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Effect of interactions on deep-hole interferences in photon-induced Auger emission from solids

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Résumé. — Les interférences de trous profonds dans l'émission Auger des solides induite par photons ont été mises en évidence dans un article précédent. Nous étudions ici l'effet des interactions sur ces interférences. Les interactions qui ne conservent pas le nombre de trous profonds ont essentiellement pour effet de limiter le temps de vie du trou. Celles (comme l'émission de phonons ou la diffusion des électrons de conduction) qui conservent le nombre de trous induisent une décorrélélation des phases des trous qui donnent lieu à l'interférence. Nous montrons que les temps de décorrélération correspondants sont beaucoup plus grands que les temps de vie de trou caractéristiques, de sorte que les effets d'interférence devraient contribuer de façon appreciable au courant Auger, sauf éventuellement dans le cas d'interférences extra-atomiques entre niveaux profonds ayant des largeurs assez faibles (de l'ordre de 50 meV).

Abstract. — Deep-hole interferences in photon-induced Auger emission from solids have been discussed in a previous paper. We study the effect of interactions on these interferences. Interactions which do not conserve the number of deep-holes essentially give rise to the hole lifetime, while those (phonon emission, conduction electron scattering) which conserve the number of holes induce a decorrelation of the phases of interfering holes. We show that the associated decorrelation times are much larger than the hole lifetime, so that interference effects should contribute non-negligibly to Auger currents, except possibly in the case of extraatomic interferences between deep levels with widths in the 50 meV range.

1. Introduction. — In a recent paper [1] hereafter referred to as (I), we have shown that photon-induced Auger emission is not simply an additive superposition of the emission of the various degenerate deep-hole levels. Indeed, the phase coherence between the holes, which results from the creation process (optical absorption of monochromatic X-rays), gives rise to interferences between Auger processes involving the various degenerate (intermediate) hole states. In (I), the effect of interactions was only treated phenomenologically, in terms of lifetimes of the various levels involved in the hole creation and Auger processes.

It was then shown that one must distinguish between two types of interferences: intraatomic ones, between degenerate hole states localized on the same atomic site, which are unaffected by lifetime effects, and extraatomic ones (occurring only if one at least of the final shallow hole states of the Auger transition lies in a wide band). Lifetime and/or mean free path effects limit their space range, for the deepest hole states, to distances of atomic order in light materials, while virtually suppressing them in heavy ones.

In this article we intend to study in more detail the effect of interactions on these interferences, in order to have a more reliable estimate of their importance in the interpretation of experimental results.

Clearly, it is out of our reach to treat properly the effect of interactions on the five levels involved in the photon-induced Auger process. This, however, is not necessary for our purpose, which is simply to understand whether interactions may decrease appreciably the relative contribution of interference terms, with respect to non-interferent ones.
Interferences result from the phase coherence between degenerate deep-hole states. It is this phase correlation which carries to the Auger process the memory of the hole creation event. Therefore (and since, as was shown in (I), the loss of phase coherence due to the finite mean free paths of the final states is in general small compared with the effect of the deep-hole lifetime $T_1$) the decorrelation effects in which we are interested are governed by the interactions involving the deep-hole. For this reason, we will only retain these in our model.

Interferences will survive if the phase coherence lasts for a time $T_2$ larger than or comparable with the hole lifetime $T_1$. This remark leads us to distinguish, as is done in magnetism when one separates longitudinal and transverse relaxations, between two types of interactions:

(i) those which do not conserve the number of deep-holes (radiative recombination and Auger processes themselves) which give rise to the longitudinal relaxation time $T_1$.

(ii) those which conserve the number of deep-holes (phonon-hole interaction and scattering of conduction electrons by the hole potential). These only contribute to the transverse relaxation time $T_2$.

Non-conserving interactions can also contribute to phase decorrelation (either by inducing indirect interactions between deep-holes or by allowing for hole creation processes involving the decay of a deeper hole). These effects, obviously, are always of higher order than those giving rise to $T_1$, and thus give decorrelation times much larger than $T_1$. Consequently, interactions of type (i) are reasonably well taken into account by the semi-phenomenological lifetime approximation of (I).

On the other hand, deep-hole conserving interactions of course modify the shape of the deep-hole spectral density, which reacts on the line shape of both interferent and non-interferent Auger terms. But their main effect, for our purpose, is to give rise to phase decorrelation between the deep-holes, i.e. to reduce the total strength of interference contributions: indeed, they can be interpreted as arising from fields (of atomic motion, or electric fields due to conduction electron motion) which act to modulate randomly the phases of the various hole states.

In paragraph 2, we derive the formal expression of the Auger current for a hole level without intratomic degeneracy and with the approximations described above. Namely, we only retain interactions involving the deep-hole, those of type (i) are treated in terms of a phenomenological lifetime, and only those of type (ii) are treated explicitly. We apply this to discuss the effect of phonons and of conduction electron scattering on extraatomic interferences. We show that, for not too narrow hole levels, the resulting decorrelation times are larger than the hole lifetime, even for nearest neighbour interference terms which should therefore, as discussed in (I), contribute non-negligibly to the Auger emission of light materials.

In paragraph 3, we discuss the same question for intraatomic interferences in the case of an atomically degenerate deep-hole level. This new degree of freedom gives rise to further complications of the Kondo type, so that only a qualitative estimate is possible. It is shown that, due to the smallness of the relevant couplings and of the corresponding random modulation of relative phases, the resulting $T_2$'s are much larger than in the non-degenerate case, giving a negligible reduction of interferences.

Finally, we briefly discuss how the existence of interference contributions, which should therefore be present as predicted in (I), could be checked experimentally.

2. Non degenerate deep-hole : effect of interactions on extraatomic interferences. — We assume the deep level $|h\rangle$ (energy $\omega_h$) to have no atomic degeneracy, so that the only possible interferences are extraatomic, arising from holes on different atomic sites.

To lowest order in the Coulomb interaction, the Auger current flowing in direction $\vec{R}$ at energy $\omega$ is described (see (I)) by the bare diagrams of figure 1, corresponding respectively to the direct (Fig. 1a) and exchange (Fig. 1b) contributions. Interferences are coming from the terms where the two deep-hole (double) lines correspond to two holes on different atomic sites. This can occur only if at least one of the final hole states of the Auger transition (lines $\omega_1$ and $\omega_2$) belongs to a wide band, which we assume from now on.

![Fig. 1. — The two lowest order diagrams describing the photon-induced Auger current. Wavy lines stand for the electromagnetic field, full lines for electron propagators, double lines for deep-hole propagators, and dotted ones represent the Coulomb interaction. (1a) Direct current ; (1b) Exchange current.](image-url)
We want to renormalize selectively the deep-hole part of these diagrams, which is obviously the same for the direct and exchange Auger terms. As stated in paragraph 1, we only treat explicitly interactions which conserve the number of deep-holes. Moreover, for all the interactions of interest here (phonon emission and absorption, conduction electron scattering), due to the negligible overlap of hole wavefunctions centred on different sites, only the terms which conserve the number of holes on a given site — i.e. diagonal in site indices — are of importance. Therefore, the site index is conserved everywhere along a given deep-hole line. (In other words, the physical connection between two holes on different sites can only be made via a phonon or a conduction electron, since hole jumps are negligible.) Each hole line therefore has no internal structure (accessible degree of freedom). In such a case, the method set up by Nozières and De Dominicis [2] to treat X-ray absorption edge singularities, and developed by Nozières and Abrahams [3] in the case, closely analogous to the present one, of Raman scattering, can be applied directly.

Since this method gives a simple expression of hole renormalization effects in the time representation, we rewrite the expression of the direct Auger current, measured at energy \( \omega \), at \( R \to \infty \) in direction \( \hat{R} \), in the following way:

\[
 j_d^{\text{Auger}}(\hat{R}, \omega) = K_{\omega} \sum_{n, p \in P} \langle np | V | h_i \phi_{> \to \to} \rangle \langle h_i | a \cdot \nabla | s \rangle \langle s | a \cdot \nabla | h_j \rangle x \int \frac{d\omega'}{2\pi} \frac{d\omega''}{2\pi} \theta(\omega - E)
\]

where the notations are those of (I), namely: \( V \) is the Coulomb interaction, \( \mu \) the Fermi energy of the solid, \( E \) the vacuum energy;

\[
 K_{\omega} = \frac{-e m}{\hbar^2} \left( \frac{e}{mc} \right)^2 k_{ao}, \quad k_{ao} = \left[ \frac{2 m}{\hbar^2} (\omega - E) \right]^{1/2},
\]

| \( h_i \rangle \) corresponds to the atomic deep-hole state localized on site \( i \), \( n \) and \( p \) run on the allowed final shallow hole states (of energies \( \epsilon_n, \epsilon_p \)) appropriate to the particular given Auger transition. \( s \) labels the primary electron (excited by the optical transition) and \( \phi_{> \to \to} \) is the LEED wavefunction corresponding to the emitted Auger electron. An analogous expression (differing from eq. (1) only by the standard interchange of Auger matrix elements, see (I)) stands for the exchange current \( j_e(\hat{R}, \omega) \).

\[
 S_{ij}(\omega_1, \omega_2; \omega_3, \omega_4) \text{ is the fully renormalized propagator of a pair of deepholes located on sites } i \text{ and } j.
\]

Fig. 2. — Schematic representation of the fully renormalized propagator of a pair of deep-holes located on sites \( i \) and \( j \):

\[
 S_{ij}(t_1, t_2; t_3, t_4) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3 S_{ij}(t_1, t_2; t_3, 0) \exp[-i\omega'(t_1 - t_2) + i\omega''t_3]
\]

where we have made use of the time translational invariance (energy conservation \( \omega_1 - \omega_2 = \omega_3 - \omega_4 \)) in the system at equilibrium to choose \( t_4 = 0 \).

In the absence of interactions, as shown in (I), \( S_{ij}(t_1, t_2; t_3, 0) \) simply reduces to the product of free hole Keldysh [4] causal and anticausal propagators:

\[
 S_{ij}^{(0)}(t_1, t_2; t_3, 0) = G_{h_{i}}^{(+)}(t_3 - t_1) G_{h_{j}}^{(-)}(t_2) = -e^{-i\omega(t_3 - t_1)} \theta(t_1 - t_3) e^{-i\omega t_2} \theta(t_2).
\]

Note that the \( \theta \)-functions in eq. (3) account for the fact that \( G_h \) describes the propagation of a hole (\( \epsilon_h < \mu \)). This also entails that, at zero temperature, the electron-occupation propagator \( G_{h_{i}}^{(+)}(t) = 0 \) for
the (h) level. Therefore, the Kjeldysh index, which is fixed by the conditions \( \omega > \mu, \epsilon_n \) and \( \epsilon_q < \mu \) to be (+) at vertex \( t_1 \) and (−) at vertex \( t_2 \), is conserved everywhere along a given hole line, even in the presence of this. This remains true in practice at all temperatures of interest, due to the high binding energy of the deep-holes.

As stated above, we include the effect of the interactions which do not conserve the number of holes as a lifetime effect, so that eq. (3) becomes :

\[
\tilde{S}_{ij}^{(0)}(t_1, t_2 ; t_3, 0) = - \exp \left[ -i g_\lambda(t_3 - t_1 + t_2 - t_3) - \frac{1}{\omega q_\lambda} \left( a_{q_\lambda} + a_{q_\lambda}^\dagger \right) g_{\lambda q_\lambda} \right] \times \frac{1}{\theta(t_1 - t_3) \theta(t_2)} .
\]

2.1 INTERACTION WITH PHONONS. — Following Hedin [5], and Hedin and Rosengren [6], the interaction Hamiltonian which describes the fluctuation of Coulomb potential at the core electrons reads :

\[
H_{\text{ph}} = \sum_{q, \lambda} \left( \frac{\hbar^2}{2 \omega_{q_\lambda} N M} \right)^{1/2} \times e^{i q \cdot R_{ij}} \psi_i^\dagger (a_{q_\lambda} + a_{q_\lambda}^\dagger) g_{\lambda q_\lambda}
\]

where \( M \) is the atomic mass ; \( \omega_{q_\lambda} \) is the frequency of a phonon of momentum \( q \) and polarization \( \lambda \), \( a_{q_\lambda} \) and \( a_{q_\lambda}^\dagger \) are the corresponding annihilation and creation operators, and \( \psi_i \) (resp. \( \psi_i^\dagger \)) annihilates (resp. creates) an electron at site \( i \) in the deep h level. \( R_{ij} \) is the position of the \( j \)th atomic site, and \( R_{ij} \) is the unit vector along the direction of vector \( \mathbf{R} \) is the derivative of the screened pseudopotential \( w \) of an ion.

Finally, \( w(R) \) is the derivative of the screened pseudopotential \( w \) of an ion.

As shown in references [2] and [3], in any diagram of renormalization of the two-hole propagator \( S_{ij}(t_1, t_2 ; t_3, 0) \) (see for example figure 3), the phase factors corresponding to the structureless hole lines factor out into a contribution \( \tilde{S}_{ij}^{(0)}(t_1, t_2 ; t_3, 0) \) common to all diagrams. The linked cluster theorem then applies to the calculation of the exact contribution of all phonon lines, giving :

\[
S_{ij}(t_1, t_2 ; t_3, 0) = \tilde{S}_{ij}^{(0)}(t_1, t_2 ; t_3, 0) \exp \left\{ C_{ij}^{(+)}(t_1 - t_3) + C_{ij}^{(-)}(t_2) + C_{ij}^{(+)}(t_1, t_2, t_3) \right\}
\]

where \( \exp \left\{ C_{ij}^{(+)}(t_1 - t_3) \right\} \) is the complete contribution of all the phonon lines which are connected to the hole line \( (i) \) only (Fig. 3).

\[
C_{ij}^{(+)}(t_1 - t_3) = -i \sum_{q_\lambda} g_{q_\lambda}^2 \int_{t_3}^{t_1} dt' D_{q_\lambda}^{(+)}(t - t')
\]

\[
D_{q_\lambda}^{(ab)}(t) \text{ is the } (a,b) \text{ component of the phonon Keldysh matrix propagator, } \{ a, b \} = \{ +, - \}. \text{ For example :}
\]

\[
D_{q_\lambda}^{(+)}(t - t') = -i \left( \frac{\hbar^2}{2 \omega_{q_\lambda} N M} \right) \langle T \{ (a_{q_\lambda}(t) + a_{-q_\lambda}(t)) (a_{q_\lambda}(t') + a_{-q_\lambda}(t')) \} \rangle .
\]

Analogously, \( \exp \left\{ C_{ij}^{(-)}(t_2) \right\} \) gives the separate renormalization of the \( (j) \) hole line :

\[
C_{ij}^{(-)}(t_2) = -i \sum_{q_\lambda} g_{q_\lambda}^2 \int_{t_0}^{t_2} dt' D_{q_\lambda}^{(-)}(t - t')
\]

while \( \exp \left\{ C_{ij}^{(+)}(t_1, t_2, t_3) \right\} \) accounts for the contribution of all the lines which connect lines \( (i) \) and \( (j) \) :

\[
C_{ij}^{(+)}(t_1, t_2, t_3) = i \sum_{q_\lambda} g_{q_\lambda}^2 e^{i q \cdot R_{ij}} \int_{t_3}^{t_1} dt' \int_{t_0}^{t_2} dt'' D_{q_\lambda}^{(+)}(t - t'')
\]

\[ R_{ij} = R_j - R_i \] is the distance between the two interfering holes.

Clearly, given a model for phonon dispersion and damping, one can straightforwardly calculate the integrals in eqs. (7), (9), (10), which gives the complete expression of \( S_{ij} \) and the corresponding effect on the Auger spectrum.

One finds, as can be expected, that the Auger line
shape is changed. In particular, phonon satellites appear on both sides of the main peak: normal satellites below the central line essentially come from the renormalization of the deep-hole spectral density; the anomalous ones (above the main peak) result from a shake-up effect—i.e. from the sudden creation of the hole, which therefore has not completely reached its phonon dressed equilibrium state when the Auger transition takes place. These anomalous satellites are analogous to those predicted by Watts [7] in his study of plasmon effects.

We will not calculate here the detailed line shape (which should in fact include the effect of the coupling of phonons to all the electronic levels involved), but, as stated in paragraph 1, will concentrate on calculating the total strength of the (deformed) Auger line. Indeed, this simpler quantity is sufficient to characterize the relative strength of interferent \((i \neq j)\) and non-interferent \((i = j)\) contributions.

We define this strength, for the given \((hNP)\) line, by the total current in direction \(\hat{R}\):

\[
J^{hNP}(\hat{R}) = \int d\omega J^{hNP}(\hat{R}, \omega).
\]

The simple result of eq. (12) shows that the relevant \(S_{ij}\)’s (see Fig. 3) reduce to those with \(t_1 = t_2, t_3= 0\) or, more exactly, \(|t_1 - t_2| \ll |t_1 - t_3|\). This is clearly not a severe physical restriction, since the important values of \(|t_1 - t_2|\) and \(|t_3|\), which are respectively the characteristic times for the Auger and optical transitions (of order \(Q^{-1}\) and \((\varepsilon_n + \varepsilon_p - \omega)^{-1}\)), are much smaller than the typical propagation time \(|t_1 - t_3|\), which is of the order of the hole lifetime \(\Gamma^{-1}\) (with \(\Gamma \lesssim 1\) eV).

\[
C_{ij}(\hat{R}) = \sum_{s,n} K_{in} \delta(\omega + Q - \varepsilon_n - \varepsilon_p - \varepsilon_s) \langle h_i | a_i \cdot V | s \rangle \langle s | a_i \cdot V | h_j \rangle \times
\]

\[
\left\{ \langle np | V | h_i \phi_{\omega} \rangle \langle \phi_{\omega} h_j | V | pn \rangle - \frac{1}{2} \langle \phi_{\omega} h_j | V | np \rangle \right\}_{\omega = \bar{\omega}}.
\]

Let us first consider the interaction correction to the non-interferent terms:

\[
S_{ij}(t) = - e^{-2\Gamma t} \theta(t) \times
\]

\[
\exp \left\{ C_{ii}^{(+)}(t) + C_{ii}^{(-)}(t) + C_{ii}^{(+)}(-t) \right\}.
\]

Noticing that the phonon propagators \(D_{q\lambda}^{(+)}(t)\) and \(D_{q\lambda}^{(-)}(t)\) [4] are even functions of time, and that \(D_{q\lambda}^{(+)}(-t) = D_{q\lambda}^{(-)}(-t)\), we get:

\[
C_{ii}^{(+)}(t) + C_{ii}^{(-)}(t) + C_{ii}^{(+)}(-t) = \frac{i}{2} \sum_{q\lambda} |g_{q\lambda}|^2 \int_0^t dr' \int_0^t dr \left[ D_{q\lambda}^{(+)}(r) + D_{q\lambda}^{(-)}(r) - D_{q\lambda}^{(+)}(-r) + D_{q\lambda}^{(-)}(-r) \right]_{t-r'}.
\]

\[
\left(16\right)
\]

since, from the definition of Kjeldysh propagators, one always has:

\[
D^{(+)} + D^{(-)} = D^{(+)} + D^{(-)}.
\]

Therefore, the phonons do not alter the strength of non-interferent Auger terms. This was to be expected since, even if a hole is submitted to a random field which modulates its energy, it remains perfectly autocorrelated. The energy modulation only affects the shape of the associated line. From this interpretation, one may anticipate that this result will be true for any hole-conserving interaction.

On the contrary, for an interferent \((i \neq j)\) term,
This expression parallels the one which is found in the semi-phenomenological treatments of motional narrowing of the nuclear relaxation [8], here $\Im D_{qz}^{(+)}(\tau)$ is the microscopic expression of the correlation function of the random field which gives rise to transverse relaxation. Following these theories, one must compare two times:

(i) The correlation time $\tau_c$ of the fluctuating field — here the phonon one — which is the time range of the phonon propagator appearing in eq. (18). That is, $\tau_c \approx \omega_0^{-1}$, where $\omega_0$ is a Debye frequency, of order $1/40$ eV.

(ii) $T_1 = (2\Gamma)^{-1}$, the lifetime of a hole. $T_1$ gives the scale of values of $t$ which are important in the integral of eq. (12b). For light elements and not too shallow $h$ levels, $\Gamma$ is typically of the order of a few tenths of eV, so that $\tau_c \gg T_1$, and one can set in eq. (19)

$$\int_{-\tau}^{\tau} d\tau \Im D_{qz}^{(+)}(\tau) \approx \tau^2 \Im D_{qz}^{(+)}(\tau = 0) = -\frac{\hbar^2}{2\omega_{qz}NM\Gamma^2(1 + 2b_q)}$$

where $b_q = (\exp(\omega_q/kT) - 1)^{-1}$ is the phonon thermal occupation number, and $\omega_{qz}$ the phonon frequency.

In this regime of slow fluctuations, the dynamics of the fluctuating field is unimportant since the two interfering holes live such a short time that they only see instantaneous configurations of the phonon field. One therefore gets:

$$F_{ij} = \int_0^\infty dt \exp[-2\Gamma t - A_{ij}t^2]$$

$$A_{ij} = \sum_{qz} |g_{qz}|^2 \frac{\hbar^2}{2\omega_{qz}NM} (1 - e^{i\mathbf{q} \cdot \mathbf{R}_{ij}})(1 + 2b_q)$$

and, in order for $F_{ij}$ to be noticeably reduced from its zeroth order value $F_{ij}^{(0)} = (2\Gamma)^{-1}$, one should have a strong modulation amplitude for the hole frequency, such that $A_{ij} \approx (2\Gamma)^2$. $A_{ij}$ can be rewritten as:

$$A_{ij} = \frac{\hbar^2}{16\pi^3\rho} \sum_{(BZ)} d^3q |g_{qz}|^2 \coth \frac{\beta\omega_{qz}}{2} (1 - e^{i\mathbf{q} \cdot \mathbf{R}_{ij}})$$

where $\rho$ is the mass density, $BZ$ the Brillouin zone and $\beta = (kT)^{-1}$. Note that $(R_{ij} \to \infty)$ is precisely the width $\Delta$ of the phonon distribution in the presence of the coupling to the core hole calculated in reference [6]. Hedin and Rosengren have shown that this quantity is, at $T = 0$ K, typically of the order of 20 meV in metals, and that its temperature variation is well represented by the expression:

$$\Delta(T) = \Delta(T = 0)A(\theta_0/T)$$

$$A(x) = \left(1 + 8x^{-4} \int_0^x du \frac{u^3}{\cosh u - 1}\right)^{1/2}$$

where $\theta_0$ is the Debye temperature of the material. This gives typically $\Delta \sim 35$ meV at room temperature.

Of course, the $A_{ij}$'s in which we are interested here are those for small $R_{ij}$'s corresponding to first or possibly second nearest neighbours. Indeed, as was shown in (1), extratomic interferences are limited by lifetime and mean free path effects to a range of atomic order. We can calculate the value $A_{01}$ of $A_{ij}$ for nearest neighbours in the simplified model defined by Hedin and Rosengren [6] (keeping only the contribution of longitudinal phonons, and calculating $\omega_{qz}$ in a nearest neighbour spherical approximation).

For a BCC lattice $(k_B T < 4.25)$ we find $A_{01} \approx 1.2 A$. So, at most, $A_{ij} \lesssim 50$ meV. Thus, for deep levels with widths larger than typically $10^{-1}$ eV, $A_{ij} \ll \Gamma$ and hole decorrelation induced by phonons is negligible.

This should be the case for all but the most shallow levels. Some of these shallow levels (e.g. the L3 level of Al) have widths in the 50 meV range. If this is the case, the approximation of eq. (20) no longer holds, since $\omega_0 \approx \Gamma$. This corresponds to the so-called incomplete relaxation regime, which has been studied extensively to analyse XPS data [9, 10]. The time integral appearing in eq. (19) should then be calculated completely, and the corresponding weight $F_{ij}$ (eq. (12b)) should be computed numerically for each particular $h$ level. At this stage, one can only predict qualitatively a non negligible decorrelation effect.

2.2 CONDUCTION ELECTRON SCATTERING. — We assume the following model: the charge localized on a deep-hole polarizes, via the Coulomb interaction, the conduction electron gas, which polarization reacts on the deep-holes themselves. We take into account the Coulomb interaction between conduction elec-
trons, but neglect the deformation of their wavefunctions by the (transient) deep-hole potential. Indeed, as is well known, the main effect of this rearrangement is to give rise, in metals, to the so-called infrared singularity, localized in the immediate vicinity of the high energy edge of the Auger line [2, 11], and which has a small relative weight in the total line strength. On the other hand, outside of this small energy range, the transient character of the hole potential becomes negligible, and one is left with standard screening effects in the presence of a localized impurity, which can be taken into account by means of a dielectric constant approximation. Since we are interested mainly in orders of magnitude of the decorrelation effect on the Auger line strength, we neglect the corrections to the screening due to the presence of the hole.

In this model, the renormalization of the two-hole propagator \( S_{ij} \) is given by the same set of diagrams as for the phonon case (see figure 3), where the wavy lines are now to be understood as propagators of fluctuations of the conduction electron gas (Fig. 4).

\[
\varphi^2 = \text{photon propagator} + \text{neutrino propagator}
\]

Fig. 4. — Diagrammatic equation for the propagator of fluctuations of the conduction electron gas, represented by a wavy line. Dotted lines stand for Coulomb interactions, and renormalized bubbles represent polarization loops.

That is, the corresponding Keldysh matrix propagator \( \overline{D}_q(\omega) \) now becomes

\[
\overline{D}_q(\omega) = \frac{4 \pi e^2}{q^2} \frac{1}{\varepsilon(q, \omega)}
\]

and

\[
\frac{1}{\varepsilon(q, \omega)} = \sigma_z + \frac{4 \pi e^2}{q^2} \phi_q(\omega)
\]

where \( \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \), and \( \phi \) is the Keldysh matrix generalization [4] of the causal density-density response function [12].

The renormalized two-hole propagator is then again given exactly (apart for the \( \lambda \)-sum) by eqs. (6), (7), (9), (10), where now

\[
g_q = \int d^3 r \ | \varphi_h(r) |^2 e^{i q \cdot r}
\]

and \( \varphi_h(r) \) is the (atomic) wavefunction of a deep-hole localized on site \( R = 0 \).

As in the phonon case, we only consider the total strength of the Auger line, and not its detailed shape. Since, as far as time ordering is concerned, density operators are equivalent to boson field operators, the generalized density response \( \overline{\rho} \) satisfies the two Keldysh relations:

\[
\phi^{(+)} + \phi^{(-)} = \phi^{(+)} + \phi^{(-)}
\]

\[
\phi^{(-)}(t) = - [\phi^{(+)}(t)]^\ast
\]

so that eqs. (16) and (17) are still satisfied in the present case. Therefore, the interaction with conduction electrons — as with phonons — does not modify the strength of non-interferent Auger terms.

For interferent terms we are left with eqs. (18-19) for the contribution to the total strength but, in contradistinction with the phonon case, the correlation time \( \tau_c \) of the fluctuating field (here the conduction electron polarization field) is now much smaller than the hole lifetime \( T_1 \). Indeed, while for light elements \( T_1 \gg 1 \text{ eV} \), \( \tau_c \) is controlled by the motion of charge fluctuations, i.e. \( \tau_c \approx \omega_{pl} / (\mu) \), where the plasmon frequency \( \omega_{pl} \) and the Fermi energy \( \mu \) are of order 10 eV.

This limit is the opposite of that for phonons, so that eq. (20) is no longer valid and has to be replaced, for \( t \gg \tau_c \), by:

\[
\int_{-t}^t dt \left[ t_1^{\ast} \right] \text{Im} D_{ij}^{(+)}(t) \approx t \text{Im} D_{ij}^{(+)}(\omega = 0) =
\]

\[
- t \frac{4 \pi e^2}{q^2} \left| \text{Im} \frac{1}{\varepsilon(q, \omega = 0)} \right|
\]

where \( \varepsilon(q, \omega) \) is now the usual physical dielectric constant.

In this regime of fast fluctuations, the holes live long enough to see only an average effect of the random field. In our model, which neglects infrared singularity corrections (which are anyhow reduced by the hole lifetime effect), this average loss of memory is measured by the dissipative part of the static dielectric constant.

The \( ij \) contribution \( F_{ij} \) to the total line strength given by eq. (12b) can therefore be written (up to errors of order max \( \tau_c, \tau_L, \beta_{ij} \)):

\[
F_{ij} = \int_0^\infty dt e^{-2r_1 - r_{ij}} = (2 \Gamma + \beta_{ij})^{-1}
\]

\[
\beta_{ij} = \sum_q |g_q|^2 (1 - e^{i \omega_{p}\tau}) \frac{4 \pi e^2}{q^2} \left| \text{Im} \frac{1}{\varepsilon(q, \omega = 0)} \right|
\]

\( \beta_{ij}^{-1} \) thus gives the decorrelation time for the relative phases of holes \( i \) and \( j \). Non-interferent terms correspond to \( \beta_{ij} = 0 \). The importance of the relative reduction of interference terms is fixed by the value of the ratio \( \beta_{ij}^2 / 2 \Gamma \) of longitudinal and transverse relaxation times.

Note that, at zero temperature, \( \text{Im} \varepsilon(q, \omega = 0) = 0 \) (real transitions with zero energy transfer are impossible). For \( T \neq 0 \text{ K} \), for metals,

\[
| \text{Im} \varepsilon(q, \omega = 0) | \propto kT/\mu < 10^{-2}
\]
while it is much smaller for semiconductors and insulators, and phase decorrelation effects are certainly always small compared with lifetime ones.

In order to get a numerical estimate of \( \beta_{ij} \), we have calculated it for two light metals (Na and Al), taking for the dielectric constant its RPA expression \(^{13}\), and setting \( q = 1 \), which is a reasonable approximation in view of the extremely localized character of deep-hole wavefunctions.

Numerical computation shows that, for the \( R_{ij} \)'s of interest in interferent terms (\( R_{ij} \geq \) nearest neighbour separation), \( \beta_{ij} \) increases with \( R_{ij} \), so that

\[
\beta_{ij} < \beta_{R_{ij} \rightarrow \infty} \equiv \beta(\infty)
\]

and we find, at room temperature \( (T = 1/40 \text{ eV}) \)

for Na : \( \beta(\infty) \simeq 2 \times 10^{-2} \text{ eV} \)

for Al : \( \beta(\infty) \simeq 10^{-2} \text{ eV} \).

So, as in the case of phonons, for not too narrow levels, the \( \beta_{ij} \)'s characteristic of extraatomic phase decorrelation are much smaller than the hole inverse lifetime \( \Gamma \gtrsim 10^{-1} \text{ eV} \), and it can be concluded that phase decorrelating interactions do not appreciably decrease extraatomic interferences. Of course, the space range of these interferences remains limited by the lifetime effects discussed in (I) to distances of atomic order.

3. Degenerate deep-hole: effect of interactions on intraatomic interferences. — Let us first, for simplicity, consider a line corresponding to three narrow levels (\( h, n, p \)), so that there are no extraatomic interferences. We assume that the deep-hole level (\( h \)) is degenerate, and call \( \lambda, \mu, \nu, \ldots \) the corresponding deep atomic orbitals (centred on a given site). As discussed in (I), the Auger current now contains contributions due to interferences between different orbitals on the same atomic site.

As in 2, we only consider interaction processes which are diagonal with respect to the site indices. Then, the Auger current \( j^{(hnp)}(R, \omega) \) is still given by the formal expression (1), where the atomic indices \( (i, j) \) must now be replaced by the orbital indices. One must therefore now study the propagator

\[
S_{\lambda \mu}(t_1, t_2 ; t_3, 0)
\]

of two holes located on the same site, in the same (\( \lambda = \mu \)) or in two different (\( \lambda \neq \mu \)) orbitals.

Note, however, that the matrix elements characteristic of the phase decorrelating interactions are not diagonal with respect to the orbital indices. Let us first consider conduction electron scattering, for which

\[
\langle q^{(z)}(\Delta) \rangle \quad \text{and} \quad \langle q^{(x)}(\Delta) \rangle
\]

For deep (i.e. very localized) orbitals, since, for typical values of \( q \leq a^{-1} \), where \( a \) is the atomic distance, \( q \ll \langle r_h \rangle^{-1} \), where \( \langle r_h \rangle \) is the average orbital radius, and \( g_1^{(z)} = 1 + \mathcal{O}(q^2 \langle r_h \rangle^2) \) while, at best, \( g_1^{(x)} = \mathcal{O}(q \langle r_h \rangle) \ll g_1^{(z)} \). However, if working to lowest order in \( q \langle r_h \rangle \), the phase decorrelation effect would be completely lost. Indeed, setting \( g_1^{(z)} = \delta_{\lambda \mu} \) means that the shift of energy, for any given configuration of the random field, is the same for all the orbitals concerned, so that their relative phases are not modified by the interactions. This is exactly what we have found, in paragraph 2.2, for \( i = j \) terms. Therefore, the intraatomic transverse relaxation rate is precisely due to deviations from the \( g_1^{(z)} = \delta_{\lambda \mu} \) approximation.

A further complication then arises, in comparison with the non-degenerate hole case, due to the non-diagonal couplings \( g_1^{(x \neq z)} \). Along a given deep-hole line, the orbital index is not conserved, i.e. an interaction process can induce a \( \lambda \rightarrow \mu \) transition: the internal structure of the hole must therefore be taken into account. Such a situation is well known to give rise to a Kondo effect (which, if, for example, \( \lambda, \mu, \ldots \) refer to an orbital momentum degeneracy, will be an orbital Kondo effect). We are therefore unable to calculate exactly the propagators \( S_{\lambda \mu}(t) \). However, the non-diagonal couplings being extremely small \((\sim \mathcal{O}(q \langle r_h \rangle/a))\), the corresponding Kondo temperature is negligibly small, and these couplings do not give rise to any singularity at the temperatures of the Auger experiments.

Therefore, to obtain an estimate of the decorrelation rate, we neglect them and only take into account diagonal couplings (the corresponding result being valid within about one order of magnitude). With this simplification, the orbital index is conserved along each deep-hole line, and it is straightforward to apply the algebra of paragraph 2.2, giving:

\[
\beta(\infty) \simeq \beta(\infty) \langle \langle r_h \rangle/a \rangle^2
\]

\[
\beta(\infty) \text{, defined by eq. (31), has been seen in paragraph 2.2 to be of order } 10^{-1} \text{ eV} \text{, } \langle \langle r_h \rangle/a \rangle \text{ being always smaller than typically } 10^{-1}, \quad \beta(\infty) \ll 10^{-5} \text{ eV} \ll 2 \Gamma.
\]

Analogously, the importance of the decorrelation due to phonons is fixed by the difference between the couplings of a given phonon to the two degenerate orbitals. Since the range of the space variations
of the screened ionic pseudopotential which gives rise to the coupling is typically of atomic order,
\[ g_{e}^{(E)} - g_{e}^{(aa)} \approx g_{e}^{\sqrt{\langle r_{h} \rangle / a}}. \]
Thus, again, the corresponding relaxation width
\[ \Delta_{\lambda} \sim \Delta (\langle r_{h} \rangle / a)^4 < 10^{-5} \text{ eV} \]
is completely negligible with respect to the longitudinal one, \( 2 \Gamma \) and, finally, intraatomic phase decorrelation is always negligible.

Let us recall that lifetime and mean free path effects do not reduce the weight of intraatomic interferences which should therefore, as discussed in (I), play an important part in Auger spectra.

Finally, the intraatomic decorrelation widths are always much smaller than the extraatomic ones. So the effect of atomic degeneracy on the phase decorrelation of two holes on different atomic sites is negligible compared with that due to the site degeneracy, and the result of the preceding section for the atomic deep level is degenerate.

4. Conclusion. — It therefore appears that the interactions which conserve the number of deep-holes do give rise to phase decorrelation between interfering holes. However, the corresponding transverse relaxation rate is in general much smaller than the longitudinal one, \( 2 \Gamma \), due to the finite hole lifetime. Thus we can conclude that:

(i) Intraatomic interferences, which occur in the case of a degenerate deep-hole, should give an important contribution to the Auger current, and in particular modify its angular distribution.

(ii) For Auger lines involving at least one propagating final hole state (atomic-atomic-V or atomic-V-V line), there are extraatomic interferences.

For light materials and not too shallow \( h \) levels (i.e. for deep-holes with linewidths in the \( 10^{-1} \) to 1 eV range) extraatomic interferences should be important. Their space range is limited by lifetime effects only to the first few nearest neighbour sites.

For the deepest levels of heavy elements (with linewidths larger than typically 1 eV) the extraatomic effects should be completely washed out by lifetime effects.

For very narrow \( h \) levels (with widths in the 50 meV range) phase decorrelation might decrease the importance of extraatomic interferences. The magnitude of this effect should be computed numerically for each particular experimental case.

Up to now, the existence of interference effects has not been checked experimentally. It seems that, before including interference contributions into the numerical computation of Auger current distributions, one should look for a qualitative proof of the presence of the effect. This could be done along two different directions:

(i) The existence of interferences is physically equivalent to a memory, in the Auger process, of the hole creation event. Such a memory implies that the Auger distribution depends on the frequency of the incident photon, or on its polarization, which could be a simpler experiment since the X-ray beams provided by synchrotron radiation have a high degree of polarization. Such dependences, corresponding to analogous memory effects, are known to be present in optical experiments (hot luminescence and resonant Raman scattering in semiconductors).

(ii) The fact that the Auger process keeps some memory of the hole creation process implies that the deephole has not enough time, between the two events, to equilibrate completely. This, as shown by Watts [7], gives rise to anomalous gain satellites, above the main Auger line, the observation of which should also be an indirect proof of the existence of the interference effect. Only plasmon satellites have a large enough energy separation to be observable. One easily shows [14] that the strength of the \( n \)th gain satellite is proportional to \( e^{-\beta} \beta^{2n} \) (where the dimensionless coupling constant \( \beta \) is of order \( r_{e}/6 \) [15], \( r_{e} \) being the interelectron spacing expressed in Bohr units) while the strength of the \( n \)th normal loss satellite is of order \( e^{-\beta} \beta^{n}/n \). Following Langreth's estimate [15], for Al this gives a strength for the first gain satellite of about 10-15 % of the main peak strength, so that it could possibly be observed.

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