range. Therefore, SE has been widely used to characterize various semiconductors.[7] For $CuIn_{1-x}Ga_xSe_2$, SE has been employed to study the composition-dependence of ε and interband critical-point (CP) energies,[8–10] effects of Cu-deficiency on the optical properties,[11] and thin-film growth mechanisms.[12]

The optical information of materials at low temperature, where the thermal broadening of optical transitions is reduced and individual optical structures are better resolved, plays an important role in understanding the material's electronic structure. Hence, the ε spectra for many elemental[13,14] and compound semiconductors[15–17] have been determined by SE at low temperature. However, low-temperature SE study of $CuIn_{1-x}Ga_xSe_2$ is rare despite the availability of rich information on its room-temperature optical properties.

Here, we apply SE to determine the ε spectra of $CuIn_{0.7}Ga_{0.3}Se_2$ at 40 K, a composition for high-performance $CuIn_{1-x}Ga_xSe_2$ PV devices. The SE data are in a good agreement with the ε spectra of $CuIn_{0.5}Ga_{0.5}Se_2$ calculated by the full-potential linearized augmented plane wave (FPLAPW) method using the generalized gradient approximation (GGA) plus an onsite Coulomb interaction U of the Cu d states.[4] The above-bandgap CP energies are obtained from a standard lineshape analysis of SE data. We discuss the probable electronic origins of the observed CP features.

A polycrystalline $CuIn_{0.7}Ga_{0.3}Se_2$ thin film was grown at 600 °C by a single-stage thermal co-evaporation method, where the fluxes of Cu, In, Ga, and Se were kept at constant during the deposition and thus the compositional homogeneity is greatly improved along the growth direction.[18] Ga and In fluxes were maintained at the Ga/(Ga+In) ratio of 0.3, and the atomic Cu ratio Cu/(Ga+In) was approximately 0.87. Soda-lime glass coated with an approximately 1-μm-thick molybdenum (Mo) film was used as the substrate. The nominal thickness of $CuIn_{0.7}Ga_{0.3}Se_2$ thin film was 1.5 μm. Chemical composition of the grown film was determined by X-ray fluorescence.

SE data were collected from 0.74 to 6.43 eV using a dual-rotating-compensators type system (J.A. Woollam Inc., RC-2 model) equipped with a variable-temperature cryostat. The angle of incidence is 68°. We reduced the microscopic roughness of $CuIn_{0.7}Ga_{0.3}Se_2$ film by chemical-mechanical polishing of the surface using a colloidal silica suspension with 0.02-μm particles.

The ε spectra for $CuIn_{0.7}Ga_{0.3}Se_2$ are obtained by the multilayer analysis of the SE data.[19] The multilayer model consists of the ambient, a surface roughness layer, the $CuIn_{0.7}Ga_{0.3}Se_2$ layer, and the Mo layer. The glass substrate is not included in the model because its presence is completely obscured by the metallic Mo layer in the spectral range used for this study. We estimate the surface-roughness layer to be ~2.2 nm thick, and modeled its response as a Bruggeman effective-medium-approximation[20] 50–50 mixture of the $CuIn_{0.7}Ga_{0.3}Se_2$ layer and void. The ε spectra for $CuIn_{0.7}Ga_{0.3}Se_2$ layer were reconstructed by the B-spline formulation.[21]

Real (ε_1) and imaginary (ε_2) parts of the modeled ε spectra for $CuIn_{0.7}Ga_{0.3}Se_2$ taken at 40 and 300 K are given in

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