Line Shifts in Electron Channeling Radiation from Lattice Vibrations

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A density-matrix formulation of the dynamical theory of spectral shifts from correlated lattice vibrations in electron channeled radiation is presented. For lower-energy channeling, in which two bound levels exist in the channeling potential, positive shifts can be demonstrated. In the case of higher-energy channeling, where the number of bound levels increases, reduced positive shifts and negative shifts are shown. The theory suggests a novel method for experimentally determining a transverse, vibration correlation length in crystals.

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Relativistic electrons propagating through crystal axial or planar channels may occupy bound energy states in the transverse direction. Spontaneous transitions between these discrete, energy eigenstates give rise to narrow-width electromagnetic radiation that is strongly peaked in the forward direction.[1–14] The x-ray spectrum observed has consistently displayed a relatively simple structure to which a quantum mechanical analysis is well suited. Spectral linewidths due to lattice vibrations have been treated through a quantal, sudden-collision approximation[1,4] or by considering the imaginary part of an optical potential.[5,15] These studies have taken into account thermal scattering by uncorrelated individual atoms in order to explain the experimental observations of the linewidths.[4,5,7] In planar channeling, satisfactory agreement has been obtained in cases where the vibrations of different atoms are uncorrelated.[4] For axial channeling or for planar channeling close to a major crystal axis in the plane, the correlation of thermal scattering from neighboring atoms can be particularly important and must be included in the analysis of linewidths.[4,8,9] Recent experimental and theoretical studies have indicated reasonable agreement on the location of spectral peaks.[1–14] However, a consistent pattern of small spectral shifts has been seen with negative shifts occurring for axial channeling,[3,11] and both positive and negative shifts for planar channeling (17, 31, and 54 MeV) in LiF,[5] diamond, and silicon.[3,11] The standard remedy of varying the thermal vibrational amplitudes that appear in the thermally adjusted channel potentials was judged as unsatisfactory for always some energies.[5,6] An alternative theory that can account for these irregularities is desired. In this Letter, we use a density-matrix approach to show that the presence of spectral shifts may be partially related to an average, interstate dynamical effect due to the interaction of channeled electrons with correlated lattice vibrations. This treatment, which includes correlations of vibrations, provides a possible explanation for the spectral line shifts for both axial channeling and planar channeling close to a major axis in the plane. It is found that lower-energy channeling, in which two bound levels exist in the channeling potential, can have a positively shifted spectral line. At higher channeling energies the bound system becomes multilevel and reduced positive shifts and negative shifts are indeed predicted. It must be emphasized that the spectral properties derived here are a consequence of our having used a dynamical description of the electron-lattice vibration interaction in lieu of a thermal modification of the static channeling potential.

The transverse Hamiltonian for axial or planar channeling is obtained by first considering the crystal potential $V(\mathbf{r},z)=\sum_l V_a(\mathbf{R}-\mathbf{R}_l-\mathbf{U}_l)$, where $V_a(\mathbf{R})$ is the atomic potential, $\mathbf{U}_l$ is the thermal displacement of the atom from site $\mathbf{R}_l$, $\mathbf{R}=(\mathbf{r},z)$, and $\mathbf{r}=(x,y)$. Here the coordinate z is chosen to lie along the channeling direction, while for planar channeling x (y) is normal (parallel) to the channeling plane. Upon averaging of the local oscillations in $V(\mathbf{r},z)$ along the z axis and expression of z as a timelike parameter $t=z/v$ (where v is the electron channeling velocity), the transverse Hamiltonian for a channeled electron bound in a potential $V(\mathbf{r},t)$ can be written as $H_t^0=H_t^{(0)}+H_t^{(1)}$, where $H_t^{(0)}=H_f+V_T(\mathbf{r})$, $H_t^{(1)}=V(\mathbf{r},t)-V_T(\mathbf{r})$, H_f is the free-particle Hamiltonian, and the channeling potential is $V_T(\mathbf{r})\equiv\langle\langle V(\mathbf{r},t)\rangle\rangle_z$. The double angular brackets denote an ensemble average over $\mathbf{U}_l$ and the subscript z denotes an average over the z dependence. In case of planar channeling $V_T(\mathbf{r})\rightarrow V_T(x)$ after averaging also over the y dependence. As considered by Andersen *et al.*,[4] $H_t^{(1)}$ can be approximated as

$$H_t^{(1)}\simeq V(\mathbf{r},)-\langle\langle V(\mathbf{r},t)\rangle\rangle\equiv\delta V(\mathbf{r},t).$$

In a representation where $H_t^{(0)}$ is diagonal, and for the lower bound levels, the matrix Hamiltonian simply becomes $H_{ij}^0=\epsilon_i^0\delta_{ij}-\hbar\Omega_{ij}^0$, where $\epsilon_i^0\delta_{ij}=H_{ij}^{(0)}$, $\hbar\Omega_{ij}^0=\delta V_{ij}$, $\delta V_{ij}=\langle i|\delta V|j\rangle$, the transverse subscript is dropped, and zero superscripts indicate laboratory-frame quantities. For planar channeling ϵ_i^0 includes the free states with different y momenta. Noting that $\langle\langle\Omega_{ij}^0\rangle\rangle=0$, we next introduce a spatial correlation along the z direction in the laboratory frame at a given time,[16]

$$\langle\langle\Omega^0(z')\Omega^0(z'')\rangle\rangle=\langle\langle[\Omega^0(0)]^2\rangle\rangle F(|z'-z''|/L_p^0),$$

where F denotes a general correlation function which is a decreasing function of the argument $|z'-z''|/L_p^0$, $F(0)=1$. Here L_p^0 is defined as a transverse-vibration correlation length and the various correlations in vibrations are taken up to several lattice spacings. Here, we remark that a further ensemble average of H_{ij}^0 at this stage would only reproduce the previous technique of thermally modifying the channel potential and the results derived therefrom.[1,14]

Upon Lorentz transformation in the beam direction, the Hamiltonian in the particle frame is given by[17] $H_{ij}=\gamma H_{ij}^0=\epsilon_i\delta_{ij}-\hbar\Omega_{ij}$, where γ is the relativistic factor $(1-v^2/c^2)^{-1/2}$. In this frame, the transverse electron wave function may be written as a complete sum over eigenstates, $|\Psi\rangle=\sum_i C_i|i\rangle$, whereupon the density-matrix elements ρ_{ij} are given by $C_iC_j^*$. These elements evolve in time according to $i\hbar\, d\rho_{ij}/dt=(H\rho-\rho H)_{ij}$. Defining $\omega_i=\epsilon_i/\hbar$, $\omega_{ij}=\omega_i-\omega_j$, using the definition of H_{ij}, and separating $\rho_{ij}(t)$ into slowly and rapidly varying factors, $\rho'_{ij}(t)\exp(-i\omega_{ij}t)$, we find that ρ'_{ij} satisfies

$$\rho'_{ij}(t)=\rho'_{ij}(0)+i\sum_k\int_0^t dt'\,[\Omega_{ik}(t')\rho'_{kj}(t')\exp(i\omega_{ik}t')-\rho'_{ik}(t')\Omega_{kj}(t')\exp(i\omega_{kj}t')]. \tag{1}$$

If we iterate and perform an ensemble average Eq. (1) becomes

$$\begin{aligned}\langle\langle\rho'_{ij}(t)\rangle\rangle=\langle\langle\rho'_{ij}(0)\rangle\rangle-\sum_{k,k'}\int_0^t dt'\int_0^{t'}dt''\,F(|t'-t''|/\tau_p)\\ \times\{[\Omega^2_{ikkk'}\langle\langle\rho'_{k'j}(t'')\rangle\rangle\exp(i\omega_{ik}t'+i\omega_{kk'}t'')-\Omega^2_{ikk'j}\langle\langle\rho'_{kk'}(t'')\rangle\rangle\exp(i\omega_{ik}t'+i\omega_{k'j}t'')]\\ -[\Omega^2_{ik'kj}\langle\langle\rho'_{k'k}(t'')\rangle\rangle\exp(i\omega_{kj}t'+i\omega_{ik'}t'')-\Omega^2_{k'kkj}\langle\langle\rho'_{ik'}(t'')\rangle\rangle\exp(i\omega_{kj}t'+i\omega_{k'k}t'')]\},\end{aligned} \tag{2}$$

where $Q^2_{ijkl}=\gamma^2\langle\langle\delta V_{ij}\delta V_{kl}\rangle\rangle/\hbar^2$, $\tau_p=L_p/c$, $L_p=L_p^0/\gamma$, and

$$\langle\langle\Omega(t')\Omega(t'')\rho'(t')\rangle\rangle\cong\langle\langle\Omega(t')\Omega(t'')\rangle\rangle\langle\langle\rho'(t')\rangle\rangle.$$

Dropping the double angular brackets, taking the time derivative of Eq. (2), and noting that the dominant contribution to the t'' integration occurs for $t''\sim t$, we find

$$\frac{d}{dt}\rho_{ij}(t)=-i\omega_{ij}\rho_{ij}(t)-\sum_k\left(\frac{\Omega^2_{ikki}}{i\tilde{\omega}_{ki}}+\frac{\Omega^2_{jkkj}}{i\tilde{\omega}_{jk}}\right)\rho_{ij}(t)+\sum_{k,k'}\Omega^2_{ikk'j}\left(\frac{1}{i\tilde{\omega}_{k'j}}+\frac{1}{i\tilde{\omega}_{ik}}\right)\delta_{\omega_{ik'},-\omega_{k'j}}\rho_{kk'}(t), \tag{3}$$

where $\rho'_{ij}(t)$ has been transformed back to $\rho_{ij}(t)$, $\delta_{\omega_{ik'},-\omega_{k'j}}$ is the Kronecker δ and arises from a time average over the oscillatory terms,

$$(i\tilde{\omega}_{ij})^{-1}\equiv\left[\int_0^t d\tau\,F(|\tau|/\tau_p)\exp(-i\omega_{ij}\tau)\right]_t,$$

and the subscript t denotes a time average over oscillatory terms. Alternatively, $i\tilde{\omega}_{ij}$ may be represented in the form $i\tilde{\omega}_{ij}=i\alpha_{ij}\omega_{ij}+\beta_{ij}/\tau_p$. For some physical cases of interest $\hbar\omega_{ij}^0$ is of the order of a few electronvolts or less and it is a good approximation to take $(\omega_{ij}\tau_p)^2<<1$. In such a case α_{ij} and β_{ij} are of the order of 1 for *general* F and they are nearly insensitive to the quantum indices i,j. For an exponential correlation function $\alpha_{ij}=\beta_{ij}=1$. For $i=j$, the population in state $|i\rangle$, ρ_{ii}, satisfies

$$d\rho_{ii}/dt=-\sum\nolimits_k(\rho_{ii}-\rho_{kk})/\tau_{ik}(1),$$

where $\tau_{ik}^{-1}(1)\equiv A_{ik}\beta_{ik}\tau_p^{-1}$, and

$$A_{ik}=2\Omega^2_{ikki}/[(\alpha_{ik}\omega_{ik})^2+(\beta_{ik}/\tau_p)^2].$$

Physically, $\tau_{ik}(1)$ represents the decay time of ρ_{ii} from lattice vibrations when a population imbalance between states $|i\rangle$ and $|k\rangle$ occurs.

For electron channeling the transition frequencies are nondegenerate, i.e., $\omega_{ik}\neq\omega_{jk'}$ for all distinct transitions. Eq. (3) then for $i\neq j$ assumes the suggestive form

$$\begin{aligned}&d\rho_{ij}(t)/dt\\ &\quad=-i(\omega_{ij}+\Delta_{ij})\rho_{ij}(t)-\rho_{ij}(t)/\tau_{ij}(2),\end{aligned} \tag{4}$$

where $\hbar\Delta_{ij}$ and $\hbar\tau_{ij}^{-1}(2)$ correspond to energy shifts and widths, respectively. Making a Lorentz transformation back to the laboratory frame via the relations $\omega_{ij}\tau_{ij}(2)=\omega_{ij}^0\tau_{ij}^0(2)$, $\omega_{ij}\tau_p=\omega_{ij}^0\tau_p^0$, $\Delta_{ij}=\gamma\Delta_{ij}^0$, $\omega_{ij}=\gamma\omega_{ij}^0$, and $A_{ij}=A_{ij}^0$, and Doppler correcting by the factor $2\gamma^2$ for the photon shifts and widths, we obtain

$$\bar{\Delta}_{ij}=\tfrac{1}{2}\sum\nolimits_k(\alpha_{ik}\bar{\omega}_{ik}A_{ik}^0+\alpha_{kj}\bar{\omega}_{kj}A_{jk}^0), \tag{5}$$

$$\bar{\Gamma}_{ij}\equiv2\gamma^2/\tau_{ij}^0(2)=\gamma^2\sum\nolimits_k(\beta_{ik}A_{ik}^0+\beta_{jk}A_{jk}^0)/\tau_p^0, \tag{6}$$

where overbars denote Doppler-corrected frequencies in the laboratory frame, and zero superscripts again indicate laboratory-frame quantities. The sums in Eqs. (5) and (6) are over all intermediate (bound and free) states k and are due to thermal transitions starting at states i and j.

For planar channeling close to a major crystal axis in the plane, the y separation of neighboring atoms is small and highly correlated.[4] One can then use in Eqs. (5) and (6) the completeness of the continuum set of y-motion wave functions to obtain for the photon shifts and widths the same form. However, the matrix elements in Eqs. (5) and (6) will now involve x-motion wave functions only with the y dependence in Ω^2_{ijji} averaged. For two bound levels in the channeling potential the photon shift and width in the case of axial or planar channeling follow directly from Eqs. (5) and (6):

$$\frac{\bar{\Delta}_{10}}{\bar{\omega}_{10}} = A^0_{10}\alpha_{10}, \quad \frac{\bar{\Gamma}_{10}}{\bar{\omega}_{10}} = \frac{A^0_{10}\beta_{10}c}{\omega^0_{10}L^0_p}, \tag{7}$$

where a strictly *positive* shift is to be noted. Although this feature qualitatively agrees with some lower-energy planar-channeling experimental results,[5,11] future experiments should investigate this effect as a function of beam incidence relative to a major axis in the plane. In the case of LiF, for example, one can obtain from Eq. (7) a relative shift $\bar{\Delta}_{10}/\bar{\omega}_{10}$ of a few percent when $\eta \equiv \alpha_{10}L^0_p/\beta_{10}a_0 \sim 4$, where a_0 is the lattice constant and the experimental values of $\bar{\Gamma}_{10}/\bar{\omega}_{10} \sim 20\%$, $\hbar\bar{\omega}_{10} \sim 20$ keV are taken from Ref. 5. For planar channeling close to a crystal axis or axial channeling the correlation length L^0_p can be several lattice spacings[4,8,9] and $\eta \sim 4$. A more definitive comparison necessarily entails a highly model-specific analysis of L^0_p, α_{ij}, and β_{ij}, that is appropriate for further studies. From Eq. (7) it is seen that the ratio of the shift to width is directly related to the microscopic quantity L^0_p. As this correlation length in the transverse direction depends only on the channeling direction, a straightforward way of measuring L^0_p as a function of crystal orientation is indicated. Finally, a direct measure of the strength of the potential, i.e., through A^0_{10}, is provided through measurement of the spectral shift.

For multiple bound levels in the channeling potential the shift in Eq. (5) may be explicitly written as

$$\frac{\bar{\Delta}_{j+1,j}}{\bar{\omega}_{j+1,j}} = A^0_{j+1,j} - \frac{[\bar{\omega}_{j+2,j+1}A^0_{j+2,j+1} + \bar{\omega}_{j,j-1}A^0_{j,j-1}]}{2\bar{\omega}_{j+1,j}}, \tag{8}$$

where terms proportional to $A^0_{i,j}$ for $|i-j| \geqslant 2$ are negligibly small and have been ignored, and α_{ij} and β_{ij} are taken as unity for simplicity. It is noticed that relative to the two-bound-state case, the shifts are significantly reduced and may even become negative. This qualitatively corresponds to the higher-energy electron planar-channeling results where the bound system is multilevel and negative shifts are generally seen.[3,5,11] With use of the thermally averaged exponential channel potential $V_T(x)$ of Pantell and Swent,[14] for example, and with the approximation $\Omega^2_{ij} \simeq (\gamma V'_{ij}/\hbar)^2 \times \langle\langle U^2_x \rangle\rangle$, where U_x is the atomic thermal displacement and $V'_{ij} = \langle i | dV_T(x)/dx | j \rangle$, a quantitative analysis shows that $A_{j,j+1}/A_{j-1,j}$ behaves as $(\omega_{j,j+1}/\omega_{j-1,j})^2$. When this relation is used in Eq. (8), for LiF,[5] diamond,[3] and silicon,[14,18] negative shifts are indeed seen to occur for all transitions except for the 1 → 0 case. When this relation is also used in Eq. (6) for the spectral widths, a general decrease in width with decreasing transition frequency is verified.

This density-matrix formulation of electron channeling in the presence of correlated lattice vibrations has simultaneously provided a general description of line shifts, widths, and population decay rates due to interstate transitions. In particular, we have shown the possibility of positive shifts in the two-bound-level case that can exist in lower-energy electron channeling. Further increasing of the channeling electron energy gives rise to multiple bound levels where reduced positive shifts or negative shifts were found. Whereas general agreement between theoretically predicted and observed spectral maxima was previously found with use of a thermally averaged, static, channeling potential, an understanding of these finer features, e.g., positive and negative shifts, may be partially furthered by a dynamical description.

A detailed phonon representation of lattice vibrations and their effect on line shifts and widths is in preparation. Additional refinement of the line shifts requires an analysis of the dynamic effects of the bound- to free-state transitions. Work on this area within the context of both electron and positron channeling is in progress.

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[1] J. U. Andersen, E. Bonderup, and R. H. Pantell, Annu. Rev. Nucl. Part. Sci. **33**, 453 (1983).

[2] R. L. Swent, R. H. Pantell, M. J. Alguard, B. L. Berman, S. D. Bloom, and S. Datz, Phys. Rev. Lett. **43**, 1723 (1979).

[3] S. Datz, R. W. Fearick, H. Park, R. H. Pantell, R. L. Swent, J. O. Kephart, R. K. Klein, and B. L. Berman, Phys. Lett. **96A**, 314 (1983); R. K. Klein *et al.*, "Electron Channeling From Diamond," to be published.

[4]J. U. Andersen, E. Bonderup, E. Laegsgaard, and A. H. Sorensen, Phys. Scr. **28**, 308 (1983).

[5]R. L. Swent, R. H. Pantell, H. Park, J. O. Kephart, R. K. Klein, S. Datz, R. W. Fearick, and B. L. Berman, Phys. Rev. B **29**, 52 (1984); R. L. Swent, Ph.D. Dissertation, Stanford University, 1982 (unpublished).

[6]B. L. Berman *et al.*, University of California, Lawrence Radiation Laboratory Report No. 89454, 1983 (unpublished).

[7]V. A. Bazylev and V. V. Goloviznin, Radiat. Eff. **60**, 101 (1982).

[8]J. U. Andersen *et al.*, Nucl. Instrum. Methods **194**, 209 (1982).

[9]O. H. Nielsen and W. Weber, J. Phys. C **13**, 2449 (1980).

[10]J. U. Andersen *et al.*, Phys. Rev. Lett. **49**, 215 (1982).

[11]H. Park, J. O. Kephart, R. K. Klein, R. L. Swent, and R. H. Pantell, "Electron Channeling Radiation Measurements," to be published.

[12]J. U. Andersen, K. R. Eriksen, and E. Laegsgaard, Phys. Scr. **24**, 588 (1981).

[13]H. J. Alguard *et al.*, IEEE Trans. Nucl. Sci. **26**, 3865 (1979); R. H. Pantell *et al.*, IEEE Trans. Nucl. Sci. **30**, 3150 (1983).

[14]R. H. Pantell and R. L. Swent, Appl. Phys. Lett. **35**, 910 (1979).

[15]P. H. Dederichs, Solid State Phys. **27**, 135 (1972).

[16]A. A. Maradudin *et al.*, *Theory of Lattice Dynamics in the Harmonic Approximation* (Academic, New York, 1971).

[17]D. M. Bird and B. F. Buxton, Proc. Roy. Soc. London, Ser. A **379**, 459 (1982).

[18]A. W. Saenz, H. Überall, and Anton Nagl, Nucl. Phys. **A372**, 90 (1981).