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THE BACK SCATTERING OF ELECTRONS BY CRYSTALS AT LOW AND HIGH ENERGIES

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Résumé. — La diffusion des électrons peut être utilisée à toutes énergies pour étudier les surfaces cristallines. Nous montrons que le contraste dû à des plans cristallins réflecteurs existe pour toutes les composantes du spectre de secondaires. Ceci permet l'utilisation du courant dû aux électrons rétrodiffusés élastiques et même aux inélastiques, pour les études métallurgiques, spécialement dans le microscope à balayage de haute résolution à haute énergie. On a mis en évidence les mêmes effets jusqu'à 100 eV, et nous proposons d'utiliser aussi la technique de balayage dans cette gamme d'énergie.

Abstract. — Electron scattering can be used as a tool for the study of crystalline surfaces, and for the study of defects in the region of the surface, at all energies. It is shown that contrast from diffraction effects appears in all parts of the secondary electron spectrum. This permits a variety of measurements of both the elastic and inelastic back scattered current to be used for metallurgical studies, especially in the high resolution scanning electron microscope at high electron energies. Scanning techniques at low energies should also prove useful, and evidence is presented for the existence of similar effects down to 100 eV.

1. Introduction. — Because of the strength of the interaction with both bound and free charges, electron scattering represents a powerful method for the investigation of the details of the density distribution of charge on and in the region of crystalline surfaces. Although the details of the transmission of electron beams through thin crystals are well understood (e. g. electron microscopy [1]), it is measurements of the back-scattered electrons (e. g. Low Energy Electron Diffraction (LEED) [2]) which primarily contain information about the surface region. Measurements of the angular and energy distribution of the back scattered electrons as a function of the parameters of the incident beam are difficult to interpret because they only indirectly contain information concerning the details of the dynamical wavefield in the crystal [3]. Furthermore, although exact theories exist, their implementation is difficult because of the length and complexity of the computations required [4] or the uncertainties in the magnitudes of the scattering potential itself, particularly at low electron energies. At high electron energies, surface topology and crystal preparation play an important role [5], while intermediate electron energies (2 000-8 000 eV) present an interesting but unexplored diffraction regime [3]. The only directly available information from back scattering is the periodicity of the crystal structure parallel to the surface from elastic measurements, and the details of impurity contamination from inelastic measurements, i. e. Auger spectroscopy.

2. Back scattering experiments. — In order for an electron to be back scattered (i. e. towards the vacuum)

by the crystal, it must undergo an elastic scattering event : inelastic processes provide only a small momentum transfer and for angles of incidence other than grazing will not deviate an electron by a sufficient amount for it to appear in the vacuum. Thus except for very low energy inelastic electrons which are the result of inelastic cascades, and Auger electrons whose direction of emission is independent of the orientation of the incident beam, the back scattered electron current contains contrast information due to elastic diffraction conditions on the primary beam. This is true for both the elastic and inelastic current, especially when either is integrated to give the net reflectivity of the crystal.

There are two experimentally different, but conceptually similar types of back scattering measurement possible. First, it is possible to measure the total reflectivity of the crystal as a function of the orientation of the incident beam at constant energy. This provides information on the details of the diffraction process and can be interpreted in terms of a suitable phenomenological (pseudo) potential [4] whose real and imaginary Fourier coefficients are related to the crystal structure and the angular dependence of the elastic and inelastic scattering. Such a potential can be used to describe the total back scattering of a perfect crystal in terms of a suitable scattering model which takes into account the details of multiple diffraction and the spectra of inelastic processes possible. Second, it is possible to consider an experiment where the orientation of the primary beams is fixed with respect to the crystal, and the beam, having a suitably small diameter and angular divergence, is scanned over the surface while an arbi-

trary diffraction condition on the incident beam is maintained constant. The resulting contrast in the elastic and inelastic back scattered current is then a function of the local orientation of the crystal or the surface normal (an equally important although unusual independent variable in dynamical diffraction theory) the variations of which are due to the presence of crystalline defects, and surface topography, respectively.

The first of these experiments is closely related to typical electron diffraction measurements [6] where the usual situation is the observation of the intensity in the specularly reflected beam as a function of electron energy (typical in LEED) or more infrequently as a function of incident orientation at constant energy, as a rocking curve (variation of diffraction angle at constant azimuth), or a rotation diagram (variation of azimuthal angle at constant polar angle). The second of these experiments is the typical situation in scanning electron microscopy, usually at high electron energies.

The following discussion is intended to demonstrate that at least in a qualitative way the same mechanism, the diffraction condition on the incident electron beam, is responsible for contrast in both scanning and diffraction measurements at high and low electron energies. In particular, the observation of defect contrast (i. e. dislocations), in the back scattering mode of the high resolution scanning electron microscope is evidence for supporting the development of scanning techniques at comparatively low electron energies for the purpose of detailed studies of crystal surfaces.

3. Electron reflectivity vs. incident beam orientation. — In recent years, considerable interest has been shown in the variation of the total secondary electron spectrum (i. e. all energy components of the electron current emitted from the crystal during electron bombardment) as a function of the orientation of the incident electron beam. Detailed investigations [7] in this laboratory has shown that as far as the position and symmetry of the contrast is concerned, a qualitative analysis points to the diffraction conditions of the primary incident beam as the underlying mechanism. Ignoring the details and fine structure of the contrast, in the energy range between 75-2 000 eV (and above) at all but grazing incidence, the total secondary electron current (i. e. total electron reflectivity) appears to be related to the excitation of a sequence of strong reflections primarily associated with crystal planes having low indices ($h^2 + k^2 + l^2$ less than 12). This effect is seen in figure 1 where the structure in the total secondary emission (here measured as the current from crystal to ground) is shown as a function of polar angle of incidence, near to normal incidence. The angular widths given are for the position of the first strong reflection, here (002), and are seen to correspond well with observed minima in the secondary current [7]. Detailed analysis of the angular dependence of various

FIG. 1. — Total secondary electron current (measured from crystal to ground) as a function of polar angle, for incident energies between 75-2 000 eV. Angular widths are for the (002)-(002) reflections. W (110) surface.

energy losses can be carried out in two ways. Figure 2 shows the angular dependence of the secondary electron spectrum integrated from zero loss up to some well defined cutoff energy, for a primary beam at 2 000 eV. In both the figures, the crystal is W ($\bar{1}10$) and the incident beam is maintained in the (110) plane. It can be seen that the structure in the secondary spectrum has the same symmetry at all cutoff energies shown (this is true qualitatively for the comparison between the elastic (zero loss) and total inelastic currents as well (not shown)). A second analysis can be made from examination of the angular variation of specific energy components of the secondary electron spectrum. In the analysis of figure 2, cutoff was determined by applying a retarding voltage to the LEED optics which were used as a collector. In this latter case, it is necessary to use phase sensitive detection (as in Auger analysis) : the results are shown in

FIG. 2. — Secondary electron current vs. polar angle, as a function of inelastic cutoff energy, for W ($\bar{1}10$) surface, at 2 000 eV primary electron energy. The current is measured with standard LEED optics, and corresponds to the secondary electron emission from totally elastic (i. e. primary energy) electrons up to electrons having lost a maximum energy given by the cutoff voltage.

figure 3, where various components of the inelastic spectrum are shown as a function of incident angle, for the surface Ni (100) : incident energy 600 eV and

FIG. 3. — Inelastic spectrum as a function of polar angle, for a given energy loss. Ni (100) surface, incident energy 600 eV, collected with the LEED optics with phase sensitive detection. Energy width approximately 5 eV.

the incident beam maintained in the (010) plane [8]. Again the symmetry of each component part of the inelastic spectrum is the same. Similar results are shown for a constant energy rotation diagram in figure 4, where various components of the secondary

FIG. 4. — Various components of the secondary electron emission, integrated over 2π solid angle, as a function of azimuthal direction ϕ , for 1500 eV incident electron energy, from the (001) face of Al, at 45° polar angle. The crystal current is the net secondary electron current, measured between crystal and ground.

spectrum, integrated over 2π solid angle, are measured as a function of azimuthal angle, for an incident beam at 45° , at 1500 eV, from the Al (100) surface (STERN, R. M., BLANC, E., DUFAYARD, D., AILLE, G., HOWIE, A., private communication, to be published). Similar contrast is found in all parts of the inelastic spectrum, except at energies below about 10 eV corresponding to true secondaries. Contrast in

the Auger signal should be noted as well : it's origin is complicated, but is at these angles primarily from the back scattered elastic current since the Auger mean free path is of the order of 5 Å in aluminium [9] (HOWIE, A., private communication).

Although a complete, quantitative analysis of the origin of the contrast in the secondary electron spectrum is complicated by the energy and angular dependence of the elastic scattering, multiple scattering, inelastic loss, and other processes which contribute to the net secondary electron current, a highly simplified model can be based on standard two beam dynamical theory. This is shown schematically in figure 5 where the amplitudes of the two types of

FIG. 5. — Behavior of the dynamical wavefield for a two beam case of a symmetrical reflection : a) Relative intensity of the two wave fields ; b) Absorption coefficient for each wavefield ; c) Net absorption $(I_1 u_1 + I_2 u_2)/(I_1 + I_2)$.

wavefields, their respective absorption, and the net absorption is plotted as a function of excitation error for a symmetrical reflection.

To a zeroth approximation, large absorption corresponds to a dynamical situation where there is a strong elastic interaction with the atomic cores, which results in the possibility of large momentum transfer, as well as a large crosssection for inelastic scattering (also with a relatively large momentum transfer possible). Thus one can expect that when the two beam theory predicts an increased absorption, they will be a corresponding increase in the back scattered current in a real

experiment. Since at the electron energies discussed here (75-2 000 eV) strong, low index reflections occur at a large angular interval, in spite of the actual multiple scattering process involved, the character of the contrast in the secondary electron emission is dominated by the strong beams. Any combination of localized absorption and dynamical diffraction will yield contrast of the form of figure 5c. This effect is demonstrated in figure 6, where in the lower curve is plotted the total crystal current (from W (110) at 2 000 eV) as a function of incident polar angle. A computer calculation is performed by superposition

FIG. 6. — Total secondary emission as a function of polar incident angle, for the (110) surface of tungsten, at 2 000 eV incident energy: (upper curve) calculated on the basis of a simple two beam model where the absorption and the secondary emission are proportional; (lower curve) measured variation (crystal current). The calculated curve is a best fit for the parameters used.

of two beam absorption curves, assigning a strength to each reflection which decreases with increasing order: the arbitrary parameters of the calculation [7] are varied until a best fit is obtained, as shown in the upper curve of figure 6. Although the parameters themselves have questionable physical significance, the basis of the model, associating the contrast with the strong reflections can be demonstrated by a prediction based on the calculated contrast at a significantly different energy. In this case, the only parameter changed is the electron energy, and the calculation and measurements of figure 6 are repeated at 500 eV and shown in figure 7. The agreement between the predic-

FIG. 7. — (Upper curve) predicted total secondary electron emission as a function of polar angle, for 500 eV incident energy, using the best fit parameters of figure 6; (lower curve) observed crystal current at 500 eV.

tion and observation is perhaps fortuitous although similar results have been obtained on nickel in the same range of energies. The phenomenological conclusion is that both the elastic and inelastic scattering undergo strong contrast variation in the vicinity of diffraction conditions of the primary beam which excite low index Laue reflections. This argument can also be based on the use of the reciprocity theorem, and the observation of the strong Kikuchi contrast observed on the screen during any of these measurements; the contrast in the secondary emission is just that given by the Kikuchi pattern [7], [9], [10].

4. Dislocation contrast during scanning electron microscopy in back scattered mode. — A constant, albeit arbitrary orientation of the incident electron beam corresponds to a constant, well defined (but arbitrary) value of the elastic and inelastic crystal electron reflectivity. During a characteristic line scan in the scanning electron microscope (SEM), the angle

of the beam with respect to the crystal axis and to the average crystal surface normal, is held constant. The signal measured in any of the back scattered modes (elastic, total inelastic, or energy analyzed) will also be constant for a perfect, flat crystal. If the crystal is slightly bent, then a linear line scan corresponds to a variation in incident orientation appropriate to the geometrical variation of local orientation with position: varying the positional coordinate is the same as varying the orientation coordinate (i. e. excitation error) of figure 5. If the crystal contains a dislocation located near the surface (i. e. within a primary extinction length) then if the diffraction conditions are exactly satisfied for a reflection having the usual contrast conditions $G \cdot b \neq 0$ far from the dislocation, the excitation error far from the dislocation is zero. As the beam is scanned across the dislocation, for a vanishingly small beam crosssection, and angular divergence, the effective excitation error is scanned, first to small positive values, and then to small negative

values (or vice versa), corresponding to the magnitude of the local variation in the orientation of the crystal lattice planes resulting from the presence of the dislocation. The details of the resulting contrast in the back scattered signal depend to a great deal on the characteristics of the electron beam, the geometry of the dislocation, and the diffraction geometry of the incident beam. Detailed, quantitative theories have recently appeared in the literature [11], and are appropriate not only for this problem at typical SEM energies, but probably for the general case of the calculation of back scattered signal at LEED energies as well, although they have not as yet been applied to measurements of the type shown in figures 1-7.

Dislocation images have so far only been observed in the back scattering mode of the high resolution SEM for relatively thin crystals [12], [13] although there is good evidence for the characteristic excess-defect type of contrast predicted by the theories [13]: these observations are reproduced in figures 8, 9. Here the

FIG. 8. — Back scattered (BEI) and transmitted (TEI) electron image, from a thin molybdenite crystal, measured in the high resolution scanning electron microscope with a conventional filament. Filament brightness 3×10^5 A/cm²-sr, beam divergence 2.6×10^{-3} , primary beam current 2×10^{-10} A (TEI) and 7×10^{-10} (BEI), scan times: 50 s/frame (TEI, SEI) and 100 s/frame (BEI).

FIG. 9. — Transmitted, Back Scattered Elastic, and Back Scattered Inelastic (Total Secondary emission) Image from a second molybdenite sample. Both figures show excess/defect (black/white) contrast expected for dislocations in both modes of the SEM.

back scattered elastic current (BEI) collected with an annular solid state detector having an aperture of 0.9π , and the transmitted current (TEI) are shown. These measurements were made in a JEOL SEM-50 A and are described in detail elsewhere [13]. Additional images are presented in the following article. The main point to be made is that there is strong defect contrast to be observed in the back scattered mode in the SEM, and that this contrast is of similar origin to that observed in typical elastic and inelastic diffraction measurements.

5. Dependence of back scattered contrast on electron energy and order of reflection. — The arguments above have been made to demonstrate that the same diffraction effects are responsible for contrast in the back scattered electron signal at low and high electron energies. It is therefore of interest to consider if there exist fundamental differences between experiments performed at different energies, and to what extent these differences are important and hence must be considered in a determination of exactly what type of experiment is to be performed with a particular goal in mind. The question as to whether or not there is a fundamental limitation on the information from a given scattering experiment must also be considered : primary consideration will be given to information concerning the details of the crystal surface.

Preliminary, qualitative arguments can be made from a determination of the average mean free path for electrons as a function of incident energy, as shown in figure 10 for aluminium. Below 1 000 eV a combi-

FIG. 10. — Average electron mean free path for Al as a function of incident electron energy, based on plasma loss and thermal diffuse scattering, only.

nation of electron-electron (plasmon excitation) scattering and thermal diffuse scattering [14] keep the mean free path below about 10 Å (although the ratio of these two loss mechanisms is rapidly changing over this energy range). Between 1 000 and 100 000 eV

the mean free path continuously increases, finally saturating at a maximum value due to relativistic effects in the region of 1 MeV. Except for the variation in the spectrum of inelastic processes involved, the mean free path is such that all measurements below 1 keV essentially sample only the crystal surface. At higher energies it is possible to obtain the same component of the mean free path normal to the surface by making measurements at increasingly more grazing angles of incidence. Measurements below 1 keV only sample the surface, but at high energies it is possible by suitable choice of geometry to make either direct surface, or bulk measurements. In the intermediate energy region, some advantage can perhaps be taken of the possibility of anomalous absorption effects permitting a selective variation of the depth of penetration of the probe with slight variation of the diffraction conditions : little is presently known however about the relative magnitude of the effect [14]. Except for the instrumental problems of beam formation, lens design and operation, etc. measurements in the 1-2 keV region should be useful for extremely surface-sensitive scanning electron microscopy at normal incidence. In particular the possibility of combining SEM optics with standard LEED geometries and instrumentation is an attractive one because of the prevalence of standard LEED systems, and the availability of commercial SEM optics as well.

A significant variation of the elastic scattering with energy can also be expected in the range (from 20-10 000 eV) between LEED and typical SEM measurements. Recent calculations of the differential cross-section based on partial wave methods show significant variations at low energies, but because of poor convergence are difficult to make in the intermediate energy range. To overcome this problem, it is convenient to take experimental data for partial crosssections where available : in one case, that of elastic electron scattering by Hg, sufficient information is available to construct the angular and energy dependence of the atomic scattering factor (HOWIE, A., private communication, HOWIE, A. and STERN, R. M., unpublished) : this is shown in figure 11 where the atomic scattering factor, plotted as $|f(S)|^2$ is shown as a function of $S = 2 \sin \theta / \lambda$, for several primary electron energies. It is expected that such a curve is characteristic for scattering from Au and is representative of the sort of variation expected for most heavy metals. The consequences of such variation is that particularly at low electron energies, the relative strengths of low order reflections are difficult to predict, and in particular, detailed dynamical diffraction calculations must take such variations into account. These large variations with energy of the elastic scattering factors, and hence the variation of intensity with energy of the low order reflections make geometrical interpretation of measurements of reflected intensity as a function of energy difficult (typical of most LEED experiments) and emphasize the utility of constant energy measurements at all energies.

FIG. 11. — Differential scattering crosssection (elastic) (shown as the square of the atomic scattering factor $|f(s)|^2$) as a function of electron energy, calculated from the measured crosssection for Hg, and assumed to be representative for heavy metals, i. e. Au.

One characteristic observation of diffraction effects in the secondary emission contrast at all energies is that there appear to be a limited number of reflections which play a role in the scattering process. A qualitative argument has been presented [14] which considers that the spectrum of reflections which do contribute to the elastic contrast depends on the relative strength of the absorption compared to the elastic scattering. A convenient measure of this is the ratio of the primary extinction length (i. e. the elastic mean free path $\Lambda = u^{-1}$) to the pendullossung period

$$\xi_g/u_0^{-1} = \exp(-2M) v_g/v_0^i$$

where M is the Debye Waller factor, v_g the elastic (real) Fourier coefficient associated with the g th reflection, and $v_0^i = 2\sqrt{E}/\lambda$, the constant imaginary part of the complex potential. It can be argued simply that those reflections which correspond to wavefields which are absorbed (in a distance Λ) before they can interfere with themselves do not contribute to the contrast. This produces an apparent upper limit to the order of the reflection which is usually observed to give contrast at low and intermediate energies in most back scattering experiments: the higher the order the weaker the contrast. An obvious exception is at very low energies where the details of the angular dependence of the scattering must be taken into account. Typically $G^2 = h^2 + k^2 + l^2 \leq 12$.

6. Localized mode contribution to contrast : surface state resonances. — It has recently been demonstrated

that considerable contrast in a particular diffracted beam can be associated with the simultaneous excitation of a surface state resonance (or with a surface step resonance: a similar localized mode having one rather than two dimensional periodicity) [16], [17]. Because the dark field image (see following paper) in transmission SEM shows similar contrast to the bright field image except for the expected contrast reversal associated with image formation for defects near to surfaces, and is similar to that for the back scattered image of the same area, one can assume that each diffracted beam contains the same contrast information. Localized mode contrast is observed in those diffracted beams having a strong coupling through a three dimensional Fourier coefficient with the surface (or step) state [17]. One could then predict that there is enhanced SEM image contrast for such crystalline features when these coupling conditions are satisfied. Thus a particular choice of incident beam geometry which couples the dark field beam used for image formation with a localized mode associated with a step should enhance the contrast of the step: the same should be true for all one and two dimensional defects. An extension of these arguments justifies considering development of high resolution SEM images in the back scattered mode made at intermediate energies (500-5 000 eV) for the detailed study of the defect content of crystalline surfaces. Since the contrast due to the excitation of localized modes is dependent only on the momentum component of the scattered beam normal to the defect, and not on the total energy of the beam, such contrast and hence defect information is contained in conventional back scattered high energy electron diffraction measurements [18]: indeed there is strong evidence that such observations are not uncommon [17].

7. Summary and conclusions. — It has been shown that the same diffraction process, associated more or less directly with the phenomenon of anomalous absorption, and which accompanies the excitation of strong forward (Laue) reflections, is responsible for contrast in the elastic and inelastic back scattered electron current at high and low energies. This mechanism ultimately results in the ability to observe dislocation images in the high resolution SEM in the back scattered mode, and is responsible for the angular variation of the entire secondary electron spectrum. There are therefore strong arguments for the development of intermediate energy SEM measurements to replace (or at least supplement) LEED as a useful tool for the study of crystalline surfaces.

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