Quantum cryptography

Idea is to use the quantum mechanical nature of light to make either (a) unbreakable codes, or (b) communications systems that are completely secure.

In this case, complete security means absolute certainty of whether any third parties are eavesdropping.

Nanoscale physics can be directly relevant to the physical implementation of these schemes.

- “No-cloning” theorem
- Basic idea of quantum cryptography
- Implementations and nanoscale physics

Key distribution

Suppose Alice and Bob want to send coded messages to each other in a secure way.

One might think that QM can help here. Suppose Alice and Bob agree to use a complicated quantum mechanical state as their key somehow. Then no one single measurement by a third party could be used to deduce the key.

Unfortunately, it's not possible for Alice and Bob to each have identical quantum states at their disposal, because of the "no-cloning" theorem.

No-cloning theorem

It is impossible to exactly duplicate an unknown quantum state.

This result is called the “no-cloning” theorem (Wootters, Zurek; Dieks 1982), and is easy to prove by contradiction.

Suppose there was a Quantum Xerox Machine that could clone quantum states.

$$|0\rangle \otimes |u\rangle \rightarrow |0\rangle \otimes |0\rangle \otimes |v_0\rangle$$
$$|1\rangle \otimes |u\rangle \rightarrow |1\rangle \otimes |1\rangle \otimes |v_1\rangle$$

state of QXM before

state of QXM after

Note that there’s no reason for the “after” states to be orthogonal to each other.

What happens if we use the QXM on a superposition?

No-cloning theorem

$$\left[a|0\rangle + b|1\rangle\right] \otimes |u\rangle \rightarrow a|0\rangle \otimes |0\rangle \otimes |v_0\rangle + b|1\rangle \otimes |1\rangle \otimes |v_1\rangle$$

Problem! This does not equal a clone of the initial state:

$$\left[a|0\rangle + b|1\rangle\right] \otimes |u\rangle \rightarrow \left[a|0\rangle + b|1\rangle\right] \otimes \left[a|0\rangle + b|1\rangle\right] \otimes |v'\rangle$$

So, cloning of a generic state is not compatible with linearity in quantum mechanics.

In fact, cloning would violate unitarity, and would also allow faster-than-light communication via EPR states.

So, it's pretty clear that cloning of unknown states can't work.

Basic idea

Suppose Alice wants to encode a message and wants Bob to be able to read it.

The most secure way to do this is the “one-time pad”. Alice and Bob have to have a shared set of randomly generated key (zeros and ones) that they use once and never again.

One problem: how to distribute this key *securely*, using potentially insecure public communications.

Another problem: how to generate the key in the first place, so that it’s truly random.

Turns out, quantum mechanics can help us with both of these.

Measurement and perturbation

Start with two possible quantum states that are *not* orthogonal, and ask how easy it is to distinguish between them.

Couple these states to some system in a way that *does not disturb* the states, and let them evolve.

$$|\phi\rangle \otimes |u\rangle \rightarrow |\phi\rangle \otimes |v\rangle$$
$$|\psi\rangle \otimes |u\rangle \rightarrow |\psi\rangle \otimes |v'\rangle$$

Inner product of initial states should = inner product of final states:

$$(\langle u| \otimes \langle \phi|)(|\psi\rangle \otimes |u\rangle) = (\langle v| \otimes \langle \phi|)(|\psi\rangle \otimes |v'\rangle)$$
$$\langle u\|u\rangle\langle \phi\|\psi\rangle = \langle v\|v'\rangle\langle \phi\|\psi\rangle$$
$$1 = \langle v\|v'\rangle$$

This just shows that any measurement that doesn't perturb the system can't let you distinguish nonorthogonal states.

The only way to distinguish nonorthogonal states is to disturb them!

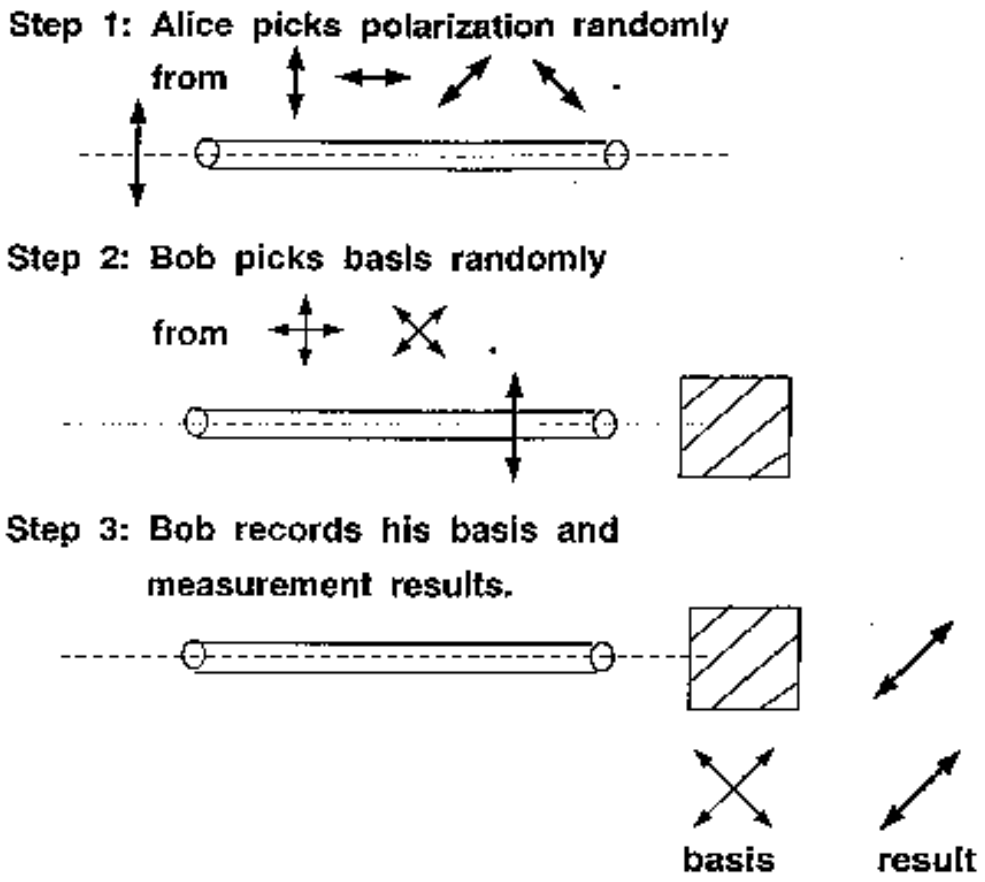
Correlations, polarization, and security

This fact makes possible a very clever cryptography scheme.

Suppose Alice sends a series of photons in four possible polarizations, randomly selected.

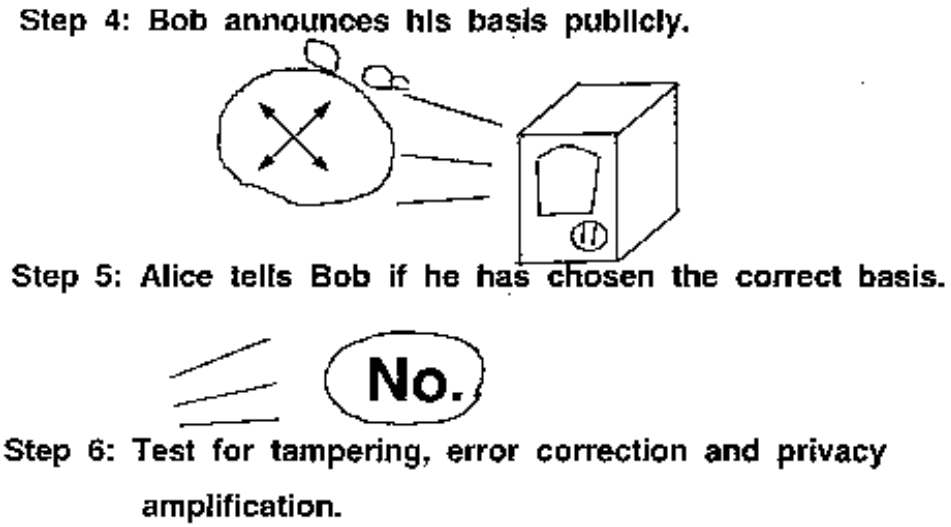
Then Bob detects in two possible bases, randomly selected.

Note that no single measurement really determines the basis Alice used….



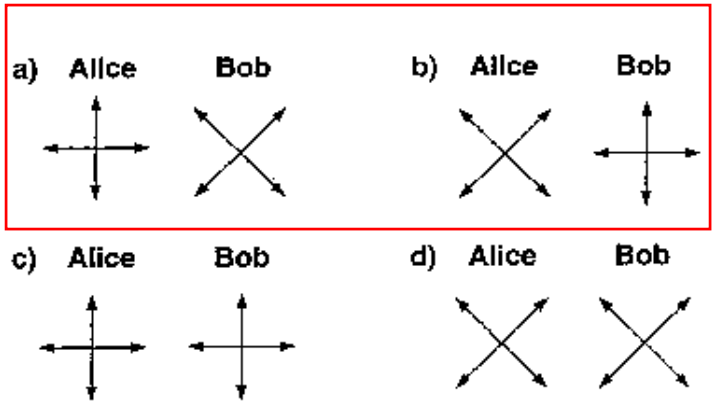
"BB84" key distribution protocol

Correlations, polarization, and security

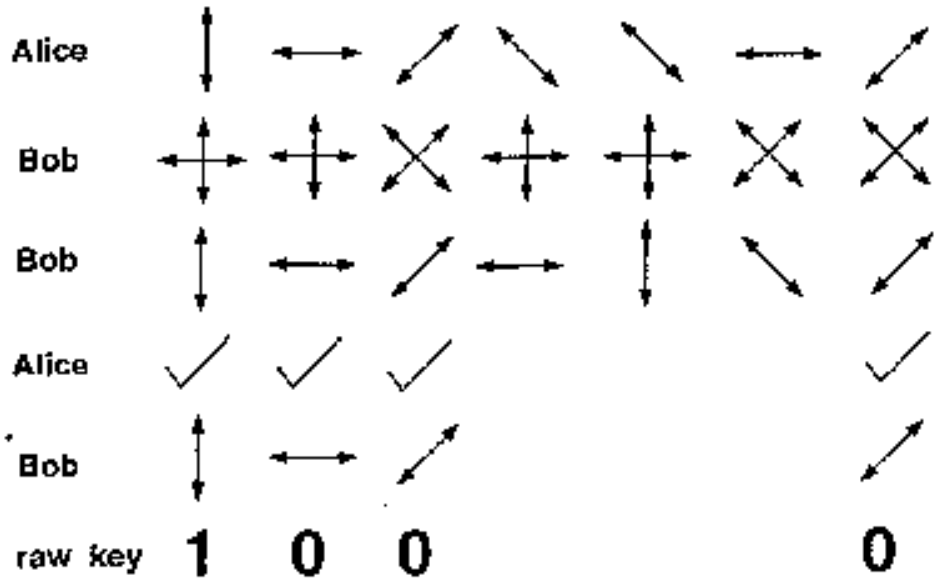


Four possibilities:

disregard



Correlations, polarization, and security



Alice and Bob can then pick a polarization direction and test it to see what the error rate is. If the error rate is too high, someone may be eavesdropping! Otherwise, they've been able to distribute a 1-time pad.

Single photon sources

Other schemes are also possible, including ones based on EPR effects using parametric down-conversion.

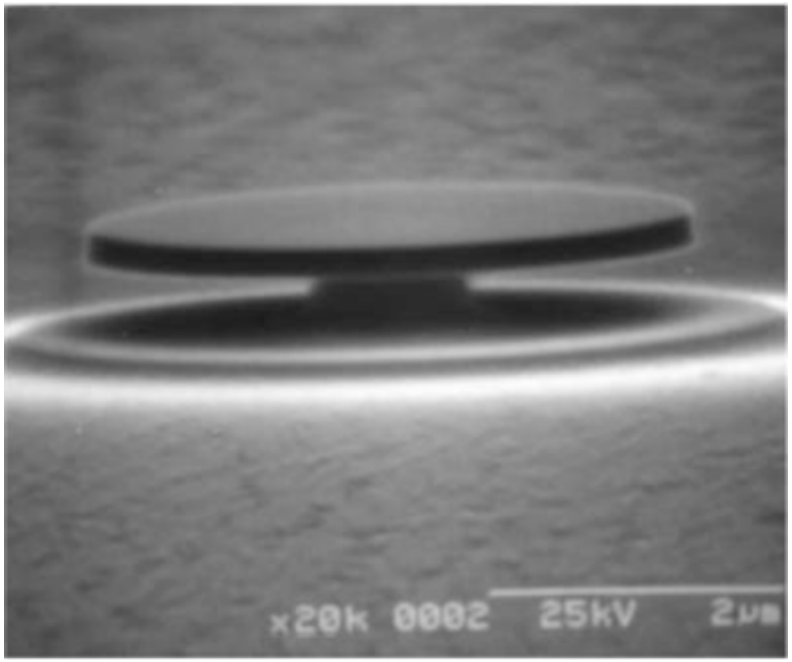
The key in all of these methods, though, is to perform measurements on single photons!

- Need single photon sources.
- Need detectors with extremely high quantum efficiencies.

Can get single photons just by attenuation, but one wants *reliable*, *reproducible* single-photon production.

The key: nanostructured sources.

Single photon sources



Michler *et al.*, **290**, 2282 (2000).

The GaAs disk resonator contains self-assembled InAs quantum dots.

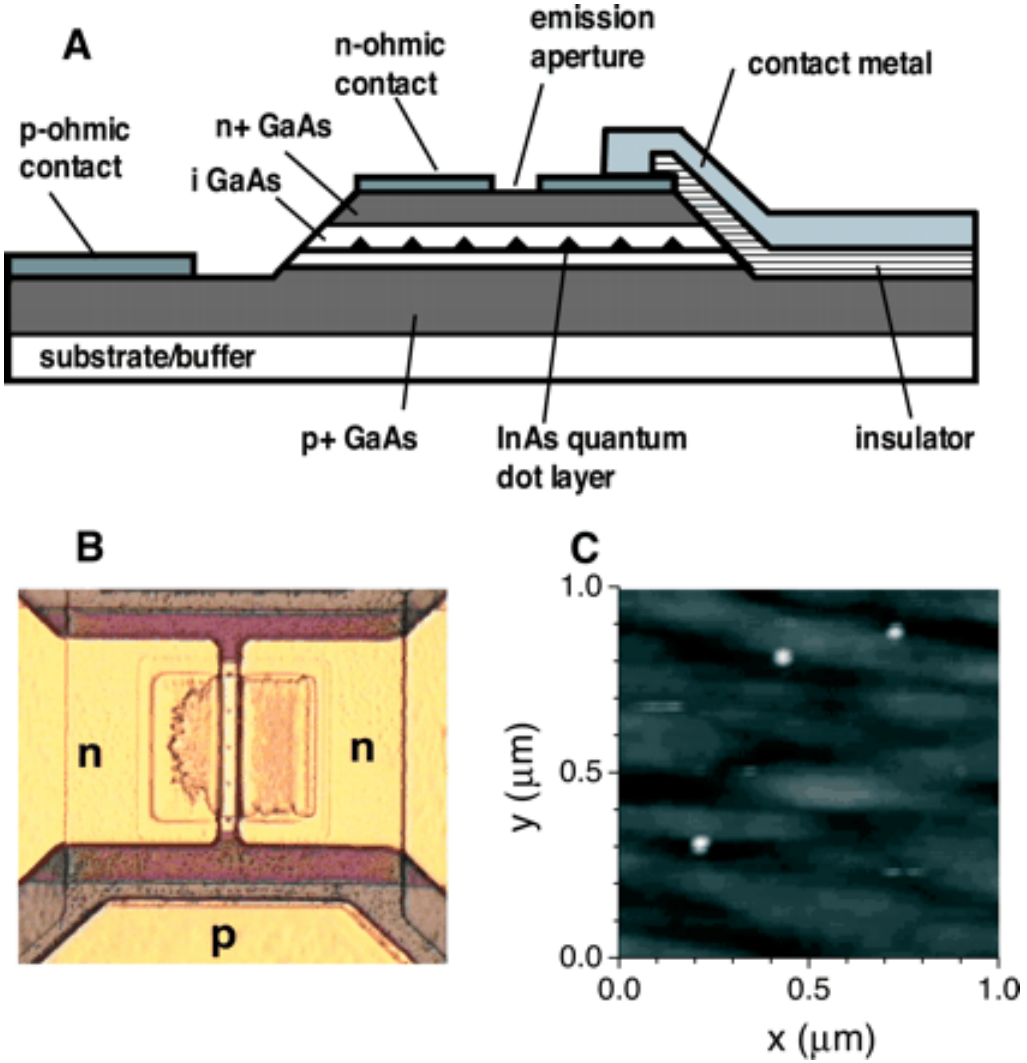
These dots are ~ 45 nm in diameter and 3 nm thick.

Quantum confinement effects make multiexciton decays very slow, and the net result is that every pulse of a mode-locked pump laser can be of the right intensity to lead to exactly one photon emitted per cycle.

Single photon sources

Electrical pumping is also possible and desirable.

Again, the basic idea is to use self-assembled quantum dots of similar sizes as those on prev. slide.



Yuan *et al.*, **295**,102 (2002).

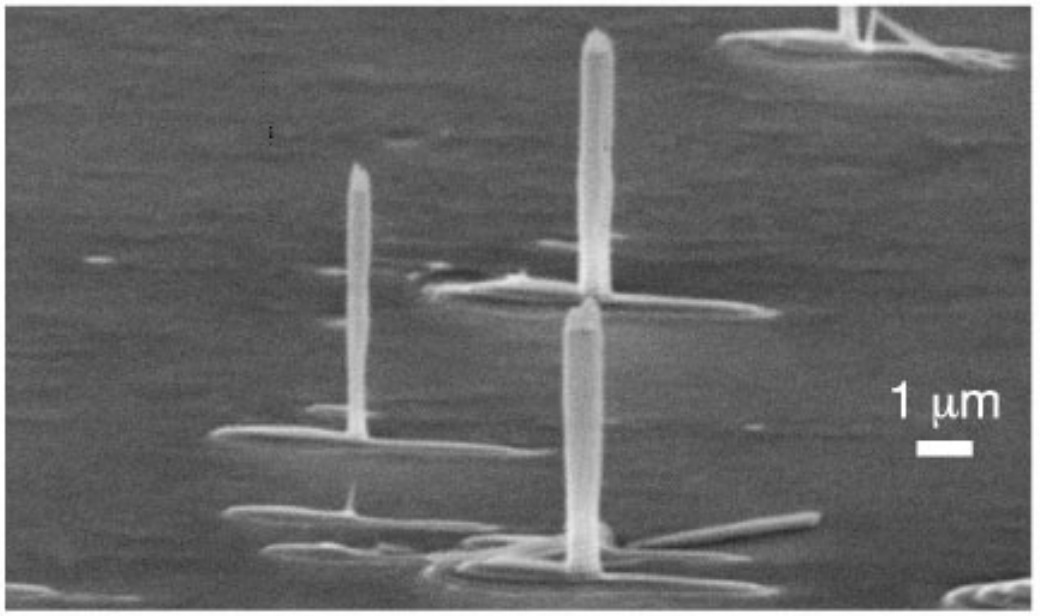
Single photon sources

Santori *et al.*, **419**, 594 (2002)

These pillars have Bragg dielectric mirrors above and below the active layer, to form the cavity.

Again, InAs dots.

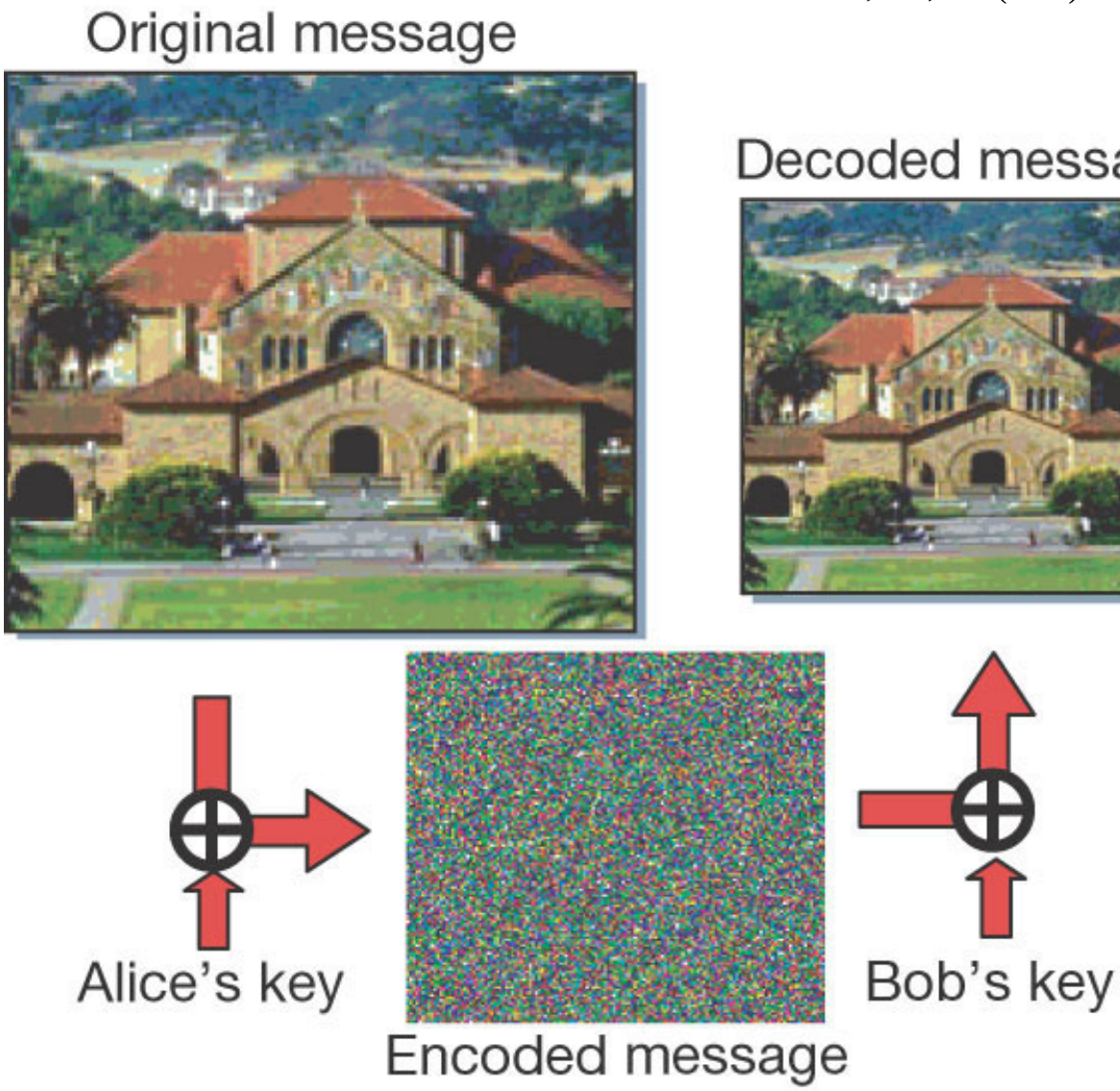
Optical pumping, with vertical etching guaranteeing only 1 or 2 dots per pillar.



This particular sample has actually been used to do exactly the key distribution scheme we talked about earlier….

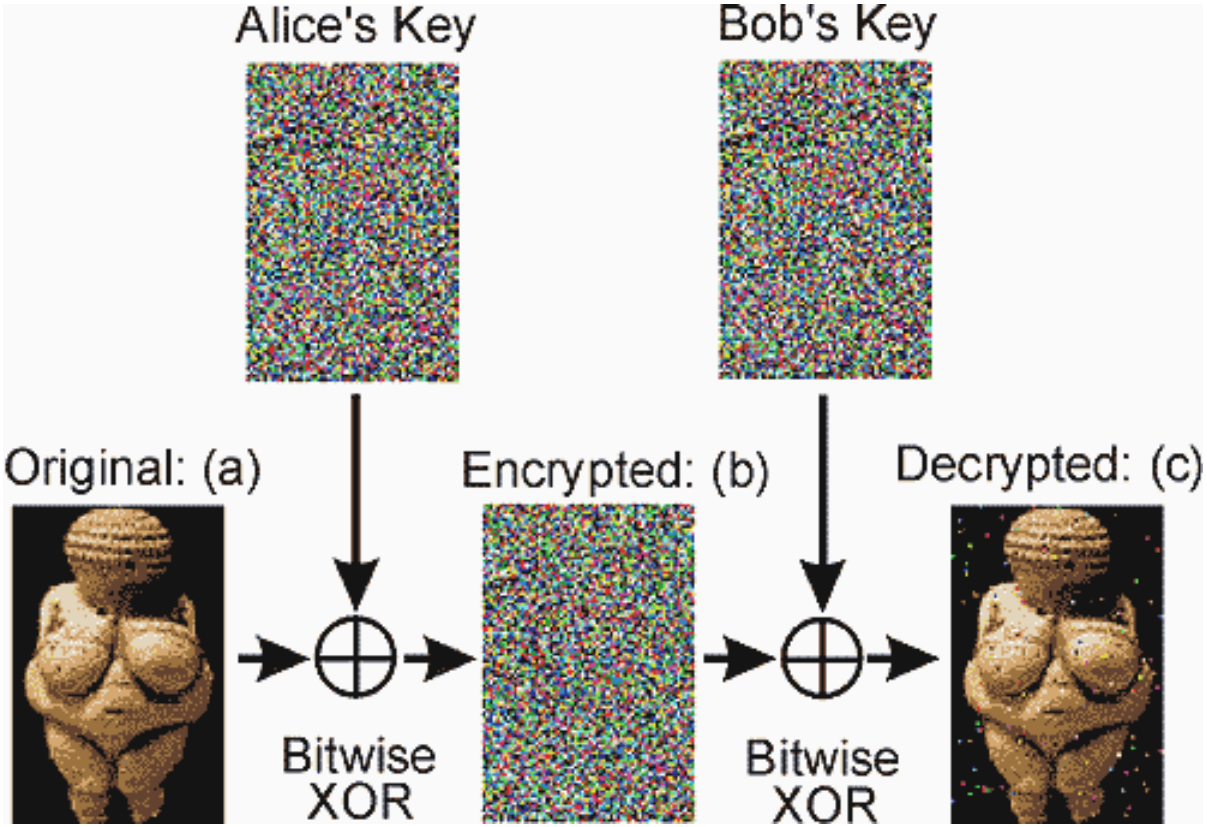
Practical demonstrations

Waks *et al.*, **420**, 762 (2002)



Practical demonstrations

Jennewain *et al.*, PRL **84**, 4729 (2000)



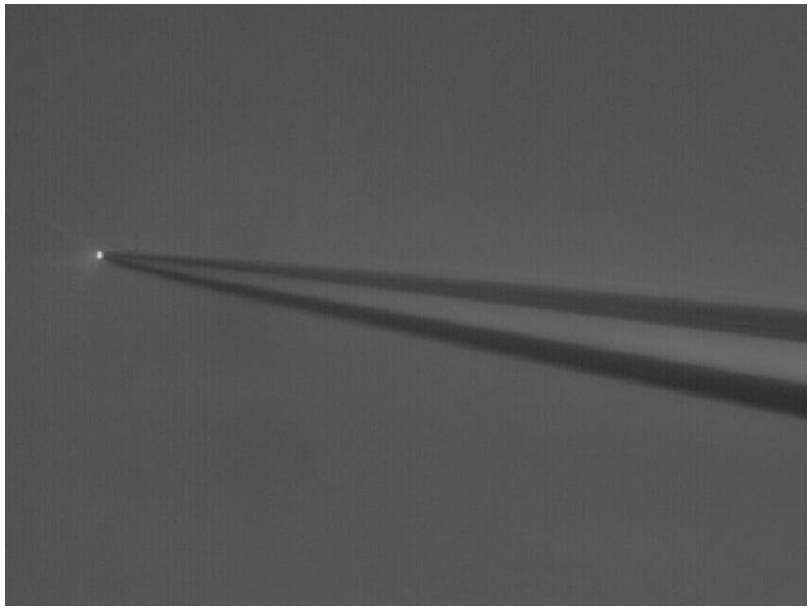
Have also demonstrated EPR-based key distribution, over 1 km of fiber.

Summary

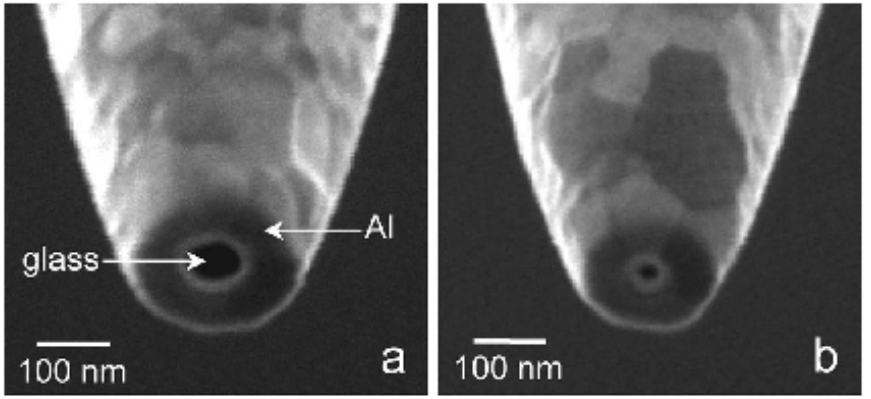
- Quantum cryptography has great potential for novel and incredibly secure communications schemes.
- Novel nanostructures are essential for simple implementations of single-photonics required for some quantum cryptography schemes.
- Real interest in controlling polarizations in fiber….

Near-field optics

- Basic physics of the near field
- Discussion of Near-field Scanning Optical Microscopy (NSOM)
- Other near-field systems



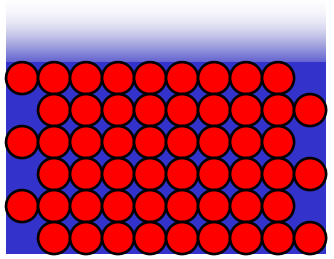
NIST NSOM group



From Hsu, MSE:R **33**, 1 (2001).

Basic idea of the near field

As we've seen in the past, it's possible for traveling wave phenomena to generate *evanescent* waves at interfaces.



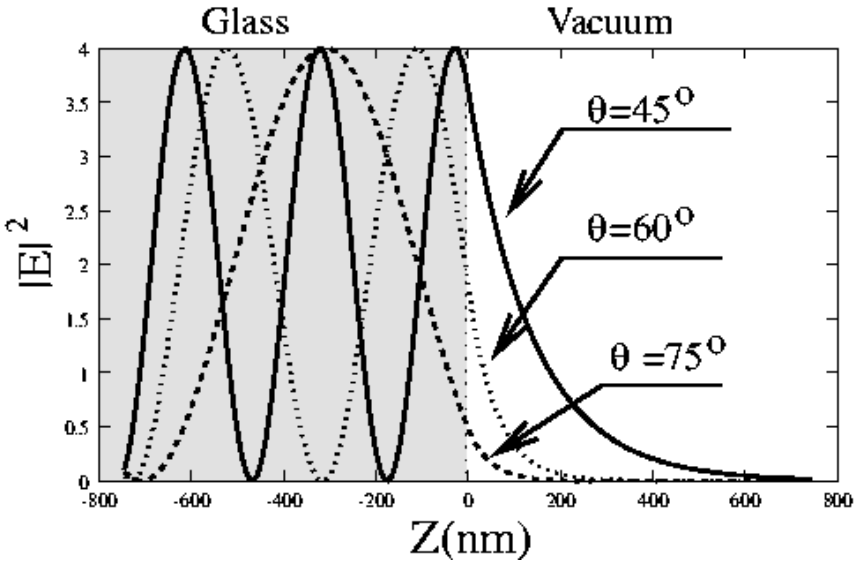
Electronic example: surface dipole as result of electronic wavefunction "spilling" beyond lattice

Optical example: exponentially decaying field strength in medium 2 in case of total internal reflection from medium 1, $n_2 < n_1$.

$$\theta_* \equiv \sin^{-1}\left(\frac{n_2}{n_1}\right)$$

$$\mathbf{E} \sim \exp(-\eta z)$$

$$\eta = \frac{\omega}{c}\left(\sin^2\theta - \sin^2\theta_*\right)^{1/2}$$

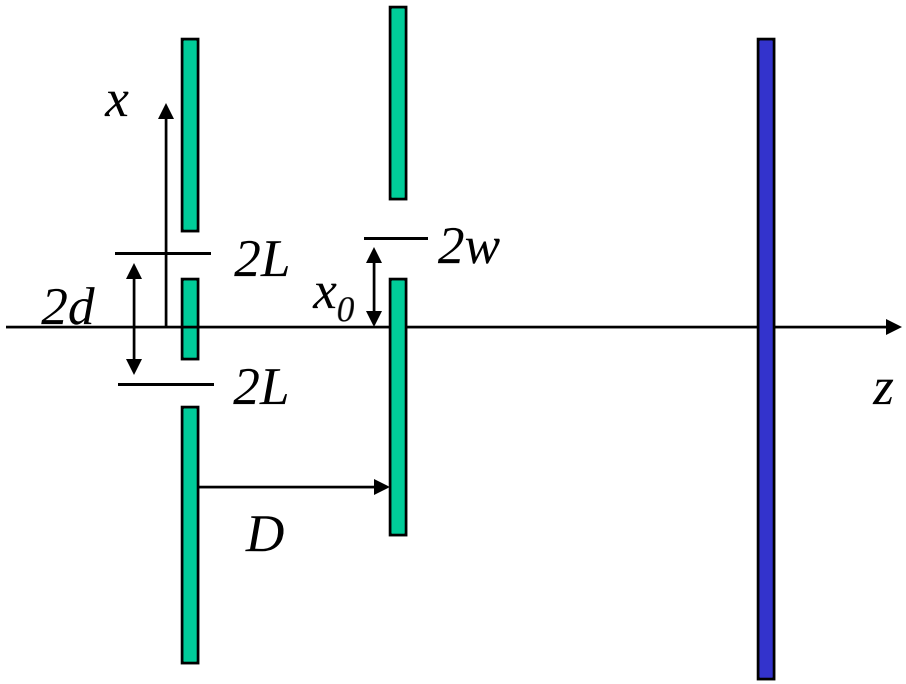


Basic idea of the near field

This exponential sort of decay happens under other circumstances. For example, trying to pass a wave through a waveguide beyond cutoff.

One limit of this is the problem of trying to pass a wave through an aperture significantly smaller than a wavelength.

We can get a sense of how this works by considering a toy model.



Illuminate screen from far -*z*. Assuming *2L* and *2w* are both subwavelength, what does the eventual detected intensity look like as a function of x_0?

Diffraction

Remember scalar diffraction theory:

$$\psi_d(\mathbf{r}) = -\frac{1}{2\pi}\oiint_S da' \; \psi_i(\mathbf{r}') \; \mathbf{n}'\cdot\left(\frac{e^{ikR}}{R}\left(ik - \frac{1}{R}\right)\hat{\mathbf{e}}_{\mathbf{R}}\right)$$

$$\mathbf{R} \equiv \mathbf{r} - \mathbf{r}'$$

In far field limit, a useful thing to compute is the Fourier transform of the illumination pattern at the screens.

For the 2-slit screen:

$$F(k_x, z=0) = \int_{d-L}^{d+L} E_0 \exp(ik_x x)dx + \int_{-d-L}^{-d+L} E_0 \exp(ik_x x)dx$$

$$= 4E_0 \cos(k_x d)\frac{\sin(k_x L)}{k_x}$$

For the 1-slit screen (our aperture):

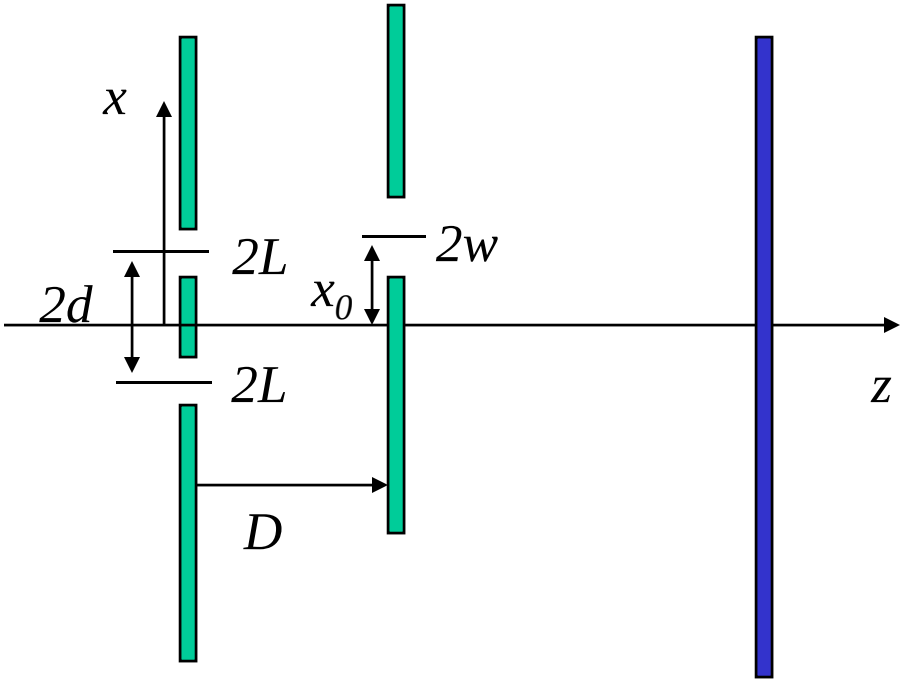
$$F(k_x, z=D) = \int_{x_0-w}^{x_0+w} E_0(x, z=D)\exp(ik_x x)dx$$

$$\approx E_0(x=x_0, z=D)\frac{\sin(k_x W)}{k_x}\exp(ik_x x_0)$$

Diffraction

Need to relate amplitude at some distance z to that at $z = 0$.

Can do this just with our definitions of Fourier transform and knowledge of propagation:



$$k_z = \sqrt{k^2 - k_x^2}$$

So, $$E_0(x,z) = \int_{-\infty}^{\infty} dk_x \left[F(k_x, z=0)\exp(ik_x x)\right]\exp(ik_z z)$$

In words: because the initial slits are sub-wavelength, the Fourier transform introduces high values of k_x (contain high lateral resolution information) components into the diffracted wave.

What happens to these components far from the initial screen?

Diffraction

$$E_0(x,z) = \int_{-\infty}^{\infty} dk_x \left[F(k_x, z=0)\exp(ik_x x)\right]\exp(ik_z z)$$

$$E_0(x, z=z_0) = \int_{-\infty}^{\infty} dk_x F(k_x, z=0)\exp(ik_x x)\exp(ik_z z_0)$$

$$= \int_{-\infty}^{\infty} dk_x F(k_x, z=0)\exp(ik_x x)\exp(i\sqrt{k^2 - k_x^2}\, z_0)$$

Note that for $k_x > \omega/c$, the quantity under the square root is *negative*, leading to exponential decay in z of those components!

As a result, for $z_0 >> \lambda$,

$$\approx \int_{-\omega/c}^{\omega/c} dk_x F(k_x, z=0)\exp(ik_x x)\exp(i\sqrt{k^2 - k_x^2}\, z_0)$$

In the far field, all the high spatial resolution information about the initial slits is lost!

Near-field aperture

By placing a near-field aperture at a distance $D << \lambda$ from the initial slits, it is possible to preserve some of that information as a modulation of the intensity eventually detected in the far field.

$$E_0(x_0, z = D) = \int_{-\infty}^{\infty} dk_x' \left[F(k_x', z = 0) \exp(ik_x' x_0) \right] \exp(i\sqrt{k^2 - k_x'^2}\, D)$$

Need to use this to plug into here:

$$F(k_x, z = D) = \int_{x_0 - w}^{x_0 + w} E_0(x, z = D) \exp(ik_x x) dx$$

To see what far field we eventually generate at $z_0 >> \lambda$.

Near-field aperture

$$E_0(x, z = z_0 >> \lambda) = \int_{-\omega/c}^{\omega/c} dk_x E_0(x = x_0, z = D) F(k_x, z = D) \exp(ik_x x) \exp(i\sqrt{k^2 - k_x^2}(z_0 - D))$$

$$= \int_{-\omega/c}^{\omega/c} dk_x \exp(ik_x x) \exp(i\sqrt{k^2 - k_x^2}(z_0 - D)) \times$$

$$\int_{-\infty}^{\infty} dk_x' \exp(ik_x' x_0) \exp(i\sqrt{k^2 - k_x'^2} D) F(k_x', z = 0) F(k_x - k_x', z = D)$$

Plugging in our expressions for the functions *F*,

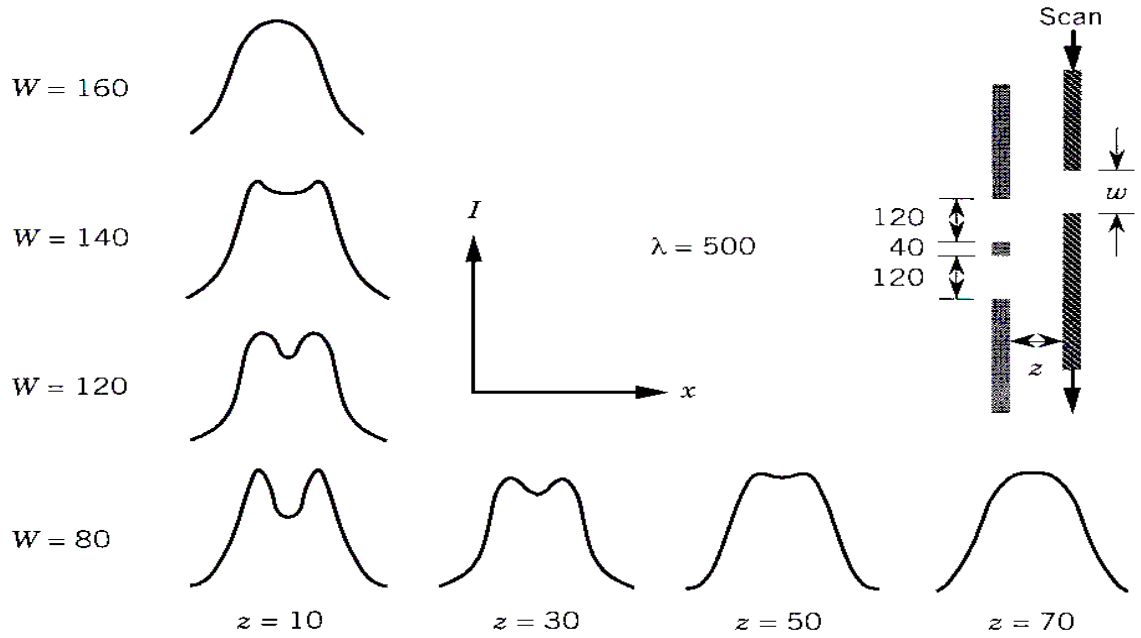
$$= \int_{-\omega/c}^{\omega/c} dk_x \exp(ik_x x) \exp(i\sqrt{k^2 - k_x^2}(z_0 - D)) \times$$

$$\int_{-\infty}^{\infty} dk_x' \exp(ik_x' x_0) \exp(i\sqrt{k^2 - k_x'^2} D) 4E_0 \cos(k_x' d) \frac{\sin(k_x' d)}{k_x'} \frac{\sin((k_x - k_x')w)}{k_x - k_x'}$$

Near-field aperture results

Point is that near-field aperture couples those short-range high resolution components near the initial screen into the final intensity that's measured:

Paesler and Moyer, *Near Field Optics*



- Resolution is best when aperture is smallest and closest to initial screen.
- Trade-off: smaller aperture leads to less overall power coming in.

Near-field scanning optical microscopy

One big application of these ideas is NSOM.

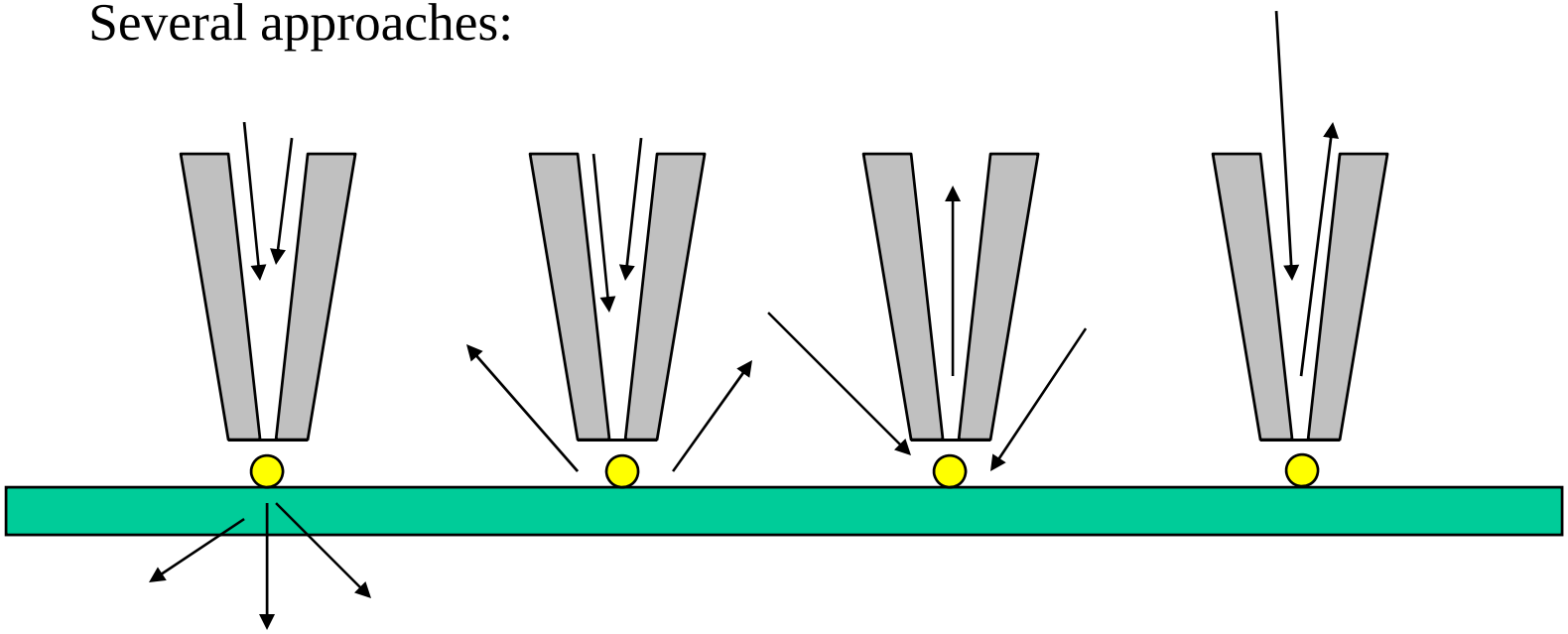
Interestingly, using subwavelength apertures for “ultramicroscopy” was first suggested in 1928 (!) by E.H. Synge, and again independently in 1956 by John A. O’Keefe.

Technology required to make this work:

- Sensitive photodetectors
- Effective optical waveguides
- Positioning control at the nm level
- Material control at the nm level (aperture, tip roughness)

NSOM

Several approaches:



transmission illumination

reflection illumination

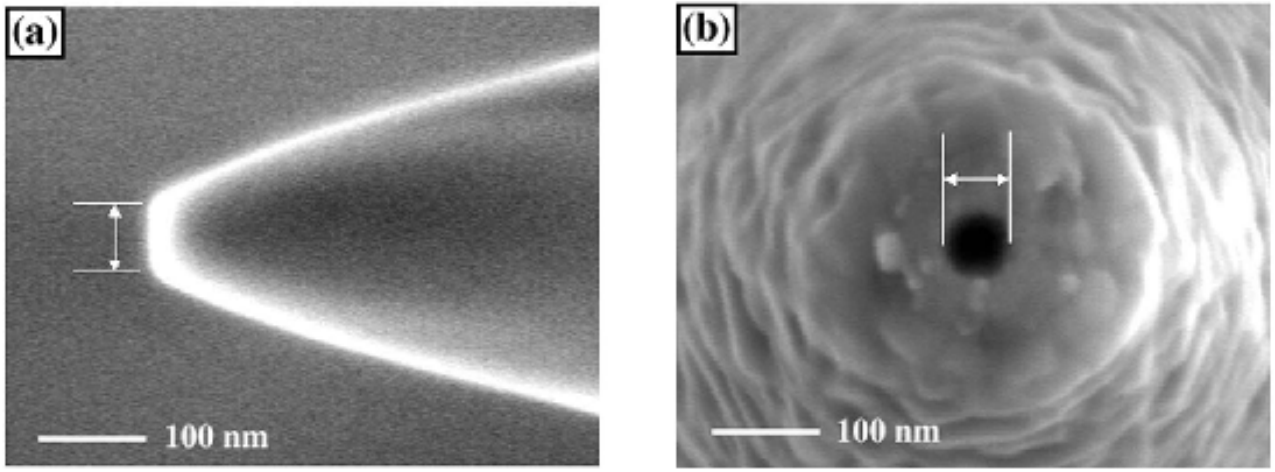
reflection collection

illumination collection

How are these implemented in practice?

NSOM tips

From Hsu, MSE:R **33**, 1 (2001).



- Drawn optical fibers.
- Coated by aluminum in thermal evaporator at an angle (to leave uncoated aperture).
- Metal needed to limit optical losses out the sides of the tapering fiber.
- Aluminum used because of ease, and minimum skin-depth in the visible and IR.

NSOM position sensing

Sensing distance between the tip and the sample is critical to maintain spacing and avoid crashing the tip.

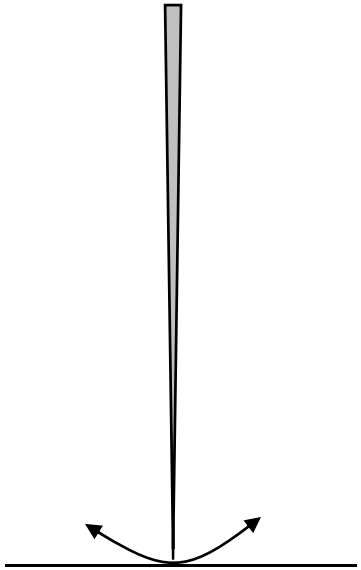
Most common approach uses *lateral force feedback*.

Tip is fixtured in a piezo-driven holder.

Tip is vibrated laterally at its natural mechanical resonance frequency.

As tip approaches surface, attractive tip-surface interactions (Van der Waals) shift the natural frequency and damp the amplitude.

By doing *z* feedback to maintain constant amplitude, topography can be measured with *z* resolution better than 0.1 nm.



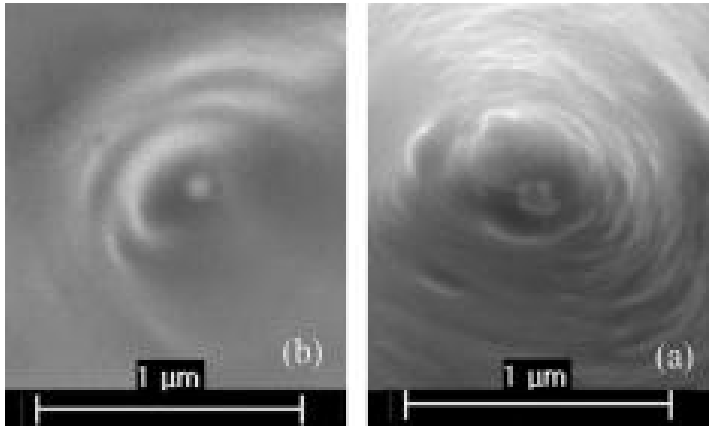
Complications

Quantitatively analyzing NSOM information can be very challenging:

- Microscopic configuration of metal and aperture are generally not well-known.
- In *I* mode, most power gets reflected back up the fiber, leading to large dissipation in the Al film: can get melting.
- Proximity between tip and sample can change properties of sample (*e.g.* shorter fluorescence lifetimes).
- Can get unusual optical interactions between tip and certain samples (coupling into prop. modes of sample).
- Polarization effects can be very complicated in the near-field in realistic systems - far-field selection rules may no longer hold.
- Resolution is tricky to determine, too - nonlinear distance effects.

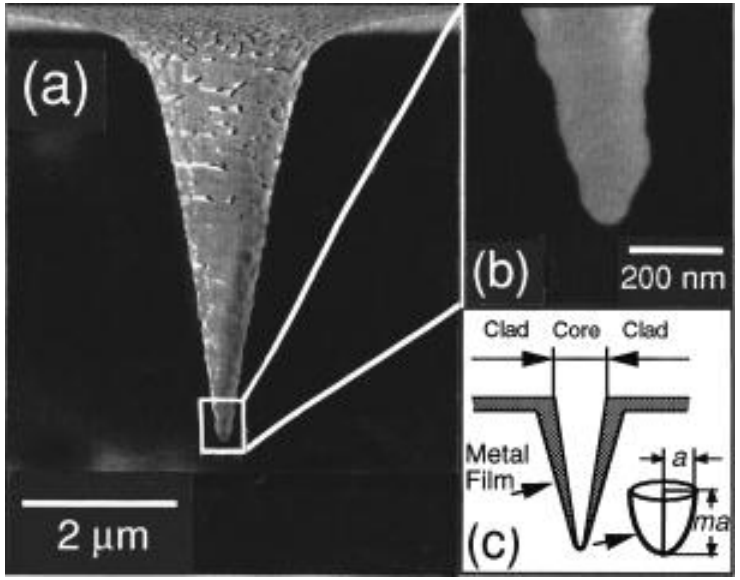
Exotic tips

Trying to attach gold nanoparticles to tip to use Au plasmon resonance to enhance coupling.



Sqalli *et al*., APL **76**, 2134 (2000)

Similar idea - Au film designed to localize a plasmon resonance at the tip.



Ashino *et al*., APL **72**, 1299 (1998)

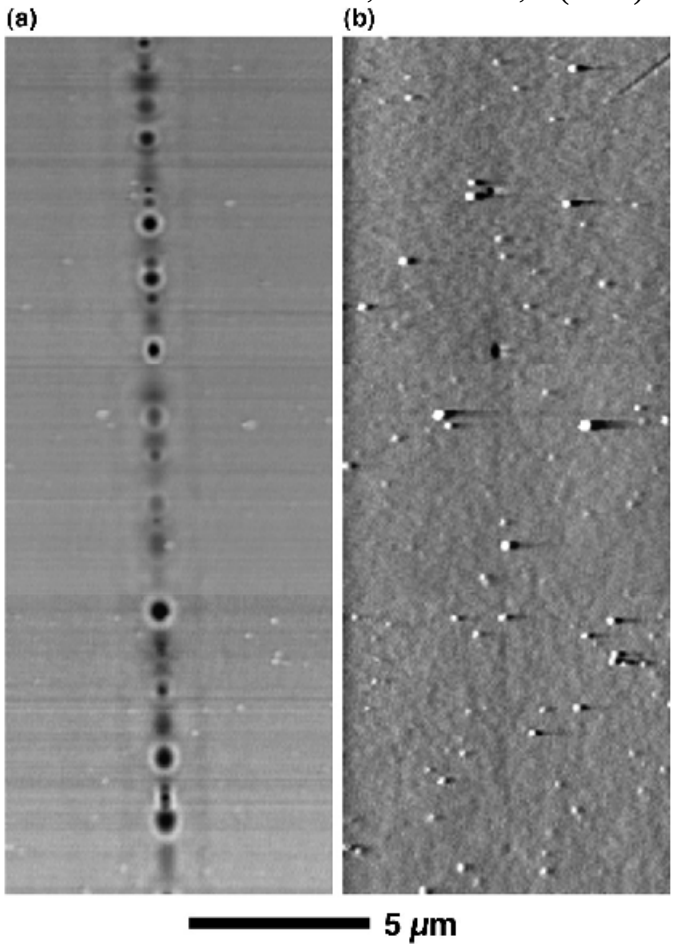
Applications: semiconductors / materials characterization

NSOM can be great for looking at defects in insulating / intrinsic material that would be extremely difficult to spot with other methods.

Example: Two $SrTiO_3$ wafers bonded together and polished.

Bicrystal defects / stacking faults show up dramatically in NSOM image because of weird local behavior of substrate dielectric response.

From Hsu, MSE:R **33**, 1 (2001).

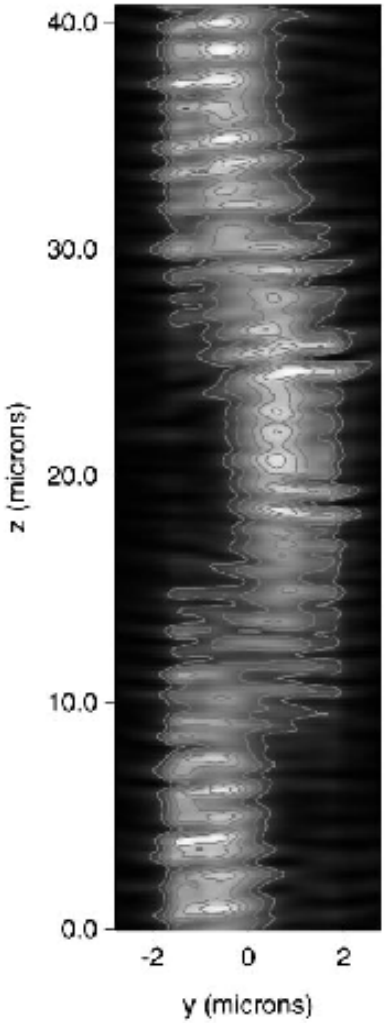


Applications: photonic systems

In collection mode, can be directly sensitive to evanescent fields.

Example: can see local intensity in a waveguide directly.

Notice beating between two modes (TE_0 and TE_1) of slightly differing propagation constants.

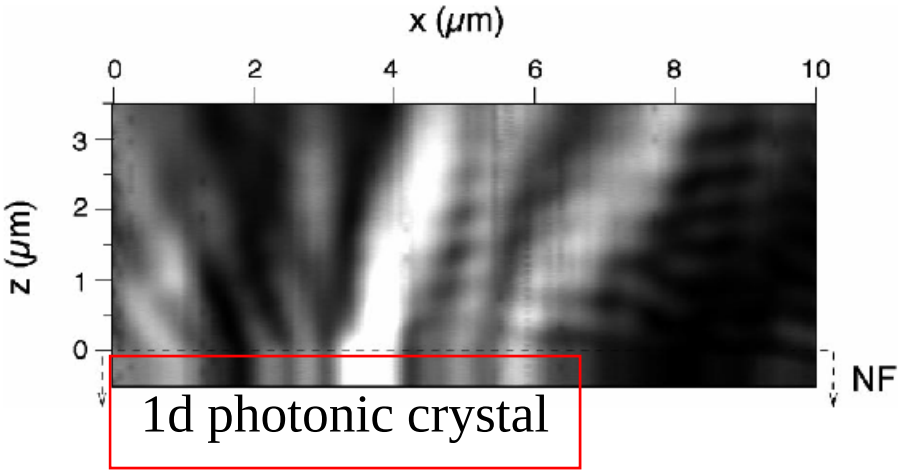


From Hsu, MSE:R **33**, 1 (2001).

Applications: photonic systems

Similarly, can probe photonic bandgap systems' properties.

Note that the sharp features here do *not* decay exponentially with distance.



From Hsu, MSE:R **33**, 1 (2001).

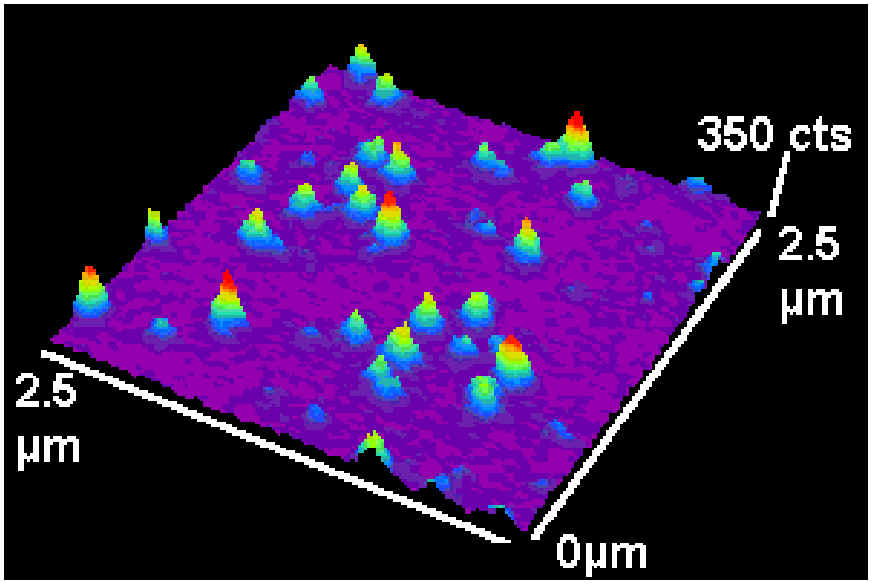
This is a result of the coherent nature of the optical scattering going on in the PBG material.

Applications: chemistry and biology

Paul Barbara, UT

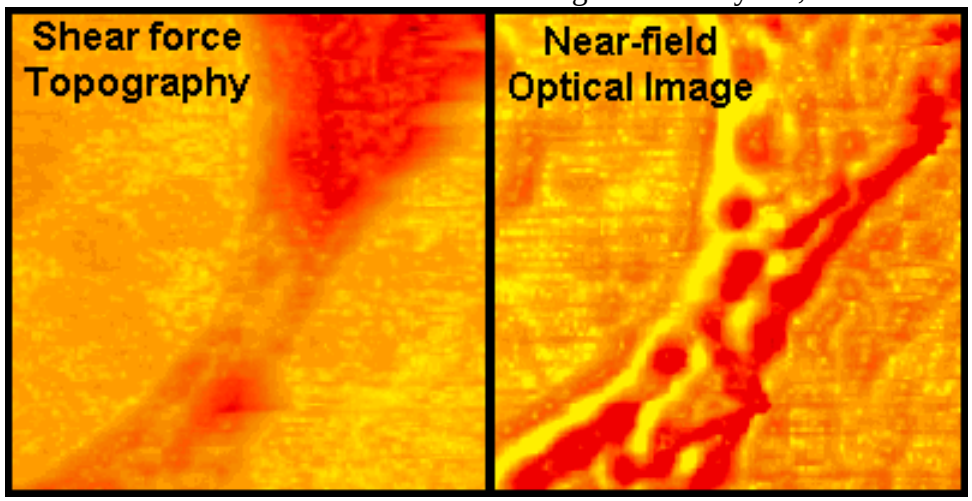
Single-molecule spectroscopy!

Image at right is of spatially resolved counts in illumination mode from individual dye molecules distributed in PMMA.



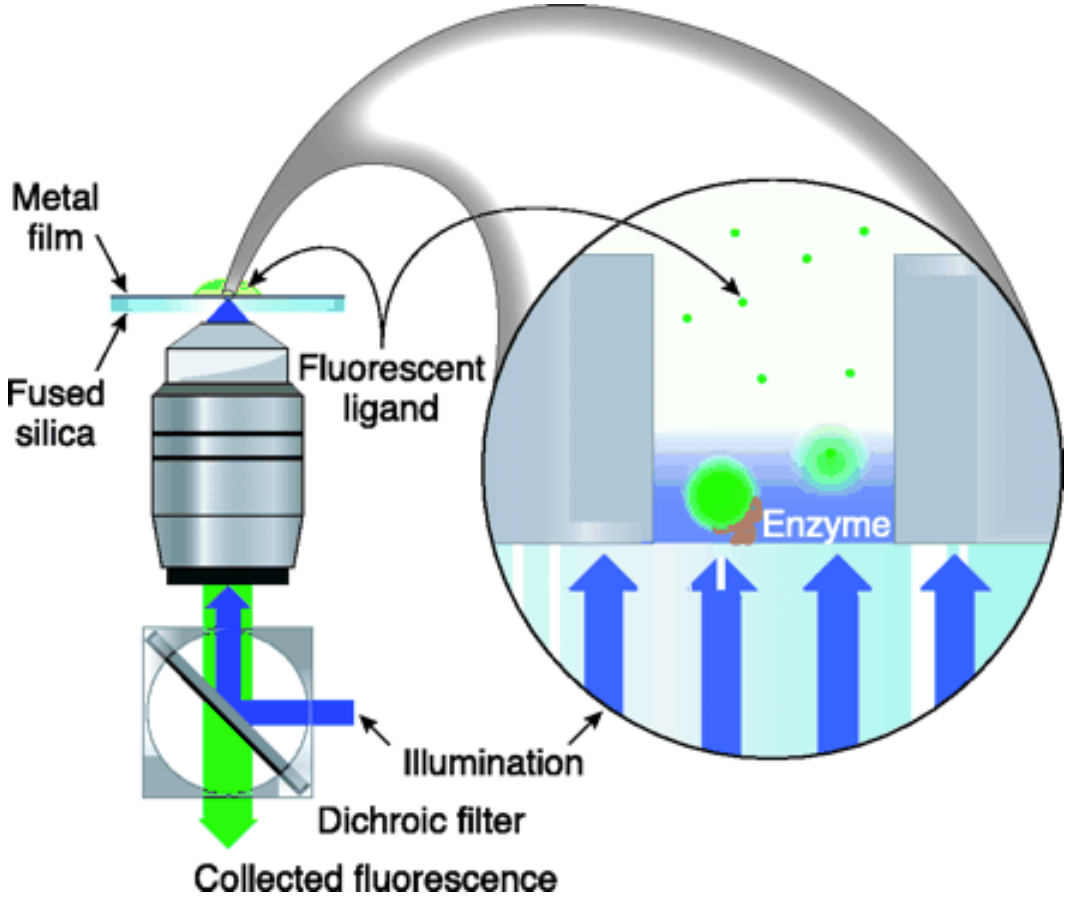
Ragona and Haydon, IA State

Also of great use in biological systems - provides qualitative contrast without necessarily staining samples.



Another near-field example

Levene *et al.*, Science **299**, 682 (2003)



Apertures are 50 nm diameter holes in Al film on coverslip.

Enables measurements of enzyme activity in 10^{-21} L effective volumes!

Summary

- Near-field optics works because near-fields can contain high lateral spatial resolution information that is exponentially attenuated at distances much longer than a wavelength.
- Key is that subwavelength aperture transmission to a far field detector can be modulated by these components.
- NSOM is the resulting technology - powerful and impressive, but quantitative analysis can be very complicated.
- Has been applied in a number of disciplines, including physics, electrical engineering, materials science, chemistry, biology.
- Enabling technologies: optical fibers, metal film fabrication, and other scanned probe microscopies.

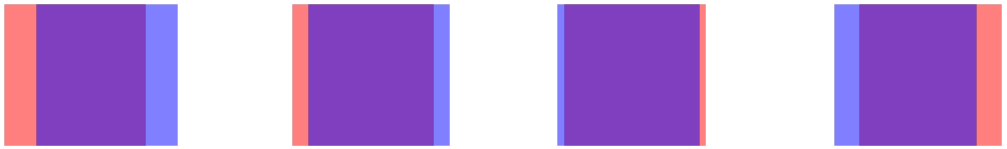
Plasmons, surface enhancements, and left-handed media

Today we're going to discuss interactions of electromagnetic waves with highly structured conductors. There are several distinct exciting areas of development that spring from this topic:

- Plasmons and nanoparticles
- Surface enhanced spectroscopies
- "Plasmonics" as optical engineering
- Metamaterials with exotic optical properties ("left-handed" media?)

Simple plasmons

Consider what happens if the electrons in a bulk material are displaced from equilibrium:



Ignoring dissipation, classically we would have

$$m\frac{d^2\mathbf{r}}{dt^2} = -e\mathbf{E}$$

Use Gauss' law to find electric field inside box that provides restoring force…

$$\mathbf{E} = \frac{ne}{\varepsilon_0}\mathbf{r}$$

Assume harmonic time variation gives:

$$\omega_p^2 = \frac{ne^2}{m\varepsilon_0}$$

This is the frequency for bulk ($q = 0$) charge density oscillations: plasmons

These excitations survive in quantum mechanical treatments, and are observed spectroscopically.

Simple plasmons

It's possible for plasmons in bulk systems to have finite wavevectors, also (as opposed to $q = 0$ uniform translations of all the electrons in the material).

For long wavelengths, the quantum mechanical result for the dispersion relation is:

$$\omega^2 = \omega_p^2 + \frac{6}{5}\frac{E_F q^2}{m}$$

Plasmons with frequencies less than the plasma frequency can't propagate (imaginary q).

This relation was again calculated by finding the dielectric response of the conducting medium.

As you might imagine, electrons in small particles (rather than bulk systems) will have their own particular oscillatory normal modes.

These will depend in detail on the geometry and electron density of the nanoparticles, and will manifest themselves in the dielectric reponse.

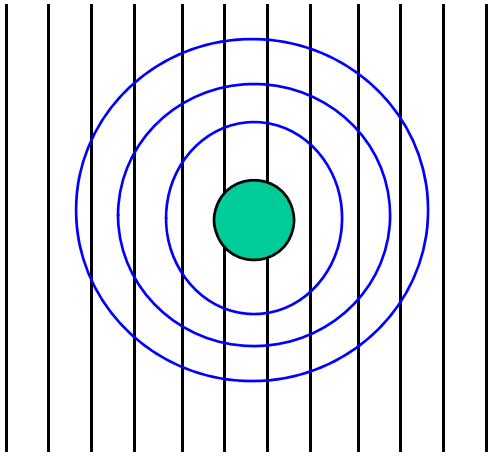
Light scattering by small particles: Mie theory

One way of probing these modes in small particles is through light scattering.

Mie (1908) developed a general treatment for (steady state) classical scattering of radiation off a homogeneous, isotropic, linear sphere with essentially arbitrary dielectric response.

Unfortunately, Mie theory is pretty gory, using vector spherical harmonics. Here I'm just going to outline the general idea.

Start with an incident plane wave, and assume that there is an internal field and a scattered field.



Mie theory

Define the vector spherical harmonics:

$$\mathbf{N}^{a}_{\ell n} = \frac{1}{k}\nabla\times(\nabla\times(\mathbf{r}f^{a}_{n\ell})), \qquad f^{a}_{n\ell} = z^{a}_{n}(kr)P^{\ell}_{n}(\cos\theta)\exp(i\ell\phi)$$

$$\mathbf{M}^{a}_{n\ell} = \nabla\times(\mathbf{r}f^{a}_{n\ell}), \qquad z^{a}_{n}(kr) = j_{n}(kr), h^{(1)}_{n}(kr), h^{(2)}_{n}(kr)$$

Can then expand fields in terms of these.

$$\mathbf{E}_{s} = \sum_{n} E_{n}[ia_{n}\mathbf{N}^{(3)}_{e\ell n} - b_{n}\mathbf{M}^{(3)}_{o\ell n}]$$

$$\mathbf{E}_{\text{int}} = \sum_{n} E_{n}[c_{n}\mathbf{M}^{(1)}_{o\ell n} - id_{n}\mathbf{N}^{(1)}_{e\ell n}]$$

$$\mathbf{E}_{i} = E_{0}\sum_{n} i^{n}\frac{2n+1}{n(n+1)}\left[\mathbf{M}^{(1)}_{o\ell n} - i\mathbf{N}^{(1)}_{e\ell n}\right]$$

As you might think, the variable *n* is called the mode order, and gives the radial dependence, while ℓ is related to the number of field maxima and minima in the angular dependence.

Mie theory

Those coefficients a_n, b_n, c_n, d_n are called the Mie coefficients, and are defined in terms of the Bessel / Hankel functions and their derivatives.

The dependence on sample properties comes down to two parameters:

$$x \equiv \frac{2\pi a}{\lambda}$$ "size parameter"

$$n(\omega) \equiv \sqrt{\frac{\varepsilon(\omega)}{\varepsilon_0}} = \sqrt{\kappa(\omega)}$$ Index of scattering medium.

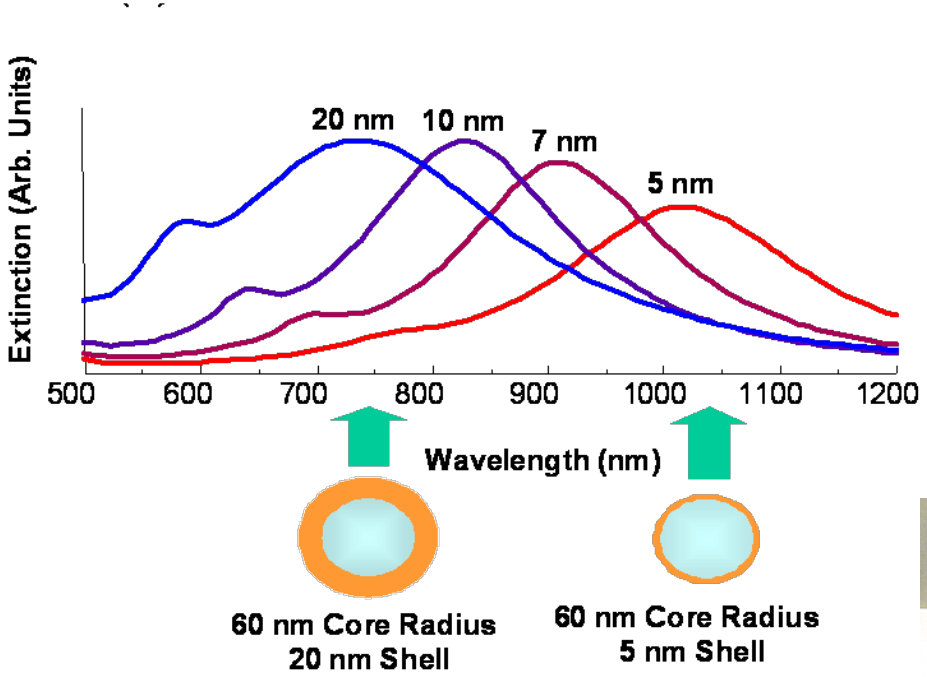
When the expansion coefficients a and c have poles, there are resonances in the scattering.

Note that the incident beam of light has picked out a particular direction, and the polarization of the incident field can lead to anisotropy in the internal and scattered fields.

Plasmons in nanoparticles

- It's possible to do variations of Mie theory to worry about the optical properties of nanoparticles, even those that are not spherical.
- The most commonly examined nanoparticles are gold clusters.
- Varying particle geometry allows tuning of optical resonances.
- For particles with broken symmetries, orientation with respect to incident light can lead to dramatic changes in amplitudes, frequencies of optical resonances.
- Plasmon resonances, associated with large time varying polarizations, can lead to very strong optical scattering coincident with extremely strong local electric fields.
- Local electric fields due to these plasmon effects are *evanescent*, and so have very short ranges.

Plasmons in nanoparticles (Halas group)

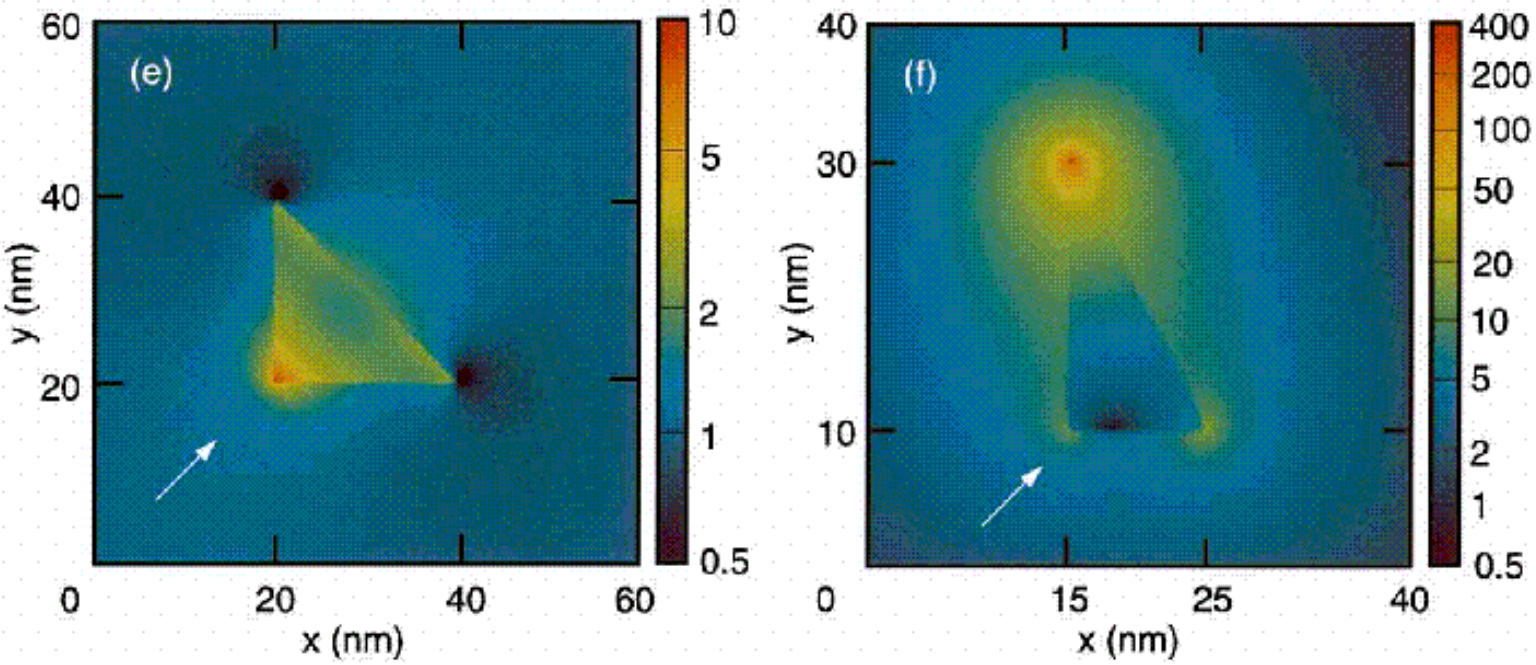


Au shell around SiO_2 core.

Tuning core-shell ratio can strongly adjust optical properties.

Local fields in nonsymmetric particles

Kottman *et al.*, CPL **341**, 1 (2001).



Calculation shows local electric fields due to 400 nm incident light scattering off conducting (plasmon resonating) triangular prism particles.

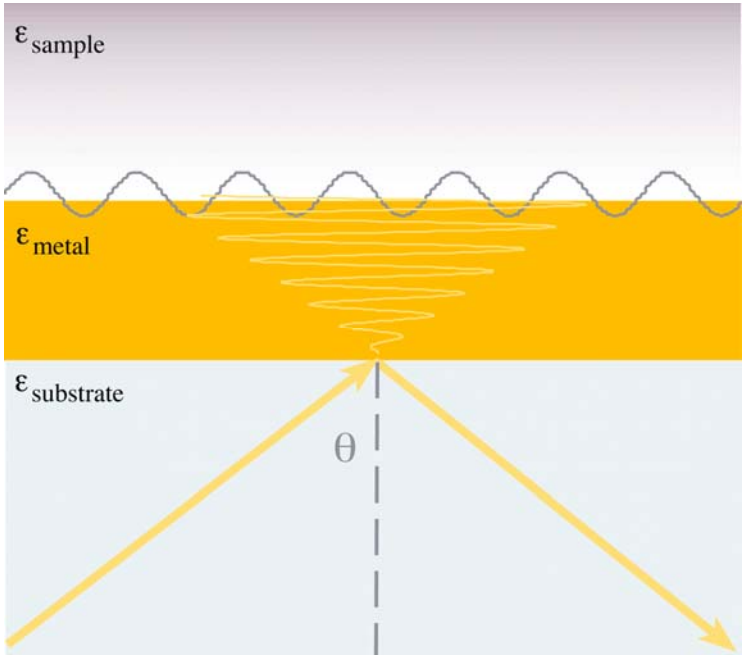
Note the logarithmic color scales.

- Very sharp features lead to extremely large ***E*** fields localized at the nanometer scale near particles like this.

Surface plasmons

We had gone from 3d (bulk) plasmons to 0d (strongly confined nanoparticle) plasmons.

What about 2d? This is actually the most widely used geometry.



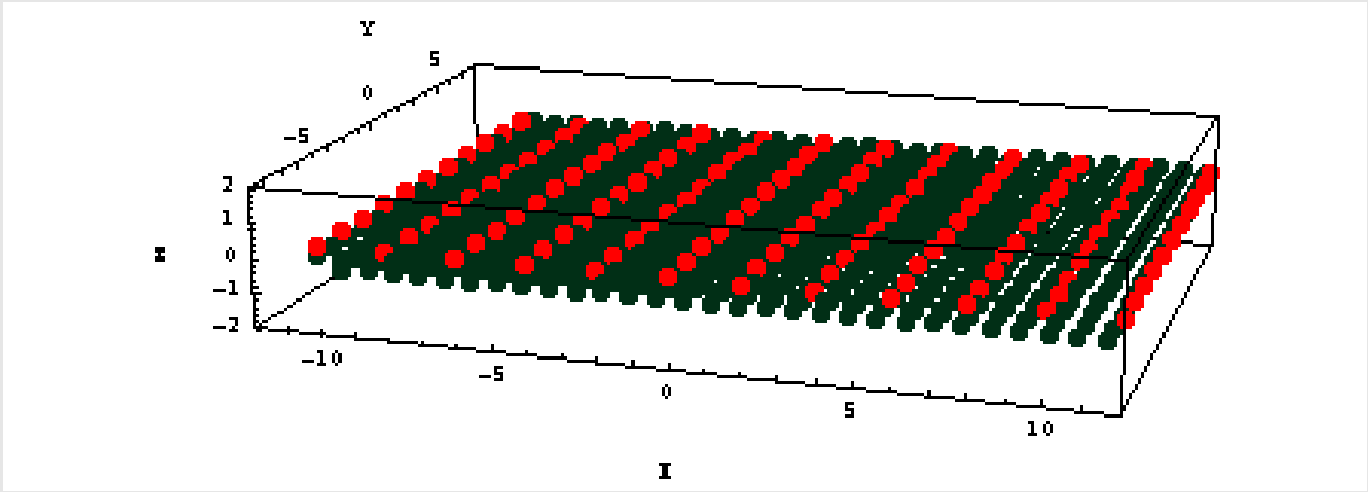
Campbell group, U. Washington

Use evanescent field from total internal reflection at the edge of a prism to excite surface plasmons in metal film.

Then make some interesting use out of the evanescent field from those surface plasmons that extends into the few nm above the metal.

Surface plasmons

Surface Plasmon Mode



Oscillating conduction electrons are shown in red, the fixed substrate of positive charge in green. The surface plasmon mode is driven by a coupled, external, electromagnetic field.

The surface plasmon has a nonzero q determined in part by the wavelength of the totally internally reflected radiation.

Surface plasmon polaritons

Some jargon to know:

Polariton: when an acoustic excitation in a solid (i.e. an optical phonon) couples strongly with radiation, the coupled excitation (light + big induced polarization due to phonon-related dielectric response) is called a polariton.

Since plasmons are somewhat analogous to sound waves in the electron gas, a strongly coupled excitation of (light + a big induced polarization due to plasmon resonance) is often called a *plasmon polariton*.

Surface enhanced spectroscopies

One extremely interesting and important phenomenon known to be related to plasmons are *surface enhanced spectroscopies*.

Discovered ~ 1974. The Raman signal from pyridine on a silver film was found to be ~ 10^6 times larger than what one would expect, based on the Raman signal for the molecule in solution.

The record enhancement inferred for SERS is now ~ 10^{14} (!!).

- Important because it allows analytical sensors of amazing sensitivity, able to perform discriminating measurements at phenomenally low analyte concentrations.
- Literally hundreds of papers published on this, particularly in the last 10 years.
- Big applications: gene sequencing, chemical analysis, explosives detection, pollution sensing

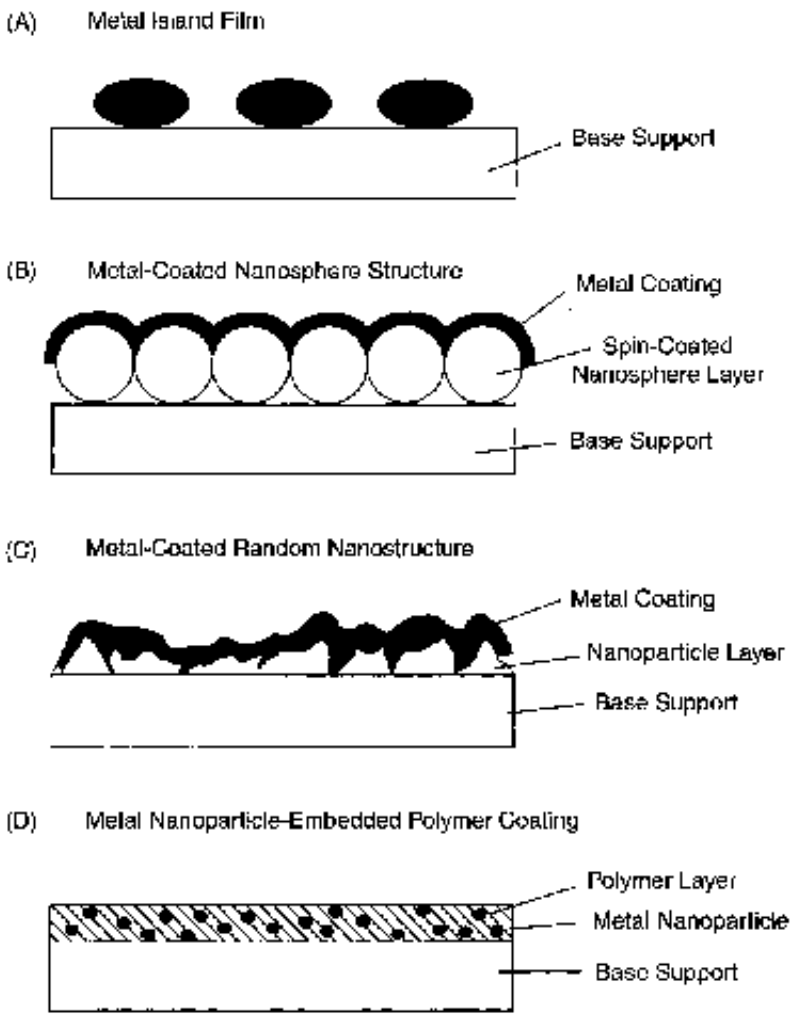
Surface enhanced spectroscopies

Not all the details of these processes are understood.

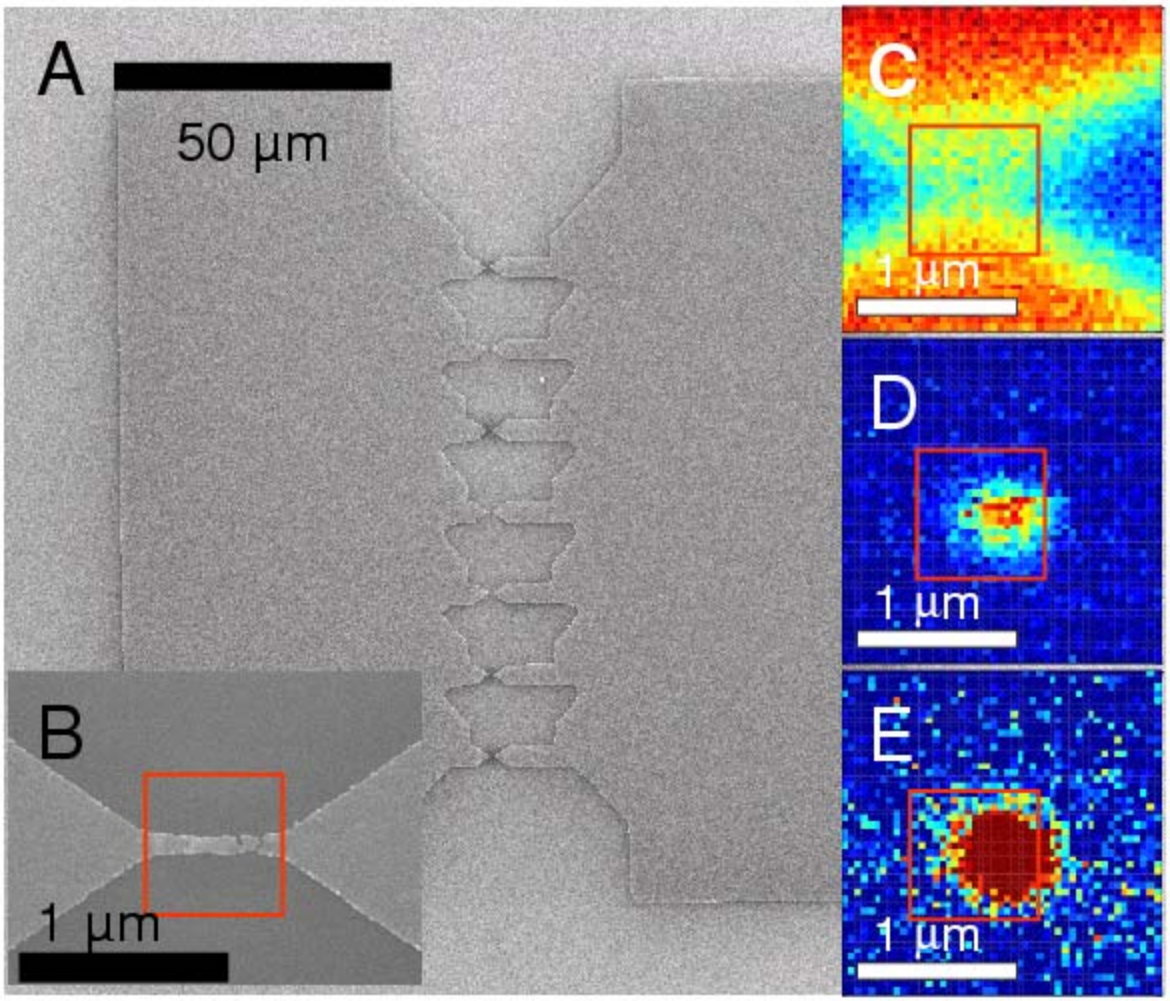
However, the basic idea is that roughened metal surfaces lead to localized "hot spots" of extremely intense electric field.

Raman scattering scales like *intensity*^{2}, so a 1000-fold increase in electric field compared to the incident radiation would lead to a 10^{12} increase in Raman signal, all other things being equal.

A number of approaches for getting big signals.

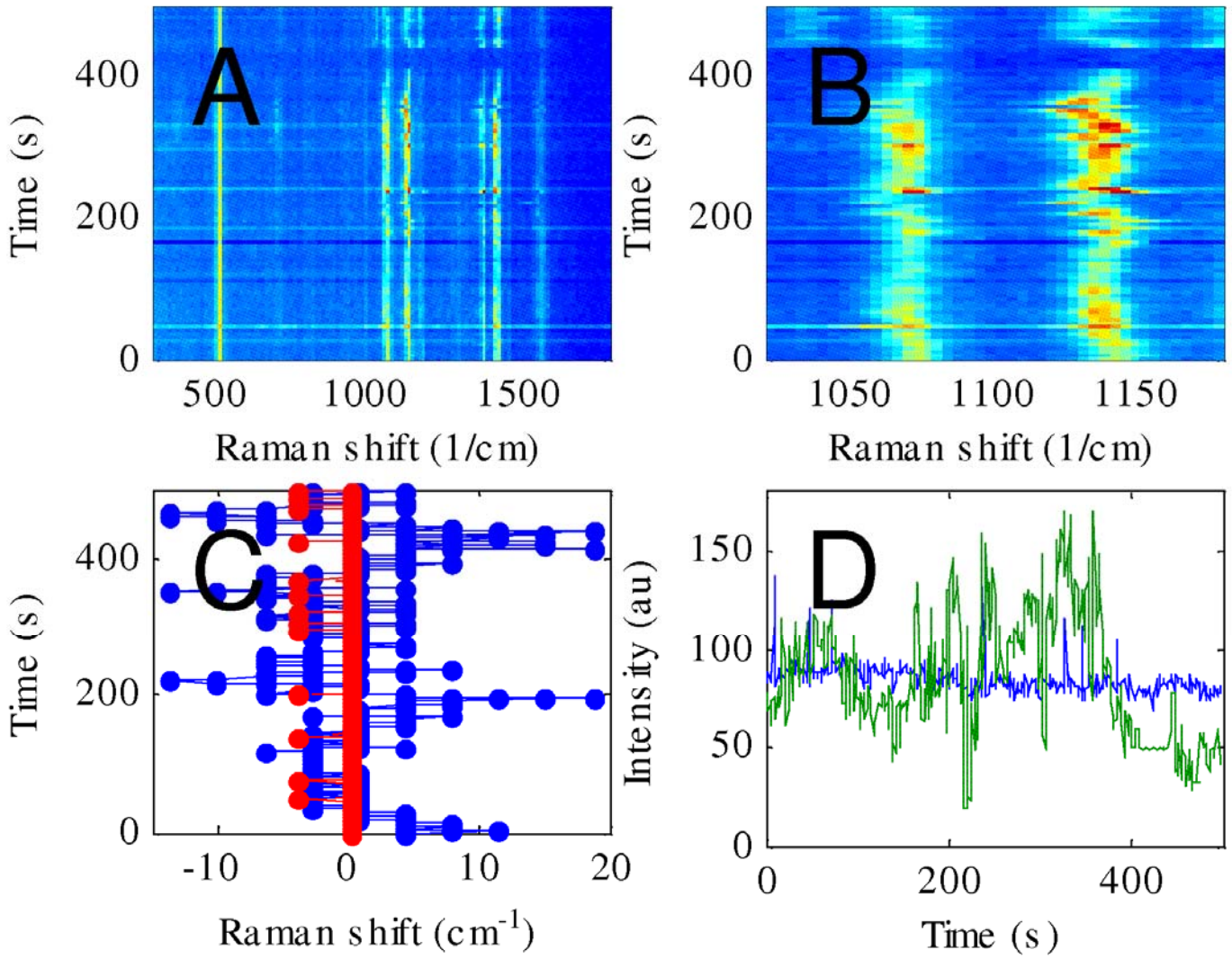


Surface enhanced spectroscopies



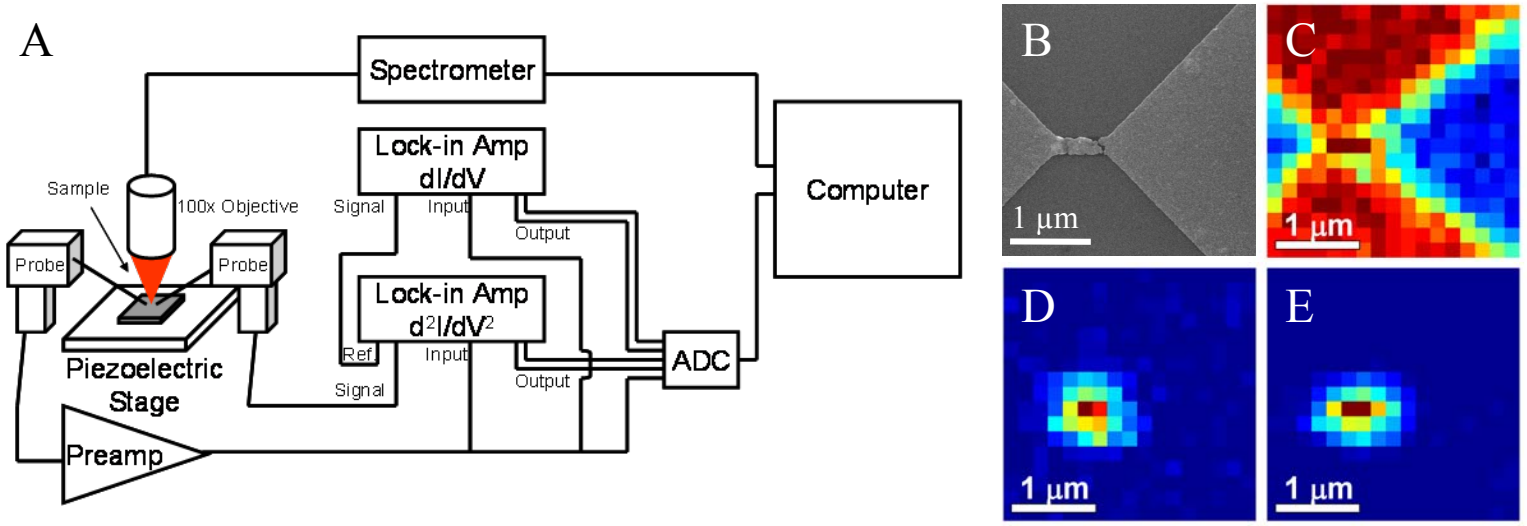
Ward *et al.*, Nano Lett. **7**, 1396 (2007)

Surface enhanced spectroscopies



Ward *et al.*, Nano Lett. **7**, 1396 (2007)

Transport + SERS

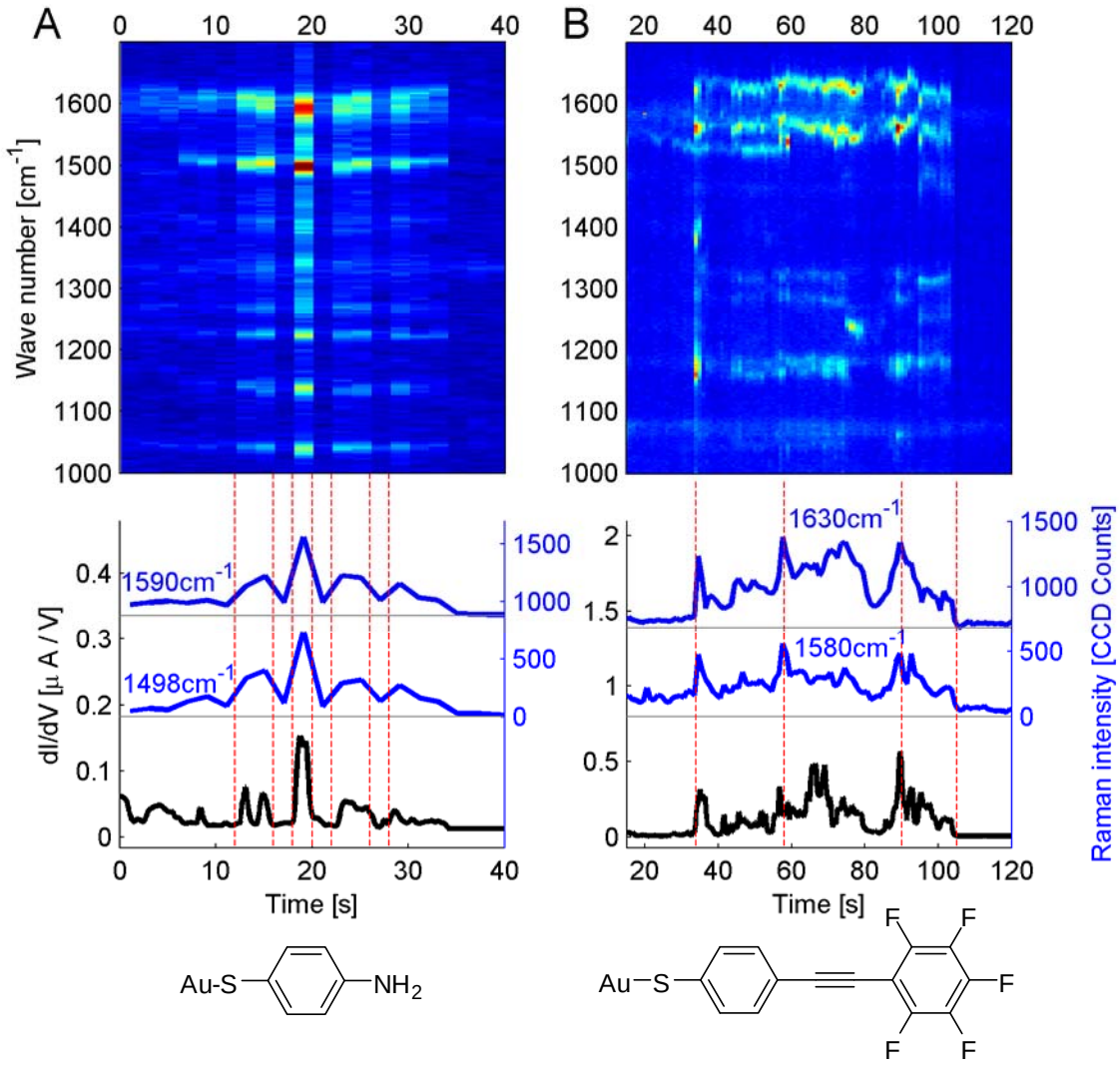


- Simultaneous transport + SERS can quantify Raman sensitivity.
- Conduction via tunneling – probes ***very*** small volume ~ 1 molecule.

Ward *et al.*, Nano Lett. **8**, in press (2008)

Transport + SERS

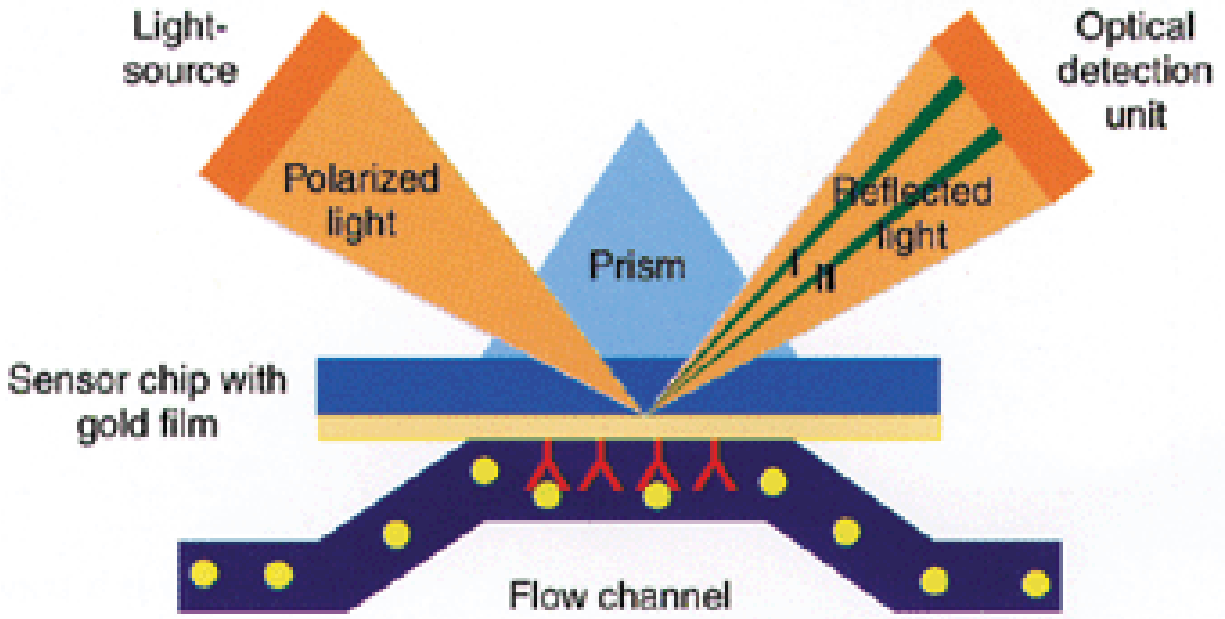
- Raman and transport correlate very strongly in time.
- **Demonstrates single-molecule Raman sensitivity.**
- **Demonstrates multifunctional sensing at single-molecule level.**



Ward *et al.*, Nano Lett. **8**, in press (2008)

Surface enhanced spectroscopies

Even without large surface enhancement effects, surface plasmon resonance sensors have become very popular (quite easy to implement).



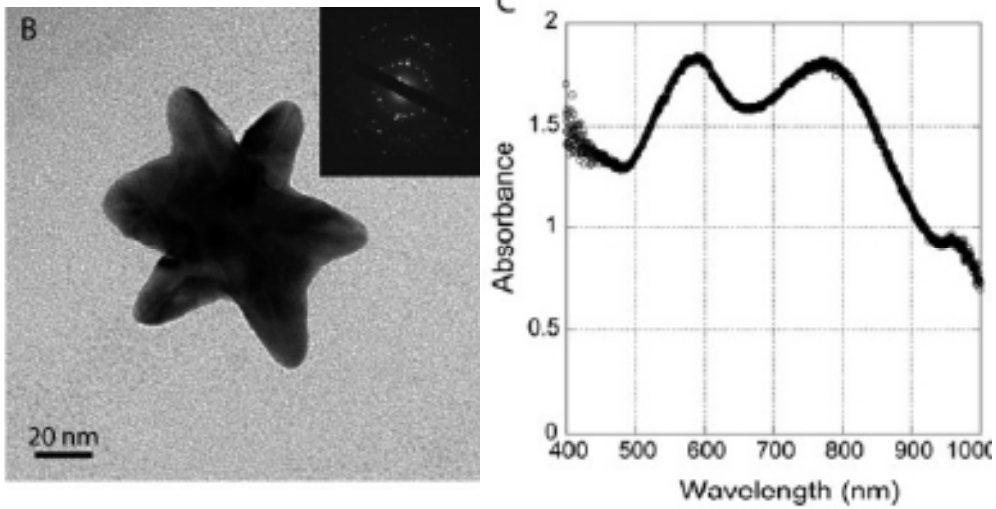
Commercial BIACore 3000 product in use at immunology center at UIUC.

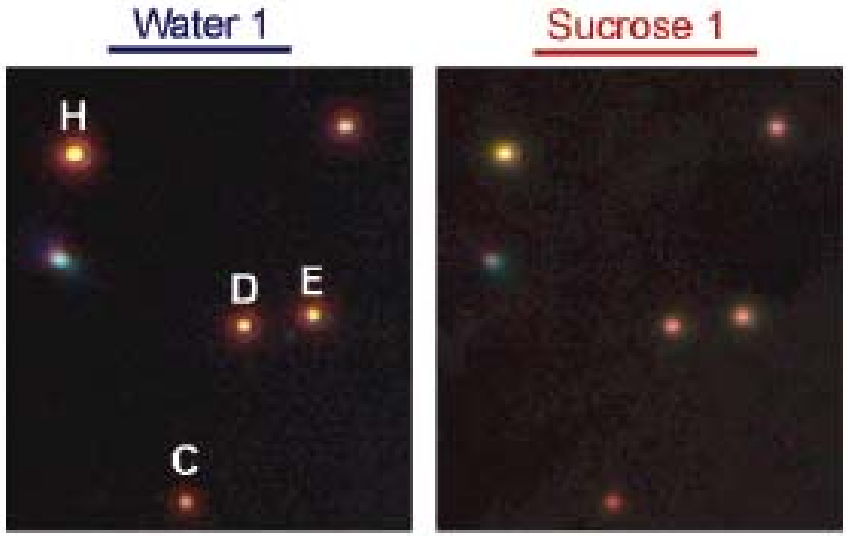
Surface enhanced spectroscopies

Single particles with sharp features in scattering spectrum due to plasmons.

Change in local dielectric environment shifts plasmon resonance.

Can see binding/unbinding events as shifts in single particle spectra.



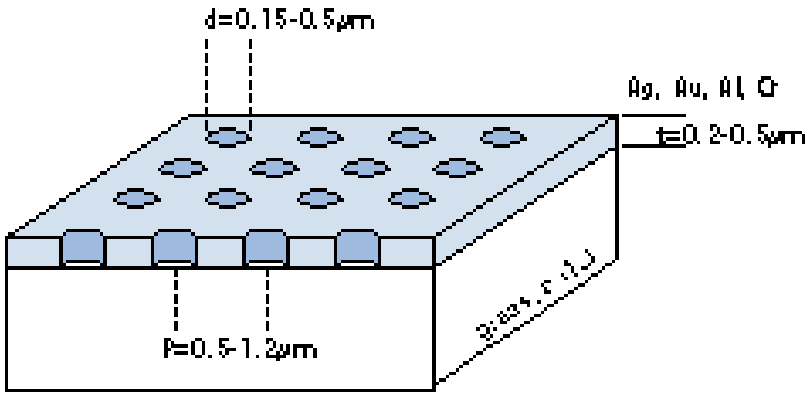


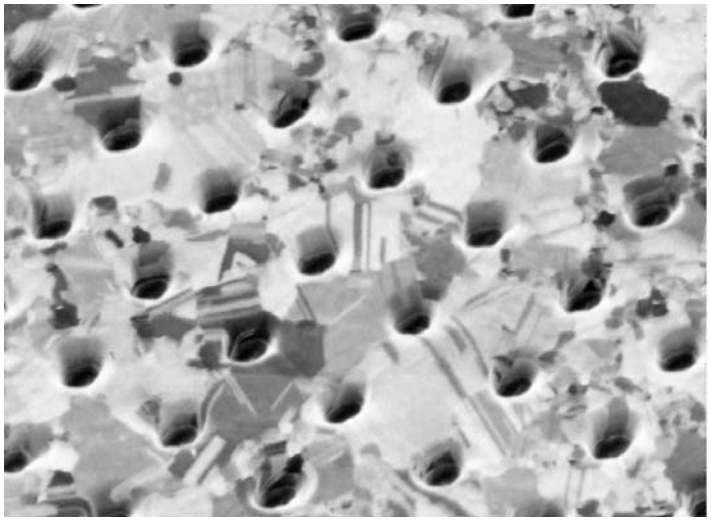
Nehl *et al*., Nano Lett. **6**, 683 (2006)

Plasmonics – transmission through apertures

Engineering nanostructures to enhance the coupling of plasmons to incident radiation has lead to a number of neat results.

Transmission through an optically thick film with a hole in it of size d is expected to scale like $(d / \lambda)^4$, and thus be very small….

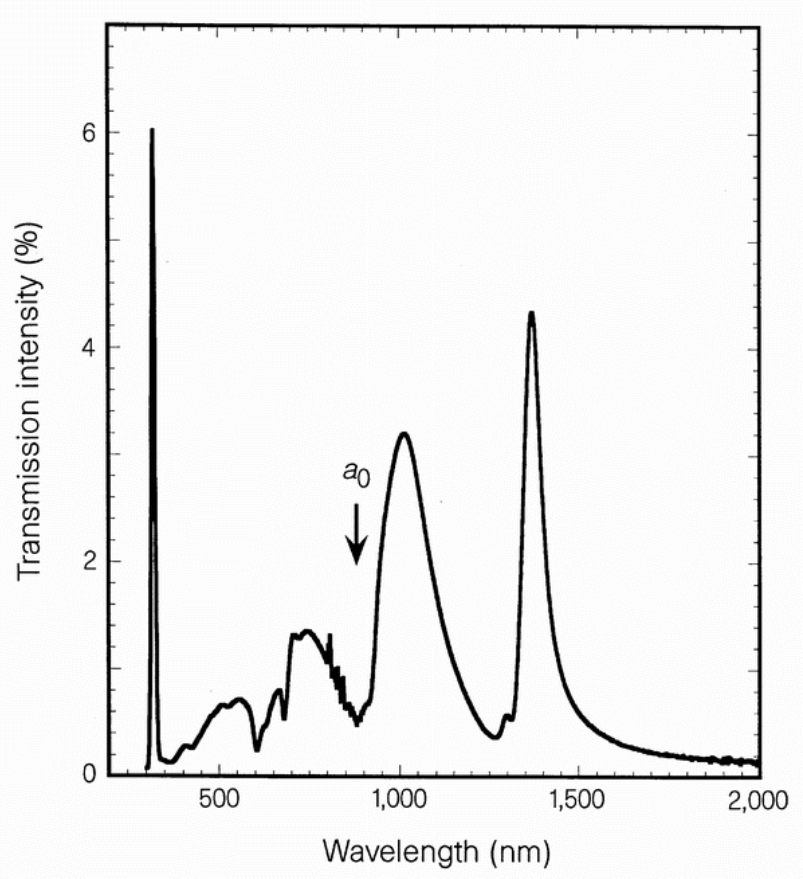




Thio, NEC

Plasmonics – transmission through apertures

Instead, for holes in a metal membrane, surface plasmons lead to much larger transmission than expected.



Shows efficiency of transmission through an array of 150 nm diameter holes in a Ag membrane.

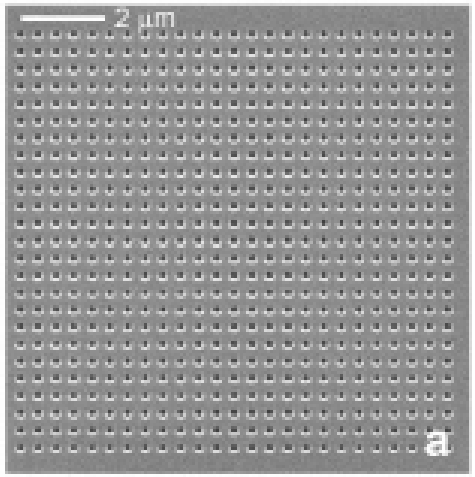
Expected efficiency is supposed to be ~ 10^{-3}.

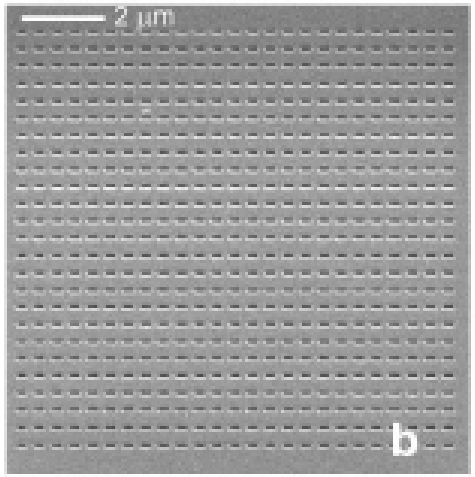
Even for wavelengths 10 times the hole diameter, transmission is 50x what one would expect.

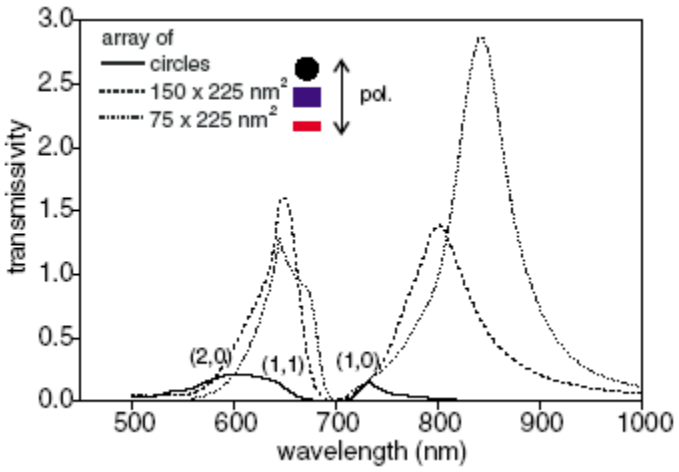
Very useful for things like NSOM….

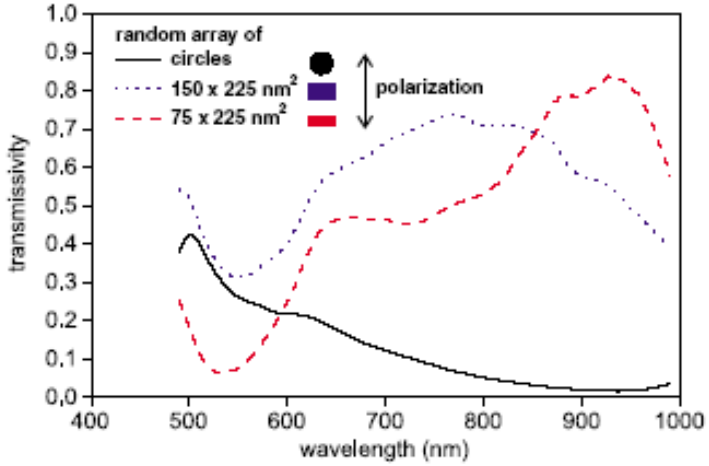
Plasmonics – transmission through apertures

Klein Koerkamp *et al.*, PRL **92**, 183901 (2004)









Shape resonances, cooperativity both matter!

Plasmonics – waveguides

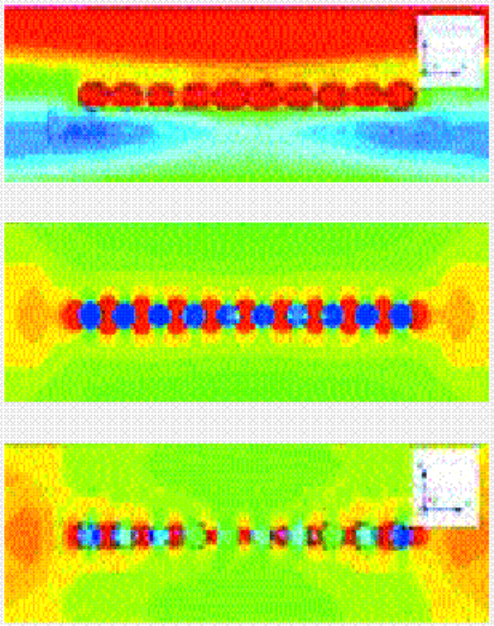


Figure 1 FDTD calculations of resonant and nonresonant excitation of plasmon wire composed of 50-nm gold particles: (a) in cross-section view of transverse mode resonant with the plasmon frequency; (b) in planar view of longitudinal mode excited resonantly at the plasmon frequency; and (c) in planar view of nonresonantly excited longitudinal mode.

Figure 2 Electron-beam lithography was used to fabricate a 90° corner (right) and a tee structure (left) of 50-nm Au particles on ITO-doped glass; particles are on a 75-nm (center-to-center) spacing.

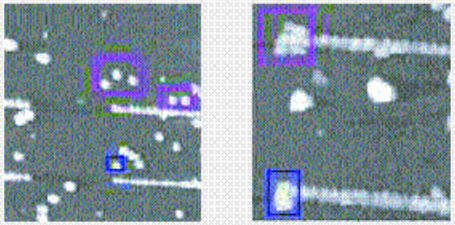
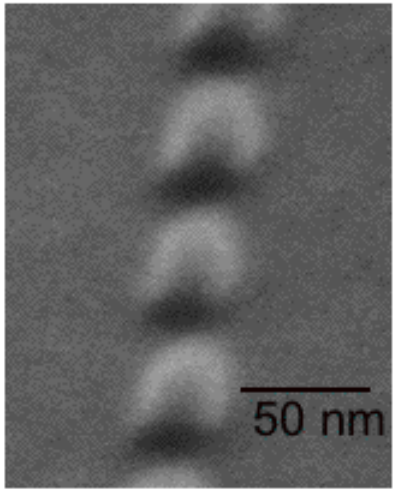


Photo credit: Caltech

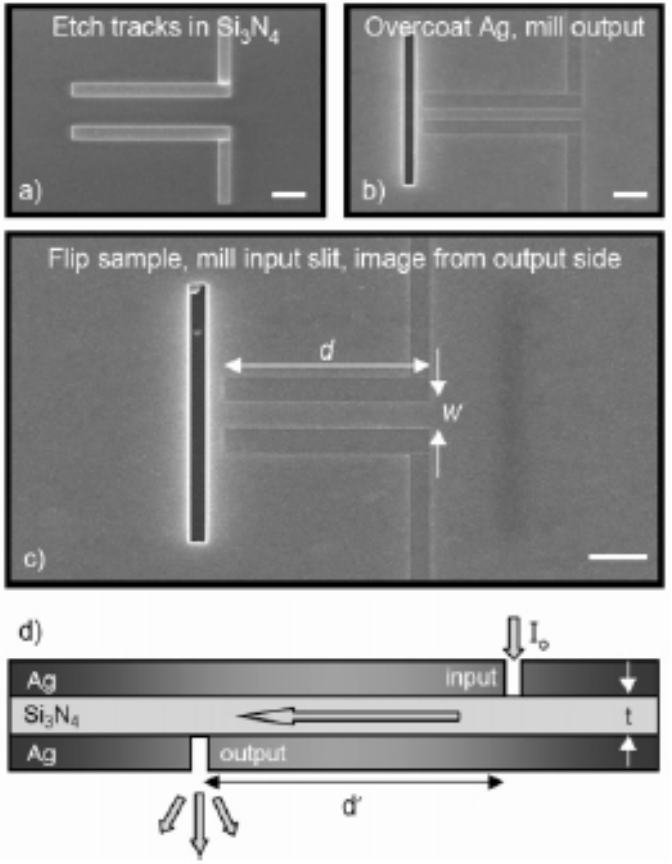


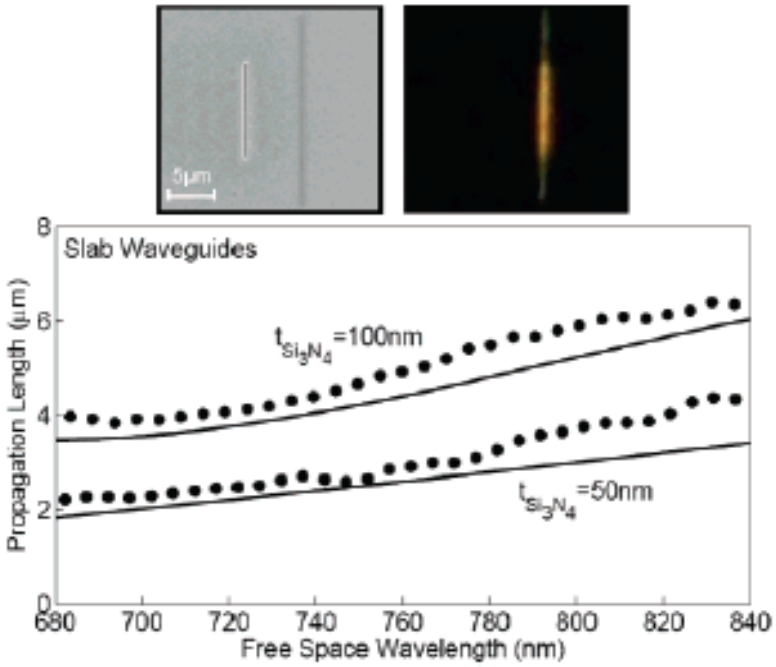
Atwater, Cal Tech

As in using defects in photonic band gap systems, there is interest in using coupled chains of metal nanoparticles to transfer electromagnetic energy.

Would all be very subwavelength, but losses are a nontrivial problem.

Plasmonics – waveguides





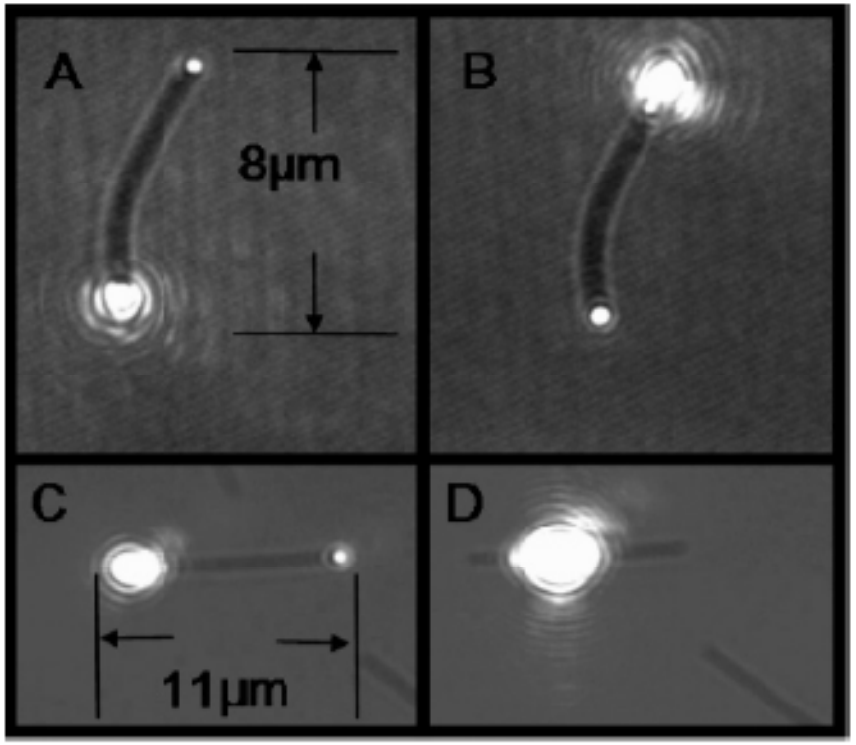
Dionne *et al.*, Nano Lett. **6**, 1926 (2006)

Can use slots in metal films to couple light into surface plasmon mode and ***steer*** or ***focus*** it at subwavelength scale.

Plasmonics – waveguides

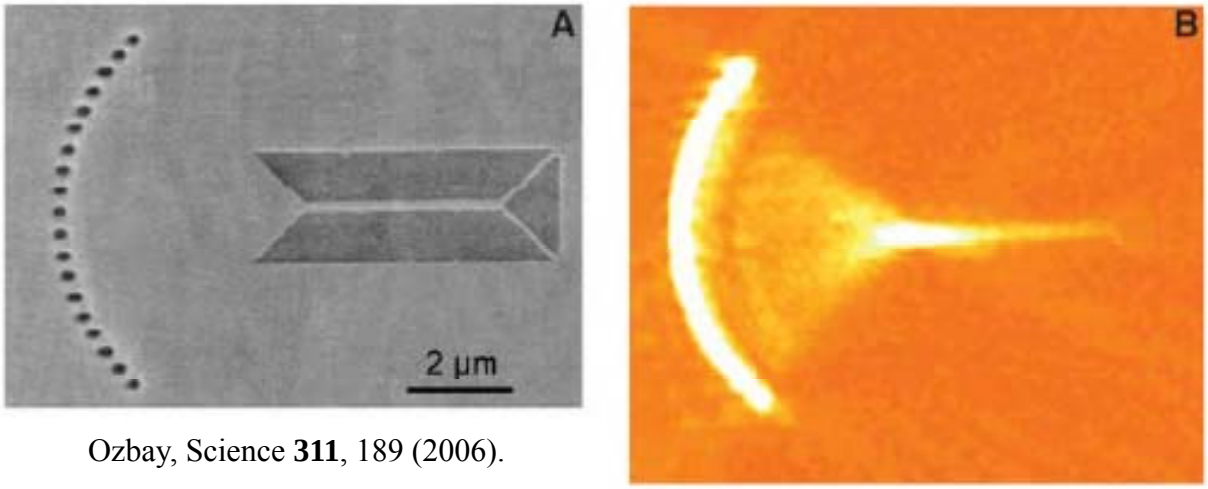
Silver nanowires can act as efficient plasmonic waveguides (!).

Need to break cylindrical symmetry to get coupling to/from the far field.



Sanders *et al*., Nano Lett. **6**, 1822 (2006)

Plasmonics – waveguides



Ozbay, Science **311**, 189 (2006).

Can do clever plasmonic lensing as well….

Metamaterials and "left-handed media"

We've already seen that nanostructuring materials can lead to dramatic changes in their optical properties, including tremendous flexibility in manipulating the effective index of refraction.

In 1964, Veselago examined the consequences of having a material in which, for some range of frequencies, ε and μ are both negative.

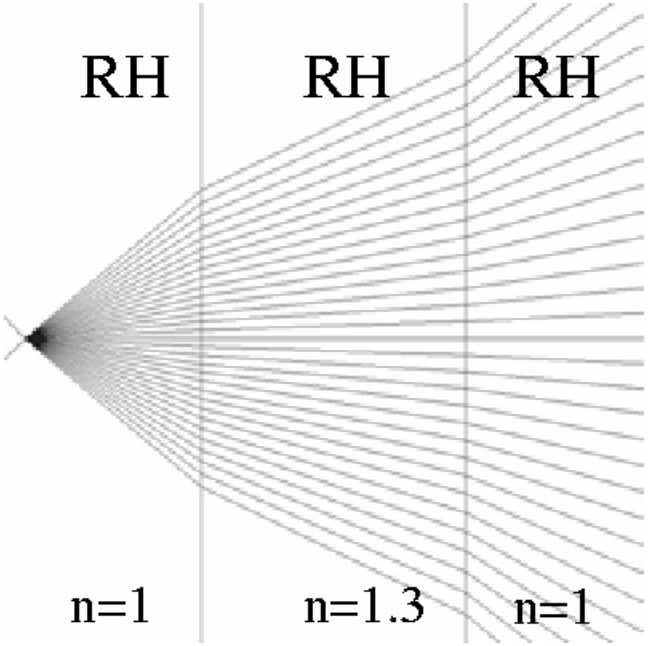
Since the product of the two is still positive, you might think this would be pretty weird by boring.

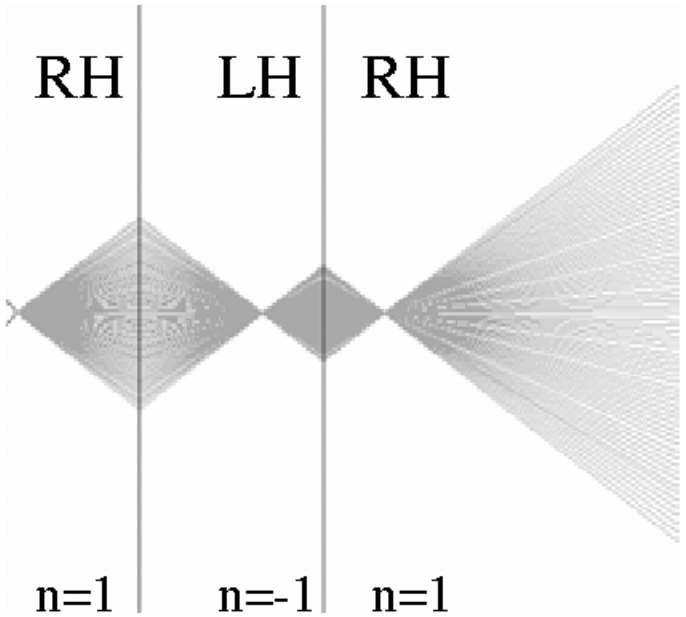
Actually has profound possibilities!

Can be interpreted (Snell's law) as having a *negative index*:

$$n(\omega) \equiv \sqrt{\frac{\varepsilon(\omega)}{\varepsilon_0}}\sqrt{\frac{\mu(\omega)}{\mu_0}} \qquad \sin\theta_i = \pm|n|\sin\theta_r$$

The idea

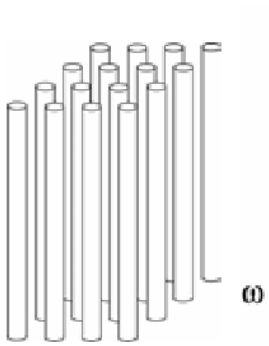


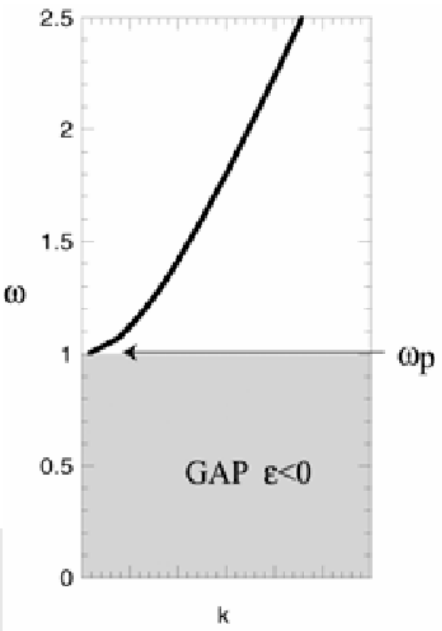


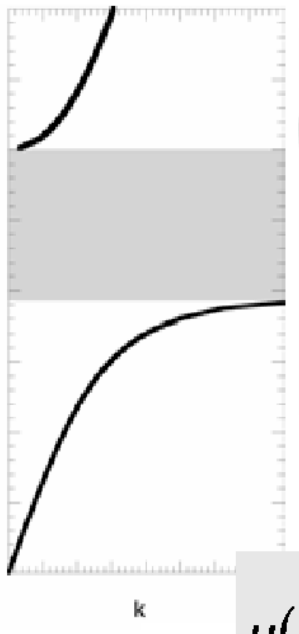
Soukoulis, IA State

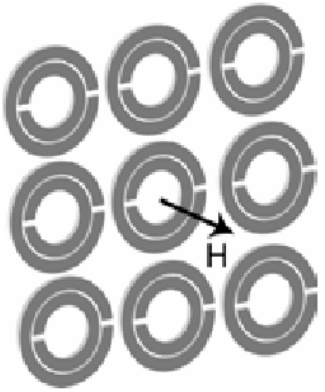
Can get focusing without lenses, and can even evade diffraction limits, if one can really get this to work.

The idea









$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2}$$

$$\mu(\omega) = 1 - \frac{\omega_p^2}{\omega^2 - \omega_0^2}$$

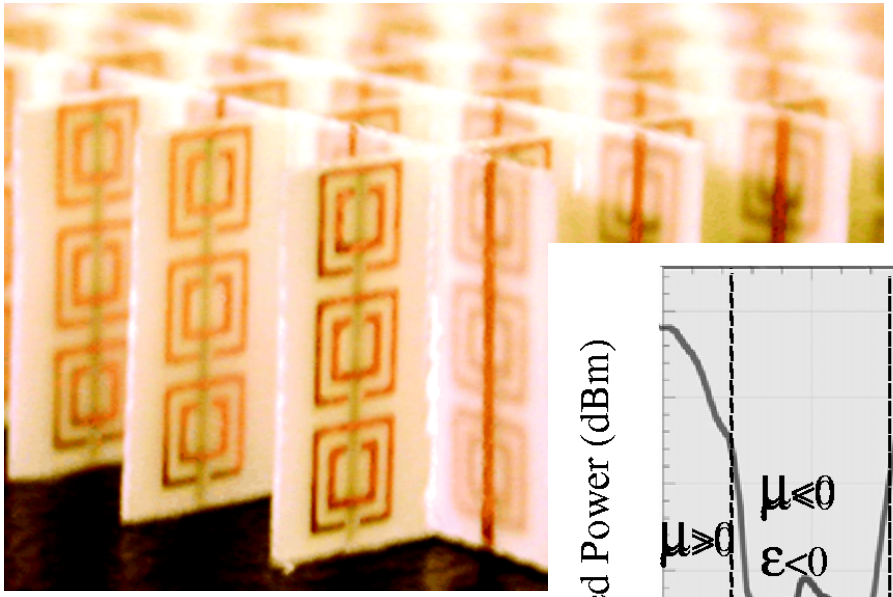
J. B. Pendry

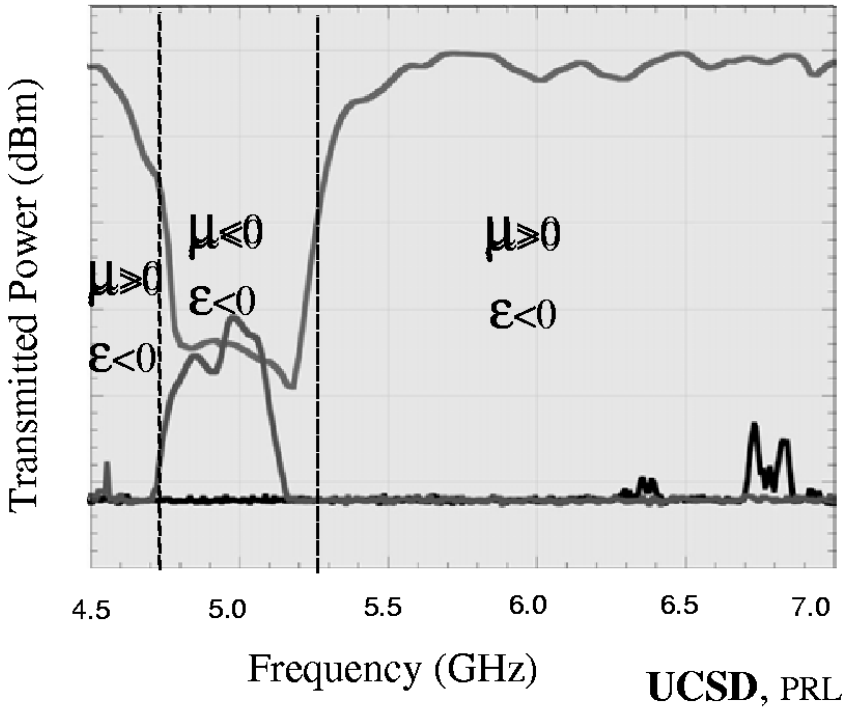
Soukoulis, IA State

Idea is to combine different metamaterials composed of metals and dielectrics to create a frequency region where the dielectric permittivity and the magnetic permeability ar simultaneously negative.

The claim

D. Smith, UCSD



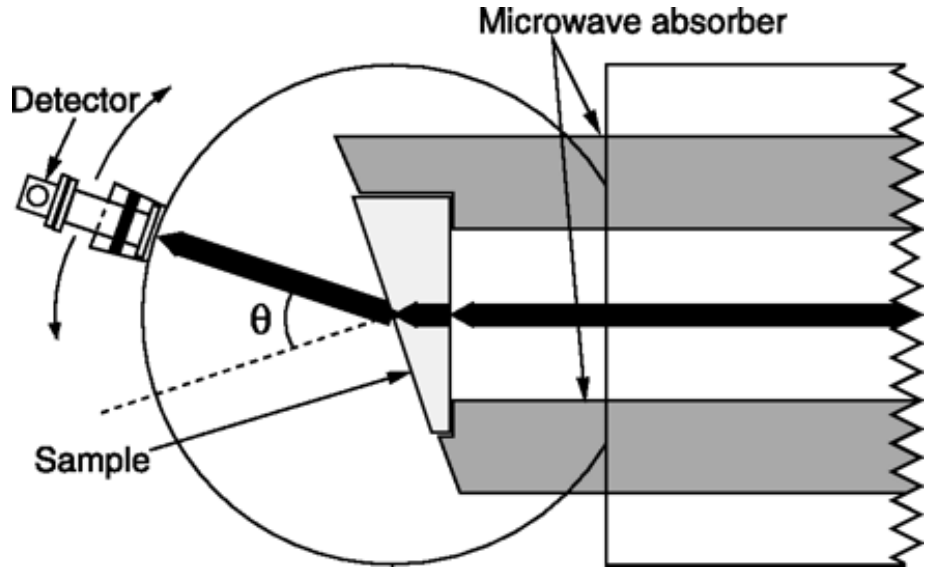


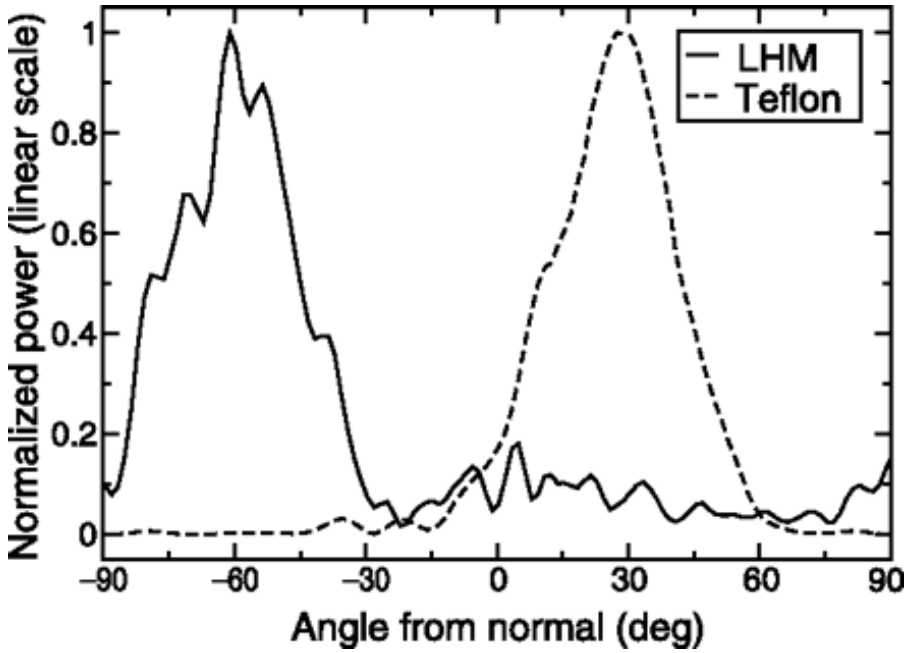
UCSD, PRL 84, 4184 (2000)

The claim

Shelby *et al.*, Science **292**, 77 (2001)

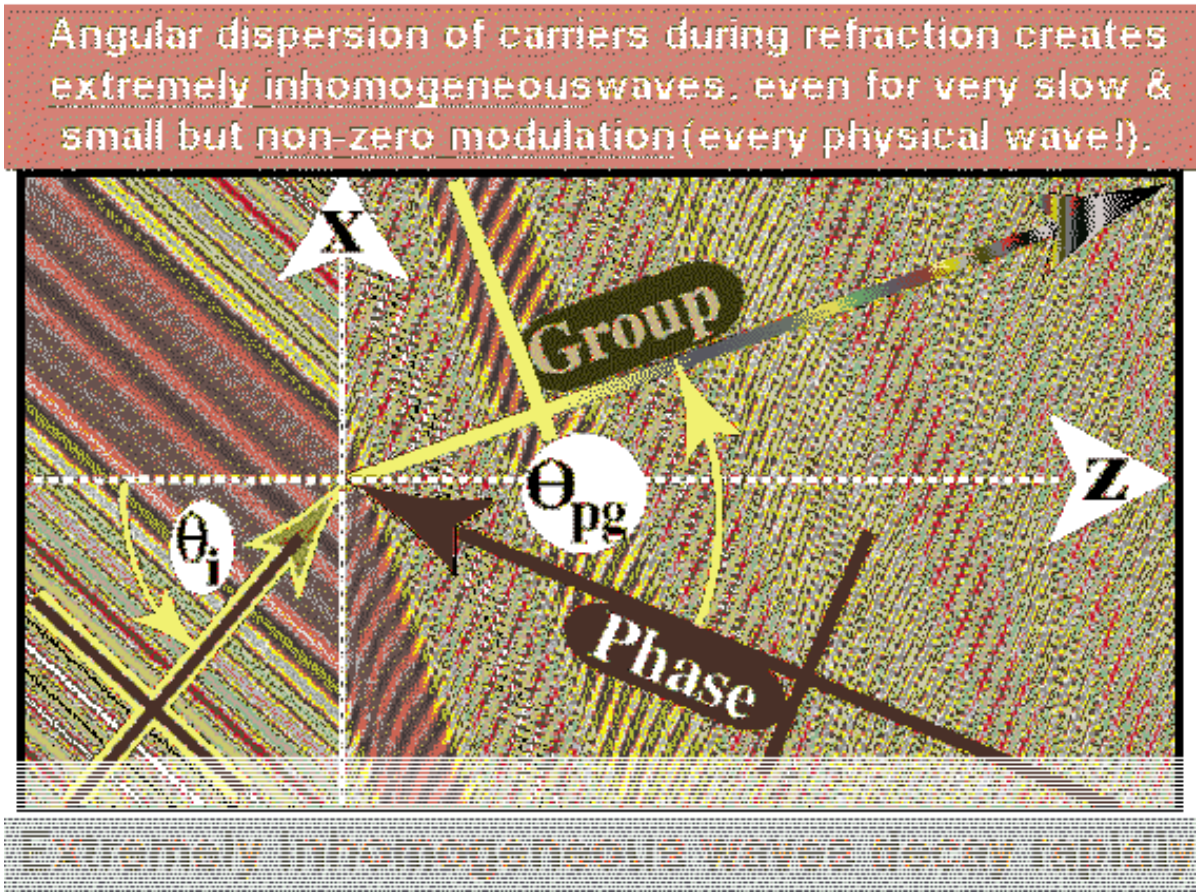
Clearly saw a microwave signal that seems to indicate the bulk of the transmitted microwave power does, indeed, come out the direction you'd expect for a negative index material!





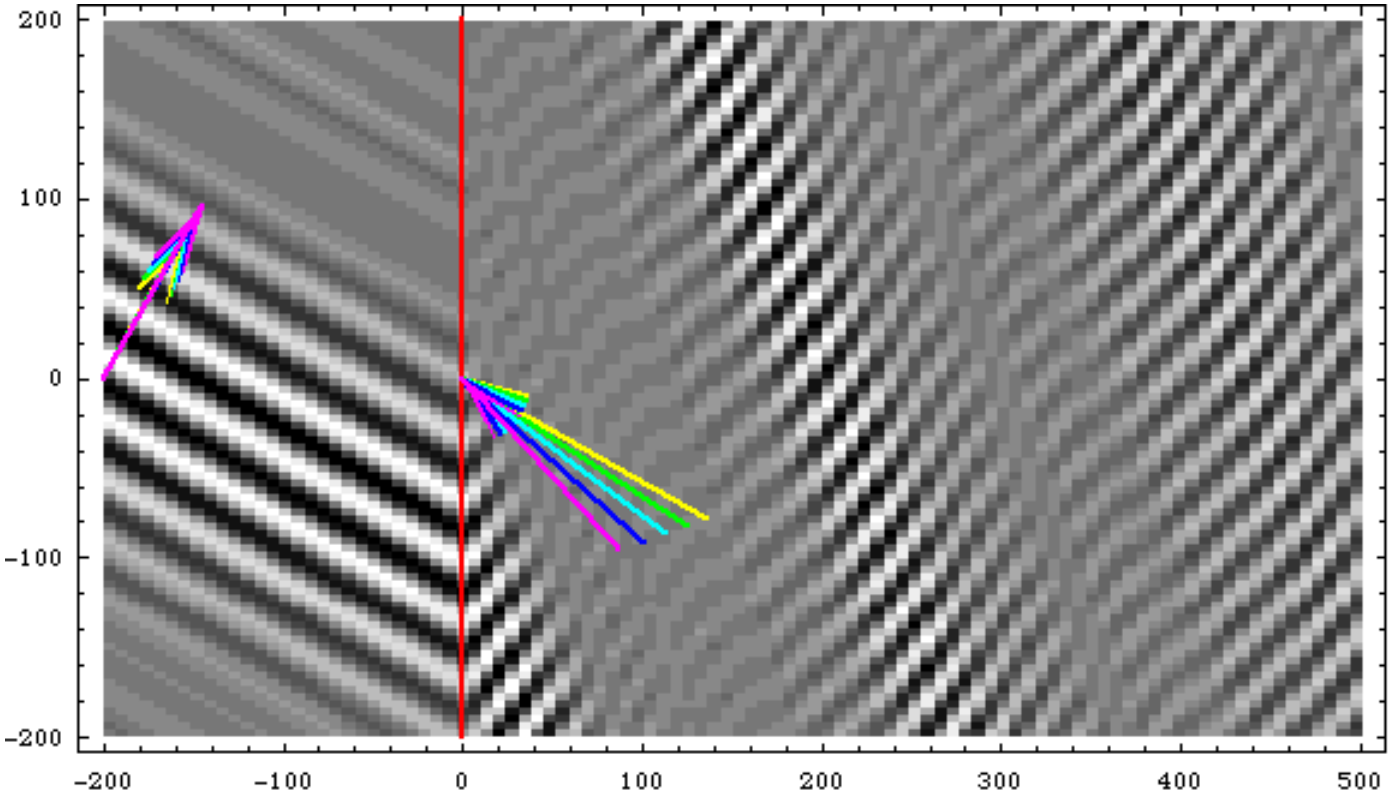
The counter-claim

http://www.utexas.edu/research/cemd/nim/index.html



Valanju *et al.* at UT claim that this is *not* useful - that dispersion makes it impossible to use this for sending signals that refract negatively.

The counter-claim



The debate rages on…..

The plot thickens

Consider a twinned crystal boundary in a uniaxial material.

$$\varepsilon_{\perp} \neq \varepsilon_{\parallel}$$

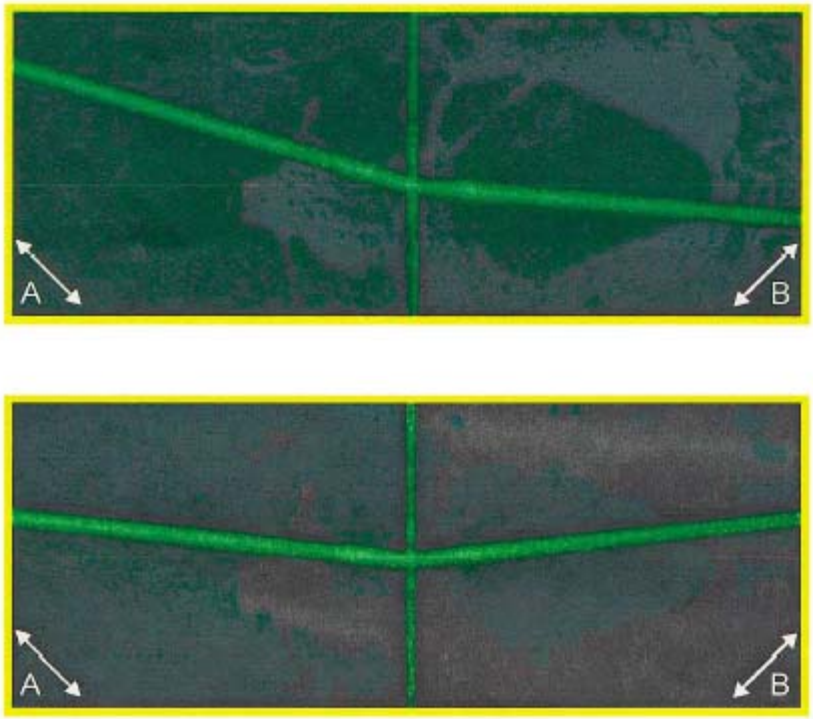
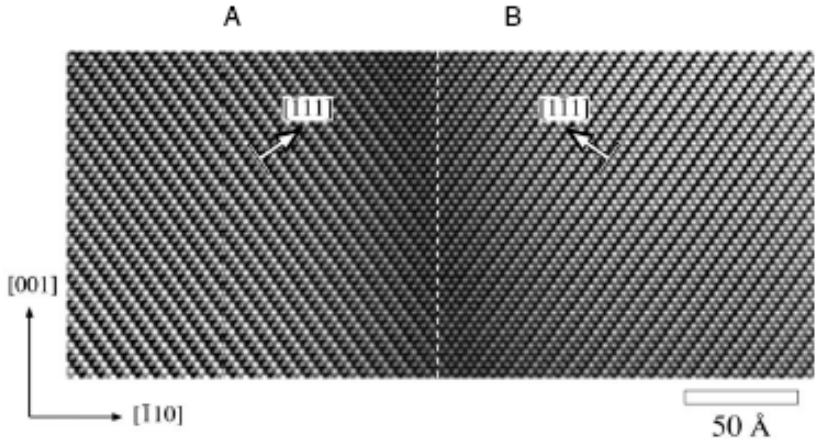


FIG. 3 (color). Images of light propagation in a YVO_4 bicrystal. The upper panel shows an example of normal (positive) refraction, the lower panel shows an example of abnormal (negative) refraction. Note that no reflection is visible at the bicrystal interface to the naked eye. The interface is illuminated by inadvertently scattered light.

Zhang *et al.*, PRL **91**, 157404 (2003)

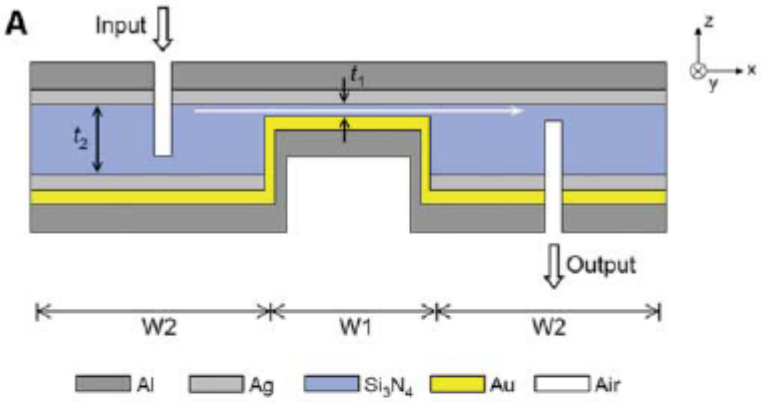


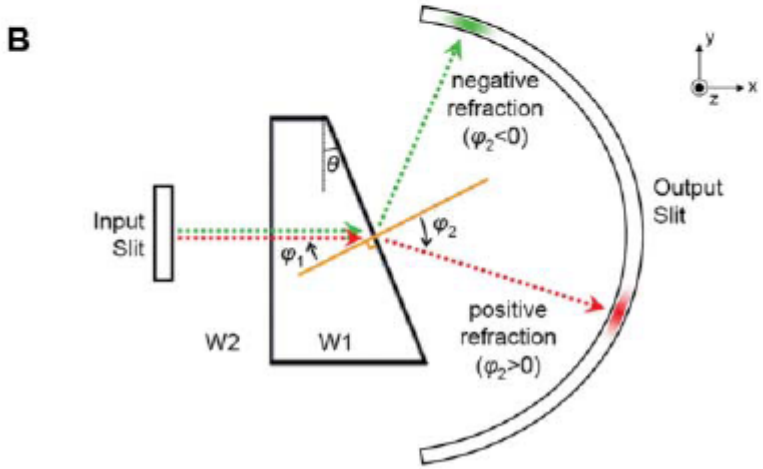
For the right circumstances, can get *negative refraction* (lower panel at left) or even *total refraction* (upper).

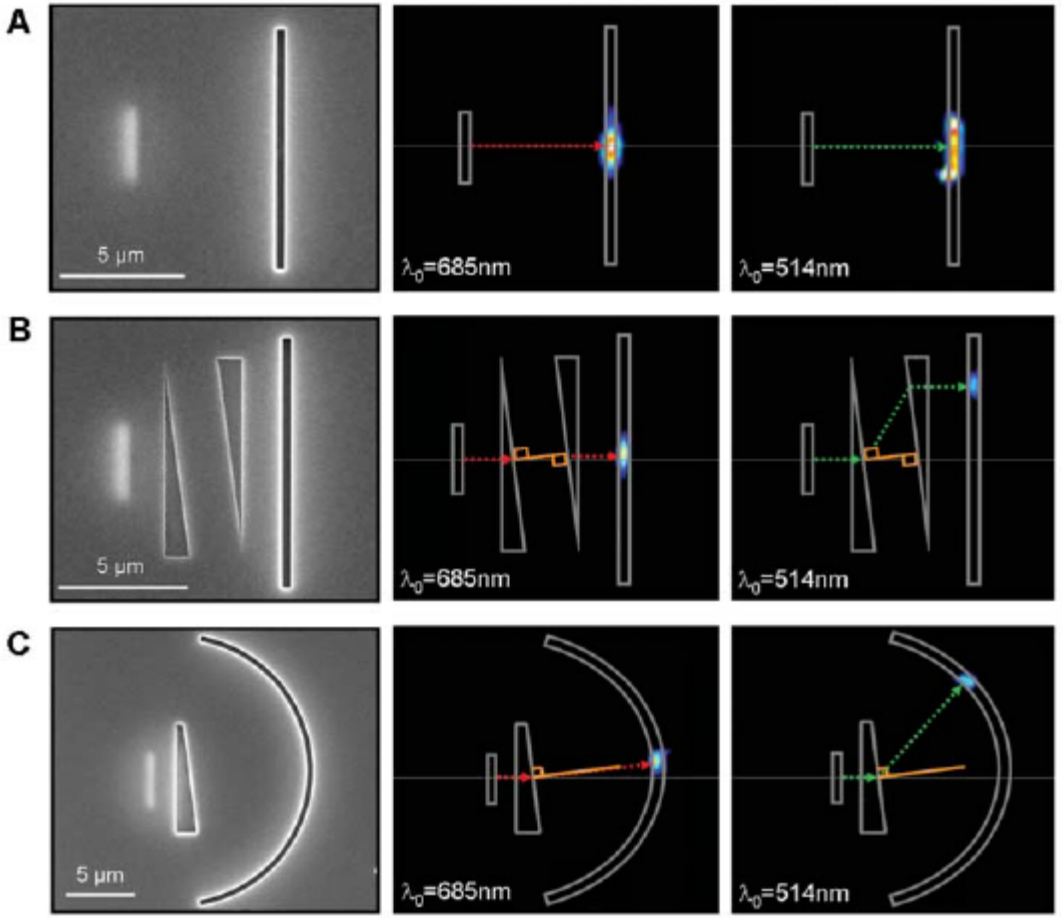
Authors point out that analogous behavior can happen for e^- at heterointerfaces, too.

Negative refraction (of surface plasmons) at visible frequencies

Lezec *et al.*, Science **316**, 430 (2007)



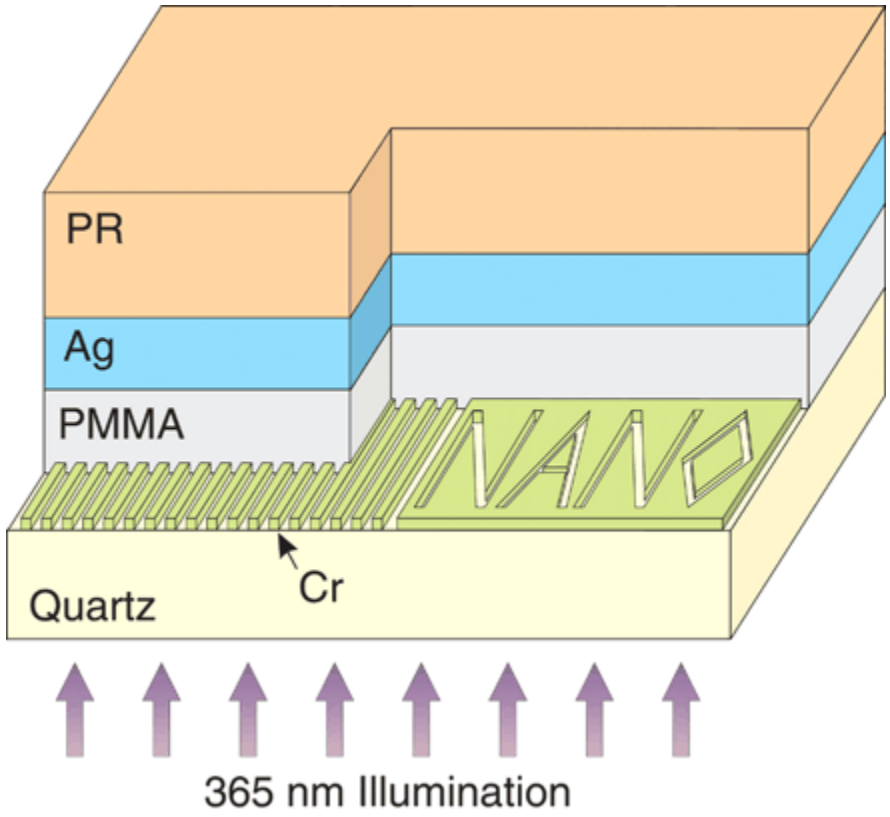




Superlenses

Fang *et al.*, Science **308**, 534 (2005)

Use plasmon properties of metal layer to enhance evanescent fields.



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Fang *et al.*, Science **308**, 534 (2005)

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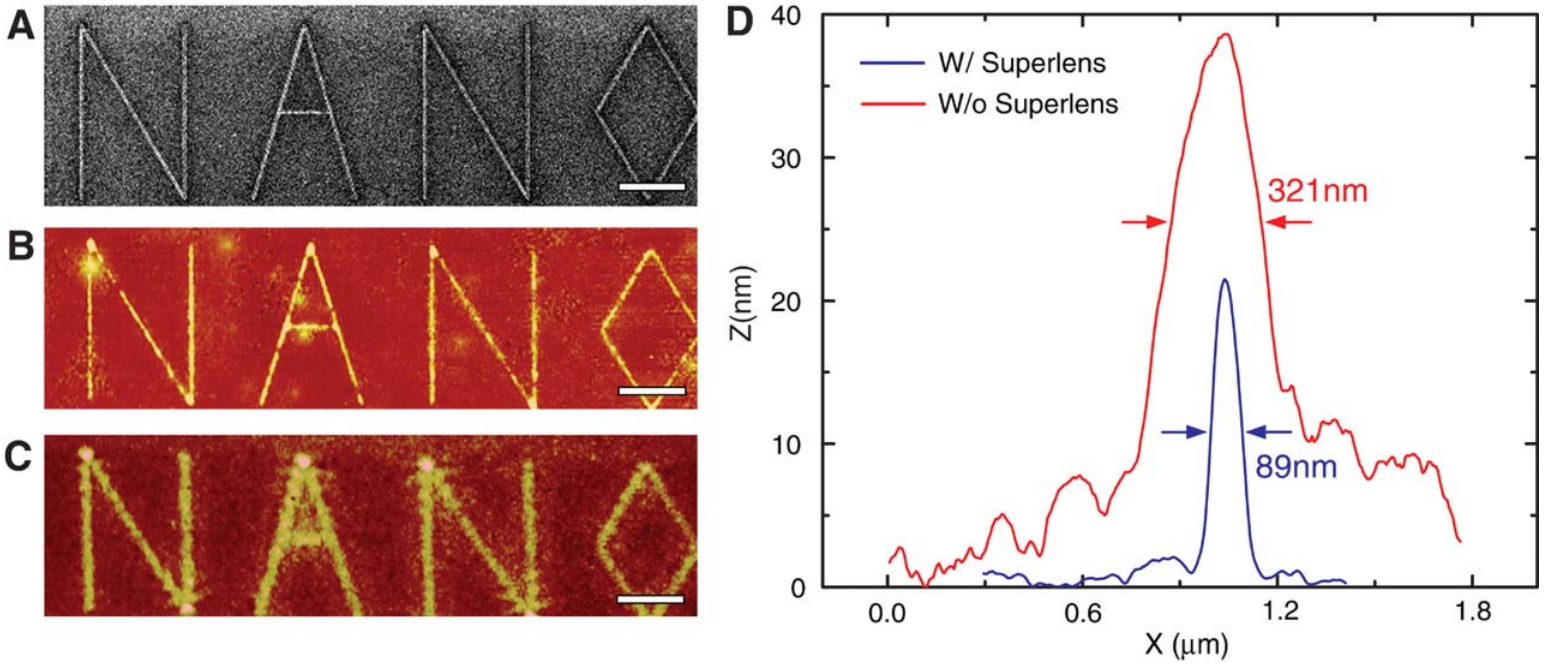
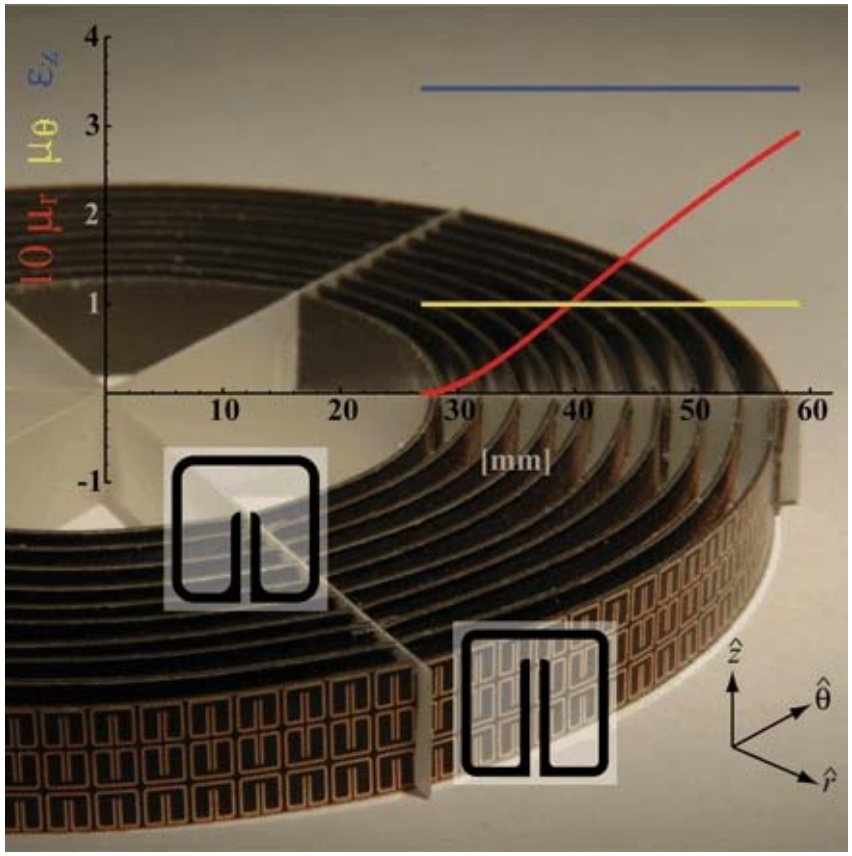
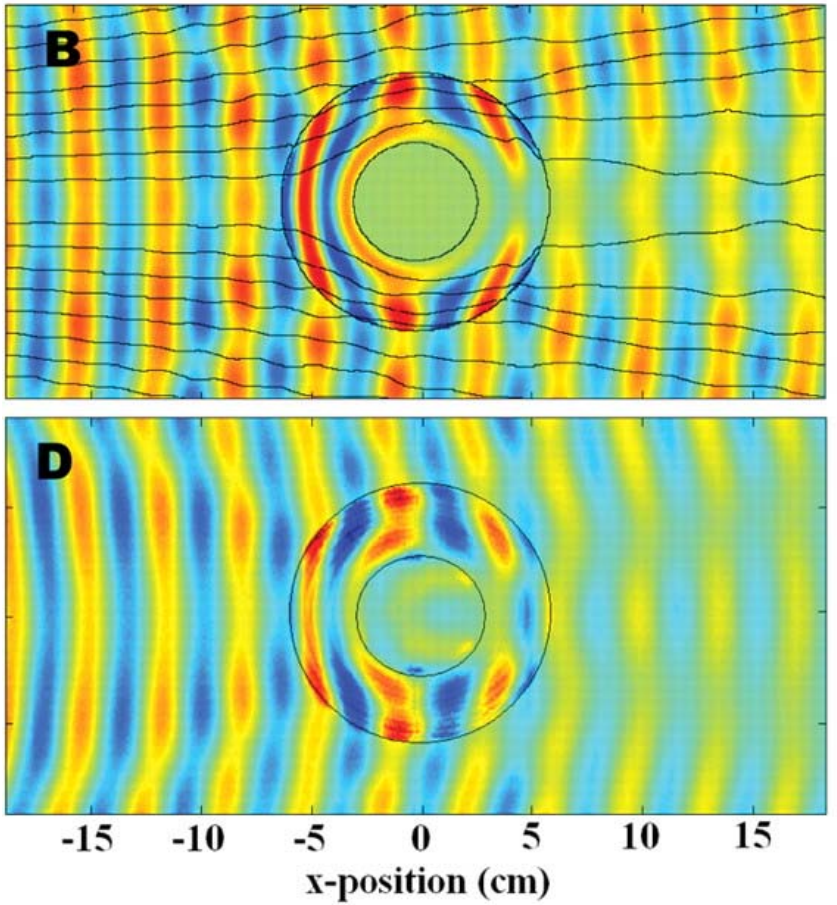


2 micron scale bar – resulting developed resist pattern is narrower when Ag layer is present than when PMMA control layer is present.

“Invisibility”

Schurig *et al.*, Science **314**, 977 (2006)





Can bend (one particular range of frequencies) completely around an object.

How well can this be done, and what are the downsides?

Summary

- Adding nanostructured conductors into the mix makes possible all sorts of useful optical phenomena.
- Plasmons can be combined with near-field effects to have enormous impact in a number of fields.
- Fundamental questions still raised, with possibility of metamaterials with quasimiraculous properties….
- Review article: Ozbay, Science **311**, 189 (2006).