

Section 4 Surfaces and Interfaces

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Chapter 1. Statistical Mechanics of Constrained Electronic Systems and Semiconductor Structures

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1.1 Introduction

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Our objectives are to produce microscopic theories and to effect the statistical mechanics for electronic systems that are constrained by finite sizes, quenched disorder, and/or surfaces and for semiconductor systems that exhibit a multiplicity of structures on surfaces or in bulk. In these systems, the microscopic degrees of freedom, electronic or structural, are strongly coupled and therefore their correlated behaviors determine the mesoscopic and macroscopic properties. Examples are the enhanced conductivity of an electronic system, the different step arrangements of a vicinal semiconductor surface, or the different surface or bulk structures of a semiconductor alloy. All of these are among the results obtained in our research. The correlated fluctuations of the degrees of freedom are dominated, at low temperatures by quantum effects, at higher temperatures by thermal effects, and in mesoscopic systems by quenched impurity and boundary effects. Furthermore, competing interactions between these degrees of freedom often cause frustration, namely an inability to satisfy all of the interactions, thereby injecting entropy into the system and causing a highly complex and degenerate free energy landscape.

Renormalization-group theory, introduced by K.G. Wilson for critical phenomena, is a microscopic theory that can address these effects. In our research we have developed this theory for electronic conduction and semiconductor systems. Renormalization-group theory is, when systemat-

ically pursued, an almost guaranteed procedure for solving a complex physical problem by repeatedly thinning out the degrees of freedom. Because the calculation remains a physical description at every length scale, it is ideally suited for the inclusion of the effects mentioned above. Another used method that is well suited for finite-size boundary constraints, quenched impurities, and frustration, is our recently introduced hard-spin mean-field theory.

1.2 Renormalization-Group Approach to Highly Correlated Electronic Systems

Project Staff

Alexis Falicov, Professor A. Nihat Berker

The metal-insulator transition, metallic magnetism, heavy-fermion behavior, and high- T_c superconductivity are all finite-temperature effects that result from the strong correlation of electrons in narrow energy bands. It is therefore of significant interest to study finite-temperature phenomena in strongly correlated electronic systems. Accordingly, we have performed the finite-temperature statistical mechanics of the tJ model of electronic conduction in d dimensions, obtaining the phase diagrams, electron densities, kinetic energies, and correlation functions using renormalization-group theory. While zero-temperature properties of the tJ model have been studied by a variety of methods, the finite-temperature behavior of the model had been essentially unexplored, especially in $d=3$, where we now obtain a rich structure.

We find that the renormalization-group method is very well suited for the statistical mechanics of an electronic conduction system. The work described

here constitutes an important step for the statistical mechanics of mesoscopic systems and quantum dots, as described in section 4.3 below.

Our renormalization-group calculation automatically yields the global finite-temperature phase diagram and statistical mechanics of a generalized tJ model, defined, on a lattice with one spherically symmetric orbital at each site i , by the following Hamiltonian:

$$-\beta\mathcal{H} = t \sum_{\langle ij \rangle, \sigma} (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma}) - J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + V \sum_{\langle ij \rangle} n_i n_j + \mu \sum_i n_i,$$

where, at each site i , $c_{i\sigma}^\dagger$ and $c_{i\sigma}$ are the electron creation and annihilation operators with z -component of spin $\sigma = |\uparrow\rangle, |\downarrow\rangle$, $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ and \vec{S}_i are the electron density and spin operators, and $n_i = n_{i\uparrow} + n_{i\downarrow}$. In this Hamiltonian, the consecutive terms respectively represent the electron kinetic energy, nearest-neighbor antiferromagnetic interaction (for $J > 0$), nearest-neighbor Coulomb interaction, and chemical potential. The traditional tJ Hamiltonian obtains for $V/J = 0.25$.

We find no finite-temperature phase transition in $d=1$ and a finite-temperature critical point terminating a first-order boundary in $d=2$ (figure 1a). The first-order boundary in $d=2$ represents phase

separation between electron-rich and hole-rich phases. As the hopping strength t is increased, this phase separation vanishes around $t/J \approx 0.25$, as seen in figure 1b. This result addresses a currently controversial issue in the theory of strongly coupled electronic systems.

In $d=3$, we find a remarkably complex multicritical phase diagram, with a novel phase and multiple reentrances at different temperature scales, as seen in figures 2 and 3. This phase diagram exhibits antiferromagnetic (a), electron-rich disordered (D) and hole-rich disordered (d) phases separated by first- and second-order phase boundary lines. These boundary lines are punctuated by critical points (C), critical endpoints (E), tricritical points (T), and special multicritical points (M).

The antiferromagnetic phase of the filled system is unstable to a small amount of doping by holes (by about 5 percent in figure 3). One novel aspect is the appearance, close to a narrow phase separation of hole-rich and electron-rich phases, of a new phase (which we have called " τ "), occurring at low temperatures in a narrow interval of doping. This phase, to our knowledge never seen before in finite-temperature phase transition theories, is the only volume of the extended phase diagram in which, after repeated length rescalings of the renormalization-group, the electron hopping strength t does not renormalize to zero. In fact, all the interaction constants (t, J, V, μ) renormalize to infinite strengths, while their ratios eventually remain constant, a typical behavior for the renormalization-group sink of a low-temperature phase. A

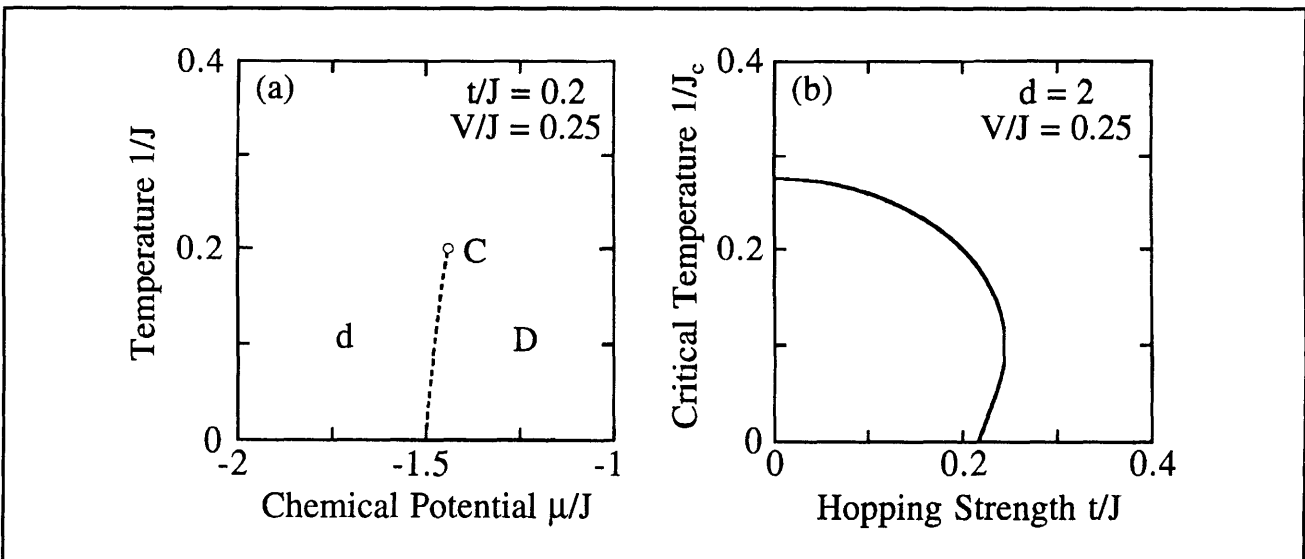


Figure 1. (a) Typical cross-section of the calculated global finite-temperature phase diagram for the two-dimensional tJ model, with $t/J=0.2$ and $V/J=0.25$. Between the electron-rich (D) and hole-rich (d) disordered phases, a phase-separation boundary terminates at a critical point (C). (b) Calculated critical temperatures $1/J_c$ as a function of relative hopping strength t/J , in two dimensions. It is thus seen that finite-temperature phase separation occurs only for low values of t/J .

distinctive feature is that at this sink, which as usual epitomizes the entire thermodynamic phase that it attracts, the electron density $\langle n_i \rangle$ has the non-unit, non-zero value of $2/3$. This feature makes strong-coupling conduction possible by having the system

non-full and non-empty of electrons, and has also not been seen previously. The occurrence and characteristics of our newly discovered phase are quite suggestive.

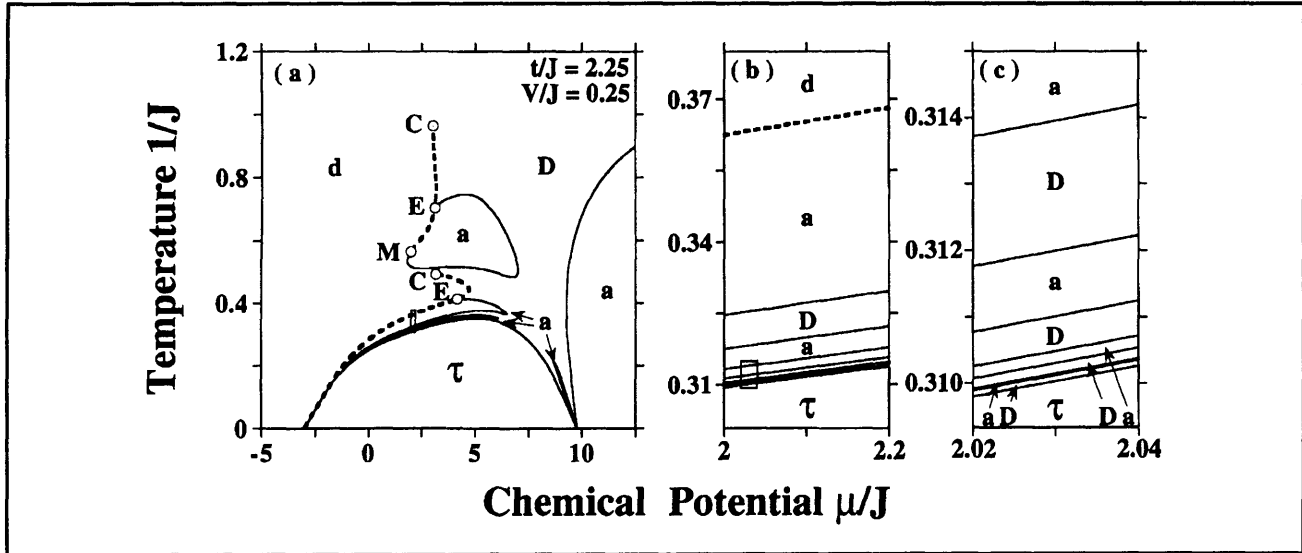


Figure 2. Typical cross-section of the calculated global finite-temperature phase diagram for the three-dimensional tJ model, in the temperature and electron chemical potential variables. First- and second-order boundaries are shown respectively with dashed and full curves. Figures 2b and 2c are respectively the blow-ups of the small rectangles in figures 2a and 2b. The corresponding temperature versus electron density phase diagram is in figure 3.

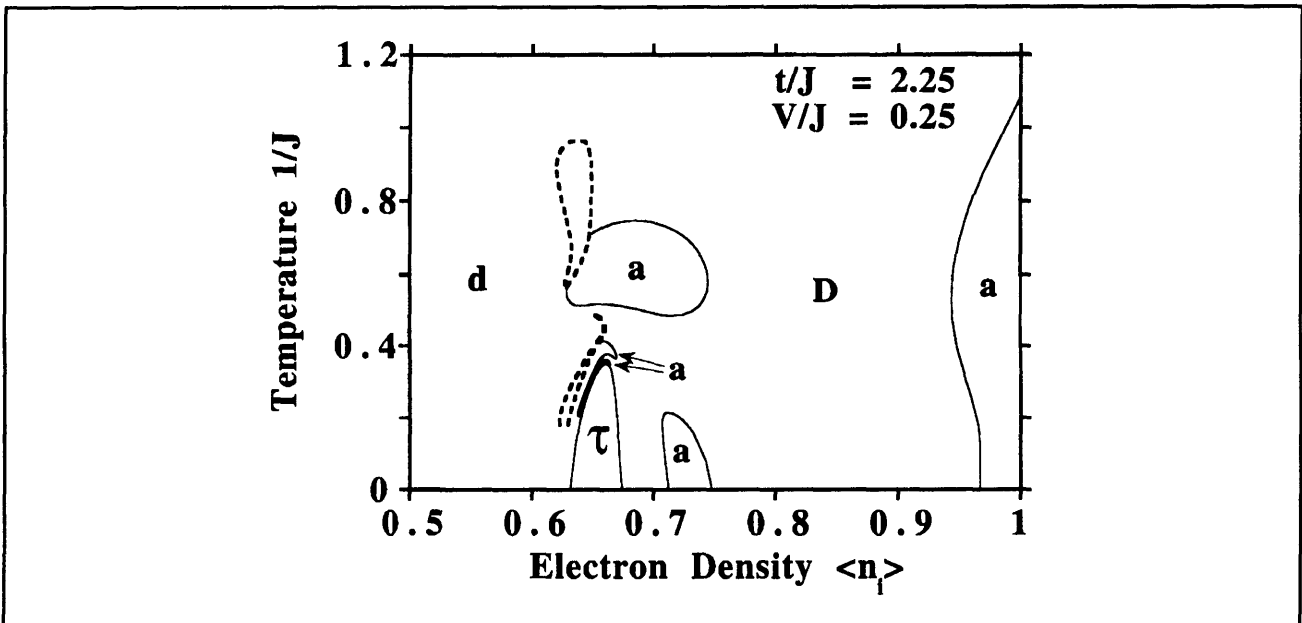


Figure 3. Typical cross-section of the calculated global finite-temperature phase diagram for the three-dimensional tJ model, in the temperature and electron density variables. The phase separation boundaries of the first-order phase transitions are drawn with dashed curves; the unmarked regions inside these boundaries are the coexistence regions of the phases marked at each side of the regions. The second-order phase boundaries are drawn with full curves. The temperature versus electron chemical potential phase diagram corresponding to this figure is in figure 2a.

Another feature is the appearance of several islands of the antiferromagnetic phase. The islands are bounded by first- and second-order phase transitions adorned by the various special points already mentioned above. Thus, a multiply reentrant phase diagram topology obtains. The antiferromagnetic phase also occurs as a narrow sliver, within the disordered phase reaching zero temperature between the antiferromagnetic and "τ" phases. The appearance of the antiferromagnetic islands at dopings in the neighborhood of the "τ" phase indicates that, when the electron hopping strength t strengthens under rescaling, antiferromagnetically long-range correlated states acquire substantial off-diagonal elements, which lowers the free energy of the antiferromagnetic phase. On the less dense side of the boundary of the "τ" phase, a lamellar sequence of antiferromagnetic slivers and disordered inlets occurs, at several temperature scales, as seen in the blow-ups of figures 2b and 2c.

We have calculated electron densities, kinetic energies

$$\langle T_{ij} \rangle =$$

$$\langle c_{i\uparrow}^\dagger c_{j\uparrow} + c_{j\uparrow}^\dagger c_{i\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow} + c_{j\downarrow}^\dagger c_{i\downarrow} \rangle ,$$

and nearest-neighbor density-density and spin-spin correlation functions by summing along renormalization-group trajectories and matching to the densities of phase sinks. The kinetic energies reflect the conductivity of the system as seen from the sum rule

$$\text{Tr} \int_0^\infty dw \text{Re}[\sigma_{\alpha\alpha}(w)] = -\frac{\pi e^2 d}{2\hbar a} \langle T_{ij} \rangle ,$$

where $\sigma_{\alpha\alpha}(w)$ is the frequency-dependent conductivity, e is the electronic charge, and a is the lattice spacing. Thus, the conductivity, as expected, goes to zero in the dilute and dense limits (figure 4).

1.3 Statistical Mechanics of Mesoscopic Systems and Quantum Dots

Project Staff

Alkan Kabakçioğlu, Alexis Falicov, Professor A. Nihat Berker

Our electronic-system renormalization-group calculations are now being merged with (1) finite-system

calculations, for a variety of boundary conditions, using methods that we have perfected in previous work on surface systems, and (2) quenched impurities, including pinning potentials, in the interior or at the boundary, using methods that we have developed in the work of section 4.4 below. Thus, the statistical mechanics of mesoscopic systems and quantum dots will be achieved.

Our calculations yield the entire statistical mechanics of the systems that are studied, including for the first time the kinetic energies (figure 4). The kinetic energy is related by a sum rule to the ac conductivities, as mentioned above. However, a direct calculation of ac and dc conductivities by renormalization-group theory would be a highly desirable and needed contribution. We intend to pursue this by combining the renormalization-group rescaling behaviors of the hopping strength and the local correlations. Using this approach, it should be possible to calculate the conductivities across a specific realization of a mesoscopic system for any arbitrary distribution of contact points at the boundary. This is made possible by the real-space formulation of the renormalization-group theory, which stays closely coupled to the physical description of the system at each step of the calculation.

The effect of a magnetic field will also be added to the Hamiltonian. Thus, for different physical realizations, the kinetic energies and conductivities will be calculated as a function of magnetic field strength and temperature, yielding an accessible first-principles study of these "magnetofingerprints" of mesoscopic systems.

1.4 Collective Phenomena in Systems with Quenched Impurities and Frustration

Project Staff

Alexis Falicov, Daniel P. Aalberts, Professor A. Nihat Berker

Our studies of systems with quenched randomness and frustration have yielded (1) new phenomena that are very generally applicable and (2) highly developed theoretical tools to deal with such effects in electronic and semiconductor systems. These theoretical tools consist of the renormalization-group theory, which we have perfected for the study of systems with quenched randomness, and the hard-spin mean-field theory, which for the first time conserves frustration and which in fact was introduced in these studies. Frustration, which signifies the lack of a unique local minimum-energy

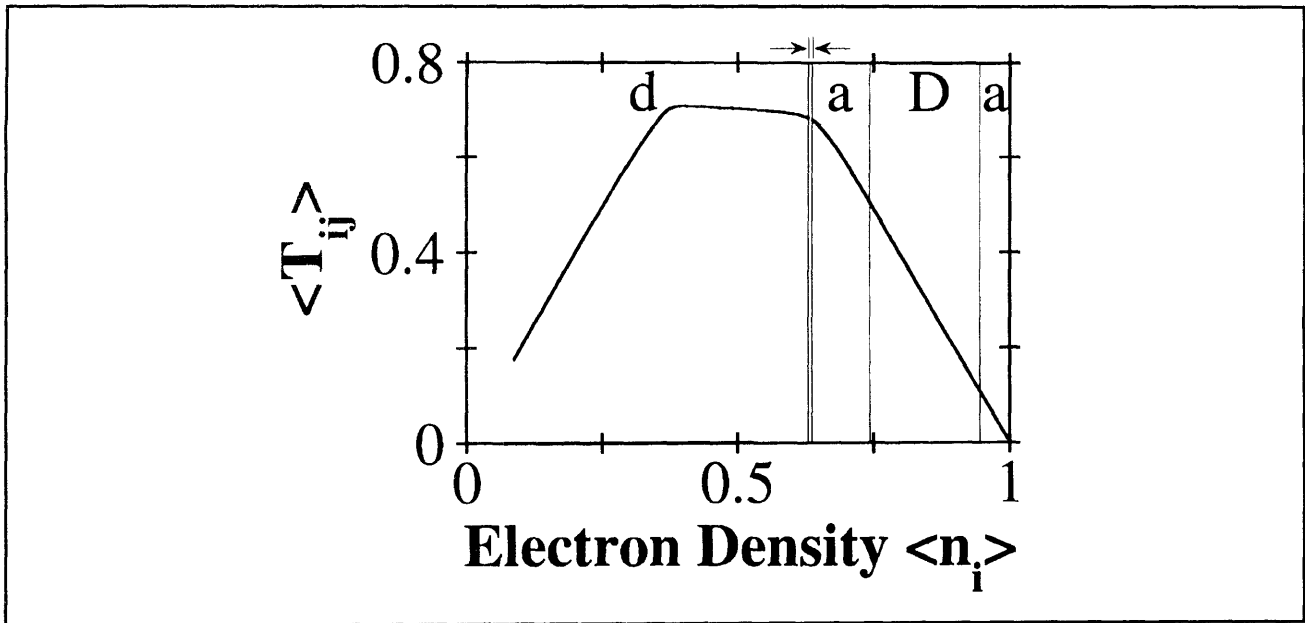


Figure 4. Electron kinetic energies in the tJ model, calculated by renormalization-group theory, at temperature $1/J=2$ across the phase diagram of figure 3. These kinetic energies are related to the conductivities by the sum rule given above.

state due to competing interactions, appears to underly phenomena in systems ranging from spin-glasses (where, as we have discovered, a chaotic rescaling behavior results) to liquid crystals and to high T_c superconductivity.

One of our general results is that, under quenched random interactions, all first-order phase transitions that involve a symmetry breaking are converted into second-order phase transitions. This amounts to the generation of infinite correlation lengths by the effect of impurities! This prediction, which, for example, raises the possibility of replacing the uncontrolled hysteresis loops of probing devices by infinite-response functions, was originally indicated by our general domain wall arguments. Most recently, our detailed renormalization-group calculations have confirmed the prediction, for example with the phase diagram of a spin-1 magnetic model (the Blume-Emery-Griffiths model), used in the study of electronic systems and of semiconductor alloys. The pure (non-random) system has first-order phase transitions at low temperatures and low densities, and second-order phase transitions at high temperatures and high densities. Our calculations show that, with the introduction of quenched randomness into the interactions, first-order transitions are converted to second-order transitions, which then reach zero temperature and low densities.

1.5 Surface and Bulk Structures of Semiconductor Alloys

Project Staff

William C. Hoston, Asad A. Naqvi, Alkan Kabakçioğlu, Professor A. Nihat Berker, in collaboration with Professor John D. Joannopoulos

Ternary and quaternary semiconductor alloys can exist in zincblende, chalcopyrite, or possibly stannite structures, involving two interpenetrating fcc lattices on which up to four atomic species exist. One atomic species occupies one of the fcc lattices while three other atomic species may (chalcopyrite, stannite) or may not (zincblende) order on the other fcc lattice. The structural phase diagram of these alloys as a function of atomic interactions, composition, and temperature is of interest. In recent work, K.E. Newman and collaborators have adapted the spin-1 Ising (Blume-Emery-Griffiths) model to the study of zincblende to chalcopyrite or stannite transitions. In this model, the three spin values are each associated with a different species of atom, A, B, or C, which exist on one of the fcc lattices. The other fcc lattice is occupied by atomic species D. The systems under consideration have the composition $[(AB)_{1-x}C_{2x}]D_2$. Precise values of the model interaction strengths applicable to the semiconductor alloy systems will be provided, in our work, from the total energy calculations of Professor J.D. Joannopoulos. The regimes of the model that are applicable to the alloy systems include negative bilinear and negative biquadratic interactions, and

therefore frustration, both features bringing interesting complications. Newman et al. obtained unusual phase diagrams using a cluster-variational theory, which does not include correlated fluctuations and frustration at the level of renormalization-group theory and hard-spin mean-field theory.

In our previous works, we have studied the Blume-Emery-Griffiths model extensively. Most recently we have studied the negative biquadratic interaction regimes, finding six new phase diagrams, with a novel multicritical topology and two new ordered phases. Thus, we have determined that the phase diagram of this simple spin system includes nine distinct topologies and three ordered phases. These results were obtained by a global mean-field theory with four independent order parameters. Our renormalization-group theory confirmed the occurrence of critical points inside an ordered phase, a question that had been raised by measurements on metamagnetic alloys.

However, since the fcc lattice is frustrated, analysis beyond standard mean-field theory is required. Our new method, "hard-spin mean-field theory", incorporates the hard-spin condition of local degrees of freedom and thereby conserves frustration. We have tested this method on frustrated triangular and stacked triangular lattices with the spin-1/2 Ising model, obtaining excellent results. We intend to apply the method to the Blume-Emery-Griffiths model on the fcc lattice, which is more frustrated. We also intend to use renormalization-group theory, which a preliminary study we have done indicates is feasible on this complicated physical system.

Furthermore, these semiconductor alloys are expected to have a variety of surface reconstruction phases on different crystal facets and a variety of step structures on vicinal surfaces. Our calculations will be extended to these effects, also using the meandering step-Hamiltonian method introduced in our previous study of vicinal silicon (100) surfaces. These calculations for bulk and surfaces of semiconductors integrate the electronic energy determinations of Professor J.D. Joannopoulos with statistical mechanics, and thereby are uniquely first-principles calculations for physical systems.

1.6 Publications

Berker, A.N. "Critical Behavior Induced by Quenched Disorder." *Physica A* 194(1): 72-76 (1993).

Berker, A.N., and K. Hui. "Phase Diagram of the Ising Model on the Square Lattice with Crossed

Diagonal Bonds." *Phys. Rev. B* 48(17): 12393-12398 (1993).

Falicov, A., and A.N. Berker. "Finite-Temperature Phase Diagram of the tJ Model: Renormalization-Group Theory." Submitted to *Phys. Rev. B*.

Netz, R.R., and A.N. Berker. "Renormalization-Group Theory of an Internal Critical Endpoint Structure: The Blume-Emery-Griffiths Model with Biquadratic Repulsion." *Phys. Rev. B* 47(22): 15019-15022 (1993).

1.6.1 Meeting Papers

Berker, A.N. "Hard-Spin Mean-Field Theory." Invited paper presented at the Topics in Statistical Physics Meeting, Antigonish, Nova Scotia, Canada, October 1-3, 1993.

Berker, A.N. "Hard-Spin Mean-Field Theory." Invited paper presented at the 70th Statistical Mechanics Meeting, New Brunswick, New Jersey, December 15-17, 1993.

Berker, A.N., and K. Hui. "Phase Diagram of the Ising Model on the Square Lattice with Crossed Diagonal Bonds." Paper presented at the American Physical Society Meeting, Seattle, Washington, March 22-26, 1993.

Berker, A.N., and R.R. Netz. "Hard-Spin Mean-Field Theory and Frustrated Systems in $d=2$ and $d=3$." Paper presented at the Gordon Research Conference on Aspects of Disorder in Condensed Matter Physics, Wolfeboro, New Hampshire, June 28-July 2, 1993.

Falicov, A., and A.N. Berker. "Finite-Temperature Phase Diagram of the tJ Model: Renormalization-Group Theory." Paper presented at the 70th Statistical Mechanics Meeting, New Brunswick, New Jersey, December 15-17, 1993.

Falicov, A., and A.N. Berker. "Finite-Temperature Phase Diagram of the tJ Model: Renormalization-Group Theory." Paper presented at the American Physical Society Meeting, Pittsburgh, Pennsylvania, March 21-25, 1994.

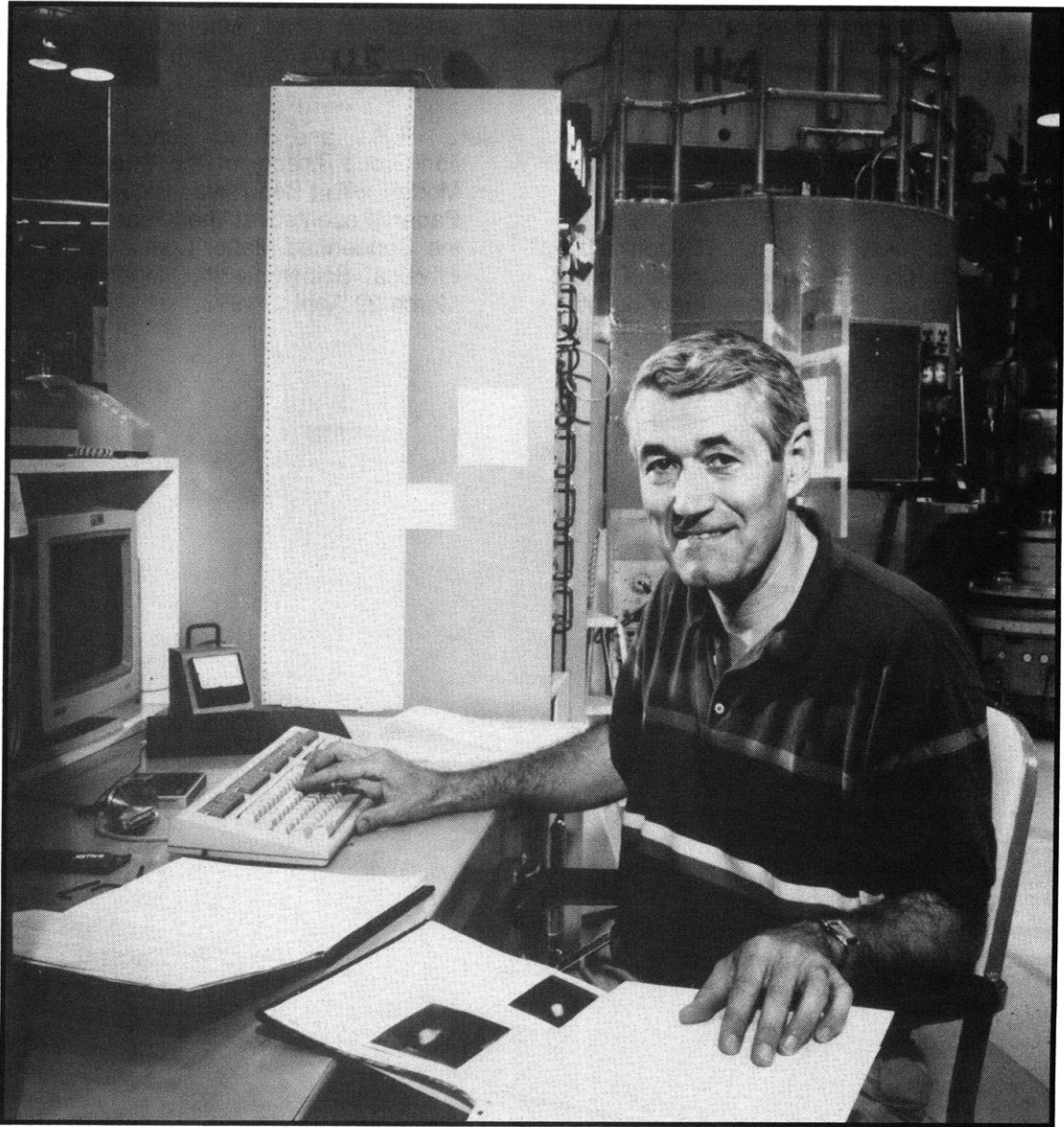
Falicov, A., and A.N. Berker. "Finite-Temperature Phase Diagram of the tJ Model: Renormalization-Group Theory." Invited paper to be presented at the Istanbul Technical University Statistical Physics Days, Istanbul, Turkey, July 14-15, 1994.

Hui, K., and A.N. Berker. "Analytic Expression for the Exact Boundary of the Superantiferromagnetic Phase of the Ising Model with Nearest- and Next-Nearest-Neighbor Interactions on the Square Lattice." Paper presented at the American Physical Society Meeting, Pittsburgh, Pennsylvania, March 21- 25, 1994.

Netz, R.R., and A.N. Berker. "Renormalization-Group Theory of an Internal Critical Endpoint Structure: The Blume-Emery-Griffiths Model with Biquadratic Repulsion." Paper pre-

sented at the American Physical Society Meeting, Seattle, Washington, March 22-26, 1993.

Netz, R.R., and A.N. Berker. "Renormalization-Group Theory of the Blume-Emery-Griffiths Model with Repulsive Biquadratic Coupling." Paper presented at the General Conference of the Condensed Matter Division of the European Physical Society, 13th, Regensburg, Germany, March 29-April 2, 1993.



Professor Robert J. Birgeneau, Dean of the School of Science. (Photo courtesy of the Photography Division of the Brookhaven National Laboratory)