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COHERENT ABSORPTION AND EMISSION PROCESSES OF ENERGETIC PARTICLES PENETRATING CRYSTALS

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INTRODUCTION

Swift charged particles moving through an ordered array of atoms in a crystal experience periodic perturbations which can give rise to coherent absorption or emission of photons. Indeed the phenomenon of coherent bremsstrahlung arising from ultrarelativistic electron penetration of crystals has been studied for many years.¹ The complexity of the Fourier components of the electric field experienced by the particle is greatly reduced if the direction of motion lies along a crystal axis or in a crystal plane. Here, however, the motion of the particle may be governed by correlated collisions with the atomic rows or planes and the particle is said to be "channeled." In this case another periodicity is involved in the motion and transitions between bound states in the transverse motion can lead to radiation of another type which has been dubbed "channeling radiation."

Channeled, highly stripped ions of sufficiently high atomic number $Z_1 \gtrsim 5$ can exist in a crystal in well defined quantum states and transitions between quantum states can be affected when resonant frequencies, dependent on crystal direction and velocity, are achieved. Studies of resonant coherent excitation of channeled ions constitute a form of absorption spectroscopy which yields information on the ionic states and the crystalline milieu.

Experimental observation of both the absorption²⁻⁵ and emission phenomena⁶⁻¹¹ in channeling has occurred only in the past two years and a brief review and collation of the various effects observed will be presented in this paper.

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Frequencies Due to the Almost Rectilinear Path of the Particle

The set of frequencies experienced by the particle arising from its almost rectilinear path depends on its velocity v_i and the distance between perturbing centers, e.g., for particles passing through a crystal parallel to a row of atoms with an interatomic spacing d ; $v = k(v/d)$ $K = 1, 2, 3, \dots$. In a real crystal the nature of the field will depend on the detailed symmetry conditions. An example is shown in Fig. 1 for a particle moving at an angle θ with respect to a $\langle 100 \rangle$ axis in the (100) plane of a face centered cubic crystal.⁵

For a simple FCC crystal we place the origin on an atom and define orthogonal U , V , and W axes directed towards the corners of a cubic unit cell. The field is the negative gradient of the potential function $\phi(U, V, W)$ which may be written as a triple Fourier sum

$$\phi(U, V, W) = \sum_{hkl} \phi_{hkl} \cos(2\pi hU/a) \cos(2\pi kV/a) \cos(2\pi lW/a) . \quad (1)$$

In the case of FCC the only non-zero coefficients ϕ_{hkl} are those in which the indices are either all even or all odd. For a particle channeled in the (100) plane on a straight line making an angle θ with W the center of the channel is the $U = \frac{1}{4}a$ plane and we suppose the ion to travel a fixed distance b_U from the midplane. The trajectory is then

$$W = vt \cos \theta , \quad (2)$$

$$V = vt \sin \theta , \quad (3)$$

$$U = \frac{1}{4}a + b_U \quad (4)$$

Fig. 1. Coordinate system for (100) planar channeling in the FCC system.

The perturbation frequency is

$$\omega^{\pm} = (\ell \cos\theta \pm k \sin\theta)(2\pi v/a) \quad (5)$$

For (100) planar channeled particles $h = 0$ and we denote the harmonic frequencies by (ℓ, k) . Physically this corresponds to the frequency of crossing (ℓ, k) planes which are orthogonal to the plane and which make an angle $\alpha = \tan^{-1}(k/\ell)$ with V .

For relativistic leptons such planar crossing frequencies are seen in the energy of the emitted coherent bremsstrahlung. For channeled positive ions the effects of these frequencies are seen in resonant coherent excitation.

Resonant Coherent Excitation (RCE) of Channeled Ions

Crucial to an understanding of RCE effects observed is a rudimentary knowledge of the incoherent processes occurring in ion channeling. The avoidance of small impact parameter collisions caused by the channeling process limits the ion to close interactions with only conduction or valence electrons. For $v_i \gg v_0$ the electrons will have velocities lower than the penetrating ion. Thus for electron capture and loss we can view the penetrating ion as stationary and being bombarded by electrons of energy $1/2 m_e v_i^2$; e.g., an ion moving with an energy 2 MeV amu can be viewed as being bombarded by electrons of ~ 1 keV of energy. The result is that a large fraction of channeled ions of sufficiently high $Z \gtrsim 5$ which have one or two $1s$ electrons bound to it can pass through a crystal (e.g., Au 5000 Å thick) without changing charge.¹² If, on the other hand, a bound electron is raised to an $n = 2$ level the ionization cross section is greatly increased (a factor of ~ 10). The experiment basically consists in measuring the charge fraction remaining in the initial charge state after passage through the crystal as a function of ion energy. A monotonic secular trend with energy is anticipated from the change in electron capture and loss cross sections. A resonant transition which excites the ion also increases the probability of collisional electron loss and is signaled by a decrease in the surviving charge fraction.

An example of the effect is shown in Fig. 2 for N^{6+} ions channeled in a (100) plane at an angle $\theta = 38^\circ$ from a $\langle 100 \rangle$ axis where we plot the fraction of N^{6+} ions transmitted through a ~ 1000 Å crystal of Au. For a given transition E_{ij} in the moving ion to occur the ion energy E_r required for RCE is

$$E_r^{1/2} = \Delta E_{ij} (a/h) (\ell \cos\theta \pm k \sin\theta) \quad (6)$$

ΔE_{ij} for the $1s \rightarrow 2p$ transition in an isolated ion is 500 eV. The arrows on the figure indicate the anticipated positions of the resonances (eq. 6).

Fig. 2. Ratio $R = N^{6+}/N^{6+} + N^{7+}$, in (100) planar channeling at $\theta = 38^\circ$ vs ion beam energy.

Two features are noteworthy and indicate the information obtainable from these experiments. 1) For the odd harmonics the peaks are shifted to lower energies. 2) The even harmonic resonance (2,2) is split into a doublet on the high and low side of the resonance. These shifts and splittings arise from the fact that the ion is not in its vacuum state.

Two principal effects perturb the energy levels of an ion moving in a crystal channel compared to its vacuum state. First, the ion is contained in a potential gradient (crystal field) formed by bounding atomic rows or planes. The results is, e.g., a reduction in the $n = 1$ to $n = 2$ spacing, and accounts for the general observation of lowered resonant velocities.

The second effect is caused by the presence of an electron "wake"; electrons scattered from the ion ($v_i \gg v_0$) create an enhanced electron density wave which follows the ion; the distance to the first node in the wave being $\sim 2\pi\hbar v_i/\omega_p$ where ω_p is the plasmon frequency. Thus the ion experiences a velocity dependent dc field which can act to Stark mix the ionic states. Since the field acts always in the z direction the mixing occurs between the $2p_z$ and $2s$ states. In $\langle 100 \rangle$ and (100) symmetries only the even harmonics give strong longitudinal (Z) Fourier components; hence the observation of a Stark split doublet in the (2,2) harmonic of Fig. 2.

Measurements of more than 50 such resonances (some deeper than 50%) for hydrogen-like B^{4+} to F^{8+} and He-like N^{5+} and O^{6+} have been made by the Oak Ridge group and much detailed information on ionic states in crystalline environments has been developed.

Frequencies Due to Oscillatory Motion of Positive Particles in Planar Channels; Radiation from Planar Channeled Positrons⁶

The containment of positive particles between planes of atoms in a crystal may be viewed as being due to the repulsion of two adjacent sheets of screened nuclei; each sheet potential being made up of an appropriate sum of the potentials of the atoms lying in the plane. The potential is symmetric about the midplane and causes oscillatory motion. From measurements of the wavelengths of planar channeled positive ions, detailed potentials have been derived.¹³ A reasonable approximation to the potential is given by $U = U_0 \cosh bx$ where b is a screening constant and x is the distance from the channel midplane. For the purpose of this discussion we will further approximate and assume a classical harmonic potential with a transition frequency $\omega_0 = (k_0/m_0)^{1/2}$ where k_0 is the force constant of the planar potential and m_0 is the particle rest mass. For positive ions at nonrelativistic velocities $\hbar\omega_0$ is quite small (e.g., k_0 is typically $\sim 40 \text{ eV/\AA}$ which for a proton mass gives $\hbar\omega_0 \sim 0.4 \text{ eV}$). For relativistic positrons, however, the photon energy can be much larger.¹⁴ Since $m = \gamma m_0$ the frequency in the laboratory frame $\omega_L = \omega_0 \gamma^{-1/2}$ and the emitted radiation is Doppler shifted to give a maximum frequency (in the forward direction) of $\omega_{\text{max}} \approx 2\gamma^{3/2} \omega_0$. Thus for 56 MeV positrons⁶ ($\gamma \approx 100$) in the (110) silicon the photon energy should be $\sim 40 \text{ keV}$. Moreover the photon energy can be varied by changing the incident-particle energy. The radiation is predicted to be highly directional (half angle $= 1/\gamma$); linearly polarized; and considerably more intense than ordinary bremsstrahlung within the bandwidth. Figure 3 shows a photon spectrum observed⁶ in the forward direction for 56 MeV positrons channeled in the (110) plane of Si (18 μm thick).

Fig. 3. Positron planar channeling radiation spectrum for 56 MeV positrons in (110) Si.

Table 1. Spectral features of positron channeling radiation.

Plane	γ	$E\gamma(\text{calc})$ (keV)	$E\gamma(\text{expt})$ (keV)	Linewidth (%)
(110)	99	33-38	36.5 ± 1.0	≈ 20
(110)	111	38-44	42.5 ± 0.5	24
(111)	111	29-35	35.3 ± 0.5	26
(100)	111	46-51	46.7 ± 0.5	18

Table 1 gives a comparison of the energies of the peaks in the photon spectra obtained for different planar channels in Si from an anharmonic-oscillator model (hence the range in calculated frequencies) based upon a planar-averaged static Moliere potential.⁷ Table 1 also gives the measured linewidths. The calculated linewidth for (110) planar channeling for a beam divergence of 1 mrad is 13%. Both the skewed line shape and the somewhat larger linewidth for (110) and (111) planar channeling probably result from Doppler broadening caused by the relatively large [≈ 7 mrad (FWHM)] beam divergence in the experiment.⁶

Radiation from Axially Channeled Positrons

Except for the rare case of hyperchanneled positrons¹³ which requires extremely tight collimation, axially channeled positrons are free to wander from one axial channel to another. Measurements of the axial channeling radiation⁶ yielded a spectrum which was explainable in terms of a mixture of planar channeling and coherent bremsstrahlung.¹⁰

Radiation from Planar Channeled Electrons⁸

Since the quantum mechanical and classical harmonic oscillators have nearly identical observable properties, both models predict the observed spectral peak from planar channeled positrons correctly. The case for planar channeled electrons, however, is quite different. The planar potential for channeling of electrons is illustrated in Fig. 4. The electron is constrained in its motion to weaving through a single atomic plane and may be viewed as being captured into a one dimensional screened Coulomb potential. The eigenstates of this potential are characterized by only one quantum number, n , and dipole selection rules give $\Delta n = 1, 3, \dots$. The radiation characteristics of plane polarization Doppler shift and angular confinement, etc., should, of course, be the same as for planar channeled positrons. In Fig. 5 we show the emission spectrum observed in the forward direction for 56 MeV electrons channeled in a (110) plane of Si (18 μm thick). The lines recorded here are all $\Delta n = 1$ and form a series not unlike the "Lyman α ," Balmer α "...

Fig. 4. Schematic potential for electrons in planar channels. The three vertical lines represent atomic planes.

Fig. 5. Electron planar channeling radiation spectrum for 56 MeV electrons in (110) Si.

of its three dimensional counterpart ($\Delta n = 3$ transitions have been observed in subsequent experiments).

We have made an analysis of the spectra observed in three different planes of Si at electron energies of 28 and 56 MeV, using the following potential:

$$V(x) = V_0 e^{-k|x|} + A, \text{ for } 0.1 \text{ \AA} \leq |x| \leq \frac{1}{2}d \quad (7)$$

and

$$V(x) = V_1 x^2 + B, \text{ for } |x| \leq 0.1 \text{ \AA} . \quad (8)$$

The parameter values for the potential were chosen to predict, as closely as possible, the spectral peaks for the (110) case at 56 MeV. The cusp-like nature of the potential gives rise to unequally spaced energy levels, resulting in many spectral lines. The calculated photon energies⁹ are given in Table 2 for comparison with the experimental values. The dependence of photon energy on electron energy has been calculated for the four lowest energy levels over the range from 10 MeV to 250 MeV. The behavior is described very well by a power law, with the exponent varying from 1.64 (for $n = 1$ to $n = 0$) to 1.90 (for $n = 3$ to $n = 2$). From the estimated linewidths, the effective coherence lengths $2\gamma^2\beta c\tau$, where $2h/\tau$ is the fwhm linewidth, have been calculated, and are included in Table 2.

Radiation from Axially Channeled Electrons¹¹

In the channeling picture crystal axes may be viewed as a set of parallel "strings" of closely packed positive atomic cores with a radially symmetric continuum potential. Injection of electrons with sufficiently low transverse energy can lead to bound states in this two dimensional screened atomic potential which are eigenstates of the Hamiltonian

Table 2. Spectral features of electron-planar-channeling radiation.

Beam Energy (MeV)	Crystal Planes	Calculated Photon Energy (keV)	Measured Photon Energy (keV $\pm$ 2 keV)	Linewidth (keV $\pm$ 2 keV)	Effective Coherence (Length μ m)
28	{001}	31	31	4	0.6
		40	40	6.5	0.4
		25	25	5.5	0.5
56	{001}	100	99	16	0.6
		65	64	11	0.9
		44	39	8	1.2
	{110}	127	128	15	0.7
		92	94	13	0.8
		68	68	8	1.2
	{111}	53	52	5	1.9
		-	99	20	0.5
		-	37	10	1.0

$$[P_{\perp}^2/2m + U(\vec{r})]\psi_2 = E_{\perp, \nu} \psi_u(\vec{r}) \quad (9)$$

where $p_{\perp}$ and r are projections of the momentum and position on a plane perpendicular to the axis and U is the string potential. Radiation from axially channeled electrons then arises from transitions between these bound states.¹⁵ After suitable relativistic transformations one obtains for the photon energy in the lab system

$$h\omega_L \approx 2\gamma\omega_R \quad (10)$$

where ω_R refers to the rest frame frequency

$$h\omega_R = E_{\perp, \nu_R} - E_{\perp, \mu_R} \quad (11)$$

Because of the relativistic increase in $m = \gamma m_0$ the number of bound states of eq. 9 increases with energy. When 28 or 56 MeV electrons were channeled in Si only a continuum with a high energy cut off⁸ was observed because of the plethora of states at these energies. Using lower energy electrons (1.5 - 4 MeV) Andersen and Laegsgaard¹¹ obtained the beautiful results for the $\langle 111 \rangle$ direction in Si shown in Fig. 6. The upper curves are the raw data (proportional counter) and the lower ones represent the extracted channeling radiation. The solid curve is identified with $2p \rightarrow 1s$ transitions. In increasing order in energy the peaks for the 4 MeV data are $2s \rightarrow 2p$, $3d \rightarrow 2p$, $2p \rightarrow 1s$, and $3p \rightarrow 1s$ transitions. The line energies and

Fig. 6. Electron axial channeling radiation for 1.5 to 4 MeV electrons incident to the $\langle 111 \rangle$ direction in Si.

e^- energy dependences agree reasonably well with many beam calculations using an approximate $U(r) \propto 1/r$ potential. These other recent results on Si $\langle 110 \rangle$ obtained at SUNY Albany are also well fit by a direct solution of the two dimensional Schrödinger equation with a string potential made up from atoms described by a Molieré potential.¹⁶

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