X-Ray Evanescent-Wave Absorption and Emission

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The presence and properties of x-ray evanescent waves are demonstrated by detecting their absorption and their excitation in the vicinity of a target surface. The role of microscopic reversibility in relating the two cases is discussed and practical applications of the phenomena to probe interfaces are suggested.

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The process whereby light is totally reflected from the boundary between two optically transparent media has long been of interest to students of optics.[1] One of the more fascinating aspects of the phenomena is the wave-optical prediction of an evanescent or exponentially damped wave that penetrates a short distance past the interface into the more optically rare (i.e., lower refractive index) medium. The latter is usually the atmosphere or vacuum and the incident and reflected light waves are confined to a dense material such as glass. For an electromagnetic wave whose frequency exceeds that associated with the binding of the majority of electrons in the medium, a phase velocity greater than c is obtained. The reflection then occurs externally with an evanescent field penetrating into this medium which now appears to be optically less dense than the vacuum. This is the case that applies to hard x rays on all solid targets and the possibility of total external reflection of such radiation has been known for some time. It is today a matter of some practical importance for x-ray telescopes and collection optics on synchrotron radiation sources.[2]

The possibility of extending the utility of total reflection to study thin epitaxial films on crystals has recently been realized by Marra, Eisenberger, and Cho[3] by combining elastic Bragg scattering and total reflection conditions to help isolate a surface signal. Vineyard[4] has recently presented a theoretical analysis of this problem which may pave the way for quantitative diffraction studies of surface reconstruction in this region. In the following we describe a series of experiments, using inelastic scattering, designed to probe the x-ray evanescent-wave properties under the reflection condition alone. First we observe the absorption of an external beam incident in the total reflection region by detecting target fluorescent x rays, and thereby demonstrate how the strength of the evanescent wave depends on the incident angle via both kinematical (variation of penetration depth) and dynamical (matching to an external standing wave) factors. Secondly and perhaps more interestingly, we observe the emission of x rays from the surface due to internal excitation of the evanescent wave. This case of emission to the evanescent wave by atoms in the near-surface region is strongly related to the previously mentioned absorption case by the principles of microscopic reversibility and reciprocity. While the main purpose of the following shall be to elucidate the physics involved in these phenomena it is perhaps not inappropriate at this point to indicate that these matters may have a highly useful aspect in providing a means of obtaining depth information in x-ray fluorescence and Compton scattering studies of materials. We return to this discussion towards the end of this Letter.

The inset in Fig. 1 shows a schematic layout of the experiment to observe the evanescent-wave absorption. A fine-focus x-ray tube (1 kW) pro-

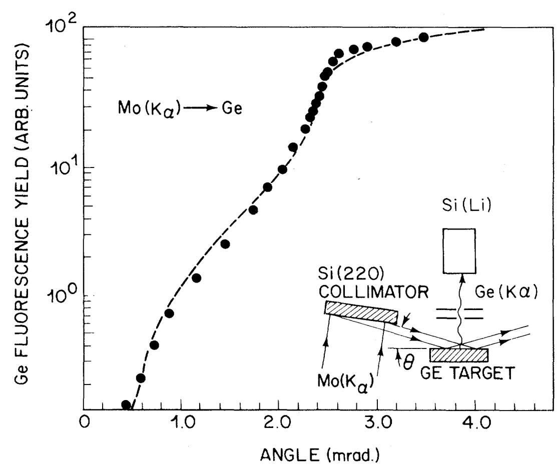

FIG. 1. X-ray evanescent-wave absorption data and Fresnel theory. The inset shows the experimental geometry.

vides a Mo($K\alpha$) beam that is collimated by an asymmetrical (220) silicon crystal to an angular divergence of 0.08 mrad with a beam width of 10 μm. This incident beam then impinges at glancing angle θ on a 50-mm-diam polished polycrystalline germanium sample from which the x-ray beam is reflected. Scattered fluorescent x rays emitted from the germanium sample are detected by a lithium-drifted silicon [Si(Li)] detector whose aperture views a small region of the illuminated target surface. As a result of the geometry virtually all absorption of the incident molybdenum radiation occurs very near to the target surface and consequently all subsequent germanium fluorescent radiation directed out of the target towards the Si(Li) detector will be registered. The dependence of the germanium fluorescent x-ray counting rate on the glancing angle θ in this geometry therefore directly depends on the incident molybdenum absorption rate. We shall therefore refer to this case as the absorption experiment, the data from which appear as the plotted points in Fig. 1. A clear and dramatic reduction in absorption rate is observed for angles decreasing below approximately 2.5 mrad. This is also observed to be the angle below which strong external reflection of the incident beam occurs.

The explanation of the gross reduction in counting rate at small angles is clear. The beam is simply expelled from the target by the reflection process before it acquires a significant chance of being absorbed. Nevertheless, even under conditions of strong reflection, there is a finite chance of an absorption occurring from the evanescent wave that penetrates a short distance into the target. Is it instructive to attempt to explain the shape of the absorption curve on the basis of the properties of this evanescent wave as predicted by the Fresnel-type[1] theory of the reflection. For angles below the critical angle for reflection θ_c, momentum and energy conservation require that the internal x-ray wave have an imaginary wave vector normal to the surface. Its value increases abruptly as the incidence angle is decreased from θ_c. The precipitous drop in the intensity between 2.4 and 2.0 mrad is accounted for by the corresponding drop in penetration depth. The theory, however, predicts that upon further decrease in angle the exponential penetration depth of the evanescent-wave intensity slowly approaches a minimum value of 25 Å. The data points, however, continue to fall strongly as the angle is further decreased. This feature is explained by the fact that the internal evanescent-wave solution must be matched smoothly to the external beam conditions. The external beam consists of both an incident and a coherently related reflected beam of nearly equal intensity. These two beams interfere and create a standing wave whose nodal planes lie parallel to the reflecting surface. The phase of this standing wave, to which the internal evanescent wave must match, varies by π as the incidence angle is decreased from the critical angle to zero. At the critical angle the antinode of the standing wave lies on the interface while at zero angle of incidence the node occupies this position. The reduction in yield for angles below 2.0 mrad is thus mainly due to standing-wave motion changing the surface intensity to which the internal wave must match. The curve drawn in Fig. 1 indicates the yield predicted by this Fresnel theory. We shall return shortly to a more detailed discussion of this exercise.

First consider a related experiment whose geometry is schematically illustrated in the inset of Fig. 2. Here an incident x-ray beam impinges on the germanium target at some large angle. This beam causes the germanium atoms to fluoresce over a wide range of depths into the target. We may now inquire into the nature of the angular yield of this emitted fluorescent radiation in the vicinity of small glancing angles to the surface. We refer to this as the emission case. For this experiment the Si(Li) detector is outfitted with a slit that insures angular resolution of 0.3 mrad, while still being larger than the target

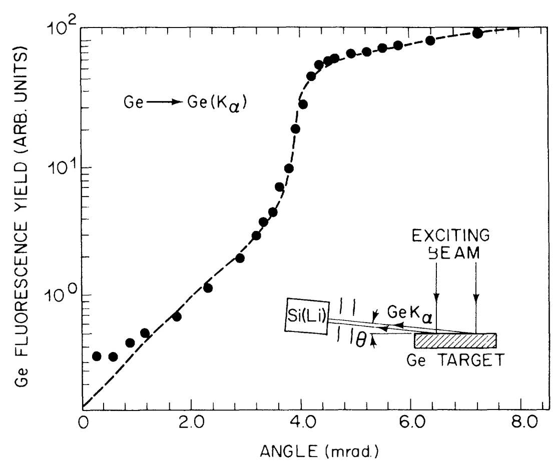

FIG. 2. X-ray evanescent-wave emission data and Fresnel theory. The inset shows the experimental geometry.

source size as it is viewed by the detector.

The results of the emission experiment are plotted in Fig. 2. They look remarkably like those of the absorption experiment discussed previously except for a change of angular scale. Again the x-ray yield drops precipitously just below what would be the critical angle for external reflection for the germanium $K\alpha$ radiation (even though there is clearly no externally reflected beam in this case). The basic reason for this drop in yield at small angles may be deduced by considering the fluorescent radiation emitted in the direction of the surface from deep within the target. For a source far from the interface, the emitted rays may be considered to be independent plane waves that approach the boundary and are reflected or refracted according to the Fresnel laws. This results in a completely inaccessible region for external emission at angles below the critical angle. This is an extremely important feature. Radiation from the bulk atoms far from the surface cannot be observed at angles less than critical. What atoms do contribute to the yield at small angles then? It must be those near to the interface.

While a detailed analytical description of the radiation pattern of a source in the vicinity of a plane interface can be effected along the lines of the classic exposition of the radio problem by Sommerfeld,[5] we can here be satisfied with a much more elementary discussion. We simply note that the principle of microscopic reversibility[6] predicts that the results of absorption- and emission-type experiments should be identical were they performed with the same wavelength radiation. It further follows that at each angle the depth distribution of atoms contributing to the yield is the same. The germanium emission results can therefore be calculated just as if it were an absorption experiment with incident germanium $K\alpha$ radiation. The curve in Fig. 2 is obtained in this way.

Figure 3 shows the angular variation in penetration length and surface intensity calculated for this case (Ge $K\alpha$ in germanium). For angles of observation below 3.0 mrad we expect to be observing emission from only the first 25 Å past the interface. Indeed, even for angles near the critical angle, confinement is still only of order 100 Å. The former length is similar to escape lengths for Auger electrons and this suggests that the evanescent-wave emission may be similarly useful as a very-near-surface probe. We have already utilized the evanescent-wave emission to limit x-ray standing-wave atom-location signals to come from the near-surface layer and these results will be reported on elsewhere. The reader should also note that practical application of the evanescent-wave emission need not use x rays as the initial exciter of the fluorescing atoms. Electrons or heavy energetic ions may be of equal utility. Furthermore, since the exciting beams need not illuminate large regions of the target surface in the emission geometry, constraints on surface flatness are considerably lessened compared with the absorption geometry.

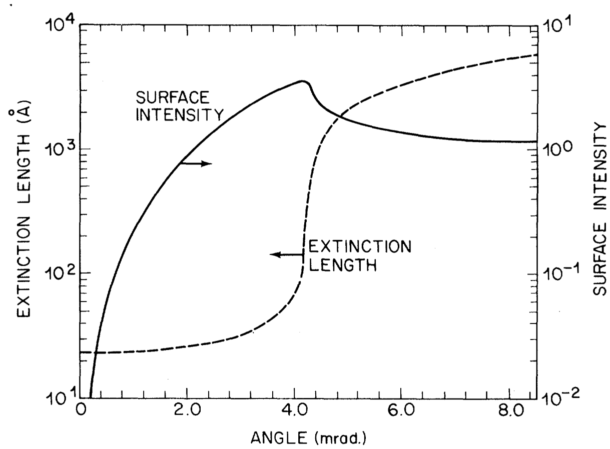

FIG. 3. Angular variation of penetration depth and surface intensity appropriate for the case of germanium.

Finally we would like to return to the Fresnel-theory curves shown in Figs. 1 and 2. The angular dependence of the yield in both cases is obtained as the product of the length and surface yield (both indicated in Fig. 3). For the absorption case, aside from a linear factor of θ to account for the fixed Si(Li) detector aperture, this is simply proportional to 1 minus the surface reflectivity. The direct application of such calculations to the emission experiment relies on the fixed illuminated area of the target surface being totally observable by the Si(Li) detector for only then are the phase-space factors in the two experiments equivalent. Furthermore, while a comparison between theory and experiment in the figures certainly bears out the general physical picture propounded by the simple Fresnel theory, a close inspection shows deviations in excess of experimental counting uncertainties. Analogous deviations have been observed in a beautiful series of reflection measurements by Bilderback.[7] He ascribes them to the presence of a graded rather than sharp interface. We believe that future studies must pay close attention to this question, even considering the inhomogeneous charge density at the surface associated with atomic structure on crystalline targets. Indeed it may well be that reasonably detailed characterization of such interfaces will result from such a project.

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Initiation of Superfluorescence in Coherently Pumped Three-Level Systems

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The fully quantum mechanical treatment of superfluorescence buildup in the linearized regime of superfluorescence is presented for coherently, optically pumped three-level systems. It is shown that to order τ_p/τ_R, where τ_p is the pump time duration and τ_R the characteristic superfluorescence time, fluorescence buildup during the dynamical pumping process contributes significantly to quantum initiation of superfluorescence.

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Superfluorescence (SF)[1] is the phenomenon whereby a collection of atoms or molecules is prepared initially in a state of complete inversion, and then allowed to undergo relaxation by collective, spontaneous decay. Since Dicke's initial work,[2] there has been a large amount of theoretical and experimental work dealing with this process.[3]

With the exception of the more recent work of Bowden and Sung[4] and Bowden and Mattar,[5] all theoretical treatments have dealt exclusively with the relaxation process from a prepared state of complete inversion in a two-level manifold of atomic energy levels, and thus do not consider the dynamical effects of the pumping process. Yet, all reported experimental work[3] has utilized optical pumping on a minimum manifold of three atomic or molecular energy levels by laser pulse injection into the nonlinear medium, which subsequently superfluoresces.

It was pointed out by Bowden and Sung[4] that for a system otherwise satisfying the conditions for superfluorescent emission, unless the characteristic superradiance time,[1] τ_R, is much greater than the pump-pulse temporal duration, τ_p, i.e., $\tau_R \gg \tau_p$, the process of coherent optical pumping on a three-level system can have dramatic effects on the SF. This is a condition which has not been realized over the full range of experimental data.[3]

It has not only been shown that the dynamics of the pumping process can have dramatic effects upon the SF pulse evolution,[4,5] but also that the

