Fine structure in coherent bremsstrahlung spectra

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Type-*B* (axial) coherent bremsstrahlung spectra have been obtained with an energy resolution of 42 eV. These show a type of fine structure, due to the contributions of Umklapp events from different reciprocal-lattice vectors in the same reciprocal-lattice plane normal to the beam, not passing through the origin. A plane-wave theory fits the data well if atomic scattering factors for isolated atoms are used. The observation of crystal bonding (solid-state relaxation) effects on the coherent-bremsstrahlung intensities requires a more sophisticated theory for the treatment of the continuous background. Bloch-wave broadening effects are not seen at this resolution.

The γ radiation which is emitted by energetic charged particles traversing natural or artificial layered crystal structures has been the subject of considerable recent research.[1] In the simplest classical picture, the transverse periodic motion of a charged particle moving through a crystal gives rise to radiation. While closely related to undulator, wiggler, transition, and free-electron laser radiation, the use of a crystal as a "wiggler" introduces features which can be understood through the use of the classical theory of particle channeling[2,3] for heavier particles, and, for electrons and positrons at lower energies, many-beam dynamical diffraction[4] or a localized transverse orbital picture.[5–7] Two types of radiation have traditionally been distinguished: channeling radiation (CR) due to transitions between the transverse eigenstates of the particle in the crystal potential,[7–10] and coherent bremsstrahlung[11–13] (CB), in which the recoil generated in the spontaneous emission process is taken up by the crystal as whole in an "Umklapp" process, as in the Mössbauer effect. The relationship between the two kinds of radiation has been extensively discussed.[14] Several uses for these polarized, tunable sources of γ radiation have been suggested.[1]

The purpose of this paper is to report features observed in high-resolution CB spectra, and to find the simplest theory which accounts for the different structure. In particular, we wish to assess the relative importance at this resolution of Bloch-wave and plane-wave effects, of the choice of crystal potential, and of the fine structure due to different reciprocal-lattice vectors in the same plane. Specifically, we have observed a single type-*B* (Ref. 15) coherent-bremsstrahlung peak from low-energy (100 kV) electrons traversing a thin silicon target in the $[\bar{1}\,\bar{1}0]$ axial orientation. As shown in Fig. 1, a wavelength dispersive crystal x-ray spectrometer has been used with a resolution of 40 eV (rather than a conventional energy-dispersive pulse-height analysis system, with typical resolution 200 eV) to obtain high-energy resolution. For this type-*B* emission, the Umklapp vector terminates in a plane of reciprocal-lattice points normal to the beam but not passing through the origin of reciprocal space. The x-ray detector is approximately at right angles to the beam, and the "dipole radiator" axis and Umklapp vector are approximately antiparallel to the beam. We first consider the quantum-mechanical contributions to CB fine structure.

A unified view of CB and CR radiation is obtained by treating it as spontaneous emission arising from transitions between the three-dimensional Bloch-wave eigenstates of the three-dimensional crystal potential.[16,17] The continuum or projection approximation is not made, and the variation of the crystal potential in the beam direction is retained. Figure 1 indicates a possible Umklapp vector **h** and photon wave vector **q**. If the three-dimensional Bloch waves excited inside the crystal have labeling wave vectors $\mathbf{k}^{i'}$ and $\mathbf{k}^{j}$ (primes denoting final states), then the conservation of *crystal* momentum requires

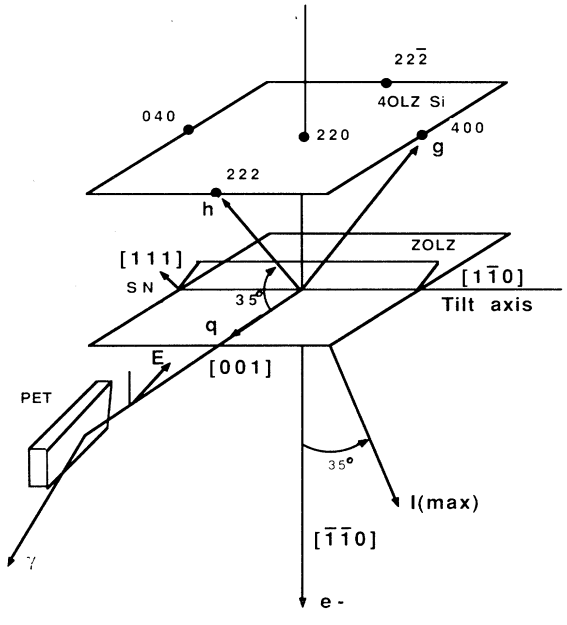

FIG. 1. Experimental geometry. The x-ray analyzer crystal (PET), sample normal (SN), Fourth-order Laue zone (4OLZ), and sample rotation (tilt) axis are labeled. Photon direction [001], beam direction $[\bar{1}\,\bar{1}0]$. Possible Umklapp events g,h indicated. The direction I(max) of maximum CB emission is also shown.

$$\mathbf{k}^{i'}=\mathbf{k}^{j}+\mathbf{g}-\mathbf{q}\ . \tag{1}$$

For an electron beam with wave vector **K**, incident at angles (α,Φ), we allow for the dispersion of the Bloch waves by introducing the eigenvalues

$$\lambda^{j}(\alpha,\Phi)=(\mathbf{k}^{j})^2-\mathbf{K}^2 \tag{2}$$

with a similar primed equation for the final state. The solution to the relativistic kinematics for three-dimensional Bloch waves obtained from (1) and (2) gives the energy of the photon as[17]

$$\varepsilon=hcq=\frac{hc\beta[g_z-(g^2/2K)-(\lambda^{j}-\lambda^{i'}/2K)]}{1-\beta\cos\theta-(g\beta\cos\phi/K)}\ . \tag{3}$$

Here θ and ϕ are the angles **q** makes with **K** and **g**, respectively. The first term here agrees with the classical expression for CB emission energy obtained by calculating the frequency at which the beam electron passes atoms in its path belonging to the family of planes **g**. The second introduces a correction, normally neglected, related to the curvature of the "Ewald sphere" used in diffraction theory. At the exact Bragg condition for reflection **g**, the first two terms sum to zero and CB is forbidden—pure CR results. The final term describes interband CR transitions between Bloch-wave states j and i' (or intraband if between j and j'). Values of the eigenvalues λ^{j} (simply related to the transverse energies of CR theory in the continuum approximation) may be obtained from numerical solutions of the relativistically corrected Schrödinger equation describing transmission electron diffraction.[18] At low energies, **q** may be larger than **g** for CB, thus the orientation of the final state **K′** depends on the unknown $\lambda^{i'}$, but may be determined approximately using plane-wave kinematics. All combinations of "free" and "bound" transitions are possible, resulting in the possibility of CR "sidebands" on CB lines.[7] Both the second and third terms contribute previously unobserved fine-structure to CB emission.

In addition to the above kinematic effects, three other effects broaden CB lines. Lifetime broadening due to dechannelling by phonon scattering may be estimated as

$$\Delta E_1=hc\beta/\Gamma(1-\beta\cos\theta)\ , \tag{4}$$

where $\Gamma=(\mu^{j}-\mu^{i})/\pi$ with μ^{j} the absorption coefficients for the two dominant Bloch waves. These are related to the imaginary parts of the optical potential used to describe the depletion of the elastic wave field by inelastic scattering.[19] Using values of μ^{j} measured in highly accurate electron-diffraction experiments on silicon,[20] we find this contribution to be 1.25 eV at room temperature. An independent effect of temperature (also included in our calculations) due to virtual phonon excitation results in a modification to the static crystal potential through the Debye-Waller factor, and so causes small shifts in the emission energy.[1] Doppler broadening across our detector (at approximately 90° relative to the beam line) contributes a broadening of

$$\Delta E_2=\varepsilon\beta\,\Delta\theta\ , \tag{5}$$

which is 40 eV for the solid angle used. This is the most severe resolution limit—a smaller solid angle results in unacceptable source intensity variation and counting statistics during our 30-min data collection time. The energy resolution of the spectrometer itself (13 eV FWHM), and that due to the divergence of the incident electron beam are both much smaller.

The results shown in Fig. 2 were obtained on a Hitachi EM500 electron microscope, operated at 100 keV and using a lanthanum hexaboride electron source. This offers excellent control of beam collimation (down to 0.1 mRad), a source energy spread of about 1 eV, a small electron probe (about 1 μm in diameter), and the ability to accurately align the target by observation of the transmission electron diffraction pattern. Electron images were also formed to ensure that the region of sample used was free of defects. A Johann-type curved crystal spectrometer was interfaced to the machine,[21] fitted with a P20 flowing gas proportional counter and pentaerythritol (PET) crystal analyzer. A single-tilt side-entry goniometer was used, with a carbon sample holder to minimize background. The sample was rotated outside the microscope until the required orientation was found by setting the $(hh0)$ Kikuchi band normal to the tilt axis. Then the [111] surface normal of the silicon slab is inclined at 35° to the analyser axis (approximately along [001]), as shown in Fig. 1. The beam energy was measured to be 99.086 kv using the Kikuchi pattern method.[22] The smallest angle between **K** (along [110]) and **q** was $88.4°=180°-\theta$. The elevation angular subtense of the spectrometer was 1.2°. Background has been subtracted using a straight-line fit.

Equation (3) shows that the 3.55-kV peak observed results from Umklapp events [**g** vectors in Eq. (3)] lying entirely in the fourth-order Laue zone of silicon along $[hh0]$. (The variation of peak energy with accelerating voltage was used to confirm that the peak is due to CB).

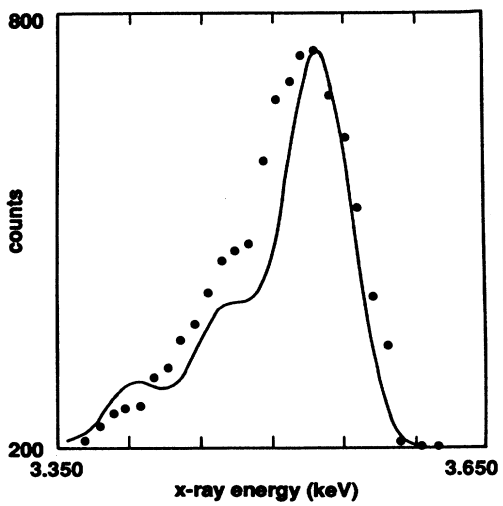

FIG. 2. Experimental CB peak from 4OLZ in Si along $[\bar{1}\,\bar{1}0]$ at 100 kV. Solid curve shows theory 60 sec per data point collection time.

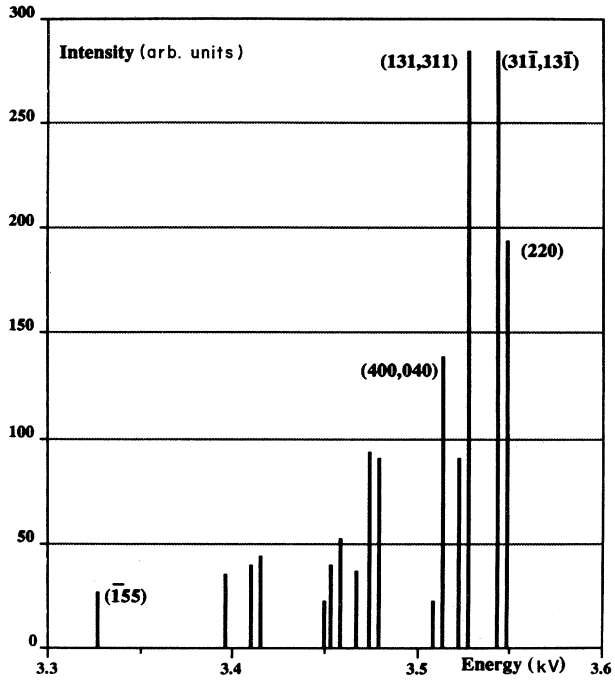

FIG. 3. Intensity and energy of the individual contributions to the spectrum of Fig. 2, obtained from isolated atom calculations. Crystal bonding effects would increase the (220) line by about 0.5%. Dependence of energy on azimuth splits degeneracy of lines with same (cubic) crystal symmetry. All lines fall in the fourth reciprocal-lattice plane from the origin, whose normal is [$\bar{1}\,\bar{1}$0].

Figure 3 shows the idealized monochromatic emission expected from the relevant reciprocal-lattice vectors in this plane, using energies from the first two terms of Eq. (3). The second term in the numerator produces different emission energies for **g** vectors in the same plane. The third ϕ-dependent term splits the degeneracy between crystallographically equivalent planes, since it depends on the detector azimuth. The peak heights in Fig. 3 were obtained from the plane-wave theory of CB, which gives the emission energy per incident electron per unit solid angle for one Umklapp event as

$$\frac{dP}{d\Omega}=\frac{\alpha^3 t(2\pi a_0 e V_g g_z)^2\sin^2\theta}{hc\beta\gamma^2(1-\beta\cos\theta)^5}\ , \tag{6}$$

where a_0 is the Bohr radius, t the sample thickness, α the fine-structure constant, and V_g a Fourier coefficient of crystal potential. Figure 3 ignores certain small cross terms between different Umklapp events which arise in finite crystals and/or in the presence of dechannelling. Values of V_g were obtained from relativistic Hartree-Fock calculations for isolated atoms taken from the International Tables for X-ray Crystallography. The effects of crystal bonding may be estimated from measurements of the relevant structure factors by electron diffraction.[20,23] For example, in Fig. 3, the effect of bonding would be to increase the (220) intensity by about 0.5%. This change, due to solid-state relaxation, is not detectable owing to uncertainties in the estimation of the background, but might readily be detectable using a more efficient spectrometer and longer counting times. More dramatic crystallographic effects are seen in CB spectra from the "contributions" of structure factors which are either zero as a result of crystal symmetry, or very small [e.g., the (222) reflection in Si, which is absent for spherical atoms].[17] The use of CB spectra for the accurate measurement of structure factors is described in more detail elsewhere.[24]

The smooth curve in Fig. 2 was obtained from Fig. 3 by including several additional effects. The x-ray signal detected is the result of the three-dimensional Bragg diffraction (at the analyser) of a polarized CB x-ray beam which originates from a moving (relativistic) electron source. This problem has been studied in detail,[25] the main effects are the following: (1) The Doppler shift across the angular subtense of the analyzer crystal. Different portions of the analyzer crystal satisfy the Bragg condition for different component wavelengths. (2) The polarization of the CB emission incident at the Bragg angle on the analyzer. (3) The intrinsic energy resolution limit of the spectrometer. (4) Energy broadening due to the incident beam divergence. This increases with the angle between **K** and **g**, from 1 eV for planes normal to the beam [**g**=(2,2,0)] to 130 eV for inclined planes whose reciprocal-lattice vector lies in the same reciprocal-lattice plane [e.g., **g**=(5,−1,5)]. The total broadening due to all these effects is 42 eV near (220), resulting in the theory curve shown.

More complete Bloch-wave calculations for CB show that the energy broadening due to deviations from plane-wave character are smaller than most of the effects listed above. (At the exact Bragg condition for planes **h**, a "nearly free" electron model gives the Bloch-wave splitting due to crystal bonding as $2V_h$, and smaller for our axial orientation.[26]) The experimental spectrum shown in Fig. 2 is seen to consist of three subpeaks, not previously resolved. We see that the plane-wave calculations reproduce these three main subpeaks of the "single" CB peak fairly well. We conclude that, at a resolution of 42 eV, the contributions from different groups of reciprocal-lattice vectors in the same reciprocal space plane can be resolved, that Bloch-wave effects are not visible at this resolution, and that relativistic Hartree-Fock calculations for isolated atoms give a sufficiently good description of the crystal potential to account for CB spectra at this resolution, where crystal bonding effects are not directly observable in CB spectra. This appears, however, to be the first observation of fine structure due to different reciprocal-lattice vectors in the same plane. Improvements in spectrometer design and beam stability should make the 0.5% effects of crystal bonding measurable by this technique.

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