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Local structure reconstruction in hydrogenated amorphous silicon from angular correlation and synchrotron diffraction studies

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Abstract

Hydrogenated amorphous silicon (a-Si:H) is a widely used thin film semiconductor material which is still incompletely understood. It is generally assumed to form a continuous random network, with a high concentration of coordination defects (dangling bonds), which are hydrogen terminated. Neither the exact nature of these sites nor the degree of medium range order has been fully determined. In this paper, we present the first results for the local structure, from a combined study using angular correlation of positron annihilation radiation (ACAR) and synchrotron radiation diffraction. Reciprocal space information is obtained directly, for the mesoscale structure and the local defect structure, from the orientation dependent diffraction and 2D-ACAR patterns, respectively. Furthermore, inversion of both patterns yields a comparison of real space information through maps of the silicon–silicon pair correlation function and the electron–positron autocorrelation function $B^{2\gamma}(r)$. From this information, it is possible to identify the dominant structural defect as a vacancy-size dangling bond cluster, around which the network strain is fully relaxed.

Keywords: Amorphous silicon; ACAR; Synchrotron; Autocorrelation; Pair correlation; Continuous random network

1. Introduction

Hydrogenated amorphous silicon (a-Si:H) is a widely used important thin film semiconductor with applications in photovoltaics [1] and transistor arrays

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for various applications including pixel switching [2]. All structural models of a-Si:H are formed from a continuous random network (CRN) built from distorted Si–Si_4 tetrahedra [3]. Such structures are highly strained with both bond lengths and bond angles varying by a few percent from crystalline silicon. This microscopic strain is relaxed by dangling bond defects which, in the real material, are mainly hydrogen terminated. In-service degradation effects, such as light induced metastability, known as the Staebler–Wronski effect [4], are known to be related to the release of hydrogen from these defects leaving localised deep levels in the band gap [5–9].

Positron annihilation, amongst other methods, has been applied by several groups to the study of structural defects in a-Si:H. However, the geometry and size of the dangling bond defects remain unclear. Besides the smaller dangling bond complexes, depending on the growth method and conditions, other defects are observed. Early studies, on thick layers found both a long lifetime and narrow momentum peak corresponding to the annihilation of positronium in large voids [10,11]. Later work on plasma enhanced chemical vapour deposition (PECVD) grown layers, of electronic quality but with a high hydrogen concentration, found evidence of hydrogen filled nanovoids [12,13]. The material which best approximates a CRN, with a low hydrogen concentration is produced by hot-wire chemical vapour deposition (HWCVD) [14]. In such material, only a single positron annihilation state, with a lifetime in the range 320–390 ps is observed [15–18]. This is a weakly localised state in which the positron exhibits hopping diffusion [19]. Our own calculations suggest that the associated defect is a small dangling bond complex, and that the positron binding energy is strongly reduced by hydrogen decoration of the dangling bonds.

Nevertheless, despite the presence of dangling bond and other defects, a-Si:H layers grown by HWCVD still possess a macroscopic residual strain, which can be observed directly by both curvature [20] and diffraction [21] methods. This strain is generally compressive in the plane of the layer, and, as with all thin films, results from a combination of thermal mismatch between the substrate and layer, and an intrinsic strain determined by the growth process [22]. At first sight, it could be expected that this strain should also be manifested in the average geometry of the dangling bond complexes.

The aim of this work is to compare the local environment sampled by the positron with the mesoscopic structure of the a-Si:H CRN, as determined by synchrotron radiation diffraction. Of particular interest are the spatial extent and average anisotropy of the correlated electron–positron state. These are investigated using two-dimensional angular correlation of positron annihilation radiation (ACAR), at the POSH source in Delft [23]. Diffraction and ACAR data effectively yield reciprocal space maps of the mesoscopic structure and the local electronic structure, respectively. Neither of these maps is a perfect representation, however, in that both are averages of an ensemble of different specific environments, and that both are modified by the nature of the experiment, i.e. the atomic form factor and enhancement due to electron–positron correlation, respectively. In particular, synchrotron diffraction is insensitive to hydrogen, and only the silicon network is observed. However, under the sensible assumption that these effects, at worst, reflect the anisotropy of the region under investigation, a comparison of the wavevector dependent scattering intensity $S(q_x, q_y)$ with the ACAR distribution $N(p_x, p_y)$ should indicate the effect of structural anisotropy on the defect structure.

Both data sets can be Fourier inverted to give real space maps of the mesoscopic and defect structure. The transform of $S(q_x, q_y)$ yields the Si–Si pair correlation function $g(x, y)$, which is equivalent to the radial distribution function [24] in isotropic homogenous media. This directly yields the directional dependence of the nearest and next nearest neighbour separations, and hence gives an indication of short and medium range order. The Fourier transform of the ACAR distribution is the electron–positron autocorrelation function $B^{2\gamma}(x, y, 0)$ [25] which indicates the spatial overlap of electron and positron wave functions in the medium. In a covalent network, this indicates the spatial extent of the localised positron states. Also for the average of localised states, any systematic anisotropy should appear in this representation.