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Small-Angle-Scattering Evidence of Voids in Hydrogenated Amorphous Silicon

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The observation of extensive small-angle x-ray scattering from hydrogenated *a*-Si indicates that the void distribution in this material is very similar to that for evaporated *a*-Si. Thus the improved electrical properties of the hydrogenated material are not due to this material having a homogeneous structure.

The doping of crystalline semiconductors with substitutional impurities for the purpose of controlling electronic properties is a most important feature of modern solid-state electronics. Prior to 1969, attempts to control the properties of amorphous tetrahedrally coordinated semiconductors in a similar manner were unsuccessful. The main reason for this failure was the high level of the localized-state density found in the mobility gap of evaporated or sputtered *a*-Si (Spear¹) and *a*-Ge. These states were thought to be related to microvoids and point defects.

It was demonstrated² in 1969 that *a*-Si prepared by the glow-discharge decomposition of silane with about 200 ppm of phosphine could lead to a hundred-fold increase in specimen conductivities. Subsequent investigations of electrical properties,^{3,4} optical properties,^{5,6} and the distribution of localized states^{1,7} have been undertaken on specimens of *a*-Si prepared from the gas-phase decomposition of silane in a rf glow discharge. These studies indicate that the gap-state densities are remarkably low in such specimens when deposited at relatively high substrate temperature (500–600 °K). Samples prepared in such a manner may contain from 18 to 50 at.% hydrogen,⁸ and are, thus, called hydrogenated *a*-Si.

This paper reports a study of the small-angle x-ray scattering (SAXS) from a 10- μ m sample of *a*-Si fabricated by the glow-discharge decomposition of silane onto a 20- μ m Al substrate. It contains 15 at.% hydrogen. The objective was to determine whether a sample so prepared was

free from microvoids that are normally associated with a high level of localized-state density. The observed SAXS, however, is very similar to that observed for evaporated *a*-Si.⁹ This indicates that samples produced from the glow discharge of silane contain a distribution of voids or holes containing an undetermined amount of hydrogen (x rays are only weakly scattered by the hydrogen relative to the Si atoms) similar to that found in evaporated samples. Thus the improved electrical properties of the hydrogenated *a*-Si are not the result of the material being free from microregions of low electron density.

The scattering of x rays is sensitive to the fluctuations in the electron density in the sample. The SAXS is determined by such fluctuations or inhomogeneities in regions measuring about 6 Å or more in size. These inhomogeneities may be due to voids or phase-separated regions dispersed in an approximately homogeneous medium. In the approximation that the inhomogeneities are widely dispersed and the approximate law of Guinier holds, the scattering is proportional to¹⁰

$$I_c(s) \propto \sum_{R_0} M(R_0) R_0^3 \exp(-R_0^2 s^2 / 3), \quad (1)$$

where $I_c(s)$ is the calculated intensity, $M(R_0)$ is the volume fraction of the inhomogeneities with radius of gyration R_0 , and s is the scattering vector ($s = 4\pi \sin\theta/\lambda$). For a spherical particle $R_0^2 = 0.6R^2$, where R is the radius of a sphere. An instrumental resolution function, $P(s)$, may be defined, the convolution of which with $I(s)$

yields a representation of the observed intensity

$$I_0(s) \approx I_c'(s) = \int I_c(s) P(s - s_0) ds_0. \quad (2)$$

A least-squares procedure¹¹ may be employed to obtain $M(R_0)$ by fitting $I_c'(s)$ to $I_c(s)$.

The experiments were carried out on a Picker FACS-1 single-crystal diffractometer using Cu $K\alpha$ radiation with a diffracted beam monochromator, a scintillation detector, and a pulse-height analyzer. Data were collected in the range $s = 0.05$ ($2\theta = 0.70^\circ$) to $s = 2.4$ ($2\theta = 34.25^\circ$) in increments of 0.05- s units. The x-ray-source size, incident- and diffracted-beam collimators, and detector-slit system were chosen so that the resolution function (the function that would be a δ function for perfect resolution) has a full width at half height of 0.015- s units in the range of data collection. In order to minimize the background scattering, the sample was placed in a He atmosphere. Diffraction data were measured for samples of α -Si on Al in He, of Al substrate in He, and of He only. Absorption measurements were then taken for the α -Si on Al in He and Al in He samples at $2\theta = 0^\circ$ to determine the attenuation of the characteristic radiation by the samples. In this way the appropriate amount of the He-only pattern was subtracted from the Al-in-He pattern to yield the Al-only pattern illustrated in Fig. 1. Likewise, an appropriate fraction of the Al-in-He pattern was subtracted to yield the α -Si-only pattern also shown in Fig. 1. A Guinier plot of the

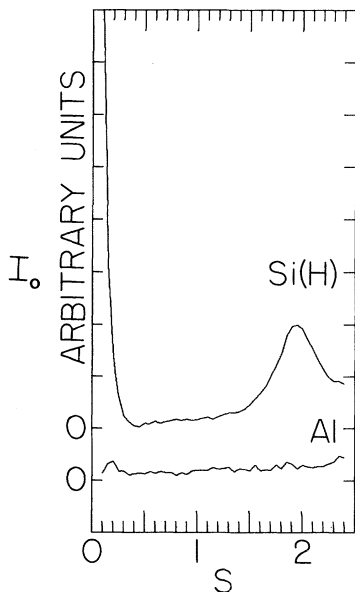


FIG. 1. The SAXS of hydrogenated α -Si and the Al substrate on which it was deposited.

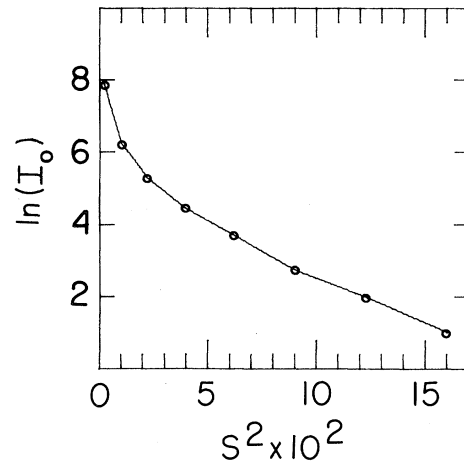


FIG. 2. A Guinier plot (Ref. 10) of the SAXS of hydrogenated α -Si.

intensity is given in Fig. 2.

Table I lists the observed and calculated intensities corresponding to the derived $M(R_0)$. The intensity is given in electron units and was scaled with the large-angle data to correspond to a scattering volume for a single silicon atom (20.02 \AA^3). The fractional void volume, X_v , was obtained¹⁰ as

$$X_v = \int s^2 I(s) ds / (2\pi^2 V_{\text{at}} \rho^2) = 0.00937, \quad (3)$$

where V_{at} is the atomic volume and ρ is the electron density of the silicon phase. The small-angle x-ray scattering for the sample starts to rise rapidly at about $s = 0.3$ just as it does for the evaporated silicon sample reported by Shevchik and Paul.⁹ The $M(R_0)$ function illustrated in Fig. 3 indicates a much sharper distribution of void

TABLE I. Observed and calculated intensities for hydrogenated α -Si. I_0 is the experimental intensity. I_c' is the calculated intensity that has been broadened with the instrumental resolution function. I_c is the unbroadened intensity calculated for the void volume distribution illustrated in Fig. 3.

s	I_0	I_c'	I_c
0.05	2664.00	2664.30	3707.64
0.10	510.00	509.30	709.95
0.15	197.98	198.88	284.96
0.20	85.34	86.02	123.22
0.25	39.59	36.41	52.10
0.30	15.36	15.69	22.46
0.35	7.43	7.07	10.13
0.40	2.77	3.35	4.81

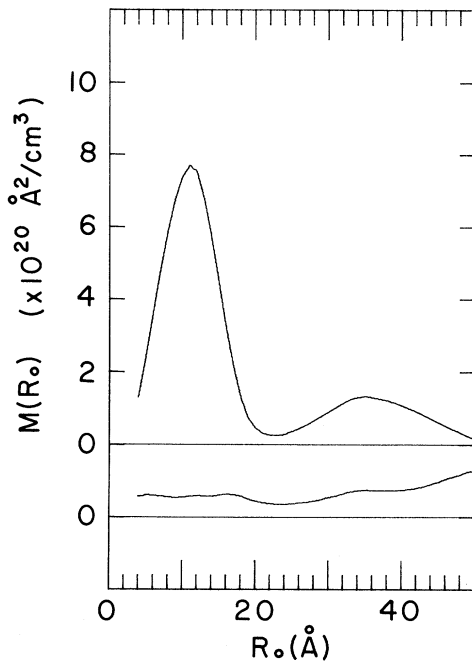


FIG. 3. Fractional void volume distribution for hydrogenated a -Si (top) and the associated standard deviation curve (bottom).

sizes than does the corresponding figure in their paper.⁹ However, greatly different experimental arrangements were employed and different methods for obtaining $M(R_0)$ were used. It would be necessary to collect data under the same conditions for both types of samples in order to draw firm conclusions on this point. It may be stated, however, that this hydrogenated a -Si sample produces a large amount of small-angle scatter-

ing very similar to that observed for evaporated a -Si. Therefore, the improved electrical properties are not due to the elimination of electron-density inhomogeneities in the hydrogenated sample.

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Critical Superflow in a Random Network

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The problem of superflow through a random bond network is formulated. The flow is taken to be limited by the critical velocity. This problem is solved for a two-dimensional system; a first approximation for attacking the problem in three-dimensional system is described. Possible applications are to superfluid flow through Grafoil.

The recent success of the Kosterlitz-Thouless theory¹⁻³ in providing an explanation of superfluid flow experiments on Mylar and other substrates has focused attention on the nature of two-dimen-

sional systems (system = helium + substrate) and their flow properties. Certainly, the most extensively studied two-dimensional system, the helium-Grafoil system,⁴ is an excellent candidate for