VII. X-RAY SCATTERING SPECTROSCOPY

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Joint Services Electronics Program (Contract DAAB07-76-C-1400)

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During the past year we have constructed and implemented a versatile high-resolution x-ray spectrometer system. This x-ray spectrometer is based on a Rigaku 12 kW rotating anode x-ray source, a Huber high-precision 2-circle goniometer and an on-line PDP 11/34 computer control system with Camac interfacing. The spectrometer angles may be scanned through 360° with a step size of 1.8 seconds of arc and an absolute accuracy of about 10 seconds. We are constructing a second spectrometer of somewhat lower resolution for surface layer studies and for experiments on structural phase transitions. As ancillary equipment we have constructed temperature control equipment which enables us to study samples at temperatures from 10 K to 500 K with a relative accuracy of 1 to 3 \times 10^{-3} K. Our initial experiments have been directed primarily toward phase transitions in liquid crystals and in lower dimensional solids. The liquid crystal experiments show special promise and, indeed, they demonstrate that high-resolution x-ray techniques will have a major impact on that area of research. We have also initiated a project on the structure and scattering phenomena in physiadsorbed layers on surfaces. We now discuss our individual projects.

1. NEMATIC-SMECTIC-A TRANSITION IN CBOOA

A number of experiments, especially in cyanobenzylideneoctyloxyaniline (CBOOA), suggested the N-A transition was characterized by two, rather than one, divergent correlation lengths. This would seem to conflict with our current ideas on scaling near second-order transitions and would greatly complicate any attempt at a theoretical description of the transition. The salient result of our experiments is that, by direct measurement with x rays, the correlation lengths along and transverse to the molecules diverge with identical exponents; thus the transition is characterized by a single length. This is perhaps the simplest example of melting in nature and we hope that these studies, besides elucidating the nature of phase changes in liquid crystals, will also give insight into the general phenomena of melting.
2. NATURE OF THE SMECTIC-A PHASE

Whether, in nature, bodies can exist for which the density functions depend not on three but only on one or two coordinates is a classic problem which has occupied theorists since the 1930's. A closely related problem is the establishment of a one-dimensional density wave in a three-dimensional system, as occurs in a smectic-A liquid crystal. It is believed that in two-dimensional systems and also in the smectic-A phase there should be a second-order transition to a state characterized by long-range correlations but without long-range order in the usual sense. Our experiments provide the first direct experimental evidence that the theoretical conjectures are correct. We found that the instantaneous density correlations for a one-dimensional density wave in a three-dimensional liquid exhibit not long-range order but rather an anisotropic power law dependence on molecular separation. Thus smectic liquid crystals are materials in which these ideas may fruitfully be studied experimentally. Further experiments and much more theoretical efforts are called for. This work is described more fully elsewhere.2

3. SPIN-PEIERLS TRANSITION IN TTF-BDT (Cu)

The organic charge transfer salt TTF-BDT (Cu) is believed to provide an ideal example of a spin-dimerization transition in an $S = 1/2$ one-dimensional antiferromagnet. This system is of interest both because of its role as an archetype for magnetism in organic materials and also because of the analogy with the more familiar electronic Peierls transition as observed, for example, in TTF-TCNQ. In a collaborative experiment with D. E. Moncton (performed at Bell Laboratories) we have succeeded in observing this structural transition at 12 K using x-ray diffuse scattering techniques. We have monitored both the magnitude and spatial variation of the critical fluctuations above $T_c = 12$ K together with the growth of the order parameter below $T_c$. The measured order parameter agrees well with that calculated for a BCS superconductor. Further, the experiments demonstrate clearly that the transition involves a fundamentally different pairing than that inferred by others from simple molecular stacking considerations. This in turn necessitates that the dominant exchange paths differ from those which are suggested by a naive consideration of the TTF molecular orbitals. This work has been published.3

4. STRUCTURAL PHASE TRANSITION IN SQUARIC ACID

The compound $\text{H}_2\text{C}_4\text{O}_4$ is a hydrogen-bonded system with a planar structure. Neutron scattering experiments have indicated that it exhibits an unusual quasi two-dimensional structural phase transition. We have performed initial x-ray measurements which show
resolvable two-dimensional critical scattering with a variety of unusual features. Further x-ray experiments are under way to elucidate the nature of this most interesting hydrogen-bonded system.

5. ELECTRONIC EXCITATIONS IN LIQUID HELIUM

We have now completed our inelastic x-ray scattering study of electronic excitations in liquid helium (collaborators: P. E. Eisenberger and W. Marra of Bell Laboratories). We find that the electron density response function $S(q, \omega)$ can be understood quantitatively by using a simple hydrogenic atomic theory. No many-body or multielectron excitation effects are observed above a significance level of approximately 10%. This is in sharp contrast to simple metals like aluminum where many-body effects play a central role and where theory is still able to provide at best a qualitative understanding. Thus this study in liquid helium provides one physical system where theory and experiment for the electron density fluctuations are in accord. This work has been published. 4

6. MEAN FIELD THEORY AND THE GINZBURG CRITERION

As part of our studies of phase transitions we have also carried out the following theoretical analysis. By applying a real space version of the Ginzburg criterion it is possible to assess the relative importance of critical fluctuations and hence the probable limitations of mean field theory for a wide variety of phase transitions. For example, for ferroelectrics and for certain soft acoustic phonon structural transitions the Ginzburg arguments predict that mean field theory should be valid for spatial dimensionalities greater than $d^* = 3$ and 2, respectively. These "marginal dimensionalities" follow directly from the geometry of the critical fluctuations in phase space. We show also that this approach provides a simple technique for assessing the self-consistency of Landau theory in a vast range of physical systems.

Publications
