than the probability for electron spin flip ($1/\tau_s \sim 10^8$ sec^{-1}). Moreover, the Zeeman interaction of a localized electron with the external magnetic field $H_0 = 4.9$ kG is stronger than its Zeeman interaction with the nucleus of the phosphorus donor atom,[9] so we can speak of the spin state of the localized electrons in the external magnetic field.

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Some features of x-ray fluorescence in metals near the absorption threshold

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An expression for the x-ray inelastic scattering cross section in metals is derived by taking into account interference of hole states localized at different centers. It is demonstrated that the shape of the spectral band depends on the initial photon frequency ω if the latter is near to the inner level ionization threshold. A shift of the fluorescence threshold is found in alkali metals.

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I. INTRODUCTION

We have previously[1] considered the dependence of the emission spectrum of a diatomic homonuclear molecule on the frequency ω of the exciting photon. This dependence is due to interference between the x-ray-excited states with holes localized on different atoms. The purpose of the present study was to investigate the influence of this interference on the shape of the x-ray emission band of a metal. The role of the interference effects was qualitatively discussed earlier by Nozieres and Abrahams,[2] who considered the scattering of a soft x-ray photon in a metal. Nozieres and Abrahams[2] investigated inelastic scattering of a photon in a metal with allowance for the interaction of the electrons of the valence band with the hole in the internal shell. In view of the complexity of the many particle formulation of the problem, Nozieres and Abrahams actually confined themselves to an investigation of the singularities of the photon scattering cross section in the region

$$\omega-\omega_0 \lesssim \gamma \sim 0.1 \text{ eV} \tag{1}$$

and (or)

$$\omega_0-\omega' \lesssim \gamma,$$

where the interference effects, in their opinion, are appreciable. Here $\omega_0 = E_F - E_0$, ω, ω' are the frequencies of the initial and final photons; E_F and E_0 are the Fermi energy and the energy of the internal level. γ is the natural width of the x-ray line. In the present paper, confining ourselves to the single-particle approximation, we investigate the cross section for the scattering of an x-ray photon in a metal outside the narrow interval (1). The existing single-particle calculations[3] of emission bands in a metal agree well with experiment, including the region (1) and the low-energy tails of the emission band. Skinner[4] attributes these tails to the energy dependence of the lifetime of the single-particle state in the valence bands; for the state at the bottom of the valence band this time is much less than near the Fermi level. Within the framework of the single-particle description, this effect can be easily taken into account by introducing the corresponding damping parameter γ.[5]

2. *K* FLUORESCENCE

I. We consider *K* fluorescence of a metal. The cross section for inelastic scattering of a photon by the system is described by the Kramers-Heisenberg formula.[6] Using the dipole approximation for the atomic matrix element and neglecting the width of the x-ray line, we obtain

$$\sigma(\omega,\omega')\sim\sum_{kk'}\Big|\sum_{n}\langle n|\mathbf{e}'\mathbf{r}_n|k'\rangle\langle k|\mathbf{e}\mathbf{r}_n|n\rangle\exp(i\mathbf{q}\mathbf{R}_n)\Big|^2\delta(E_1-E_{k'})$$
$$\times\delta(E_2-E_k)\theta(E_F-E_{k'})\theta(E_k-E_F), \tag{2}$$

where

$$\theta(x)=\begin{cases}1, & x\geqslant 0\\ 0, & x<0\end{cases},$$

$E_1=E_0+\omega'=k'^2$; $E_2=E_0+\omega=k^2$; $\mathbf{r}_n=\mathbf{r}-\mathbf{R}_n$; $\mathbf{e}$ and $\mathbf{e}'$ are the polarization vectors of the incident and scattered photons, respectively, $|k\rangle$, E_k, and $|n\rangle=R_{1s}(r_n)Y_{00}$, E_0 are respectively the vectors of the state and the energies of the valence electron and of the K electron of the atom n; $\mathbf{q}$ is the change of the photon momentum; $\mathbf{R}_n$ is the radius vector of the n-th atom.

We investigate (2) in the muffin-tin (MT) potential approximation. As the zero of the energy we take the potential outside the MT spheres. Introducing the single-electron Green's functions $G_E^{\pm}=(E-H\pm i0)^{-1}$ with the aid of the relation

$$\sum_k|k\rangle\delta(E-E_k)\langle k|=\frac{i}{2\pi}(G_E^{+}-G_E^{-})$$

we rewrite (2) in the form

$$\sigma(\omega,\omega')\sim-\frac{1}{4\pi^2}\sum_{nn'}\exp(i\mathbf{q}\mathbf{R}_{nn'})\langle n|\mathbf{e}'\mathbf{r}_n(G_{E_1}{}^{+}-G_{E_1}{}^{-})\mathbf{e}'\mathbf{r}_{n'}|n'\rangle$$
$$\times\langle n'|\mathbf{e}\mathbf{r}_{n'}(G_{E_2}{}^{+}-G_{E_2}{}^{-})\mathbf{e}\mathbf{r}_n|n\rangle\theta(E_F-E_1)\theta(E_2-E_F), \tag{3}$$

where

$$\mathbf{R}_{nn'}=\mathbf{R}_n-\mathbf{R}_{n'}.$$

If we use the node representation for the retarded Green's function[7] $(L=lm)$

$$G^{+}(\rho+\mathbf{R}_n,\rho'+\mathbf{R}_{n'})=k(1-\delta_{nn'})\sum_L R_L(\rho_<)N_L(\rho_>)$$
$$+k\sum_{LL'}[\exp(i\delta_l)G_{LL'}^{nn'}\exp(i\delta_{l'})-i\delta_{LL'}\delta_{nn'}]R_L(\rho)R_{L'}(\rho')$$

and the identity

$$G_E^{-}(\mathbf{r},\mathbf{r}')=(G_E^{+}(\mathbf{r}',\mathbf{r}))^{*}=(G_E^{+}(\mathbf{r},\mathbf{r}'))^{*},$$

then we can express the cross section σ in the form

$$\sigma(\omega,\omega')\sim kk'\left(\frac{P(E_1)P(E_2)}{6\pi}\right)^2\sum_{nn'}\sum_{\substack{m_1m_2\\m_3m_4}}e_{m_1}'e_{m_2}'e_{m_3}e_{m_4}$$
$$\times\exp(i\mathbf{q}\mathbf{R}_{nn'})\Lambda_{m_1m_2}^{nn'}(E_1)\Lambda_{m_3m_4}^{n'n}(E_2). \tag{4}$$

Here

$$G_{LL'}^{nn'}=g_{LL'}^{nn'}+\sum_{n''L''}g_{LL''}^{nn''}(\pi t_{l''})G_{L''L'}^{n''n'}$$
$$P(E)=\int_0^{\infty}R_1(r,E)R_{1s}(r)r^3\,dr,$$
$$R_L(\mathbf{r})=R_l(r)Y_L(\hat{r}), \tag{5}$$
$$\Lambda_{m_1m_2}^{nn'}(E)=2\delta_{m_1m_2}\delta_{nn'}+i[S(E)G_{1m_11m_2}^{nn'}(E)-(S(E)G_{1m_21m_1}^{n'n}(E))^{*}]$$
$$=2\,\mathrm{Im}[i\delta_{m_1m_2}\delta_{nn'}-S(E)G_{1m_11m_2}^{nn'}(E)],$$
$$S(E)=\exp(2i\delta_l(E)),\quad \pi t_l=-\sin(\delta_l(E))\exp(i\delta_l(E)),$$

$R_l(r,E)$, $(N_l(r,E))$, and $\delta_l(E)$ are the regular (irregular) solutions of the Schrödinger equation inside the MT sphere and the phase shift corresponding to this solution. $g_{LL'}^{nn'}$ is the node Green's function of the free electron, equal to zero at $n=n'$.

In the traditional theory of x-ray emission spectra of metals[3,7] no account is taken of the interference between the hole states localized at various atoms, i.e., it is implicitly assumed that

$$\Lambda_{m_3m_4}^{n'n}(E_2)=2\delta_{m_3m_4}\delta_{nn'}.$$

It is easy to note that this approximation is valid only for sufficiently high frequencies of the initial photon, $R_{nn'}(\omega-|E_0|)^{1/2}\gg 1.$[1] Indeed, at $kR\gg 1$ we have

$$\Lambda_{m_3m_4}^{n'n}-2\delta_{m_3m_4}\delta_{nn'}\sim G_{1m_31m_4}^{n'n}(E_2)\sim(1-\delta_{nn'})(kR_{nn'})^{-1}\to 0.$$

It can be shown that in this limit the K fluorescence is described by formulas (27) and (48) of[7]. However, near the ionization threshold of the K level $(\omega\sim E_F-E_0)$ the terms $\Lambda_{m_3m_4}^{n'n}$, which are off-diagonal in the lattice sites, being of the same order as the diagonal ones,[1] greatly influence the shape of the emission band. Let us investigate this question in greater detail.

II. In view of the translational symmetry of the Hamiltonian of the conduction electrons, it is convenient to change over to the Fourier transform of the node Green's function

$$G_{LL'}^{nn'}(E)=\frac{1}{\tau}\int_B d^3p\,G_{LL'}(\mathbf{p},E)\exp(i\mathbf{p}\mathbf{R}_{nn'}),$$

where the integration is carried out over the first Brillouin zone with volume τ. Then Eq. (5) is rewritten in the form

$$G_{LL'}(\mathbf{p}.E)=g_{LL'}(\mathbf{p},E)+\sum_{L''}g_{LL''}(\mathbf{p}.E)(\pi t_{l''})G_{L''L'}(\mathbf{p},E),$$

or in the matrix representation:

$$G(\mathbf{p},E)=[g^{-1}(\mathbf{p}.E)-t]^{-1}.$$

Using transformations analogous to those in[7], we obtain

$$\Lambda_{m_1m_2}^{nn'}(E)=\frac{\tau^{-1}}{\sin^2\delta_1(E)}\int_B d^3p\exp(i\mathbf{p}\mathbf{R}_{nn'})$$
$$\times[([g(\mathbf{p}.E)-t^{-1}]^{-1})_{1m_11m_2}-([g(\mathbf{p}.E)-t^{-1}]^{-1})^{*}_{1m_21m_1}].$$

Since the matrix $[g(\mathbf{p},E)-t^{-1}]^{-1}$ is Hermitian, it follows that contributions to $\Lambda_{m_1m_2}^{nn'}(E)$ are made only by those points of reciprocal space at which the equality

$$D(\mathbf{p},E)=\det[g(\mathbf{p}.E)-t^{-1}]=0$$

is satisfied. The latter is none other than the Kittel-Kassuya-Rudeman equation for the dispersion of $E(\mathbf{p})$. Since we are operating with the retarded Green's function G_E^{+}, the last expression for $\Lambda_{m_1m_2}^{nn'}$ should be rewritten in the form

$$\Lambda_{m_1m_2}^{nn'}(E)=\frac{2\tau^{-1}}{\sin^2\delta_1(E)}\int_B d^3p\exp(i\mathbf{p}\mathbf{R}_{nn'})M_{1m_11m_2}(\mathbf{p}.E)\,\mathrm{Im}\left(\frac{1}{D(\mathbf{p},E+i0)}\right)$$
$$=-\frac{2\tau^{-1}}{\sin^2\delta_1(E)}\int_B d^3p\exp(i\mathbf{p}\mathbf{R}_{nn'})M_{1m_11m_2}(\mathbf{p}.E)\delta(D(\mathbf{p}.E))\,\mathrm{sign}\frac{\partial}{\partial E}D(\mathbf{p},E),$$

where we used the definition

$$([g(\mathbf{p},E)-t^{-1}]^{-1})_{LL'}=\frac{M_{LL'}(\mathbf{p},E)}{\det[g(\mathbf{p},E)-t^{-1}]}.$$

As a result we obtain the following expression for the scattering cross section

$$\sigma(\omega,\omega')\sim Nkk'\left(\frac{\tau^{-1}P(E_1)P(E_2)}{3\pi\sin(\delta_1(E_1))\sin(\delta_1(E_2))}\right)^2$$
$$\times\sum_{\substack{m_2\geqslant m_1\\ m_4\geqslant m_3}}\rho(m_1m_2m_3m_4)\int_B d^3p\int_B d^3p'\,\Lambda_{m_1m_2}(\mathbf{p},E_1)\Lambda_{m_3m_4}(\mathbf{p}',E_2)S(\mathbf{p}-\mathbf{p}'+\mathbf{q}),\tag{6}$$

where

$$\Lambda_{m_1m_2}(\mathbf{p},E)=\delta(D(\mathbf{p},E))\,\mathrm{sign}\left(\frac{\partial}{\partial E}D(\mathbf{p},E)\right)\mathrm{Re}\,M_{1m_11m_2}(\mathbf{p},E)$$
$$=\sum_i\delta(E-E_i(\mathbf{p}))\left|\frac{\nabla_\mathbf{p}E_i(\mathbf{p})}{\nabla_\mathbf{p}D(\mathbf{p},E)}\right|\mathrm{sign}\left(\frac{\partial}{\partial E}D(\mathbf{p},E)\right)\mathrm{Re}\,M_{1m_11m_2}(\mathbf{p},E),\tag{7}$$
$$\rho(m_1m_2m_3m_4)=e_{m_1}{}'e_{m_2}{}'e_{m_3}e_{m_4}(2-\delta_{m_1m_2})(2-\delta_{m_3m_4}),\tag{8}$$
$$S(\mathbf{p})=\frac{1}{N}\sum_{nn'}\exp(i\mathbf{p}\mathbf{R}_{nn'})=\tau\sum_\mathbf{g}\delta(\mathbf{p}-\mathbf{g}),$$

g is the reciprocal-lattice vector, N is the number of atoms in the crystal, i is the number of the band, and $S(\mathbf{p})$ is the structure factor.

III. At

$$q\ll p_F=\sqrt{E_F}\tag{9}$$

it follows from (6) and (8) that

$$\sigma(\omega,\omega')\sim\sum_{fi}\int_B d^3p\,Q_{f}\qquad E_2-E_f(\mathbf{p}))\delta(E_1-E_i(\mathbf{p})),\tag{10}$$
$$E_i(\mathbf{p})\leqslant E_F<E_f(\mathbf{p}).$$

Here

$$Q_{fi}(\mathbf{p})=Nkk'\tau^{-1}\left(\frac{P(E_1)P(E_2)}{3\pi\sin(\delta_1(E_1))\sin(\delta_1(E_2))}\right)^2$$
$$\times\sum_{\substack{m_2\geqslant m_1\\ m_4\geqslant m_3}}\rho(m_1m_2m_3m_4)A^i_{m_1m_2}(\mathbf{p}E_1)A^f_{m_3m_4}(\mathbf{p},E_2),$$
$$A^i_{m_1m_2}(\mathbf{p},E)=\left|\frac{\nabla_\mathbf{p}E_i(\mathbf{p})}{\nabla_\mathbf{p}D(\mathbf{p},E)}\right|\mathrm{sign}\left(\frac{\partial D(\mathbf{p},E)}{\partial E}\right)\mathrm{Re}\,M_{1m_11m_2}(\mathbf{p},E).$$

The condition (9) is satisfied for ultrasoft x radiation, $\omega\lesssim 0.1$ keV. In the case of hard x-ray photons, the inequality (9) is valid only for small angle scattering ($\theta\ll 1/137\omega R$) near the absorption threshold. If the quantities $A^i_{m_1m_2}(\mathbf{p},E)$ are expressed directly in terms of the expansion coefficients $a^L_{\mathbf{p},i}$ of the Bloch wave function[7]

$$\psi_{\mathbf{p},i}(\mathbf{r})=\sum_L a^L_{\mathbf{p},i}R_l(r,E_i(\mathbf{p}))Y_L(\hat{r}),$$

then it is easy to verify that the quantity $Q_{fi}(\mathbf{p})$ is a smooth function of the quasimomentum **p**. In other words, the singularities of the scattering cross section (10) are determined only by the points of tangency of the surfaces $E_1=E_i(\mathbf{p})$ and $E_2=E_f(\mathbf{p})$.[5,8] The tangency points are divided into critical points of the extremal and saddle types and symmetrical critical points.[5,8] Near the critical point $\mathbf{p}=\mathbf{p}_0$ the cross section (10) can be rewritten in the form

$$\sigma(\omega,\omega')\sim\sum_{fi}Q_{fi}(\mathbf{p}_0)\int_B d^3p\,\delta(E_2-E_f(\mathbf{p}))\delta(E_1-E_i(\mathbf{p})).$$

Thus, the cross section $\sigma(\omega,\omega')$ has the same singularities as the quantity

$$\varphi(\omega,\omega')=\sum_{fi}\int_B d^3p\,\delta(E_2-E_f(\mathbf{p}))\delta(E_1-E_i(\mathbf{p})).\tag{11}$$

It is obvious that the singularities in the $L, M,\ldots$ fluorescence spectra of metals will also be determined by the singularities of $\varphi(\omega,\omega')$. The function $\varphi(\omega,\omega')$ is connected with the interband density of states $\rho(E,\omega)$[5] by the relation

$$\varphi(\omega,\omega')=\rho(\omega'+E_0-E_F,\omega-\omega').\tag{12}$$

The singularities of the interband density of states $\rho(E,\omega)$ were investigated earlier in works on the theory of the photoemission spectra of metals.[5,8] We therefore leave out in the present paper the analysis of the singularities of $\varphi(\omega,\omega')$. For a semiquantitative estimate of the dependence of the shape of the emission band on the frequency of the initial photon we can assume that

$$\sigma(\omega,\omega')\sim\varphi(\omega,\omega').$$

Figure 1 shows the function $\varphi(\omega,\omega')$ for silver. Calculation of the interband density of states with allowance for the finite lifetime of the single-particle state ($\gamma=0.3$ eV), carried out in[9], has enabled us to reconstruct the $\varphi(\omega,\omega')$ curve for constant ω in the region $E=E_1-E_2<-2$ eV. The strong dependence of the shape of the emission band on ω indicates that the traditional description[3,7] of x-ray fluorescence does not hold near the absorption threshold.

3. ALKALI METALS

Consider the case of alkali metals, in which the Fermi surface is in the first band. We write for them the function $\varphi(\omega,\omega')$ in the free-electron approximation:

$$\varphi(\omega,\omega')=\sum_\mathbf{g}\int d^3p\,\delta(E_1-p^2)\delta(E_2-(\mathbf{p}+\mathbf{g})^2)$$
$$\sim\sum_g\int_{-1}^{1}dx\,\delta\left(x-\left(\frac{E_2-E_1-g^2}{2g\sqrt{E_1}}\right)\right).$$

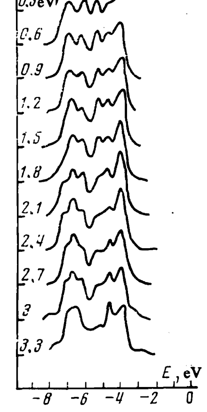

FIG. 1. The function $\varphi(\omega,\omega')$ for silver; $E=\omega'+E_0-E_F$.

It is obvious that $\varphi(\omega, \omega')$ will be different from zero if

$$|E_2-E_1-g^2|\leqslant 2\sqrt{E_1}g. \tag{13}$$

Rewriting (13) in the form

$$E_F\geqslant E_1\geqslant(\sqrt{E_2}-g)^2,$$

we determine the width Γ of the emission band by the relation

$$\Gamma=\mathrm{Max}\,[E_F-(\sqrt{E_2}-g_n)^2]\geqslant 0, \tag{14}$$

where we have numbered the reciprocal-lattice vectors in the sequence

$$0<g_1<g_2\ldots\,.$$

It follows directly from (14) that

$$\begin{aligned}\Gamma=0 \quad &\text{for } E_F\leqslant E_2\leqslant(g_1-\sqrt{E_F})^2,\\ \Gamma=\Gamma_1 \quad &\text{for } (g_1-\sqrt{E_F})^2\leqslant E_2\leqslant{}^1/_4(g_1+g_2)^2,\\ \Gamma=\Gamma_n \quad &\text{for } {}^1/_4(g_{n-1}+g_n)^2\leqslant E_2\leqslant{}^1/_4(g_n+g_{n+1})^2,\ n=2, 3,\ldots\,.\end{aligned} \tag{15}$$

Here

$$\Gamma_n=E_F-(\sqrt{E_2}-g_n)^2.$$

A plot of (15) is shown in Fig. 2. Attention should be called to the fact that in alkali metals the interference leads to a shift of the threshold of the fluorescence yield. Emission takes place only at $\Delta\omega>\Delta$ and not at $\Delta\omega>0$, where

$$\Delta=g_1(g_1-2\sqrt{E_F}), \qquad \Delta\omega=\omega+E_0-E_F.$$

For example, $\Delta\sim 2.4$ eV for lithium. The shift of the threshold means that in lithium in the region $\Delta\omega<\Delta$ the interference blocks most radiative decay channels of the state with a hole in the K level. Recognizing that for alkali metals the free-electron approximation within the limits of the first zone describes well the dispersion of $E(\mathbf{p})$, we can assume that this result is reliable enough.

4. CONCLUSIONS

Just as in the cross section for the photoelectron yield,[5,8] the dynamics of the singularities of the cross section $\sigma(\omega, \omega')$ as functions of ω yields information on the topology of the equal-energy surfaces in p space. Unlike photoemission, however, the value of $\sigma(\omega, \omega')$ does not depend on the surface properties of the sample. Thus, a joint investigation of the spectra of metals by these two methods makes it possible to separate experimentally the surface singularities[8] from the singularities of the interband density of states.

Let us dwell briefly on the possibilities of experimentally verifying the effects considered in this paper. In the experiment, the following conditions should be satisfied.

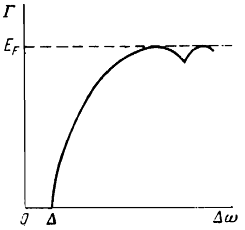

FIG. 2. Dependence of the width Γ of the emission band on Δω.

1. The degree of monochromatization ($\delta\omega$) of the exciting radiation in the region $\omega\lesssim 0.1$ keV should not be worse than 0.1 eV.

2. The monochromatic photon beam incident on the sample should have a sufficiently high intensity I_ω.

Condition 1 can be easily attained.[10,11] The second condition is much more difficult. The intensity $I_\omega\sim 10^{12}$ photons/cm^2 sec with $\delta\omega\sim 0.1$ eV, which is realistic for modern electron storage rings,[10,11] is sufficient for the registration of fluorescence spectra in the region $\omega'\lesssim 40$ eV. This range contains, for example, the L band of Na and the $M_{\mathrm{II,III}}$ bands of Ti, V, Cr, and Mn. To proceed into the harder region, more intense radiation sources are needed.

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