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Electron and Field Interactions with Thin Films and Surfaces

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The research under this grant has focussed on a considerable number of different aspects of the interactions of surfaces with electrons or x-rays. In the summary below, we will briefly describe each problem under investigation, the results obtained, and, where applicable, the work in progress that is still continuing to complete the tasks begun under this project. References in each paragraph are to publications and reports listed at the back.

1) Size Effects in Metals

This long standing problem deals with the details of the collisions of electrons with boundary surfaces, and how these special collisions affect the electrical transport properties, especially in samples thin enough so that an appreciable fraction of all electron scattering involves the surfaces of the sample. (Size effect). We have completed the following specific problems:

a. A new theory of surface scattering has been developed which applies to geometrically rough surfaces, and which predicts a detailed angular dependence of the surface scattering cross section.(5)

b. An experimental investigation of single crystals of Zinc at helium temperatures was carried out to measure the conductivity and galvanomagnetic coefficients in fields up to 14 kG. The data have shown that the two types of carriers in zinc have sufficiently different interactions with the surface so that the compensation known to exist in bulk is destroyed. This has opened the possibility that surface scattering may be used as a filter to suppress or enhance the effectiveness of specific groups of carriers.(7)

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c. Extensive experimental work on thin epitaxially grown single crystal films of silver has, first of all, resulted in procedures for producing thin films of the highest quality, and has furthermore been used to show that existing interpretations of transport data in thin films have not been consistent. Careful analysis requires, for instance, that a contribution to the surface scattering cross section is definitely temperature - dependent. (2,25)

d. The theory and experimental technique have been worked out to determine the angular dependence of the surface specularly parameter by analyzing the harmonic content of the magnetic field dependence of magnetomorphic oscillations. The results show that such angular dependence can indeed be identified in the experimental data for Cd, and that under proper conditions, this technique can give appreciably detailed information on this point. (13)

e. A search for a size effect in the strain dependence of the resistivity of thin films was unsuccessful. The results obtained are the first systematic study of this dependence over a large range of temperature. They show that, within fluctuations from sample to sample, strain dependent scattering in thin epitaxial films is comparable to that in bulk material, and that the surface does not contribute any sizeable anomaly. (14,20)

2.) Metallic Field Effect

An experimental and theoretical program has been developed to study the electric field effect in metals, as a possible new tool for investigating in detail the interaction of electrons and adsorbed gases with metallic surfaces. Experimentally, this has been a hard task because the small size and sensitivity of the effect has required the development of rigorous and careful techniques in an area in which there had not been any previous systematic work. The following problems have been studied:

a. The field effect of epitaxial silver on mica has been studied extensively as a function of temperature, and of film thickness, in order to identify size effects by two different techniques of altering the ratio of thickness to mean free path. Elaborate procedures were developed to perform the entire sequence of experiments without breaking vacuum. The data show a definite and systematic change in the field effect with thickness and temperature, and, for the first time, are sufficiently comprehensive to allow an analysis in terms of more than the simplest theory. They indicate that electric field modulated surface scattering is one of the primary sources of the field effect, and furthermore, that non current carrying surface states may play a significant role at the silver-mica interface. (25)

b. Field effect studies in gold have shown that the gold-mica interface has very different properties, which are probably associated with the lack of a good gold-mica bond. As a result we have carried out various studies on the growth of single crystal gold on mica. (19)

c. A theory of the metallic field effect has been developed in terms of three mechanisms involving surface charging, surface scattering,

and potential shifts. This theory includes explicitly the influence of size effects, and considers also the possibility of angle-of-incidence dependent surface scattering.(4)

d. The field effect has been measured in magnetic materials in order to assess the role of spin-dependent surface scattering. The experiments show that in nickel and permalloy the metallic field effect has a different magnitude from that of the noble metals, and is due to a different mechanism. Furthermore, it shows a very pronounced magnetization dependence. Experimental work and analysis of this latter effect are still in progress.(18,30)

3) Spin Wave Boundary Conditions

The question of spin wave boundary conditions in thin ferromagnetic films has been one which has occupied many researchers, since it has to be answered in order that unequivocal interpretation of experiments probing the details of exchange interactions in ferromagnetic metals is possible. We have designed and carried out a combination of measurements involving resonance absorption in homogeneous and inhomogeneous microwave fields, as well as detection of dc effects in magnetic resonance, to show that a determination of spin wave boundary conditions of the longest spin wave modes is indeed possible. The agreement with theory is excellent. This work has also identified a possible spin wave surface mode, which can be analyzed fully within the same theoretical framework.(21)

4) Scattering of x-rays from surfaces and thin films

X-rays, representing electromagnetic waves of wavelengths comparable to those of metallic electrons, exhibit analogous interactions with surfaces. They were studied to obtain additional insight into the coupling of waves to surfaces and thin films, made possible because the interaction strength is much smaller.

a. A detailed study of the reflection of x-rays from thin metallic films above the critical angle has shown that the fringe pattern observed at a fixed angle of reflection can be associated kinematically with non-specular coherent scattering of the beams on the two film surfaces, with the same phase shift as in the specular reflection. The method gives a simultaneous determination of film thickness, and of the interface phase shifts, and the intensity distribution is a measure of the "surface roughness".(3)

b. A similar study at optical wave lengths on barium stearate confirmed the basic interpretation, but gave additional information about the double layer spacing of these multilayer films, since each interface acted as a new source of non-specular scattering.(1,15)

c. A specific theory has been developed for the origin of the observed "surface roughness" in the x-ray scattering. The model assumes that, because of crystallite sizes and orientations, the dielectric constant is a fluctuating function of position throughout the film thickness.

It can explain successfully the intensity distributions observed in the fringes, cut-offs, and other "anomalous" results reported in the literature. As a new tool for studying the perfection of thin structures, it can be expected to have wide application.(10,22)

d. As a byproduct of x-ray studies, we have developed a method for obtaining strong reflections from thin films for quick determination of crystallinity. The method uses the very high intensity of one of the characteristic lines in the spectrum (Copper K-alpha coincides with the (111) spacing of copper, silver, and gold) to overcome the generally insufficient intensity for such studies.

5) Anomalous transmission of x-rays in perfect crystals

Since we have been able to prepare thin films of high perfection it became necessary to consider the anomalous dynamic effects which waves encounter in such structures (Borrmann effect), and since furthermore, at the normal electron wavelengths many beams are excited simultaneously, the work had to be extended considerably beyond what was known, both in experiment and theory.

A systematic study was carried out on the simultaneous transmission of three anomalous beams in silicon, with special emphasis on the polarization, intensity and absorption of the various interlocking wave components. Several new features of the phenomenon were discovered - which could be confirmed by theory - and they have, among other things, led to a more general characterization of the Borrmann effect, as it may manifest itself in a variety of applications of x-ray diffraction. While this work as such has been completed, it has had a large stimulus on further investigations in this area which are being pursued now.(11,23)

6) Theories of Low Energy Electron Diffraction (LEED)

While much of the work reported above was concerned with the interaction of internal electrons with surfaces, a nearly equally important phase attacked the theory of external electron scattering (LEED) as a required tool for understanding the information about surfaces obtainable by that method. Our studies have revealed that, at least on crystals with clean surfaces, low energy electrons interact strongly with both the surface and the bulk below it, and that the interpretation of LEED intensities cannot proceed without a clear understanding of both interactions. We have developed two specific theories of LEED, one emphasizing the most direct way of obtaining diffraction intensities, the other stressing the physical understanding of the strong coupling accompanying electron scattering.

a. A theory has been developed to determine LEED intensities by the use of a new variational principle relating to reflection coefficients. The theory obviates the need for an exact knowledge of the electron wave functions inside the crystal (band structure), and separates structural and dynamic effects in a clear cut fashion. It is particularly applicable to crystals with differing surface layers.(12,27)

b. A theory has been worked out based on an explicit analytic formulation of multiple scattering in a periodic medium. The formal structure of the theory leads automatically to the prediction of anomalous beams and surface resonances, and it allows to study the interaction to any degree of multiple scattering, indicating where these effects predominate, and where a simpler formulation suffices.(26)

c. In support of the other phases of this program, an investigation was carried out on the effect of surfaces on the propagation constants of Bloch waves in crystals. In particular, emphasis was given to the appearance of complex propagation constants, i.e. evanescent waves, in the presence of a surface, because these waves play an important role in a quantitative determination of the reflection and diffraction intensity of external waves.(6)

7) Tunneling Phenomena

In the area of tunneling the broad aim of our research was to perform tunneling experiments in such geometries and otherwise well specified situations so that one could truthfully compare the results to the idealized theories which have been proposed. Despite considerable effort, however, we have not had very good success. It has turned out the clean, and nearly perfect crystal surfaces which we have prepared either on bulk crystals or on thin films are chemically so inert that they do not form good tunneling junctions, while tunneling between polycrystalline or contaminated surfaces does not present any problems. This has been particularly true for superconducting materials.(16,29)

Consequently, we have developed a program to study tunneling between single crystals through vacuum. This experiment is still in progress.(28)

8) Field Emission from Organic Molecules

In order to understand the tunneling of electrons through adsorbed molecules in the high electric fields of the field emission microscope, we have carried out a theoretical study of a number of aspects which enter into this problem, such as the molecular distortion in the intense electric field, the orientation and shift in vibrational frequencies of a molecule under these conditions, and the adsorption equilibrium in the presence of an electric field.

As part of the development of the mathematical techniques needed, an intensive effort was spent on calculating the shift in energy and other properties of the hydrogen atom and the hydrogen molecule in very intense electric fields, and we now have, at least in the former case, a very reliable answer to a problem which had been solved only partially a number of times in the past. (Incidentally, this development led, indirectly, to a re-examination and generalization of the entire problem of the WKB approximation) In the case of H_2 , we have obtained dissociation energies which explain the observations, and have predicted shifts in the vibrational spectrum which should become observable in the interaction of H_2 with intense infrared light.(9,17,24)

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