A study of electron field emission as a function of film thickness from amorphous carbon films

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The electron field-emission properties of hydrogenated amorphous carbon and nitrogenated tetrahedral amorphous carbon thin films are examined by measuring the field-emission current as a function of the applied macroscopic electric field. The experimental results indicate the existence of an optimum film thickness for low-threshold electron field emission. The predictions of various emission models are compared to the experimental results. © *1998 American Institute of Physics.* [S0003-6951(98)03351-8]

Currently, there is much interest in electron field emission as a potential flat panel display (FPD) technology.[1–3] Thin amorphous carbon films are a possible cold cathode material[4–6] for use in FPDs. For this to come to fruition the lifetime and emission site density of these materials needs to be maximized for low operating electric fields to levels at which a display device may be technically and commercially viable. One of the most important experimental parameters examined in the research conducted is the emission threshold electric field of a thin film. This threshold field is defined here as the applied macroscopic electric field at which a close to steady-state emission current of 1 nA is measured for a conditioned film. In this letter we report on the dependence of the threshold field of hydrogenated amorphous carbon (*a*-C:H) and nitrogenated tetrahedral amorphous carbon (*ta*-C:N) thin films on film thickness. We examine the experimental results in terms of various emission models proposed for thin amorphous semiconductors.

a-C:H films were deposited using a radio-frequency (RF) capacitively coupled Plasmatech DP800 plasma-enhanced chemical-vapor deposition (PECVD) system configured with a water-cooled, earthed substrate table. The substrate material was phosphorus-doped, *n*-type, 1–2 Ω cm, ⟨100⟩ silicon. A He plasma preclean was performed at ambient temperature on the substrates with 75 sccm helium flow at a pressure and RF power of 200 mTorr and 200 W, respectively. The depositions were carried out using CH_4:He feed gases with flow rates of 30:75 sccm at ambient temperature at a pressure and RF power of 200 mTorr and 200 W. A set of samples with different film thicknesses were grown by varying the deposition time for each sample. The thickness of the film on each sample was then measured using ellipsometry, to a precision of ±3 nm. These films were then subject to field-emission testing.

The field-emission properties of the *a*-C:H films were investigated using a ''sphere-to-plane'' electrode configuration with a 5 mm diam stainless-steel ball anode, 40 μm from the film, in series with a 100 μH inductor in a vacuum better than 5×10^{-6} mbar. Samples were subject to a forward bias until an emission current of 500 nA was detected or a maximum macroscopic field of 90 V/μm was reached. The voltage cycling and current/voltage measurements were computer controlled to ensure repeatability of the testing procedure. A relatively high applied field was required for emission to be initiated for all the *a*-C:H films in the first cycle and a considerable hysterisis in the current/applied field characteristics was observed, as shown in Fig. 1. For successive voltage cycles, this hysterisis diminished with each successive cycle—a conditioning process. Typically, this hysterisis was no longer present after 3 or 4 voltage cycles. Reverse bias tests were also performed after each set of measurements on every sample to check for short circuits and to validate the results. Testing was performed on five different areas of each sample to gauge the repeatability of the results. The conditioned electric field for these devices was measured and a standard deviation for the threshold electric field was no more than 5 V/μm. Films deposited under identical conditions on subsequent runs resulted in threshold fields that were in agreement with the error bars shown in Fig. 2.

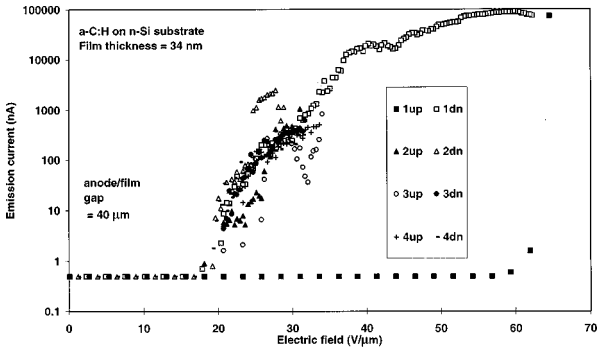

FIG. 1. A typical emission current vs applied electric-field characteristic for a 34 nm thick *a*-C:H film. The applied voltage was cycled four times, until the threshold electric field approached a limiting value. The labels 1up, 1dn, 2up,..., refer to the voltage cycle (first, second,...) and to whether the applied voltage was increasing (up) or decreasing (dn).

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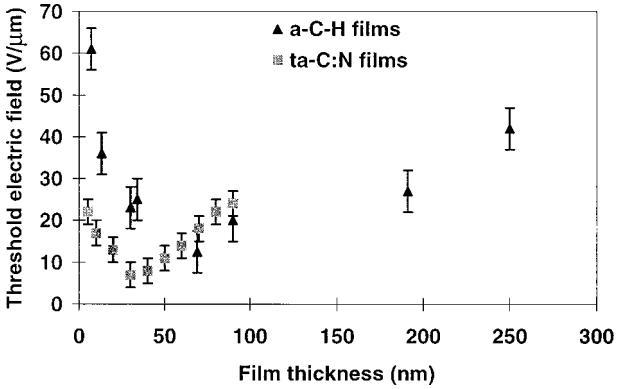

FIG. 2. The threshold electric field (the applied electric field for which the conditioned films produce an emission current of 1 nA) as a function of film thickness for the *a*-C:H (▲) and *ta*-C:N films (■).

After field-emission testing, the surfaces of the *a*-C:H films were imaged using a Cambridge Instruments Stereoscan 250 scanning electron microscope (SEM). No morphological changes were apparent on the micron scale and no evidence of any difference in surface roughness between the different film thicknesses was detected. A set of similarly prepared samples were examined in an atomic force microscope (AFM) and exhibited rms surface roughness values of less than 5 Å with no discernible relationship between the thickness and roughness. A similar study on nitrogen-doped tetrahedral amorphous carbon (*ta*-C:N) films was carried out, independent of the *a*-C:H work, by Cheah and Shi. These films were deposited using a filtered cathodic vacuum arc (FCVA) system at an energy of 100 eV on n^{++}-Si substrates. The nitrogen was obtained from a 100 eV ion source with a 20 mA beam current resulting in a bulk nitrogen content of 30 at. %. The testing regime employed and hysteresis effects observed were similar to that described for the *a*-C:H films.

The data shown in Fig. 2 show a strong dependence of the conditioned threshold field on film thickness. More specifically, the data for both types of films show that there is a minimum turning point in the threshold field with film thickness. For the *a*-C:H films this minimum occurs around 60 nm and for the *ta*-C:N films the minimum occurs at around 30 nm. This indicates that there is a window for optimizing the electron field-emission properties of these films. It is interesting that this thickness dependence phenomenon has been observed for two different types of amorphous carbon (*a*-C) with vastly differing microstructural properties, deposited and tested independent of each other. In order to attempt to explain these experimental results we examine the different models that have been proposed to describe electron field emission from thin films.

The classical explanation for electron field emission from a surface is Fowler–Nordheim (FN) tunneling,[7] which is generally aided by high-field enhancement factors (β). Here, electrons tunnel from the Fermi level at the film surface through an approximately triangular potential barrier into vacuum. This model can be described by the simplified FN equation.[5]

$$I = \frac{B\beta^2 V^2 A}{\Phi} \exp\left[-\frac{C\Phi^{3/2}}{\beta V}\right], \tag{1}$$

where, I is the emission current (amps), β is a geometrical factor (cm^{-1}), V is the applied potential (volts), A is the emission area (cm^2), ϕ is the work function (eV), and B and C are numerical constants. This emission mechanism applied to a nominally flat cathode requires surface features, or protrusions, to produce a large enough field enhancement (β) to allow the local electric field to be great enough to permit the tunneling. For this model to predict the observed trend either ϕ or β, or both, needs to change significantly and systematically with film thickness. There is little reason to expect, or evidence to support, the possibility that the work function (ϕ) would depend significantly on film thickness. As stated previously, the SEM and AFM images offered no evidence in support of the existence of surface morphological changes with film thickness and, hence, changes in β cannot account for the observed threshold field dependence on film thickness. Although it may be, as previously reported,[8] that there are nanoprotrusions on the film surface, it is difficult to see how these would vary in systematic manner to explain the particular threshold field/film thickness relationship observed. Fowler–Nordheim calculations performed on the *a*-C:H data, assuming a work function of 2 eV for these films, required field enhancement factors greater than 1000 to satisfy the FN equation. Atomistic bond reordering on the surface of the emitting films giving rise to high β factors and possible changes in the threshold field could not be easily examined. Consequently, FN theory does not appear to successfully explain the experimental results observed in Fig. 2.

There are other possible emission mechanisms, for example, Krauss *et al.*[9] suggest that the emission could be due to quantum effects from sp^2 cluster edges. It is not obvious how this model could account for the results reported in this study as it does not appear to be thickness dependent. A change in the sp^2/sp^3 content of the film as a function of thickness could, in theory, affect the emission properties in the Krauss model. However, for our films, measured parameters such as refractive index and optical band gap do not show any significant variation with film thickness, and therefore, it is unlikely that the sp^2/sp^3 varies as a function of thickness in our films. Robertson[10] proposed an emission mechanism for diamond-like carbon films based on nonuniform hydrogen termination at the film surface. In this work, emission is postulated as occurring mainly from nanometer-sized surface regions unterminated by hydrogen. However, we do not expect the hydrogen termination at the surface of our films to be thickness dependent, and therefore, do not believe the Robertson model to be applicable to this work.

A space-charge-induced bandbending interlayer model was also proposed for field emission from *a*-C films.[4,11] A similar model was originally proposed by Latham and Bayliss to explain emission from metal/insulator structures.[12] An important feature that distinguishes the space-charge-induced bandbending model for semiconductors from that proposed by Latham is that these films, unlike insulators, can sustain a thickness-dependent electric field. In the proposed model it is the heterojunction and bandbending within the film that are important.[11] In our model, the bandbending occurs as a result of carrier depletion across the whole thickness of the *a*-C film, which gives rise to a maximum internal electric field close to the heterojunction. The band diagrams under com-

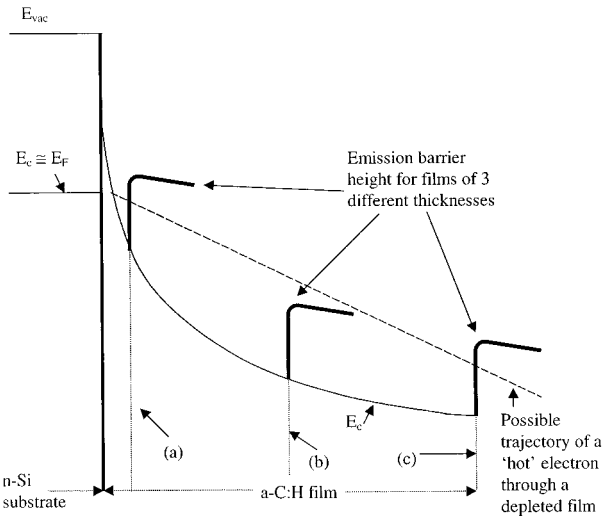

FIG. 3. Band diagram illustrating the case of hot electron transport through a depleted *a*-C film under the influence of a given applied electric field. The valence bands have been omitted to minimize the complexity of the diagram. The emission barrier is shown for three different film thicknesses (cases a, b, and c) along with a possible path taken by the hot electrons. The diagram demonstrates that emission will not occur for the thinnest and thickest films (cases a and c), but it may for the intermediate film thickness (case b).

parable situations have been simulated by Lerner *et al.*,[13,14] who have once more shown the internal field to be greater than the applied external field. The phenomenon of the local electric fields in the film being much greater than the applied electric field was qualitatively described by Amaratunga and Silva[4] and also shown experimentally by Schlesser *et al.*[15] In the case of the work reported by Schlesser *et al.*,[15] they showed the internal field within the material as approximately a factor of 10 greater than the applied external field. In the proposed model, Fig. 3, electrons tunnel from the conduction band of the *n*-doped silicon substrate into the highly curved conduction band of the *a*-C:H film. Conduction-band emission has also been proposed by Schlesser *et al.*[15] and Choi *et al.*[16] These electrons then become ''hot'' while traversing the film, as shown by Fitting *et al.*,[17] and possibly, depending on the film thickness, retain enough energy to overcome the surface barrier of the *a*-C:H films (2–3 eV) to vacuum. It should also be noted that the only value available for the effective tunneling mass of electrons in *a*-C:H is approximately $0.07\,m_e$.[18] This may give rise to energy relaxation lengths (which are typically several mean-free paths in magnitude) greater than those usually associated with amorphous materials.

A possible trajectory for hot electrons generated by the presence of the highly curved local electric fields in the *a*-C:H is shown in Fig. 3. The rate of electron energy loss to the lattice with thickness is taken as a constant to a first approximation in Fig. 3. The schematic path for electrons indicated by the dashed line is shown to illustrate that the emission from these films could occur as a function of film thickness. It should be noted that despite the loss of energy to the lattice with thickness, the electrons gain energy with respect to the conduction band at low thicknesses. It highlights the fact that there possibly is an optimum thickness for field-emission threshold fields (case b) and that if the film thickness is too large (case c), the *a*-C:H films would not be fully depleted, and therefore, the energy loss of the hot electrons (relative to the conduction band) would be large and thus prevent it from escaping from the surface. If the film thickness is too thin (case a), electrons that are emitted from the heterojunction (either thermally or by tunneling) will not gain enough energy relative to the conduction band for them to be able to surmount the emission barrier to vacuum. The variation of the threshold field discussed qualitatively as a function of thickness is observed in the experimental data presented for two sets of differing carbon films shown in Fig. 2. This gives us confidence that the electron emission from *a*-C may be explained using a space-charge-induced band-bending ''interlayer'' model, in which the real cathode is the *n*-Si substrate with the *a*-C film acting as an ''interlayer.''

In conclusion, we have presented field-emission data from *a*-C:H and *ta*-C:N with a distinctive film thickness/threshold field dependence. We have examined our data with respect to different field-emission models, and while more characterization is needed for a definitive model, we believe a space-charge-induced ''interlayer'' model can best explain these results.

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