Low Energy Electrodynamics in Solids 2002

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Abstracts
& Participants

Co-Chairs:
László Mihály (Stony Brook)
G. Lawrence Carr (BNL NSLS)
Peter D. Johnson (BNL Physics)
Welcome to Montauk and the LEES ’02 conference. Like previous LEES meetings [this being the fifth, following those in Bad Honnef (Germany, 1993) Trest (Czech Republic, 1995) Ascona (Switzerland, 1997) and Pécs (Hungary, 1999)], the program focuses on the science of low energy electron dynamics in condensed matter, including correlation effects and collective phenomena. In addition to infrared, THz, microwave and Raman studies, this year's meeting includes other synchrotron radiation probes (such as photoemission and inelastic scattering) of low-energy behavior. Sessions focusing on complex metals and oxides, synthetic metals, magnetic materials, and the techniques for studying them (such as high-field ESR, coherent THz spectroscopy, and synchrotron-based source technology) are included in the schedule. We hope you find the meeting program interesting and that you enjoy your stay in beautiful Montauk.

The Conference Co-Chairs:
László Mihály (Stony Brook)
G. Lawrence Carr (BNL NSLS)
Peter D. Johnson (BNL Physics)

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The organizers gratefully acknowledge the support of our commercial sponsors.

LEES ’02

ABSTRACTS
An Infrared Probe of Itinerant Ferromagnetism in III-V Semiconductors.

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The doping and temperature dependence of the complex conductivity is determined for the ferromagnetic semiconductor Ga$_{1-x}$Mn$_x$As. A broad resonance develops with Mn doping at an energy scale close to 200 meV, well within the GaAs band gap. Possible origins of this feature are explored in the context of a Mn induced impurity band and inter valence band transitions. From a sum rule analysis of the conductivity data the effective mass of the itinerant charge carriers is found to be at least a factor of three greater than what is expected for hole doped GaAs. In the ferromagnetic state a significant decrease in the effective mass is observed, demonstrating the role played by the heavy carriers in inducing ferromagnetism in this system. E.J. Singley et al. Physical Review Letters 89, 097203 (2002).

This work is a collaboration with E.J. Singley (University of California, San Diego), R. Kawakami and D.D. Awschalom (University of California, Santa Barbara).
Magneto-optical properties of metallic (III,Mn)V magnetic semiconductors

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Metallic diluted magnetic semiconductors (DMS's) materials offer a much wider spectrum of magneto-transport and magneto-optic effects than conventional itinerant electron ferromagnets, mostly due to the greater tunnability of the Mn moments ordered state through growth conditions, doping, gates, and light. The magneto-optical effects have not been explored as intensely as its transport counterpart in spite of the obvious potential for novel physical behavior and more precise characterization of the underlying physics. Here we study the infrared magneto-optical properties together with some thermodynamic and transport properties of (III,Mn)V semiconductors based on the itinerant hole-fluid model. Within this phenomenological approach we consider the virtual crystal approximation, applicable to the cleaner metallic samples, including disorder scattering within a Born type approximation, and a finite size self-consistent calculation where disorder and interactions are treated in an equal footing. We will discuss the different results and predictions of these calculations. We also demonstrate the possibility of using optical absorption as a tool to measure the itinerant carrier density.
Manipulating magnetism with light in ferromagnetic semiconductors and related nanostructures

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Because of moderate carrier concentration, semiconductors and associated nanostructures are suitable electronic systems to control both charge and spin by electromagnetic means. Particularly in magnetic semiconductors, manipulation of carrier spin can result in the cooperative and amplified effects through the spin exchange interaction between carrier spin $s$ and local spin $S$. One precursory demonstration is the manipulation of magnetism with light in ferromagnetic III-V alloy semiconductors (In,Mn)As and (Ga,Mn)As.

In this presentation, we discuss the experimental results on collective rotation of ferromagnetically coupled spins and resultant change in magnetization orientation in ferromagnetic (Ga,Mn)As, caused by circularly polarized cw- or pulsed-light excitations without a magnetic field. In particular, a change in magnetization orientation in picosecond time domain is the subject of current interest. Light-induced change in magnetic susceptibility observed at room temperature in GaAs-Fe composite structures that contain magnetic nanoparticles is another interesting subject. Introduction as to the development of ferromagnetic III-V alloy semiconductors shall also be given.
Interplay between the numerous degrees of freedom in CMR manganites

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In dilute magnetic semiconductors (DMS), the interactions between carrier and spin are known to play important roles. The physical properties of these intriguing materials have similarities as well as difference with those of the CMR manganites, such as (La,Ca)MnO$_3$. In this talk, we will focus on how the optical spectroscopy technique can be used to probe the interplay between these degrees of freedom in the CMR manganites. Especially, we will address how the carrier and the spin can be strongly coupled, and how the lattice degree of freedom can form a lattice polaron, which modifies the strong carrier-spin coupling [1]. We will also show how the spin/orbital ordering pattern can affect the optical properties of the manganites, such as (Nd,Sr)MnO$_3$ [2]. Then, we will also discuss on possible implications of our results in understanding the physics of DMS.


Electronic Excitations in Low Dimensional Cuprates: Resonant Inelastic X-ray Scattering Studies

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Resonant inelastic x-ray scattering studies of the electronic excitations in a series of low-dimensional cuprates are reported. These experiments utilize the enhancements in the inelastic cross-section obtained when the incident photon energy is tuned to, in this case, the Cu K-edge. This technique is discussed first in the context of the quasi-one dimensional, edge-sharing, material, CuGeO₃, for which an enhancement of a factor of 300 is observed over the non-resonant scattering. This enhancement allows high-resolution (0.3 eV) experiments to be performed. In CuGeO₃, such measurements revealed the presence of three excitations and the dispersion along the chain direction is reported for each. While no dispersion is observed for the gap excitation, at 3.8 eV, the charge-transfer excitation (6.4 eV) exhibits some dispersion (0.15 eV) along the chain direction. The role of the geometry (connectivity of the CuO₄ plaquettes) and dimensionality in these results is emphasized by comparison with other cuprates. These include LiCuO₂, a more ideal edge-sharing chain compound, which exhibits no measurable dispersion of the gap excitation and SrCuO₂, a zig-zag chain system comprised of two corner-sharing chains which exhibits considerable dispersion (1.5 eV). These results are contrasted with detailed studies of the two dimensional system, La₂₋ₓSrₓCuO₄ for which two excitons are observed with differing dispersions in the 2D Brillouin zone. Doping carriers into this system is observed to fill the gap. The increased screening that arises in the resulting metallic-like state washes out the lower energy, 2eV exciton. In contrast, the higher-energy, 4eV excitation appears to survive in the metallic state largely unaffected.

Work performed at BNL under DOE contract no. DE-AC02-98CH10886.
Low Energy Phenomena at High Magnetic Fields: A Facility Overview*

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There have been many investments with more planned in high magnetic field capabilities across the world. The National High Magnetic Field Laboratory (NHMFL), which is funded by the National Science Foundation, is only one example of the new capabilities to explore physical phenomena at the edge of an ever-expanding parameter space, i.e., pressure, temperature, magnetic fields and orientation. The Netherlands will dedicate a new magnet laboratory in 2003 and new capacity is being developed in France and Germany. In addition, many other countries are exploring options for new high magnetic field facilities. These investments are being driven by the exciting scientific opportunities in condensed matter and biological sciences. In addition to the need for continued investment in high magnetic fields, there is a complementary need to advance instrumentation responding to the energy scales dominantly impacted by current and future magnet facilities, e.g., advanced approaches to make mm-wave and far infrared spectral regions more accessible to researchers working at high magnetic fields. This presentation will provide a worldview of high magnetic field facilities and survey some of the exciting new science opportunities driving this interest.

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Phase separation in the charge ordered compounds
Nd(Pr)$_{1-x}$Ca$_x$MnO$_3$: an ESR study

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High frequency (9.4 GHz - 475 GHz), high magnetic field (0 – 12 T) Electron Spin
Resonance has been used to study the magnetic excitations and possible phase separation
effects in the charge ordered manganites Nd(Pr)$_{1-x}$Ca$_x$MnO$_3$. In the Nd compounds, in
form of powders, we show [1] that the Nd ions are weakly coupled to the Mn ions via
ferromagnetic exchange and are responsible for the peculiar ferromagnetic resonance
observed in the FM phase of both compounds (ground state below 120K for x=0.3, high
field state for x=0.5). We also show that there is no trace of the FM state imbedded in the
low field, CO phase in Nd$_{0.5}$Ca$_{0.5}$MnO$_3$. On the contrary, in a 250 nm thick
Pr$_{0.5}$Ca$_{0.5}$MnO$_3$ film grown on LaAl$_2$O$_3$ substrate, we show evidences for the presence of
a FM phase within the CO phase in form of very thin layers, with the ferromagnetic easy
axis at 45° from the film plane. The coupling of this FM phase with the CO phase
depends on the orientation of the applied magnetic field (parallel or perpendicular to the
film plane).

High-field ESR on the spin dynamics in La$_{1-x}$Sr$_x$MnO$_3$

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Sr-substitution to the parent antiferromagnet LaMnO$_3$ leads to a rich phase diagram ranging from an antiferromagnetic insulator up to a ferromagnetic metal. The spin dynamics of the resulting compounds can be well studied using high-field electron spin resonance (ESR). We present results of high-field ESR experiments in La$_{1-x}$Sr$_x$MnO$_3$ obtained within different quasioptical arrangements for frequencies $40 \leq \nu \leq 700$ GHz and for magnetic fields $B \leq 12$ T. A splitting of the antiferromagnetic resonance (AFMR) mode is observed in the magnetic field for the parent compound LaMnO$_3$ in agreement with the antiferromagnetic structure of this material. Abrupt changes in the AFMR frequencies have been observed around $x \approx 0.025$ and were attributed to a transition between a pure antiferromagnetic and a canted state. For increasing Sr-doping the AFMR modes are split even in zero field, which can be naturally explained using the concept of a canted magnetic structure for $x < 0.1$. The high-field ESR experiments thus contradict the scenarios of electronic phase separation in low-doped manganites. In La$_{0.825}$Sr$_{0.175}$MnO$_3$ the ESR spectra are consistent with the ferromagnetic and metallic state. The lines of ferromagnetic resonance and ferromagnetic antiresonance can be clearly observed. For intermediate concentrations $0.1 \leq x \leq 0.15$ the ESR spectra become complicated, which can be well explained by a single ferromagnetic resonance mode and taking electrodynamic effects into account.
Recent Results of EPR Studies of La$_{2-x}$Sr$_x$CuO$_4$

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Electron Paramagnetic Resonance (EPR) is a powerful tool in solid state physics, which allows the study of electronic and magnetic properties on a microscopic level. However, the application of EPR to high-T$_c$ cuprates was restricted, owing to the absence of intrinsic EPR signals in these compounds. We present the new model of Cu spin relaxation which leads to the extremely large linewidth and provides an explanation for the long-standing problem of EPR silence in high-T$_c$ cuprates.

Another approach in the application of EPR to high-T$_c$ superconductors is to dope these compounds with small amounts of some paramagnetic ions which are used to probe the intrinsic behavior. One of the best candidates in Mn, which in the 2$^+$ valent state gives a well defined EPR signal and substitutes for the Cu$^{2+}$ in the CuO$_2$ plane. It was found that the Mn ions are strongly coupled to the collective motion of the Cu spins (the so called bottleneck regime). The bottleneck regime allows to obtain substantial information on the dynamics of the copper electron spins in the CuO$_2$ plane. We present a survey of recent results obtained by EPR of the Mn$^{2+}$-doped La$_{2-x}$Sr$_x$CuO$_4$ concerning the microscopic electronic phase separation, charge-spin dynamics and unusual oxygen isotope effects.
Magnetic Phase Transition and Hole Localization in Lightly Doped YBa$_2$Cu$_3$O$_6$

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The structure of the magnetic elementary cell is well established in YBa$_2$Cu$_3$O$_6$, the antiferromagnetic (AF) parent compound of a high temperature superconductor, but little is known about the structure of AF domains and virtually nothing about how introducing holes affects the magnetic structure. AF domains in YBa$_2$Cu$_3$O$_{6+\delta}$ were first reported$^1$ in neutron diffraction experiments in 1998 and were independently observed$^2$ in Gd$^{3+}$ ESR studies. ESR data in undoped crystals were interpreted assuming domain walls separating domains along the $c$ crystallographic direction. On the other hand, in lightly hole doped antiferromagnets, conducting "stripes" formed from segregated lines of localised holes and running within the ($a,b$) plane were suggested to explain µSR$^3$ and resistance$^4$ data.

In this talk the temperature and magnetic field dependence of the antiferromagnetic domain structure in a lightly hole doped Ca$_x$Y$_{1-x}$(Gd)Ba$_2$Cu$_3$O$_6$ copper oxide single crystal with $x \approx 0.008$ will be discussed. The domain structure is observed using a Gd$^{3+}$ ESR probe at several frequencies. The ESR satellites from Gd at first neighbor to Ca sites show that holes are not localized to the neighborhood of Ca$^{2+}$ ions at 2.5 K or above. This is consistent with the idea that localized holes form an ordered structure at low temperatures. The AF structure is static up to high temperatures. Hole doping changes the antiferromagnetic structure at low temperatures: the easy direction of the sublattice magnetization, $M_s$, is along [110] in the ground state in contrast to [100] in the undoped crystal. Increasing the temperature from 6 K to 100 K, a gradual magnetic phase transition to the usual [100] easy direction is observed. The transition is tentatively attributed to strain fields arising from holes localizing into an unknown structure.

High field EPR investigations of quantum and environmental effects in single molecule nanomagnets

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We report high frequency electron paramagnetic resonance (EPR) investigations of a series of high spin (total spin up to $S = 10$) manganese, iron, cobalt and nickel complexes which have been shown to exhibit single molecule magnetism, including low temperature (below ~ 1K) hysteresis loops and resonant magnetic quantum tunneling. A cavity perturbation technique enables high sensitivity oriented single crystal EPR measurements spanning a very wide frequency range (16 to 200+ GHz). Fitting of the frequency and field orientation dependence of EPR spectra allows direct determination of the effective spin Hamiltonian parameters. Studies for a range of materials with varying (approximately axial) site symmetries facilitates an assessment of the role of transverse anisotropy (terms in the Hamiltonian that do not commute with $S_z$) in the magnetic quantum tunneling phenomenon. We also examine quantitatively the temperature dependence of the EPR linewidths and line shifts, for fixed frequency measurements with an applied magnetic field along the easy axis. Simulations of the obtained experimental data take into account intermolecular spin-spin interactions (dipolar and exchange), as well as distributions in both the uniaxial anisotropy parameter $D$ and the Landé g-factor. These findings could have important implications for the mechanism of quantum tunneling of magnetization in nanomagnets, as well as providing deeper insights into the interactions which give rise to quantum decoherence.

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HIGH FREQUENCY MAGNETIC SPECTROSCOPY ON MOLECULAR MAGNETS

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New type of high frequency magnetic spectroscopy is utilized to study the crystal field transitions, tunneling and relaxation phenomena in Fe$_8$ [1] and Mn$_{12}$ac [2] magnetic clusters. Dipolar transitions within the ground $S=10$ multiplets split by the crystal field are directly observed (at frequencies below 5 cm$^{-1}$ for Fe$_8$ and below 10 cm$^{-1}$ for Mn$_{12}$ac) and studied in the equilibrium state: temperature and magnetic field dependences of eigenfrequencies, magnetic contributions and lineshapes. The relaxation of magnetization is studied in Mn$_{12}$ac by polarizing the spin subsystem, reversing the magnetic field and watching the time evolution of the magnetic dipolar transitions. For small temperatures, below 2 K, a resonant quantum tunneling is directly observed as a “magnetic hole burning” in the absorption spectra of a polycrystalline Mn$_{12}$ac sample. We develop a microscopic model which allows to quantitatively describe all observations. Several additional absorption lines are detected at elevated frequencies which could originate from transitions between different multiplets or lattice vibrations.

References
Electrodynamics of electron glasses

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Electron states, localized by disorder can assume various configurations, displaying different responses to electromagnetic fields. When electrons are near to being delocalized, a quantum critical state develops. In the well localized regime, and in the absence of electron-electron interactions Fermi statistics prevail, leading to a Fermi Glass (FG). Interactions lead to a Coulomb Glass (CG) phase, with a crossover to a FG behavior with increasing energy scales of the relevant probes. In all cases, in the low energy limit the complex conductivity can be described by a power law behavior, with different exponents. I will discuss the characteristic electrodynamical response of these states in NbSi and in Si:P. These experiments firmly establish the existence of the various states, and also lead to a crossover-diagram in the frequency-concentration phase space.
Quasiparticles in 1D Peierls systems: coexistence of conventional and unexpected features

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We utilized photoelectron spectroscopy (ARPES) with very high energy and momentum resolution to investigate the electronic states of two typical quasi-one dimensional systems: (TaSe₄)₂I [1,2] and the blue bronze K₀.₃MoO₃ [3]. Both materials exhibit metal-insulator Peierls transitions to a charge density wave (CDW) ground state. We find 1D features whose dispersion and symmetry properties are consistent with band structure calculations, but also with the existence of the CDW superlattice. We also observe characteristic spectral changes across the Peierls transitions. In spite of these 'conventional' features, the spectral lineshapes reveal very unconventional quasiparticles, with large masses and short coherence lengths of just a few angstrom. We suggest that the strong coupling to the lattice is responsible for the heavy renormalization, and that the ARPES spectra mainly reflect the incoherent nature of these polaronic excitations.

Spin dynamics and sliding density-wave in Sr$_{14}$Cu$_{24}$O$_{41}$ ladders

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Key words: Spin ladders; charge density waves; superconductivity; Raman spectroscopy

Sr$_{14}$Cu$_{24}$O$_{41}$ ladder compounds contain linear fragments of copper oxide planes. In contrast to the two-dimensional antiferromagnetic cuprates the spin $\frac{1}{2}$ two-leg ladders have short-range magnetic order and a spin gap. Holes doped into these ladders pair and superconduct at high doping concentrations, while insulators are known to result from low hole concentrations. The competition between insulating states and superconductive pairing has emerged as a key feature of the high-$T_c$ problem, but the character of the insulating states has remained elusive. Here, using transport and Raman scattering data, we identify the insulating state of self-doped two-leg spin ladders of Sr$_{14}$Cu$_{24}$O$_{41}$ as a weakly pinned, sliding density wave. This collective density-wave state exhibits a giant dielectric response, non-linear conductivity, and persists to well above room temperature [1].

Our results have quantitative parallels with sliding density wave transport phenomena observed in established charge/spin density wave materials, yet there is a number of important microscopic differences from conventional weak-amplitude charge- and spin-density waves. The density wave correlation in Sr$_{14}$Cu$_{24}$O$_{41}$ is a high temperature phenomena that we observe up to the highest measured temperature, above 630 K. Such high temperature correlations can not be supported by phonons and suggest that the charge/spin correlations arise from strong spin exchange interactions with characteristic energy scale $J \simeq 1300$ K [2]. Theoretical calculations for a doped two-leg spin ladder suggest that the holes are paired in a state of approximate $d$-wave symmetry with a few lattice spacings in size. The superconducting condensation of bound pairs is competing with a crystalline order of these pairs in a density wave state.

References


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18
Ubiquitous generalized ARPES signatures of electron fractionalization in quasi-low-d metals

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Many quasi-low-dimensional (quasi-low-d) metals show ARPES lineshape anomalies that contrast greatly with the spectra of the quasi-2d Fermi liquid reference material TiTe\textsubscript{2} and hint at the electron fractionalization of the Tomonaga-Luttinger (TL) model. But with the exception of quasi-1d Li\textsubscript{0.9}Mo\textsubscript{6}O\textsubscript{17}, whose ARPES line shapes have most of the features predicted for the TL model, in general these anomalies do not show detailed agreement with the simple model lineshapes. This situation has generated skepticism as to the occurrence of fractionalization in quasi-low-d metals and so motivates the use of generalized spectral signatures of fractionalization abstracted from the TL and other models. These signatures are found in the normal metallic states of quasi-1-d K\textsubscript{0.3}MoO\textsubscript{3} and of quasi-2-d systems like NaMo\textsubscript{6}O\textsubscript{17} which display hidden one-dimensionality, and arguably also of quasi-2d superconducting cuprates, if the effects of disorder and the intrinsic nature of k-independent “background” spectral weight are recognized.

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Condensation energy and optical sum rule in strongly coupled superconductors.

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I plan to discuss the origin and the magnitude of the condensation energy in the cuprates. I adopt the magnetic scenario and show that for large spin-fermion couplings, the condensation energy $E_c$ results from the feedback on spin excitations, while the electronic contribution to $E_c$ is positive due to an 'undressing' feedback on the fermions. The same feedback effect accounts for the gain of the kinetic energy at strong couplings. I show that the condensation energy comes from frequencies comparable to the pairing gap, but the behavior of the optical conductivity and the relaxation rate is influenced by strong coupling effects up to much larger frequencies $\sim 1-2eV$. I discuss the relation of these results to the specific heat and optical conductivity data for the cuprates.
Optical Conductivity and Correlated Electron Physics

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The uses of optical conductivity in elucidating the physics of correlated electron systems are outlined. The low energy version of the f-sum rule is derived and its limitations are discussed. The importance of the CMR manganites as a model system is explained. Errors in previous work of the authors are corrected. Additional applications to double perovskites and to the c-axis conductivity of high T_c superconductors and other materials are presented.
Coherence-Incoherence and Dimensional Crossover in Layered Strongly Correlated Metals

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Many “quasi-low-dimensional” materials consist of 1D or 2D building blocks, loosely connected into a 3D whole. Their physical properties are therefore highly anisotropic. Within the Fermi liquid picture, the currents are carried by quasiparticles (QP) and resistivity is proportional to the QP scattering rate. The anisotropy is then a consequence of different hopping integrals for different directions and is usually small and essentially T-independent. Problems with this picture appear when conductivities become uncoupled, i.e. when the anisotropy is strongly T-dependent. We have studied single-particle excitations in ARPES in several layered systems (Sr$_2$RuO$_4$, NaCo$_2$O$_4$ and (Bi,Pb)$_2$Ba$_3$Co$_2$O$_9$) that display a crossover in the c-axis transport, from insulating-like, at high temperatures, to metallic-like at low temperatures, while being metallic over the whole temperature range in the plane. We have found sharp, QP-like excitations in the low-temperature, 3D-like ($\rho_c(T) \propto \rho_{ab}(T)$) phase, and their absence in the effectively 2D, high-temperature phase. Similar dimensional crossovers to coherent, 3D low-temperature states have been identified in some other layered materials. The high temperature superconductors, showing a sharp transition from an incoherent 2D-like ($\rho_{ab}(T) > 0$, $\rho_c(T) < 0$) normal state into the coherent 3D superconducting state will be discussed.

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Mapping the quasiparticle dynamics in high temperature superconductors

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Detailed study of temperature, doping and momentum dependence of the quasiparticle dynamics in high temperature superconductors is shown. We present direct evidence of coupling of quasiparticle to phonons and we discuss the peculiar doping dependence observed in different materials, there where the critical temperature varies by a factor of three or more. The existence of a universal behavior for the nodal Fermi velocity in high temperature superconductors is discussed.
Phonons in high-temperature superconductors

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In good conductors optical phonons are usually screened, and therefore not observed. However, sharp features due to infrared-active (q ∼ 0) modes in the copper-oxygen planes are observed in the optical conductivity of the electron and hole-doped cuprates Pr$_{1.85}$Ce$_{0.15}$CuO$_4$ and YBa$_2$Cu$_3$O$_{6.95}$, respectively.$^1$ Oscillator strengths indicate that despite screening lengths of ≲ 1 Å, the screening of these modes is either poor or totally absent. In addition, there is little indication of electron-phonon coupling. These materials are compared with the two-dimensional material η-Mo$_4$O$_{11}$, in which lattice modes appear suddenly below the charge-density wave transition, but where the Fermi surface is not fully gapped and the resistivity continues to decrease with temperature.$^2$ It is proposed that poor screening in the cuprates originates from charge inhomogeneities in the copper-oxygen planes. The general role that phonons play in these systems will be discussed.

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The role of magnetism in forming the $\alpha$-axis spectral peak at 400 cm$^{-1}$ in high temperature superconductors.

T. Timusk and C. C. Homes

We discuss the peak at 400 cm$^{-1}$, which is seen in $\alpha$-axis conductivity spectra of underdoped high temperature superconductors. The model of van der Marel and Munzar, where the peak is the result of a transverse plasmon arising from a low frequency conductivity mode between the closely spaced planes, fits our data well. Within the model we find that the temperature dependence of the peak amplitude is controlled by in-plane scattering processes. The temperature range where the mode can be seen coincides with $T_\alpha$, the spin gap temperature, which is lower than $T^*$, the pseudogap temperature. As a function of temperature, the amplitude of the mode tracks the amplitude of the 41 meV neutron resonance and the spin lattice relaxation time, suggesting to us that the mode is controlled by magnetic processes and not by superconducting fluctuations which have temperature scale much closer to $T_c$, the superconducting transition temperature.
ARPES of Pb-Bi2212: new views on old problems.

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An overview of the recent photoemission results from the superstructure-free Pb-Bi2212 is presented. We re-address several important for HTSC questions intensively studied by the ARPES in the past, paying a special attention to the effects originating from the c-axis bilayer splitting. In particular, we derive precise set of the tight-binding parameters from the experimentally determined Fermi surfaces and velocities for different doping levels. We show that each of the bilayer split Fermi surface sheets supports a superconducting gap and these gaps are identical within the experimental error bars. The famous peak-dip-hump line shape observed near the \((\pi, 0)\)-point in the superconducting state turns out to be strongly excitation energy dependent and its origin is different in under- and overdoped cases. We find a convincing evidence of the coupling between the electrons and a sharp collective mode below \(T_c\) and estimate the strength of such coupling as a function of doping.

Order induced spectral weight transfer in correlated matter.

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The optical spectra of a number of solids are known to exhibit a shift of spectral weight over a large energy scale when a phase transition occurs. Often this turns out to be an energy scale far exceeding the characteristic temperature of the phase transition. A notorious example of a spectral weight shift over more than 1 eV upon opening a correlated insulating gap is FeSi. In the cuprates spectral weight shift upon entering the superconducting state has been associated with the condensation of Cooper-pairs. In this talk I will discuss some of these examples, and relate the spectral weight shift to the change in physical state. In particular I will discuss if, and to what extent, correlations are responsible for the spectral weight transfer.
Talk by Campuzano – abstract not available at time of printing.
Electro-Optical Studies of Charge-Density-Wave Conductors

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We have observed that the infrared transmittance ($\tau$) of the charge-density-wave (CDW) conductor $K_{0.3}\text{MoO}_3$ (blue bronze) is affected by the depinning of the CDW by an electric field. The changes vary spatially across the sample, with the transmittance increasing at the positive contact and decreasing at the negative (typically by $\sim 0.3$ % for a $\sim 10 \, \mu\text{m}$ thick sample and light polarized perpendicularly to the conducting chains) [1], and were associated with deformations of the CDW in the applied field. We have resolved two contributions to $\Delta\tau$: a sluggish “bulk” component which varies gradually with position and a “fast” contribution which is localized within 100 $\mu\text{m}$ of the contacts [2]. We have used tunable diode lasers to study the $\Delta\tau$ spectra: both spatial contributions have identical spectra consisting of a broad continuum below the CDW gap energy, associated with changes in the density of quasiparticles screening the CDW deformations, and modulations of phonon frequencies (typically $\Delta\nu \sim 0.01 \, \text{cm}^{-1}$), linewidths ($\Delta\Gamma \sim 0.01 \, \text{cm}^{-1}$) and oscillator strengths ($\sim 0.1\%$) [3] No “static” [3] or oscillating [4] changes in spectra that could be associated with either midgap states or oscillations of the quasiparticle density at the CDW “washboard” frequency have been observed. New experiments using an IR microscope are underway on the related CDW compounds, TaS$_3$ and NbSe$_3$. This research has been supported by the U.S. National Science Foundation.

Pressure-Induced Phase Transitions in Ca\textsubscript{n+1}RuO\textsubscript{3n+1} (n=1,2) and TiSe\textsubscript{2}: Raman Spectroscopic Studies

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We have developed a new system capable of making inelastic light scattering measurements simultaneously at high-pressures (100 kbar), low-temperatures (3.2K), and in a magnetic field (8T). We present results from the ruthenates and TiSe\textsubscript{2}, a CDW material. The ruthenate, Ca\textsubscript{3}Ru\textsubscript{2}O\textsubscript{7}, undergoes a metal-insulator transition at 56K and a paramagnetic-antiferromagnetic transition at 48K at atmospheric pressure. Our results reveal a pressure-induced decrease in the metal-insulator transition all the way down to a T~0 phase transition, with a concomitant decrease and eventual destruction of the antiferromagnetic state. The results on the bi-layered Ca\textsubscript{3}Ru\textsubscript{2}O\textsubscript{7} are compared to the single layered Ca\textsubscript{2}RuO\textsubscript{4}, where 5 kbar drives the system into a metallic state while higher pressures reveal a co-existence of ferromagnetism and antiferromagnetism. TiSe\textsubscript{2} develops a commensurate Charge Density Wave (CDW) below 200K at atmospheric pressure and our preliminary results show the destruction of the CDW insulating state at 3.5K at a pressure of 20 kbar.
In-plane charge dynamics of \( \text{La}_{2-x-y}\text{Nd}_y\text{Sr}_x\text{CuO}_4 \) and \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \)

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Using infrared spectroscopy, we found that changes in the in-plane charge dynamics attributable to static stripe order in \( \text{La}_{1.4-x}\text{Nd}_{0.6}\text{Sr}_x\text{CuO}_4 \) or superconductivity in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) \((x = 0.08\) and \(0.125)\) are confined to energies smaller than 200 cm\(^{-1}\). An absorption peak in the low-frequency conductivity of the Nd-doped compounds is suggestive of localization effects due to disorder. This result is consistent with the reduced dimensionality of the electronic transport in the static stripe ordered state. This peak is absent in the Nd-free compound. Below \( T_c \approx 30 \text{ K} \), we observed a distinct decrease of conductivity in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) at \( \omega < 200 \text{ cm}^{-1} \). The energy scale attributable to localization in \( \text{La}_{1.4-x}\text{Nd}_{0.6}\text{Sr}_x\text{CuO}_4 \) appears to be comparable to the magnitude of the superconducting gap in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \). This may explain anomalous sensitivity of superconductivity in La-based cuprates to static stripe ordering. Neither superconductivity nor static stripe ordering have noticeable effect on the depression of the scattering rate at \( \omega < 1000 \text{ cm}^{-1} \) reminiscent of the pseudogap state in other classes of moderately doped cuprates. In \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) the characteristic temperatures and the energy scales of pseudogap and superconducting gap differ about one order of magnitude.

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OPTICAL RESPONSE OF SLIDING CHARGE DENSITY WAVE IN K$_{0.3}$MoO$_3$

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We measured the optical response of K$_{0.3}$MoO$_3$ single crystal along the conducting b-axis in the charge density wave (CDW) ground state and found two optically active modes in the millimeter- and very far-infrared wavelength ranges, at 2 cm$^{-1}$ and 10 cm$^{-1}$, which we assign to the amplitudon and phason CDW excitations, respectively [1]. Applying an external electric field which exceeds the threshold for the nonlinear conductivity due to the CDW depinning, we observe a new mode appearing inbetween, around 4 cm$^{-1}$, which exhibits an additional fine structure. The origin of the mode can be related to the sliding CDW condensate which interacts with the underlying lattice or pinning centers. We observe no changes in the phason mode under bias and only a small loss of the spectral weight is seen for the amplitudon mode.

Reference
Superconducting Magnets and Low Temperature Environments for Spectroscopy Applications

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Oxford Instruments is a world leader in research instrumentation, with over 40 years experience in cryogenics, superconductivity and vacuum technology. Latest products and developments will be presented such as the MicroStat HiRes, the first microscope cryostat to offer sub-micron spatial resolution and interface to a large range of microscopes. The Microstat BT provides a unique environment for magneto-optical microscopy whilst the Spectromag offers a versatile platform magneto-optical spectroscopy in a wide range of application areas.
Soft chiral excitations in triangular antiferromagnets.

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Single crystals of LuMnO$_3$ are studied by Raman spectroscopy. We investigate the magnetic properties of the spin S = 2 two-dimensional (2D) triangular antiferromagnetic Mn - O layers above and below the magnetic transition temperature $T_N \approx 90$ K. Just below $T_N$ a new mode with a soft character appears in the Raman spectra and hardens to about 22 cm$^{-1}$ at $T = 10$ K. Symmetry analysis and magnetic field effects show that the low energy feature is a chiral excitation in the singlet sector transforming like $S_i (S_j \times S_k)$, first discussed as a relevant effective Raman spin operator in the context of 2D insulating Cu-O layers characteristic of high $T_c$ superconductors [1]. The critical exponents obtained by power law fits to the peak energy and weight are close to Monte Carlo predictions for the 3D or XY Heisenberg models in triangular lattices. Surprisingly, if compared to all previous studies, we find that the soft chiral mode is a sharp and well defined magnetic feature at low temperatures. The scattering width and strength demonstrate a distinct relaxation mechanisms and stronger coupling to the radiation field of what may be a ubiquitous feature of frustrated triangular antiferromagnets with out-of-plane spin canting. In the symmetric Raman channel, excitations which could probably be associated with spin pair scattering are observed as a low energy continuum in the 0 – 200 cm$^{-1}$ range in the fluctuation regime around $T_N$.

Far-infrared vibrational spectroscopy of $C_{60}$ in different symmetry-breaking environments

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The high icosahedral symmetry of the $C_{60}$ cage makes for an especially simple vibrational spectrum, consisting of four $T_{1u}$ intramolecular modes [1]. When the $C_{60}$ molecule undergoes symmetry lowering due to excess charge, bonding, or an asymmetric charge environment, changes in the vibrational spectrum are expected. We have studied the far and middle infrared vibrational spectra of $C_{60}$/polymer blends to look for such effects. In composites of $C_{60}$ with polymethylmethacrylate (PMMA), we observe a downshift of the $T_{1u}$ modes with increasing concentration, reflecting the decreasing isolation of the $C_{60}$ molecule. In the same blends, the lowest energy fundamental $T_{1u}$ mode exhibits weak splitting (observable at room temperature but better defined at low temperature), consistent with symmetry lowering and activation of forbidden excitations in these materials [2]. Similar room temperature splitting of $T_{1u}$ (1) is observed in $C_{60}$/polystyrene blends. When films of $C_{60}$/PMMA are heated, the decomposition of PMMA is delayed to higher temperatures than in the pristine polymer [3]. By tracking the vibrational spectrum of $C_{60}$/PMMA at different heat treatment temperatures, we can attribute the increased stability of the polymer to bonds formed between it and $C_{60}$, as evident in the growth of new methanofullerene vibrational modes.

Far-infrared and submillimeter-wave conductivity in electron-doped cuprate $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$

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We performed far-infrared and submillimeter-wave spectroscopy in the electron-doped cuprates $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$. The onset of the absorption in the superconducting state appears gradual in frequency and is inconsistent with a BCS gap. Instead, a narrow quasiparticle peak is observed at zero frequency and a second peak at finite frequencies. The infrared conductivity as well as the suppression of the quasiparticle scattering rate below $T_c$ are qualitatively similar to the results in the hole-doped cuprates.

In addition, the conductivity has been investigated for $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ films tilted $1^\circ$ off from the $ab$-plane. The effective conductivity measured in this geometry reveals an intensive peak at finite frequency ($\nu \sim 50\text{ cm}^{-1}$) even in the normal state, which is due to a mixing of the in-plane and out-of-plane responses. The peak disappears for the pure in-plane response transforming into a Drude-like contribution. Comparative analysis of the mixed and the in-plane contributions allows to extract the $c$-axis conductivity which shows a Josephson plasma resonance in the superconducting state.
The far-infrared reflectance of heavy-fermion superconductor UBe\textsubscript{13} has been measured as a function of temperature. For the first time measurements are carried out below the superconducting transition temperature. At low temperatures the coherent state is characterized by an optical conductivity that exhibits a renormalized Drude peak at low frequencies superimposed on a broad background, giving rise to a minimum near 75 cm\textsuperscript{-1}. Invoking an extended Drude analysis gives rise to the emergence of a peak in the frequency-dependent scattering rate which, in analogy with other heavy-fermion materials, can be associated with the characteristic energy separating coherent behaviour from single impurity scattering. Both the energy of the peak and the temperature at which the behaviour manifests itself are found to correspond to a temperature of ~45 K although the resistivity of UBe\textsubscript{13} continues to increase with decreasing temperature down to ~2.5 K.

The superconducting state is characterized by a noticeable decrease in spectral weight over a wide frequency range and a scattering rate which is depressed at low frequencies, suggesting that the formation of the condensate alters dramatically the interaction between the heavy electrons.

Work at Brock was supported by the Natural Sciences and Engineering Research Council of Canada and that at Los Alamos under the auspices of the U.S. Dept. of Energy.
Magneto-optical far-infrared Spectroscopy at U12IR beamline of the NSLS

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We describe the magneto-optical far-infrared spectroscopy equipment installed at the U12IR beamline of the National Synchrotron Light Source (NSLS) at Brookhaven National laboratory. The instrumentation allows to study optical properties of materials in magnetic fields up to 16 Tesla and is well suited for observing and measuring electron spin resonance in solids. We present the demonstration experiments on the paramagnetic and antiferromagnetic state of LaMnO₃.

² Work at the NSLS is supported by U.S. DOE under contract DE-AC02-98CH10886.
Structural Incommensurability in the Spin-Peierls Compound CuGeO₃: High-Field Electron Spin Resonance Study.

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A new facility for high-field millimeter and submillimeter wave spectroscopy was completed at the NHMFL (DC Field Facility), Tallahassee. A technical description and first applications of the facility are reported. In the spin–Peierls compound CuGeO₃, the dimerized – solitonlike – plane wave structural phase transitions are fully monitored in a frequency range of 140-700 GHz (≫ 4.7-23.3 cm⁻¹), and in fields up to 25 T. The ESR data confirm a continuous change from the solitonlike lattice close to the dimerized – incommensurate phase transition to the plane wave phase at higher fields. Significant line broadening observed in the vicinity of the dimerized – solitonlike and solitonlike – plane wave phase transitions (Bₜₜ ≫ 12.5 T and Bₚₚ ≫ 16 T, respectively) is explained in terms of the incommensurate superstructure evolution in CuGeO₃ in high magnetic fields. In addition, ESR spectra for 0.4%Si-doped samples are studied. The doping suppresses magnetic hysteresis effects, observed with ESR in undoped CuGeO₃ in the incommensurate phase. Remarkably, CuGeO₃+0.4%Si undergoes transition into the plane wave phase at higher fields (Bₚₚ ≫ 20 T) without passing through the intermediate solitonlike lattice phase. Difference in the high-field behavior of the pure and diluted CuGeO₃ is discussed.
Spin-dependent effects in the optical study of hexagonal LuMnO$_3$

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The colossal magnetoresistance compounds based on doped pseudo-cubic LaMnO$_3$ have excited much attention because of their interesting physical properties and potential applications. Another series of RMnO$_3$ materials, where R is one of the heavier lanthanides (Ho, Er, Tm, Yb, and Lu) have smaller radius R$^{3+}$ ions and crystallize in the hexagonal lattice. The hexagonal manganites are interesting as a limiting case of a huge Jahn-Teller distortion of the orthorhombic MnO$_6$ complex, and they are examples of multiferroics — they are both ferroelectric ($T_c$~900 K) and antiferromagnetic ($T_N$~90 K) and the antiferromagnetism is strongly frustrated. One of the manifestations of coupling between ferroelectric and magnetic order parameters is an anomaly in the temperature dependence of the static dielectric constant. In the present work, we analyze the contributions of the phonons and the electronic transitions to the dielectric function. We find that the phonons are mainly responsible for the anomaly in the static dielectric constant. We also find that both Mn phonons and the lowest 1.7 eV electronic excitation exhibit identical temperature dependent shifts in their resonance energies and that these shifts have the character of the nearest neighbor spin-spin correlation function $<S_iS_j>$. We will also discuss the Mn d-level splitting in this system, the assignment of observed electronic transitions and the mechanism of their temperature dependencies. This work has been supported by the NSF-MRSEC at the University of Maryland, DMR#0080008.
Exploring the phase diagram of cuprates via Inelastic Light Scattering

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We report electronic Raman scattering studies on high-$T_c$ cuprates. Inelastic light scattering has proven itself to be a powerful tool for exploring the phase diagram and the symmetry of the order parameter of the cuprates. Indeed by choosing different incoming and outgoing light polarisations Raman scattering allows to study electronic excitations in the the normal and superconducting state in different symmetries (known as $B_{1g}$, $B_{2g}$ and $A_{1g}$) corresponding to different directions in k-space: the $B_{1g}$ and $B_{2g}$ symmetries probe respectively the $(\pi, 0)$ (hot spot) and $(\pi, \pi)$ (cold spot) directions while the $A_{1g}$ symmetry is fully symmetric and thus probe the entire Fermi surface. Here we explore the cuprates phase diagram in both bilayer Y-123 and single-layered Hg-1201 focusing on the energy scales revealed by Raman scattering in the superconducting state. Using Ni impurity in the Y-123 system we show that the superconductivity-induced peak observed in the fully symmetrical $A_{1g}$ channel tracks the energy of the resonance peak observed by inelastic neutron scattering. Our data indicates the presence of two distinct energy scales in the optimally doped regime of the cuprates, one is seen in $B_{1g}$ symmetry and corresponds to the maximum of the $d_{x^2-y^2}$ gap while the other one (seen in $A_{1g}$ symmetry) intriguingly tracks that of the neutron resonance. In the underdoped regime the Raman scattering intensity shows an abrupt decrease of intensity in the $B_{1g}$ and $A_{1g}$ channel in Y-123 and Hg-1201. In both compounds superconductivity-induced peaks disappear immediately below optimal doping in both channels while intensity in the $B_{2g}$ channel seems unaffected. The similar behavior towards underdoping of orthorhombic bilayer Y-123 and tetragonal single-layered Hg-1201 points out the existence of an generic transition occurring in the superconducting state of the cuprate near optimal doping at the hot spots of the Fermi surface.

Searching for the Slater Transition in the Pyrochlore Cd$_2$Os$_2$O$_7$
With Infrared Spectroscopy

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Infrared reflectance measurements were made on the single crystal pyrochlore Cd$_2$Os$_2$O$_7$ in order to examine the transformations of the electronic structure and crystal lattice across the boundary of the metal insulator transition (MIT) at $T_{MIT} = 226$K. All predicted IR active phonons are observed in the conductivity over all temperatures and the oscillator strength is found to be temperature independent. These results indicate that charge ordering plays only a minor role in the MIT and that the transition is strictly electronic in nature. The conductivity shows the clear opening of a gap with $2\Delta = 5.2k_BT_{MIT}$. The gap opens continuously, with a temperature dependence similar to that of BCS superconductors, and the gap edge having a distinct $\sigma(\omega) \sim \omega^{1/2}$ dependence. All of these observables support the suggestion of a Slater transition in Cd$_2$Os$_2$O$_7$. 
Far-infrared Microspectroscopy with Synchrotron Radiation

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Synchrotron radiation serves as an IR source that is several orders of magnitude brighter than traditional thermal sources, including in the far-infrared spectral range. High-brightness is crucial for throughput limited techniques such as microspectroscopy. Whereas a conventional source is not sufficient below a few hundred wavenumbers, the synchrotron source allows infrared microspectroscopy to be performed into the THz range. We report the performance of an IR microspectrometer, installed at NSLS beamline U4IR, at frequencies below 30 cm$^{-1}$ and demonstrate its capabilities for performing transmission and reflection studies of solids only 100 microns in size.

Work at the NSLS is supported by U.S. DOE under contract DE-AC02-98CH10886, and LDRD funds at Brookhaven Lab.
Spin gap excitations in $\alpha$-NaV$_2$O$_5$ studied by far-infrared spectroscopy

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Sodium vanadate, $\alpha$-NaV$_2$O$_5$, is a quarter-filled spin-ladder compound which undergoes a phase transition at $T_c = 34\, \text{K}$. Below $T_c$ a new phase with a different lattice, charge and magnetic order emerges. New lattice order appears as a doubling of the unit cell along ladder legs (b-axis) and rungs (a-axis) and quadrupling in the c-direction. Rung-centered $V^{4+}$ charge order changes to zig-zag order of $V^{4+}$ and $V^{5+}$ charges. In the magnetic excitation spectrum spin gap opens and two branches are observed by inelastic neutron scattering. The lower branch has been identified as a singlet to triplet excitation by high field electron spin resonance. We study lattice, charge and magnetic excitations by far-infrared spectroscopy between 3 and $200\, \text{cm}^{-1}$ and in magnetic fields up to $12\, \text{T}$. In zero magnetic field the triplet resonance appears at $65.4\, \text{cm}^{-1}$. By applying magnetic field we observe all three transitions from the singlet ground state to the triplet sublevels $S_z = -1, 0, +1$ and study their selection rules in Faraday and Voigt configurations. The zero-field splitting between the triplet state sublevels is less than $0.2\, \text{cm}^{-1}$. It appears that the dominant interaction responsible for the oscillator strength of otherwise forbidden singlet to triplet transition is the coupling of the triplet state to other excitations.
The dynamics of pressure- and field-tuned transitions in strongly correlated systems: Raman spectroscopic studies*

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I will describe recent Raman spectroscopic studies of spin-, charge-, and orbital-organization near low temperature pressure- and field-tuned phase transitions in various strongly correlated systems. In particular, I will describe Raman studies of: (a) the evolution of spin- and lattice dynamics through the T~0 pressure-tuned collapse of the Mott-like state in layered ruthenates, which reveal evidence for a pressure-tuned 'disordered' phase involving the coexistence of antiferromagnetic insulator and ferromagnetic metal phases; and (b) the temperature- and field-induced nucleation and evolution of spin polarons near the Curie temperature of magnetic semiconductors such as EuO and EuB$_6$, which demonstrate the importance of spin cluster formation on the "colossal" sensitivities of these systems to applied magnetic fields. These studies illustrate the power of light scattering for probing the dynamics of pressure- and field-tuned quantum phase transitions in correlated systems.

*Work supported by the Department of Energy under Grant DEFG02-96ER45439.
Electromagnetic response of cuprates in the regime of spin/charge stripe order

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We report on the anisotropy of both in-plane electronic conductivity and lattice dynamics examined in the lightly doped phase of a prototypical high-$T_c$ system La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.03$ and $0.04$). Using infrared spectroscopy, we show that spin self-organization in untwinned La$_{2-x}$Sr$_x$CuO$_4$ crystals has profound consequences for the dynamical conductivity. Our data for phonon modes indicate that spin stripes are accompanied by quasi-static charge ordering. We also find significant anisotropy of the electronic response within the CuO$_2$ planes with enhancement of the conductivity along the stripe direction ($\sigma_a/\sigma_b \leq 2$) in the limit $T, \omega \rightarrow 0$. The Drude-like frequency dependence of the optical conductivity at moderate temperatures and the linear progression of electronic spectral weight with doping ($0.03 < x < 0.125$) suggest band-like electronic transport even in lightly doped LSCO. The peak feature observed in $\sigma_1(\omega)$ at finite frequencies below 80 K can be attributed to localization of charge carriers due to the reduced dimensionality of the electronic transport in the stripe ordered state$^1$. The energy scales associated with the localization of charge carriers for transport parallel and perpendicular to the stripes differ by a factor of two. The moderate anisotropy in the charge dynamics of lightly doped LSCO is consistent with the idea of transverse mobility of stripes. Our results uncover a complex electronic behavior due to stripes that is beyond an idealized picture of strictly 1D charge channels embedded into AFM insulator.

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Raman spectroscopy in the superconducting state: comparisons of n-type and p-type cuprates

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Raman spectroscopic studies of n-type Pr$_{1.85}$Ce$_{0.15}$CuO$_4$ and Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ crystals reveal features unique among the cuprate superconductors. When compared with the p-type cuprates Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ and La$_{2-x}$Sr$_x$CuO$_4$, we observe that the pairing is much weaker, yielding $2\Delta_{\text{max}}/k_B T_c = 4.1$ for n-type but $2\Delta_{\text{max}}/k_B T_c = 8$ for p-type. While both types exhibit gap anisotropy consistent with $d_{x^2-y^2}$ order parameter including nodes along $\Gamma \rightarrow (\pi, \pi)$, the n-type have $2\Delta_{B2g} > 2\Delta_{B1g}$, contrary to p-type. We interpret this using a non-monotonic $d_{x^2-y^2}$ form for the n-types. The effect of applied magnetic field is also different, with the $2\Delta$ feature shifting toward lower energy in the n-type while only the $2\Delta$ intensity changes for the p-type.

References:
Local Stripe Order in the Cuprate Superconductors

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The implications of a local tendency to stripe order in the high temperature superconductors are explored. Specifically, as time permits, I will discuss the following topics: 1) The nature of the broken symmetry phases that occur naturally in such systems; 2) Experimental strategies for detecting local (or fluctuating) stripe order; 3) Some aspects of the interplay between stripe and superconducting order, with special emphasis on what aspects remain unsettled; 4) Some of the effects of local stripe order on other properties of the system, especially the single particle spectrum.
Talk by Sawatzky – abstract not available at time of printing.
Fourier-Transform Scanning Tunneling Spectroscopy (FT-STS)
- A new window on the electronic structure of correlated-electron materials.

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When elastic scattering mixes the electronic eigenstates $\vec{k}_1$ and $\vec{k}_2$, of a system, a quantum interference pattern appears in the norm of the wavefunction at the wavevector $\vec{q} = \vec{k}_2 - \vec{k}_1$ (or equivalently at spatial wavelength $\lambda = 2\pi/|\vec{q}|$. This produces modulations with wavelength $\lambda_i$ in the local density of electronic states LDOS(E) which can be observed by STM as modulations of the differential tunneling conductance. STM studies of such conductance modulations allowed the first direct probes of the quantum interference of electrons in metals[1].

Equivalent phenomena have long been predicted to occur in the high-$T_c$ cuprates when quasiparticles are scattered from impurities[2]. I will introduce high-resolution Fourier-transform scanning tunneling spectroscopy (FT-STS) as a new spectroscopic technique for study of cuprate quasiparticles[3]. By studying the interference patterns produced by scattering, we report discovery of a characteristic ‘octet’ of quasiparticle states that determine the quasiparticle scattering processes. By analyzing the wavevectors of these interference patterns we determine the normal-state Fermi-surface and the superconducting energy gap $|\Delta(\vec{k})|$. These are in excellent agreement with angle resolved photoemission spectroscopy (ARPES). This provides the first direct connection between the tunneling and photoemission spectroscopies in the cuprates and, by virtue of the good agreement, strongly supports the validity of the respective conclusions.

Broadband Microwave Spectroscopy of YBCO


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Employing a microwave spectroscopic technique that uses bolometric detection, we have been able study the microwave conductivity in the superconducting state of YBCO in unprecedented detail, over the range 0.2 to 20 GHz, and T = 1.3 to 6.7 K. For YBCO$_{6.5}$ in the highly oxygen ordered Ortho-II phase ($T_c = 55$K), we have clearly observed for the first time the characteristic frequency and temperature dependencies of weak (Born) scattering in a d-wave superconductor. For YBCO$_{6.99}$, the Born scattering limit is not seen until the temperature is below 4K, a result that is not understood at present.

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Anomalous far-infrared response in (La,Sr)$_2$CuO$_4$

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Recently there have been accumulated the experimental results indicating that the spectral weight over a wide energy range contributes to the superconductivity condensate in high-$T_c$ cuprates.[1-3] The advantages of our method are the growth of a large crystal with single grain, the accurate spectrum measurement over a wide energy range down to 8cm$^{-1}$, and the simultaneous measurement of $\mu$SR on the same sample used for optical measurement. As a result, we found a substantial suppression of far-infrared missing area in La$_{1.85}$Sr$_{0.15}$CuO$_4$, in comparison with the superfluid density determined by $\mu$SR, which implies a high-energy spectral contribution to the superconducting condensate. Measuring a spectrum down to a very low frequency, we also found an anomalous Drude-like peak remaining in the superconducting state that affects seriously the estimation of missing area. This residual conductivity is commonly observed in almost all high-$T_c$ cuprates, and might be the evidence for the inhomogeneous electronic state, namely, coexistence of the normal and superconducting regions.

This is a result of collaboration with Y. Fudamoto at SRL-ISTEC, T. Kakeshita and S. Uchida at the Univ. of Tokyo, B. Gorshunov and M. Dessel at University Stuttgart. This work was supported by New Energy and Industrial Technology Development Organization in Japan.

References  
Electrodynamics at microwave frequencies of superconducting cuprates and diborides

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A systematic set of microwave measurements on several oxides - cuprate superconductors, CMR manganites and striped nickelates - has yielded new insights into their electrodynamic properties. A cohesive picture is emerging that the electrodynamics at these sub-optical frequencies is dominated by collective charge transport and also manifests signatures of lattice instabilities at specific temperatures.

In the superconducting state below $T_c$ of all the cuprates, including BSCCO, HgBa$_2$Ca$_2$Cu$_3$O$_{8+y}$, HgBa$_2$CuO$_{4+d}$, Tl$_2$Ba$_2$CuO$_{6+d}$ and YBa$_2$Cu$_3$O$_{7-\delta}$, the microwave data clearly are incompatible with the quasiparticle response of a pure d-wave superconductor. Instead, we have successfully modeled the data in terms of a non-quasiparticle scenario, according to which the electrodynamic response is ascribed to a collective mode, similar to the dynamic response of a density wave in low dimensional materials. Such a density wave can well arise from an inhomogeneous electronic state.

The collective mode dynamics is also observed above $T_c$, in terms of an anomalous plasmon-like response implying negative permittivities (Re($\epsilon(\omega)$) < 0) at microwave frequencies in the pseudogap state of the cuprate superconductors. The microwave plasmon arises from a charge collective mode characterized by a low plasma frequency and extremely low damping, distinctly different from those observed at optical frequencies.

Another feature common to several oxides, including the parent members of the cuprate superconductors and the nickelates, is the observation of dielectric transitions at characteristic temperatures that we associate with lattice instabilities. In La$_2$CuO$_{4+x}$ and La$_{5/3}$Sr$_{1/3}$NiO$_4$, these occur at common temperatures 32K and 245K, and are signatures of local lattice octahedra instabilities occurring in these isostructural perovskite oxides. Similar dielectric transitions are also observed in non-superconducting YBa$_2$Cu$_3$O$_{6,0}$ at 65K and 110K - the latter correlates well with other reports of changes in the buckling angle and NQR reports of CDW formation. The microwave results are correlated with a variety of other measurements and reveal new aspects of the phase diagram of the perovskite cuprates and nickelates. These results also indicate that inhomogeneous electronic states, such as charge stripes and oxygen ordering, are strongly connected to underlying lattice instabilities.

MgB$_2$ presents a considerably different picture in terms of the microwave response in the superconducting state. Sample quality has been rapidly improving and in some high quality films we observe directly an exponential dependence on temperature of the surface resistance and the penetration depth. The resulting gap obtained from the measurements is approximately 1.92 meV, substantially smaller than the mean field value, but consistent with value of the smaller of the two gaps reported by other techniques.

Work supported by Office of Naval Research
Raman-scattering evidence for a metal-insulator transition in strongly overdoped cuprates

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We discuss results from inelastic light scattering experiments on electronic excitations in cuprates over a wide range of doping. At high doping well beyond the level optimal for \(T_c\), the quasiparticle dynamics are isotropic and similar to those expected for conventional metals. At lower doping strong anisotropies in the quasiparticle relaxation and pronounced discrepancies between single- and many-particle probes develop. The results can be interpreted in terms of an unconventional metal-insulator transition with an anisotropic gap which disappears for doping levels above approximately 0.22 holes/CuO\(_2\). We try to make a connection between this phenomenon and various other anomalies in underdoped cuprates such as the pseudogap or charge ordering.
LOW-ENERGY EXCITATIONS IN MOLYBDENUM SULPHIDE NANOTUBES

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Molybdenum sulphide nanotubes MoS$_{2-y}$ grow in bundles$^1$ with unusually weak coupling between individual nanotubes$^2$. Consequently, the electrodynamical properties of these NTs are expected to exhibit strongly one-dimensional behaviour, more so than either quasi-1D crystals, or even carbon nanotube ropes. In many respects the chalcogenide NTs show similar properties as single-wall carbon nanotubes (e.g. field emission, strong mechanical properties etc.). However, transport and magnetic properties of pristine and doped$^3$ nanotubes – which will be reviewed - show some peculiar effects, such as a giant magnetic susceptibility, a tendency toward ferromagnetic spin ordering and strongly anisotropic transport properties. The undoped NTs are apparently metallic along the tube axis, but insulating when transport is measured perpendicular to the tube axis. The implications of these observations will be discussed in terms of the expected physics of 1D systems.


2. A. Kis et al., to be published (2002)

JAHN-TELLER EFFECT IN A MONOVALENT FULLERIDE SALT STUDIED BY INFRARED SPECTROSCOPY

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Jahn-Teller (JT) distortion of fulleride molecular ions enters in many recent theories of electronic structure of these materials. Direct structural evidence for a distortion is scarce, however, because it is expected to be around the detection limit of diffraction methods, and because the effect may be dynamic. Vibrational spectroscopy can contribute to the detection of structural changes through the differences in molecular symmetry.

We will present infrared spectra of the insulating fulleride salt (Ph₄P)₂C₆₀I and their temperature dependence, with the intention to determine the changes in molecular symmetry from the splitting of vibrational bands. Based on our earlier work on the infrared spectra of fullerene polymers and their explanation by quantum molecular dynamics calculations, we suggest that the symmetry point group of the C₆₀⁻ anion in the low-temperature phase is D₂h, the JT distorted state with the lowest possible symmetry. Both fulleride and counterion modes show changes around 150 K, where a weak structural phase transition has been found recently.

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Dynamical Response of quasi one-dimensional Mott and CDW insulators.

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We calculate the low temperature spectral function of one-dimensional incommensurate charge density wave (CDW) states and half-filled Mott insulators (MI). The former is characterized by a gap in the spin sector and a gapless charge sector. At $T = 0$ there are two dispersing features associated with the spin and charge degrees of freedom respectively. We show that already at very low temperatures (compared to the gap) one of these features gets severely damped. We comment on implications of this result for photoemission experiments. We present a scenario for the evolution from a Mott insulating state to a Fermi liquid as the interchain hopping is increased in a system of weakly coupled chains.

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New low-energy phenomena probed with terahertz pulses

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Both the coherent and pulsed nature of short terahertz (THz) electric field transients open new pathways to investigating low-energy excitations and their dynamics. This talk presents studies that probe, in solids, intrinsic excitations spanning the range from collective phenomena to atom-like transitions. (i) conductivity gap in superconducting MgB$_2$: we study the complex, frequency-dependent THz conductivity of MgB$_2$ films. The imaginary part exhibits an inductive response due to the emergence of the superconducting condensate. The real part - obtained on equal footing - reveals the opening of a superconducting energy gap characterized by strong depletion of the oscillator strength near 5 meV. An anomalously small gap ratio $2D/k_B T_c$ 2 points to complex physics in accordance with the much-discussed two-gap model. (ii) exciton dynamics probed via internal THz transitions: a novel, ultrafast THz probe is employed to investigate the dynamical interplay of optically-induced excitons and unbound electron-hole pairs in GaAs quantum wells. Resonant creation of excitons induces a low-energy oscillator linked to transitions between the internal exciton degrees of freedom. Time-resolved pump-probe studies under various conditions reveal population relaxation, thermal ionization and the formation of excitons from unbound electron hole pairs.
Optical Studies of Electron-boson Interactions in Superconductors

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Electron-phonon coupling and in general electron-boson coupling, are essential for the formation of Cooper-pairs in superconductors. Infrared spectroscopy is a direct probe to study such interactions and their influences on superconductivity. In this talk, three representative superconductors will be discussed: Nb (Tc = 9.3 K), MgB2 (Tc = 39.6 K) and OP Bi2212 (Tc = 91.5 K). The robustness of a second derivative technique will be established first in Nb by determining \( a^2F(w) \) in this classic BCS superconductor. The electron-phonon interaction is more intriguing in MgB2 and \( l_\nu = 0.13 \) is determined from our optical data. This surprising result can be understood since the electron-phonon interaction is highly anisotropic in MgB2. An electron-boson spectral function peaked at 43 meV is found in OP Bi2212 using the second derivative technique and is shown to be involved in the pair formation. In contrary to the recent suggestion, our data indicate that the zone-boundary phonons (~ 80 meV) alone are not sufficient to explain the superconducting properties in high-Tc cuprates and our data are consistent with the spin-fluctuation models.

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Ferromagnetism and Superconductivity in Boride Systems

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The boride materials raised quite a bit of interest recently because of the wealth of interesting and puzzling phenomena characterizing their physical properties. Ferromagnetism at high Curie temperature (like in some rare earth hexaborides or CaB$_2$C$_2$) or giant magnetoresistive behaviour, as in the ferromagnet EuB$_6$, as well as superconductivity in MgB$_2$ are some examples. In my talk, particular emphasis will be devoted to the discussion of the electrodynamic response of EuB$_6$ and of the novel MgB$_2$ superconductor.

I will present our magneto-optical investigations (reflectivity and Kerr rotation) of EuB$_6$ from the far infrared up to the ultraviolet spectral range as a function of temperature and external magnetic field. We find a remarkable blue shift of the plasma edge both with decreasing temperature at zero field or with increasing field at constant temperature. In coincidence with the plasma edge we observe a sharp structure in the Kerr rotation, which gets stronger with increasing field at low temperature. From the Kerr rotation we gain spectroscopic insight about magnetism involving both the itinerant and localized charges.

I will also present magneto-optical reflectivity results in the basal-plane of the hexagonal MgB$_2$. The data were collected on a mosaic of MgB$_2$ single crystals with $T_c$=38 K from the ultraviolet down to the far infrared as a function of temperature and magnetic field oriented along the c-axis. In the far infrared there is a clear signature of the superconducting gap with a gap-ratio $2\Delta/k_BT_c$~1.2, well below the weak-coupling value. The gap is suppressed in an external magnetic field, which is a function of temperature. We extract the upper critical field $H_{c2}$ along the c-axis. The temperature dependence of $H_{c2}$ is compatible with the Helfand-Werthamer behaviour.
Talk by Marsiglio – abstract not available at time of printing.
Electron Spin Resonance and Microwave Conductivity Studies of MgB₂

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We report the observation of conduction electron spin resonance (CESR) in MgB₂ fine powder samples using multifrequency (3-225 GHz, 0.1-8 T) ESR technique. The normal state spin-susceptibility, \( \hat{\chi}_S \), measured from the CESR signal intensity agrees with band structure calculations, confirming that MgB₂ is a weakly correlated metal. The CESR linewidth, that measures the spin-lattice relaxation, follows the normal state resistivity, which also shows a conventional metallic behavior. Below \( T_c \), we observe a peculiar magnetic field dependence of the CESR signal: above 75 GHz (2.7T Zeeman field) we find that part of the sample is in the normal state in addition to the superconducting material. The normal state fraction grows with magnetic field at the cost of the superconducting one. We conclude that a significant \( H_{c2} \) anisotropy (\( H_{c2}^{a,b} : H_{c2}^c = 8 \)) can explain the observations. Field and temperature dependent magnetization data confirms this explanation [1]. Since our original report on powders, this has been confirmed by measurements on good quality single crystals. We also observe an anomalously large magnetic field induced \( \hat{\chi}_S \) below \( T_c \) at low magnetic fields, where only the superconducting phase is present. At the lowest magnetic field (0.14 T, 0.38 GHz), the opening of the gap is observed and the temperature dependence of \( \hat{\chi}_S \) is compatible with the two-gap superconductivity model suggested for MgB₂. For fields above 0.2 T there is an anomalously large field induced \( \hat{\chi}_S \) and microwave conductivity that can not be explained by the current theories.
Condensate and quasiparticle dynamics in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ and NbN probed by time-resolved THz spectroscopy

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The dynamics of charge and spin on short timescales is of increasing relevance for understanding the pairing mechanism in high-$T_c$ cuprates and the unusual metallic state from which it forms$^{1,2,3}$. Here, the ultrafast dynamics of the superconducting condensate in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO) and in NbN are directly probed via optical-pump terahertz-probe spectroscopy. We discuss the picosecond reformation kinetics of the condensate and the relaxation of quasiparticles in each case. For optimally-doped BSCCO, after optical excitation an ultrafast change due to nonequilibrium reduction of the condensate density is followed by a picosecond recovery. The recovery rate increases strongly both with temperature and excitation density. Significantly, for low density and low temperature, the dynamics is seen to be nonexponential and approaches a bimolecular kinetics. The observation of a bimolecular picosecond recovery is surprising in view of the expected bottleneck due to the d-wave energy-linear density of states near the superconducting gap nodes. This surprising behavior in BSCCO can be contrasted with that of conventional superconductors. In particular, it can be seen from our studies of NbN films that the condensate recovery occurs over comparatively long timescales (~600 ps) consistent with the limitations imposed in the phonon-bottleneck scenario. Moreover, the dynamics in NbN is exponential with a relaxation time independent of excitation density. Additional features of the complex conductivity dynamics of the conventional superconductor around its energy gap are discussed which further throw into relief the novel physical mechanisms at play in the cuprates.

Doping Dependence of the Coupling to MFL Fluctuations from Bi$_2$Sr$_2$CaCu$_2$O$_{8-\delta}$ Reflectance Data

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We use 300 K Bi$_2$Sr$_2$CaCu$_2$O$_{8-\delta}$ reflectance data to investigate the coupling to Marginal Fermi Liquid (MFL) fluctuation. We calculate the coupling constant at 300 K from the reflectance data by using the MFL theory. The coupling constant, $\lambda(p)$, appears to vary smoothly with the hole doping level, $p$. Although we analyze reflectance data at 300 K, a temperature that is well above the superconducting transition temperature ($T_c$) and the pseudogap temperature ($T^*$), we can observe some indications of phase changes. We introduce a doping dependent parameter $\alpha$ to see how well the reflectance of a given system agrees with the MFL theory. The parameter $\alpha$ is a constant close to the MFL value ($\alpha=1$) in the underdoped region ($p \leq 0.16$) and a little bit higher and more variable in the overdoped region ($p \geq 0.16$); this difference between underdoped and overdoped cuprates is probably related to the pseudogap. We also observe a possible sign of a competing new phase setting in from a rapid drop of $\alpha$ above a doping level of $p = 0.21$.

PACS numbers:

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The optical response of the spin-ladder compound Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$ (x=0, 3, 9) has been studied in a broad frequency range from radiofrequencies up to the infrared [1]. At temperatures below 250 K an absorption peak appears around 12 cm$^{-1}$ in Sr$_{14}$Cu$_{24}$O$_{41}$ for the radiation polarized along the chains/ladders, E||c. In addition, a strongly temperature dependent dielectric relaxation is observed in the kHz - MHz range. We explain this behavior by a charge density wave which develops in the ladders subsystem and produces a phason mode pinned at 12 cm$^{-1}$. With increasing x the mode shifts up in frequency and eventually disappears for x=9 because the dimensionality of the system crosses over from one to two dimensions, giving way to the superconducting ground state under pressure.

Reference
Assignment of the Raman spectra of some deuterated-BEDT-TTF superconductors

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We present the room-temperature Raman spectra of both the protonated and deuterated forms of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, and β-(BEDT-TTF)$_2$I$_3$. Along with data for the neutral BEDT-TTF molecule these spectra are used to assign the many features in the spectra of the deuterated compounds.
Far-infrared studies of charge ordering and superconductivity in layered organic conductors $\alpha$-(BEDT-TTF)$_2X$

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A comparative study of the low-energy optical properties of the metallic $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ and the superconducting $\alpha$-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ was performed by polarized reflection measurements at temperatures $5 \leq T \leq 300$ K. The optical conductivity of $\alpha$-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ steadily increases with decreasing frequency as expected for a metal. However, for the non-superconducting K-analog we find in both orientations of the highly conducting plane the gradual development of a pseudogap in the optical conductivity around 200 cm$^{-1}$ as the temperature is reduced below 200 K; a narrow Drude-like contribution remains. We assign the observed behavior to the proximity of a correlation induced metal-insulator transition.

The far-infrared in-plane reflectivity of superconducting $\alpha_t$-(BEDT-TTF)$_2$I$_3$ was measured down to very low frequencies (10 cm$^{-1}$). We determined an energy gap of 25 cm$^{-1}$ which opens below $T_c = 8$ K; $2\Delta/k_BT_c = 4.4$ is in good agreement with moderate coupled BCS superconductor. A considerable low-frequency absorption remains in the superconducting state. From our analysis we estimated a London penetration depth $\lambda_L \approx 6 \mu$m.

Additionally, we present data on the optical properties of the organic metal (BEDT-TTF)$_4$Ni(dto)$_2$ which strongly indicate that a correlation gap opens at low temperatures.

New Josephson Plasma Modes in Underdoped YBa$_2$Cu$_3$O$_{6.6}$ Induced by Parallel Magnetic Field

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We have measured c-axis reflectivity of underdoped YBa$_2$Cu$_3$O$_{6.6}$ under in-plane magnetic fields up to 7T [1]. The Josephson plasma edge at ~40 cm$^{-1}$ for H=0 splits into two edges under the parallel fields, indicating an existence of two kinds of Josephson couplings in the energy scale of the inter-bilayer coupling. We presume that the two modes originate from the insulating layers with and without Josephson vortices. The 400 cm$^{-1}$ peak of optical conductivity gains the spectral weight under the parallel magnetic field, supporting an idea that this mode involves the intra-bilayer Josephson coupling as has been proposed by van der Marel and A. Tsvetokov [2] and D. Munzar [3]. We found that the sum-rule is conserved between the superconducting condensate weight, new (transverse) Josephson plasma mode, and the enhancement of the 400 cm$^{-1}$ peak.

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Doping dependence of the Fermi Surface in the electron doped superconductor Nd$_{2-x}$Ce$_x$CuO$_{4+x}$

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Using the mean field spin density wave (SDW) formalism augmented with self-consistent renormalization (SCR) computations within the framework of the one-band $t-t'-t''-U$ Hubbard model Hamiltonian, we explain the doping dependence of the Fermi surface (FS) maps and spectral intensities determined recently in the electron doped compound Nd$_{2-x}$Ce$_x$CuO$_{4+x}$ (NCCO) via high resolution ARPES measurements$^1$.

The crossover of the FS from small electron pockets centered at $(\pi,0)$ at low dopings, to large sheets at optimal doping is a consequence of the collapse of the correlation induced Mott pseudogap just above optimal doping. This collapse reflects in part a reduction of the effective Hubbard coupling constant $U$ with increasing electron doping.

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Investigation of Superconducting State of $\text{Sr}_2\text{RuO}_4$ by Far Infrared Spectroscopy

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The ratio of the far infrared reflectance of $\text{Sr}_2\text{RuO}_4$ in the superconducting state to that in the normal state has been measured along the a- and c-axes using an ac-plane oriented crystal and within the ab-plane using an ab-plane oriented crystal. The measurements were taken above and below the 1.42 K superconducting transition temperature of both the samples using a helium-3 cryostat and step-and-integrate Martin-Puplett type polarizing interferometer with 1 cm$^{-1}$ resolution. For light polarized within the ab-plane, the superconducting state reflectance is enhanced over that of the normal state below ~10 cm$^{-1}$. This energy interval coincides with the superconducting energy gap established via tunneling measurements [1]. The enhancement is significantly larger than that expected for a BCS s-wave superconductor. Along the c-axis no such enhancement is observed within the energy range investigated.

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**Inelastic X-ray scattering in correlated (Mott) insulators.**

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We calculate the inelastic light scattering from X-rays, which allows the photon to transfer both energy and momentum to the strongly correlated charge excitations. We find that the charge transfer peak and the low energy peak both broaden and disperse through the Brillouin zone similar to what is seen in experiments in materials like \( \text{Ca}_{2}\text{CuO}_{2}\text{Cl}_{2} \).
Optical properties of MgB$_2$ thin films

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(October 8, 2002)

The optical conductivity, loss function and other optical quantities of polycrystalline MgB$_2$ thin films have been determined using Kramers-Kronig analysis of the reflectance measured in the broad spectral range 30 cm$^{-1}$ - 110 000 cm$^{-1}$. This procedure was supplemented with ellipsometric measurements in the range 8 000 cm$^{-1}$ - 40 000 cm$^{-1}$. The plasma edge in the reflectivity spectrum and the peak in the loss function at 13 500 cm$^{-1}$ is in a good agreement with crossing of $n$ and $k$ obtained by ellipsometry. The position of the plasma edge and the other features, which appear in the spectra, depend on the conditions of film preparation and its history (exposition to the air). The analysis of the ellipsometric data reveals an oxide layer on the top of the films. In the far-infrared region we have observed a pronounced rise of the reflectance below 60 cm$^{-1}$, which appears below T$_c$ and can be associated with superconducting state. The real and imaginary parts of the conductivity show there also behavior, which is consistent with superconducting-gap opening.
Non-equilibrium electron dynamics in MgB$_2$
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We have performed time-resolved measurements of photoexcited carrier relaxation dynamics on recently discovered MgB$_2$ superconductor by using femtosecond real-time optical techniques. In particular, we have measured photoinduced THz conductivity dynamics and induced changes in reflectivity at 1.5 eV on timescales spanning from femtoseconds to nanoseconds, enabling direct studies of Cooper pair braking rates and subsequent recombination dynamics of photoexcited quasiparticles back to the condensate. The results show strong temperature dependence of both the pair braking rate and the recombination dynamics. Moreover, the pair breaking dynamics was found to be two-exponential and strongly photoexcitation intensity dependent, which is not observed neither on conventional nor on high-temperature superconductors. The observation of photoexcitation intensity and temperature dependence of Cooper pair breaking rates is inconsistent with the expected behavior for a conventional BCS single-gap superconductor, but suggests the presence of two distinct energy gaps on two weakly coupled Fermi surfaces, consistent with recently proposed two-gap models.

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Infrared transmittance of free-standing single-wall carbon nanotube films

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The infrared transmittance of free-standing single-wall carbon nanotube films has been studied over 30–30,000 cm\(^{-1}\) (4 meV–4 eV) at temperatures between 20 and 300 K. The optical constants were calculated by Kramers-Kronig analysis of the transmittance. Results for the conductivity and the low energy spectral weights show that there is a significant fraction of metallic tubes, but also show evidence of a pseudogap around 10–15 meV. Conductivity peaks corresponding to transitions between density-of-states peaks of semiconducting as well as metallic tubes are seen in the near-IR–visible region. Their energy locations are consistent with electronic structure calculations for 1.4 nm diameter tubes.

The far-IR data suggest a dc conductivity of around 500–1000 S/cm. The low-energy conductivity has an overall width of around 200 cm\(^{-1}\). With decreasing temperature, the conductivity increases, but only by about 30% between 300 K and 20 K.

Work supported by DARPA through grant DAAD19-00-1-0002, by the NSF through grant INT-9902050, and by OTKA grant No. 032613.
Terahertz Spectroscopy of Metamaterials

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Infrared Terahertz (THz) imaging systems have applications for explosives detection, aircraft guidance and landing in zero-visibility weather conditions, as well as terrestrial and astronomical remote sensing. These critical applications need a variety of optical elements in the THz frequency, which has yet to be explored. We demonstrate a high pass THz filter which utilizes the lowered plasma frequency of thin metal wire structures. A micro-stereolithographic technique is applied to fabricate the 2D lattice of thin metal cylinders. The reflection property of the filter is characterized by FTIR, and the plasma frequency is determined at 0.7 THz, which agrees with the approximate theory.
Evidence for Multiple Gaps in MgB$_2$ from Far-infrared Photoreflectance


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We have studied the photoexcitation and relaxation of excess quasiparticles in a film of superconducting MgB$_2$ by time-resolved far-infrared reflectance spectroscopy. A short pulse of near-IR light is used to excite the film, breaking Cooper pairs to produce excess quasiparticles. The resulting shift in the spectroscopic energy gap is monitored as a function of time using pulsed infrared synchrotron radiation. A decay time of about 1 ns is observed, consistent with a phonon bottleneck and phonon transport from the film to the underlying substrate. The observed gap shift occurs at a frequency lower than what is observed in equilibrium measurements, suggesting that at least two energy gaps exist in MgB$_2$.

Work at the NSLS supported by U.S. DOE under contract DE-AC02-98CH10886.
**Coherent THz Sources at the NSLS**

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Coherent THz pulses are emitted when electrons experience a common, rapid acceleration that persists for 1 ps or less. The most common method for producing such pulses is from laser excitation of strongly biased photoconductive switches using femtosecond pulses of light. Linear accelerators can also produce short bunches of electrons that will emit coherently in the THz range. Though the number of accelerated electrons can be similar to that for a photoconductive switch, the THz emission from the accelerator is typically larger due to relativistic enhancements of the radiated output. For example, the coherent THz pulses produced by the NSLS Source Development Lab (SDL) linear accelerator can exceed 1 microjoule energies. Such high intensity pulses present opportunities for pump-probe and non-linear spectroscopies as well as THz imaging.

Work at the NSLS supported by U.S. DOE under contract DE-AC02-98CH10886.
Energy localization in condensed matter systems using very high power radiation

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It had been known for some time that nonlinearity and discreteness can play important roles in many branches of condensed matter physics as evidenced by the appearance of domain walls, kinks and solitons. In the last ten years it has been demonstrated that some localized vibrations in perfectly periodic crystals can be stabilized by lattice discreteness[1]. This talk focuses on the experimental investigation of nonlinear nanoscale localization phenomena in condensed matter physics. One way to produce such intrinsic energy localization over a nanoscale region is by driving the modulational instability of specific large amplitude nonlinear plane wave excitations. To better understand the pathways from plane wave instability to nanoscale energy localization in crystals we describe three different classes of experiments, which operate in three different frequency regions.


This work was supported by NSF-DMR.
Time-resolved optical spectroscopy of quasiparticle dynamics

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We present an overview of time-resolved pump-probe experiments on the nonequilibrium state of superconductors. In each study the superconductors are excited by a 100 fs pulse of 1.5 eV photons. The resulting nonequilibrium population of quasiparticles is probed by a second pulse, either a second visible pulse, or a synchronous optoelectronically generated terahertz transient. By contrasting the results of the same experiment on NbN and BSCCO, we show that thermalization and recombination are qualitatively different in the high-Tc cuprates. We show that the decay of the excited state in BSCCO and YBCO (Ortho II) proceeds without limitation imposed by the phonon bottleneck. This allows direct measurement of the microscopic or 'bare' recombination rate of superconducting quasiparticles in these materials.
Ultrafast Terahertz Spectroscopy of CMR Manganites

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Ultrafast optical techniques can serve as an important tool in elucidating quasiparticle behavior in complex materials since the dynamics can be temporally resolved at the fundamental timescales of electronic and nuclear motion. We have extended these time-resolved techniques to probe ultrafast dynamics in the far-infrared (~0.2-2.5 THz) using time-domain terahertz spectroscopy with a view towards probing low energy excitations of doped transition metal oxides and other complex materials. Specifically, we measure the optically induced changes in the far-infrared conductivity with picosecond resolution. For $La_{0.3}Mn_{0.7}O_3$, $La_{0.3}Ca_{0.7}MnO_3$, and $Nd_{0.3}Sr_{0.7}MnO_3$ thin films a two-component decrease in the conductivity is observed below $T_c$ and a fast increase is observed above $T_c$. These measurements demonstrate the ability to separate and characterize spin and phonon dynamics below $T_c$ [1], as well as indicate the presence of photo-assisted hopping of small polarons near $T_c$. Commensurate with the optically induced decrease in the far-infrared conductivity, we observe an increase in the reflectivity at 1.5 eV with nearly identical dynamics. For a decrease in the far-IR conductivity there is a commensurate increase in the absorption at 1.5 eV (and vice versa), indicative of a dynamic spectral weight transfer consistent with the temperature dependence of the optical conductivity [2,3].


Probing Electronic Conduction in Insulators by THz Spectroscopy

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In this paper we describe the use of THz time-domain spectroscopy to examine the nature of charge transport in insulators. The method has been applied to model insulating systems, including non-polar liquids, such as hexane, and crystalline solids, such as sapphire. The approach permits one to follow the temporal evolution of the density of charge carriers subsequent to their generation by an ultrashort excitation pulse. Most distinctively, the method, by virtue of its ability to probe the complex frequency-dependent conductivity in the THz frequency range, allows one to determine carrier scattering rates in a direct fashion. Recent results on the rates and mechanism for carrier scattering in sapphire will be highlighted.

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**Very high Power THz radiation from Relativistic Electrons**

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We report the production of high power (20 watts average, ~1 Megawatt peak) broadband THz light from the coherent emission off sub-picosecond bunches of relativistic electrons. The work was done at the Thomas Jefferson National Accelerator Facility, Ref. 1. Electrons are produced by photoexcitation in GaAs, and are extracted and accelerated to 40 MeV. On passage through a dipole magnet, the electron bunches exhibit super-radiant synchrotron radiation emission at THz wavelengths when the bunch length is shorter than the wavelength of the emitted light. The radiation is essentially similar to the THz radiation produced by ultrafast laser techniques - spatially coherent, short duration pulses with transform-limited spectral content. The high intensity is easily understood from Larmor’s formula as being due to the relativistic enhancement. We will describe the source in detail, presenting theoretical calculations and their experimental verification. High peak and average power THz sources are also critical in driving new non-linear phenomena with excellent signal to noise, and for pump-probe studies of dynamical properties of novel materials.


This work was supported by the U.S. Dept. of Energy, the Office of Naval Research, the Air Force Research Laboratory, the Commonwealth of Virginia and the Laser Processing Consortium.
Performance of infrared beamline U12IR at the NSLS

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Beamline U12IR (National Synchrotron Light Source) utilizes infrared synchrotron radiation from a bending magnet. A combination of beamline design features and spectroscopic instrumentation allows the facility to reach the extremely low frequency limit of ~2 cm$^{-1}$ (≡ 60 GHz or 250 µeV) at rather high resolution. A 16 T magnet is also available.

The high brightness of the synchrotron emission yields substantial benefits for the study of small (mm-sized) samples. Below 20 cm$^{-1}$ the synchrotron radiation is more intense than that from a high-pressure mercury lamp.

A key feature of the beamline is a facility for sub-nanosecond time-resolved (pump-probe) infrared spectroscopy. (This capability may also be applied to the infrared-visible at U10B.) A mode-locked Ti:sapphire laser produces near-IR or (with doubling) visible pump pulses synchronized to probe pulses from the synchrotron. The broadband infrared from the synchrotron allows the entire spectral range from 2–20,000 cm$^{-1}$ (0.25 meV–2.5 eV) to be probed. A temporal resolution of 200 ps, limited by the infrared synchrotron-pulse duration, may be achieved. A maximum time delay of 170 ns is available without gating the infrared detector.

Data from the beamline from studies of semiconductors and superconductors will be presented.

This research has been supported by the U.S. Department of Energy through contracts DE-FGO2-96ER45584 and DE-FG02-02ER45984 at the University of Florida and DE-AC02-98CH10886 at the NSLS.
A dedicated storage ring for coherent THz synchrotron radiation at the ALS

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We present the concepts for a synchrotron storage ring dedicated to and optimized for the production of stable coherent synchrotron radiation (CSR) in the far-infrared (THz) wavelength range from about 100 microns to 5 mm. The 66 m circumference ring will be located above the existing ALS booster ring and will use the ALS injection system parasitically. This area provides enough room for both the ring and far-infrared scientific beamlines. CSR can be produced two ways: by shortening the electron bunches to lengths smaller than the wavelengths desired, or by longer but non-Gaussian longitudinal bunch distributions. This ring will be designed to operate in both modes. Additionally, special vacuum chambers with very large opening angles are being designed and tested to allow for efficient collection of the CSR radiation for the beamlines out to the longest possible wavelengths. The intensity of this source is predicted to be 5 to 6 orders of magnitude higher than conventional thermal or synchrotron far-IR sources. Additionally, the time structure of light will be \( \sim 200 \) fsec pulses with up to 1.5 GHz repetition rates, allowing for the use of coherent detection and/or pump-probe techniques. This new high-power THz source will enable a wide range of new science.
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