Exploring the Limits of Sensitivity for Strain Gauges of Graphene and Hexagonal Boron Nitride Decorated with Metallic Nanoislands

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Figure S1: TEM image of Gr/Pd. Transmission electron microscopy (TEM) image of palladium of a nominal thickness of 6 nm on top of single layer graphene.



Figure S2: SEM image of 2D/Au. Scanning electron microscopy (SEM) images of different nominal thicknesses of gold supported by either graphene (Gr) or hexagonal boron nitride (hBN).



Figure S3: TEM image of 2D/Pd. Transmission electron microscopy (TEM) images of different nominal thicknesses of palladium supported by either graphene (Gr) or hexagonal boron nitride (hBN).



Figure S4: Piezoresistive response of Gr/M and hBN/Pd samples under 1 ppm strain. Plots compare signal to noise of a disconnected metal film on graphene to a percolated subcontiguous film on graphene and hBN.

Strain-Induced Changes in the 2D Density of States (DOS)

While a thorough exploration of the theoretical origins of the piezoresistance seen in these composite materials is beyond the scope of this work, it is worth briefly discussing the mechanisms we believe are at play, which arise from the mismatch in quantization of energy levels in different regions of the metallic network. The low nominal thicknesses of the metallic films are well within the regime where metals show quantization of energy levels,¹ and, for illustrative purposes, we assume individual nanoislands show behavior reminiscent of thin films. We believe this to be a reasonable assumption given that their lateral dimensions are generally significantly larger than their nominal thicknesses. We can use the prototypic 1D quantum well as a starting point to guide the discussion, for which the spacing between allowed energy levels demonstrates a $1/a^2$ dependence, where a is the width of the well. This model finds its real-world analogue in uniform thin films, where each of these quantized energy levels (confined along the z-direction) correspond to a subband edge, which (if the electrons population near the subband edge can be approximated as a Fermi gas) displays parabolic dispersion along the in-plane (xand y-) directions. Correspondingly, this results in a DOS(E) which is a sum of step functions, where each step occurs at the energy pertaining to the next quantized energy level.^{2,3} It may start becoming clear how, in a percolated network primarily composed of thin-film-like regions of variable thickness, there are many scattering opportunities as the charge is transferred from one region to the next.

Consider two thin, semi-infinite films which are of different thicknesses and lie in the same plane, such that they share an edge where charge is transferred between the two films as it propagates along the plane (in contrast to transverse to it, as is usually considered in typical

heterostructures). The steps in the DOS(E) are shifted to higher energies for the thinner film than the thicker one. If charge is to transfer from the thicker region to the thinner one, there will be some energy ranges where there are more occupied states in the thicker region than are available in the thinner one. This will be true even if we ignore the occupancy of states in the thinner region, because the steps in the DOS(E) are shifted to higher energies in the thinner region as compared to the thicker one. So, if electron energies are conserved, this will result in backscattering at the interface, as there is a fundamental deficiency of available states to accommodate those incoming electrons. This pertains to the real systems at hand in that, under some applied tensile strain, it can be reasonably expected that stress is concentrated in the thinner regions of the percolated network, further decreasing their critical dimension and forcing the subband edges to higher energies, exacerbating the effect described above and further frustrating charge transfer from adjacent thicker regions. The reasoning here bears resemblance to the theory of quantized conductance, developed by Landauer et al [cite].⁴⁻⁶ According to this theory, discrete conductance channels originate from the subbands of 1D conductors, and discrete steps in the conductance correspond to the populating of these subbands. We suggest analogous channels are at play in our percolated networks, where access to these channels is regulated by an applied strain instead of applied bias.

We have arrived at this description from an extremely simple thought experiment which only considers the variations in the DOS(E) of dissimilar thin films.Considering added complications such as additional conserved quantities (e.g. *k*-vector components), the necked regions between nano-islands (which will behave more like 1D conductors than thin-films), increased surface area/scattering centers, etc. only adds additional mechanisms by which scattering would be promoted as tensile stress is applied. Thus, we postulate the decrease of the critical dimensions of dissimilar regions in the percolated network under an applied tensile stress as a likely mechanism driving the overall increase in electrical resistance of our systems.

References

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