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14. ABSTRACT Progress has been made in several areas of research undertaken by the PI and co-PIs. Research areas include the development of rewritable nanoelectronic devices with potential for quantum information processing, optical characterization of coherent phonons in GaAs, synthesis of monodisperse crystalline colloidal arrays, and development of plasmonic nanoscale waveguides for spectroscopy on a chip applications.					
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Report Title

Quantum Information Processing with Ferroelectrically Coupled Quantum Dots: Final Report

ABSTRACT

Progress has been made in several areas of research undertaken by the PI and co-PIs. Research areas include the development of rewritable nanoelectronic devices with potential for quantum information processing, optical characterization of coherent phonons in GaAs, synthesis of monodisperse crystalline colloidal arrays, and development of plasmonic nanoscale waveguides for spectroscopy on a chip applications.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Y. Xi, Y. S. Jung, and H. K. Kim, "Interaction of light with a metal wedge: the role of diffraction in shaping energy flow", Optics Express 18, 2588-2600 (2010).

Y. S. Jung, J. Wuenschell, H. K. Kim, P. Kaur, and D. H. Waldeck, "Blue-shift of surface plasmon resonance in a metal nanoslit array structure", Optics Express 17, 16081-16091 (2009).

Y. S. Jung, Y. Xi, J. Wuenschell, and H. K. Kim, "Near- to far-field imaging of phase evolution of light emanating from a metal nanoslit", Optics Express 16, 18881-18888 (2008).

Number of Papers published in peer-reviewed journals: 3.00

(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

S. Srisophonpan, H. Park, Y. S. Jung, and H. K. Kim, "Ballistic transport of electrons in nanoscale void channels formed in the oxide layer of a MOS structure," Proceedings of IEEE Nano 2010, the 10th International Conference on Nanotechnology, Seoul, Korea, August 17-20, 2010.

Number of Papers published in non peer-reviewed journals: 1.00

(c) Presentations

Jeremy Levy, APS 2007 March Meeting, "Oxide-Semiconductor Materials for Quantum Computing", Denver, CO, March 7, 2007.

Jeremy Levy, National University of Singapore, "Opportunities in Oxide Nanoelectronics", Singapore, June 12, 2007.

Jeremy Levy, National University of Singapore, "Quantum Computing with Electron spins in Semiconductors", Singapore, June 13, 2007.

Jeremy Levy, University of Delaware, "Nanoscale Control of a Metal-Insulator Transition a Room Temperature", Newark, DE, September 19, 2007.

Jeremy Levy, CMU Quantum Information Lecture Series, "Designer Quantum Materials", Pittsburgh, PA, May 2, 2008.

Jeremy Levy, Birla Institute of Technology and Science, Pilani, "Quantum Computing", Pilani, India, Jan 9, 2009.

Jeremy Levy, MRS Spring Meeting, "Oxide Nanoelectronics On Demand", San Francisco, CA, April 7, 2010.

Number of Presentations: 7.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

S. Srisophonpan, H. Park, Y. S. Jung, and H. K. Kim, "Ballistic transport of electrons in nanoscale void channels formed in the oxide layer of a MOS structure," Proceedings of IEEE Nano 2010, the 10th International Conference on Nanotechnology, Seoul, Korea, August 17-20, 2010.

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

1

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

0

(d) Manuscripts

Y. Xi, Y. S. Jung, and H. K. Kim, "Interaction of light with a metal wedge: the phase relationship of boundary diffraction waves" (in preparation).

A. Basak, H. Petek, K. Ishioka, and C. Stanton, "Ultrafast Carrier-phonon coupling with high photoexcitation in GaAs," Phys. Rev. B (in preparation).

A. Basak, H. Petek, K. Ishioka, and C. Stanton, "Anisotropic Coupled Plasmon-Phonon Coupling in GaAs under Intense Photoexcitation," Phys. Rev. B (in preparation).

K. Ishioka, A. Basak, H. Petek,, "Optical Detection Mechanisms of Coherent Phonons and Plasmon-Phonon Coupled Mode in GaAs Excited near the E0 Gap," Phys. Rev. B (in preparation).

Number of Manuscripts: 4.00

Patents Submitted

Patents Awarded

Awards

Sandy Asher: Spectroscopy Society of Pittsburgh, Pittsburgh Spectroscopy Award (2008).

Jeremy Levy: APS Fellow (2010), Nano50 innovation award (2008).

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Yun Suk Jung	0.10
Yonggang Xi	0.40
Amlan Basak	0.25
Yun Sun Jung	0.10
FTE Equivalent:	0.85
Total Number:	4

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME

PERCENT SUPPORTED

FTE Equivalent:

Total Number:

Names of Under Graduate students supported

NAME

PERCENT SUPPORTED

FTE Equivalent:

Total Number:

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in
science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue
to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 0.00

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The number of undergraduates funded by your agreement who graduated during this period and intend to
work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive
scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME

PERCENT SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

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Quantum Information Processing with Ferroelectrically Coupled Quantum Dots: Final Report

Statement of the Problem

The basis of this proposal originated from a DARPA grant (DAAD-19-01-1-0650) led by **Jeremy Levy**, the purpose of which was to develop quantum computing technology using ferroelectrically coupled quantum dots. Significant progress has been made in the development of oxide-semiconductor materials for this application, including techniques for growing ordered arrays of sub-10-nm Ge islands on Si substrates, for growing out-of-plane oriented ferroelectrics epitaxially on Si, electrical gating methods for single spin quantum gating.

In this proposed extension, the focus will be to broaden and expand on some of the most successful avenues of research, and to include new research directions that possess potential benefit for the Army and the soldier.

The principal investigators for this project are listed below. We note that Albert Heberle left the University of Pittsburgh.

Investigator	Department
Sandy Asher	Chemistry
Albert Heberle	Physics and Astronomy
Hong Koo Kim	Electrical and Computer Engineering
Jeremy Levy	Physics and Astronomy
Hrvoje Petek	Physics and Astronomy

Table 1. Participating faculty members and their affiliation at the University of Pittsburgh.

Our team is composed entirely of faculty members at the University of Pittsburgh (see Table 1), and includes the two co-directors of the University of Pittsburgh's Petersen Institute for NanoScience and Engineering (PINSE), **Hong Koo Kim** and **Hrvoje Petek**. Much of the research related to the development of shared resources at the Nanoscale Fabrication and Characterization Facility (NFCF). A Collaborative Research And Development Agreement (CRADA) had been set up through the University of Pittsburgh to help guide the research and shape its relevance to AMRDEC research activities and goals.

The proposed research falls under three main themes: (1) quantum information technology, (2) photonics and (3) novel spectroscopies. Below we describe five projects that can broadly be characterized within these three major research areas.

Quantum Information Technology (Jeremy Levy)

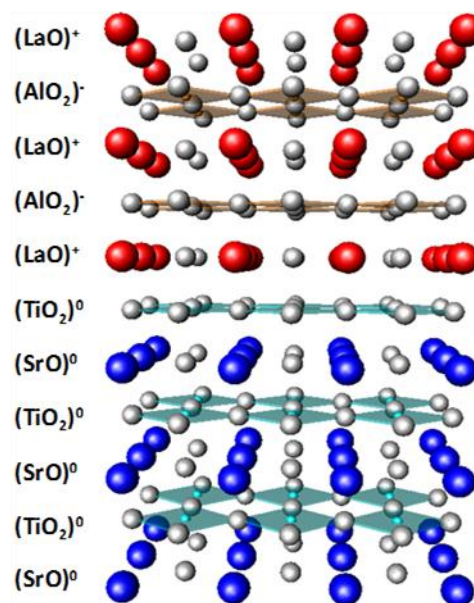
Two-Dimensional Electron Gas

The two-dimensional electron gas (2DEG) that forms at the interface between two heterogeneous semiconductors, or between a semiconductor and oxide, provides a platform for some of the most useful and prevalent electronic devices. Silicon-based metal-oxide-semiconductor field-effect transistors, GaAs-based high-mobility transistors, solid-state lasers and photodetectors are but a few examples of technologies that have emerged from semiconductor interfaces. Herb Kroemer, when receiving the Nobel Prize in Physics for the invention of the semiconductor heterostructure, famously stated that “the interface is the device” [1].

Complex oxides

Complex oxides differ in many ways from traditional semiconductors that are used to form 2DEG devices [2]. Both LaAlO_3 and SrTiO_3 are compatible with a large class of materials that, collectively, exhibit a broad range of behaviors that include ferroelectricity, magnetism, multiferroic behavior, and superconductivity. The development of complex oxides over the past fifteen years has raised the prospect for new classes of electronic devices [2,3]. In 2004, Ohtomo and Hwang published their seminal discovery that a high-mobility 2DEG can form at the interface between LaAlO_3 and SrTiO_3 [4] (Figure 1). Since then a number of striking properties of this interface have been discovered and explored, including interfacial superconductivity [5,6] and the ability to reversibly create conducting nanostructures at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface using conductive atomic-force microscopy (c-AFM) [7,8].

Figure 1. Oxide heterostructure consisting of 3 unit cells of polar LaAlO_3 grown on TiO_2 -terminated (non-polar) SrTiO_3 . The polar discontinuity supports the formation of a conducting interface when the top LaAlO_3 surface is positively charged. Adapted from Ref. [9].



$\text{LaAlO}_3/\text{SrTiO}_3$ structure and properties

LaAlO_3 and SrTiO_3 both possess a perovskite crystal structure. SrTiO_3 , a non-polar oxide, is composed of alternating, stacked layers of $(\text{SrO})^0$ and $(\text{TiO}_2)^0$. SrTiO_3 is pseudo-cubic at room temperature with a lattice constant $3.905 \text{ \AA}$. It is a band insulator with a band gap of 3.25 eV , is chemically inert, and has been a substrate of choice for the growth of many other oxides and high- T_C superconductors [3,10,11]. LaAlO_3 , a polar oxide, is composed of alternating stacked layers of $(\text{LaO})^+$ and $(\text{AlO}_2)^-$. LaAlO_3 has a pseudo-cubic structure with a lattice constant of $3.789 \text{ \AA}$ at room temperature. It is a Mott insulator [12] with a wide band gap of 5.6 eV . Films of LaAlO_3 thinner than ~ 15 monolayers (ML) [13] can be grown coherently strained to SrTiO_3 . The manageably small lattice mismatch between LaAlO_3 and SrTiO_3 (about 3%) enables high-quality

epitaxial growth of LaAlO_3 on SrTiO_3 via pulsed laser deposition (PLD) [9] or molecular-beam epitaxy (MBE) [14].

Polar catastrophe model

Epitaxial growth of LaAlO_3 on SrTiO_3 [4] can lead to an unusual and energetically unstable charge distribution. For the case of TiO_2 -terminated SrTiO_3 (Figure 2(a)), the first-grown LaO layer will have a positive charge (+1/unit cell), after which the charge of each layer alternates between -1 and +1. This alternating charge density can be integrated to reveal both the internal electric field E and voltage V across the LaAlO_3 layer. The magnitude of the voltage increases linearly with the thickness of the LaAlO_3 layer. The divergence of the electrostatic energy can lead to a “polar catastrophe” and an associated electronic reconstruction in which the polarization is screened (in part) by the formation of a 2DEG at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface (Figure 2(c)).

Top LaAlO_3 surface

One missing component of this model is a discussion of what happens at the top LaAlO_3 surface. The absence of charge neutrality (apparent in Figure 2(c)) leads to a much more severely catastrophic situation than a finite, but diverging, polarization. To achieve charge neutrality, the top surface must also be compensated with charge. One such scenario that was invoked to explain metastable writing at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface (described in more detail below) is the formation of oxygen vacancies at the top LaAlO_3 surface. Another scenario, described by Son *et al* [15], involves the adsorption of hydrogen at the top AlO_2 surface. The hydrogen could be formed by spontaneous dissociation of H_2O molecules or through direct adsorption of H_2 in gas form. The charged top surface is expected to retain its insulating properties, but donation of electrons to the $\text{LaAlO}_3/\text{SrTiO}_3$ interface can be achieved for TiO_2 -terminated SrTiO_3 .

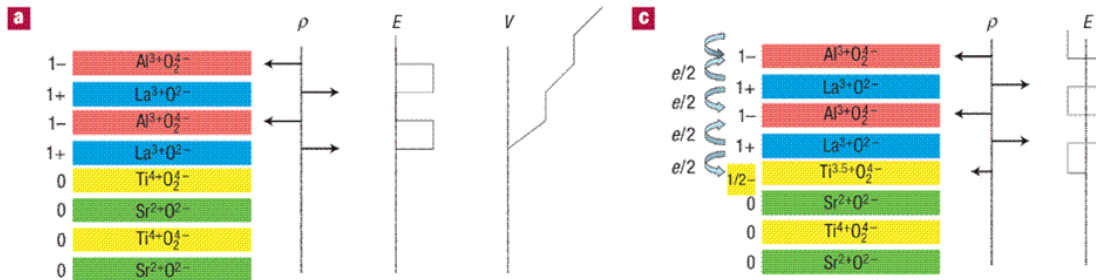


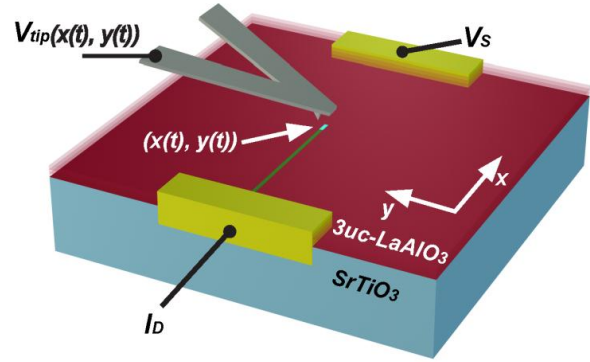
Figure 2. Illustration of polar discontinuity contributing to the formation of conductivity at $\text{LaAlO}_3/\text{SrTiO}_3$ interface. (a) Non-reconstructed LaO/TiO_2 interface. A positive potential exists in the LaAlO_3 layer and diverges with film thickness. (c) At the LaO/TiO_2 interface, adding $1/2$ electron per unit cell to the top TiO_2 layer can minimize the potential in LaAlO_3 . (Adapted from Ref. [4].)

Metal-insulator transition in $\text{LaAlO}_3/\text{SrTiO}_3$

One predicted consequence of the polar discontinuity between LaAlO_3 and SrTiO_3 is an interfacial insulator-to-metal transition that is dependent on the LaAlO_3 thickness. This effect, first reported in 2006 by Thiel *et al* [9] and confirmed by many groups, occurs in structures of LaAlO_3 grown on TiO_2 -terminated SrTiO_3 . When LaAlO_3 is grown with greater than a critical thickness $d_c = 3$ unit cell (uc), the interface between LaAlO_3 and SrTiO_3 is found to be conducting. When the thickness of LaAlO_3 is smaller than d_c the interface remains insulating.

Most importantly, in samples grown with approximately 3 uc of LaAlO_3 (normally insulating), the interface can be reversibly switched between the insulating and conducting states by applying a voltage to the back of the SrTiO_3 substrate [9]. This transition is a hysteretic function of the applied electric field.

Figure 3. Schematic illustration of the nanowriting process at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface. Au electrodes (shown in yellow) are electrically contacted to the $\text{LaAlO}_3/\text{SrTiO}_3$ interface. The AFM tip with an applied voltage is scanned once between the two electrodes with a voltage applied $V_{\text{tip}}(x(t), y(t))$. Positive voltages locally switch the interface to a conducting state, while negative voltages locally restore the insulating state. Here, a conducting nanowire (shown in green) is being written. The conductance between the two electrodes is monitored by applying a small voltage bias on one of the two gold electrodes (V_s) and reading the current at the second electrode (I_D). (Adapted from Ref. [16].)



Reconfigurable nanoscale devices

A powerful method for creating nanoscale devices at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface involves metastable charging of the top LaAlO_3 surface with a c-AFM probe [7,8]. By locally and reversibly controlling a metal-insulator transition, the creation of both isolated and continuous conducting features has been demonstrated with length scales smaller than 2 nm. These structures can be erased and rewritten repeatedly. As a result of the enormous flexibility in controlling electronic properties at near-atomic dimensions, a variety of nanoscale devices can be realized.

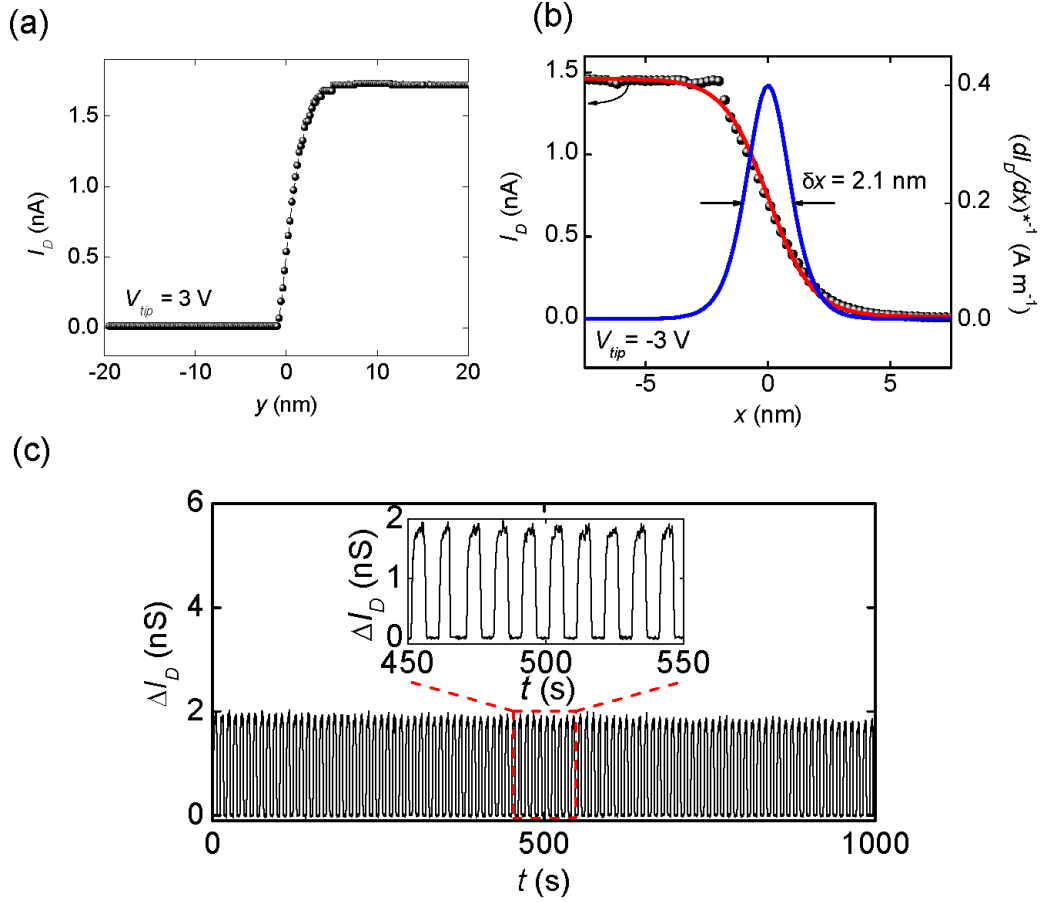


Figure 4. Writing and erasing nanostructures at the LaAlO₃/SrTiO₃ interface. (a) Conductance between the two electrodes measured with a lock-in amplifier as a function of the tip position while writing a conducting wire with $V_{tip} = +3$ V. A steep increase in conductance is observed when the tip reaches the second electrode. (b) Conductance drops when the wire is cut with $V_{tip} = -3$ V. (c) Repeated cutting and restoring the conductance of a 12 nm-wide nanowire using $V_{tip} = \pm 10$ V. (Adapted from Ref. [7] and [8].)

Nanoscale writing and erasing

To create conducting nanostructures, a c-AFM tip is placed in contact with the top LaAlO₃ surface and biased at V_{tip} with respect to the interface, which is held at electrical ground (

Figure 3). Positive tip voltages locally produce a metallic interface, while negative tip voltages locally restore the insulating state. During the writing and erasing process, the conductance is monitored between buried Au electrodes that directly contact the interface. Figure 4 (a) illustrates the result of writing with $V_{tip} = +3$ V. A sharp increase in conductance is observed when a conducting path is obtained between the two monitored electrodes ($y = 0$ nm).

To provide a measure of the transverse dimension of the conducting wire, and to demonstrate that the writing process is reversible, the wire can be “cut” with a reverse voltage $V_{tip} = -3$ V (Figure 4 (b)). A sharp reduction in current is observed, comparable in abruptness to the one found for the writing process. Assuming that the erasure process has a resolution comparable to

the writing process, one can infer the nanowire width from the deconvolved differential profile full width at half maximum (FWHM).

The writing and erasing process can be repeated hundreds of times without noticeable degradation of the conducting properties. Figure 4 (c) illustrates the repeated erasing and writing of a nanowire using 100 ms voltage pulses. The inset shows that alternating conducting and insulating states are consistently achieved.

Photonics

Asher

We developed new methods to synthesize monodisperse highly surface charged silica spheres which self assemble into colloidal arrays that diffract light in the UV spectral region. The objective is to synthesize spheres that did not absorb for wavelength greater than 200 nm. We monitored the diffraction from these particles and the magnitude of the repulsive interactions between them. The objective is to use these particles to construct a photonic crystal deep UV Rayleigh rejection filter.

We developed a synthesis for monodisperse silica colloids that will be used for the deep UV photonic crystals. We then developed a synthetic method to functionalize the surface so that the particles would repel each other and form crystalline colloidal arrays that diffract deep UV light.

Petek

Coherent phonon spectroscopy on variously doped GaAs samples was performed with 10 fs pulse excitation at 800 nm and 400 nm. The dynamics observed strongly depend on the probing wavelength on account of different penetration depths. With 800 nm probing, the experiment is sensitive to the coherent phonon interactions with the bulk carriers. By contrast, at 400 nm the penetration depth is <100 nm and dynamics reflect the hole-phonon interactions, because of fast electron diffusion from the surface region.

Novel Spectroscopies

Hong Koo Kim

We observed electrically-induced, explosive atomization of metals and analytes at nanoscale. The phenomenon is found to involve formation of highly-localized nanoscale leakage channels in the oxide layer of a metal-oxide-semiconductor (MOS) structure under pulsed drive, ballistic transport of injected electrons in the nanoscale void channels, impact ionization of metal atoms, and explosive atomization of metal and adjacent analyte materials. The fragmented atoms produce atomic luminescence from radiative transitions in the relaxation process. This electrically induced explosive atomization through nanochannels offers a potential for nanoscale elemental/trace analysis on a chip.

When a light wave hits a metal wedge structure, the metal surfaces respond to the incident light by generating both free-space and surface-bound waves. Here we present a physical model that elucidates electromagnetic interactions of an incoming planar wave with a simple semi-infinite 90° metal wedge. We show that a metal wedge structure possesses an intrinsic capability of directing the incident power around the corner into the forward direction. Interplay of the

boundary diffraction wave and the incident and reflection waves in the near field region of a metal corner is found to form a basis of the funneling phenomena that are commonly observed in metal nanoslit structures. Theory and experiment reveal that the incident wave propagating parallel to the sidewall destructively interferes with the boundary diffraction wave forming a depleted-energy-flow region along the glancing angle direction. A physical understanding of various electromagnetic phenomena associated with a metal wedge structure confirms rich potential of the simple structure as an elemental building block of complex metal nanostructures.

The adsorption of a self-assembled monolayer of molecules on a metal surface commonly causes a red-shift in its surface plasmon resonance. We report that the anomalous dispersion of surface plasmons in a Au nanoslit array structure can cause a blue-shift of optical transmission upon adsorption of a non-absorbing self-assembled monolayer of molecules. We develop a simple model that explains the blue-shift observed in the transmission spectra with monolayer adsorption in terms of the interplay of anomalous dispersion and the cavity resonance of surface plasmons in the nanoslit array.

We report near- to far-field measurement of optical wavefronts emanating from a nanoslit formed in a thin (50 nm thick) Ag film. The evolution of optical phases is imaged using a self-interference technique in conjunction with a scanning probe method. The phase relationship of the slit-transmitted waves with respect to the direct transmission through the thin metal film is quantitatively established. The singular-phase points resulting from the interplay of slit diffraction and surface plasmons are identified in the intermediate-field region.

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