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## Valence-band plasmon effects on line shifts and widths in positron planar-channeling radiation

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A dynamical description of spectral shifts and widths of positron planar-channeled radiation is presented. The presence of well-defined valence electron collective oscillations or plasmons as documented in electron-loss spectroscopy is proposed as one mechanism for experimentally observed negative shifts. Qualitative comparison with the experimental data at 55 MeV for diamond, silicon, and LiF is indicated.

### I. INTRODUCTION

Relativistic planar-channeled positrons in crystals may occupy bound energy eigenstates in the transverse direction. Spontaneous transitions between these discrete states yield intense, narrow-width, strongly forward-peaked x-ray radiation.<sup>1,2</sup> Channeling radiation from positrons of approximately 55 MeV has been observed in silicon, diamond, germanium, and LiF.<sup>3-6</sup> According to the harmonic potential model of Pantell and Alguard,<sup>2</sup> a single spectral envelope should be observed. This composite structure results from a set of closely spaced spectral lines arising from weak anharmonicity and the associated natural line-broadening mechanisms. In most experiments to date on positron channeling,<sup>3-6</sup> such a structure has indeed been observed but with a perceptible negative shift. This paper examines these negative shifts, as well as the accompanying spectral widths, from a dynamical approach by using a density-matrix treatment.

We have recently shown that in the case of high-energy electron planar channeling, the presence of *nondegenerate* transition frequencies can yield negatively shifted spectral lines.<sup>7</sup> Specifically, each bound eigenstate effectively repels the adjacent level through intermediate or virtual transitions by an amount significantly depending on the bound electron energy level. In the case of positron channeling, however, the transition frequencies are nearly degenerate, and the effects of intermediate bound-bound (*b-b*) transitions on spectral shifts nearly cancel. This motivates our investigation here of the influence of intermediate bound-free (*b-f*) transitions on spectral line shifts.

In previous studies of electron planar channeling, the electric field fluctuations associated with the thermal motion of the ions, i.e., phonons, were emphasized because the channeled electrons are localized near the ionic planes.<sup>1,7</sup> For positron channeling, however, the relevant electric field fluctuations are principally associated with the valence electrons since the positrons are confined near the ionic midplanes.<sup>8-10</sup> Electron-loss spectroscopic (ELS) studies have documented that the valence electrons collectively behave in the form of relatively long-lived plasmon excitations for the crystalline samples we shall

consider.<sup>9</sup> These experimental results are of essential importance in our identification of the electric field fluctuations with coherent plasmon phenomena. Because the samples are ordinarily around room temperature ( $\frac{1}{40}$  eV) or less and the valence plasmon energies are typically greater than 15 eV, the valence electrons have only zero-point collective motions with plasmon occupation number zero. As the channeling positron traverses the crystal, virtual processes result where plasmons are created and subsequently annihilated. The effect of these virtual transitions on the spectral characteristics of the channeling radiation is to yield dynamical line shifts and widths. These are understood most conveniently through a density-matrix formulation which we employ here.<sup>11</sup> For the three low-*Z* samples to which we apply this simple formalism, qualitative agreement with the experimentally observed negative shifts and widths is found. In the case of higher-*Z* elements such as germanium<sup>3</sup> or tungsten (W) (Ref. 12), the presence of more complicated band structures prevents us from reliably applying the theory as presented here.

The paper is organized as follows. In Sec. II a density-matrix equation of motion is obtained for the channeling positrons interacting with the valence electrons of the crystal. In Sec. III the line shifts and widths of the channeling radiation are obtained from the ensemble-averaged density-matrix equation of motion. Section IV uses a simplified density fluctuation model for comparing our results with the experimental data at 55 MeV for diamond, silicon, and LiF.

### II. THE DENSITY-MATRIX EQUATION OF MOTION

We consider a system of channeling positrons interacting with the valence electrons of the crystal. The Hamiltonian for the channeling positrons may be written as

$$H = H_0 + \delta V, \quad (1)$$

where  $H_0$  describes the channeling in the ensemble-averaged planar potential:

$$H_0 = \sum_{\alpha, \mathbf{q}} \hbar[\omega_{\alpha} + \omega(\mathbf{q})] a_{\alpha}^{\dagger}(\mathbf{q}, t) a_{\alpha}(\mathbf{q}, t). \quad (2)$$

Here  $a_\alpha^\dagger(\mathbf{q}, t)$  [ $a_\alpha(\mathbf{q}, t)$ ] is the creation (annihilation) operator of the transverse state  $\alpha$  in the  $x$  direction with momentum  $\mathbf{q}$  in the  $y$ - $z$  plane,  $z$  is the main channeling direction, and  $\omega_\alpha$  and  $\omega(\mathbf{q})$  represent the directional positron energy.<sup>13</sup> The perturbation term  $\delta V$  represents the interaction potential of the valence electron density oscillations or plasmons and the positrons:

$$\delta V = \int d\mathbf{r} \int d\mathbf{r}' V(|\mathbf{r}-\mathbf{r}'|) \delta n_v(\mathbf{r}') n_c(\mathbf{r}), \quad (3)$$

where  $n_v(\mathbf{r})$  and  $n_c(\mathbf{r})$  are the density operators for valence electrons and channeling positrons, respectively.<sup>13</sup> The fluctuating valence electron density operator  $\delta n_v(\mathbf{r})$  is given by

$$\delta n_v(\mathbf{r}) = n_v(\mathbf{r}) - \langle n_v(\mathbf{r}) \rangle, \quad (4)$$

where angular brackets denote an ensemble average. We assume in Eq. (4) that  $\langle n_v(\mathbf{r}) \rangle \simeq \langle n_v(\mathbf{r}) \rangle_p$ , where  $\langle \rangle_p$  indicates an ensemble and planar average, independent of the low-current positron beam.<sup>14</sup>

Using the field operators  $\Psi_v(\mathbf{r})$  and  $\Psi_c(\mathbf{r})$  of the valence and channeling particles, we may rewrite the density operators in second quantized language:

$$n_v(\mathbf{r}) = \Psi_v^\dagger(\mathbf{r}) \Psi_v(\mathbf{r}), \quad n_c(\mathbf{r}) = \Psi_c^\dagger(\mathbf{r}) \Psi_c(\mathbf{r}). \quad (5)$$

In the momentum representation  $\Psi_v(\mathbf{r})$  is given by

$$\Psi_v(\mathbf{r}) = \sum_{\mathbf{q}} \zeta_{\mathbf{q}}(\mathbf{r}) b_{\mathbf{q}}(t), \quad (6)$$

where  $b_{\mathbf{q}}^\dagger$  ( $b_{\mathbf{q}}$ ) is the creation (annihilation) operator for a valence electron with momentum  $\mathbf{q}$ .<sup>13</sup> The wave function  $\zeta_{\mathbf{q}}(\mathbf{r})$  is chosen as<sup>15</sup>

$$\zeta_{\mathbf{q}}(\mathbf{r}) = \frac{1}{\sqrt{N_0}} \sum_l e^{i\mathbf{q} \cdot \mathbf{R}_l} W(\mathbf{r} - \mathbf{R}_l), \quad (7)$$

where  $N_0$  is the number of lattice points, and  $W(\mathbf{r} - \mathbf{R}_l)$  is the Wannier function localized around the lattice site  $\mathbf{R}_l$ . The Wannier function is related to the atomic wave function and forms an orthonormal set. For the channeling positron we use

$$\Psi_c(\mathbf{r}) = \sum_{\alpha, \mathbf{p}} e^{i\mathbf{p} \cdot \boldsymbol{\rho}} \zeta_\alpha(x) a_\alpha(\mathbf{p}, t), \quad (8)$$

where  $\zeta_\alpha(x)$  is the transverse wave function and  $\boldsymbol{\rho}$  is the projection of  $\mathbf{r}$  onto the  $(y, z)$  plane. For a bound (free) positron,  $\zeta_\alpha(x)$  is the one-dimensional harmonic oscillator (plane) wave function.<sup>16</sup>

In the  $y$ - $z$  plane the wave function is a plane wave with momentum  $\mathbf{p} = (p_y, p_z)$ , and the crystal area in the  $y$ - $z$  plane as well as the crystal volume are taken as unity for simplicity. Using the Fourier expansion,

$$V(|\mathbf{r}-\mathbf{r}'|) = \sum_{\mathbf{Q}} V(\mathbf{Q}) e^{i\mathbf{Q} \cdot (\mathbf{r}' - \mathbf{r})},$$

substituting Eqs. (5), (6), and (8) in Eq. (3) and taking  $\mathbf{k} \rightarrow -\mathbf{k}$ , the interaction potential  $\delta V$  in the momentum representation becomes

$$\delta V = \sum_{\substack{\mathbf{p}, \mathbf{k} \\ \alpha, \alpha'}} \hbar \Omega_{\alpha\alpha'}(\mathbf{k}, t) a_{\alpha'}^\dagger(\mathbf{p} + \mathbf{k}, t) a_\alpha(\mathbf{p}, t), \quad (9)$$

where

$$\hbar \Omega_{\alpha\alpha'}(\mathbf{k}, t) = V_{\alpha\alpha'}(\mathbf{k}) \delta n_{\mathbf{k}}(t), \quad (10)$$

and

$$V_{\alpha\alpha'}(\mathbf{k}) = V(k) F(\mathbf{k}) \int dx e^{ik_x x} \zeta_\alpha^*(x) \zeta_{\alpha'}(x). \quad (11)$$

In Eq. (10),  $\delta n_{\mathbf{k}}$  is the density fluctuation operator for the valence electrons given by

$$\delta n_{\mathbf{k}}(t) = n_{\mathbf{k}}(t) - \langle n_{\mathbf{k}}(t) \rangle,$$

where

$$n_{\mathbf{k}}(t) = \sum_{\mathbf{q}} b_{\mathbf{q}}^\dagger(t) b_{\mathbf{q}+\mathbf{k}}(t).$$

In addition,

$$\Omega_{\alpha\alpha'}^\dagger(\mathbf{k}, t) = \Omega_{\alpha\alpha'}(-\mathbf{k}, t), \quad \delta n_{\mathbf{k}}^\dagger(t) = \delta n_{-\mathbf{k}}(t), \quad (12)$$

and  $F(\mathbf{k})$  is a form factor [ $|F(\mathbf{k})|^2 \sim 1$ ] given by

$$F(\mathbf{k}) = \int d\mathbf{r} W^*(\mathbf{r}) W(\mathbf{r}) e^{-i\mathbf{k} \cdot \mathbf{r}}. \quad (13)$$

The density fluctuation operator  $\delta n_{\mathbf{k}}$  represents the creation and annihilation operators for plasmons with momentum  $\mathbf{k}$  and energy  $\hbar\omega_p$ , where  $\omega_p = (4\pi e^2 n_e / m_e)^{1/2}$  is the plasma frequency.<sup>17</sup> The density  $n_e$  is the average valence electron density and  $m_e$  and  $-e$  are the mass and charge of the electron, respectively. Also,  $\delta n_{\mathbf{k}}$  can be explicitly represented as

$$\delta n_{\mathbf{k}}(t) = c_{\mathbf{k}}(t) + c_{-\mathbf{k}}^\dagger(t), \quad (14)$$

where  $c_{\mathbf{k}}^\dagger$  ( $c_{\mathbf{k}}$ ) is the creation (annihilation) operator of a plasmon. Thus the valence electron plasmon free Hamiltonian  $H_p^0$  becomes

$$H_p^0 = \sum_{\mathbf{k}} \hbar\omega_p [c_{\mathbf{k}}^\dagger(t) c_{\mathbf{k}}(t) + \frac{1}{2}]. \quad (15)$$

The density matrix for the positrons is defined as<sup>11</sup>

$$\rho_{ij}(\mathbf{p}', \mathbf{p}, t) = a_i^\dagger(\mathbf{p}', t) a_j(\mathbf{p}, t). \quad (16)$$

Using the usual anticommutation relations for the positrons

$$[a_i(\mathbf{p}', t), a_j^\dagger(\mathbf{p}, t)]_+ = \delta_{ij} \delta_{\mathbf{p}', \mathbf{p}},$$

$$[a_i(\mathbf{p}', t), a_j(\mathbf{p}, t)]_+ = 0,$$

and the Heisenberg equation of motion,

$$i\hbar \frac{\partial}{\partial t} \rho_{ij}(\mathbf{p}', \mathbf{p}, t) = [\rho_{ij}(\mathbf{p}', \mathbf{p}, t), H], \quad (17)$$

together with the Hamiltonian [Eqs. (1), (2), and (9)], a kinetic equation for the channeling positrons is obtained:

$$\begin{aligned} \frac{\partial}{\partial t} \rho_{ij}(\mathbf{p}', \mathbf{p}, t) = & i[\omega_{ij} + \omega(\mathbf{p}', \mathbf{p})] \rho_{ij}(\mathbf{p}', \mathbf{p}, t) \\ & + i \sum_{\mathbf{k}, \alpha} [\Omega_{\alpha i}(\mathbf{k}, t) \rho_{\alpha j}(\mathbf{p}' + \mathbf{k}_1, \mathbf{p}, t) \\ & - \rho_{i\alpha}(\mathbf{p}', \mathbf{p} - \mathbf{k}_1, t) \Omega_{j\alpha}(\mathbf{k}, t)], \quad (18) \end{aligned}$$

where

$$\omega_{ij} = \omega_i - \omega_j, \quad \omega(\mathbf{p}', \mathbf{p}) = \omega(\mathbf{p}') - \omega(\mathbf{p}), \quad (19)$$

and  $\mathbf{p}', \mathbf{p}, \mathbf{k}_1$  are all taken to be in the  $y$ - $z$  plane ( $\mathbf{k}_1 = \mathbf{k} - k_x \hat{\mathbf{x}}$ ).

In Sec. III we develop an equation of motion for the ensemble-averaged density matrix and obtain expressions for the channeling radiation line shifts and widths.

### III. LINE SHIFTS AND WIDTHS OF CHANNELING RADIATION

Of primary interest is the time evolution of the quasi-static portion  $\rho'_{ij}(\mathbf{p}', \mathbf{p}, t)$  of the ensemble-averaged density matrix, where

$$\rho_{ij}(\mathbf{p}', \mathbf{p}, t) \simeq \rho'_{ij}(\mathbf{p}', \mathbf{p}, t) e^{i[\omega_{ij} + \omega(\mathbf{p}', \mathbf{p})]t}. \quad (20)$$

We need to find the oscillatory dependence of the operators in Eq. (18). For the plasmon operators  $c_{\mathbf{k}}(t)$  and  $c_{\mathbf{k}}^\dagger(t)$ , the oscillatory behavior is obtained using the equation of motion [Eq. (17)] with the Hamiltonian  $H_p^0$  [Eq. (15)] and the anticommutation relations to yield directly, for  $k_z > 0$ ,

$$c_{\mathbf{k}}(t) \sim e^{-i\omega_p t}, \quad c_{\mathbf{k}}^\dagger(t) \sim e^{i\omega_p t}. \quad (21)$$

For a collective mode of energy  $\hbar\omega_p$ , changing  $\mathbf{k} \rightarrow -\mathbf{k}$  corresponds to  $\omega_p \rightarrow -\omega_p$  from a reality consideration of the physical fields.<sup>18</sup> Thus for all  $\mathbf{k}$  we can write

$$c_{\mathbf{k}}(t) \sim e^{-i\eta_{\mathbf{k}}\omega_p t}, \quad c_{\mathbf{k}}^\dagger(t) \sim e^{i\eta_{\mathbf{k}}\omega_p t}, \quad (22)$$

where

$$\eta_{\mathbf{k}} = \begin{cases} 1, & k_z > 0 \\ -1, & k_z < 0. \end{cases}$$

The operators appearing in Eq. (18) are  $\Omega(\mathbf{k}, t)\rho_{ij}(\mathbf{p}' + \mathbf{k}_1, \mathbf{p}, t)$  and  $\Omega(\mathbf{k}, t)\rho_{ij}(\mathbf{p}', \mathbf{p} - \mathbf{k}_1, t)$ . From Eqs. (10) and (14), we need in particular to find the oscillatory part of the operators:

$$\begin{aligned} & c_{\mathbf{k}}(t)\rho_{ij}(\mathbf{p}' + \mathbf{k}_1, \mathbf{p}, t), \quad c_{\mathbf{k}}^\dagger(t)\rho_{ij}(\mathbf{p}' - \mathbf{k}_1, \mathbf{p}, t), \\ & c_{\mathbf{k}}(t)\rho_{ij}(\mathbf{p}', \mathbf{p} - \mathbf{k}_1, t), \quad c_{\mathbf{k}}^\dagger(t)\rho_{ij}(\mathbf{p}', \mathbf{p} + \mathbf{k}_1, t). \end{aligned}$$

Using the equation of motion, Eq. (17), with the Hamiltonian  $H_0 + H_p^0$  gives for  $k_z > 0$  the following oscillatory dependence:

Integrating over time in Eq. (27) and taking  $\mathbf{k} \rightarrow -\mathbf{k}$  gives

$$\rho'_{ij}(\mathbf{p}', \mathbf{p}, t) = \rho_{ij}(\mathbf{p}', \mathbf{p}, 0) + i \sum_{\mathbf{k}, l} \int_0^t dt' [\tilde{\Omega}_{il}^\dagger(\mathbf{k}, t') \rho_{ij}(\mathbf{p}' - \mathbf{k}_1, \mathbf{p}, t') e^{i\omega_{il} t'} - \rho_{ij}(\mathbf{p}', \mathbf{p} + \mathbf{k}_1, t') \tilde{\Omega}_{ij}^\dagger(\mathbf{k}, t') e^{i\omega_{ij} t'}]. \quad (29)$$

Upon iterating by using Eq. (29) in the rhs of Eq. (18), ensemble averaging, and dropping the angular brackets in  $\rho'$  we obtain

$$\begin{aligned} \frac{\partial}{\partial t} \rho'_{ij}(\mathbf{p}', \mathbf{p}, t) = & - \sum_{l, l', \mathbf{k}} \int_0^t dt' [\langle \tilde{\Omega}_{il}(\mathbf{k}, t) \tilde{\Omega}_{l'l'}^\dagger(\mathbf{k}, t') \rangle \rho'_{l'l'}(\mathbf{p}', \mathbf{p}, t') e^{i\omega_{il} t + i\omega_{l'l'} t'} \\ & - \langle \tilde{\Omega}_{il}(\mathbf{k}, t) \tilde{\Omega}_{l'l'}^\dagger(\mathbf{k}, t') \rangle \rho'_{l'l'}(\mathbf{p}' + \mathbf{k}_1, \mathbf{p} + \mathbf{k}_1, t') e^{i\omega_{il} t + i\omega_{l'l'} t'} \\ & - \langle \tilde{\Omega}_{il}^\dagger(\mathbf{k}, t') \tilde{\Omega}_{jl}(\mathbf{k}, t) \rangle \rho'_{l'l'}(\mathbf{p}' - \mathbf{k}_1, \mathbf{p} - \mathbf{k}_1, t') e^{i\omega_{jl} t + i\omega_{l'l'} t'} \\ & + \langle \tilde{\Omega}_{l'l'}^\dagger(\mathbf{k}, t') \tilde{\Omega}_{jl}(\mathbf{k}, t) \rangle \rho'_{l'l'}(\mathbf{p}', \mathbf{p}, t') e^{i\omega_{jl} t + i\omega_{l'l'} t'} ], \quad (30) \end{aligned}$$

$$\begin{aligned} c_{\mathbf{k}}(t)\rho_{ij}(\mathbf{p}' + \mathbf{k}_1, \mathbf{p}, t) & \sim c_{\mathbf{k}}(t)\rho_{ij}(\mathbf{p}', \mathbf{p} - \mathbf{k}_1, t) \\ & \sim e^{-i(\omega_p - k_z c)t} e^{i[\omega(\mathbf{p}', \mathbf{p}) + \omega_{ij}]t}, \quad (23) \end{aligned}$$

$$\begin{aligned} c_{\mathbf{k}}^\dagger(t)\rho_{ij}(\mathbf{p}' - \mathbf{k}_1, \mathbf{p}, t) & \sim c_{\mathbf{k}}^\dagger(t)\rho_{ij}(\mathbf{p}', \mathbf{p} + \mathbf{k}_1, t) \\ & \sim e^{i(\omega_p - k_z c)t} e^{i[\omega(\mathbf{p}', \mathbf{p}) + \omega_{ij}]t}, \quad (24) \end{aligned}$$

where we used that for a highly relativistic positron  $\omega(p) \simeq pc$  and  $\omega(\mathbf{p} + \mathbf{k}_1, \mathbf{p}) = k_z c$ . In Eq. (23) the exponent  $-\omega_p + k_z c$  represents the annihilation of a virtual plasmon  $\hbar\omega_p$  and the gain in energy  $\hbar k_z c$  of the channeling particle, while in Eq. (24) the exponent  $\omega_p - k_z c$  indicates the generation of a virtual plasmon and the loss of channeling particle energy. To extend Eqs. (23) and (24) for  $k_z < 0$ , reality considerations give<sup>18</sup>

$$\begin{aligned} c_{\mathbf{k}}(t)\rho_{ij}(\mathbf{p}' + \mathbf{k}_1, \mathbf{p}, t) & \sim c_{\mathbf{k}}(t)\rho_{ij}(\mathbf{p}', \mathbf{p} - \mathbf{k}_1, t) \\ & \sim e^{-i\eta_{\mathbf{k}}(\omega_p - k_z c)t} e^{i[\omega(\mathbf{p}', \mathbf{p}) + \omega_{ij}]t}, \quad (25) \end{aligned}$$

$$\begin{aligned} c_{\mathbf{k}}^\dagger(t)\rho_{ij}(\mathbf{p}' - \mathbf{k}_1, \mathbf{p}, t) & \sim c_{\mathbf{k}}^\dagger(t)\rho_{ij}(\mathbf{p}', \mathbf{p} + \mathbf{k}_1, t) \\ & \sim e^{i\eta_{\mathbf{k}}(\omega_p - k_z c)t} e^{i[\omega(\mathbf{p}', \mathbf{p}) + \omega_{ij}]t}. \quad (26) \end{aligned}$$

For  $k_z < 0$ , the term  $(\omega_p - k_z c)(-)$  in Eq. (25) [Eq. (26)] represents the annihilation (creation) of a plasmon and a decrease (increase) of the channeling particle energy.

The equation for  $\rho'_{ij}(\mathbf{p}', \mathbf{p}, t)$  is obtained by using Eq. (20) in the left-hand side (lhs) of Eq. (18) and Eqs. (25) and (26) in the right-hand side (rhs) of Eq. (18):

$$\begin{aligned} \frac{\partial}{\partial t} \rho'_{ij}(\mathbf{p}', \mathbf{p}, t) & = i \sum_{\mathbf{k}, l} [\tilde{\Omega}_{il}(\mathbf{k}, t) \rho'_{ij}(\mathbf{p} + \mathbf{k}_1, \mathbf{p}, t) e^{i\omega_{ij} t} \\ & - \rho_{il}(\mathbf{p}', \mathbf{p} - \mathbf{k}_1, t) \tilde{\Omega}_{jl}(\mathbf{k}, t) e^{i\omega_{jl} t}], \quad (27) \end{aligned}$$

where

$$\hbar \tilde{\Omega}_{ij}(\mathbf{k}, t) \equiv V_{ij}(\mathbf{k}) [c_{\mathbf{k}}(t) e^{i\eta_{\mathbf{k}} k_z c t} + c_{-\mathbf{k}}^\dagger(t) e^{i\eta_{\mathbf{k}} k_z c t}] \quad (28)$$

and the oscillatory parts of  $c_{\mathbf{k}}(t)$  and  $c_{-\mathbf{k}}^\dagger(t)$  are given in Eq. (22),  $V_{ij}(\mathbf{k})$  is found from Eq. (11), and

$$\tilde{\Omega}_{ij}^\dagger(-\mathbf{k}, t) = \tilde{\Omega}_{ji}(\mathbf{k}, t).$$

where we use the approximation

$$\langle \tilde{\Omega}(\mathbf{k}, t) \tilde{\Omega}^\dagger(\mathbf{k}', t') \rho' \rangle \simeq \langle \tilde{\Omega}(\mathbf{k}, t) \tilde{\Omega}^\dagger(\mathbf{k}, t') \rangle \langle \rho' \rangle .$$

In obtaining Eq. (30) we have assumed that the plasmons are almost independent modes and the average  $\langle \tilde{\Omega}(\mathbf{k}, t) \tilde{\Omega}^\dagger(\mathbf{k}', t') \rangle$  differs from zero only for  $\mathbf{k} = \mathbf{k}'$  and depends on  $t - t'$ .

Changing the integration variable in Eq. (30) from  $t' \rightarrow \tau = t - t'$ , we note that because of the oscillatory terms in Eq. (30), the main contribution to the  $\tau$  integration occurs for  $\tau \sim 0$ . Thus we can use that  $\rho_{ij}(\mathbf{p}', \mathbf{p}, t - \tau) \simeq \rho_{ij}(\mathbf{p}', \mathbf{p}, t)$ . Integrating over  $\tau$  and time averaging over the fast oscillations give

$$\begin{aligned} \frac{\partial}{\partial t} \rho_{ij}(\mathbf{p}', \mathbf{p}, t) &= i[\omega_{ij} + \omega(\mathbf{p}', \mathbf{p})] \rho_{ij}(\mathbf{p}', \mathbf{p}, t) \\ &- \sum_{i, \mathbf{k}} [G_{ii}(\mathbf{k}) + G_{ij}^\dagger(\mathbf{k})] \rho_{ij}(\mathbf{p}', \mathbf{p}, t) + \sum_{i, l', \mathbf{k}} [G_{iil'}(\mathbf{k}) + G_{iil'j}^\dagger(\mathbf{k})] \delta_{\omega_{ii}, \omega_{l'j}} \rho_{iil'}(\mathbf{p}' + \mathbf{k}_1, \mathbf{p} + \mathbf{k}_1, t), \end{aligned} \quad (31)$$

where  $\rho'_{ij}(\mathbf{p}', \mathbf{p}, t)$  has been transformed back to  $\rho_{ij}(\mathbf{p}', \mathbf{p}, t)$  by Eq. (20),  $\delta_{\omega_{ii}, \omega_{l'j}}$  is the Kronecker  $\delta$ ,  $\tilde{\Omega}_{ij}^\dagger(\mathbf{k}, t) = \tilde{\Omega}_{ji}(-\mathbf{k}, t)$  is used,

$$G_{ij}(\mathbf{k}) = \left[ \int_0^t d\tau \langle \tilde{\Omega}_{ij}(\mathbf{k}, t) \tilde{\Omega}_{ij}^\dagger(\mathbf{k}, t - \tau) \rangle e^{i\omega_{ij}\tau} \right]_t, \quad (32)$$

$$G_{iil'j}(\mathbf{k}) = \left[ \int_0^t d\tau \langle \tilde{\Omega}_{ii}(\mathbf{k}, t) \tilde{\Omega}_{l'j}^\dagger(\mathbf{k}, t - \tau) \rangle e^{i\omega_{l'j}\tau} \right]_t, \quad (33)$$

and the subscript  $t$  denotes a time average over oscillator terms.

Including in Eq. (31) only the terms proportional to  $\rho_{ij}(\mathbf{p}', \mathbf{p}, t)$  for bound states  $i \neq j$ ,  $\rho_{ij}$  is found to obey

$$\begin{aligned} \frac{\partial}{\partial t} \rho_{ij}(\mathbf{p}', \mathbf{p}, t) &\simeq i[\omega_{ij} + \Delta_{ij} + \omega(\mathbf{p}', \mathbf{p})] \rho_{ij}(\mathbf{p}', \mathbf{p}, t) \\ &- \frac{1}{\tau_{ij}(2)} \rho_{ij}(\mathbf{p}', \mathbf{p}, t), \end{aligned} \quad (34)$$

where  $\hbar\Delta_{ij}$  and  $\hbar/\tau_{ij}(2)$  correspond to energy shifts and widths, respectively, in the transition between states  $i$  and  $j$ . Doppler correcting by a factor  $2\gamma^2$  for the emitted photon shifts  $\bar{\Delta}_{ij}$  and widths  $\bar{\Gamma}_{ij}$ , where  $\bar{\Delta}_{ij} = 2\gamma^2\Delta_{ij}$  and  $\bar{\Gamma}_{ij} = 2\gamma^2/\tau_{ij}(2)$ , we obtain

$$i\bar{\Delta}_{ij} - \bar{\Gamma}_{ij} = -2\gamma^2 \sum_{i, \mathbf{k}} [G_{ii}(\mathbf{k}) + G_{ij}^\dagger(\mathbf{k})], \quad (35)$$

where  $\bar{\Gamma}_{ij}$  is the half-width at half maximum (HWHM). The terms  $G_{ij}$  for  $i \simeq j$  represent momentum change without transitions between transverse states. Thus to eliminate nonradiative transitions from Eq. (35) we assume in the following  $G_{ii}(\mathbf{k}) = 0$ .

To evaluate the term  $G_{ij}(\mathbf{k})$  in Eq. (32) we approximate Eq. (11) by expanding  $\exp(ik_x x) \simeq 1 + ik_x x$  for a bound state  $\alpha$  or  $\alpha'$  with  $\alpha \neq \alpha'$ . This yields the following dipole interaction approximation for  $V_{\alpha'\alpha}(\mathbf{k})$ :

$$V_{\alpha'\alpha}(\mathbf{k}) \simeq ik_x X_{\alpha'\alpha} V(k) F(\mathbf{k}), \quad (36)$$

where  $X_{\alpha'\alpha}$  is the dipole moment,

$$X_{\alpha'\alpha} = \int dx \zeta_{\alpha'}^*(x) x \zeta_{\alpha}(x). \quad (37)$$

$$\langle \delta \tilde{n}_{\mathbf{k}}(t) \delta \tilde{n}_{\mathbf{k}}^\dagger(t - \tau) \rangle = \langle [c_{\mathbf{k}}(t) e^{i\eta_{\mathbf{k}} k_z c t} + c_{-\mathbf{k}}(t) e^{-i\eta_{\mathbf{k}} k_z c t}] [c_{\mathbf{k}}^\dagger(t - \tau) e^{-i\eta_{\mathbf{k}} k_z c (t - \tau)} + c_{-\mathbf{k}}^\dagger(t - \tau) e^{i\eta_{\mathbf{k}} k_z c (t - \tau)}] \rangle. \quad (44)$$

The explicit oscillatory dependence of Eq. (44) can be obtained by using Eq. (22). A decay term can be introduced from electron-loss spectroscopic experiments,<sup>9</sup> which phenomenologically represents the various plasmon interactions in the

Thus  $\tilde{\Omega}_{\alpha'\alpha}(\mathbf{k}, t)$  [Eq. (28)] can be represented as

$$\tilde{\Omega}_{\alpha'\alpha}(\mathbf{k}) = e \delta E_x(\mathbf{k}, t) X_{\alpha'\alpha} F(\mathbf{k}), \quad (38)$$

where  $\delta E_x(\mathbf{k}, t)$  is the plasmon electric field in the  $x$  direction as seen by the channeling positrons,

$$e \delta E_x(\mathbf{k}, t) = ik_x V(k) \delta \tilde{n}_{\mathbf{k}}(t) \quad (39)$$

and

$$\delta \tilde{n}_{\mathbf{k}}(t) = c_{\mathbf{k}}(t) e^{i\eta_{\mathbf{k}} k_z c t} + c_{-\mathbf{k}}^\dagger(t) e^{-i\eta_{\mathbf{k}} k_z c t}. \quad (40)$$

Inserting Eq. (38) in Eq. (32) then simply gives

$$\begin{aligned} G_{ij}(\mathbf{k}) &= \left[ \frac{e}{\hbar} \right]^2 |X_{ij}|^2 |F(\mathbf{k})|^2 \\ &\times \left[ \int_0^t d\tau \langle \delta E_x(\mathbf{k}, t) \delta E_x^\dagger(\mathbf{k}, t - \tau) \rangle e^{i\omega_{ij}\tau} \right]_t. \end{aligned} \quad (41)$$

Upon using Eq. (41) in Eq. (35), the photon shifts and widths are expressly given in terms of the plasmon correlation function.

For  $i = j$  in Eq. (31) a rate equation for the population  $\rho_{ii}(\mathbf{p}, t)$  in state  $|i\rangle$  is obtained:

$$\begin{aligned} \frac{\partial}{\partial t} \rho_{ii}(\mathbf{p}, t) &= - \sum_{i, \mathbf{k}} [\rho_{ii}(\mathbf{p}, t) \\ &- \rho_{ii}(\mathbf{p} + \mathbf{k}, \mathbf{p} + \mathbf{k}, t)] / \tau_{ii}(\mathbf{k}), \end{aligned} \quad (42)$$

where

$$\frac{1}{\tau_{ii}(\mathbf{k})} = G_{ii}(\mathbf{k}) + G_{ii}^\dagger(\mathbf{k}) \quad (43)$$

represents the reciprocal decay time of  $\rho_{ii}$  when a population imbalance between states  $|i\rangle$  and  $|l\rangle$  occurs.

#### IV. RESULTS AND DISCUSSION

To calculate the channeling radiation line shifts and widths from Eqs. (35) and (41) we need to know the plasmon correlation function  $\langle \delta E_x(\mathbf{k}, t) \delta E_x^\dagger(\mathbf{k}, t - \tau) \rangle$  or [by using Eqs. (39) and (40)]  $\langle \delta \tilde{n}_{\mathbf{k}}(t) \delta \tilde{n}_{\mathbf{k}}^\dagger(t - \tau) \rangle$ , where

crystal. For silicon, LiF, and diamond, well-defined plasmons with frequency  $\omega_p$  due to four valence electrons per atom are documented in these experiments. The plasmon lifetime  $\tau_p$  is found to be comparatively long so that  $(\omega_p \tau_p)^2 \ll 1$  up to some plasmon momentum  $k_c$  with  $k \leq k_c$ . With these remarks, Eq. (44) becomes

$$\langle \delta \tilde{n}_{\mathbf{k}}(t) \delta \tilde{n}_{\mathbf{k}}^\dagger(t-\tau) \rangle = e^{-\tau/\tau_p} \langle [c_{\mathbf{k}}(0) e^{-i\eta_{\mathbf{k}}(\omega_p - k_z c)t} + c_{-\mathbf{k}}^\dagger(0) e^{-i\eta_{\mathbf{k}}(\omega_p + k_z c)t}] \times [c_{\mathbf{k}}^\dagger(0) e^{i\eta_{\mathbf{k}}(\omega_p - k_z c)(t-\tau)} + c_{-\mathbf{k}}(0) e^{i\eta_{\mathbf{k}}(\omega_p + k_z c)(t-\tau)}] \rangle. \quad (45)$$

The plasmon operators  $c_{\mathbf{k}}(0), c_{\mathbf{k}}^\dagger(0)$  are related to the occupation number  $n_{\mathbf{k}}$  of plasmons with momentum  $\mathbf{k}$  by

$$\langle c_{\mathbf{k}}(0) c_{\mathbf{k}}^\dagger(0) \rangle = n_{\mathbf{k}} + 1, \quad \langle c_{-\mathbf{k}}^\dagger(0) c_{-\mathbf{k}}(0) \rangle = n_{-\mathbf{k}}, \quad (46)$$

and satisfy

$$\langle c_{\mathbf{k}}(0) c_{\mathbf{k}}(0) \rangle = 0, \quad \langle c_{-\mathbf{k}}^\dagger(0) c_{-\mathbf{k}}^\dagger(0) \rangle = 0.$$

The crystals normally considered are around room temperature ( $\frac{1}{40}$  eV) or less and the valence-band plasmon energies are typically greater than 15 eV. Thus the valence electrons have only zero-point collective motions with plasmon occupation number zero. Thus, from Eqs. (45) and (46) with  $n_{\mathbf{k}}=0$  we obtain

$$\langle \delta \tilde{n}_{\mathbf{k}}(t) \delta \tilde{n}_{\mathbf{k}}^\dagger(t-\tau) \rangle = e^{i\eta_{\mathbf{k}}(k_z c - \omega_p)t - \tau/\tau_p}. \quad (47)$$

Using Eqs. (39) and (47) in Eq. (41), we find

$$G_{ij}(\mathbf{k}) = \left[ \frac{e}{\hbar} \right]^2 |X_{ij}|^2 |F(\mathbf{k})|^2 \times \left[ \int_0^t d\tau e^{i[\omega_{ij} + \eta_{\mathbf{k}}(k_z c - \omega_p)]\tau - \tau/\tau_p} \right]_t \times \langle |\delta E_x(\mathbf{k}, 0)|^2 \rangle. \quad (48)$$

Since  $(\omega_p \tau_p)^2 \ll 1$ , the fluctuation dissipation theorem<sup>8</sup> may be straightforwardly applied to give  $\langle |\delta E_x(0)|^2 \rangle / 8\pi = \hbar \omega_p / 2$ . Using this in Eq. (48) and integrating over  $\tau$  finally gives

$$G_{ij}(\mathbf{k}) = - \left[ \frac{8\pi e^2}{\hbar^2} \right] |F(\mathbf{k})|^2 \left[ \frac{\hbar \omega_p}{2} \right] \times \left[ \frac{|X_{ij}|^2}{i[\omega_{ij} + \eta_{\mathbf{k}}(k_z c - \omega_p)] - \frac{1}{\tau_p}} \right]. \quad (49)$$

Inserting Eq. (49) in Eq. (35) and taking  $k_z \rightarrow -k_z$  for  $k_z < 0$  yields, for the line shifts and widths,

$$\bar{\Delta}_{ij} = -2\gamma^2 \left[ \frac{8\pi e^2}{\hbar^2} \right] |F(\mathbf{k})|^2 \left[ \frac{\hbar \omega_p}{2} \right] \sum_{\substack{l, \mathbf{k} \\ n=\pm 1}} \Theta(k_z) \times \left[ |X_{li}|^2 \frac{u_{ij}(\mathbf{k}, n)}{[u_{li}(\mathbf{k}, n)]^2 + \left[ \frac{1}{\tau_p} \right]^2} - |X_{lj}|^2 \frac{u_{ij}(\mathbf{k}, n)}{[u_{lj}(\mathbf{k}, n)]^2 + \left[ \frac{1}{\tau_p} \right]^2} \right], \quad (50)$$

$$\bar{\Gamma}_{ij} = 2\gamma^2 \left[ \frac{8\pi e^2}{\hbar^2} \right] |F(\mathbf{k})|^2 \left[ \frac{\hbar \omega_p}{2} \right] \sum_{\substack{l, \mathbf{k} \\ n=\pm 1}} \Theta(k_z) \frac{1}{\tau_p} \times \left[ |X_{lj}|^2 \frac{1}{[u_{lj}(\mathbf{k}, n)]^2 + \left[ \frac{1}{\tau_p} \right]^2} + |X_{ij}|^2 \frac{1}{[u_{ij}(\mathbf{k}, n)]^2 + \left[ \frac{1}{\tau_p} \right]^2} \right], \quad (51)$$

where

$$u_{ij}(\mathbf{k}, n) = \omega_{ij} + k_z c - \eta_n \omega_p \quad (52)$$

and

$$\eta_n = \begin{cases} +1, & n = 1 \\ -1, & n = -1 \end{cases} \quad (53)$$

$$\Theta(k_z) = \begin{cases} 1, & k_z > 0 \\ 0, & k_z < 0. \end{cases}$$

From Eqs. (50) and (51) one explicitly sees how the line shifts and widths depend on the transverse energies  $\omega_{ij}$ , the dipole moment  $X_{ij}$  which can be obtained from the thermally averaged potential, and on the plasmon energy  $\hbar \omega_p$  and lifetime  $\tau_p$ .

The photon line shifts in Eq. (50) are obtained by summing over transverse intermediate bound and free states  $|l\rangle$ . It can be shown that for nearly degenerate transition frequencies, as in positron channeling,  $|X_{l+1, l}|^2$  is proportional to the quantum number  $l$  and the bound transition contributions to the line shifts nearly cancel. The bound-free contribution is obtained by summing in Eq. (50) over all transverse free momentum states  $\mathbf{q}$ , where  $|l\rangle \equiv |\mathbf{q}\rangle$  are taken as plane-wave states. We use the

Fourier transform of the harmonic oscillator eigenfunctions to obtain the dipole moments,<sup>16</sup>

$$|X_{qj}|^2 = \frac{\pi\hbar}{\gamma m \omega_0} [\sqrt{j+1}\Psi_{j+1}(q) + \sqrt{j}\Psi_{j-1}(q)]^2, \quad (54)$$

where  $\{\Psi_j(q)\}$  form a complete set of orthonormal functions. Upon transforming the sum over  $\mathbf{k}$  in Eq. (50) to an integral over the degrees of freedom by  $\sum_{\mathbf{k}} \rightarrow N_v/(2\pi)^3 \int d\mathbf{k}$  for  $k \leq k_c$  where  $N_v$  is the number of atoms per unit cell, the relative photon shifts  $\bar{\Delta}_{j+1,j}^{bf}/\bar{\omega}_{j+1,j}$  due to virtual bound-free transitions are obtained:

$$\frac{\bar{\Delta}_{j+1,j}^{bf}}{\bar{\omega}_{j+1,j}} \simeq -\mu \left[ \sum_{n=\pm 1} \ln \left( \frac{K^2}{(M-j-\eta_n j_p)^2 + \delta^2} \right) \right]. \quad (55)$$

Here

$$\mu = \frac{e^2 k_c^2 \omega_p}{2\pi c m \gamma \omega_0^2} = \frac{\alpha}{2\pi} \left( \frac{\omega_0^*}{\omega_0} \right)^2, \quad (56)$$

$\omega_0$  is the oscillator frequency,  $\alpha = e^2/\hbar c = \frac{1}{137}$  is the fine-structure constant,  $\omega_0^* = \omega_p/\sqrt{\gamma} = \theta(\omega_0)$ ,  $N_v \approx 2$ , and the maximum allowed collective plasmon wave number  $k_c$  is chosen as<sup>10</sup>

$$k_c = (4\pi n_e e^2/\hbar \omega_p)^{1/2}. \quad (57)$$

Also

$$K = ck_c/\omega_0, \quad j_p = \omega_p/\omega_0, \quad \delta \approx 1/\omega_0 \tau_p, \quad (58)$$

where  $\delta^2 \ll j_p^2$ ,  $|F(k)|^2$  is taken as unity,  $j$  is a integer label for the quantum bound state,  $M$  is a maximum quantum number corresponding to the absolute depth of the channeling potential and  $K \gg j_p, M$ . For a given channel half-width  $d$ ,  $M$  is roughly estimated as  $2\pi n_e e^2 d^2/\hbar \omega_0$ . In addition, eigenstates near the top of the well will have relatively short lifetimes due to wave-function overlap with the thermally vibrating atomic planes. Such states are not expected to significantly contribute to the sharp line spectrum. An estimate of the critical quantum number  $j_c$  below which the states are effectively long-lived involves the Thomas-Fermi screening length  $a_{TF}$  through the expression  $j_c \sim 2\pi n_e e^2 (d - a_{TF})^2/\hbar \omega_0$ .<sup>19</sup> For 55-MeV channeled positrons in (110) silicon, LiF, and diamond,  $j_c$  ( $M$ ) equals 6 (10), 3 (9), and 3 (8), with volume plasmon spectral peaks  $\hbar \omega_p$  (Refs. 9 and 20) situated near 16.5, 25.3, and 34.0 eV, respectively. Inserting these values in Eq. (55), one finds that the corresponding  $j_c \rightarrow j_c - 1$  transitions are maximally shifted by the amount:  $-2.3\%$ ,  $-3.5\%$ , and  $-3.6\%$ . As these transitions represent the most intense spectral lines, it is presumed that the maxima of the spectral envelopes are correlated with the position of these dominant lines. To compare these dynamic shifts with experiment, it is necessary to only superimpose them on the previously found static shifts due to anharmonicity.<sup>2,19</sup> For silicon, LiF, and diamond, the agreement between theory and experiment is qualitatively improved.<sup>3-6</sup>

In the following, we determine the plasmon contribution to the spectral widths of the photon spectrum. The plasmon excitations defined by the regime  $k \leq k_c$  are

reasonably long-lived as documented in ELS. For example, in silicon, LiF, and diamond, volume plasmon spectral widths are observed with full width at half maximum (FWHM) 3.6, 4.7, and 14 eV, respectively.<sup>9,20</sup> This corresponds to a plasmon lifetime  $\tau_p$  of about  $10\omega_{pe}^{-1}$  for silicon,  $13\omega_{pe}^{-1}$  for LiF, and  $5\omega_{pe}^{-1}$  for diamond.

The spectral widths (HWHM) from the  $b$ - $f$  and the  $b$ - $b$  virtual transitions are obtained from Eq. (51). Noting that in Eq. (51) terms with  $|j-j'| \geq 2$  are negligibly small, we find for the relative widths:

$$\bar{\Gamma}_{j+1,j}^{bf}/\bar{\omega}_{j+1,j} \simeq 4\mu(j+1) \{ \pi + \tan^{-1}[(j_p - M + j)/\delta] - \tan^{-1}[(j_p + M - j)/\delta] \}, \quad (59)$$

$$\bar{\Gamma}_{j+1,j}^{bb}/\bar{\omega}_{j+1,j} \simeq 4\mu(j+1)\pi, \quad (60)$$

where  $\tan^{-1}[(K + M - j_p)/\delta] \simeq \pi/2$ , and  $\bar{\Gamma}^{bf} \simeq \bar{\Gamma}^{bb}$ . For the  $b$ - $f$  and  $b$ - $b$  cases, maximal widths are associated with the highest allowed transition. For 55-MeV channeled positrons in the (110) direction the corresponding  $b$ - $f$  ( $b$ - $b$ ) widths for silicon, LiF, and diamond are 7.0 (7.5), 5.5 (6.0), and 6.0 (6.5) percent. Thus the total linewidths are 14.5%, 11.5%, and 12.5%. For lower-level transitions, both types of width are considerably reduced. In the case of LiF, these dynamic widths are masked by strong anharmonic effects, but in diamond the  $3 \rightarrow 2$  transition could significantly contribute to the composite spectral width.

## V. CONCLUSIONS

In summary, the system of valence electrons behaves collectively to produce a zero-point level of electric field fluctuations effectively near the channeling midplane. The dynamical effect of these fluctuations on the positron static channeling potential may be sufficiently large to induce observable shifts associated with the composite spectra. For silicon, LiF, and diamond the agreement between theory and experiment on shifts is qualitatively improved. The dynamic widths for diamond may be particularly important since minimal anharmonicity occurs.

Although a plasmon feature has been proposed to partially explain the anomalous spectral shifts, additional refinement may require consideration of phonons and excitons for example. In the case of LiF, sharp excitonic structure is seen in ELS studies,<sup>20</sup> thereby possibly obscuring the interpretation of the broad central peak as a sole volume plasmon feature.<sup>21</sup>

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