ON THE EFFECT OF INTERELECTRON INTERACTION IN METALS ON THE FINE STRUCTURE OF X-RAY SPECTRA

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Absorption of an x-ray quantum in a metal, with simultaneous excitation of a plasma, is considered for wavelengths close to the absorption edge. The calculations performed are valid for transition metals in the Fe group. It is demonstrated that the effect under consideration is very probable, the probability decreasing from Ti to Fe. A new interpretation of some short-wave satellites of the extreme short-wave emission line of the series is proposed.

1. INTRODUCTION

THE correspondence between the fine structure of x-ray absorption spectra and the characteristic energy levels of the electrons in metals has been noted by many researchers[1-4] who treated the characteristic electron energy losses from the point of view of the single-electron mechanism of transitions between bands. The fine structure of the x-ray spectra was considered from this same point of view. Establishment of a correspondence between the two phenomena has confirmed, in the opinion of many authors, the single-electron interpretation of the characteristic losses.

Ferrell[5] advanced the hypothesis that the excitation of plasma oscillation of the electron gas in a metal upon absorption of an x-ray quantum can complicate the fine structure of the x-ray absorption spectrum. Sobel'man and Feĭnberg[6] considered theoretically the effects of the excitation of plasma oscillations of the electron gas in a metal on the fine structure of the x-ray absorption spectrum for the region far from the absorption edge, that is, for the case when $E_f - E_F \gg \hbar\omega$, where E_f is the energy of the final state of an electron that has absorbed an x-ray quantum, E_F is the electron energy on the Fermi surface, and $\hbar\omega$ is the plasmon energy.

The results of the experimental investigations have pointed, however, to the necessity of investigating the question of the absorption of an x-ray quantum with simultaneous excitation of a plasmon (or with simultaneous single-electron transition between bands) for the region close to the absorption edge, that is, where the inequality given above is not satisfied and where, consequently, the deductions of Sobel'man and Feĭnberg cannot be directly applied. As will be made clear in the sequel, the process of absorption of an x-ray quantum with simultaneous excitation of the plasmon is of particular significance for transition metals.

2. FORMULATION OF THE PROBLEM

Our problem will consist of calculating the ratio of the coefficient of x-ray absorption, τ, with allowance for a simultaneous production of a plasmon, and the coefficient of absorption τ_0, given by the single-electron theory of x-ray spectra. In other words, the problem consists of finding the absorption coefficient with allowance for the interaction between an electron that has absorbed an x-ray quantum with all the free electrons of the metal (which interact with each other).

Let us consider a system consisting of the following: 1) An electron located in the periodic field of the ions (with a radius vector $\mathbf{R}$); in the initial state the electron is in the K shell (the generalization to other cases is easy), and goes over during the process of absorbing the x-ray quantum to the free states of the metal above the Fermi surface. 2) A gas of N free electrons of the metal, the common negative charge of which is offset by the positive background of the metal ions. 3) The field of the x-ray photons.

The Schrödinger equation of such a system will be written in the form

$$i\hbar\partial\varphi/\partial t = \mathcal{H}\varphi, \tag{1}$$

$$\mathcal{H} = H_1(\mathbf{R}) + H_e(\mathbf{r}_1, \ldots, \mathbf{r}_N) + H_{ph} + H_2,$$

$$H_1 = -\frac{\hbar^2}{2m}\nabla_{\mathbf{R}}^2 + U(\mathbf{R}), \qquad H_2 = \sum_j \frac{e^2}{|\mathbf{r}_j - \mathbf{R}|} + \hat{V},$$

$$H_e = -\sum_j \frac{\hbar^2}{2m}\nabla_j^2 + \frac{1}{2}\sum_{j_1 \neq j_2} \frac{e^2}{|\mathbf{r}_{j_1} - \mathbf{r}_{j_2}|} + H_{ei}.$$

Here H_{ph} is the self energy of the x-ray photons, $U(\mathbf{R})$ is the periodic lattice potential, H_{ei} is the energy operator of the interaction between the electrons and the positive background,* and $\hat{V}$ is the energy operator of the interaction between the electron ($\mathbf{R}$) and the x-ray photon.

Following Zubarev,[9] we introduce the particle-density operator $\rho(\mathbf{r}) = \Sigma_j \delta(\mathbf{r} - \mathbf{r}_j)$. Its Fourier amplitudes are

$$\rho_k = N^{-1/2} \sum_j \exp\{-i\mathbf{k}\mathbf{r}_j\}, \qquad k < k_c,$$

where k_c is the maximum wave number of the plasma wave. The operators

$$b_k = \tfrac{1}{2}(\partial/\partial q_{-k} + q_k), \qquad b_k^+ = \tfrac{1}{2}(-\partial/\partial q_k + q_{-k}) \quad (2)$$

satisfy the commutation relations of the Bose creation and annihilation operators. Here

$$q_k = \rho_k/\sqrt{2}\,\lambda_k, \qquad \lambda_k = \sqrt{(\hbar^2k^2/2m)/\hbar\omega_p},$$

$$\omega_p = \sqrt{4\pi Ne^2/Vm}.$$

The operators a and a^+ are the Fourier amplitudes of the Fermi-field operators

$$\hat{\psi}(\mathbf{r}, s_z) = \frac{1}{\sqrt{V}} \sum_{\sigma p} a_{p\sigma} e^{i\mathbf{p}\mathbf{r}} S_\sigma(s_z),$$

$$\hat{\psi}^+(\mathbf{r}, s_z) = \frac{1}{\sqrt{V}} \sum_{\sigma p} a_{p\sigma}^+ e^{-i\mathbf{p}\mathbf{r}} S_\sigma(s_z). \quad (2')$$

We now change over to a mixed second-quantization representation, using expression (2) and (2′). Then H_2 is transformed, after expansion of the first term in a Fourier series in plane waves, into the form

$$H_2 = \frac{\sqrt{N}}{V} \sum_{k<k_c} \frac{4\pi e^2}{k^2} \lambda_k (b_k + b_{-k}^+) e^{i\mathbf{k}\mathbf{R}} + \frac{1}{V} \sum_{\substack{k>k_c \\ p}} \frac{4\pi e^2}{k^2} e^{i\mathbf{k}\mathbf{R}} a_{p-k}^+ a_p + \hat{V}, \quad (3)$$

and the initial equation (1), after canonical transformations similar to those introduced by Zubarev,[9] becomes

$$i\hbar\,\partial\Psi/\partial t = \mathcal{H}_1\Psi,$$

where

$$\mathcal{H}_1 = H_0 + H, \qquad H_0 = H_1 + H_B + H_F + H_{ph},$$

$$H = H_2 + H_{BB} + H_{BF} + H_{FF},$$

$$H_B = \sum_{k<k_c,\ k\neq 0} \hbar\omega(k) b_k^+ b_k, \quad H_F = \sum_{p\sigma} \frac{\hbar^2 p^2}{2m} a_{p\sigma}^+ a_{p\sigma}. \quad (4)$$

*The energy of interaction between the electrons and the homogeneous positive background is equal and opposite to the zeroth term (k = 0) of the Fourier expansion for the energy

$$\tfrac{1}{2} \sum_{j_1 \neq j_2} e^2/|\mathbf{r}_{j_1} - \mathbf{r}_{j_2}|.$$

Here H_B is the self energy of the Bose excitations of the electron gas (plasmons); H_F is the self energy of the electron gas; H_{BB}, H_{BF}, and H_{FF} describe the interaction between the plasmons themselves, the plasmons and the electrons, and the electrons themselves.

The problem of finding the probability of the absorption of an x-ray quantum with simultaneous excitation of a plasmon (or the creation of an electron-hole pair) reduces the problem to finding the probability of transition between the eigenstates of the Hamiltonian H_0 under the influence of the perturbation H. The problem is solved by the methods of the non-stationary perturbation theory with allowance for damping.[10]

3. CALCULATION OF THE MATRIX ELEMENTS

We consider transitions between the following eigenstates Ψ_μ of the operator H_0:

1. The ground state $\Psi_0 = \psi_0 \Phi_{B_0} \Phi_{F_0} \Phi_{ph}$. The function $\psi_0(\mathbf{R})$ describes the state of an absorbing electron in the K shell of the atom, Φ_{B_0} corresponds to the absence of plasmons, Φ_{F_0} corresponds to the ground state of the Fermi-particle state, and Φ_{ph} corresponds to a certain photon distribution.

2. Intermediate state, $\Psi_\nu = \psi_n \Phi_{B_0} \Phi_{F_0} \alpha_\lambda \Phi_{ph}$. It corresponds to a transition of an absorbing electron into the intermediate state ψ_n with absorption of an x-ray quantum, characterized by the index λ. This index takes into account the wavelength of the absorbed photon, the direction of propagation, and the state of polarization; α_λ is the annihilation operator of such a photon.

3. Final state

$$\Psi_{\varphi_1} = \psi_f b_\varkappa^+ \Phi_{B_0} \Phi_{F_0} \alpha_\lambda \Phi_{ph} \quad \text{or} \quad \Psi_{\varphi_2} = \psi_f \Phi_{B_0} a_{p_1}^+ a_{p_2} \Phi_{F_0} \alpha_\lambda \Phi_{ph}.$$

We are interested in the transition from the ground state to the final state. The investigated process should be considered in the second order of perturbation theory, and its probability per unit time is

$$w_{\varphi 0} = 2\pi\hbar^{-1} |\widetilde{\mathcal{H}_{\varphi 0}}|^2 \delta(E_\varphi - E_0),$$

$$|\widetilde{\mathcal{H}_{\varphi 0}}| = \sum_\nu \langle\varphi|H|\nu\rangle\langle\nu|H|0\rangle (E_\varphi - E_\nu + \tfrac{1}{2}i\hbar\gamma_\nu)^{-1}. \quad (5)$$

Here γ_ν is a total width of the level ν or a quantity reciprocal to the lifetime of the system in the state Ψ_ν; E_φ and E_ν are eigenvalues of the operator H_0 respectively for the functions Ψ_φ and Ψ_ν.

Using relations (3) and (4), we obtain

$$\langle\varphi_1|H|\nu\rangle = \frac{\sqrt{N}}{V} \frac{4\pi e^2}{\varkappa^2} \lambda_\varkappa \langle\psi_f|e^{i\varkappa\mathbf{R}}|\psi_n\rangle, \qquad \varkappa < k_c, \quad (6)$$

$$\langle \varphi_2 | H | \nu \rangle = \frac{1}{V} \frac{4\pi e^2}{|\mathbf{p}_2 - \mathbf{p}_1|^2} \langle \psi_f | \exp\{i(\mathbf{p}_2 - \mathbf{p}_1)\mathbf{R}\} | \psi_n \rangle,$$

$$|\mathbf{p}_2 - \mathbf{p}_1| > k_c, \tag{7}$$

$$\langle \nu | H | 0 \rangle = \langle \psi_n \left| -\frac{ie\hbar}{mc} \mathbf{A}_0 \nabla \right| \psi_0 \rangle, \tag{8}$$

where $\mathbf{A}_0$ is the amplitude of the vector potential of the electromagnetic wave.

Inasmuch as we are directly interested in the fine structure of the main absorption edge, the eigenvalue of the energy of the absorbing electron in the final state is $E_f \sim E_F$. In addition, it follows from (5) that the greatest contribution to the effect under consideration would be given by those intermediate states ψ_n, for which the relation $E_n - E_f \sim \hbar\omega$ is satisfied, that is, $E_n - E_F \sim 10$ ev, since $\hbar\omega \sim 10$ ev.

The curve of the energy distribution of the free-electron states N(E) is shown in the figure for transition metals of the Fe group.[11] The Fermi level passes through the region of high state density, corresponding to the 3d band of the metal, whose wave functions correspond, as is well known, to the strong-coupling approximation.

The intermediate states ψ_n, for which $E_n - E_F \sim 10$ ev, correspond to the region of considerably smaller values of N(E) (the 4p band), and here N(E) is a slowly-varying function of E. The latter indicates that ψ_n should be considered in the weak-coupling approximation, the limiting case of which is a plane wave. Therefore, for a concrete calculation of (6), (7) and (8), we specify the wave functions:

$$\psi_0 = -(Z_0/a^3\pi)^{1/2} \exp\{-Z_0 R/a\}, \tag{9}$$

i.e., the 1s state of the electron in an atom with atomic number Z_0; furthermore

$$\psi_n = V^{-1/2} \exp\{i\mathbf{k}_2\mathbf{R}\},$$
$$\psi_f = G^{-1/2} \sum_j \varphi_{3d}(\mathbf{R} - \mathbf{R}_j) \exp\{i\mathbf{k}_1\mathbf{R}_j\}, \tag{10}$$

i.e., the final state of the absorbing electron, ψ_f, is represented in the strong-coupling approximation, φ_{3d} is the atomic function describing the 3d state, $\mathbf{R}_j$ is the radius vector of the j-th atom, and G is the number of atoms in the lattice.

We shall make the calculations for the final state Ψ_{φ_1}, and drop the subscript 1 from now on. Substitution of (9) and (10) in (6) and (8) yields

$$\langle \varphi | H | \nu \rangle = 1.1\pi^{3/2} \frac{\sqrt{NG}}{V^{3/2}} \frac{e^2\hbar}{\varkappa\sqrt{2m\varepsilon_p}} \left(\frac{Z}{a}\right)^{9/2} (\mathbf{k}_2 + \boldsymbol{\varkappa})^{-6} \times \left[1 + \frac{Z^2}{9a^2(\mathbf{k}_2 + \boldsymbol{\varkappa})^2}\right]^{-4} \delta_{\mathbf{k}_2 + \boldsymbol{\varkappa} - \mathbf{k}_1}, \tag{11}$$

$$\langle \nu | H | 0 \rangle = 8\sqrt{\frac{\pi}{V}} \frac{\hbar e}{mck_2^3} A_0 \left(\frac{Z_0}{a}\right)^{5/2} (1 + Z_0^2/k_2^2 a^2)^{-2}. \tag{12}$$

Considering that $Z_0^2/k_2^2a^2 \gg 1$, expression (12) can be written in the form

$$\langle \nu | H | 0 \rangle = 8\sqrt{\frac{\pi}{V}} \frac{\hbar e k_2}{mc} A_0 \left(\frac{Z_0}{a}\right)^{-3/2} \tag{13}$$

Here a is the Bohr radius, Z the effective charge of the nucleus for 3d electrons, and $\epsilon_p = \hbar\omega_p$.

4. CALCULATION OF THE PROBABILITY OF ABSORPTION OF AN X-RAY QUANTUM

The coefficient of absorption of x-rays is determined by the value W_q of the total probability of absorption of an x-ray quantum by an atom of matter per unit time. In our case, W_q is given by

$$W_q = \int \frac{2V}{(2\pi)^3} d^3k_1 \cdot \frac{V}{(2\pi)^3} d^3\varkappa \cdot w_{\varphi 0}, \tag{14}$$

where $w_{\varphi 0}$ is determined from (5).

We introduce the notation

$$M^2 = \frac{V}{(2\pi)^3} \int d^3\varkappa \, |\tilde{\mathcal{H}}_{\varphi 0}|^2. \tag{15}$$

Then

$$W_q = \frac{2\pi}{\hbar} \overline{M^2} N_{3d}(E), \qquad N_{3d}(E) = \frac{2V}{(2\pi)^3} \int_S dS / |\nabla_{\mathbf{k}_1} E_{3d}|, \tag{16}$$

where $N_{3d}(E)$ is the state density in the 3d band, $\overline{M^2}$ denotes averaging of M^2 over the surface S, defined by the equation $E_{3d}(\mathbf{k}_1) = \text{const}$, and satisfying the law of conservation of energy.

Substituting the expressions (11) and (13) into (15), we obtain after several simplifications the following estimate of the order of magnitude of W_q:

$$W_q \sim \frac{10^{10}\pi^7 NG}{V^3} \frac{e^6 A_0^2 a^{10} k_c}{\hbar m^2 c^2 \varepsilon_p Z_0^3 Z^7 \gamma^2 d^4} (E_{3d} + \varepsilon_p) N_{3d}, \tag{17}$$

where $\epsilon_p \equiv \hbar\omega_p$, and d is the parameter of the crystal lattice.

In the derivation of (17) the following simplifying assumptions were made: 1) the width of the intermediate state γ_ν is constant and is independent of ν, that is, $\gamma_\nu \equiv \gamma$; 2) $\hbar\omega(\kappa) = \hbar\omega_p = \epsilon_p$; 3) $k_1 \sim \pi/d$ on the surface S (over which M^2 is averaged).

Let us compare now the expression (17) with the total probability W_q^0 of absorption of an x-ray quantum by an atom of matter per unit time as a result of the direct transition $\psi_0 \rightarrow \psi_n$:

$$W_q^0 = \frac{2\pi}{\hbar} \overline{|H_{\nu 0}|^2} N_{4p} = 64 \frac{\pi^3}{V} \frac{\hbar^2 e^2 A_0^2}{m^2 c^2 d^2} \left(\frac{a}{Z_0}\right)^3 N_{4p}. \tag{18}$$

Here N_{4p} is the density of states in the 4p band of the metal.

The ratio $\chi = \tau/\tau_0 = W_q/W_q^0$ of the coefficient of absorption τ with simultaneous plasmon crea-

tion taken into account and of the coefficient of absorption τ_0 given by the single-electron theory is obtained from (17) and (18):

$$\chi \sim \frac{3\cdot 10^7 \pi^3 NG}{V^2} \frac{e^4 a^7 k_c}{\gamma^2 d^2 \hbar^2} \frac{E_{3d}+\varepsilon_p}{\varepsilon_p} \frac{N_{3d}}{N_{4p}} \frac{1}{Z^7}. \quad (19)$$

We assume the following estimates: the volume of the system $V = 1\ cm^3$, $N = G = 10^{23}$, $k_c = 1\,A^{-1}$, $d = 3\,A$, $(E_{3d} + \epsilon_p)/\epsilon_p \sim 1$, $N_{3d}/N_{4p} = 5$ (see figure). To estimate the width of the excited state γ, we can use the results from reference 12, from which it follows that for electrons whose energy exceeds the Fermi energy by ~ 10 ev, the width of the state will be on the order of several electron volts. We therefore assume $\gamma = 3 \times 10^{15}\ sec^{-1}$, corresponding to width of ~ 2 ev of the state. Then an estimate of the order of magnitude of (19) yields

$$\chi \sim 10^5/Z^7. \quad (20)$$

The value of Z — the effective charge of the nucleus for the 3d electrons of the atom — can be estimated following Slater;[13] we then obtain the following values of χ for Ti, Cr and Fe:

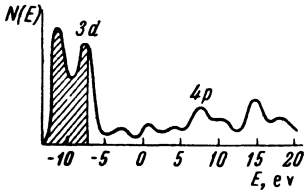
	Z (after Slater)	χ
Ti	4	5
Cr	5	1
Fe	7	0.1

5. DISCUSSION OF THE RESULTS

The results obtained show that the probability of absorption of x-ray quanta with simultaneous excitation of plasmons is of the same order of magnitude as the probability of direct transition without plasmon excitation; this transition pertains to the region of the main edge of the absorption spectrum. This effect becomes particularly significant for transition metals, since the factor N_{3d}/N_{4p}, which enters into the expression (19) for χ, may reach large values for these metals (see diagram).

The very strong dependence of χ on Z, given by (20) is connected with the choice of ψ_n in the form of a plane wave. Another form of intermediate-state function was considered

$$\psi_n = \frac{1}{\sqrt{G}} \sum_j \varphi_{4p}(\mathbf{R} - \mathbf{R}_j) \exp\{i\mathbf{k}_2\mathbf{R}_j\},$$



Energy distribution of the density of states of the free electrons for the transition elements of the iron group. The shaded area denotes the occupied states for the case of iron.[11]

since the other extreme case, that of a strong coupling with the lattice, was assumed. In this case $\chi \sim 100/Z^2$ where Z is the effective averaged charge of the nucleus for the 3d and 4p electrons. Obviously, χ remains of the same order of magnitude, but its dependence on Z becomes weaker.

Thus, we can conclude with regard to the absorption spectrum that the experimentally observed absorption spectrum should be a superposition of the true absorption spectrum (which would be produced only by direct transitions $\psi_0 \rightarrow \psi_n$), and of the same spectrum, but shifted by ϵ_p towards shorter wavelengths.

An experimental confirmation of this deduction is seen in the results of Borovskiĭ and Batyrev,[7] who investigated the fine structure of the x-ray absorption spectra of the elements of the iron group: Ti, V, Cr, Fe, Co, Ni, Cu. The details of the fine structure (which are considered by the authors as a plasma repetition) manifest themselves most clearly in titanium and chromium; in iron and chromium they are much less clearly pronounced, and dissappear completely in the case of nickel. This corresponds to our conclusion that χ decreases gradually on going from titanium through chromium to iron. Another experimental confirmation of our conclusion with regard to the absorption spectra has been obtained by Borovskiĭ and the author[8] in an investigation of the temperature dependence of the fine structure of the absorption spectrum of iron, and its connection with the characteristic losses of electron energy.

We note still another possible application of the results obtained here to the theory of emission spectra. All the calculations remain in force if we consider the reverse transition $\Psi_\varphi \rightarrow \Psi_\nu \rightarrow \Psi_0$, that is, if the initial state is considered to be a hole in the K shell, and if we consider the plasma excitation of the electron gas (appearing simultaneously with the production of the hole). Since the lifetime of the hole in the K shell and the lifetime of the plasmon are of the same order of magnitude ($\sim 10^{-16}$ sec), the existence of such a state is quite feasible. The transition to the intermediate state ψ_n is executed by the electron of the conduction band with absorption of a plasmon, after which it fills the hole in the K shell with emission of a short-wave satellite of the last (short-wave) emission line of the series. In this case the energy distance between the short-wave satellite and the generating line coincide with the magnitude of the characteristic energy loss of the electrons in the given substance. The following table, based on literature data, does not pretend to be systematic

Element	Distance from line to satellite, ev (reference 14)	Characteristic energy losses, ev (reference 15)	Element	Distance from line to satellite, ev (reference 14)	Characteristic energy losses, ev (reference 15)
Mg	$K_{\beta_1}-K_{\beta}^{V}$ 9.1	10.2	Fe	$K_{\beta_5}\ K-_{\beta}^{III}$ 7.9	7.5
Al	$K_{\beta_1}-K_{\beta}^{IV}$ 14.8	14.6	Mo	$L_{\beta_2}-L_{\beta_2}^{I}$ 24.8	24.7
K	$K_{\beta_5}-K_{\beta}^{III}$ 31.3	32.0	Sb	$L_{\beta_7}-L_{\beta_2}^{II}$ 18.0	18—20
Ca	$K_{\beta_5}-K_{\beta}^{III}$ 27.0	29.0	Te	$L_{\beta_7}-L_{\beta_2}^{II}$ 18.5	17.9

or complete, and is given merely to illustrate this conclusion. In this table the energy distance of the considered satellite from the last short-wave line of the series is compared with the characteristic loss for the given material. All the satellites have an energy greater than the energy of the corresponding edge of the absorption band.

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