CMSN Newsletter

Newsletter of the
Computational Materials Science Network

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Division of Basic Energy Sciences
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University of Washington, Seattle

CMSN News

The major developments in CMSN in the last year revolve around the three-year review of CMSN, held in Chicago at the Four Points O'Hare Sheraton on April 5, 2003. The day-long meeting featured presentations from each of the four active CMSN Cooperative Research Teams (CRTs) as well as presentations from five prospective teams. The meeting resulted in funding for the four continuing CRTs plus the addition of a fifth team.

New CRT on Strongly Correlated Systems

We are very pleased to announce the addition of a new CRT to the CMSN community. The team, led by Warren Pickett and Richard Scalettar of the University of California at Davis, is titled *Predictive Capability for Strongly Correlated Systems*. The CRT is organized into three subteams, each with a different research focus:

1. Density Functionals: Correlated Bands and Charge/Spin Response
2. Dynamical Mean Field Theory and its Cluster Extensions
3. Emerging Downfolding Approaches to Strong Correlations

The scientific purpose of the team is summarized on the CRT's website, and is excerpted here.

> There are classes of materials that are important to DOE and to the science and technology community in general which have proven very difficult to understand and to simulate in a material-specific manner. These range from actinides, which are central to the DOE mission, to transition metal oxides, which include the most promising components of new spin electronics applications, to intermetallic compounds whose quantum critical behavior has given rise to some of the most active areas in condensed matter theory. After decades of study from a variety of often quite approximate viewpoints, a material-specific, predictive capability for many of these correlated electron systems is now a realistic goal. This exciting possibility is based on (1) new theoretical innovations, (2) coupling of experts in many-body theory with electronic structure practitioners, and (3) development of novel computational algorithms to solve the resulting equations.

We encourage you the visit the *Predictive Capability for Strongly Correlated Systems* website at http://leopard.ucdavis.edu/rts/cmsn/index.html.

Updates from Other Teams

Four of the five CMSN teams summarized in Table 1 will continue to pursue the goals they articulated in the initial CMSN cycle, which began in 2000. One of these four is changing its focus slightly. The CRT led by Anthony Rollett (Carnegie-Mellon) and Alain Karma (Northeastern), and formerly titled *Microstructural Evolution of Fundamental Interfacial Properties*, will now be known as *Fundamentals of Dirty Interfaces: From Atoms to Alloy Microstructures*. The scientific focus of the team will shift to examining the role of impurities in determining materials properties, as explained in the following excerpt from the team's proposal.

> The reality of everyday materials is that, with a few notable exceptions such as semiconductor substrates, useful materials are impure. Moreover, the impurities have often drastic effects on the properties of the materials and, in particular, on the properties of the interfaces inside the materials.

Table 1. Summary of Current Cooperative Research Teams.

CRT	Leaders
Magnetic Materials Bridging Basic and Applied Science	Bruce Harmon (Ames) and Malcolm Stocks (ORNL)
Excited State Electronic Structure and Response Functions	John Rehr (Washington) and Steven Louie (UC-Berkeley)
Fundamentals of Dirty Interfaces: From Atoms to Alloy Microstructures	Alain Karma (Northeastern) and Anthony Rollett (Carnegie-Mellon)
Microstructural Effects on the Mechanics of Materials	Dieter Wolf (ANL) and Richard Lesar (Sandia)
Predictive Capability for Strongly Correlated Systems	Richard Scalettar and Warren Pickett (UC-Davis)

The Scientific Oversight Committee

A new feature of the CMSN organization was unveiled at the review meeting: the CMSN Scientific Oversight Committee (SOC). The SOC's responsibility is primarily to provide reviews of CMSN and the CRTs to the DOE every three years. The SOC's additional duties include providing timely advice to the Coordinators and Steering Group on new research directions to pursue, new CRTs to solicit, and the like. The SOC is currently composed of seven eminent scientists drawn from diverse fields. Table 2 below summarizes the SOC's membership.

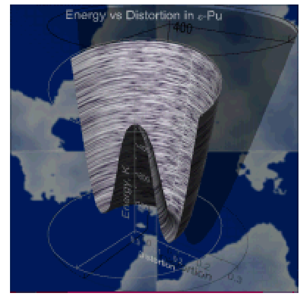
Table 2. Membership of the CMSN Scientific Oversight Committee.

Member	Affiliation
Ellen Stechel (chair)	Ford Motor Company
Samuel Trickey	University of Florida
Juan Sanchez	University of Texas-Austin
James Davenport	Brookhaven National Lab
George Samara	Sandia National Lab
Hamish Fraser	Ohio State University
Roberto Car	Princeton University

Research Highlights

In this issue of the CMSN newsletter we feature highlights from our newest CRT, Predictive Capabilities for Strongly Correlated Systems, and from the Excited States CRT.

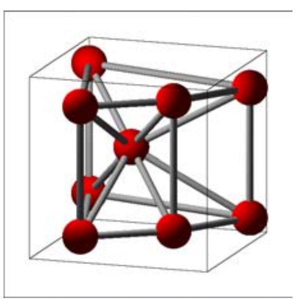
Phonons and Phases of Plutonium



Cooperative Research Team on

Predictive Capabilities for Strongly Correlated Systems

Plutonium, a key element of the energy industry, is not well studied either experimentally or theoretically due to its toxicity, radioactivity, and importance of many body electronic correlations. We develop a new approach to study its properties based on a combination of dynamical mean field and linear response theories. This allows us for the first time to look into vibrational spectra of Pu. Very recently our predictions have been confirmed experimentally.



Plutonium (Pu) is a material with very unusual solid-state properties. Despite its scientific and technological importance, many of its key properties, such as the spectrum of lattice vibrations, remain uninvestigated. It has not been possible to measure that spectrum experimentally because of Pu's extreme toxicity and radioactivity. It has not been possible to compute the spectrum theoretically, because Pu is strongly correlated, and the traditional electronic structure methods fail to describe it even qualitatively. These studies are, however, essential to be able to address the factors that govern the lattice stability of Pu, an issue that is important for Pu's storage and disposal over long time scales.

In order to treat structural, spectroscopical and lattice dynamical properties of materials such as Pu we have developed an *ab initio* approach which takes into account many-body Coulomb correlations among the electrons [1]. Our method allows us to include dynamical self-energy effects in calculating total energies, spectra, and phonon dispersions [1,2]. Its foundation is provided by the dynamical mean field theory (DMFT), which treats systems with competing localization and delocalization tendencies of the electrons, where such methods as the density functional theory in its local density approximation (LDA) or generalized gradient approximations (GGAs) have limited applicability.

Fig. 1 shows a comparison between calculated (dashed lines) phonon dispersion relations for the delta-phase of Pu (May 9, 2003 issue of ***Science,*** [2]) which only very recently were confirmed (circles) by the experiments conducted at Lawrence Livermore National Laboratory (August 22, 2003 issue of ***Science***, [3]).

We have further studied the lattice dynamical properties of the highest temperature bcc phase of Pu, which is called ε-phase. Several soft phonon modes has been predicted which point out that ε-Pu has strongly anharmonic lattice vibrations. The corresponding total energy calculation using a dynamical mean field method reveals a double well behavior (See. Fig. 2) and emphasizes the importance of the phonon entropy in understanding variety of structures in the phase diagram of Pu.

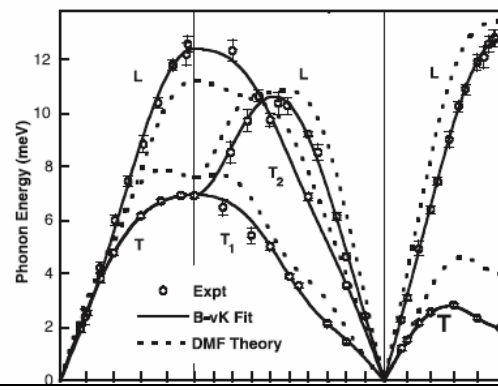


Fig. 1 Measured phonon frequencies of δ-Pu (circles connected by full lines), which have confirmed our theoretical predictions [2] (dashed lines). Data are reproduced from the experimental work [3].

Studies of other correlated materials and developments of novel robust algorithms are under way. The members of our cooperative research team are working together on correlated band structures and new density functionals (collaborations between Eguiluz, Martin, Martin, Mazin, Pickett, Scuseria, Singh, Spalding), dynamical mean field theories and its cluster extensions (collaborations between Dean, Gubernatis, Kotliar, Ku, Jarrell, McMahan, Savrasov, Scalettar, van Schilfgaarde, Schulthess, Zhang) as well as downfolding approaches to strong correlations (collaborations between Antropov, Cox, Eguiluz, Kotliar, Ku, Umrigar, White, Wilkins, Zhang).

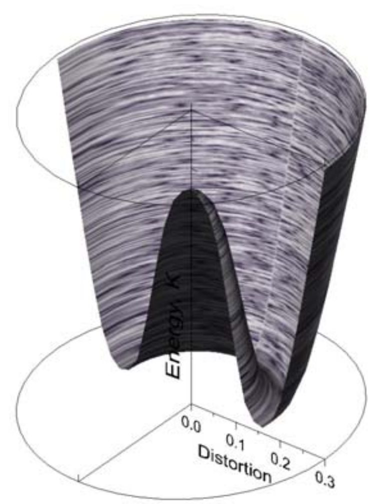


Fig. 2 Total energy vs. distortion for one of the soft phonon modes in ε-Pu which reveals a double well behavior and highlights the importance of anharmonic effects. After [2].

References

[1] S. Savrasov, G. Kotliar, and E. Abrahams, "Electronic correlations in metallic Plutonium within dynamical mean-field picture", Nature **410**, 793 (2001).
[2] X. Dai, S. Y. Savrasov, G. Kotliar, A. Migliori, H. Ledbetter, E. Abrahams "Calculated Phonon Spectra of Plutonium at High Temperatures", Science **300**, 953-955 (2003).
[3] J. Wong, M. Krisch, D. L. Farber, F. Occelli, A. J. Schwartz, T.-C. Chiang, M. Wall, C. Boro, R. Xu, "Phonon Dispersions of fcc-Plutonium-Gallium by Inelastic X-ray Scattering", Science **301**, 1078-1080 (2003).

Contact: Sergej Savrasov, New Jersey Institute of Technology

More information can be found at this CRT's website, at http://leopard.ucdavis.edu/rts/cmsn/index.html.

Core-hole Effects in X-ray Spectra

J. J. Rehr

Excited State Electronic Structure and Response Functions

The question of how to treat the core-hole effects in x-ray absorption spectra (XAS) has long been unsettled, and is a major focus of one sub-project in the Excited State Electronic Structure and Response Functions CMSN CRT. The Team members working on this subproject include J. Rehr (Univ. of Washington), E. Shirley (NIST), S. Pantelides (Vanderbilt U), Z. Levine (NIST) and shared postdoctoral research scientists A. Soininen, A. Ankudinov, and A. Francheschetti.

Within the independent-electron approximation, the core hole is often treated within the "final-state rule," (FSR) i.e., the one-electron golden rule with final-state potentials calculated in the presence of a fully relaxed core hole. This approach provides a well established and efficient method for calculating x-ray absorption spectra and also includes electron damping effects by virtue of a complex electron self-energy [1]. Simpler one-electron approximations have also been proposed, including for example, the "$Z + 1$" [2] and "transition state" approaches.

More detailed treatments of the core-hole interaction require approximations that go beyond the independent-electron approximation. For example, a two-particle Bethe-Salpeter Equation (BSE) approach has recently been applied to XAS [3]. This approach explicitly treats particle-hole excitations and includes a self-energy based on Hedin's GW approximation [4]. Approximate methods that go beyond the independent electron model include time-dependent density functional theory (TDDFT), as discussed recently by Ankudinov *et al* [5].

Recent work has aimed at elucidating the formal relationships between these various approaches for calculations of deep core XAS. It has been found that BSE for deep-core XAS and FSR are closely similar, apart from differences in their practical implementations. Their similarities help explain the numerical agreement often observed in calculations. Their formal differences rest largely in the treatments of local-field effects and exchange. These differences help explain why the effect of the core-hole varies from case to case, and also suggest how these approaches can be improved. This work has been presented at the recent International Conference on X-ray Absorption Fine Structure (Malmo, Sweden). The work is aimed at improved calculations of x-ray spectra and other optical constants from the UV to the hard x-ray limit. For example, Fig. 1 shows calculations both of the x-ray absorption coefficient and the loss function $-\mathrm{Im}(\varepsilon^{-1})$.

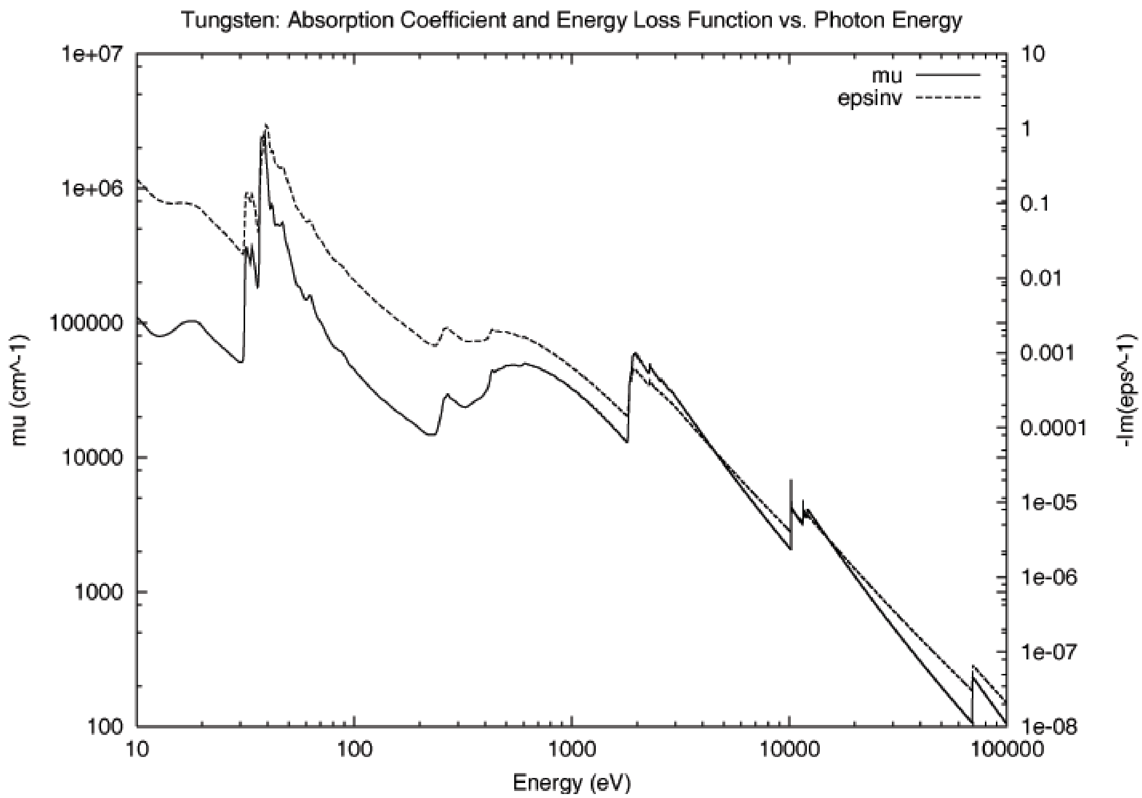


Figure 1. Tungsten optical constants: x-ray absorption coefficient (solid, left axis), and energy loss function (dashed, right axis) vs. photon energy (eV).

Further improvements based on an effective Hamiltonian with improved treatments of the core-hole are in progress [6]. They suggest that it may be preferable in calculations of deep core XAS to replace the FSR Hamiltonian with its deep-core BSE counterpart h_{eff}.

As discussed by Z. Levine [7], accurate measurements of the mass absorption coefficient are important for technological purposes, e.g., in measuring the thickness of integrated circuit interconnects. The discrepancy between theory and experiment in Fig. 2 is likely due to local field effects which can be treated within the TDDFT. A robust, quantitative theory is clearly necessary for understanding these important technological applications as well as for a general understanding of the details of x-ray absorption phenomena.

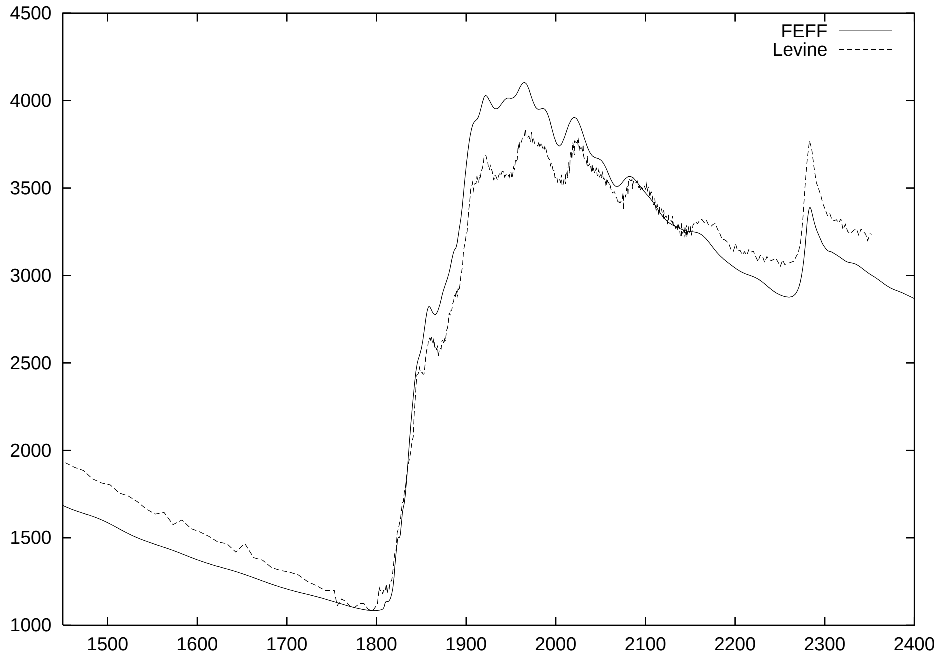


Figure 2. Tungsten x-ray absorption spectrum vs. photon energy (eV). The solid line is the FSR calculation with the UW FEFF code; the dashed line is Levine's experimental data. The discrepancy is likely due to local field effects which can be treated with TDDFT methods.

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[1] J. J. Rehr and R. C. Albers, *Rev. Mod. Phys.* **72**, 621 (2000).
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Contact: John J. Rehr, Department of Physics, University of Washington, Seattle.

More information can be found on this CRT's website, at
http://www.phys.washington.edu/~cmsn/CRTs/ESESRF/

Conferences and Workshops

Three CMSN CRTs have held or will be holding fall workshops. These include the following:

- 8th Meeting of the *Fundamentals of Dirty Interfaces* CRT, October 2-3, 2003, at the Colorado School of Mines, Golden, Colorado.
- *Predictive Capability for Strongly Correlated Systems*, First Workshop, November 7-9, 2003, at the University of Tennessee at Knoxville. The meeting program, participant lists, and talks are now available at the meeting's website: http://cst-www.nrl.navy.mil/~singh/pcscs-workshop/.
- *Excited State Electronic Structure and Response Functions*, Fourth Annual Workshop, December 19, 2003, at NIST, Gaithersburg, Maryland. The meeting website is at http://www.phys.washington.edu/~cmsn/CRTs/ESESRF/WorkshopDec03/.

Please consult the CMSN website for updated information on conferences and workshops, at http://www.phys.washington.edu/~cmsn/.

Postdoctoral Position Announcements

1. This announcement comes from Michel Van Hove at Lawrence Berkeley National Lab.

Soon available will be a postdoctoral position at Lawrence Berkeley National Laboratory for a project on optimization in diffraction and total-energy calculations in nanoscience. The purpose is to improve, in collaboration with mathematicians, methods to locate local and global minima.

In the case of diffraction calculations (e.g. low-energy electron diffraction, photoelectron diffraction, x-ray absorption fine structure), the optimization finds structural and non-structural parameters that best fit experimental data. For total-energy calculations (e.g. density-functional theory, embedded atom model), the lowest-energy structure is sought.

The position is initially for one year with the possibility of renewal.

Candidates should send a *curriculum vitae* and three recommendation letters by mail, e-mail, or fax, to:

Dr. Michel A. Van Hove
Mailstop 2-100
Lawrence Berkeley National Lab
1 Cyclotron Road
Berkeley, CA 94720
USA

Fax: 1-510-486-5530
Email: MAVanHove@lbl.gov
Web site: http://electron.lbl.gov/

2. The following announcement comes from Zachary Levine and Eric Shirley (NIST), members of the Excited States team.

NIST has begun a five year joint experimental, computational, and theoretical program to obtain three-dimensional maps of elements and chemicals in nanoscale material samples using analytical scanning transmission electron microscopy, specifically including electron energy loss spectroscopy and energy dispersive x-ray spectroscopy. Here, we seek a candidate with a strong background in electronic structure calculations and method development to calculate spectra, using and extending methods as the Bethe-Salpeter approximation and FEFF for application to electron microscopy. The work will be applied to image processing and tomography, although the candidate need not have experience in these areas.

US citizens are encouraged to apply (with a sponsor, named below) through the NIST/National Research Council Postdoctoral fellowship program. Opportunities exist outside this program for both US and foreign nationals, possibly with the participation of a host university. More senior candidates may request consideration for a 1-2 year term appointment, in lieu of a post-doctoral appointment.

The NIST/NRC application deadline is February 1, 2004, and carries a salary of $55,700, an additional allocation of $5,500 for research expenses, and other benefits. See http://www.nist.gov/oiaa/postdoc.htm for details.

Faculty Position Announcement

Colorado School of Mines
Division of Engineering
Assistant or Associate Professor
Quantum/Stochastic Control of Nano-Scale Materials Phenomena

The Division of Engineering invites applications for a new anticipated tenure-track faculty position at the level of Assistant or Associate Professor in material physics with special emphasis on the control of the micro/nano-mechanics of materials processing. Individuals with more experience may also be considered.

Because of the recent explosion of nano-scale engineering activities, there are enormous opportunities for guided or templated assembly of material systems at the micron, nano and quantum length scales, with applications ranging from material property design to MEMS/NEMS devices to quantum computing. This emerging discipline bridges materials processing and control theory and is in line with the synergistic overlap of two of the primary groups within our division—Material Mechanics and Automation/Sensing.

The Division of Engineering has a wide range of interdisciplinary research programs. Those most closely related to the new position are solid-state physics, computational mechanics, optical diagnostics, process control/optimization, electronic thin film synthesis, and combustion processing of nano-powders and films. Faculty in these areas conduct joint investigations of materials processing which integrate design, simulation, sensing, and control.

Responsibilities: The successful candidate will teach mechanical engineering classes at both the undergraduate and graduate levels and will be expected to develop a research program focused on the control of small-scale processes within solid-state materials. Further, the successful candidate will be expected to work effectively in a collaborative, interdisciplinary, environment within the Division of Engineering as well as with researchers within our Materials Science, Physics, and Chemical Engineering programs.

Qualifications: An individual is being sought who will complement and enhance the existing research expertise of the faculty by concentrating on the effects of nano/meso-scale mechanical state on the processing of advanced materials. The successful candidate must have a Ph.D. in Mechanical Engineering, Electrical Engineering, Physics, Materials Science or in a field closely related to one of these. Applicants should have: an understanding of teaching principles or a record of successful teaching; a record of research accomplishments commensurate with experience; the ability to develop external research funding; and superb interpersonal and communication skills. Preference will be given to those applicants with demonstrated expertise in both materials science and quantum/stochastic control theory.

To Apply: Interested individuals must submit a letter of application, curriculum vita and contact information for three references to: Colorado School of Mines, Office of Human Resources, Search # 03-011830, Golden, CO 80401. Review of applications will begin on February 15, 2004.
CSM is an EEO/AA employer and is committed to enhancing the diversity of its faculty and staff. Women, minorities, veterans, and individuals with disabilities are encouraged to apply.

CMSN Information

CMSN Coordinators	John J. Rehr (Washington) George F. Bertsch (Washington)
CMSN Steering Committee	Ellen Stechel (Ford) Bruce Harmon (Ames Lab) Malcolm Stocks (ORNL)
CMSN DOE Contact	Dale Koelling (DOE-BES)
CMSN Editor & Administration	Jesse Canterbury (Washington)

For inquiries regarding CMSN, the CMSN Teams, the Newsletter and Newsletter submissions, send email to *cmsn@phys.washington.edu*. More information can be found on the web at http://www.phys.washington.edu/~cmsn/.