Channeling radiation beyond the continuum model: The phonon "Lamb shift" and higher-order corrections

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The quantum theory for channeling radiation from mega-electron-volt electrons is extended to include higher-order corrections from phonon scattering. The formalism developed to describe radiation line profiles may be applied to any radiating system exposed to external perturbations. For channeling radiation, it is shown that at electron energies above a few mega-electron-volts, the dominant correction to line energies is of third order in the strength of the interaction giving rise to phonon scattering. Detailed analytical expressions as well as simple estimates are derived for both the second- and third-order thermal corrections. Their magnitude, relative to line energies, is typically of the order of 10^{-3} or less. Corrections due to the periodic structure of axial and planar potentials are shown to be of the same order or smaller. Hence the present investigation results in a firm theoretical foundation for channelingradiation spectroscopy which may then be used as a valuable probe for solid-state parameters, potentially in the study of vibrational anomalies in high- T_c superconductors. For electrons of a few megaelectron-volts, a shift due to photon refraction is important which previously has been included in an *ad hoc* way. The formalism contains this effect and the semiclassical formula for photon refraction in a free-electron gas is rederived within a fully-quantum-mechanical framework.

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I. INTRODUCTION

Channeling radiation is emitted by electrons (or positrons) moving through a crystal nearly parallel to a plane or axis, with transverse motion confined by the averaged planar or axial potential [2]. The frequency spectrum reflects the periods of this motion or, in a quantum description, differences between energy levels of the oneor two-dimensional transverse motion. The number of confined "bound" states increases with the relativistic mass of the electrons and becomes so large at GeV energies that a classical description is appropriate. However, we shall here be concerned only with electrons in the MeV region where transitions between the few quantum states can give rise to a spectrum of sharp, separated lines.

Channeling radiation may be regarded as a kind of coherent bremsstrahlung with the special property that the discrete frequencies are determined by the strength of the lattice potential and not by the periodicity. Precise observations of this radiation—channeling spectroscopy—can therefore be used as a tool for the determination of crystal properties such as electron density [3,4] and thermal vibrations [5,6]. However, this also requires a very accurate theoretical description for the interpretation of experiments, and it is the object of the present paper to give a careful evaluation of corrections to the standard continuum model [1].

In order to define the framework and notation, we summarize in Sec. II the relevant aspects of the basic channeling theory [7]. The picture of electron motion in continuum, thermally averaged axial or planar potentials emerges from a systematic series of approximations, leaving corrections which may be reintroduced as perturbations. The most important ones, which will be studied here, are the periodic structure of the potentials and the fluctuations due to thermal vibrations. It is the applicability of a perturbation expansion, owing to the small charge and high velocity of the projectiles, which makes it possible to aim at much higher accuracy than usual in the description of channeling.

In the following two sections, we develop the formalism to give a systematic account, through an expansion to third order in the perturbation strengths, of the line broadening and line shifts caused by the perturbations. The formalism is general and may be applied to study line profiles for any radiating system exposed to external perturbation. Scattering in the initial and final states of the radiation process must be treated in a coupled manner, and this is accomplished through the introduction of product states, as in the density-matrix formalism, and of associated projection operators.

The effects of thermal scattering are discussed in Sec. V. Higher-order perturbation calculations often become very messy, with extensive formulas and complicated numerical evaluations, and the present are no exception. However, with crude but reasonable approximations, quite simple and transparent expressions are obtained for the leading terms. Albeit small, the corrections turn out often to be at a level ($\sim 10^{-3}$) where more careful evaluation is necessary and we therefore supplement the simple estimates with more accurate formulas (Appendix B) and numerical results for a few examples (Appendix C). Typically, the simple estimates are confirmed within a factor of 2 to 3.

A peculiar phenomenon is encountered in connection with the perturbation expansion: The third-order term contributes a line shift which is often larger than the second-order shift. The reason is that the third-order term contributes to lower order in a second expansion parameter which is essentially the phase difference between initial and final states of the scattering process, developed during the collision. This double expansion is closely related to the sudden-collision expansion in [8], and, in fact, the two formalisms are for the present purpose equivalent [1].

The periodic perturbations, from atomic structure of strings and from string structure of planes, are treated in Sec. VI by a similar double expansion. Again, application of the sudden-collision expansion is an alternative, equivalent method [1]. The level shifts may also be obtained from the Kapitza potential [9] associated with the periodic perturbation.

Although we are mainly concerned with interactions of the projectile with the lattice and their influence on the emitted radiation, it is interesting to note that the formalism also contains the well-known effects of the coupling between the lattice and the radiation field, and our procedure applied to this coupling reproduces the established formulas for photon refraction, due to polarization of the crystal, and for the analog of the atomic Lamb shift due to emission and absorption of virtual photons (Appendix A). Refraction is of direct interest in channeling spectroscopy, leading to important corrections to line frequencies [10,11]. To second order in the coupling between the particle and the radiation field, a Lamb shift for channeling radiation is also obtained, but this is entirely negligible compared to the "phonon Lamb shift" due to emission and absorption of virtual phonons (the second-order thermal shift discussed above).

The concluding remarks in Sec. VII contain partly a brief discussion of electronic excitations, which have been neglected in this paper, partly a comparison with the earlier treatment of line shifts from lattice vibrations by Strauss *et al.* [12]. For the second-order line shift, our estimates differ by an order of magnitude, and this is shown to be due to a strong overestimate—in their treatment—of the correlation of vibrations of different atoms.

II. BASIC CHANNELING THEORY

Most detailed quantum treatments of channeling radiation are based on the Dirac equation which describes the relativistic behavior of spin- $\frac{1}{2}$ particles. However, radiative matrix elements involving "spin flips" are negligible and, furthermore, the matrix elements without spin flip contain spatial wave functions which are solutions of a Klein-Gordon-type equation with a spin-orbit term of relative magnitude 10^{-5} to 10^{-6} for channeled electrons of a few mega-electron-volts [13]. One may therefore apply the much simpler Klein-Gordon equation, for a spinless particle of mass *m* and charge -e, to obtain stationary states Ψ corresponding to energy *E* of the total system,

$$\left[\left[-i\hbar\nabla_{\mathbf{R}} + \frac{e}{c}\mathbf{A}(\mathbf{R},\ldots)\right]^{2}c^{2} + m^{2}c^{4}\right]\Psi(\mathbf{R},\ldots)$$
$$= \left[E - V(\mathbf{R},\ldots) - H_{l} - H_{r}\right]^{2}\Psi(\mathbf{R},\ldots) . \quad (2.1)$$

Here, H_l and H_r denote Hamiltonians of the lattice and the radiation field which interact with the projectile through the crystal potential $V(\mathbf{R}, ...)$ and the vector potential $\mathbf{A}(\mathbf{R}, ...)$. Only the projectile coordinates \mathbf{R} are shown explicitly.

At high energies, scattering angles are small and the momentum transfers to the projectile are mainly transverse. Accordingly, we separate a factor $\exp(iKz)$ from the wave function [8],

$$\Psi(\mathbf{R},\ldots)=e^{iKz}w(\mathbf{r},z,\ldots), \quad \mathbf{R}=(\mathbf{r},z) , \qquad (2.2a)$$

with

$$E^2 = \hbar^2 K^2 c^2 + m^2 c^4 . ag{2.2b}$$

In the equation for w, obtained by inserting (2.2) into (2.1) and dividing by $2E = 2m\gamma c^2$, we neglect terms $(\hbar^2 c^2/2E)\partial^2/\partial z^2$ and $(V+H_l+H_r)^2/2E$. These combine to give radiation-energy corrections of which the most important may be interpreted as a Doppler correction, accounting for a finite angle between the directions of the projectile and of the emitted photon [7]. This correction becomes important only at very high projectile energies ($E \gtrsim 1$ GeV) where a quantal treatment of channeling is no longer required.

In the Coulomb gauge $(\nabla \cdot \mathbf{A}=0)$, which will be used throughout, we then obtain

$$i\hbar\frac{\partial}{\partial t}w(\mathbf{r},t,\ldots) = \left[-\frac{\hbar^2}{2m\gamma}\nabla_{\mathbf{r}}^2 + V(\mathbf{r},t,\ldots) + H_l + H_r + H_{e,r}\right]w , \qquad (2.3)$$

$$H_{e,r} = \frac{e}{m \gamma c} \mathbf{A}(\mathbf{R},\ldots) \cdot \mathbf{P} + \beta e A_z(\mathbf{R},\ldots), \quad \mathbf{P} = -i \hbar \nabla_{\mathbf{R}} .$$

The depth parameter z has here been replaced by a "time" parameter t = z/v, where $v = \beta c$ is given by the relation $\hbar K = m \gamma v$, and the resulting equation for w has the form of a time-dependent Schrödinger equation.

The coupling of the projectile coordinates to the degrees of freedom of the radiation field is contained in $H_{e,r}$. We are interested here in spontaneous, radiative *transitions* induced by $H_{e,r}$, and hence contributions of order A^2 to this operator have been neglected.

In a precise description, the coupling between the crystal and the radiation field is included in the Hamiltonian operator for the lattice, H_l . This coupling is treated in Appendix A where it is shown that considerations of the induced shifts lead to the formulas for photon refraction. For these studies, the A^2 terms are essential, resulting in a first-order energy shift which gives the main contribution to the refractive index for photon frequencies far from atomic resonances. Note also that magnetic excitations are a result of scattering processes induced by a combination of $H_{e,r}$ and the coupling between lattice and radiation field [7]. Here we disregard this coupling, except for the correction for refraction introduced in Sec. II B.

The coupling of the projectile to the radiation field, $H_{e,r}$, may be treated as a perturbation which to zeroth or-

der is neglected. The solutions of (2.3) are then products of stationary photon states and solutions ψ of

$$i\hbar\frac{\partial}{\partial t}\psi(\mathbf{r},t,\ldots) = \left[-\frac{\hbar^2}{2m\gamma}\nabla_{\mathbf{r}}^2 + V(\mathbf{r},t,\ldots) + H_l\right]\psi .$$
(2.4)

Also the coupling in (2.4) of the particle to the lattice degrees of freedom may be treated as a perturbation and to zeroth order, we replace the potential V by the thermally averaged lattice potential $V_c^T(\mathbf{r},z)$ which is the expectation value of $V(\mathbf{r}, z, ...)$ in the ground state for the target electrons and in the vibrational states of the lattice, weighted by Boltzmann factors. When the average of Vin the electronic ground state is approximated by a sum of atomic potentials, centered on nuclei displaced from their equilibrium positions, the average over vibrational states corresponds to a convolution of each atomic potential with the thermal distribution function for atomic displacements. Within the harmonic model for the crystal, the convolution involves a Gaussian distribution function and corresponds in reciprocal space to multiplication of the Fourier components of the atomic potential by Debye-Waller factors.

A. Continuum model

The final simplification is obtained when $V_c^T(\mathbf{r}, z)$ is replaced by the continuum potential which, for an axis in the z direction, is given by

$$U_T(\mathbf{r}) = \frac{1}{d} \int_0^d dz \ V_c^T(\mathbf{r}, z) \ , \qquad (2.5)$$

where d denotes the lattice period along the axis. The solutions of (2.4) may in this case be chosen as product functions,

$$\psi^{c}(\mathbf{r},t,\ldots) = u(\mathbf{r})|\chi_{l}\rangle \exp(-i\varepsilon t/\hbar) , \qquad (2.6)$$

where the lattice part $|\chi_l\rangle$ is an eigenstate of the Hamiltonian H_l . The energy ε is a sum of the eigenvalue of this operator and the energy E_{\perp} of the transverse motion of the projectile which is described by a stationary Schrödinger equation,

$$\left[-\frac{\hbar^2}{2m\gamma}\nabla_{\mathbf{r}}^2 + U_T(\mathbf{r})\right] u(\mathbf{r}) = E_{\perp}u(\mathbf{r}) . \qquad (2.7)$$

Note that the energy E enters (2.7) through the relativistic mass $m\gamma$ of the projectile. The potential U_T has the translational symmetry of a two-dimensional Bravais lattice, and accordingly, the solutions $u(\mathbf{r})$ may be chosen as Bloch waves

$$u_{n,\mathbf{k}}(\mathbf{r}) = \frac{1}{(L_x L_y)^{1/2}} e^{i\mathbf{k}\cdot\mathbf{r}} \sum_{\alpha,\beta} C^n_{\alpha,\beta}(\mathbf{k}) e^{i(\alpha \mathbf{g}_1 + \beta \mathbf{g}_2)\cdot\mathbf{r}} , \qquad (2.8)$$

where \mathbf{g}_1 and \mathbf{g}_2 are primitive basis vectors for the twodimensional reciprocal lattice, the wave vector \mathbf{k} is chosen in the first Brillouin zone, n is a band index, and $L_x L_y$ a quantization area. The corresponding Fourier expansion of the potential is given by



FIG. 1. Bloch bands for planar channeling of 4-MeV electrons along the (100) plane of nickel at 500 K. Also shown is the continuum potential. The values indicated on the x axis are in units of the basis vector for the one-dimensional reciprocal lattice and of the distance between planes, respectively.

$$U_T(\mathbf{r}) = \sum_{\alpha,\beta} U_T(\alpha,\beta) e^{i(\alpha \mathbf{g}_1 + \beta \mathbf{g}_2) \cdot \mathbf{r}} , \qquad (2.9)$$

where the coefficients $U_T(\alpha,\beta)$ are Fourier components of the thermally averaged lattice potential, corresponding to wave vectors perpendicular to the axis. When the average of the lattice potential in the electronic ground state is approximated by a sum of atomic potentials, the crystalline Fourier components may be expressed in terms of the components $V_{aT}(k)$ for the thermally averaged atomic potential. In the simplest case, when all atoms contribute in phase, the relation is

$$V_c^T(\mathbf{k}) = N_d V_{aT}(k) . (2.10)$$

Here, N_d is the density of atoms and $V_{aT}(k)$ an atomic Fourier component, $V_a(k)$, multiplied by the Debye-Waller factor $\exp(-k^2\rho^2/2)$, where ρ is the onedimensional root-mean-square vibrational amplitude.

In the planar case, the continuum potential V_T depends on one coordinate only,

$$V_T(x) = \frac{1}{d_s} \int_0^{d_s} dy \ U_T(\mathbf{r}) , \qquad (2.11)$$

where d_s is the spacing of axes in the plane. The transverse wave function $u(\mathbf{r})$ separates into a product of a one-dimensional Bloch wave, which describes the motion perpendicular to the plane, and a plane wave in the y direction parallel to the plane. The Bloch bands for the one-dimensional "crystal" are shown in Fig. 1. (Note

that the z momentum of the projectile is given by $(\hbar K - \epsilon/v)$ [see (2.2a) and (2.6)]: the larger the transverse energy, the smaller the z momentum. Electron microscopists traditionally consider z momenta instead of transverse energies, and accordingly they obtain diagrams as Fig. 1 turned upside down.)

According to this description, the solutions (2.6) will be indexed as ψ_{nkl}^c , which is convenient in Sec. III where non-continuum-model solutions of (2.4) are expanded in the basis set of functions (2.6).

B. Channeling spectroscopy

We now consider radiative transitions between solutions of (2.4) with a continuum potential. The transition probabilities may be obtained from a standard perturbation treatment of the coupling term $H_{e,r}$ in (2.3). Since, for t=0, the transverse state of the particle is a plane wave, and the radiation field is in the ground state $|0\rangle$, the first-order amplitude for finding the particle (and the lattice) at time t=T in a state $\psi_{f}^{c}(t=T)$ together with a photon of momentum $\hbar\kappa$ and energy $\hbar\omega$ is given by

$$a_{f,\kappa}^{(1)}(t=T) = \frac{1}{i\hbar} \int_0^T dt \left\langle \psi_f^c(t), \kappa \exp(-i\omega t) \middle| e \left[\beta A_z + \frac{1}{m\gamma c} \mathbf{A} \cdot \mathbf{P} \right] \middle| \sum_n A_n \psi_{n\mathbf{k}l_i}^c(t), 0 \right\rangle , \qquad (2.12)$$

where A_n is the overlap of the initial plane wave $|\mathbf{k}\rangle$ with the Bloch state $u_{n,k}$, and $|\chi_{l_i}\rangle$ is the initial lattice state.

We shall consider emission in the forward (z) direction. The photon polarization vector ε_{μ} is then in the transverse (xy) plane, and the relevant matrix element of the radiation field, quantized within a volume L_0^3 , is given by [14]

$$\langle \mathbf{\kappa} | \mathbf{A} | 0 \rangle = \left[\frac{2\pi c^2 \tilde{\mathbf{\hbar}}}{L_0^3} \right]^{1/2} \frac{1}{\sqrt{\omega}} \boldsymbol{\varepsilon}_{\mu} e^{-i\kappa z} .$$
(2.13)

The final state ψ_f^c is chosen from the basis set of stationary solutions $\psi_{n'\mathbf{k}'l}^c$ of which only the functions $\psi_f^c = \psi_{n'\mathbf{k}l_i}^c$ contribute to the radiation probability. According to the definition of the pseudotime *t*, the *z* parameter of (2.13) may be written as z = vt. Introducing the photon energy-momentum relation

 $\omega = \kappa c / n_r , \qquad (2.14)$

where the index of refraction n_r has been included in order to take into account the change of the excitation quantum $\hbar\omega$ of the radiative mode κ due to the interaction between the radiation field and the lattice (electrons), we see that the *n*th term of expression (2.12) for the emission amplitude equals

$$\frac{1}{i\hbar} \frac{e}{m\gamma c} \left[\frac{2\pi c^2 \hbar}{L_0^3} \right]^{1/2} \frac{1}{\sqrt{\omega}} \langle u_{n',\mathbf{k}} | \boldsymbol{\varepsilon}_{\mu} \cdot \mathbf{p} | u_{n,\mathbf{k}} \rangle A_n \int_0^T dt \exp[i(E_{\perp_{n',\mathbf{k}}} - E_{\perp_{n,\mathbf{k}}})t/\hbar] \exp(i\omega t) \exp(-i\omega n_r \beta t) .$$
(2.15)

Here, the transverse momentum **p** is defined by $\mathbf{p} = (p_x, p_y) = -i\hbar\nabla_r$. The absolute square of (2.15) is proportional to

$$T^{2} \frac{\sin^{2} x}{x^{2}}$$

with $x = \frac{T}{2} \{ \omega (1 - n_{r}\beta) - (E_{\perp_{n,k}} - E_{\perp_{n',k}}) / \hbar \}$, (2.16)

where the peak value is obtained for x=0. This determines the photon energy in terms of the transverseenergy difference,

$$\hbar\omega = \frac{1}{1 - n_r \beta} (E_{\perp_{n,\mathbf{k}}} - E_{\perp_{n',\mathbf{k}}})$$

$$\simeq (1 + \beta) \gamma^2 \frac{E_{\perp_{n,\mathbf{k}}} - E_{\perp_{n',\mathbf{k}}}}{1 + (1 + \beta) \gamma^2 \delta} , \qquad (2.17)$$

where $n_r \beta \simeq 1$ and $\delta \equiv 1 - n_r$ have been used. Photon energies, corresponding to optical transitions in the transverse potential, are pushed into the keV region (for MeV electrons) by the $2\gamma^2$ factor. This is a result of photon recoil which must be included due to the relativistic longitudinal projectile motion.

The same effect explains the presence of the index of refraction in the channeling frequency condition. For precision channeling spectroscopy, the correction for refraction is important, amounting at low radiation energies ($\sim 2 \text{ keV}$) to a few percent. As shown in Appendix A, the effect of refraction is part of the formalism developed, being due to the interaction between the radiation field and the target electrons. This interaction also leads to x-ray absorption which contributes to the linewidth of channeling radiation through broadening of photon states. However, the absorption lengths are typically longer by two orders of magnitude than decay lengths for channeling states, and hence this contribution is negligible.

The linewidth, W, is then in the continuum model given by the reciprocal emission time,

$$W \simeq \frac{1}{1 - n_r \beta} \frac{2\pi}{T} , \qquad (2.18)$$

in analogy to the line broadening encountered for colliding atoms in a gas (pressure broadening). For large T, the radiation spectrum consists of a series of sharp lines, and each line corresponds to a unique transition $(n,\mathbf{k}) \rightarrow (n',\mathbf{k})$ when interference between different terms

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can be neglected.

In the following sections, we shall for simplicity set the refractive index equal to one. The channeling spectrum will be investigated in the neighborhood of lines corresponding to transitions between low-lying, very narrow Bloch bands; throughout we shall use the $1 \rightarrow 0$ line as a specific example. With the definition $\hbar\omega_{10} = E_{\perp_1} - E_{\perp_0}$, the amplitude for emission into photon mode κ in the forward direction and within the $1 \rightarrow 0$ line may be written

$$a_{f,\kappa_{1\to0}}^{(1)}(t=T) = \frac{1}{i\hbar} \frac{e}{m\gamma c} \left[\frac{2\pi c^2\hbar}{L_0^3} \right]^{1/2} \frac{1}{\sqrt{\omega}} \langle u_0 | \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{p} | u_1 \rangle \\ \times \int_0^T dt \exp\{i[\omega(1-\beta) - \omega_{10}]t\} A_1;$$
(2.19)

 $f = (0\mathbf{k}l_i)$ is the only final state contributing. Note that for low-lying bands, the matrix element in (2.19) is independent of Bloch vector \mathbf{k} , and the functions u_0 and u_1 may be chosen as localized states, bound to a single string or plane.

To obtain the number of photons emitted during passage of a crystal of thickness L = Tv within the $1 \rightarrow 0$ line and within a solid angle $d\Omega$ around the forward direction, we multiply the absolute square of (2.19) by a photon phase-space factor and integrate over frequency with the result

$$dN_{10} = 2(1+\beta)\alpha\omega_{1\to 0}T(mc)^{-2}|\langle u_0|\epsilon_{\mu}\cdot\mathbf{p}|u_1\rangle|^2$$
$$\times |A_1|^2 \frac{d\Omega}{4\pi}, \qquad (2.20)$$

where $\omega_{1\to 0}$ is given by (2.17) with n = 1 and n' = 0, α is the fine-structure constant, and $|A_1|^2$ is the probability at the front face for populating the first Bloch band.

III. LINE PROFILES

When the coupling between projectile and lattice coordinates is included, the states ψ^c in (2.12), which are given by (2.6), must be replaced by solutions ψ of the timedependent Schrödinger equation (2.4) with the full lattice potential $V(\mathbf{r}, t, ...)$. Considering at first axial channeling, we express the potential V as a sum of the continuum potential U_T and of various scattering potentials to be treated by perturbation theory,

$$V = U_T + \delta V_1 + \delta V_2 + \delta V_3 . \tag{3.1}$$

The crystal potential may be approximated by a sum of atomic potentials V_a , and the different terms in (3.1) may then be written as

$$\delta V_{1} = \sum_{i} \left[\left[\frac{-Ze^{2}}{|\mathbf{R} - \mathbf{R}_{i} - \mathbf{U}_{i}|} + \sum_{j=1}^{Z} \frac{e^{2}}{|\mathbf{R} - \mathbf{R}_{i,j}|} \right] - V_{a}(\mathbf{R} - \mathbf{R}_{i} - \mathbf{U}_{i}) \right],$$

$$\delta V_{2} = \sum_{i} \left[V_{a}(\mathbf{R} - \mathbf{R}_{i} - \mathbf{U}_{i}) - V_{a,T}(\mathbf{R} - \mathbf{R}_{i}) \right],$$

$$\delta V_{3} = \sum_{i} V_{a,T}(\mathbf{R} - \mathbf{R}_{i}) - U_{T}(\mathbf{r}),$$

(3.2)

where Ze is the nuclear charge, U_i is the displacement vector for the nucleus with equilibrium position \mathbf{R}_i , and $\mathbf{R}_{i,j}$ is the coordinate vector of the *j*th electron in the *i*th atom. In the following, excitations of lattice electrons will be neglected. In (3.2) only the first perturbation, δV_1 , couples to excited states of the target electrons, and since the expectation value of δV_1 is zero in the electronic ground state, this term does not give scattering without electronic excitation.

The second perturbation, δV_2 , gives rise to "thermal diffuse scattering," mostly with phonon emission or absorption. In contrast, the last term, δV_3 , does not couple to vibrational coordinates, and since it is periodic, scattering is confined to reciprocal-lattice vectors. In diffraction theory, the technical term is HOLZ (higher-order Laue zone) scattering, referring to nonzero momentum transfer parallel to the axis [15].

For planar channeling with incidence at a fairly small angle to a strong axis in the plane, it is natural to express the planar continuum potential as an average of axial potentials, as in (2.11), and U_T in (3.1) is split into

$$U_T(\mathbf{r}) = V_T(x) + \delta V_4 . \qquad (3.3)$$

The potential $V_T(x)$ contains only "systematic" scattering corresponding to reciprocal-lattice vectors perpendicular to the plane while δV_4 introduces the "nonsystematic" reflections with momentum transfers perpendicular to the axis.

The influence of the individual scattering components on the channeling-radiation spectrum will be treated on the basis of the general formalism developed in the following sections.

A. Radiation amplitudes

We consider radiative transitions between solutions of (2.4) where the potential V is a sum of the axial (planar) continuum potential and the perturbation $\delta V = \delta V_1 + \delta V_2 + \delta V_3 (+ \delta V_4)$. The first-order amplitude for finding the particle and the lattice at time t = T in a state $\psi_f(t=T)$ together with a photon of momentum $\hbar \kappa$ and energy $\hbar \omega$ is given by [compare with (2.12)]

$$a_{f,\kappa}^{(1)}(t=T) = \frac{1}{i\hbar} \int_0^T dt \left\langle \psi_f(t), \kappa \exp(-i\omega t) \middle| e \left[\beta A_z + \frac{1}{m\gamma c} \mathbf{A} \cdot \mathbf{P} \right] \middle| \psi_i(t), 0 \right\rangle , \qquad (3.4)$$

where ψ_i is the solution of (2.4), which at the front face matches the incoming plane wave and the initial lattice state, and where ψ_f is picked out from a basis set specified at the front surface.

Expanding the wave functions ψ_i and ψ_f in terms of the stationary solutions ψ_{nkl}^c of the continuum model, we obtain for emission in the forward direction

$$a_{f,\kappa}^{(1)}(t=T) = \frac{1}{i\hbar} \frac{e}{m\gamma c} \left[\frac{2\pi c^2 \hbar}{L_0^3} \right]^{1/2} \frac{1}{\sqrt{\omega}} \sum_{n,n'} \sum_{\mathbf{k},l} \langle u_{n',\mathbf{k}}(\mathbf{r}) | \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{p} | u_{n,\mathbf{k}}(\mathbf{r}) \rangle \int_0^T dt \exp\{i[\omega(1-\beta) + \Delta E_{\perp}/\hbar]t\} \langle \psi_f(t) | \psi_{n'\mathbf{k}l}(t) \rangle \times \langle \psi_{n\mathbf{k}l}^c(t) | \psi_i(t) \rangle , \qquad (3.5)$$

with

$$\Delta E_{\perp} = E_{\perp_{n',\mathbf{k}}} - E_{\perp_{n,\mathbf{k}}}$$
(3.6)

We shall assume that for ω in the neighborhood of, say, the $1 \rightarrow 0$ line, the time integration in (3.5) eliminates all terms except those with n'=0 and n=1. This requires the separation between lines from different transitions to be large compared to the linewidths. We then obtain for the contribution from the state ψ_f to the $1 \rightarrow 0$ -line amplitude

$$a_{f,\kappa_{1\to0}}^{(1)}(t=T) = \frac{1}{i\hbar} \frac{e}{m\gamma c} \left[\frac{2\pi c^2 \hbar}{L_0^3} \right]^{1/2} \frac{1}{\sqrt{\omega}} \langle u_0 | \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{p} | u_1 \rangle \int_0^T dt \exp\{i[\omega(1-\beta) - \omega_{10}]t\} \\ \times \left[\sum_{\mathbf{k},l} \langle \psi_f(t) | \psi_{0\mathbf{k}l}^c(t) \rangle \langle \psi_{1\mathbf{k}l}^c(t) | \psi_i(t) \rangle \right].$$
(3.7)

Here, we have used the fact that for low-lying, narrow bands, the transition frequency ω_{10} and the dipole matrix element $\langle u_0 | \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{p} | u_1 \rangle$ are independent of the summation variables \mathbf{k}, l .

B. Projection-operator formalism

The influence of the perturbation δV is contained in the last factor of the time integrand, the sum over \mathbf{k}, l . To study the complicated time development of this sum, it is convenient to introduce a more compact notation. We shall not discuss the electronic excitations induced by the perturbation δV_1 in (3.2), and hence we may ignore the electronic degrees of freedom. The atomic variables, ..., \mathbf{U}_i , ..., are denoted by \mathbf{U} , and the quantum numbers \mathbf{k}, l are combined into one symbol q. Furthermore, we may reduce the product of two matrix elements into a single one by introducing a function space Ω , spanned by product eigenstates,

$$\Psi_{n_{1}q_{1},n_{2}q_{2}}^{c}(\mathbf{r}_{1},\mathbf{U}_{1},\mathbf{r}_{2},\mathbf{U}_{2},t)$$

$$\equiv \psi_{n_{1}q_{1}}^{c*}(\mathbf{r}_{1},\mathbf{U}_{1},t)\psi_{n_{2}q_{2}}^{c}(\mathbf{r}_{2},\mathbf{U}_{2},t) . \quad (3.8)$$

The initial and final states are then combined into one function,

$$\psi_{fi}(\mathbf{r}_1, \mathbf{U}_1, \mathbf{r}_2, \mathbf{U}_2, t) = \psi_f^*(\mathbf{r}_1, \mathbf{U}_1, t) \psi_i(\mathbf{r}_2, \mathbf{U}_2, t) , \quad (3.9)$$

and the sum in (3.7) may be expressed in terms of the projection of this wave function onto a subspace Ω_{01} defined by

$$\Omega_{01} = \operatorname{span}_{q} \{ \Psi_{0q,1q}^{c}(t) \} .$$
 (3.10a)

The corresponding projection operator P_{01} is given by

$$P_{01}\Psi(t) = \sum_{q} \langle \Psi_{0q,1q}^{c}(t) | \Psi(t) \rangle \Psi_{0q,1q}^{c}(t) , \qquad (3.10b)$$

and the sum in (3.7) takes the form

$$\sum_{q} \langle \psi_{f}(t) | \psi_{0q}^{c}(t) \rangle \langle \psi_{1q}^{c}(t) | \psi_{i}(t) \rangle$$
$$= \sum_{q} \langle \Psi_{0q,1q}^{c}(t) | P_{01} \Psi_{fi}(t) \rangle . \quad (3.11)$$

The wave function Ψ_{fi} satisfies the time-dependent Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\Psi_{fi} = \left[\left[\frac{-\hbar^2}{2m\gamma} \Delta_{\mathbf{r}_2} + U_T(\mathbf{r}_2) + H_I(\mathbf{U}_2) \right] - \left[\frac{-\hbar^2}{2m\gamma} \Delta_{\mathbf{r}_1} + U_T(\mathbf{r}_1) + H_I(\mathbf{U}_1) \right] + \left\{ \delta V(\mathbf{r}_2, t, \mathbf{U}_2) - \delta V(\mathbf{r}_1, t, \mathbf{U}_1) \right\} \right] \Psi_{fi}(t)$$

$$\equiv H_{12}(t)\Psi_{fi}(t) , \qquad (3.12a)$$

and the stationary basis states $\Psi_{n_1q_1,n_2q_2}^c$ are solutions of the corresponding zeroth-order Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\Psi^{c} = H_{12}^{c}\Psi^{c}, \qquad (3.12b)$$

with

$$H_{12}(t) \equiv H_{12}^{c} + \{\delta V(\mathbf{r}_{2}, t, \mathbf{U}_{2}) - \delta V(\mathbf{r}_{1}, t, \mathbf{U}_{1})\} . \qquad (3.12c)$$

The effects of the perturbation $\{\delta V(\mathbf{r}_2, t, \mathbf{U}_2) - \delta V(\mathbf{r}_1, t, \mathbf{U}_1)\}$ in (3.12c) on the expression (3.11) may qualitatively be split into three categories: (1) Reduction of the projection of Ψ_{fi} onto Ω_{01} by scattering out of the states 0 and 1, (2) change in phase of $P_{01}\Psi_{fi}$, and (3) increase of the projection of Ψ_{fi} onto Ω_{01} by feeding of the states 0 and 1. The first two effects contribute line broadening and line shifts and are our main concern here,

while the third effect only influences the line intensity.

To make this separation explicit, we introduce a division of the time T for passage through the crystal into finite time intervals of length Δt and centered on times t_n , n = 1, 2, ..., N. The change of Ψ_{fi} over one interval is determined by a propagator $D(t_n)$,

$$\Psi_{fi}(t_{n+1}) = D(t_n)\Psi_{fi}(t_n) , \qquad (3.13a)$$

which may be expressed in terms of a product of differential propagators

$$\delta(t) = \left[1 + \frac{dt}{i\hbar} H_{12}(t) \right] . \tag{3.13b}$$

The time integration in (3.7) may be replaced by a summation over the finite number of intervals, provided that Δt is small on the time scale for scattering into or out of Ω_{01} . On the other hand, Δt is chosen large enough that

contributions to the radiation intensity from different intervals add incoherently. We return to this critical point later.

The component of Ψ_{fi} , which contributes to the radiation amplitude in the *n*th time interval, consists partly of the component contributing in the preceding interval, depleted by scattering out of Ω_{01} , partly of a term originating from feeding in from outside the space Ω_{01} . This is expressed compactly by the following equation:

$$P_{01}\Psi_{fi}(t_n) = P_{01}D(t_{n-1})P_{01}\Psi_{fi}(t_{n-1}) + P_{01}D(t_{n-1})(1-P_{01})\Psi_{fi}(t_{n-1}) , \qquad (3.14)$$

which follows from the definition (3.13a) and linearity of the operator $D(t_{n-1})$.

Using the iterative equation (3.14), we may show by induction that

$$\int_{0}^{1} dt \exp\{i[\omega(1-\beta)-\omega_{10}]t\} \sum_{q} \langle \Psi_{0q,1q}^{c}(t)|P_{01}\Psi_{fi}(t)\rangle$$

$$\approx \sum_{n=1}^{N} \sum_{m\geq n}^{N} \Delta t \exp\{i[\omega(1-\beta)-\omega_{10}]t_{m}\}$$

$$\times \sum_{q} \langle \Psi_{0q,1q}^{c}(t_{m})|P_{01}D(t_{m}-\Delta t)P_{01}\cdots P_{01}D(t_{n})P_{01}D(t_{n-1})(1-P_{01})\Psi_{fi}(t_{n-1})\rangle . \quad (3.15)$$

For compactness, we have introduced here the notation $D(t_0)(1-P_{01})\Psi_{fi}(t_0) \equiv \Psi_{fi}(t_1)$ for the special term n = 1. The terms n > 1 contribute to the integration from the time interval around t_n , starting with a feeding-in (or feeding-back) event in the preceding time interval, which is followed by an "unbroken chain" $\cdots P_{01}DP_{01}DP_{01}\cdots$. This chain takes into account the depletion and the phase shift of the original projection onto Ω_{01} (at $t = t_n$), and we may write

$$\left\langle \sum_{q} \Psi_{0q,1q}^{c}(t_{m} + \Delta t) | P_{01} D(t_{m}) P_{01} D(t_{m} - \Delta t) P_{01} \cdots \right\rangle = e^{-i\Delta\omega_{10}\Delta t} e^{-\Delta t/2\tau_{10}} \left\langle \sum_{q} \Psi_{0q,1q}^{c}(t_{m}) | P_{01} D(t_{m} - \Delta t) P_{01} \cdots \right\rangle, \quad \Delta t/\tau_{10} \ll 1 . \quad (3.16)$$

The decay constant $1/2\tau_{10}$ is not a simple sum of separate decay constants for the two states 1 and 0 since a transition in either state to another set of quantum numbers q does not lead to depletion of the matrix element if the same q transition takes place in the other state. Thus there is a need for treating the decay constant (and the phase shift $\Delta \omega_{10}$) in a coupled manner and for this purpose, the product space is well suited. Be-

sides, the projection operator P_{01} is a convenient tool for writing down very compactly the different contributions to the radiation amplitude, with an easy interpretation of each term.

The contribution from the state ψ_f to the amplitude for forward emission within the $1 \rightarrow 0$ line we obtain using (3.7), (3.11), (3.15), and (3.16),

$$a_{f,\kappa_{1\to0}}^{(1)}(t=T) \approx \frac{1}{i\hbar} \frac{e}{m\gamma c} \left[\frac{2\pi c^{2}\hbar}{L_{0}^{3}} \right]^{1/2} \frac{1}{\sqrt{\omega}} \langle u_{0} | \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{p} | u_{1} \rangle \\ \times \sum_{n} \beta_{n} \int_{t_{n}}^{T} dt \exp\{i[\omega(1-\beta) - \omega_{10}]t\} \exp[-i\Delta\omega_{10}(t-t_{n})] \exp[-(t-t_{n})/2\tau_{10}], \qquad (3.17a)$$

where

$$\beta_n = \sum_{q} \left\langle \Psi_{0q,1q}^c(t_n) | P_{01} D(t_{n-1}) (1 - P_{01}) \Psi_{fi}(t_{n-1}) \right\rangle \,.$$

(3.17b)

Compare (3.17a) with (2.19) of the continuum model: The loss of stability of transverse states with introduction of the scattering potential δV is reflected in the replacement in (3.17a) of A_1 , the population amplitude at the front face of the right initial and final states, by a summation over all depths $t = t_n$ of the crystal, weighted by the population amplitudes β_n . Furthermore, in the continuum model, the coherent emission time extends over the full crystal thickness T but in (3.17a), the instability introduces an effective emission-time limit of order τ_{10} .

C. General remarks on line profiles and line intensities

We may calculate the line profile of well-separated lines in the radiation spectrum, obtained for emission in the forward direction, by multiplying the absolute square of (3.17a) by a phase-space factor. The result is

$$\frac{d^{2}}{d\omega d\Omega} N_{10}(\omega)_{f} \approx \frac{\alpha}{4\pi^{2} (m\gamma c)^{2}} |\langle u_{0}|\epsilon_{\mu} \cdot \mathbf{p}|u_{1}\rangle|^{2} \\ \times \omega \left|\sum_{n} \beta_{n} e^{i[\omega(1-\beta)-\omega_{10}]t_{n}}\right|^{2} \mathcal{L}(\omega) .$$
(3.18)

 $\mathcal{L}(\omega)$ denotes the Lorentzian profile,

$$\mathcal{L}(\omega) = \frac{1}{[\omega_{10} + \Delta\omega_{10} - \omega(1 - \beta)]^2 + 1/4\tau_{10}^2} , \qquad (3.19)$$

which is centered on the shifted frequency

$$\omega_{1\to 0} = \frac{\omega_{10} + \Delta\omega_{10}}{1-\beta} \tag{3.20a}$$

and has a full width at half maximum (FWHM) W given by

$$W = \frac{1}{1 - \beta} \frac{1}{\tau_{10}} . \tag{3.20b}$$

To obtain the separation between contributions to the line profile and the line intensity, the interval Δt must be chosen large enough that interference terms vanish in (3.18). In that case the total line profile is the same as the (skew) Lorentzian profile obtained individually for each term n. The feeding in during an interval Δt contributes only to the line intensity.

Here, we shall not attempt a complete analysis of the interference between terms in (3.18) [1], which correspond to feeding at different depths, but we may give two arguments for incoherence of these terms. First, particle propagation in different channeling eigenstates gives rise to phase differences. When linewidths are small as compared to line separations, the interval Δt may be chosen long enough for such phase differences to be large. Second, lattice excitations associated with the feeding are fairly localized, and hence feeding at different depths will normally be associated with orthogonal lattice states. For thermal scattering, coherence will be limited to the correlation length for thermal vibrations, which is typically of the order of a few lattice spacings only [16].

The center frequency and width of the Lorentzian profile are given by the shift $\Delta \omega_{10}$ and decay constant $1/2\tau_{10}$, respectively, and these quantities will be studied in the following section.

A few comments will be given on the intensity of the $1 \rightarrow 0$ line. The contribution from the final state ψ_f is obtained by an integration of (3.18) over photon frequencies

$$dN_{10}^{f} = \int d\omega \frac{d^{2}}{d\omega d\Omega} N_{10}^{f}(\omega) d\Omega$$

$$\approx \cdots \int d\omega \, \omega \mathcal{L}(\omega)$$

$$= 2(1+\beta)\alpha \omega_{1\to 0} \tau_{10} (mc)^{-2} |\langle u_{0}| \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{p} | u_{1} \rangle|^{2}$$

$$\times \sum_{n} |\beta_{n}|^{2} \frac{d\Omega}{4\pi} . \qquad (3.21)$$

Note the strong similarity between (3.21) and (2.20) of the continuum model. Again the instability of the transverse states leads to a replacement of T, the time for passage through the crystal, by the effective emission time, of order τ_{10} , and to a replacement of $|A_1|^2$, the probability for population of the right initial and final states at the front face, by the sum over $|\beta_n|^2$ where β_n is the amplitude for populating the right states by feeding at depth $z = vt_n$.

The yield of the $1 \rightarrow 0$ line may also be found directly from (3.5) with n = 1 and n' = 0 by taking the absolute square, multiplying the result by a phase-space factor, and integrating over frequency. Starting out by performing the ω integration, we obtain a δ function in the two times occurring, and the result is

$$dN_{10}^{f} = 2(1+\beta)\alpha\omega_{1\to0}T(mc)^{-2}|\langle u_{0}|\epsilon_{\mu}\cdot\mathbf{p}|u_{1}\rangle|^{2}\frac{d\Omega}{4\pi}$$
$$\times \frac{1}{T}\int_{0}^{T}dt \left|\sum_{q}\langle \Psi_{0q,1q}^{c}(t)|\Psi_{fi}(t)\rangle|^{2}.$$
(3.22)

By summing over all final states ψ_f and applying closure, we obtain the total intensity in the $1 \rightarrow 0$ line,

$$dN_{10}^{\text{total}} = 2(1+\beta)\alpha\omega_{1\to 0}T(mc)^{-2}$$

$$\times |\langle u_0|\epsilon_{\mu} \cdot \mathbf{p}|u_1\rangle|^2 \frac{d\Omega}{4\pi}$$

$$\times \frac{1}{T} \int_0^T dt \sum_q |\langle \psi_{1q}^c(t)|\psi_i(t)\rangle|^2 . \qquad (3.23)$$

The population of the first Bloch band at the front face, $|A_1|^2$, which was encountered in the continuum-model expression (2.20) for the total intensity in the $1 \rightarrow 0$ line, has been replaced by the average over the entire crystal of the first-Bloch-band population.

IV. TREATMENT OF LINE SHIFTS AND LINEWIDTHS

According to Eqs. (3.18)-(3.20), the line profile is defined by the shift $\Delta \omega_{10}$ and the decay constant $1/2\tau_{10}$ which in turn are given by Eq. (3.16) as

)

$$\exp\left[\left[-i\Delta\omega_{10} - \frac{1}{2\tau_{10}}\right]\Delta t\right] = \frac{\sum_{q} \langle \Psi_{0q,1q}^{c}(t_{m} + \Delta t)|P_{01}D(t_{m})P_{01}\Psi\rangle}{\sum_{q} \langle \Psi_{0q,1q}^{c}(t_{m})|P_{01}\Psi\rangle} .$$
 (4.1)

In the present section, we obtain general expressions for the shift and decay constant of first, second, and third order in the scattering potential δV , and identify the scattering processes which contribute to the shift and broadening, respectively. In the following sections, specific expressions are obtained for the individual perturbations δV_2 , δV_3 , and δV_4 , defined in (3.2) and (3.3).

On the right-hand side of (4.1), $D(t_m)$ is written as a product of differential propagators. We expand both sides of (4.1) in orders of the perturbation $\{\delta V(\mathbf{r}_2, t, \mathbf{U}_2) - \delta V(\mathbf{r}_1, t, \mathbf{U}_1)\}$ and obtain with the definitions

$$\hbar\gamma_{10} \equiv \hbar\Delta\omega_{10} - i\frac{\hbar}{2\tau_{10}} \tag{4.2}$$

$$\rho_q \equiv \langle \Psi_{0q,1q}^c(t_m) | \Psi \rangle , \qquad (4.3)$$

to first order,

$$\hbar \gamma_{10}^{(1)} = \sum_{q} \frac{\rho_{q}}{\sum_{q} \rho_{q}} \frac{1}{\Delta t} \int_{t_{m}}^{t_{m+1}} dt \left(\langle 1q | \delta V(t) | 1q \rangle - \langle 0q | \delta V(t) | 0q \rangle \right), \quad (4.4)$$

where $|nq\rangle = \psi_{nq}^{c}(t=0)$. Note that $\gamma_{10}^{(1)}$ is real and that the first-order energy shift of the state $|nq\rangle$ is given simply by the expectation value of the perturbation δV averaged over the time interval Δt .

The second-order terms of (4.1) lead to

$$\begin{split} \hbar \gamma_{10}^{(2)} &= \sum_{q} \sum_{n_{2},q_{2} \neq 1,q} \frac{1}{\Delta t} \int_{t_{m}}^{t_{m}+1} dt \left\langle \psi_{1q}^{c}(t) | \delta V(t) | \psi_{n_{2}q_{2}}^{c}(t) \right\rangle \frac{1}{i\hbar} \int_{t_{m}}^{t} dt' \left\langle \psi_{n_{2}q_{2}}^{c}(t') | \delta V(t') | \psi_{1q}^{c}(t') \right\rangle \frac{\rho_{q}}{\left[\sum_{q} \rho_{q}\right]} \\ &+ \sum_{q} \sum_{n_{1},q_{1} \neq 0,q} \frac{1}{\Delta t} \int_{t_{m}}^{t_{m}+1} dt \left\langle \psi_{0q}^{c}(t) | \delta V(t) | \psi_{n_{1}q_{1}}^{c}(t) \right\rangle^{*} \frac{1}{i\hbar} \int_{t_{m}}^{t} dt' \left\langle \psi_{n_{1}q_{1}}^{c}(t') | \delta V(t') | \psi_{0q}^{c}(t') \right\rangle^{*} \frac{\rho_{q}}{\left[\sum_{q} \rho_{q}\right]} \\ &- \sum_{q} \sum_{q' \neq q} \frac{1}{\Delta t} \int_{t_{m}}^{t_{m}+1} dt \left\langle \psi_{1q'}^{c}(t) | \delta V(t) | \psi_{1q}^{c}(t) \right\rangle \frac{1}{i\hbar} \int_{t_{m}}^{t} dt' \left\langle \psi_{0q'}^{c}(t') | \delta V(t') | \psi_{0q}^{c}(t') \right\rangle^{*} \frac{\rho_{q}}{\left[\sum_{q} \rho_{q}\right]} \\ &- \sum_{q} \sum_{q' \neq q} \frac{1}{\Delta t} \int_{t_{m}}^{t_{m}+1} dt \left\langle \psi_{0q'}^{c}(t) | \delta V(t) | \psi_{0q}^{c}(t) \right\rangle^{*} \frac{1}{i\hbar} \int_{t_{m}}^{t} dt' \left\langle \psi_{0q'}^{c}(t') | \delta V(t') | \psi_{0q}^{c}(t') \right\rangle \frac{\rho_{q}}{\left[\sum_{q} \rho_{q}\right]} . \end{split}$$

$$\tag{4.5}$$

The various second-order terms are illustrated in Fig. 2, where the straight lines indicate Bloch bands and the wavy lines changes of Bloch vector and/or of vibrational state of the lattice. The horizontal axis represents time, and the time ordering of the interactions is indicated by the dots. The coupling terms correct for the fact that a

lattice excitation or change of Bloch vector is unimportant when it is common to the initial and final states.

The third-order terms are illustrated in Figs. 3(a) and 3(b). They fall into three groups of two, six, and eight terms, respectively. In the following formula, we give only the first term in each group:

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$$\times \frac{1}{i\hbar} \int_{t_m}^t dt' \langle \psi_{1q}^c(t') | \delta V(t') | \psi_{1q}^c(t') \rangle \\ \times \frac{1}{i\hbar} \int_{t_m}^{t'} dt'' \langle \psi_{n_2q_2}^c(t'') | \delta V(t'') | \psi_{1q}^c(t'') \rangle \frac{\rho_q}{\sum_q \rho_q} + \cdots$$
(4.6)

Within the first two groups, the terms differ only by time ordering or by interchange of the initial and final states together with a complex conjugation. The last eight terms all include a "transition" without change of state, i.e., an expectation value of the scattering potential. Therefore they should usually be negligible, and they are ignored in the following. Also the exclusion of initial states in the summations will be neglected. Thereby sum rules become applicable, and this allows for analytical calculations and closed expressions for the width and shift. Conversion of the time-ordered integrals in (4.5) and (4.6) into square and cube integrals facilitates identification of the scattering processes which contribute to the linewidth and line shift, respectively.

As an example, we shall consider a separate term of second order, the first term in (4.5). The imaginary part may be written directly in terms of the corresponding square integral and the real part as a square integral with the sign function included in the integrand. Neglecting the q average over initial states, we obtain for (minus) the imaginary part

$$\left[\frac{\hbar}{2\tau_1}\right]^{(2)} = \sum_{n_2, q_2 \neq 1, q} \frac{1}{2} \frac{1}{\hbar} \frac{1}{\Delta t} \int_{t_m}^{t_m + 1} dt \langle \psi_{1q}^c(t) | \delta V(t) | \psi_{n_2 q_2}^c(t) \rangle \int_{t_m}^{t_m + 1} dt' \langle \psi_{n_2 q_2}^c(t') | \delta V(t') | \psi_{1q}^c(t') \rangle , \qquad (4.7a)$$

and for the real part

$$(\hbar\Delta\omega_1)^{(2)} = \sum_{n_2, q_2 \neq 1, q} \frac{1}{2i} \frac{1}{\hbar} \frac{1}{\Delta t} \int_{t_m}^{t_m+1} dt \langle \psi_{1q}^c(t) | \delta V(t) | \psi_{n_2 q_2}^c(t) \rangle \int_{t_m}^{t_m+1} dt' \langle \psi_{n_2 q_2}^c(t') | \delta V(t') | \psi_{1q}^c(t') \rangle \operatorname{sgn}(t-t') .$$
(4.7b)

These expressions are the starting point for further expansions in the phonon case to be described in Sec. V.

For the physical interpretation of Eqs. (4.7), the sign function will be written in terms of its Fourier transform which is proportional to Cauchy's principal value. Since all time integrations of (4.7) extend over complete intervals, they may be written in terms of Fourier components of the scattering potential,

$$\hbar \gamma_1^{(2)} \equiv \hbar \Delta \omega_1^{(2)} - i \left[\frac{\hbar}{2\tau_1} \right]^{(2)}$$

$$= -\sum_{n_2, q_2 \neq 1, q} \frac{1}{\hbar} \frac{1}{\Delta t} \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega' \langle 1q | \delta V(-\omega') | n_2 q_2 \rangle \langle n_2 q_2 | \delta V(\omega') | 1q \rangle \left\{ \frac{P}{\omega' - \omega_{1n}} + i\pi \delta(\omega' - \omega_{1n}) \right\},$$
(4.8)

where $\hbar\omega_{1n}$ is the energy difference between the states $|1q\rangle$ and $|n_2q_2\rangle$, and an ω' integration with inclusion of Dirac's δ function has been introduced into the equation for $(\hbar/2\tau_1)^{(2)}$. As in (4.4), $|n_2q_2\rangle = \psi_{n_2q_2}^c$ (t=0).

>(2)

The δ function of (4.8) requires that scattering processes contributing to the decay constant obey the energyconservation law. In the expression for $(\hbar\Delta\omega_1)^{(2)}$, however, the δ function is replaced by the principal value, and the energy shift of a state is caused by scattering to intermediate states without conservation of energy. In the phonon case ($\delta V = \delta V_2$), the shift is due to scattering to intermediate, transverse electron states with simultaneous emission and absorption of virtual *phonons* and is thus fully analogous to the Lamb shift which is caused by coupling of the electron to the radiation field and is due to scattering to intermediate electron states with simultaneous emission and absorption of virtual *photons*.

Already at this point, we may comment upon certain characteristics of the second-order level shift which are revealed by the following more compact version of the real part of Eq. (4.8):

$$\hbar\Delta\omega_1^{(2)} = -\sum_{n_2,q_2\neq 1,q} \frac{1}{\hbar} \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega' f(\omega') \frac{P}{\omega' - \omega_{1n}} , \quad (4.9a)$$

with

$$f(\omega') \equiv \frac{1}{\Delta t} |\langle n_2 q_2 | \delta V(\omega') | 1q \rangle|^2 .$$
(4.9b)



FIG. 2. Second-order terms. Straight lines represent Bloch bands and wavy lines changes in Bloch vector and/or lattice state at the interaction points indicated by dots. The horizontal axis represents time. The first two drawings illustrate secondorder contributions obtained individually for the initial and final states of the radiation process. The last two drawings show the coupling terms. Emission and absorption of lattice excitations are treated equivalently, and emission of a lattice excitation in one state is followed by absorption of the same excitation in the other state.



FIG. 3. Third-order terms. Symbols as in Fig. 2. The first two drawings in (a) show the individual third-order corrections for the initial and final states. The next six drawings illustrate terms which take into account effects of coupled behavior in the two states. They differ only by time ordering of the scattering processes, and each term has a counterpart which is obtained by interchanging initial and final states together with a complex conjugation. In (b) are shown corrections to the terms in (a). They all involve a "transition" without a change of state.

The shift is obtained by folding a frequency-dependent function $f(\omega')$ with the "atomic" dispersion $1/(\omega'-\omega_{1n})$. This convolution is well known from the study of the ac-Stark shift which may in fact be calculated directly in the semiclassical case (i.e., the atom is treated quantum mechanically, whereas a classical description is applied for the external electromagnetic field) by use of the formalism developed above [1].

The sign of the shift (4.9) is in general dependent on how the spectrum is weighted relative to the atomic dispersion. For the cases to be considered, the "spectrum" $f(\omega')$ is symmetric around $\omega'=0$, and we have typically a form as shown in Fig. 4. A continuous distribution of frequencies ω' clearly results in a positive value for the ω' integration of (4.9a). This is valid for the phonon case. In the cases $\delta V = \delta V_3$ and $\delta V = \delta V_4$, the integral is replaced by a sum over discrete frequencies $\omega' \simeq n \cdot 2\pi c / d,$ $n=\pm 1,\pm 2,\ldots$ Since typically $\omega_{ni} \ll 2\pi c / d$, the magnitude of the negative terms is seen in Fig. 4 to be larger than the magnitude of the corresponding positive values, and the sum will be negative. Hence the sign of the second-order periodic shift will be opposite to the sign of the second-order phonon shift.

In recent years, there has been a lot of interest in studying changes of Lamb shifts for atoms in cavities, induced by the restriction of radiation modes: The smaller the cavity volume, the larger the distance between the allowed modes. By exactly the same kind of reasoning as used above, it may be shown that the sign of the Lamb shift changes at certain volume sizes [17].

As will turn out, the decay constant is dominated by contributions from the second-order terms, whereas the shift has similar contributions of second and third order. Hence only the real part of the third-order terms will be considered.

In contrast to the separate second-order shifts just described, the separate third-order shifts have contributions from both energy conserving and nonconserving processes.



FIG. 4. Atomic dispersion function centered at the resonance frequency ω_{1n} and typical spectral distribution $f(\omega')$. Calculation of the second-order shift involves a convolution integral between the two. The spectrum $f(\omega')$ is the absolute square of the Fourier expansion of the perturbation.

V. THERMAL SCATTERING

Based upon the general expressions of Sec. IV, explicit evaluations of the widths and shifts, induced by the phonon coupling operator δV_2 , will be performed in this section. The periodic perturbations $\delta V = \delta V_3$, δV_4 will be treated with similar methods in Sec. VI.

Introducing rough approximations in terms of unscreened Coulomb potentials, we obtain simple formulas which reveal explicitly the dependence on basic physical parameters. Since the shifts turn out to be very small, these simple estimates usually suffice. To check the accuracy of the approximations, we have also carried out a very precise analytical calculation with the Doyle-Turner approximation for atomic potentials [18]. The resulting formulas are given in Appendix B, while some numerical results for specific cases are given in Appendix C.

Introduction of second expansion parameter

In Sec. IV an expansion in the scattering-potential strength was performed. The dimensionless expansion parameter is of order $Z\alpha$, where Z is the atomic number of the crystal and α the fine-structure constant. To perform the summation over intermediate states analytically, we shall introduce a new expansion parameter $\lambda \equiv \Delta\omega \Delta t$, where $\hbar\Delta\omega$ is a typical transverse-energy transfer. Since typical momentum transfers are of order \hbar/ρ , we obtain

$$v/\Delta\omega \sim v \frac{2m\gamma\rho^2}{\hbar} \approx 2\gamma a_0 \alpha^{-1} \left[\frac{\rho}{a_0}\right]^2 \sim 2\gamma \text{ Å}.$$
 (5.1)

For $\gamma \gg 1$, it is thus possible to choose an interval length Δt small enough to make the expansion parameter small compared to unity, *and* large enough to include correlations of thermal displacements.

In all cases, expansion in the parameter λ simplifies to expansion of the exponential function $\exp[i\Delta\omega(t-t_m)]$ $-\Delta t/2$)] which for the separate second-order terms appears in the phase factors of (4.7a) and (4.7b).

We shall start out by expanding $\hbar \gamma_{10}^{(2)}$ in the new parameter. To zeroth order in λ , the dependence on the energy of intermediate states disappears. Neglecting the exclusion of the term accounting for scattering to the state itself, we may apply closure of intermediate states and hence obtain for twice the separate term in Eq. (4.7a)

$$\left[\frac{\hbar}{\tau_1}\right]^{(2)} = \frac{\hbar}{\Delta t} \langle 1q | (iQ_{10})^2 | 1q \rangle , \qquad (5.2a)$$

where the notation

$$Q_{10} = \frac{1}{i\hbar} \int_{t_m}^{t_{m+1}} dt \, \delta V_2(t)$$
 (5.2b)

has been introduced to express similarity to "the suddencollision expansion" outlined in [8]. The potential average Q_{10} defined here equals Q/i used in [19]. The two subscripts of Q in the sudden-collision expansion correspond to the two expansions introduced above, with the first subscript of Q denoting the order in the perturbation and the second the order in λ .

The second-order shift (4.7b), expanded to zeroth order in λ , vanishes.

The sum of the two coupling terms of Eq. (4.5), expanded to zeroth order in λ , equals

$$(\hbar\Delta\gamma_{10})^{(2)} = i\hbar\sum_{q'} \frac{1}{\Delta t} \langle 0q | iQ_{10} | 0q' \rangle \langle 1q' | iQ_{10} | 1q \rangle ,$$
(5.3)

which is purely imaginary as is easily seen by writing the Bloch states as sums of localized "atomic" states.

Hence, to obtain a nonvanishing shift contribution of second order in the perturbation, we need to expand to first order in λ . For the separate part,

$$(\hbar\Delta\omega_1)^{(2)} = \sum_{n_2, q_2 \neq 1, q} \frac{1}{2} \frac{1}{\hbar} \frac{1}{\Delta t} \omega_{1q, n_2 q_2} \int_{t_m}^{t_{m+1}} dt \langle 1q | \delta V_2(t) | n_2 q_2 \rangle \int_{t_m}^{t_{m+1}} dt' \langle n_2 q_2 | \delta V_2(t') | 1q \rangle | t - t' | , \qquad (5.4)$$

which clearly reveals the proportionality of the shift to the lifetime of the intermediate state, |t-t'|. We shall write $|t-t'| = \operatorname{sgn}(t-t')(t-t')$ in terms of a derivative of the Fourier expansion of the sign function. To perform the summation over intermediate states, the energy difference between the initial and an intermediate state, $\hbar\omega_{1q,n_2q_2}$, which is by far dominated by the energy difference between the transverse electron states, will be written in terms of a commutator between the scattering potential and the continuum-model Hamiltonian operator. As shown in Appendix D, this leads to the sum rule (D1) which we shall apply here with the result

$$(\hbar\Delta\omega_1)^{(2)} = \frac{1}{\Delta t} \frac{1}{2\pi} \int d\omega \frac{P}{\omega} \frac{d}{d\omega} \frac{1}{2m\gamma} \langle 1q | [\nabla_{\perp} \delta V_2(-\omega)] \cdot [\nabla_{\perp} \delta V_2(\omega)] | 1q \rangle .$$
(5.5)

The sum of the real parts of the second-order coupling terms in (4.5) may be shown to vanish also to first order in the expansion parameter λ .

We finally need to consider the shift expanded to third order in the perturbation and to zeroth order in λ . For

the real part of the first separate term in (4.6), we get

$$\hbar \Delta \omega_1^{(3)} = -\frac{1}{6} \frac{1}{\Delta t} \hbar \langle 1q | (iQ_{10})^3 | 1q \rangle .$$
 (5.6)

The sum of coupling terms in (4.6) expanded to zeroth

order in λ equals

$$\hbar \Delta \gamma_{10}^{(3)} = \sum_{q'} \frac{\hbar}{\Delta t} \{ \langle 0q | iQ_{10} | 0q' \rangle \langle 1q' | \frac{1}{2} (iQ_{10})^2 | 1q \rangle \\ - \langle 0q | \frac{1}{2} (iQ_{10})^2 | 0q' \rangle \langle 1q' | iQ_{10} | 1q \rangle \} .$$
(5.7)

By now it is clear that the necessity for the expansion of the level shift to third order in the perturbation originates in the competition between the two expansion parameters: The third-order shift has nonvanishing contributions of zeroth order in λ , whereas the second-order shift has contributions only from first order in λ . Thus the magnitude of the first nonvanishing part of the second-order shift relative to the first nonvanishing contribution to the third-order shift is given by the ratio between the parameter λ and the perturbation-expansion parameter,

$$\frac{\Delta\omega\Delta t}{(Ze^2/\rho)(1/\hbar)\Delta t} \approx \frac{1}{Z\alpha} \frac{\hbar c}{2m\gamma c^2 \rho} = \frac{1}{2Z\gamma} \frac{a_0}{\rho} , \qquad (5.8)$$

which is smaller than unity for electrons of a few MeV in Si.

We shall introduce a thermal average over initial lattice states, and hereby the first-order shift in (4.4) vanishes, $\langle \hbar \gamma_{10}^{(1)} \rangle_{\rm th} = 0$, by definition of δV_2 . Furthermore, with an expansion of the potential δV_2 in atomic contributions, the thermal average ensures that scattering back and forth takes place only on atoms which are vibrationally correlated.

To illustrate how approximate expressions in terms of unscreened Coulomb potentials may be obtained for the various contributions to the level widths and shifts, the separate second-order level shift in (5.5) will be considered. In the transverse matrix element, the scattering potential will be written as a sum of atomic contributions which in turn are Fourier expanded,

$$\partial V_2(\mathbf{R}) = \sum_i \frac{1}{(2\pi)^3} \int d\mathbf{K} \{ V_a(K) \exp(-i\mathbf{K} \cdot \mathbf{U}_i) - V_{aT}(K) \} \exp[i\mathbf{K} \cdot (\mathbf{R} - \mathbf{R}_i)] .$$
(5.9)

The product $\nabla_{\perp} \partial V_2(-\omega) \cdot \nabla_{\perp} \partial V_2(\omega)$ is simplified if we first perform the average $\langle \rangle_{U_T}$ which consists of an integration over atomic variables, combined with a thermal average denoted $\langle \rangle_{\text{th}}$. By using that the average of exponentials of operators, linear in the positions and momenta of a harmonic crystal, may be transformed into an exponential of averages of the operators [20],

$$\langle e^{A}e^{B}\rangle_{\mathbf{U}_{T}} = \exp\{\frac{1}{2}\langle A^{2}\rangle_{\mathbf{U}_{T}} + \frac{1}{2}\langle B^{2}\rangle_{\mathbf{U}_{T}} + \langle AB\rangle_{\mathbf{U}_{T}}\},$$

(5.10)

we obtain

$$\langle \{ V_a(K_2) \exp(-i\mathbf{K}_2 \cdot \mathbf{U}_{i_2}) - V_{aT}(K_2) \} \{ V_a(K_1) \exp(-i\mathbf{K}_1 \cdot \mathbf{U}_{i_1}) - V_{aT}(K_1) \} \rangle_{\mathbf{U}_T}$$

= $V_{aT}(K_2) V_{aT}(K_1) \{ \exp[-\mathbf{K}_2 \cdot \mathbf{K}_1 \beta_c (\mathbf{R}_{i_2} - \mathbf{R}_{i_1}) \rho^2] - 1 \} , \quad (5.11)$

where ρ is the one-dimensional root-mean-square vibrational amplitude and $\beta_c(\mathbf{R}_{i_2} - \mathbf{R}_{i_1})$ the correlation coefficient for thermal displacements of atoms i_2 and i_1 . In deriving (5.11), we have assumed isotropy of the crystal,

$$\langle U_{i_{2}x}U_{i_{1}x}\rangle = \langle U_{i_{2}y}U_{i_{1}y}\rangle = \langle U_{i_{2}z}U_{i_{1}z}\rangle = \beta_{c}(\mathbf{R}_{i_{2}} - \mathbf{R}_{i_{1}})\rho^{2} .$$
(5.12)

In the special case $i_2 = i_1$, the definition of ρ implies that $\beta_c(0) = 1$. Equation (5.11) is furthermore based upon an assumption of vanishing correlation between perpendicular displacements.

For axial channeling we obtain

$$\langle \hbar \Delta \omega_{1}^{(2)} \rangle_{\text{th}} = -\frac{1}{2m\gamma v} N_{d} L_{x} L_{y} \frac{1}{(2\pi)^{5}} \\ \times \int dk_{1x} \int dk_{2x} \int dk_{1y} \int dk_{2y} \langle u_{1} | \exp[i(\mathbf{k}_{1} + \mathbf{k}_{2}) \cdot \mathbf{r}] | u_{1} \rangle \langle k_{1x} k_{2x} + k_{1y} k_{2y} \rangle \\ \times \int d\omega \frac{P}{\omega} \frac{d}{d\omega} V_{aT}(K_{1}) V_{aT}(K_{2}) \\ \times \sum_{\mathbf{R}_{i}} \{ \exp[-\rho^{2}\beta_{c}(\mathbf{R}_{i})\mathbf{K}_{1} \cdot \mathbf{K}_{2}] - 1 \} \exp(-iK_{1z}z_{i}) , \qquad (5.13a)$$

where N_d is the density of atoms in the crystal and u_1 a two-dimensional Bloch state normalized over the quantization area $L_x L_y$. The vectors \mathbf{R}_i are in this case parallel to the axis, $\mathbf{R}_i = (0, 0, z_i)$, and $\mathbf{K}_1 = (k_{1x}, k_{1y}, \omega/\nu)$, $\mathbf{K}_2 = (k_{2x}, k_{2y}, -\omega/\nu)$. The factor $k_{1x}k_{2x} + k_{1y}k_{2y}$ is a result of applying the transverse-gradient operators of Eq. (5.5). Similarly, for the planar case,

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$$\langle \hbar \Delta \omega_{1}^{(2)} \rangle_{\text{th}} = -\frac{1}{2m\gamma v} N_{d} L_{x} \frac{1}{(2\pi)^{4}} \int dk_{1x} \int dk_{2x} \langle u_{1} | \exp[i(k_{1x} + k_{2x})x] | u_{1} \rangle$$

$$\times \int dk_{y} (k_{1x} k_{2x} - k_{y}^{2}) \int d\omega \frac{P}{\omega} \frac{d}{d\omega} V_{aT}(K_{1}) V_{aT}(K_{2})$$

$$\times \sum_{\mathbf{R}_{i}} \{ \exp[-\rho^{2}\beta_{c}(\mathbf{R}_{i})\mathbf{K}_{1}\cdot\mathbf{K}_{2}] - 1 \}$$

$$\times \exp(-i\mathbf{K}_{1}\cdot\mathbf{R}_{i}) , \qquad (5.13b)$$

where u_1 denotes a one-dimensional Bloch state normalized over the quantization length L_x , $k_{1y} = -k_{2y} \equiv k_y$, and the summation is over Bravais-lattice vectors in the plane, $\mathbf{R}_i = (0, y_i, z_i)$. The condition of y momentum conservation is obtained from the y integration in the transverse matrix element.

So far no approximations have been made. In Appendix B, the Doyle-Turner approximation [18] is applied for the atomic Fourier coefficients. This leads after lengthy calculations to the very precise formulas given there. Here we shall make somewhat crude approximations for the atomic scattering factors.

Consider the term $\mathbf{R}_i = \mathbf{0}$: Scattering back and forth on the same atom. We shall first evaluate (5.13a) and (5.13b) for a plane wave u_1 . The coordinate integration then gives a δ function requiring $\mathbf{k}_2 = -\mathbf{k}_1$, and the level shift becomes

$$\langle \hbar \Delta \omega^{(2)} \rangle_{\text{th}} = -\frac{1}{2m\gamma v^2} N_d \frac{1}{(2\pi)^3} \times \int d^3 \mathbf{k} (-k^2 + k_z^2) 2 \frac{\partial}{\partial (k_z^2)} V_a^2(k) \times [1 - \exp(-k^2 \rho^2)], \quad (5.14)$$

for plane wave.

For large k, the potential approaches the unscreened Coulomb potential from the nucleus,

$$V_a(k) \approx -\frac{4\pi Z e^2}{k^2}, \quad k > 1/a_{\rm TF} ,$$
 (5.15)

where the Thomas-Fermi screening distance is given by $a_{\rm TF} = 0.8853 a_0 Z^{-1/3}$ (a_0 is the Bohr radius, $a_0 = 0.529$ Å). For small k, $k < 1/\rho$, the increase of the integrand in (5.14) is cut off partly by the last factor, proportional to k^2 , partly by screening. As a rough approximation, we may apply the unscreened potential (5.15) in (5.14), with a lower limit of integration $k_{\rm min} = 1/\rho$. This leads to a very simple formula

$$\langle \hbar \Delta \omega^{(2)} \rangle_{\rm th} \simeq -10 \alpha^2 \gamma^{-1} Z^2 e^2 (N_d a_0 \rho), \quad v \simeq c$$
, (5.16)

for plane wave. For bound electron-channeling states, there is a strong enhancement of the flux near atoms, and we may expect (5.16) to be replaced by

$$\langle \hbar \Delta \omega^{(2)} \rangle_{\rm th} \simeq -10 \alpha^2 \gamma^{-1} Z^2 e^2 (N_d a_0 \rho) O$$
, (5.17)

where the overlap factor O accounts for the increased flux. This formula may be derived from (5.13a) and (5.13b) by using that momenta in the integral are typically large, $k \ge 1/\rho$, compared to wave vectors of Fourier

components contained in the projectile wave function, i.e., the density $|u_1|^2$ varies little over distances ρ . The overlap is given by

$$O = \frac{1}{(2\pi)^2} L_x L_y \int d^2 \mathbf{K} \exp(-\frac{1}{2}K^2 \rho^2) \langle u_1 | \exp(i\mathbf{K} \cdot \mathbf{r}) | u_1 \rangle$$

= $\frac{1}{2\pi\rho^2} L_x L_y \langle u_1 | \exp(-r^2/2\rho^2) | u_1 \rangle$, (5.18a)

for axial channeling, and by

$$O = \frac{L_x}{2\pi} \int dK \exp(-\frac{1}{2}K^2 \rho^2) \langle u_1 | \exp(iKx) | u_1 \rangle$$

= $\frac{1}{\sqrt{2\pi}} \frac{L_x}{\rho} \langle u_1 | \exp(-x^2/2\rho^2) | u_1 \rangle$, (5.18b)

for planar channeling. Comparison of the simple estimate (5.16)-(5.18) with the more exact evaluation in Appendix B in a few cases showed good agreement (see Appendix C).

From (5.11) it is seen that scattering back and forth takes place only on vibrationally correlated atoms. To have contributions from scattering back and forth on different atoms, a nonvanishing transverse overlap between the scattering potentials, localized around the atomic equilibrium positions, is required. In the planar case, the z axis is chosen parallel to the projection of the initial projectile momentum onto the plane. Thereby correlated scattering on different atoms takes place only if the y distance between the atoms is less than twice the extension of the scattering potential. This corresponds to an upper limit $\varphi \approx \rho/d$ for angles of incidence to the string containing the atoms, and as soon as the angles to all major axes in the plane are larger than a few degrees, we need only include scattering sequences on single atoms. The dependence of the linewidth of planarchanneling radiation on the angle of incidence to a major axis in the plane was studied in [19], and it was found that the effect of vibrational correlations is significant at angles less than a few degrees.

For axial channeling, the correlation of thermal scattering from different atoms along the string is important. As discussed in [21], the decay constant may increase by up to a factor of ~ 2 when correlations are included, and the effect is even more dramatic for the shift. The reason is that the contribution to the shift from a virtual transition back and forth to an intermediate state is proportional to the lifetime of the intermediate state, i.e., in our description with a pseudotime z/v, it is proportional to the distance Δz between the two scattering events. For thermal scattering on one atom, this distance is $\Delta z \leq \rho$, while for correlated scattering on neighboring atoms along the string, $\Delta z = d \gg \rho$.

The axial formula (5.13a) with one term $\mathbf{R}_i = (0, 0, nd)$ in the sum may be approximated by an expression similar to formulas (5.17) and (5.18) as

$$\langle \hbar \Delta \omega^{(2)} \rangle_{\text{th},nd} \simeq -\pi \beta_c \alpha^2 \gamma^{-1} Z^2 e^2 (N_d a_0 | nd |) O$$
 (5.19)

In this case we have used that the Debye-Waller factors in the scattering factors of the thermally averaged atomic potential, combined with part of the factor in curly brackets limit the integration to momenta small compared to $1/[\rho(1-\beta_c)^{1/2}]$, and this permits a first-order expansion of the remaining factor in curly brackets when $\beta_c \ll 0.5$ which is usually the case. Furthermore, the last exponent has the strongest variation with ω , and the differentiation is confined to this factor. The resulting ω dependence limits the integration over ω to very small values, $\omega \leq v/|nd|$, whereby the ω integration separates, and Eq. (5.19) is obtained for small β_c . [In the opposite limit of $\beta_c \approx 1$, the integration is limited to $k \geq 1/\rho$ in analogy to (5.14) and estimate (5.19) is obtained also in this case.]

Comparing Eq. (5.19) with (5.17), we see that a factor ρ has been replaced by |nd| as predicted qualitatively above. Since in the normal case, $\beta_c \ll 1$, the integral receives the main contributions from $k \leq 1/\rho$, the accuracy of our procedure (neglect of screening and overlap approximation) may be somewhat poorer than for the previous case (5.17). This is confirmed by the comparisons in Appendix C which indicate that (5.19) overestimates the shift by up to a factor of 2.

To obtain the total second-order shift, (5.19) should be summed over $n = \pm 1, \pm 2, \ldots$ The (transverse) correlation coefficient β_c decreases with distance, but at high temperatures only as $\beta_c \propto 1/|nd|$, according to the Debye model. Hence the magnitude of (5.19) becomes independent of distance, and the sum diverges. An estimate for the effective cutoff of the sum is obtained from the condition of validity for the expansion leading to formula (5.5), $\lambda \equiv \Delta\omega\Delta t < 1$, where $\hbar\Delta\omega$ is a typical transverse-energy transfer, and $\Delta t = |nd|/v$ is the intermediate-state lifetime. According to (5.1), this leads to $|nd| \le 2\gamma$ Å.

This cutoff has a simple interpretation. After scattering on a string atom with momentum transfer $\hbar k \approx \hbar/\rho$, the projectile moves at an angle $\Theta \approx \hbar/(m\gamma v\rho)$ to the string. Since the perturbing potential has a range $\sim \rho$, the projectile cannot "see" atoms at distances larger than $\sim 2\rho/\Theta$.

Similar overlap estimates are obtained easily for the level width (5.2) by restricting the integration to momenta $k \ge 1/\rho$ and compensating this underestimation by application of unscreened Coulomb potentials. By furthermore assuming that the wave function contains momenta only which are small compared to typical momentum transfers \hbar/ρ , we obtain

$$\left\langle \left[\frac{\hbar}{\tau}\right]^2 \right\rangle_{\rm th} \simeq (\hbar v) N_d \sigma_{\rm th} O$$
, (5.20)

with a cross section for thermal scattering given by

$$\sigma_{\rm th} = \frac{1}{(2\pi\hbar\nu)^2} \int d^2\mathbf{k} |V_a(k)|^2 [1 - \exp(-k^2\rho^2)]$$

\$\approx 4\pi Z^2 \alpha^2 \rho^2 , \$(5.21)

where only scattering back and forth on one atom has been included. In silicon at room temperature, the estimate (5.21) is low by $\approx 35\%$ (see Appendix C). The accuracy depends on $\rho/a_{\rm TF}$ being better for larger values of this parameter.

It may be useful to express the second-order level shift (from scattering on one atom) in terms of this level width, and we obtain, from (5.20), (5.21), and (5.17),

$$\langle \hbar \Delta \omega^{(2)} \rangle_{\rm th} / \langle \hbar / \tau \rangle_{\rm th} \simeq -\alpha \gamma^{-1} \left[\frac{a_0}{\rho} \right] .$$
 (5.22)

We note that the ratio decreases with increasing projectile energy, and already for $\gamma \approx 10$, it is of order 10^{-2} only.

Finally, for the third-order level shift we obtain with similar but more complicated considerations

$$\langle \hbar \Delta \omega^{(3)} \rangle_{\rm th} \simeq 2\pi \alpha^2 Z^3 e^2 (N_d \rho^2) O$$
 (5.23)

This also includes scattering on one atom only and agrees in the planar case within a factor of 2 or 3 with the precisely calculated third-order shift (Appendix C).

For both the linewidth and the third-order line shift, there are important corrections from correlation between scattering in the initial and final states of the radiative transition. In Appendix C precise formulas are given not just for the second- and third-order level shifts, but also for the correlation terms of third order (the linewidth was treated in [19]).

It is of interest to compare the third-order level shift in (5.23) to the second-order level shift in (5.17) (scattering back and forth on the same atom),

$$\langle \hbar \Delta \omega^{(3)} \rangle_{\rm th} / \langle \hbar \Delta \omega^{(2)} \rangle_{\rm th} \simeq -\frac{2}{3} Z \gamma \left[\frac{\rho}{a_0} \right].$$
 (5.24)

`

The two shifts are of opposite sign, and the third-order shift will dominate for large values of γ . [Compare also to the estimate (5.8).] It should be stressed that this result does not indicate a break-down of the perturbation expansion. The third-order shift is important because it has a contribution of zeroth order in the expansion parameter λ , while the second-order shift (second order in the Born parameter $Z\alpha$) contains contributions only of first and higher order in λ . All further terms in the expansion are of higher order in one or both of the expansion parameters. Note further that for axial channeling, thermal-vibrational correlation may give a large increase in the second-order level shift according to (5.19).

VI. PERIODIC PERTURBATIONS

The line shifts induced by the discrete components δV_3 and δV_4 will be treated by a similar method as used for the phonon coupling. In this case, scattering back and forth may take place with infinitely large separation and, to include all possible scattering processes, we must extend Δt to cover the full crystal thickness. Hence the expansion in a parameter λ proportional to Δt cannot be applied. However, it is still possible to introduce an expansion parameter λ' defined similarly to λ but with the full interval length Δt replaced by an effective distance between scattering events.

A. Atomic structure of strings

As will turn out, the only important contributions to the coupled quantity $\hbar\gamma_{10}$ are the independent, secondorder level shifts of the initial and final states for the radiation process. Starting out by a treatment of the scattering potential δV_3 , which is regarded as a perturbation to the axial continuum-model operator, we immediately obtain from Eq. (4.8)

$$\hbar\Delta\omega_{1}^{(2)} = -\sum_{|n_{2}\mathbf{k}_{2}\rangle \ (\neq u_{1})} \frac{1}{\hbar} \frac{1}{(\Delta t)^{2}} \times \sum_{m \ (\neq 0)} |\langle n_{2}\mathbf{k}_{2}|\delta V_{3}(\omega')|u_{1}\rangle|^{2} \times \frac{1}{\omega' - \omega_{1n}} ,$$
$$\omega' = m \frac{2\pi}{d/\nu} , \quad (6.1)$$

where the lattice parts of the wave functions are omitted $(|nq\rangle \rightarrow |nk\rangle)$ since δV_3 depends only on the projectile coordinates. The scattering component contains no constant term (zero-frequency component) due to the definition of δV_3 as the difference between the crystal and the continuum potential.

We shall now introduce the substitution for the expansion parameter λ of the phonon case. Writing the last factor of Eq. (6.1) as

$$\frac{1}{\omega' - \omega_{1n}} = \frac{\omega' + \omega_{1n}}{\omega'^2 - \omega_{1n}^2} , \qquad (6.2)$$

and noting that $|\langle n_2 \mathbf{k}_2 | \delta V_3(\omega') | u_1 \rangle|^2$ is symmetric in ω' , we may immediately neglect the antisymmetric ω' term. The frequencies ω' are much larger than typical frequencies ω_{1n} , and hence we may approximate the denominator of the right-hand side of (6.2) by ω'^2 . This corresponds to an expansion of (6.2) to first order in the parameter $\lambda' \equiv |\omega_{1n}/\omega'|$ where the full interval Δt , occurring in the definition of λ , has been replaced by the time separation between atoms along the string.

To estimate the magnitude of λ' , we note that the matrix element in (6.1) becomes exponentially small for momentum transfers larger than \hbar/ρ due to Debye-Waller factors. Using the corresponding energy, $E_{\perp} \sim (\hbar/\rho)^2/2m\gamma$, as an upper estimate for $|\hbar\omega_{1n}|$, we obtain

$$\lambda' \lesssim \frac{1}{4\pi} \left[\frac{a_0 d}{\rho^2} \right] \alpha \gamma^{-1} \simeq \gamma^{-1} .$$
 (6.3)

In (6.2), the energy difference $\hbar \omega_{1n}$ may be written as a commutator, repeating the method used for the second-

order phonon shift. Applying again the sum rule (D1), we obtain

$$\hbar\Delta\omega_1^{(2)} = \frac{1}{2m\gamma} \frac{1}{(\Delta t)^2} \sum_{m \ (\neq 0)} \langle u_1 | |\nabla_1 \delta V_3(\omega')|^2 | u_1 \rangle \frac{1}{\omega'^2} ,$$
$$\omega' = m \frac{2\pi}{d/\nu} . \quad (6.4)$$

With the terminology of [9], the level shift is given as the expectation value of the sum over Kapitza potentials associated with the various Fourier components of the perturbation.

For the Fourier coefficient $\delta V_3(\omega')$, the following approximation may be introduced for well localized atomic potentials,

$$\delta V_{3}(\omega') = \int_{t_{m}}^{t_{m+1}} dt \exp(-i\omega't) \delta V_{3}(t)$$

= $\frac{\Delta t}{d/\nu} \int_{0}^{d/\nu} dt \exp(-i\omega't) \delta V_{3}(t)$
 $\approx \Delta t \sum_{i=1}^{N} \exp(-i\omega't_{i}) U_{T}(\mathbf{r}) \frac{1}{N} \quad (a \ll d) , \quad (6.5)$

where the summation extends over the N atoms contained in one period of the string, and a is the extension of the atomic potential being of the order of the Thomas-Fermi screening length. When only one atom per period is encountered (as in the case of the $\langle 100 \rangle$ axis in silicon), we may write the second-order shift as

$$\hbar \Delta \omega_1^{(2)} = \frac{(d/v)^2}{24m\gamma} \langle u_1 | (\nabla_1 U_T)^2 | u_1 \rangle .$$
 (6.6)

Note that it is immaterial whether we consider u_1 to be a single-string state or a Bloch state.

The magnitude of the string potential U_T is of order Ze^2/d , and an upper estimate of the gradient in (6.6) is $|\partial U_T/\partial r| < Ze^2/d\rho$, attained at distances $r \sim \rho$. We use half of this value to obtain a simple estimate

$$\hbar\Delta\omega^{(2)} \sim 10^{-2} \alpha^2 \gamma^{-1} Z^2 e^2 a_0 / \rho^2$$
, (6.7)

which may be compared with the second-order thermal shift (5.17),

$$\langle \hbar \Delta \omega^{(2)} \rangle_{\rm th} | / \hbar \Delta \omega^{(2)} \sim 10^3 N_d \rho^3 O$$
 (6.8)

The right-hand side is typically of order unity. It is easy to make an accurate evaluation of expression (6.6), and numerical results are given in Appendix C.

In the slightly more complicated case of, say, the $\langle 111 \rangle$ axis of silicon, which will be treated in Appendix C, we have two atoms, which are separated by the distance d/4, per lattice period along the axis, and thereby

$$\hbar\Delta\omega_1^{(2)} = \frac{7}{16} \frac{(d/v)^2}{24m\gamma} \langle u_1 | (\nabla_\perp U_T)^2 | u_1 \rangle .$$
 (6.9)

The estimate (6.7) should for this case be multiplied by $\frac{28}{16}$ since there are two atoms per period d in the $\langle 111 \rangle$ strings.

Turning now to the decay constant, we may write expression (4.7a) in the form of the "golden rule" [8],

$$\left[\frac{\hbar}{\tau_1}\right]^{(2)} = \sum_{\omega \ (\neq 0)} \sum_{|n_2 \mathbf{k}_2\rangle \ (\neq u_1)} \frac{2\pi}{\hbar} |\langle n_2 \mathbf{k}_2 | \Omega_3(\omega) | u_1\rangle|^2 \delta(\omega_{1n} - \omega) , \qquad (6.10a)$$

where

$$\Omega_3(\omega) = \frac{v}{d} \int_0^{d/v} dt \,\,\delta V_3(\mathbf{r},t) \exp(-i\omega t) \,\,. \tag{6.10b}$$

The discrete component contributes only weakly to line broadening since the requirement of energy conservation causes the transition matrix elements to be very small. As shown by the estimate (6.3), energy conservation requires momentum transfers larger than \hbar/ρ , for which the matrix elements are strongly reduced by the Debye-Waller factor.

There is no contribution from coupling terms of either second or third order in the axial-channeling case since they both include a first-order amplitude factor containing the vanishing zero-frequency Fourier component of the scattering potential.

To complete the treatment of the perturbation δV_3 , we must consider the separate third-order level shift. The contribution from scattering processes with energy conservation in the transition to intermediate states is negligible. The nonconserving processes contribute a term which involves a double integration (summation) over frequency with two principal values in the integrand. By expanding to zeroth order in the parameter λ' and applying closure, we see that the shift—as in the phonon case—is proportional to the third power of the time-averaged scattering potential. In the present case, however, the time average of the perturbation vanishes, and no thirdorder shift of zeroth order in λ' is obtained.

B. String structure of planes

We next consider the scattering component δV_4 which is a perturbation to the planar continuum-model operator. Viewing a crystal plane as a "string of strings," we may directly take over the considerations above for the independent second- and third-order shifts. As mentioned earlier, the y and z axes are usually defined in the planar-channeling case by requiring the y direction to be parallel to the projection of the initial momentum, whereby the perturbation δV_4 turns out to be time (z) dependent. It is convenient here to define an alternative set of coordinate axes (z', y') with z' directed along a major axis within the plane. We denote by φ the angle between the z and z' directions. Considering only simple planes, where the strings are equidistantly spaced, we obtain the analog of expression (6.6) for the second-order shift expanded to first order in λ' ,

$$\hbar\Delta\omega_1^{(2)} = \frac{\left[\frac{d_s}{(v\sin\varphi)}\right]^2}{24m\gamma} \left\langle u_1 \left| \left[\frac{d}{dx}V_T\right]^2 \right| u_1 \right\rangle.$$
(6.11)

Note that the discrete frequencies contained in δV_4 are separated by $\omega = 2\pi v \sin \varphi / d_s$, and hence $\lambda' = |\Delta \omega / \omega|$, where $\Delta \omega$ is a typical transverse-energy transfer induced by δV_4 . Compared to the estimate (6.3), the value of λ' is larger by a factor $\sim (\sin \varphi)^{-1}$, but λ' remains small for not too small values of γ and φ . For an order-ofmagnitude estimate, we may set $|dV_T/dx| \sim Nd_p Ze^2$, where d_p is the planar spacing, leading to a formula similar to (6.7),

$$\hbar\Delta\omega^{(2)} \sim 4 \times 10^{-2} \alpha^2 \gamma^{-1} Z^2 e^2 a_0 (N_d d_s d_p)^2 \varphi^{-2} . \qquad (6.12)$$

For a simple lattice, $d = (N_d d_s d_p)^{-1}$ is the atomic spacing along the z' axis, and in comparison to (6.7), the factor ρ^{-2} has been replaced by $(d\varphi/2)^{-2}$.

We may also repeat the arguments about the independent, third-order level shift which may then be neglected. Thus we need to consider the coupling terms only, which to second order in the perturbation strength may be written as a product of two first-order transition amplitudes and to third order as the product of a second-order term $(\sim \hbar \gamma_i^{(2)})$ and a first-order transition amplitude. Since energy conservation is required for these first-order transitions, the coupling terms turn out to be very small.

An alternative to the perturbation treatment here of the corrections δV_3 and δV_4 is to include in the diagonalization of the axial and planar Hamiltonian operators the most important Fourier components of δV_3 and δV_4 , respectively, those corresponding to the smallest reciprocal-lattice vectors. Such a calculation was carried out in [22] and [19] for the axial and planar case, respectively, and we compare results in Appendix C. It turns out that the estimate (6.11) is not very accurate. In fact, the approximation (6.5) may be expected to be less accurate in the planar case since the string potential is less peaked at small distances than the atomic potential. However, the estimate is still a useful guide to the magnitude of the correction, in particular to the strong dependence on the angle φ .

Also the contribution to level widths from the perturbation δV_4 was treated in [19], and we shall therefore not discuss it in detail here. We note that this type of scattering is often called "doughnut" scattering and is observed in transmission as scattering into a circle around an axis. It is important only at small angles to an axis, where the planar-channeling states become unstable.

VII. CONCLUDING REMARKS

A central point in the present paper is formula (4.1) which phrases precisely the essential assumption in physics of exponential decay. It results from the general treatment in Sec. III of line profiles for a radiating system externally perturbed and coupled to another system through a time-dependent interaction. The general formalism developed includes the use of a product space and of projection operators to treat the coupling of scattering in the initial and final states for a radiative transition. This coupling is of importance for linewidths and line shifts in radiation spectra.

The central formula is the basis for the first careful

evaluation of line shifts in channeling radiation, associated with corrections to the continuum model for channeling. An earlier attempt to calculate line shifts in electron-channeling radiation from lattice vibrations has been made by Strauss *et al.* [12] who estimated shifts of several percent for some cases. This result was not based on an explicit evaluation but was derived from a measured line broadening of ~20% and a simple formula for the ratio between the line shift and the linewidth, which in our notation becomes

$$\Delta \omega_{10}^{(2)} \tau_{10} \simeq \frac{L_p^0 \hbar \omega_{10}}{2\hbar c} . \tag{7.1}$$

The quantity L_p^0 was introduced as a "transversevibration correlation length" and was claimed—with reference to our Ref. [16]—to be several lattice spacings.

It is instructive to compare this formula to our results for the ratio between the thermal second-order shift (5.19) and the decay constant (5.20) and (5.21) which may be written as

$$\Delta \omega^{(2)} \tau \simeq \frac{\beta_c |nd|}{2\hbar c} \left[\frac{\hbar}{\rho}\right]^2 \frac{1}{2m\gamma} . \tag{7.2}$$

The difference between the two formulas has a simple qualitative interpretation. In [12], only scattering between the two levels 0 and 1, with energy difference $\hbar\omega_{10}$, was considered. This strongly underestimates the thermal scattering which predominantly populates free states with transverse energies up to $(\hbar/\rho)^2/2m\gamma$, corresponding to a momentum transfer $\sim \hbar/\rho$ [19]. In (7.2), this energy hence replaces $\hbar\omega_{10}$, leading to an *increase* in the estimated shift $\Delta\omega_{10}^{(2)}$. However, this is more than compensated for by the replacement of L_p^0 by $\beta_c |nd|$, a natural measure of the correlation length. As calculated in [16], $\beta_c |nd|$ is typically a fraction of an Å and not several lattice spacings, and hence the estimate of $\Delta\omega_{10}^{(2)}$ is *reduced* by one to two orders of magnitude. The combined result is a reduction by about an order of magnitude.

According to (7.2), the ratio of line shift to line broadening is determined by an effective collision time multiplied by a characteristic value of the energy jump in virtual transitions. We may use this general result for a second-order perturbation calculation to estimate the line shifts from excitations of target electrons. The contribution from electronic scattering to the linewidth is small [19,23], because the cross section for Coulomb scattering is proportional to the square of the charge. However, for distant collisions (small momentum transfers), the product analogous to (7.2) is of order unity since the energy jump comprises the excitation energy $\hbar \omega_{\rm el}$, and the effective collision time is of order $\omega_{\rm el}^{-1}$. Hence the contribution to level shifts should be relatively larger. Nevertheless, the resulting line shifts will be negligible because the distant collisions are nearly independent of the channeling state of the projectile and therefore do not affect transition energies.

We may therefore conclude that the present investigation of higher-order corrections to the simple continuum channeling model, from thermal scattering and periodicity of the crystal potential, results in a very precise theoretical description for channeling radiation. Channeling-radiation spectroscopy may then be used for studies of solid-state parameters: Electron densities, thermal-vibrational amplitudes, and potentially for studying vibrational anomalies in high- T_c superconductors.

It may also be noted that essentially the same theoretical model is the basis for analysis of so-called criticalvoltage measurements of electron diffraction which have been used to derive very accurate low-order Fourier components of the electron density in crystals [24]. Our results may be applied to estimate corrections from level shifts due to thermal scattering and periodic perturbations.

APPENDIX A: REFRACTION

The precise Hamiltonian operator contains a coupling between the charged lattice constituents and the radiation field. In the following, the effects of the coupling operator will be investigated, and for this purpose, we may neglect the incoming, channeled particle.

The coupling operator will introduce energy shifts of the unperturbed states $|i;N_{\lambda}\rangle = |i\rangle|N_{\lambda}\rangle$ where $|i\rangle$ denotes the electron wave function of the lattice and $|N_{\lambda}\rangle$ the state of the radiation field with excitation number N_{λ} for the mode λ . Here λ defines the wave vector κ as well as the polarization index μ .

It turns out that for an expansion to lowest order in the fine-structure constant α , the level shift may be written as the sum of a term proportional to the excitation numbers N_{λ} and a constant part, which may be associated with the electronic state of the lattice, the analog of the Lamb shift for atoms [25].

The linear term, on the other hand, may be described as a photon-energy shift and is thus related to the refractive index of the medium. We shall use this connection to calculate the refraction, and the result will be compared to Lindhard's calculation of the transverse dielectric constant [26]. With the same approximation (free electrons described in the Hartree picture), the two different approaches to the problem agree.

Calculation of energy shift

The electrons may be described nonrelativistically since typical Fermi velocities are of the order of 1% of the velocity of light. Accordingly, the coupling operator is given by

$$H_1 = \sum_j \frac{1}{m} \frac{e}{c} \mathbf{A}_j \cdot \mathbf{P}_j + \frac{1}{2m} \frac{e^2}{c^2} \mathbf{A}_j^2 , \qquad (A1)$$

where the summation is over all electrons in the crystal. The vector potential \mathbf{A}_i is given by

$$\mathbf{A}_{j} = \sum_{\lambda} \left[\frac{4\pi c^{2} \check{n}}{V 2 \omega} \right]^{1/2} \boldsymbol{\epsilon}_{\mu} \{ a_{\lambda} e^{i \boldsymbol{\kappa} \cdot \mathbf{R}_{j}} + a_{\lambda}^{+} e^{-i \boldsymbol{\kappa} \cdot \mathbf{R}_{j}} \} , \qquad (A2)$$

where \mathbf{R}_j is the position of the *j*th electron and V the quantization volume.

It might be tempting to neglect the A_i^2 terms since the

coefficient in front is proportional to α and is $\sqrt{\alpha}$ times the coefficient of $\mathbf{A}_j \cdot \mathbf{P}_j$. However, \mathbf{A}_j^2 gives a first-order energy shift, whereas $\mathbf{A}_j \cdot \mathbf{P}_j$ contributes to second order only. In effect, both terms induce shifts proportional to α . Transition probabilities, on the other hand, are dominated by the $\mathbf{A}_j \cdot \mathbf{P}_j$ terms since two-photon processes induced by \mathbf{A}_j^2 are weaker by a factor of α than the onephoton processes: spontaneous and stimulated emission and absorption.

The first-order energy shift is given by

$$\hbar \Delta \omega_{i,N_{\lambda}}^{(1)} = \langle i; N_{\lambda} | H_{I} | i; N_{\lambda} \rangle$$

$$= \frac{1}{2m} \frac{e^{2}}{c^{2}} \frac{4\pi c^{2} \hbar}{2V} \mathcal{N} \left[\frac{1}{\omega} (2N_{\lambda} + 1) + \sum_{\lambda' \ (\neq \lambda)} \frac{1}{\omega'} \right],$$
(A3)

where \mathcal{N} is the total number of electrons.

From (A3) we obtain the shift of the electronic state $|i\rangle$ by inserting $N_{\lambda} = 0$,

$$\Delta_i^{(1)} = \mathcal{N}\left[\frac{4\pi e^2 \hbar}{4m} \frac{1}{(2\pi c)^3} 2 \int_0^\infty d\omega' \omega' 4\pi\right], \qquad (A4)$$

where the mode summation has been replaced by an integration. Since this expression is independent of the state $|i\rangle$, exactly the same result will obtain for free and for bound electrons, and it is therefore natural to regard it as part of the observed electron mass.

The energy shift of the radiation field is given by

$$\Delta_{N_{\lambda}}^{(1)} = \hbar \Delta \omega_{i,N_{\lambda}}^{(1)} - \Delta_{i}^{(1)} = N_{\lambda} \left[\omega_{0}^{2} \frac{\hbar}{\omega} \right] \frac{1}{2} , \qquad (A5)$$

where $\omega_0 = (4\pi e^2 \rho / m)^{1/2}$ is the plasma frequency expressed in terms of the electron density ρ .

The energy shift to second order in H_I and to first order in α is given by the $A_j \cdot P_j$ terms,

$$\hbar\Delta\omega_{i,N_{\lambda}}^{(2)} = \Delta_{N_{\lambda}}^{(2)} + \Delta_{i}^{(2)} , \qquad (A6)$$

where

$$\Delta_{N_{\lambda}}^{(2)} = N_{\lambda} \frac{4\pi e^{2}}{m^{2} 2 V} \frac{1}{\omega} \sum_{n} \left[\frac{\left| \left\langle n \left| \sum_{j} e^{-i\kappa \cdot \mathbf{R}_{j}} \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{P}_{j} \right| i \right\rangle \right|^{2}}{\omega_{in} - \omega} + \frac{\left| \left\langle n \left| \sum_{j} e^{i\kappa \cdot \mathbf{R}_{j}} \boldsymbol{\epsilon}_{\mu} \cdot \mathbf{P}_{j} \right| i \right\rangle \right|^{2}}{\omega_{in} + \omega} \right],$$
(A6a)

and

$$\Delta_{i}^{(2)} = \frac{4\pi e^{2}}{m^{2} 2 V} \sum_{n} \sum_{\lambda'} \frac{\left| \left\langle n \left| \sum_{j} e^{-i\kappa' \cdot \mathbf{R}_{j}} \boldsymbol{\epsilon}_{\mu'} \cdot \mathbf{P}_{j} \right| i \right\rangle \right|^{2}}{\omega_{in} - \omega'} \frac{1}{\omega'} \,.$$

(A6b)

The term $\Delta_i^{(2)}$ is the energy shift assigned to the electronic state. The corresponding shift for free particles equals

$$\Delta_i^{(2)}(\text{free}) = -\frac{2e^2}{3\pi\hbar c (mc)^2} (\hbar\omega_{\text{max}}) \sum_j \mathbf{P}_j^{(2)} , \qquad (A7)$$

where the electronic energy change, $\hbar\omega_{in}$, has been neglected in the denominator of (A6b). The change of kinetic energy for the *j*th electron with fixed momentum \mathbf{P}_j is negative and may be interpreted as a positive change of the electron mass due to the electromagnetic interaction (self-energy of the electron),

$$\Delta m = \frac{4}{3\pi c^2} \alpha(\hbar \omega_{\rm max}) . \tag{A8}$$

Here, $\hbar \omega_{max}$ is the upper limit for the integration over photon frequencies and is of the order of mc^2 . The limit is introduced by noting that within a nonrelativistic treatment of electrons it has no meaning to consider emission of virtual photons with momenta larger than mc since the electron recoil would cause the picture to break down.

To obtain the observable energy shift of a bound electron, we have to subtract from (A6b) the free-particle energy shift $\Delta_i^{(2)}$ (free) since this part is already included in the observed electron mass (mass renormalization).

However, our main concern is the energy shift of the electromagnetic field, which we obtain by summing the two contributions (A5) and (A6a),

$$\Delta_{N_{\lambda}} = \Delta_{N_{\lambda}}^{(1)} + \Delta_{N_{\lambda}}^{(2)} . \tag{A9}$$

Introducing now the index of refraction n_r [which changes the photon energy-momentum relation in the crystal, in the sense that the modes—the κ vectors, which are determined by boundary conditions—are fixed, whereas the energies of the corresponding excitation quanta (the frequencies) are changed], we may write the energy shift of the radiation-field state with excitation number N_{λ} of mode λ as

$$\Delta_{N_{\lambda}} = N_{\lambda} \hbar \omega \left[\frac{1}{n_{r}} - 1 \right] . \tag{A10}$$

Using (A9) and (A10), we obtain the dependence of the index of refraction on the radiative mode λ . The assumption of free electrons, described in the Hartree model, results in

$$\frac{1}{n_{r}} - 1 = \frac{1}{2} \frac{\omega_{0}^{2}}{\omega^{2}} \left[1 - \frac{1}{\mathcal{N}} \sum_{j} \left[k_{j}^{2} - \left[\frac{\kappa}{\kappa} \cdot \mathbf{k}_{j} \right]^{2} \right] \times \left[\frac{1}{\kappa^{2} - 2\kappa \cdot \mathbf{k}_{j} + \frac{2m}{\hbar} \omega} + \frac{1}{\kappa^{2} + 2\kappa \cdot \mathbf{k}_{j} - \frac{2m}{\hbar} \omega} \right] \right], \quad (A11)$$

where an average over the two polarization directions $(\frac{1}{2}\sum_{\mu})$ has been included, and where $\hbar k_j$ is the momen-

tum of the *j*th electron in the state $|i\rangle$. This may be compared with the expression for the transverse dielectric constant ϵ^{tr} , defined as

$$\left[k^{2} - \frac{\omega^{2}}{c^{2}}\epsilon^{\text{tr}}\right]\mathbf{A}(\mathbf{k},\omega) = \frac{4\pi}{c}\mathbf{j}_{0}^{\text{tr}}(\mathbf{k},\omega)$$
(A12)

[expression (1.3) in [26]], where A is the vector potential, regarded as a classical field and induced by the transverse part of the *external* current density, j_0^{tr} . The effect of the induced current density in the material is contained in the dielectric constant.

In the case under consideration, where no external charges appear, the source term j_0^{tr} is zero. Thus the excited modes \mathbf{k}, ω of the electromagnetic field must satisfy the energy-momentum relation,

$$\omega = kc / (\epsilon^{\rm tr})^{1/2} . \tag{A13}$$

Comparing (A13) with (A10), we obtain

$$\frac{1}{n_r} = \frac{1}{(\epsilon^{\text{tr}})^{1/2}} ,$$

$$\frac{1}{n_r} - 1 \simeq \frac{1}{2} (1 - \epsilon^{\text{tr}}), \quad n_r, \epsilon^{\text{tr}} \approx 1 .$$
(A14)

Using (A11) and (A14), we obtain Lindhard's equation (3.14) [26] (except for a missing factor $\frac{1}{2}$ in the first term in curly brackets therein). Lindhard used the same approximations in deriving his expression (3.14), namely, free electrons in the Hartree picture.

It is worthwhile to calculate the frequency dependence of the refractive index in the case of channeling radiation from silicon crystals ($\hbar\omega \gtrsim 1.5$ keV). The K absorption edge is found just below 2 keV so we are above the region of atomic resonances. In this limit, it seems reasonable to use a free-electron model, and for the electronic ground state, we obtain from (A11)

$$\frac{1}{n_r} - 1 \simeq \frac{1}{2} \frac{\omega_0^2}{\omega^2} \left\{ 1 + \frac{V}{(2\pi)^3} 2 \frac{1}{\mathcal{N}} \int_0^{k_F} k_j^2 dk_j \int_0^{\pi} \sin(\theta) d\theta \, 2\pi \left[k_j^2 - \left[\frac{\kappa}{\kappa} \cdot \mathbf{k}_j \right]^2 \right] \frac{2(\kappa^2 + 2\kappa \cdot \mathbf{k}_j)}{[(2m/\hbar)\omega]^2 - (\kappa^2 + 2\kappa \cdot \mathbf{k}_j)^2} \right\}.$$
(A15)

In the denominator, we may neglect the term $(\kappa^2 + 2\kappa \cdot \mathbf{k}_j)$ since

$$\frac{\hbar^2}{2m} |\kappa^2 + 2\kappa \cdot \mathbf{k}_j| \le \hbar\omega \left[\frac{\hbar\omega}{2mc^2} + \frac{v_F}{c} \right] \ll \hbar\omega \qquad (A16)$$

for the frequencies considered.

As a result,

$$\frac{1}{n_r} - 1 \simeq \frac{1}{2} \frac{\omega_0^2}{\omega^2} \left[1 + \frac{1}{5} \left[\frac{v_F}{c} \right]^2 \right], \qquad (A17)$$

where k_F and v_F are the wave vector and velocity at the Fermi surface, respectively.

This result should be compared with Lindhard's equation (3.19) [26], obtained for

$$\frac{\omega}{kv_F} > 1 + \frac{k}{2k_F} , \qquad (A18)$$

which is exactly the last inequality of (A16). Both the main term $\frac{1}{2}(\omega_0^2/\omega^2)$, resulting from the \mathbf{A}^2 term in H_I , and the much smaller correction term $\frac{1}{2}(\omega_0^2/\omega^2)\frac{1}{5}(v_F/c)^2$ agree with the Lindhard expression. The correction term was obtained as a combination of the "stimulated virtual emission and absorption" terms of (A6a), induced by the $\mathbf{A} \cdot \mathbf{P}$ part of H_I .

APPENDIX B: THERMAL SHIFTS WITH DOYLE-TURNER POTENTIAL

In this appendix, we present more precise analytical formulas for the thermal shifts of second and third order. Numerical results based on these formulas are given for a few cases in Appendix C.

Very accurate numerical results may be obtained with the "many-beam" solution (2.8) for the wave function u_1 and Doyle-Turner-type potential coefficients [18]. The Fourier components of the atomic potential are related to the electronic-scattering factors f_{el} ,

$$V_{a}(K) = \frac{-2\pi\hbar^{2}}{m} f_{\rm el}(s = K/4\pi) , \qquad (B1)$$

which in the Doyle-Turner approximation are given as a sum of four Gaussian functions,

$$f_{\rm el}(s) = \sum_{l=1}^{4} a_l \exp(-b_l s^2)$$
 (B2)

Fourier components for the thermally averaged atomic potential may be written in the same form,

$$V_{aT}(K) = \sum_{l=1}^{4} A_l \exp[-K^2(B_l/4 + \rho^2/2)]$$

= $\sum_{l=1}^{4} A_l \exp(-K^2 \mathcal{B}_l/4)$, (B3)

with the definition of \mathcal{B} given in (B5c), and where the potential parameters used in Eq. (B5a) are defined in terms of the fitting parameters (a_i, b_i) by

$$(A_l, B_l) = \left[\frac{-2\pi \hbar^2}{m} a_l, b_l / (4\pi^2) \right].$$
 (B4)

It is important in calculations of thermal scattering to apply potential parameters specially chosen to represent the atomic potential down to small distances, i.e., up to high K values. Hence new fits with four Gaussian functions have been performed [21,19], based upon electronicscattering factors, given by the Doyle-Turner approximation (with their fitting parameters) for $K \le K_0 = 2(4\pi)$ Å⁻¹ and by a pure Coulomb transform proportional to Ze^2/K^2 for $K \ge K_0$ (avoiding the region just above K_0). Explicit formulas, which express the second-order thermal level shift in terms of Doyle-Turner-type potential parameters (A_1, B_1) and Fourier coefficients of the wave function are obtained from Eqs. (5.13a) and (5.13b) by a rather lengthy derivation [1]. In the planar case, the result is

$$\langle \hbar \Delta \omega_{1}^{(2)} \rangle_{\text{th}} = \frac{N_{d}}{m \gamma v^{2}} \sum_{\mathbf{R}_{i}} \sum_{l,m} A_{l} A_{m} \sum_{\alpha,\beta} C_{\alpha}^{1} C_{\beta}^{1} \{F_{\text{pl}}(\beta_{c}(\mathbf{R}_{i})) - F_{\text{pl}}(\beta_{c}=0)\}$$
(B5a)

with

$$F_{pl}(\beta_{c}) = \{ (D_{lm}/\pi)^{1/2} \exp(-z_{i}^{2}/D_{lm}) + z_{i} \Phi[z_{i}/(D_{lm})^{1/2}] \} \exp(-y_{i}^{2}/D_{lm})/\pi D_{lm}^{2} \\ \times \exp\left[-(\alpha - \beta)^{2}g^{2} \frac{\mathcal{B}_{m}\mathcal{B}_{l} - 4\beta_{c}^{2}\rho^{4}}{4D_{lm}} \right] \\ \times \left[y_{i}^{2}/D_{lm} - 1 + (\alpha - \beta)^{2}g^{2} \left[\frac{\mathcal{B}_{m}\mathcal{B}_{l} - 4\beta_{c}^{2}\rho^{4}}{4D_{lm}} - \beta_{c}\rho^{2}/2 \right] \right],$$
(B5b)

where g is the primitive basis vector for the one-dimensional reciprocal lattice, and

$$\mathcal{B}_{l} \equiv B_{l} + 2\rho^{2} ,$$

$$D_{lm} \equiv \mathcal{B}_{l} + \mathcal{B}_{m} - 4\beta_{c}\rho^{2} ,$$

$$\Phi(x) \equiv \frac{2}{\sqrt{\pi}} \int_{0}^{x} dt \exp(-t^{2}) ,$$
(B5c)

which is known as the probability integral.

The planar Bravais-lattice vector \mathbf{R}_i in Eq. (B5a) is the distance between the two atoms on which scattering back and forth takes place. The factor $\exp(-y_i^2/D_{lm})$ introduces an effective upper limit of order 2ρ for the atomic distance along the y axis. This reflects the localized nature of the scattering potentials and is in accordance with the statement in Sec. V that correlation of thermal vibrations is important only when the angle of incidence to a major axis is smaller than ρ/d .

The corresponding expression in the axial-channeling case is given by

$$\langle \hbar \Delta \omega_1^{(2)} \rangle_{\text{th}} = \frac{N_d}{m \gamma v^2} \sum_{\mathbf{R}_i} \sum_{l,m} A_l A_m \sum_{\alpha,\beta,\alpha',\beta'} C^1_{\alpha,\beta} C^1_{\alpha',\beta'} \{ F_{\text{ax}}(\beta_c'(\mathbf{R}_i),\beta_c'(\mathbf{R}_i)) - F_{\text{ax}}(\beta_c'=0,\beta_c'=0) \} , \qquad (B6a)$$

where

$$F_{ax}(\beta_{c}^{r},\beta_{c}^{t}) = \{ (D_{lm}^{r}/\pi)^{1/2} \exp(-z_{i}^{2}/D_{lm}^{r}) + z_{i} \Phi[z_{i}/(D_{lm}^{r})^{1/2}] \} / \pi (D_{lm}^{t})^{2} \\ \times \exp\left[-(\mathbf{k} - \mathbf{k}')^{2} \frac{\mathcal{B}_{m} \mathcal{B}_{l} - 4(\beta_{c}^{t})^{2} \rho^{4}}{4D_{lm}^{t}} \right] \\ \times \left[-1 + (\mathbf{k} - \mathbf{k}')^{2} \left[\frac{\mathcal{B}_{m} \mathcal{B}_{l} - 4(\beta_{c}^{t})^{2} \rho^{4}}{4D_{lm}^{t}} - (\beta_{c}^{t}) \rho^{2}/2 \right] \right].$$
(B6b)

Here a distinction has been made between radial- and transverse-correlation coefficients where radial correlations typically are much stronger than the correlation of transverse displacements. The reason why we did not incorporate two independent coefficients above is that for the planar case, the effect of correlations is relatively small, and furthermore, the dependence of $\langle \hbar \Delta \omega^{(2)} \rangle_{\rm th}$ on the radial-correlation coefficient is very weak. It is thus tacitly understood that the transverse-correlation coefficient is to be inserted for $\beta_c(\mathbf{R}_i)$ in Eqs. (B5). One should also note that, apart from the inclusion of two correlation coefficients, the axial function $F_{\rm ax}$ is identical

to the planar one, F_{pl} , with obvious substitutions: The one-dimensional reciprocal-lattice vector $(\alpha - \beta)g$ should be replaced by the two-dimensional vector $(\mathbf{k} - \mathbf{k}')$, which is expressed in terms of the primitive basis vectors \mathbf{g}_1 and \mathbf{g}_2 as $(\mathbf{k} - \mathbf{k}') = (\alpha - \alpha')\mathbf{g}_1 + (\beta - \beta')\mathbf{g}_2$, and y_i should equal zero since in the axial case, the atomic string defines the z direction.

Here just a few points of the derivation of Eqs. (B5) and (B6) will be mentioned. The first factor in curly brackets in expressions (B5b) and (B6b) originates in the integration over ω and is based on the relation

$$\int_{-\infty}^{\infty} d\omega \frac{P}{\omega} \frac{\partial}{\partial \omega} \exp(-\omega^2 a) \exp(-i\omega z/v) = -\left[2\sqrt{\pi a} \exp\left[-\frac{(z/v)^2}{4a}\right] + \pi(z/v) \Phi\left[\frac{z/v}{2\sqrt{a}}\right]\right] \quad (a > 0) , \qquad (B7)$$

where the first term on the right-hand side is easily obtained from the differentiation of $\exp(-\omega^2 a)$. The last term is due to the differentiation of the factor $\exp(-i\omega z/v)$ and is obtained by noting that the Fourier transform of $\exp(-\omega^2 a)\exp(-i\omega z/v)$ is a displaced Gaussian. In the ω integration, we may then transfer the Fourier-transformation operator to the principal value, according to the definition of Fourier transforms of generalized functions (distributions), whereby the sign function results.

It is possible to reduce all the k integrations to the Gaussian Fourier transform,

$$\int_{-\infty}^{\infty} dk \, \exp(-k^2 a) \exp(-ikb) \\ = \sqrt{\pi/a} \, \exp(-b^2/4a) \quad (a > 0) , \quad (B8)$$

by noting that the factors $(k_{1x}k_{2x})$ and $(k_{1y}k_{2y})$ may be obtained by suitable differentiations of the integrands.

For the third-order thermal shift, explicit analytical expressions of the type (B5a) and (B5b) for planar channeling and without inclusion of thermal-vibrational correlations were derived in [27] for both the independent shifts of the two levels and for the coupling terms.

We quote here the result, in the present notation, which for the independent term (5.6) is

$$\langle \hbar \Delta \omega_{1}^{(3)} \rangle_{\text{th}} = -\frac{2}{3} \frac{1}{(2\pi)^{3}} \frac{N_{d} d_{p}}{(\hbar v)^{2}} \times \sum_{l,m,n} A_{l} A_{m} A_{n} \sum_{\alpha,\beta} C_{\alpha}^{1} C_{\beta}^{1} \bigg[[B_{l} B_{m} + B_{m} B_{n} + B_{n} B_{l}]^{-1} \exp[-\frac{1}{2} (\beta - \alpha)^{2} g^{2} \rho^{2}] \times \exp[-\frac{1}{4} (\beta - \alpha)^{2} g^{2} B_{l} B_{m} B_{n} (B_{l} B_{m} + B_{m} B_{n} + B_{n} B_{l})^{-1}] - 3[B_{l} B_{m} + B_{m} B_{n} + B_{n} B_{l} + 4\rho^{2} (B_{m} + B_{n})]^{-1} \times \exp[-\frac{1}{4} (\beta - \alpha)^{2} g^{2} (B_{m} B_{n} + 2\rho^{2} (B_{m} + B_{n})) \mathcal{B}_{l} \times (B_{l} B_{m} + B_{m} B_{n} + B_{n} B_{l} + 4\rho^{2} (B_{m} + B_{n}))^{-1}] + 2[\mathcal{B}_{l} \mathcal{B}_{m} + \mathcal{B}_{m} \mathcal{B}_{n} + \mathcal{B}_{n} \mathcal{B}_{l}]^{-1} \times \exp[-\frac{1}{4} (\alpha - \beta)^{2} g^{2} \mathcal{B}_{l} \mathcal{B}_{m} (\mathcal{B}_{l} \mathcal{B}_{m} + \mathcal{B}_{m} \mathcal{B}_{n} + \mathcal{B}_{n} \mathcal{B}_{l})^{-1}] \bigg],$$
(B9)

and similarly for the shift of the final state, $\langle \hbar \Delta \omega_0^{(3)} \rangle_{\text{th}}$. The shift of the transition frequency also contains two coupling terms with correlated scattering in states 1 and 0. The first term of Eq. (5.7) becomes

$$\left(\pi \Delta \gamma_{10}^{(3)} \right)_{\text{th}}^{I} = \frac{1}{\sqrt{\pi}} \frac{1}{(2\pi)^{3}} \frac{N_{d}d_{p}}{(\pi_{0})^{2}} \\ \times \sum_{l,m,n} A_{l} A_{m} A_{n} \sum_{\alpha,\beta,\alpha',\beta'} C_{\alpha}^{0} C_{\beta}^{0} C_{\alpha'}^{1} C_{\beta'}^{1} \exp\left[-\frac{1}{4}(\beta-\alpha)^{2}g^{2} \mathcal{B}_{l}\right] \\ \times \left[(\mathcal{B}_{m} + \mathcal{B}_{n})^{-1/2} \exp\left[-\frac{1}{4}(\beta'-\alpha')^{2}g^{2}(\mathcal{B}_{m}\mathcal{B}_{n}(\mathcal{B}_{m} + \mathcal{B}_{n})^{-1} + 2\rho^{2})\right] \\ \times \left\{ (\mathcal{B}_{l}\mathcal{B}_{m} + \mathcal{B}_{m}\mathcal{B}_{n} + \mathcal{B}_{n}\mathcal{B}_{l})^{-1/2} \exp\left[-(\beta-\alpha)(\beta'-\alpha')g^{2}\rho^{2}\right] \\ - (\mathcal{B}_{m}\mathcal{B}_{n} + \mathcal{B}_{l}(\mathcal{B}_{m} + \mathcal{B}_{n}))^{-1/2} \right\} \\ - 2(\mathcal{B}_{m} + \mathcal{B}_{n})^{-1/2} \exp\left[-\frac{1}{4}(\beta'-\alpha')^{2}g^{2}\mathcal{B}_{m}\mathcal{B}_{n}(\mathcal{B}_{m} + \mathcal{B}_{n})^{-1}\right] \\ \times \left\{ (\mathcal{B}_{l}\mathcal{B}_{n} + \mathcal{B}_{m}(\mathcal{B}_{l} + \mathcal{B}_{n}))^{-1/2} \\ \times \exp\left[(\beta-\alpha)g^{2}\rho^{2}(\mathcal{B}_{m} + \mathcal{B}_{n})^{-1}((\beta-\alpha)\rho^{2} - (\beta'-\alpha')\mathcal{B}_{m})\right] \\ - (\mathcal{B}_{l}\mathcal{B}_{m} + \mathcal{B}_{m}\mathcal{B}_{n} + \mathcal{B}_{n}\mathcal{B}_{l})^{-1/2} \right\} \right].$$
(B10)

TABLE I. Overlap with nuclei for 3.5-MeV electrons in Si, channeled along a major axis. The vibrational amplitudes at the two temperatures are $\rho = 0.0563$ and 0.0793 Å, respectively, and 441 beams were used in the calculation (the Fourier components included in the Bloch-wave expansion of the wave function are referred to as "beams").

	< 1	11>	< 10	00>
State	110 K	298 K	110 K	298 K
1 <i>s</i>	52	36	41	29
2 <i>p</i>	4.4	5.5	2.8	3.7

The second term is obtained by a change in the sign of this formula together with an interchange of the two states, i.e., $C^0_{\alpha}C^0_{\beta}C^1_{\alpha'}C^1_{\beta'} \rightarrow C^1_{\alpha}C^1_{\beta}C^0_{\alpha'}C^0_{\beta'}$.

APPENDIX C: NUMERICAL RESULTS

In this appendix, numerical values for line shifts are given for a few cases where line energies have been measured. The calculations are based on the analytical formulas in Appendix B, and the results are compared with simple estimates given in the text.

Axial channeling

We shall first consider channeling of 3.5-MeV electrons along $\langle 111 \rangle$ and $\langle 100 \rangle$ axes in silicon. The corrections calculated for the 2*p*-1*s* transition in the $\langle 111 \rangle$ axial potential have been used in [6], where accurate values of the vibrational amplitude were derived from measurements of channeling radiation.

As discussed in Appendix B, it is necessary to apply a modified set of Doyle-Turner coefficients in calculations of thermal scattering, and we have applied the values given in [21]. A more accurate potential was used to calculate continuum-model energies and wave functions, and two slightly different vibrational amplitudes were used for the atomic nuclei and for the surrounding electron clouds, but for the corrections, this is not important.

The simple estimates of second- and third-order thermal shifts (5.17), (5.19), and (5.23), are proportional to the overlap of the projectile flux with vibrating nuclei, and the calculated overlaps are given in Table I. Table II contains values of the width and of the third-order thermal shift for a plane-wave state. Within a factor of 2,

TABLE II. Level width and third-order level shift (in eV) for a plane wave, multiplied by $2\gamma^2$. In parentheses are given estimates (5.20) and (5.23), respectively.

Temperature	110 K	298 K
Third-order shift	0.21(0.26)	0.38(0.52)
Width	11(5)	16(10)

there is agreement with the simple estimates given in parentheses. In this and the following tables, all energies are in eV, except where noted, and we have multiplied the shifts given in the text by a factor $(1-\beta)^{-1} \simeq 2\gamma^2$ to obtain contributions to radiation line shifts, as given in Table V. The third-order corrections in this table have been calculated from the values given in Tables I and II, with a 30% reduction accounting approximately for correlation terms and for the overlap approximation.

In Table III, thermal second-order shifts are given for the individual states, partly for local scattering back and forth on one atom, partly for correlated scattering on neighboring atoms along a string. For the $\langle 111 \rangle$ axis, only the (one) nearest neighbor was included, while for the $\langle 100 \rangle$ axis, scattering on the two nearest atoms on the string was taken into account. The simple estimates given in parentheses are seen to be reasonably accurate.

Corrections from the periodic variation of the string potential along the axis are given in Table IV. The simple estimate (6.7) for a closely bound state is given in parentheses for the 1s state, and it reproduces somewhat accidentally—the values quite accurately. The values are also in agreement with results in [22], based upon a diagonalization of the Hamiltonian operator with three-dimensional periodicity. Finally, in Table V, all corrections to the 2p-1s transition energies are given. The total correction is quite small, of relative order 10^{-3} . An increase of the thermal second-order correction by a factor of ~ 2 , due to correlation with second- and third-nearest neighbors, would clearly not change this conclusion.

Planar channeling

We consider two cases, 4-MeV e^- on Ni {100} and {111}, corresponding to the measurements in [19], and 54-MeV e^- along {110} and {100} planes in Si, corre-

TABLE III. Thermal second-order shifts (in eV), from scattering on one atom (local) and from correlated scattering on neighboring atoms on a string (correl.). For the $\langle 111 \rangle$ axis, only the nearest neighbor at a distance of 2.35 Å was included, with correlation coefficients $\beta_c = 0.24$ (298 K) and $\beta_c = 0.18$ (110 K), while for the $\langle 100 \rangle$ axis, two neighbors at a distance of 5.43 Å were included, with correlation coefficient $\beta_c = 0.07$ (298 K) and $\beta_c = 0.05$ (110 K) (from Ref. [16]). Other parameters as for Table I. The values in parentheses are the estimates (5.17) and (5.19) multiplied by $2\gamma^2$.

	<1	11>	(10	00>
State and method	110 K	298 K	110 K	298 K
1s local	-1.3(1.7)	-1.2(1.7)	-1.1(1.4)	-1.0(1.4)
1s correl.	-2.7(4.1)	-2.0(3.7)	-2.5(4.1)	-2.0(4.1)
2p local	-0.15(0.15)	-0.21(0.26)	-0.09(0.09)	-0.15(0.17)
2p correl.	-0.33(0.34)	-0.38(0.57)	-0.25(0.28)	-0.32(0.52)

TABLE IV. Level shifts (in eV and multiplied by $2\gamma^2$) from periodic potential variation along the axis. In parentheses are given the estimate (6.7), for the $\langle 111 \rangle$ axis with the correction in (6.9) and below. Calculations performed with 625 beams.

TABLE V. Corrections (in eV) to the 2p-1s line energy for
3.5-MeV electrons in Si from calculation with 441 or 625 beams.
The thermal third-order correction was obtained from Tables I
and II, with an estimated reduction discussed in the text.

	<1	11>	〈100〉		
State	110 K	298 K	110 K	298 K	
1 <i>s</i>	8.6(6.5)	3.8(3.3)	4.5(3.7)	2.1(1.9)	
2 <i>p</i>	2.5	1.7	1.1	0.8	

	< 1	11>	〈100〉	
Correction	110 K	298 K	110 K	298 K
Continuum model	4577	3855	3902	3351
Thermal 2nd order	4	3	4	3
Thermal 3rd order	7	- 8	-6	-6
Periodic	-6	-2	-3	-1

TABLE VI. Overlap for 4-MeV electrons in Ni, channeled along two planes in the two lowest levels 0 and 1. The vibrational amplitudes are $\rho = 0.049$ Å (100 K), $\rho = 0.074$ Å (300 K), $\rho = 0.095$ Å (500 K), corresponding to a Debye temperature of 375 K [19] (13 beams).

		{100}			{111}		
State	100 K	300 K	500 K	100 K	300 K	500 K	
0	5.6	4.9	4.4	6.7	5.9	5.2	
1	0.44	0.80	1.1	0.59	1.1	1.4	

TABLE VII. Second-order thermal shifts (in eV) for 4-MeV electrons in Ni, for scattering back and forth on one atom. The values in parentheses are from the estimate (5.17), multiplied by $2\gamma^2$.

{ 100 }			{111}			
State	100 K	300 K	500 K	100 K	300 K	500 K
0	-1.0(1.3)	-1.2(1.8)	-1.2(2.0)	-1.2(1.6)	-1.4(2.1)	-1.5(2.4)
1	-0.11(0.11)	-0.23(0.29)	-0.34(0.51)	-0.15(0.14)	-0.31(0.40)	-0.44(0.65)

TABLE VIII. Transition energy and shifts (in eV) for radiation from 4-MeV electrons in Ni. The second-order thermal correction contains a contribution from correlated scattering on two adjacent atoms on a $\langle 110 \rangle$ string (compare with Table VII). The low- or high-temperature limits of the Debye correlation coefficient were used [28], $\beta_c = 0.14$ (100 K, 300 K) and $\beta_c = 0.36$ (500 K). In parentheses are given the estimates (5.17) and (5.23), multiplied by $2\gamma^2$.

	{100}			{111}		
Correction	100 K	300 K	500 K	100 K	300 K	500 K
Continuum model	4256	3932	3641	4824	4436	4093
Thermal (2nd) 4°	0.68(1.2)	1.0(1.5)	1.5(1.5)	0.80(1.5)	1.2(1.7)	1.8(1.8)
Thermal (3rd)	- 8.9(19)	-13(34)	-14(45)	-10(22)	-14(40)	-15(52)
Period 2°	-62	-15	4.7	-48	-4.7	12

4031

electrons in Si (13 beams).						
	{1	{110}				
State	110 K	298 K	298 K			
0	7.9	6.8	4.6			
1	1 0	26	1.6			

TABLE IX. Overlaps from planar channeling of 54-MeV electrons in Si (13 beams).

sponding to measurements by Park *et al.* [5]. For Ni, the Doyle-Turner coefficients given in [19] were used in the calculations.

The relevant overlaps are given in Tables VI and IX for the two cases. The accuracy of the overlap approximation for the second-order thermal shift (without correlation of vibrations) is analyzed in Table VII for the lowenergy case, and again the simple estimates agree within a factor of 2. In Table VIII, all corrections to the $1 \rightarrow 0$ transition energy are given. The second-order term here includes correlated scattering on neighboring atoms along a $\langle 110 \rangle$ axis in the plane, and the angles given in the first column are defined relative to this axis. At 4°, this contribution is similar in magnitude to that from local scattering on one atom. In the third-order terms, coupling terms have been included. The simple estimate is then too large by a factor of 2-3. For a plane wave, the estimate (5.23) is high by about 75% at room temperature.

Close to an axis, the largest correction is from the periodic string structure of the plane. For the {100} plane at room temperature, we can compare with [19], where the transition energy was calculated, with inclusion of nonsystematic Fourier components of the lattice potential. From Fig. 10 of [19], the correction at 2° is seen to be about -80 eV, which is much larger than the -15 eV given in Table VIII. However, this value is a difference between shifts of 125 and 110 eV for the two states 0 and 1, respectively. As noted in Sec. VI, the approximation (6.5) leading to the simple formula (6.6) and the analogous formula (6.11) may not be very accurate, in particular at some distance from the plane, where the state $|1\rangle$ has its maximum density. The near cancellation of the two shifts is reflected in the variable sign of the correction in Table VIII. The simple estimate (6.12) gives 30 eV, a useful estimate of the magnitude of the effect; also the predicted dependence on angle agrees well with Fig. 10 of [19]. At larger angles, all corrections are again quite small, of relative order 10^{-3} only.

Similar results are given for 54-MeV electrons in Si in Tables IX and X. The third-order term is proportional to γ^2 and is now the dominant correction. It is for these cases overestimated by a factor of 3-4 by the simple formula. This results from a combination of three effects:

TABLE X. Transition energy and shifts (in keV) for planarchanneling radiation from 54-MeV electrons in Si. The thermal scattering does not include correlations. In parentheses are given the estimates (5.17) and (5.23), multiplied by $2\gamma^2$.

	{1	{100}	
Method	110 K	298 K	298 K
Continuum model	145.1	128.7	96.9
Thermal (2nd)	2.1(2.7)	1.9(2.7)	1.3(1.9)
Thermal (3rd)	-90(286)	-85(397)	-76(284)
Period 2°	74	74	68

First, for a plane wave, the estimate is somewhat high, similar to the results in Table II. Second, the ratio between the shifts of states 1 and 0 is closer to $\frac{1}{2}$ than to the ratio $\sim \frac{1}{3}$ of the overlaps in Table IX, and, third, the coupling terms (B10) give a reduction by $\sim 30\%$. For the correction for periodic structure, there is again considerable cancellation, and the two individual shifts are larger by about a factor of 3. The estimate (6.12) gives 38 eV for the {100} and 67 eV for the {110} plane, in the latter case with a correction analogous to (6.9).

APPENDIX D: SUM RULE

We obtain the formula

$$\sum_{n} \langle i | \delta V(-\omega) | n \rangle \langle n | \delta V(\omega) | i \rangle \hbar \omega_{in}$$
$$= \frac{-\hbar^2}{2m\gamma} \langle i | [\nabla_{\perp} \delta V(-\omega)] \cdot [\nabla_{\perp} \delta V(\omega)] | i \rangle \quad (D1)$$

by writing the energy difference as a commutator which involves the stationary operator of the continuum model,

$$\hbar\omega_{in}\langle i|U|n\rangle\langle n|Y|i\rangle = \frac{1}{2}(\langle i|[H^{c},U]|n\rangle\langle n|Y|i\rangle - \langle i|U|n\rangle\langle n|[H^{c},Y]|i\rangle),$$

where U and Y are operators. Applying closure of intermediate states, making use of the fact that

1

$$[H^{c}, \delta V(-\omega)]|i\rangle = \frac{1}{2m\gamma} \{\mathbf{p}_{\perp} \cdot [\mathbf{p}_{\perp}, \delta V(-\omega)] + [\mathbf{p}_{\perp}, \delta V(-\omega)] \cdot \mathbf{p}_{\perp} \}|i\rangle$$
$$= \frac{1}{2m\gamma} \{\mathbf{p}_{\perp} \cdot (-i\hbar \nabla_{\perp} \delta V(-\omega)) + (-i\hbar \nabla_{\perp} \delta V(-\omega)) \cdot \mathbf{p}_{\perp} \}|i\rangle,$$

and noting that the left-hand side of (D1) is real and hence equals the average of itself and its complex conjugate, we may easily show Eq. (D1).

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