Importance of vertex corrections for obtaining correct selection rules in the theory of photoemission

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It is shown that the commonly used approximation for the no-loss angle-resolved photocurrent from solids in general violates the optical selection rules if the final-state inverse low-energy electron diffraction orbital is taken to be damped. The violations occur in the velocity formula, which is inconsistent with the corresponding and commonly used acceleration formula in which the matrix elements are evaluated with respect to the gradient of the one-electron potential. It has recently been shown that, in a correct description of the no-loss photocurrent, the bare matrix elements should be replaced by the appropriate vector-coupling vertex function. We demonstrate that this modification is needed in order to have correct selection rules and consistency between the velocity and acceleration formulas for photoemission and photoabsorption. Our formalism further allows us to interpret the commonly used acceleration formula in terms of a well-defined approximation which leaves out local-field effects, and to give approximations which obey the selection rules.

I. INTRODUCTION

Photoemission spectra of solids can often be successfully interpreted on the basis of one-electron concepts. However, in a pure one-electron theory the photoyield would be limited by the penetration depth of the incident radiation, whereas in reality it is limited by the escape depth of the photoelectrons, which often is only a few atomic distances. Thus, in real solids only those photoelectrons which have been intrinsically created near the surface contribute to the observed spectrum. The current way to remedy one electron from this deficiency is to include the mean-free-path effects as an afterthought and take the photoelectron orbital to be damped inside the solid.[1] In quantitative terms this means that the photocurrent (J_p) is calculated from the expression

$$J_p = 2\pi \sum_n^{\text{occ}} |\langle \chi_p^{(-)} | \Delta | \phi_n \rangle|^2 \delta(\varepsilon_p - \varepsilon_n - \omega) . \quad (1)$$

Here ϕ_n and $\chi_p^{(-)}$ are initial- and final-state orbitals of energies ε_n and ε_p, respectively (p is short for momentum and spin), $\Delta = -i\mathbf{A}\cdot\nabla$ is the coupling to the optical field, and ω is the photon energy. (We use units such that $\hbar = e^2 = m = 1$.) The effects of the mean free path are introduced by solving the photoelectron orbital $\chi_p^{(-)}(\mathbf{r})$ in the effective potential seen by a quasiparticle. This potential consists of the total Coulomb potential (V_C) and the dynamical self-energy (Σ). The latter term is not Hermitian and gives rise to a damping inside the solid.

It is known, however, that by introducing self-energy effects in response functions without the corresponding vertex corrections one violates macroscopic conservation laws.[2,3] We show here that the theory summarized in Eq. (1) also leads to inconsistencies in that it violates the optical selection rules, and that these problems are resolved by a more accurate description of the photoemission process given in Ref. 4, henceforth to be referred to as I. The fact that Eq. (1) violates the optical selection rules has also been observed by Hedin and Nykvist.[5]

The origin of the selection rule violations is that the damped photoelectron orbital is not an eigensolution of the same one-electron Hamiltonian as the initial-state orbitals, and thus such violations do not occur within strict one-electron theory. If we, on the other hand, use the "acceleration" formula for the matrix elements and represent the optical field by $i\mathbf{A}\cdot\nabla V/\omega$ we obtain an expression

$$J_p = \frac{2\pi}{\omega^2} \sum_n^{\text{occ}} |\langle \chi_p^{(-)} | \mathbf{A}\cdot\nabla V | \phi_n \rangle|^2 \delta(\varepsilon_p - \varepsilon_n - \omega) . \quad (2)$$

This expression manifestly obeys correct selection rules also when $\chi_p^{(-)}$ is taken to be damped. Unfortunately, previous theoretical work gives no fundamental arguments for choosing the acceleration formula above instead of the velocity formula in Eq. (1), despite the fact that they are not at all equivalent.

In the present paper we will confine ourselves to the no-loss photoelectron current, except for a short discussion of optical absorption, and the dipole (long-wavelength) approximation. As the ambiguities arise from introducing the photoelectron self-energy in an uncritical way, a proper explanation must be based on a full many-electron description of the photoemission process. We here follow our earlier work, I. Equation (1) closely resembles an expression for the no-loss part of the photoelectron current $J_p^{(e)}$ given in I,

$$J_p^{(e)} = \int d\varepsilon\, 2\pi\delta(\varepsilon_p - \varepsilon - \omega) \langle \chi_p^{(-)} | \Lambda_v(\varepsilon+\omega, \varepsilon) A_<(\varepsilon) \Lambda_v^\dagger(\varepsilon+\omega, \varepsilon) | \chi_p^{(-)} \rangle . \quad (3)$$

Our results above involve two modifications compared to the approximation in Eq. (1): (a) the density of occupied states, $\sum_n |\phi_n\rangle\delta(\varepsilon-\varepsilon_n)\langle\phi_n|$ inherent in Eq. (1), has been replaced by its interacting counterpart $A_<(\varepsilon)$, and (b) the bare coupling $\Delta=-i\mathbf{A}\cdot\nabla$ has been replaced by the vector-coupling vertex function Λ_v. [In Eq. (3) the energy integration is confined to the quasiparticle part of $A_<$.]

As we will show in detail, the vertex correction in Eq. (3) is quite substantial and almost entirely removes the violation of the selection rules. There remain, however, small terms which are proportional to the lifetime width $\Gamma(\varepsilon)$ of the hole left behind. Thus one can say that Eq. (3) is correct to the same precision as the no-loss current itself can be defined. When $\Gamma(\varepsilon)$ is small, the final states of the solid left behind are almost stable, and we have no difficulty in distinguishing between the elastic and the loss parts of the photocurrent. When $\Gamma(\varepsilon)$ is large, on the other hand, we are in spectral regions where strong loss satellites are overlapping with the "elastic" contributions, and the distinction between a loss and an elastic part becomes ambiguous and less useful. In a metal, hole states with energies ε above the first plasmon satellite usually have only a very small lifetime width. The same situation occurs in semiconductors and insulators with energies less than a band-gap energy below the top of the valence band. Thus, for such final hole states the no-loss current is rather well defined and correctly described by Eq. (3).

In the velocity formulation of the photoemission process, where the coupling is taken as $\mathbf{A}\cdot\mathbf{P}$, correct selection rules are obtained only by including a consistent description of the relevant vertex function. It is quite difficult to obtain good approximations for such vertex parts, and thus the velocity formulation is rather awkward to use in practice. This has motivated the second part of this work, namely the derivation of correct many-electron counterparts to the acceleration formula in Eq. (2). In practical application by Pendry and by others the potential V in Eq. (2) has been taken as the total effective one-electron potential. As we will show, in a many-electron description one should instead take the matrix elements with respect to $\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w$, where w is the potential from only the nuclei and where $\epsilon(\omega)$ is the dynamical screening function. Note that $\mathbf{A}\cdot\nabla w$ gives the polarization obtained by displacing the nuclei by an amount $\mathbf{A}$ and that such a displacement gives rise to a *finite* potential, despite the fact that w itself is infinite. A vertex function enters also in the correct acceleration formula, but it plays no crucial role for establishing correct optical selection rules.

We derive the acceleration formulas in two different ways. One is based on commutation relations, and one is based on the Bethe-Salpeter equation for particle-hole excitations. The latter method allows us to give criteria for having a consistent description in approximate schemes and to give simple low-order approximations. It also allows us to give a physical interpretation of the approximation in Eq. (2). As we will show, replacing $\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w$ by $\mathbf{A}\cdot\nabla V$ in the proper acceleration formula for the elastic photocurrent corresponds to neglecting the local-field effects. Local-field effects, which are connected to the spatial variation of the induced part of the field, should be particularly important in the surface layer. Thus we expect the approximation in Eq. (2) to be less useful for describing the surface photoelectric effect.

The expression in Eq. (3) for the elastic photocurrent correctly transforms into the corresponding acceleration formula in spectral regions where the concept of a no-loss current is meaningful. The approximation in Eq. (1), on the other hand, does not have this property, which again illustrates the necessity of vertex corrections. As we will discuss at the end of this paper, similar problems occur in approximate treatments of optical absorption. Thus, approximations analogous in Eq. (1) which include self-energy renormalizations but no vertex part also violate the optical selection rules (as well as the f-sum rule), and are inconsistent with the corresponding acceleration formulas.

II. DISCUSSION OF A SIMPLE MODEL

To illustrate the principles of how selection rules are violated in the approximation in Eq. (1), we consider here a simple model with translational invariance along the surface. We assume that the system is confined to the region $z<0$ and expose it to radiation at normal incidence:

$$\delta H=\sum_{m,n}\Delta_{mn}c_m^\dagger c_n e^{-i\omega t}+\text{H.c}\ . \tag{4}$$

Here c_n is an electron operator, and

$$\Delta_{mn}=\langle m\,|\,-iA_x\nabla_x\,|\,n\rangle\ . \tag{5}$$

As is well known, the perturbation in Eq. (4) does not give rise to any transitions, owing to the translational invariance along the x direction, and the total photocurrent is zero. Equation (1), however, gives a nonzero current as soon as the photoelectron damping is introduced. The initial (ϕ_n) and final ($\chi_p^{(-)}$) states obey the one-electron equations

$$(h+v_{\text{xc}})\phi_n=\varepsilon_n\phi_n\ , \tag{6}$$

$$[h+\Sigma^\dagger(\varepsilon_p)]\chi_p^{(-)}=\varepsilon_p\chi_p^{(-)}\ , \tag{7}$$

and can be taken as

$$\phi_{\nu\mathbf{p}_{||}}(\mathbf{r})=f_\nu(z,\mathbf{p}_{||})\exp(i\mathbf{p}_{||}\cdot\mathbf{r}_{||})\ , \tag{8}$$

$$\chi_p^{(-)}(\mathbf{r})=g_p(z)\exp(i\mathbf{p}_{||}\cdot\mathbf{r}_{||})\ . \tag{9}$$

[Here $h=-\nabla^2/2+V_C$, v_{xc} is the exchange-correlation potential used for occupied states, and $\Sigma^\dagger(\varepsilon)$ is the time-reversed self-energy. The label $||$ indicates components parallel with the surface.] An easy calculation shows that Eq. (1) gives the current

$$J_p=2\pi(A_xp_x)^2\sum_\nu|\langle g_p\,|\,f_\nu\rangle|^2\delta(\varepsilon_p-\varepsilon_{\nu\mathbf{p}_{||}}-\omega)\ . \tag{10}$$

On a one-electron level v_{xc} has to be chosen real and thus different from $\Sigma^\dagger(\varepsilon_p)$, which has an anti-Hermitian part of several eV at typical photoelectron energies. Thus, in general $\langle g_p\,|\,f_\nu\rangle\neq0$ and $J_p>0$.

One might ask if these problems disappear if the damping effects are included also for the initially occupied states. The proper way to include these effects is to replace the independent-electron density of occupied states

by its interacting counterpart $A_<(\varepsilon)$ and thus to replace Eq. (1) by

$$J_p = \int d\varepsilon\, 2\pi\delta(\varepsilon_p-\varepsilon-\omega)\langle\chi_p^{(-)}\,|\,\Delta A_<(\varepsilon)\Delta^\dagger\,|\,\chi_p^{(-)}\rangle\ , \quad (11)$$

where $\chi_p^{(-)}(\mathbf{r})$ is the same time-reversed low-energy electron diffraction (LEED) orbital as in Eq. (7). Making use of Eq. (9) and introducing a complete set of orbitals $\{\phi_n\}$ we obtain from Eq. (11)

$$J_p = 2\pi(A_x p_x)^2 \sum_{m,n}\langle\chi_p^{(-)}\,|\,m\rangle\langle m\,|\,A_<(\varepsilon_p-\omega)\,|\,n\rangle \times\langle n\,|\,\chi_p^{(-)}\rangle\ . \quad (12)$$

Since the state density $A_<$ is confined to the solid, we can restrict the sum in Eq. (12) to orbitals ϕ_n which vanish for, say, $z>z_0$. Now, the state density $\langle m\,|\,A_<(\varepsilon)\,|\,n\rangle$ is a positive-definite matrix. Further, the LEED orbital $\chi_p^{(-)}$ penetrates an escape depth into the solid. Thus, in general $\langle n\,|\,\chi_p^{(-)}\rangle\neq 0$. As a consequence, also the approximation in Eq. (10) in general violates the selection rule of a vanishing photocurrent at normal incidence.

The selection rule violations show up in a very clearcut way in the simple model considered above. Similar violations also occur in more complex systems and may give, for example, incorrect behavior of peak intensities with respect to polarization angles. As we will show in the next section, these problems disappear, to within the accuracy discussed in the Introduction, when the correct vector-coupling vertex function is included.

III. ACCELERATION FORMULAS FOR THE PHOTOEMISSION PROCESS

A. General

In one-electron theory one easily obtains the acceleration formula in Eq. (2) from commutator relation $[\mathbf{p},h]=-i\nabla V$. We now wish to derive a corresponding expression for the no-loss photocurrent from an interacting solid.

In the dipole approximation one is effectively taking the optical perturbation as

$$\delta H = \mathbf{A}\cdot\mathbf{P}e^{-i\omega t}+\text{H.c.}\ , \quad (13)$$

where $\mathbf{P}=\sum_j \mathbf{p}_j$ is the total momentum and where $\mathbf{A}$ is the applied, external field. (Note that only the external field is assumed to be spatially homogeneous. The corresponding total effective field exhibits a rapid variation in each unit cell as well as at the surface and has a finite penetration depth.) The Hamiltonian of the solid is of the form

$$H = T+\sum_{j<i} v(ij)+\sum_j w(j)\ .$$

The second interaction term is translationally invariant just like the kinetic energy T. Thus,

$$[\mathbf{P},H]=\left[\mathbf{P},\sum_j w(j)\right]=-i\sum_j \nabla w(j)\ ,$$

which gives

$$\omega\langle N,s\,|\,\mathbf{P}\,|\,N\rangle = -\langle N,s\,|\,[\mathbf{P},H]\,|\,N\rangle = i\left\langle N,s\left|\sum_j \nabla w(j)\right|N\right\rangle$$

for final N-electron states $|\,N,s\,\rangle$ with an energy ω above the ground state $|\,N\,\rangle$. Thus, we can instead take the coupling to the radiation field as

$$\delta H' = (i/\omega)\sum_j \mathbf{A}\cdot\nabla w(j)e^{-i\omega t}+\text{H.c}\ . \quad (14)$$

Going through the same procedure as described in I we obtain the no-loss photocurrent as

$$J_p^{(e)} = \int d\varepsilon\, 2\pi\delta(\varepsilon_p-\varepsilon-\omega)\frac{1}{\omega^2}\langle\chi_p^{(-)}\,|\,\Lambda_a(\varepsilon+\omega,\epsilon)A_<(\varepsilon)\Lambda_a^\dagger(\varepsilon+\omega,\varepsilon)\,|\,\chi_p^{(-)}\rangle\ , \quad (15)$$

where the vertex Λ_a gives the response in the inverse of the Green's function G to the scalar perturbation $-\mathbf{A}\cdot\nabla w$:

$$\langle\mathbf{x}\,|\,\Lambda_a(\varepsilon+\omega,\varepsilon)\,|\,\mathbf{x}'\rangle = \int dt\,dt'd^3y[-\delta G^{-1}(\mathbf{x}t,\mathbf{x}'t')/\delta w(\mathbf{y},0)]e^{i(\varepsilon+\omega)t-i\varepsilon t'}[-\mathbf{A}\cdot\nabla w(\mathbf{y})]\ . \quad (16)$$

Introducing the usual screened, scalar vertex

$$\tilde{\Lambda}_0(12;3) = -\delta G^{-1}(12)/\delta V_C(3)\ , \quad (17)$$

which gives the response to an increment in the total Coulomb potential V_C, and the dielectric function ϵ $[\epsilon^{-1}(12)=\delta V_C(1)/\delta w(2)]$, we can rewrite Λ_a as

$$\langle\mathbf{x}\,|\,\Lambda_a(\varepsilon+\omega,\varepsilon)\,|\,\mathbf{x}'\rangle = \int d^3y\,d^3z\,\tilde{\Lambda}_0(\mathbf{x}\varepsilon+\omega,\mathbf{x}'\varepsilon;\mathbf{y})\epsilon^{-1}(\mathbf{y},\mathbf{z};\omega)[-\mathbf{A}\cdot\nabla w(\mathbf{z})]\ . \quad (18)$$

All matrix elements are now expressed in ∇w, and correct selection rules are obtained regardless of the approximations used for the LEED orbital, state density $A_<$, and the vertex function. Neglecting the vertex correction we obtain the simple expression

$$J_p^{(e)} = \frac{2\pi}{\omega^2}\langle\chi_p^{(-)}\,|\,[\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w]A_<(\varepsilon_p-\omega)[\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w]\,|\,\chi_p^{(-)}\rangle\ ,$$

which is analogous to the independent-electron result in Eq. (2). Thus, we see that to a first approximation the matrix elements in the acceleration formula should be taken with respect to the effective perturbation $\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w$. The vertex corrections could be estimated by, say, time-dependent local-density theory if need be, but they need not be included in order to have correct selection rules.

The result that acceleration formulations involve $\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w$ has been obtained earlier by Hermeking,[6] but no explicit result for the no-loss part of the current has been given. Instead, Hermeking obtains an expression of similar structure as our Eq. (3) [Eq. (3.15) in Ref. 6] by neglecting three-particle correlations and claims that it describes the *entire* photocurrent. [Hermeking's expression includes first-order vertices of all possible orderings and not just the time-ordered part as in our Eq. (3).] We do not agree on this statement. To the contrary, we believe Eq. (3.15) in Ref. 6 leaves out the processes responsible for the extrinsic losses, i.e., those loss diagrams which survive in the limit of high photoelectron energies.[7]

The applied field in the acceleration formula, $-\mathbf{A}\cdot\nabla w$, corresponds to displacing all nuclei by an amount $-\mathbf{A}$ while keeping the electron charges fixed. On a microscopic level this field is rapidly varying, but the corresponding averaged macroscopic field corresponds to a polarization of $-Z\mathbf{A}$ per atom, where Z is the charge of a nucleus. Consequently, no divergency occurs when summing up the contributions from all constituting atoms. The dynamical screening $\epsilon^{-1}(\omega)$ transforms this field to a screened effective field. Owing to the anti-Hermitian part in ϵ^{-1} this effective field is spatially damped inside the solid with a decay length equal to the penetration depth of the incident radiation. When ω is small compared to all core-electron energies, it is reasonable to assume the screening of the core electrons to be instantaneous, which suggests the approximation

$$\epsilon^{-1}(\omega)\nabla w=\epsilon_v^{-1}(\omega)\nabla w_{\rm ion}\ . \tag{19}$$

Here $w_{\rm ion}$ is the total Coulomb potential from the ion cores, and ϵ_v gives the screening from only the valence electrons.

B. Mutual consistency of velocity and acceleration formulations

In a correct theory the velocity and acceleration formulations should of course be equivalent. Unfortunately, the approximation in Eq. (1), or its extrapolation to include interaction effects on the initially occupied states, Eq. (11), fails badly in this respect. The velocity and acceleration formulas for the no-loss current proposed here, on the other hand, we here show to be equivalent to within terms of the order of the hole lifetimes.

Let us first consider the approximation in Eq. (11) without vertex corrections. In this approximation the contribution to the photocurrent from initially occupied states of energy $\varepsilon=\varepsilon_p-\omega$ is given by

$$2\pi\langle\chi_p^{(-)}\,|\,\Delta A_<(\varepsilon)\Delta^\dagger\,|\,\chi_p^{(-)}\rangle\ .$$

To transform this expression to an acceleration form we use the Dyson equations for $\chi_p^{(-)}$ and $A_<$. We obtain

$$\begin{aligned}\langle\chi_p^{(-)}\,|\,\Delta A_<(\varepsilon)&=\frac{1}{\omega}\langle\chi_p^{(-)}\,|\,[(\varepsilon+\omega)\Delta-\Delta\varepsilon]A_<(\varepsilon)\\&=\frac{1}{\omega}\langle\chi_p^{(-)}\,|\,\{[h+\Sigma(\varepsilon+\omega)]\Delta-\Delta[h+\Sigma(\varepsilon)+G^{-1}(\varepsilon)]\}A_<(\varepsilon)\ .\end{aligned}\tag{20}$$

[Note that $\langle\chi_p^{(-)}\,|\,G^{-1}(\varepsilon+\omega)=0.$] The density of occupied states we write as

$$A_<(\epsilon)=\frac{1}{2\pi}G(\varepsilon)\Gamma(\varepsilon)G^\dagger(\varepsilon)\ ,\tag{21}$$

where

$$\Gamma(\varepsilon)=\frac{1}{i}[\Sigma(\varepsilon)-\Sigma^\dagger(\varepsilon)]\tag{22}$$

is an Hermitian positive-definite matrix in one-electron labels which gives the hole lifetimes due to the many-electron interactions. Using the explicit forms of h and Δ we can now rewrite Eq. (20) as

$$\langle\chi_p^{(-)}\,|\,\Delta A_<(\varepsilon)=\frac{1}{i\omega}\langle\chi_p^{(-)}\,|\,[-\mathbf{A}\cdot\nabla V_C+\Sigma(\varepsilon+\omega)\mathbf{A}\cdot\nabla-\mathbf{A}\cdot\nabla\Sigma(\varepsilon)]A_<(\varepsilon)-\frac{1}{2\pi i\omega}\langle\chi_p^{(-)}\,|\,\mathbf{A}\cdot\nabla\Gamma(\varepsilon)G^\dagger(\varepsilon)\ .\tag{23}$$

Thus we see that the velocity formula without vertex corrections does not transform into anything like the acceleration formula of the previous subsection. First, the gradient of the total Coulomb potential appears in place of the dynamically screened gradient of the external potential. Second, a large term $\Sigma(\varepsilon+\omega)\mathbf{A}\cdot\nabla-\mathbf{A}\cdot\nabla\Sigma(\varepsilon)$ which violates the selection rules has appeared. To verify that these terms indeed break the symmetry we temporarily go back to the simple model discussed in Sec. II. In this model Eq. (23) simplifies to

$$\langle\chi_p^{(-)}\,|\,\Delta A_<(\varepsilon)=\frac{1}{\omega}A_xp_x\langle\chi_p^{(-)}\,|\,[\Sigma(\varepsilon+\omega)-\Sigma(\varepsilon)]A_<(\varepsilon)-\frac{A_xp_x}{2\pi\omega}\langle\chi_p^{(-)}\,|\,\Gamma(\varepsilon)G^\dagger(\varepsilon)\ .$$

We note that the anti-Hermitian part in $\Sigma(\varepsilon+\omega)$ is large at typical photoelectron energies and that it is of opposite sign as the anti-Hermitian part in $\Sigma(\varepsilon)$. We also note that the Hermitian part in $\Sigma(\varepsilon)$ is in general positive and large for localized hole states, whereas the Hermitian part in $\Sigma(\varepsilon+\omega)$ is in general negative. Finally there is in Eq. (23) also a term proportional to the hole lifetime involving $\Gamma(\varepsilon)$. As discussed in the Introduction, a clear identification of a no-loss part of the photoelectron spectrum requires $\Gamma(\varepsilon)$ to be small, and consequently we will neglect it in the spectral region under discussion.

We now turn to the full velocity formula in Eq. (3) for the no-loss current. To transform this expression we need to express the vector-coupling vertex function Λ_v in terms of the scalar vertex Λ_a. The vertices Λ_v and Λ_a give, respectively, the response in the inverse Green's function to the perturbation δH in Eq. (13) and $\delta H'$ in Eq. (14). These perturbations we rewrite here as

$$\delta H = \mathbf{A}\cdot\mathbf{P}f(t)+\text{H.c.} ,$$

$$\delta H' = \mathbf{A}\cdot\dot{\mathbf{P}}g(t)+\text{H.c.} ,$$

and study the response functions

$$R_v(x,x';t'') \equiv \delta G(x,x')/\delta f(t'') = -\langle T\psi(x)\psi^\dagger(x')\mathbf{A}\cdot\mathbf{P}(t'')\rangle ,$$

$$R_a(x,x';t'') \equiv \delta G(x,x')/\delta g(t'') = -\langle T\psi(x)\psi^\dagger(x')\mathbf{A}\cdot\dot{\mathbf{P}}(t'')\rangle .$$

(Here ψ is the electron field operator, and x and x' represent spatial and time coordinates.) We readily find

$$\frac{\partial}{\partial t''}R_v(x,x',t'') = -\langle T[\mathbf{A}\cdot\mathbf{P}(t),\psi(x)]\psi^\dagger(x')\rangle\delta(t-t'') - \langle T\psi(x)[\mathbf{A}\cdot\mathbf{P}(t'),\psi^\dagger(x')]\rangle\delta(t'-t'') + R_a(x,x';t'') .$$

We next use the relations $[\mathbf{P}(t),\psi(x)]=i\nabla\psi(x)$ and perform a Fourier transformation. This gives

$$R_v(\varepsilon+\omega,\varepsilon) = \frac{1}{i\omega}[\mathbf{A}\cdot\nabla G(\varepsilon) - G(\varepsilon+\omega)\mathbf{A}\cdot\nabla + R_a(\varepsilon+\omega,\varepsilon)] . \tag{24}$$

[In Eq. (24) the Fourier transforms are defined as in Eq. (16), and all quantities are to be interpreted as matrices in one-electron labels.] To find the relation between the corresponding vertex functions we note that $\delta G^{-1} = -G(\delta G)G$ and multiply Eq. (24) with $G^{-1}(\varepsilon+\omega)$ from the left and width $G^{-1}(\varepsilon)$ from the right to obtain

$$\Lambda_v(\varepsilon+\omega,\varepsilon) = \frac{1}{i\omega}[G^{-1}(\varepsilon+\omega)\mathbf{A}\cdot\nabla - \mathbf{A}\cdot\nabla G^{-1}(\varepsilon) + \Lambda_a(\varepsilon+\omega,\varepsilon)] . \tag{25}$$

Equation (3) for the no-loss current involves the quantity $\langle\chi_p^{(-)}|\Lambda_v(\varepsilon+\omega,\varepsilon)A_<(\varepsilon)$, which we now can transform as

$$\langle\chi^{(-)}|\Lambda_v(\varepsilon+\omega,\varepsilon)A_<(\varepsilon) = \frac{1}{i\omega}\left[\langle\chi^{(-)}|\Lambda_a(\varepsilon+\omega,\varepsilon)A_<(\varepsilon) - \frac{1}{2\pi}\langle\chi^{(-)}|\mathbf{A}\cdot\nabla\Gamma(\varepsilon)G^\dagger(\varepsilon)\right] . \tag{26}$$

Compared to the corresponding result in Eq. (23) obtained without vertex corrections, we see that now the large symmetry-breaking terms have disappeared. We also note that Eq. (26) correctly involves the dynamically screened potential $\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w$ via the scalar vertex Λ_a. However, a small term symmetry-breaking term proportional to the hole lifetime still remains. By neglecting this last term and by transforming $\Lambda^\dagger$ in a similar way as above, we obtain

$$\langle\chi_p^{(-)}|\Lambda_v(\varepsilon+\omega,\varepsilon)A_<(\varepsilon)\Lambda_v^\dagger(\varepsilon+\omega,\varepsilon)|\chi_p^{(-)}\rangle = \frac{1}{\omega^2}\langle\chi^{(-)}|\Lambda_a(\varepsilon+\omega,\varepsilon)A_<(\varepsilon)\Lambda_a^\dagger(\varepsilon+\omega,\varepsilon)|\chi^{(-)}\rangle + O(\langle\Gamma(\varepsilon)\rangle) , \tag{27}$$

i.e., our expression in Eq. (3) gives a correct description of the selection rules and transforms to its acceleration-formula counterpart to within terms of the order of the hole lifetime width.

C. Alternative treatment based on the Bethe-Salpeter equation

The equation-of-motion method used in the previous subsection to relate the vector-coupling and scalar vertices is simple, but it gives little hint if and when the important relation in Eq. (25) holds in approximate schemes. This relation is in turn the necessary and sufficient condition for having correct selection rules. We shall now give an alternative derivation based on the Bethe-Salpeter equation which allows us to give criteria for the validity of Eq. (25).

The vector-coupling vertex Λ_v fulfills a Bethe-Salpeter equation of the form[8]

$$\Lambda_v(12) = \Delta(12) - i\int d(4567)I(14;25)\times G(64)G(57)\Lambda_v(76) \tag{28}$$

(1, 2, etc., represent space, spin, and time coordinates). Here

$$\Delta(12) = -i\delta(12)\mathbf{A}(1)\cdot\nabla_1 , \tag{29}$$

and

$$I(12;34) = i\delta[\delta(13)V_H(1)+\Sigma(13)]/\delta G(42) \tag{30}$$

is the irreducible scattering function. ($V_H = V_C - w$ is the Hartree potential from the electrons.) Going over to Fourier space and a convenient matrix notation we can

rewrite Eq. (28) as

$$\Lambda_v(\omega)=\Delta-iI(\omega)(GG)_\omega\Lambda_v(\omega)$$
$$=\{1-iI(\omega)(GG)_\omega+[-iI(\omega)(GG)_\omega]^2+\cdots\}\Delta\ . \tag{31}$$

Here $\Lambda_v(\omega)$ is a vector labeled by $(\mathbf{x},\mathbf{x}',\varepsilon)$ and with the components

$$\langle\,\mathbf{x},\mathbf{x}',\varepsilon\,|\,\Lambda_v(\omega)\,\rangle=\int dt\,dt' e^{i(\varepsilon+\omega)t}e^{-i\varepsilon t'}\Lambda_v(\mathbf{x}t,\mathbf{x}'t')\ ,$$

and $I(\omega)$ and $(GG)_\omega$ are matrices with first and second labels $(\mathbf{x},\mathbf{x}',\varepsilon)$ and $(\mathbf{y},\mathbf{y}',\varepsilon')$ with the components

$$I(\mathbf{x},\varepsilon+\omega,\mathbf{y}',\varepsilon';\mathbf{x}',\varepsilon,\mathbf{y},\varepsilon'+\omega)$$

and

$$2\pi\delta(\varepsilon-\varepsilon')G(\mathbf{x},\mathbf{y},\varepsilon+\omega)G(\mathbf{y}',\mathbf{x}',\varepsilon)\ ,$$

respectively. Matrix multiplication involves integration with respect to energies and spatial coordinates (ω is a parameter).

We begin by considering the quantity

$$\langle\varepsilon'\,|\,(GG)_\omega\Delta\,\rangle=-iG(\varepsilon'+\omega)\mathbf{A}\cdot\nabla G(\varepsilon)\ .$$

Taking $\mathbf{A}$ to be constant and using the equation of motion for the Green's function we obtain

$$(GG)_\omega\Delta=\frac{1}{i\omega}(R_a+R_b+R_c)\ ,$$

where

$$\langle\varepsilon'\,|\,R_a(\omega)\,\rangle=\mathbf{A}\cdot\nabla G(\varepsilon')-G(\varepsilon'+\omega)\mathbf{A}\cdot\nabla\ ,$$
$$\langle\varepsilon'\,|\,R_b(\omega)\,\rangle=G(\varepsilon'+\omega)[\Sigma(\varepsilon'+\omega)\mathbf{A}\cdot\nabla-\mathbf{A}\cdot\nabla\Sigma(\varepsilon')$$
$$-\mathbf{A}\cdot(\nabla V_H)]G(\varepsilon')\ ,$$
$$\langle\varepsilon'\,|\,R_c(\omega)\,\rangle=G(\varepsilon'+\omega)[-\mathbf{A}\cdot(\nabla w)]G(\varepsilon')\ .$$

We next make use of the fact that I is the variational derivative of $V_H+\Sigma$ with respect to G [Eq. (30)]. (The functional $\Sigma[G]$ is defined via the expansion in skeleton diagrams.) This gives

$$-iI(\omega)R_a(\omega)=-(GG)_\omega^{-1}R_b(\omega)\ . \tag{32}$$

We now rearrange the expansion in Eq. (31) as follows:

$$\Lambda_v(\omega)=\frac{1}{i\omega}\sum_0^\infty[-iI(\omega)(GG)_\omega]^n(-\mathbf{A}\cdot\nabla w)$$
$$+\frac{1}{i\omega}[i\omega\Delta+\mathbf{A}\cdot\nabla w-iI(\omega)R_a(\omega)]$$
$$-\frac{1}{\omega}I(\omega)\sum_0^\infty[-i(GG)_\omega I(\omega)]^n$$
$$\times[R_b(\omega)-i(GG)_\omega I(\omega)R_a(\omega)]\ .$$

The third term above vanishes by virtue of Eq. (32). The second term gives

$$\Delta-(GG)_\omega^{-1}R_b(\omega)+\mathbf{A}\cdot\nabla w\ ,$$

and the first term gives the scalar vertex function Λ_a defined in Eq. (18). We write the final result in our earlier notation with Green's functions and vertices regarded as matrices in one-electron labels:

$$\Lambda_v(\varepsilon+\omega,\varepsilon)=-i\mathbf{A}\cdot\nabla+\frac{1}{i\omega}[\mathbf{A}\cdot\nabla\Sigma(\varepsilon)-\Sigma(\varepsilon+\omega)\mathbf{A}\cdot\nabla$$
$$+\mathbf{A}\cdot\nabla V_C+\Lambda_a(\varepsilon+\omega,\varepsilon)]\ . \tag{33}$$

This expression is easily seen to be equivalent to Eq. (25) of the previous subsection.

The derivation above relies on having the vertex functions Λ_v and Λ_a properly defined as variational derivatives of the Green's function. This condition is thus necessary and sufficient in order to have correct selection rules and correct relations between the velocity and acceleration formulas.

D. Approximation without local-field effects

In the foregoing analysis the vector potential $\mathbf{A}$ represented the applied external field rather than the total screened field $\mathbf{A}_{\rm eff}(\mathbf{r})$. A slight modification of the method used in Sec. III C gives an approximate acceleration formula for the no-loss current where the spatial dependence of $\mathbf{A}_{\rm eff}$ and thus the local field effects are neglected. In the present case we write the Bethe-Salpeter equation in terms of the effective field,

$$\Lambda_v(12)=\tilde{\Delta}(12)-i\int d(4567)\tilde{I}(14;25)G(64)G(57)\Lambda_v(76),$$

where

$$\tilde{\Delta}(12)=-i\delta(12)\mathbf{A}_{\rm eff}(1)\cdot\nabla_1\ ,$$

and where

$$\tilde{I}(12;34)=i\Sigma(13)/\delta G(42) \tag{34}$$

is the screened counterpart to I. An analysis analogous to that in Sec. III C gives

$$\Lambda_v(\omega)=\frac{1}{i\omega}\sum_0^\infty[-i\tilde{I}(\omega)(GG)_\omega]^n(-\mathbf{A}_{\rm eff}\cdot\nabla V_c)$$
$$+\frac{1}{i\omega}[i\omega\tilde{\Delta}+\mathbf{A}_{\rm eff}\cdot\nabla V_c-i\tilde{I}(\omega)R_\alpha(\omega)]$$
$$-\frac{1}{\omega}\tilde{I}(\omega)\sum_0^\infty[-i(GG)_\omega\tilde{I}(\omega)]^n$$
$$\times[R_\beta(\omega)-i(GG)_\omega\tilde{I}(\omega)R_\alpha(\omega)]\ . \tag{35}$$

Here

$$\langle\varepsilon'\,|\,R_\alpha(\omega)\,\rangle=\mathbf{A}_{\rm eff}\cdot\nabla G(\varepsilon')-G(\varepsilon'+\omega)\mathbf{A}_{\rm eff}\cdot\nabla\ ,$$
$$\langle\varepsilon'\,|\,R_\beta(\omega)\,\rangle=G(\varepsilon'+\omega)[\Sigma(\varepsilon'+\omega)\mathbf{A}_{\rm eff}\cdot\nabla$$
$$-\Sigma(\varepsilon')\mathbf{A}_{\rm eff}\cdot\nabla]G(\varepsilon')\ , \tag{36}$$

and $\mathbf{A}_{\rm eff}$ is now taken to be constant. From Eq. (34) we find as above

$$-i\tilde{I}(\omega)R_\alpha(\omega)=-(GG)^{-1}_\omega R_\beta(\omega)\ ,$$

and thus the third term in Eq. (35) vanishes. The first term gives the response in G^{-1} to an increment $\mathbf{A}_{\rm eff}\cdot\nabla V_C$ in the *total* potential (V_C), and we obtain the second term as in Sec. III C. The final result is

$$\Lambda_v(\varepsilon+\omega,\varepsilon)=\frac{1}{i\omega}[G^{-1}(\varepsilon+\omega)\mathbf{A}_{\rm eff}\cdot\nabla - \mathbf{A}_{\rm eff}\cdot\nabla G^{-1}(\varepsilon)+\tilde{\Lambda}_a(\varepsilon+\omega,\varepsilon)]\ . \tag{37}$$

Here

$$\langle\,\mathbf{x}\,|\,\tilde{\Lambda}_a(\varepsilon+\omega,\varepsilon)\,|\,\mathbf{x}'\,\rangle=\int d^3y\,\tilde{\Lambda}_0(\mathbf{x}\varepsilon+\omega,\mathbf{x}'\varepsilon;\mathbf{y})\times[-\mathbf{A}_{\rm eff}\cdot\nabla V_C(\mathbf{y})] \tag{38}$$

involves $\mathbf{A}_{\rm eff}\cdot\nabla V_C$, whereas the exact expression in Eq. (19) involves $\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w$.

Going through the same steps as in Sec. III C, we now obtain the following approximate acceleration formula for the no-loss photocurrent:

$$J_p^{(e)}=\int d\varepsilon\,2\pi\delta(\varepsilon_p-\varepsilon-\omega)\frac{1}{\omega^2}\langle\chi_p^{(-)}\,|\,\tilde{\Lambda}_a(\varepsilon+\omega,\varepsilon)A_<(\varepsilon)\tilde{\Lambda}_a^\dagger(\varepsilon+\omega,\varepsilon\,|\,\chi_p^{(-)}\rangle+O(\langle\Gamma(\varepsilon)\rangle)\ . \tag{39}$$

Thus, neglecting local-field effects amounts in the acceleration formula to replacing $\epsilon^{-1}(\omega)\mathbf{A}\cdot\nabla w$ by $\mathbf{A}_{\rm eff}\cdot\nabla V_C$.

IV. CONSISTENT APPROXIMATIONS FOR THE PHOTOEMISSION PROCESS

As shown above, approximations to the no-loss current obtained by choosing the vertex consistent with the approximate Green's function used give correct selection rules and equivalence between the velocity and acceleration formulas. This requirement is fulfilled for so-called Φ-derivable approximations.[3] In such an approximation the self-energy $\Sigma=\Sigma[G]$ is derivable from a scalar functional $\Phi[G]$, $\Sigma(12;G)=\delta\Phi[G]/\delta G(21)$. The self-energy thus defined is furthermore required to be consistent with G, i.e.,

$$G^{-1}=G_0^{-1}-\Sigma\ , \tag{40}$$

where G_0 is a suitably defined zero-order Green's function. The vertices are then taken as the appropriate variational derivatives, which leads to Bethe-Salpeter equations with approximate kernels (I). The simplest approximations of this kind are the time-dependent Hartree-Fock[9] and local-density[10] approximations, which correspond to

$$\Phi[G]=\tfrac{1}{2}\int d(12)iG(12)G(21)v(12)$$

and

$$\Phi[G]=\int d(1)\varepsilon_{\rm xc}(-iG(11^+))\ ,$$

respectively. [Here $v(12)$ is the Coulomb interaction and $\varepsilon_{\rm xc}$ is the exchange-correlation energy density in the homogeneous electron gas.] However, neither of these approximations accounts for the important damping effects, and Φ-derivable approximations beyond these simplest examples are hard to evaluate.

The condition of Φ derivability is actually too strong. For instance, we can consider the zero-order propagator G_0 as a functional of the total effective field, take Σ as a functional of G_0, and finally define G via the Dyson equation (40). In this way G depends on the external field only via its dependence on G_0. We then take, as before, the vertex functions as the correct variational derivatives. This guarantees the validity of Eq. (25) or Eq. (37). To obtain a consistent approximation to the no-loss photocurrent we must then clearly define the density of occupied states [$A_<(\varepsilon)$] and the photoelectron orbital via the same G as that used for the vertex part. A sensible approximation of this sort with damping effects is the time-dependent version of the "G_0W" approximation,[11] where

$$\Sigma(12;G_0)=iG_0(12)W(21)\ ,$$

$$W=v(1-Pv)^{-1}\ ,$$

and

$$P(12)=-iG_0(12)G_0(21)\ .$$

The time-dependent G_0W approximation gives correct selection rules, but its vertex function involves W to second order and is thus rather difficult to evaluate in practice. A simpler approximation is obtained by neglecting the functional dependence of W on G_0, i.e., by taking W as given. As will be seen shortly we must then take W as translationally invariant and frequency independent in order to have the correct relation between Λ_v and Λ_a or $\tilde{\Lambda}_a$ [Eq. (25) or (37)]. If the ω dependence in W is kept we obtain an approximation where the selection rule violations are much reduced compared to the theory in Eq. (1). Let us, for simplicity, confine ourselves to the approximation without local-field effects, and define G_0 by the implicit relation

$$\left[i\frac{\partial}{\partial t_1}+\tfrac{1}{2}\nabla_1^2-V_C(1)\right]G_0(12)=\delta(12)\ .$$

The Bethe-Salpeter equation is now replaced by the much simpler

$$\Lambda_v(12)=\tilde{\Delta}(12)+\int d(4567)[\delta\Sigma(12)/\delta G_0(54)]\times G_0(64)G_0(57)\tilde{\Delta}(76)\ ,$$

and a corresponding equation for $\tilde{\Lambda}_a$, where, in the present case,

$$\delta\Sigma(13)/\delta G_0(42)=iW(31)\delta(14)\delta(23)\ .$$

We explicitly find

$$\langle \mathbf{x} \mid \Lambda_v(\varepsilon+\omega,\varepsilon) \mid \mathbf{x}' \rangle = \langle \mathbf{x} \mid -i\mathbf{A}_{\text{eff}}\cdot\nabla \mid \mathbf{x}' \rangle + i\int \frac{d\varepsilon'}{2\pi} \langle \mathbf{x} \mid W(\varepsilon-\varepsilon') \mid \mathbf{x}' \rangle \langle \mathbf{x} \mid G_0(\varepsilon'+\omega)(-i\mathbf{A}_{\text{eff}}\cdot\nabla)G(\varepsilon') \mid \mathbf{x}' \rangle ,$$

and using the relation

$$G_0(\varepsilon'+\omega)(-i\mathbf{A}_{\text{eff}}\cdot\nabla)G(\varepsilon') = \frac{1}{i\omega}[\mathbf{A}_{\text{eff}}\cdot\nabla G_0(\varepsilon) - G_0(\varepsilon'+\omega)\mathbf{A}_{\text{eff}}\cdot\nabla + G_0(\varepsilon'+\omega)\mathbf{A}_{\text{eff}}\cdot\nabla V_C G_0(\varepsilon')]$$

we then readily verify Eq. (37) when W is translationally invariant and ω independent. If the ω dependence is kept, Eq. (37) would not be exactly fulfilled, but the violation could be shown to be small compared to the symmetry-breaking term $\mathbf{A}\cdot\nabla\Sigma(\varepsilon) - \Sigma(\varepsilon+\omega)\mathbf{A}\cdot\nabla$ inherent in Eq. (1).

V. RELATION TO OPTICAL ABSORPTION

Our analysis so far has shown the importance of mutually consistent approximations for self-energies and the corresponding vertex functions in the description of the photoemission process. The situation is, of course, rather similar for the case of optical absorption. It follows from Refs. 2 and 3 that consistent approximations are needed in order to fulfill the condition of particle conservation and thus the f-sum rule. We now explicitly show that consistent approximations are equally important in order to have the optical selection rules obeyed in the velocity formula for photoabsorption by inhomogeneous systems.

The velocity formula for photoabsorption can be written

$$I(\omega) = \frac{1}{\pi}\int_{-\infty}^{\infty} d\varepsilon \,\mathrm{Re}\,\mathrm{Tr}[-i\mathbf{A}\cdot\nabla G(\varepsilon+\omega)\Lambda_v(\varepsilon+\omega,\varepsilon)G(\varepsilon)] . \tag{41}$$

In order to bring out the selection rules, we transform this expression to its acceleration-formula counterpart. Using Eq. (25) we obtain $I(\omega) = I_1(\omega) + I_2(\omega)$, where

$$I_1(\omega) = \frac{1}{\pi}\int_{-\infty}^{\infty} d\varepsilon \frac{1}{\omega}\mathrm{Re}\,\mathrm{Tr}[(-i\mathbf{A}\cdot\nabla)^2 G(\varepsilon) - (-i\mathbf{A}\cdot\nabla)G(\varepsilon+\omega)(-i\mathbf{A}\cdot\nabla)] ,$$

and where

$$I_2(\omega) = \frac{1}{\pi}\int_{-\infty}^{\infty} d\varepsilon \,\mathrm{Re}\frac{1}{i\omega}\mathrm{Tr}[-i\mathbf{A}\cdot\nabla G(\varepsilon+\omega)\Lambda_a(\varepsilon+\omega,\varepsilon)G(\varepsilon)] . \tag{42}$$

By using the cyclic property of the trace we see that $I_1 = 0$. Since Λ_a involves $\mathbf{A}\cdot\nabla w$, we have already verified that any approximation which satisfies Eq. (25) also gives correct selection rules. To obtain the final acceleration formula we rewrite the trace in Eq. (42) in the matrix notation used earlier and use the Bethe-Salpeter equation to obtain [Δ_a is a vector with components $-\mathbf{A}\cdot\nabla w(1)\delta(12)$]

$$\mathrm{Tr}[-i\mathbf{A}\cdot\nabla G(\varepsilon+\omega)\Lambda_a(\varepsilon+\omega,\varepsilon)G(\varepsilon)] \equiv \Delta(GG)_\omega\Lambda_a(\omega) = \Delta\sum_0^\infty [-i(GG)_\omega I(\omega)]^n (GG)_\omega\Delta_a = \Lambda_v(-\omega)(GG)_\omega\Delta_a$$

$$\equiv \mathrm{Tr}[\Lambda_v(\varepsilon,\varepsilon+\omega)G(\varepsilon+\omega)(-\mathbf{A}\cdot\nabla w)G(\varepsilon)] .$$

We then apply Eq. (25) once more to obtain

$$I(\omega) = \frac{1}{\pi\omega^2}\int_{-\infty}^{\infty} d\varepsilon \,\mathrm{Re}\,\mathrm{Tr}[\Lambda_a(\varepsilon,\varepsilon+\omega)G(\varepsilon+\omega)(-\mathbf{A}\cdot\nabla w)G(\epsilon)] . \tag{43}$$

Thus we see that the acceleration and velocity formulas are equivalent in approximations where the vertices are variational derivatives of the Green's function. In particular, they are equivalent in the time-dependent Hartree-Fock and local-density approximations.

By leaving out the vertex part we evidently obtain symmetry-breaking terms involving $\Sigma(\varepsilon+\omega)\mathbf{A}\cdot\nabla - \mathbf{A}\cdot\nabla\Sigma(\varepsilon)$ in a similar way as was found in the case of photoemission. Thus, the vertex part Λ_v is indeed needed in the velocity formula of photoabsorption, as soon as self-energy effects are included, in order to have the selection rules obeyed.

In a similar way as in Sec. III D, we can also obtain an approximate acceleration formula without local-field effects. The derivation is obvious, so we just state the result:

$$I(\omega) = \frac{1}{\pi\omega^2}\int_{-\infty}^{\infty} d\varepsilon \,\mathrm{Re}\,\mathrm{Tr}[\tilde{\Lambda}_a(\varepsilon,\varepsilon+\omega)G(\varepsilon+\omega)(-\mathbf{A}\cdot\nabla V_C)G(\varepsilon)] . \tag{44}$$

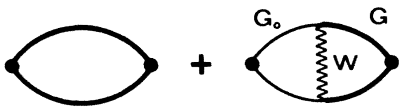

FIG. 1. Simple approximation to optical absorption without local-field effects.

In the simplified time-dependent G_0W scheme discussed at the end of the previous section, we obtain the approximation illustrated in Fig. 1.

VI. CONCLUDING REMARKS

We have in this work shown that the current practice to compute photoelectron spectra of real materials involves quite severe inconsistencies, and we have shown how the problems may be resolved on an *a priori* level. Our analysis has been based on an expression for the no-loss photocurrent given in I involving only the Green's function and its first-order response to the external optical field.

We have also discussed various approximate schemes and given criteria for having a consistent description and correct selection rules. We have made connections between the commonly used acceleration formula for photoemission and well-defined approximations which leave out the local-field effects, and obtained a simple first-order approximation based on a time-dependent G_0W approach.

The concept of a no-loss current requires the hole lifetime width to be small in the spectral region under discussion. The energy losses then enter only as virtual processes which modify the distribution of unscattered electrons. In spectral regions where the hole lifetime width is large, on the other hand, real energy losses are also both possible and important. In that case we cannot see any obvious way to identify a no-loss part, neither theoretically nor experimentally. Instead one must then analyze the photocurrent as a whole. The approach in I allows for such a description, but, in that case, true second-order responses also enter.

ACKNOWLEDGMENT

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