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# Interface and contact structures for nanoelectronic devices using assemblies of metallic nanoclusters, conjugated organic molecules and chemically stable semiconductor layers

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Self-assembly ('building') approaches can provide well-controlled structures and assemblies at the nanometer scale, but typically do not provide the specific structures or functionalities required for robust nanoelectronic circuits. One approach to realize high-density nanoelectronic circuits is to combine self-assembly techniques with more conventional semiconductor device and circuit approaches ('chiseling') in order to provide suitable functionality and arbitrary circuit functions. An interesting challenge is to find approaches where these techniques can be combined to realize suitable device structures. This paper describes recent work which combines self-assembly techniques involving metal nanoclusters and conjugated organic molecules with semiconductor interface and device structures to form structures of interest for nanoelectronics. One key requirement for this approach is the availability of a chemically stable semiconductor surface layer, which can provide a low-resistance interface between the metallic nanostructure and the semiconductor device layers following room-temperature, *ex situ* processing. As an illustration of the structures which can be realized, we describe a nanometer-scale ohmic contact to n-type GaAs which utilizes low-temperature-grown GaAs as the chemically stable interface layer. Contact structures have been realized using both isolated (sparse) clusters and using close-packed arrays of clusters on the surface. The low-resistance contacts between the nanoclusters and the semiconductor device layers indicates that relatively low surface barriers and high doping densities have been achieved in these *ex situ* structures. The general conduction model

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for this contact structure is described in terms of the interface electrical properties and the contributions from the various components are discussed.



**Key words:** nanoelectric devices, metallic nanoclusters, conjugated organic molecules, chemically stable semiconductor layers.

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The traditional way to fabricate microelectronic devices and circuits involves 'chiseling', i.e. the application of selective deposition or removal of material in lithographically defined areas. Alternatively, it is possible to 'build' well-controlled structures at the nanometer scale using chemical self-assembly techniques. There are a number of self-assembly techniques which have been developed to provide uniform nanometer-scale elements and assemblies of these elements. Of particular interest to this study are structures comprised of metal or semiconductor nanoclusters [1–3]. An interesting challenge is to find approaches where the structures available from 'building' can provide functionality comparable to that realized by 'chiseling' of semiconductor circuits. This paper describes recent work which combines self-assembly techniques involving metal nanoclusters and conjugated organic molecules with semiconductor interface and device structures to form structures of interest for nanoelectronics. We have developed *ex situ* fabrication techniques which are compatible with the self-assembly techniques, as well as suitable low-resistance interface/contact structures at the nanometer scale. We will present a brief summary of the various components and will describe the fabrication, performance and general conduction model for a low-resistance nanoscale ohmic contact structure which has been constructed using this approach.

The general approach described in this paper combines self-assembled nanostructures formed from metallic nanoclusters and conjugated organic molecules with suitable semiconductor heterostructures to realize structures of interest for nanoelectronic applications. The structure of the low-resistance nanoscale ohmic contact which is constructed using this approach is illustrated in Fig. 1. Since the chemical self-assembly techniques are generally not compatible with high-temperature post-processing, and since the successful interfacing of nanometer-scale metallic clusters with the surface requires a well-defined nanometer-scale interface, an essential feature for this hybrid approach is the availability of a chemically stable semiconductor surface layer which does not rapidly oxidize and which remains electrically active during air exposures of at least several minutes. In the current study, surface layers of low-temperature-grown GaAs (LTG:GaAs) [4], i.e. GaAs grown by molecular-beam epitaxy at substrate temperatures in the range of 250–300 °C, are used to provide the requisite surface stability.

The particular structures utilized in this study are based on Au nanoclusters which are approximately 4 nm in diameter. These clusters can be formed into uniform 2D arrays, and the conductance between adjacent clusters can be varied by changing the conjugated organic molecules which are used to link adjacent clusters [1]. The clusters are synthesized in an aerosol reactor [5], and are typically coated with an alkanethiol such as dodecanethiol in order to prevent agglomeration when the clusters are placed into a colloidal suspension. Individual clusters are fcc single crystals in the shape of a truncated octahedron. The well-defined facets of these gold nanocrystals can be observed in high-resolution transmission electron microscope (TEM) images; their influence on the structure of assemblies can also be observed in arrays of the clusters when relatively short linking molecules are used to join adjacent clusters [6]. In contrast to gold clusters which are synthesized in aqueous solution as charged particles, these clusters are charge neutral, which facilitates the formation of closely packed cluster arrays and eliminates offset charge problems when interfaced with device structures. The use of a mono-thiol/unconjugated encapsulant such as dodecanethiol also allows the insertion of conjugated di-thiol linking molecules between adjacent clusters or between a cluster and a semiconductor (or metal) surface, as is accomplished in the work reported here.

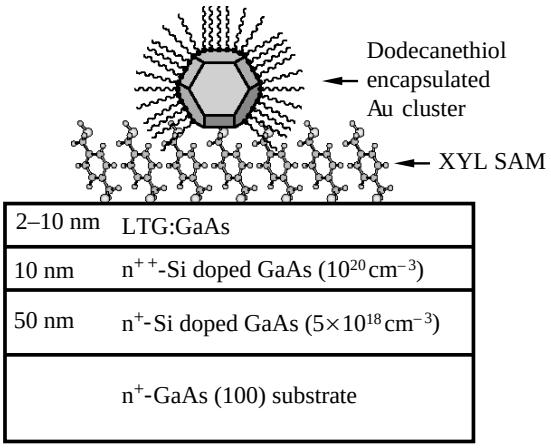


**Fig. 1.** A schematic diagram of the nanocontact structure utilizing a 4 nm diameter Au cluster, a self-assembled monolayer of xylyl dithiol and a GaAs heterostructure with a chemically stable surface layer.

Conjugated organic molecules that are difunctional such as aryl dithiols can provide mechanical linking/tethering between a cluster and another surface (a cluster or a semiconductor/metal surface) as well as a path for electronic conduction between the respective conductors. The specific molecule used for linking the metal nanoclusters to the semiconductor surface in this study is xylyl dithiol ($HS–CH_2–C_6H_4–CH_2–SH$) which will be referred to as XYL for brevity. This molecule has a length of approximately 1 nm and has thiol (–SH) end groups on each end. These thiol groups can chemically bond to the GaAs surface and to the gold nanoclusters. A self-assembled monolayer (SAM) of XYL is grown on the GaAs surface; given the size of the molecule and the expected packing density, it is believed that a number of XYL molecules (approximately 30) are packed within the area of a facet on a 4 nm diameter Au cluster. The interface between a nanocluster and the GaAs surface therefore consists of a small bundle of XYL molecules.

XYL is one of a class of conjugated organic molecules that have been studied as molecular conductors [7–9]. Such experimental studies typically involve the formation of a self-assembled monolayer (SAM) of the desired molecule on a relatively flat Au surface, with a top contact formed either by metal evaporation, deposition of a metal nanocluster, or by a scanning probe tip (e.g. a scanning tunneling microscope (STM) tip). Theoretical descriptions of the current–voltage relationships have been developed based on the scattering theory of transport. With proper treatment of the Fermi level position in the molecule, of the coupling strength to the contacts and of the capacitive division of the applied voltage, the low-field resistances and current–voltage characteristics for the various experimental reports can be adequately described by this modeling approach [7, 9]. If strong bonding (i.e. strong electronic coupling) to the metallic contacts is realized, vertical transport through a SAM of a short molecule such as XYL should have a specific resistance on the order of $1 \times 10^{-8}\ \Omega\ \mathrm{cm}^2$.

Since the self-assembly approaches involving nanoclusters and conjugated organic molecules cannot withstand the process temperatures typically associated with the annealing steps of alloyed contacts or activation of implanted dopants, it is essential that suitable semiconductor device structures are employed to provide low-resistance interfaces between the nanostructure and the doped semiconductor layers. In order to illustrate the need for a suitable surface, consider how ohmic contacts are typically made to n-type GaAs. Since it is difficult to make a contact structure in which the conduction band in n-type GaAs lines up with the Fermi level in the contact metal, tunneling type contacts are typically employed. In principle, it is simple to design a low-resistance tunneling contact: the activated donor density near the surface should be as high as possible and the surface barrier as low as possible, presumably through the use of a relatively low work-function

metal. In practice, these goals are rather difficult to achieve in *ex situ* contacts due to the limited activated donor densities which can be obtained in bulk GaAs and to the mid-gap surface Fermi level pinning which is associated with rapid oxidation of GaAs upon exposure to air. In large-area devices, alloyed contacts such as Au/Ge/Ni are used to overcome these limitations, at the expense of rather deep and nonplanar metal to semiconductor interfaces. Such approaches are not suitable for nanometer-scale ohmic contacts, particularly those involving a nanocluster/semiconductor interface, due to the nonplanar nature of alloyed contacts and to the limited thermal budget dictated by the nature of the self-assembled structures.

Large-area, low-resistance nonalloyed contacts to n-type GaAs layers have been demonstrated using semiconductor heterostructures comparable to that illustrated in Fig. 1 [10]. It is informative to describe the performance and mechanisms of the large-area contacts in order to illustrate the essential features of the semiconductor layers in the nanocontact structure. In the large-area contact structures, the LTG:GaAs layer thickness is generally between 2 and 5 nm and the contact metal (typically Ti) is deposited by evaporation shortly after an oxide-strip step. The large-area contact is therefore a metal–semiconductor (M–S) structure. The ohmic contact structure [10] employs a surface layer of 'as-grown' LTG:GaAs, in which the ∼1–2% *excess* arsenic incorporated during growth is distributed primarily as arsenic antisite defects. The high concentration ($\sim 1.0 \times 10^{20}$ cm$^{-3}$) of point defects results in short minority carrier lifetimes and bulk Fermi level pinning [4]. These defects are observed as a band of states located approximately mid-gap in the GaAs [11]. Previous studies indicate that these states prevent the GaAs surface from rapidly oxidizing due to the relatively low concentration of minority carrier holes in the surface layer [12, 13]. As a result, the presence of the gap states can be observed using STM even following brief air exposure of the samples [12].

In the large-area contact studies, the *ex situ*, nonalloyed ohmic contacts employing a LTG:GaAs surface layer and Ti metallization can provide specific contact resistivities ($\rho_c$) as low as $3 \times 10^{-7}$ Ω cm$^2$ [10]. Applications of this contact to shallow device layers and studies of their temperature stability have been reported [14]. Since these contacts do not suffer from the deep interface and spatial nonuniformity of alloyed contacts, they are of interest for nanometer-scale device applications. This type of contact structure and the chemically stable LTG:GaAs surface layer are also compatible with chemical self-assembly techniques, particularly the nanocluster/molecule-based structures employed in this study [1].

The nanocontact structure illustrated in Fig. 1 employs the same basic heterostructure as the large-area nonalloyed contact, but the nature of the interface between the metal and the semiconductor surface are different in the two cases. In particular, the nanocontact requires a molecular tether layer (the XYL) in order to provide suitable mechanical stability. Without this layer, the clusters are too mobile to form a stable contact structure. As will be described later, the XYL monolayer also plays an important role in achieving a low-barrier height and low tunneling resistance through the nanocontact. The nanocontact structure is therefore best described as a metal–'insulator'–semiconductor (M–I–S) structure, in which the XYL represents a very thin, and relatively leaky, insulator. The thickness of the low-temperature-grown GaAs surface layer is 10 nm for the nanocontact structures described in this work. In these studies, a controlled area nanocontact is formed by a single crystal, 4 nm diameter Au nanocluster deposited on the GaAs surface, which has previously been coated with a SAM of XYL. Details of the procedure have been presented elsewhere [15, 16]. The area of the conducting path for the nanocontact ($1 \times 10^{-13}$ cm$^2$) is defined by the area of a facet on the 4 nm diameter cluster. The current–voltage characteristics of the nanocontact structures have been measured using ultra-high-vacuum (UHV) STM current–voltage spectroscopy in the near-contact regime, i.e. a regime in which the resistance between the STM tip and the cluster is reduced by bringing the tip in close proximity to the cluster [15, 16]. STM tips used in this study have typical end shapes with diameters less than 15 nm, as observed from TEM micrographs. Representative current–voltage curves are shown in Fig. 2A for a nanocontact consisting of an isolated cluster, with curves shown for cases where the tip is over the cluster ('A'), i.e. conduction through the cluster-based nanocontact structure, and where the tip is over a region of XYL coated GaAs, but not over a cluster ('B'). As illustrated in the figure, $I(V)$ curves measured over the cluster indicate a significant enhancement in the relative conduction at low bias voltages as compared

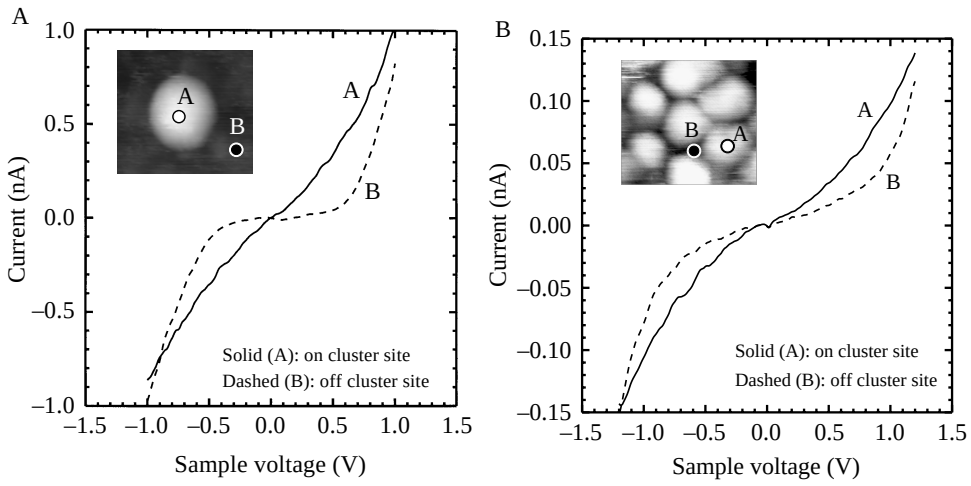


**Fig. 2.** Measured current–voltage relationships from UHV STM characterization of nanocontact structures with cross-section depicted in Fig. 1. In each figure, curves are shown for $I$–$V$ relationships both on a cluster and over the XYL-coated surface, with the respective curves corresponding to measurements at the locations indicated on the inset images. A, Representative curves acquired at a set-point of $I_{\text{set}} = 0.8$ nA, $V_{\text{set}} = -1.0$ V for an isolated nanocluster tethered to the XYL-coated heterostructure, as illustrated in the inset (a 20 × 20 nm STM topographic image of Au cluster, acquired with $I_{\text{set}} = 1.0$ nA and $V_{\text{set}} = -1.0$ V). B, Representative curves acquired at a set-point of $I_{\text{set}} = 0.15$ nA, $V_{\text{set}} = -1.2$ V for a structure with a close-packed array of Au nanoclusters on the XYL-coated heterostructure, as illustrated in the inset (a 25 × 25 nm STM topographic image of array of tethered clusters, acquired with $I_{\text{set}} = 0.1$ nA and $V_{\text{set}} = -1.2$ V).

with the corresponding curves for the measurement over the XYL coated surface (without cluster). The curves shown in Fig. 2 are taken at relatively low set-point currents, corresponding to relatively large tip-to-sample distances. In order to determine the resistance of the nanocontact, i.e. the resistance between the Au nanocluster and the doped layers in the semiconductor structure, it is necessary to reduce the tip-to-cluster distance, and thereby the tip-to-cluster resistance. This has been accomplished by monitoring the current ($I$) at constant voltage as the tip height ($z$) is decreased with respect to the height at the low-current set-point. For a structure in which the LTG:GaAs layer is undoped (n-type), the measured $I(z)$ relationship saturates as the tip is brought closer to the cluster. In the saturation regime, the overall resistance should be dominated by the cluster-to-substrate resistance, so the desired nanocontact resistance can be determined from the ratio of the applied voltage to the the saturation current [15]. For this case, the specific contact resistance is approximately $1 \times 10^{-6}$ Ω cm$^2$ and the maximum current density is approximately $1 \times 10^6$ A cm$^{-2}$ [15]. For a structure in which the LTG:GaAs layer is heavily doped with Be, as is the case in Fig. 2, values of $\rho_c$ of approximately $1 \times 10^{-7}$ Ω cm$^2$ and the maximum current density of approximately $1 \times 10^7$ A cm$^{-2}$ are obtained. Both of these values are comparable to the values achieved in high-quality large-area ohmic contacts to n-type GaAs [16]. In this case, the measurement system limits the minimum $\rho_c$ which can be resolved, so the contact resistance may be somewhat lower than this value. Thus, the measured specific contact resistances for the best nanocontact structures are somewhat better than those achieved in the large-area contact studies, indicating that high-performance nanocontacts can be formed to GaAs device layers using this nonalloyed ohmic contact approach.

We have also characterized structures in which the clusters are deposited as close-packed arrays on the XYL coated LTG: GaAs surface. A STM topographic image of an array of 5 nm diameter clusters formed on the XYL-coated LTG:GaAs structure is shown in Fig. 3. This image illustrates the good local ordering achieved through self-assembly; the faceted nature of the clusters can also been seen. Representative $I$–$V$ curves are shown in Fig. 2B, again for the case where the STM tip is over a cluster ('A') and for the case where

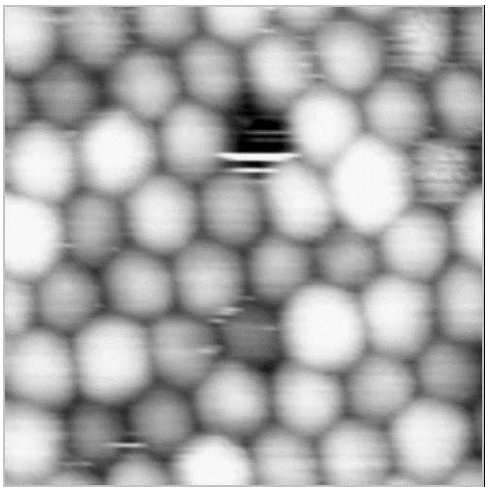

**Fig. 3.** A 50 × 50 nm UHV STM topographic image of close-packed 2D array of Au nanoclusters tethered to the XYL-coated semiconductor heterostructure with Be-doped LTG:GaAs surface layer, acquired with $V_{set} = -1.2$ V and $I_{set} = 0.1$ nA.

the STM tip is positioned between clusters ('B'). As was observed in the experiment with isolated clusters, an enhancement in low-field conduction is observed when the tip is positioned over a cluster. The 'set-points', i.e. the current/voltage pairs used to establish the relative tip height with respect to the local surface, are approximately the same in the 'on-cluster' and 'off-cluster curves', in order to allow reasonable comparison between the low-field current values. While the nature of the STM technique makes it difficult to ascribe absolute conductance values to the data, the fact that these trends persist to relatively high current levels (when the tip is brought closer to the cluster) indicates that the conduction is enhanced when the conduction is through a cluster. The current–voltage relationships for clusters within an array are comparable to those obtained on isolated clusters. This observation is consistent with the fact that the cluster array used in this study is 'unlinked', i.e. adjacent clusters within this array are separated by the dodecanethiol encapsulant but not linked by conjugated molecules. In this case, the coupling to the semiconductor substrate is much stronger than the intercluster (resistive) coupling. If adjacent clusters within the array were linked with a conductive molecule, it is expected that the intercluster resistance could be made comparable to the cluster-to-substrate resistance. In this case, the conduction path to the substrate would be through a number of clusters. The difference in shapes of the $I(V)$ curves when the STM tip is located on and off a cluster are not as dramatic in the case of the cluster within an array. Since the end size of the tip is 10–15 nm, it is likely that there is some conductivity through adjacent clusters for the 'off-cluster' curve in the array sample (Fig. 2B).

As a starting point for modeling the nanocontact performance, consider a quantitative conductance model for the large-area ohmic contact (M–S structure) which has recently been developed [17]. This analysis calculates the conduction band profile for the semiconductor structure (solution of Poisson equation and Fermi statistics), using parameters for the mid-gap and shallow acceptor states in the LTG:GaAs which are consistent with experimental observations. A calculated profile for a contact structure with a LTG:GaAs layer thickness of 3 nm, a barrier height of 0.5 eV and an activated donor density of $1 \times 10^{20}$ cm$^{-3}$ is shown in Fig. 4. Also shown in the figure is the band profile for a uniformly doped Schottky barrier (assuming complete ionization) with the same barrier height and comparable depletion depth. Since this profile is reasonably approximated by a parabolic profile, the contact resistance can be calculated by applying expressions for a uniformly doped Schottky contact with equivalent barrier heights and effective depletion depths. This model adequately predicts experimental trends for specific contact resistance versus LTG:GaAs layer thickness and

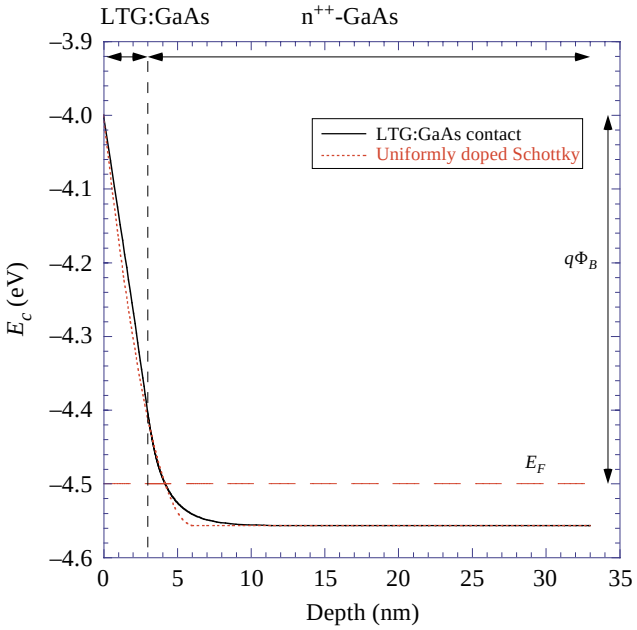


**Fig. 4.** Calculated conduction band profile for ohmic contact structure (metal/GaAs) with a 3 nm thick LTG:GaAs layer on the surface. The curve is for a structure with a barrier height of 0.5 eV, a doping density in the $n^+$-GaAs layer of $1 \times 10^{20}$ $cm^{-3}$ and a temperature of 300 K. Also shown is the parabolic conduction band profile for an equivalent uniformly doped Schottky barrier, assuming complete ionization.

versus temperature. Based on comparisons between the experimental results and the predictions, it appears that the surface barrier heights are well below mid-gap values and that activated donor densities above the bulk amphoteric limit have been achieved in the space charge region [17]. It is typically not possible to achieve either of these effects in *ex situ* contacts to n-type GaAs, due to the surface Fermi level pinning associated with rapid surface oxidation and the bulk amphoteric doping limit in stoichiometric layers. The achievement of both effects in this structure is attributed to the passivating effects of the thin LTG:GaAs layer and to the associated Fermi level control. The ability to achieve an activated donor density within the surface space-charge region which is higher than the bulk amphoteric limit has been explained in terms of the control of Fermi level during growth and the passivation effects of the LTG:GaAs layer [10]. It should be noted that the semiconductor heterostructure is typically exposed to air for prolonged periods before contact processing, so a portion of the LTG:GaAs layer does oxidize during this storage period. Since this oxidation appears to cause mid-gap surface Fermi level pinning even in LTG:GaAs following prolonged air exposure [18], the surface oxide must be stripped shortly before contact metallization. It is believed that the oxide strip restores the surface Fermi level to an unpinned state, with a re-oxidation time constant of hours.

For the nanocontact structure (M–I–S), this conduction model must be modified to incorporate the effects of the layer of XYL between the cluster and the GaAs surface. Qualitatively, this layer can be described as a leaky dielectric, since there are states associated with the molecular levels (HOMO and LUMO states). Generally, one of these two levels will be within 0.5–1 eV of the Fermi level when the molecules are sandwiched between two metallic electrodes [7]. Since the dielectric constant of this layer (expected to be around 2) is considerably lower than that of GaAs, a relatively large portion of the electrostatic potential difference between the metal cluster and the semiconductor bulk is dropped across the XYL layer. This effect reduces the barrier at the surface of the GaAs, again with respect to the GaAs bulk, and makes the barrier relatively more

transparent in comparison with a case without the organic monolayer. This effect is especially important in light of the fact that the workfunction of Au is higher than that of the Ti metallization used in large-area contacts, which would tend to raise the barrier height in the M–I–S with respect to that of the M–S structure. In order to explain the low specific contact resistance in both the large-area contact and in the nanocontact, it is necessary to assume a relatively low density of interface charge at the GaAs surface. Since the surface oxide is chemically stripped from the semiconductor surface immediately before growth of the XYL monolayer, it is believed that the surface Fermi level is also unpinned in the nanocontact structure. Independent observations of the relative stabilities of organic monolayers on undoped and Be-doped layers of LTG:GaAs indicate that the Be-doped layers are more stable. Therefore, it is likely that the nanocontact samples with Be doping in the LTG:GaAs surface layer have a lower interface state density than those in the samples with undoped surface layers. This trend could explain the superior contact resistance properties of the samples with Be-doped surface layers.

In addition to the cluster-based nanocontacts, we have demonstrated other structures which utilize either the chemically stable LTG:GaAs surface layer for device structures or patterned organic monolayers on GaAs, which can provide templates for approaches which combined self-assembly for well-ordered nanometer-scale assemblies and lithographically based techniques for arbitrary global ordering [19, 20]. These structures and approaches represent a toolbox which may eventually be suitable for self-assembling a device structure with functionality comparable to that of a transistor.

A number of devices have been reported in which some feature is in the nanometer scale, including a few recent examples [21–24]. Frequently the overall device dimensions are much larger than this minimum feature size, particularly in structures employing semiconductor channels and source/drain regions. This size discrepancy is largely due to the need for ohmic contact structures which are in the micrometer scale in lateral extent and typically 100 nm or greater in depth. In contrast, an ohmic contact technology which can provide nanometer contact dimensions, both laterally and vertically, could allow the demonstration of the high circuit densities promised by nanometer-scale device concepts.

In conclusion, we have described a nonalloyed contact structure which may be suitable for high-density nanoelectronic device applications and discussed several experiments aimed at developing nanoscale functional devices. In the nanocontact structure, the controlled dimension contact is formed by a gold nanocluster with well-defined crystal facets. Strong mechanical tethering of the cluster to the semiconductor surface is provided by a self-assembled monolayer of a conjugated organic molecule (xylyl dithiol). The low-resistance contact between the nanocluster and the doped semiconductor layer is attributed to the chemical stability of the low-temperature-grown GaAs surface layer, which results in a low interface density, and to the passivation effects and conduction properties of the xylyl dithiol. The ability to form stable, low-resistance interfaces between metallic nanoclusters and semiconductor device layers using *ex situ* processing allows chemical self-assembly techniques to be utilized to form interesting nanoscale semiconductor devices. These demonstrations provide device approaches and fabrication techniques which can be integrated to develop a high-density nanoelectronic device technology with high-throughput fabrication processes.

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