

Better Comparison of Theory with Experiment in Ultraviolet Interactions

Recent years have seen significant advances in the physics of solids in the vacuum-ultraviolet region, particularly within those areas in which theorists and experimentalists have collaborated. Calculations of the electronic band structure of solids, both complex and elemental, are now of such sophistication that the derived optical properties are detailed enough to allow, in many cases, a point-by-point comparison with experiment. Until quite recently intercomparison of data, even among experimentalists, was difficult; however, refined techniques for the study of reflection spectra and photoemission properties, and the advent of easily attainable ultrahigh vacuums that minimize sample contamination, largely eliminate earlier differences. We now have a clearer understanding of the role of excitons (metastable states of a bound electron-hole pair within the lattice structure), plasmons (states in which the electronic excitations are correlated) and interband transitions, in determining the fundamental absorption spectra of many solids. Plasmons, of both the volume and surface varieties, are being increasingly used to explain resonance phenomena in photon emission and absorption as well as in photoemission. The older, but still fruitful, techniques of studying discrete energy losses by direct transmission or reflection measurements are now supplemented by optical studies of the complex dielectric constant from which the energy-loss spectra can be extracted. Developments in these and allied areas were the subject of the 2nd International Conference on Vacuum Ultraviolet Radiation Physics—Interactions with Solids held in Gatlinburg, Tenn. in May.

Theory. The great debt of workers in this field to H. R. Philipp and E. A. Taft¹ for their pioneering work with silicon and germanium was acknowledged by J. C. Phillips (Chicago) as he urged the continued development of a balanced theory. He defined such a theory as one in which the solution to the Schrödinger equation was followed by a calculation of the optical spectra detailed enough to permit intercomparison with experiment. He

discussed the pseudopotential and model-potential calculations and emphasized that the small differences could be understood in the main by different distribution of covalent and metallic (free) electrons and lowered screening of the valence electrons for large momentum transfer. Phillips emphasized the importance of calculations like those of M.H. Cohen and T.K. Bergstresser that illustrate the "transferability" of the form factor (from which the detailed optical spectra are derived) computed for one crystal structure to another, and he expressed his belief that geometrical intricacies of reciprocal space for different related crystal structures could be untangled with some simplification.

Frank Herman (Lockheed) described the computer codes developed first at the RCA laboratories and continued at Lockheed; it was most impressive to hear that, of the 50 or so crystals studied in this way over the past two years, half had been investigated in the preceding two months. The ambitious current goal is to clarify band structures (by a combination of theoretical and adjustive techniques using experimental results) to an accuracy of several tenths of an electron volt. Herman emphasized that peaks in the absorption spectrum could arise not only from the near vicinity of critical points in reciprocal space but also from extended regions over which lower and upper energy surfaces were approximately parallel. He also pointed out the importance of photoemission experiments, of the type pioneered by William Spicer, in providing absolute values with respect to the vacuum level of certain critical energies.

John W. Cooper (National Bureau of Standards) gave a very clear exposition of the differences between the conventional ways of expressing the optical absorption coefficient in atomic and solid-state physics, emphasizing that the "density-of-states" factor is absorbed in the normalization when free-state wave functions, normalized to unit energy, are used. He also directed attention to the importance of the centrifugal barrier for final states

in producing a rapid change in phase shift for energies near the barrier height, with accompanying rapid change of partial-wave amplitude. Cooper discussed in some detail the subshell contributions in xenon, cesium, sodium, lithium and calcium; he emphasized the need for a better understanding of many-body (atomic-plasma) effects, of the effect of exchange on final-state wave functions, and of two-electron excitations.

Optical properties. Heinz Raether (Hamburg) presented a tutorial introduction to the correlation between optical constants and electron-energy losses. He pointed out that, whereas in a free electron gas the optical information agrees well with electron energy-loss data, in semiconductors there are significant differences that may, however, be entirely due to differences in sample preparation. This point came up in several subsequent papers too, and it seems to be one of the major sources of discrepancies among different authors. The plasmon interpretation appears to have very good foundation and seldom provokes discussion. However, in insulators, where part of the energy-loss spectrum can be attributed to excitons and part to interband transitions, the energy-loss function becomes more complicated. The result is good agreement in the low-energy range but somewhat poorer agreement at higher energies.

A very great variety of experimental results on many substances was presented by T. Sagawa (Tokyo); he compared the different absorption spectra and made an attempt to assign some of the observed transitions to different combinations of expected transitions. Apparently an enormous amount of experimental material is available. Phillips pointed out, in the discussion, that theoretical interpretation is still lacking, and he called for an attempt at such an interpretation.

A discussion of the alkali halides, centered primarily on those of rubidium and cesium, was presented by K. J. Teegarden (Rochester). He made a strong plea for consideration of the role that orbitals play in the observed phenomena and gave con-

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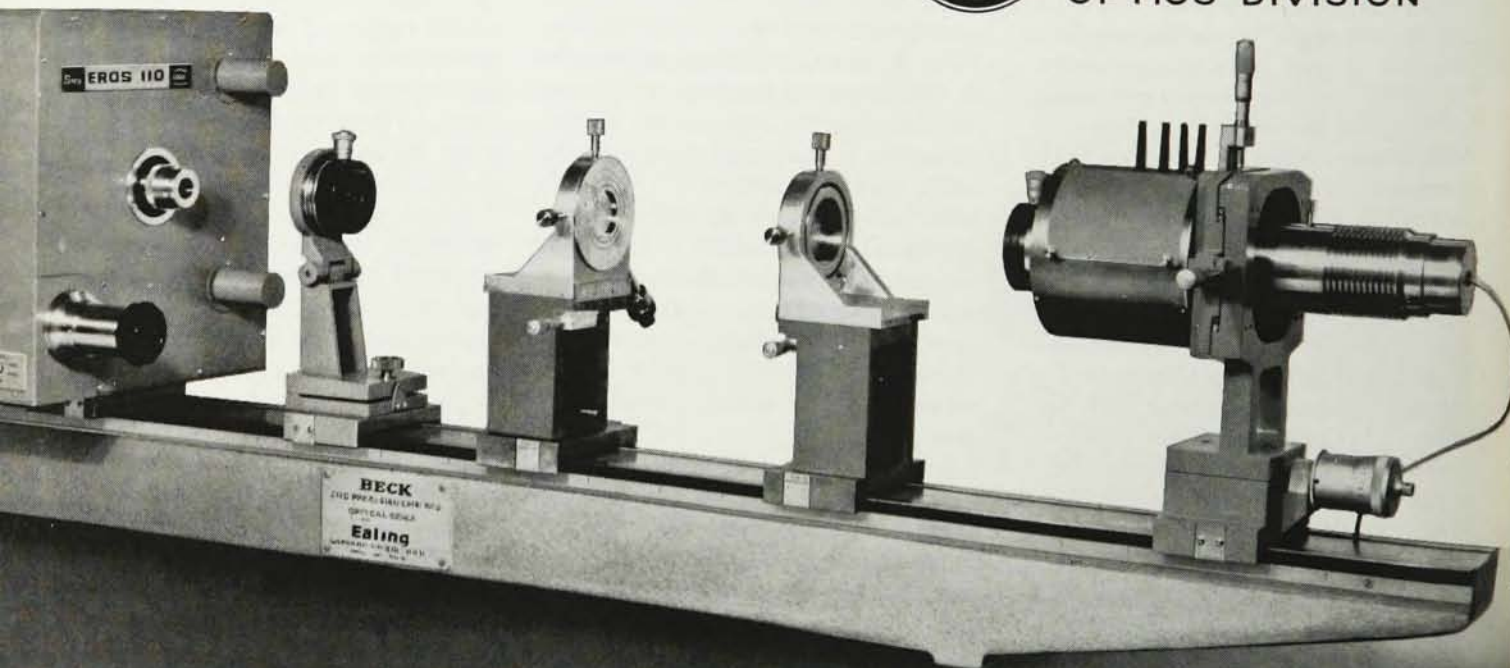
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vincing arguments that the d orbital is responsible for two lines in the rubidium-iodide spectrum, whereas the s orbital does not show up. The excitation lines are observed as closely spaced doublets.

Photoemission. William E. Spicer reviewed the recent work of his group at Stanford in which photoemissive yields and electron-energy distributions were used to probe the electronic structure of solids. He indicated that a principal advantage of this technique over the standard optical techniques is that the absolute energies of the levels involved in an electronic transition can be determined; the standard optical experiments determine only the energy differences between two states separated by $h\nu$. Experimental results from several materials illustrated this review, with considerable detail for the results on gallium arsenide. In this material 12 symmetry points have been located experimentally over a 16-eV energy range; most of them have been located for the first time. Spicer discussed the copper-nickel alloy studies and showed that in copper-rich alloys the top of the copper d band could be clearly located by photoemission and by optical experiments.

The band remains fixed at 2.1 ± 0.1 eV below the Fermi level, independent of nickel concentration in the composition range studied (up to 23% nickel). At the same time new and well-defined states were seen to build up between the top of the copper d states and the Fermi level as nickel is added. These results were taken as strong evidence against the rigid-band model for these alloys, but they agree with the virtual-bound-state model.

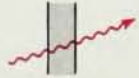
Techniques. Florin Abelès (Paris), who discussed methods for determination of optical constants, defined the best method as one that gives the most accurate results for the complex dielectric function. There is no one best method because the final accuracy depends on the values of the optical constants themselves. Abelès showed for typical values of the optical constants how an error in, for example, the measurement of transmission or reflection is propagated by the different methods of data reduction and how the accuracy of the dielectric function is affected by these errors. The influence of an oxide layer on final accuracy was also shown to depend on the experimental method and on the optical constants.

Instrumentation. A broad selection of commercial monochromators and at-

tachments meet exacting standards, as pointed out by James A. R. Samson (GCA). Improvements recently reported include a Seya-Namioka monochromator equipped with curved slits, which enhance the spectral resolution of the instrument. Commercial normal-incidence monochromators are usable at wavelengths as short as 35 nanometers, due to improvements in the coating of the grating. However, for special purposes it might be still better to use monochromators designed and built in the laboratory. One interesting example of a special design, discussed at this meeting, is a monochromator to be used with distant sources; it consists of a plane and a concave mirror and a plane grating, all working at grazing incidence. The instrument is now under construction and will be used with the DESY accelerator in Hamburg; it should be able to cover the whole ultraviolet spectrum down to a wavelength of 3 nm.

Plasma effects. The history of transition radiation was reviewed by G. Sauerbrey (Berlin); he started with the classic paper by I. M. Frank and V. L. Ginzburg², which predicted that a charged particle would radiate in passing from a medium of one dielectric constant to a medium of another.

PLASMON EXCITATION PROCESSES

EXCITED BY	k CONSERVATION	DECAYING INTO	EFFECT	SCHEMA
Electron	Yes	Electron	Characteristic energy loss	
Electron	Yes	Photon	Plasma radiation	
Electron	No	Photon	Radiative decay of surface plasmon	
Photon	Yes	Photon	Optical plasma resonance in optical transmission	
Photon	Yes	Photon	Optical plasma resonance in reflection	
Photon		Electron	Optical plasma resonance in photoemission	
Photon	No	Photon	Plasma resonance excited by light (PREL) and decaying by light emission	

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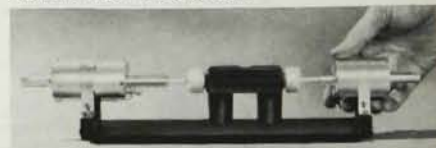
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The need for a boundary between the two optical media distinguishes transition radiation from other radiations such as bremsstrahlung, Cerenkov radiation and ordinary fluorescence. Transition radiation may be distinguished from bremsstrahlung by the energy dependence, which for bremsstrahlung is approximately $1/E$, and by the incomplete polarization of bremsstrahlung. Sauerbrey spoke of two important theoretical contributions; that of R. H. Ritchie, who calculated the dependence of the absolute emission intensities expected on the dielectric constants of the medium, and that of R. A. Ferrell, who predicted that plasmons might decay by nearly monochromatic photon emission. Both theories have been amply verified. Sauerbrey pointed out that the tangential surface plasmons first postulated by Ritchie were thought to be nonradiative, at least if the surface were completely smooth. He gave evidence to show that, for real surfaces, the surface plasmons may indeed radiate, and presented detailed evidence for this radiation in the case of silver. The possible role of surface roughness in the radiative decay of surface plasmons was suggested by E. A. Stern about two years ago.

A discussion of the optical plasma resonances was given by Wulf Steinmann (European Space Research Labs); he cataloged all the plasmon excitation and decay processes shown in the figure. Plasmons were first discovered in electron-transmission experiments, the so-called "characteristic energy loss experiments" (first row of figure). An electron beam was directed through a foil and the energy-loss spectra of the emerging electrons was noted. In the second row of the figure are shown the plasma-radiation experiments described by Sauerbrey. In these experiments an electron passes through a thin foil and the plasmon-decay radiation is observed either on the incident or exit side of the foil. Steinmann divided all optical plasma-resonance experiments into two classes, those in which the k vector of the plasmon is conserved between the excitation and the decay process, and those in which it is not. The surface plasmon-dispersion relation for the finite foil, first deduced by Ritchie and H. B. Eldridge, has been of considerable value in understanding these plasma-resonance effects. Much data have

been accumulated for the optical transmission of thin foils near the plasma frequency. A dip in the transmission was first noted by S. Yamaguchi for silver, the dip being attributed to photons that have excited plasma resonances in the thin foil. Since Yamaguchi's work similar structure has been seen in many metals. Structure is also found in optical reflection experiments, which avoid the serious experimental problem of maintaining a thin, self-supported, unoxidized foil. This resonance in reflection was first suggested by Ferrell and Stern at the first conference in this series; it has been seen recently with 10-nm-thick aluminum layers evaporated on a glass slide at ultrahigh vacuum.

An alternative to plasmon light emission is photoelectric emission. This process, of course, can occur only if the plasmon that gives its energy to the photoelectron has an energy exceeding the work function of the metal. Steinmann referred to the paper by B. Feuerbacher, M. Skibowski, and R. P. Godwin at this conference in which the photoelectric yield is reported to exhibit a strong maximum at the plasma wavelength.

In the second class of plasma resonances, those in which the k vector of the plasmon is not conserved, Steinmann referred first of all to the PREL ("plasma resonance excited by light") experiments. In these experiments light is emitted by a radiative surface plasmon, which has a wave vector different from that of the plasmon created by the incident photon. Thus light is observed in a direction that coincides neither with the incident nor the specular-reflection direction. Stern attributes the change in wave vector to scattering on surface irregularities. Results on PREL were reported by Raether on the metals silver and potassium, but Steinmann said that PREL radiation in aluminum has not so far been observed.

Energy loss. A description of the effects of neighboring interband transitions on the energy, intensity and damping of plasmon excitations was given by Cedric Powell (National Bureau of Standards). He showed that the contributions to ϵ_1 and ϵ_2 , the real and imaginary parts of the dielectric permittivity, by an interband transition can be computed in terms of three transition parameters, namely the oscillator strength, energy and half width of the transition. These contributions

may be incorporated into a free-electron-gas model of a solid, and the loss function, $-\text{Im}(1/\epsilon)$, can then be calculated for comparison with energy-loss data. Although the source of structure in the loss function is not immediately obvious one can fit the experimental data with a reasonable model and with suitable adjustment of the appropriate parameters. Powell presented several cases in which such fittings (by computer techniques) gave new information on interband transitions and in addition were in good agreement with observed reflectance data; useful comparison with the theoretical band structure is also possible, and this structure in turn may be related to measured secondary-electron spectra.

Several possible nonlinear interactions between plasmons, photons and electrons were discussed by R. H. Ritchie (Oak Ridge National Laboratory). The cross section is small for most of these interactions, although several processes are possibly within the limits of experimental observation under conditions of intense particle bombardment and irradiation with laser beams; the most promising process appears to be electron-energy gain from a photon through the intermediary of a radiative surface plasmon. Current experimental interest in surface-plasmon radiation from rough surfaces has led to study of the interaction of plasmons and photons with inhomogeneities in a solid by a quantum-theoretical approach. Ritchie suggested that a thin film deposited on an optical grating would have the desired properties for study of the dispersion characteristics of both tangential and normal surface plasmons.

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2. I. M. Frank, V. L. Ginzburg, *J. Phys. USSR* **9**, 353 (1945).

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L. C. EMERSON
Oak Ridge National Laboratory □