# Auger resonances in Compton scattering

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Resonances are observed in inelastic scattering of $\gamma$ quanta by heavy atoms for energy transfers of the order of the differences of ionization potentials of inner shells. The resonances are attributed to the effect of Auger relaxation on the dynamics of the electromagnetic transition.

## 1. INTRODUCTION

In the spectrum of photon Compton scattered by electrons bound in an atom, in contrast to the scattering by free electrons, the energy of the photon scattered at a given angle varies from zero up to the maximum value $\hbar\omega_i - E_b$, where $\hbar\omega_i$ is the energy of the incident photon and $E_b$ is the binding energy of the atomic electron. As a result the $\delta$-shaped Compton line is broadened into the bell-shaped "Compton profile." The study of the form of the "Compton profile" is widely used for the determination of the momentum distribution of electrons in solids.[1]

The study of the fine details of the spectra of inelastic scattering of $\gamma$ quanta by bound atomic electrons is fraught with considerable experimental difficulties, caused by the low intensity of the scattering process. Threshold singularities were observed in these spectra,[2] caused by the "cutoff" of the contribution of $K$-electrons to Compton scattering for photon energy losses $\Delta E \equiv \hbar\omega_i - \hbar\omega_f = I_K$, where $I_K$ is the ionization potential of the $K$-shell. Peculiarities in the formation of peaks of x-ray flourescence under threshold excitation conditions (x-ray resonance Raman scattering) were studied both experimentally and theoretically.[3,4]

We have first observed resonances in the spectra of photons undergoing inelastic scattering off many-electron atoms of lanthanides (Gd, Tb, Ho, Tm) for photon energy losses $\Delta E$ of the order of the differences in the ionization potentials of inner atomic shells ($\Delta E \approx I_K - I_L$) with relative intensity $\sim 10^{-4}$ and depth of modulation in excess of three standard deviations. The measurements were performed on an apparatus of high luminosity[5,6] with increased radiation shielding in the emitter–detector interval. By means of control measurements of the natural background it was established that the observed peculiarities were not connected with admixtures of components of the uranium-thorium series in the samples used.

The appearance of resonances for the energy transfer $\Delta E \equiv \hbar\omega_i - \hbar\omega_f \approx I_K - I_L$, corresponding to the transition of a $K$-electron to the $L$-shell, is unexpected since in the atoms under consideration the $L$-shell is full, and such a transition if forbidden by the Pauli principle. One may suppose that the discovered peculiarities are due to a stepwise process, in which first an electron from the $L$-shell is ionized and then during the lifetime of the $L$-vacancy a second photon is scattered by the atom with a transition of an electron from the $K$- to the $L$-shell. The relative contribution of such a process equals

$$\delta = \frac{(d\sigma/d\Omega_f)_{1s-2s}}{(d\sigma/d\Omega_f)_{nres}}\,\sigma_L\tau_L F,$$

where $(d\sigma/d\Omega_f)_{1s-2s}$, $(d\sigma/d\Omega_f)_{\rm nres}$, $\sigma_L$ are respectively the cross sections for $1s-2s$ Raman scattering, background Compton scattering, and photoionization from the $L$-shell, $F$ is the photon flux, $\tau_L$ is the lifetime of the vacancy in the $L$-shell. For the emitter used with activity of 1 Ci one has $F\approx 10^9$ cm$^{-2}$ sec$^{-1}$ and an estimate gives $\delta \sim 10^{-21}$–$10^{-23}$, which is by many orders smaller than the relative intensity of the observed peaks. This shows that the observed resonances are connected with inelastic scattering of a single photon by an atom with filled inner shells.

The region of energy loss $\Delta E \approx I_K - I_L$, in which the resonances were observed, is characteristic of rearrangement of electron shells of an atom with deep inner vacancy. This leads one to suppose that the observed resonances are due to relaxation processes of atomic electrons, that "wedge" into the dynamics of two-photon electromagnetic transitions. Such a mechanism, which is resonant and gives rise to the appearance of peaks for $\omega_i - \omega_f \approx I_K - I_L$, is shown in the diagram (see Fig. 1): the quantum $\hbar\omega_i$ absorbed by the atom causes the production of the autoionized state with vacancy in the $K$-shell, which then decays with the emission of an Auger electron $\mathbf{k}_f$ before the reradiation of the secondary quantum.

The calculation of the relative intensities of the resonances for the indicated mechanism is given in Sec. 3. The experimental technique is described in Sec. 2, and Sec. 4 contains a discussion of the results and conclusions.

## 2. EXPERIMENT

The spectra for inelastic scattering of $\gamma$ radiation on lanthanides (rolled metal Gd, Tb, Ho and Tm 300 $\mu$m thick of high chemical purity) are measured on a high luminosity setup.[5,6] The main feature of the apparatus (Fig. 2) is the use of a collimator (1) in the form of two coaxial disks with the source of $\gamma$ radiation (2) located in the center of the gap between them. The sample, in the form of a closed band of rolled metal, is placed on a cylindrical surface with the same axis as the collimator. The beam of scattered photons is analogously collimated on the detector (4). In comparison with known setups equipped with cylindrical collimators, the

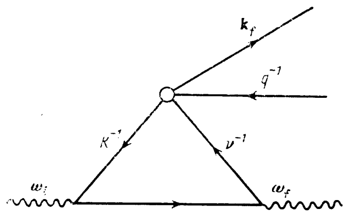


FIG. 1. The diagram defining the resonance contribution to the Compton scattering amplitude.


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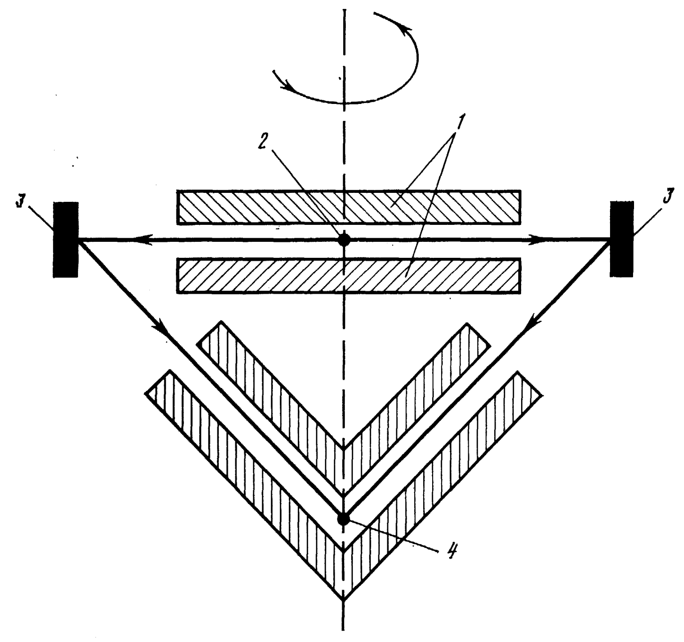


FIG. 2. Schematic of the axial spectrometer for Compton scattering (for notation see text).

spectrometer used here has approximately 200 times the luminosity for the same angular resolution. To suppress the background of photons inelastically scattered from the materials of the construction, the setup was equipped with improved radiation shielding in the interval between the emitter and the detector.

The measurements were performed in the region of strong violation of the conditions for the impulse approximation, and the dominating role of photoabsorption was compensated by the high luminosity of the setup. The radionuclide $^{123}$Te served as the source, with photon energies $\hbar\omega_i = 159.00$ keV with initial activity $\gtrsim 1$ Ci. The scattering angle was 161.5°, the opening of the collimator gap was $\pm 2°$, the special axially sensitive Ge–Li detector had an energy resolution of 600–650 eV (in the 122 keV line of $^{60}$Co) and the nonlinearity of the amplifier-analyzer tract did not exceed 0.2%. Continuous control of the quality of the measurement was achieved with the help of two connected-in-parallel amplitude analyzers AI-1024 using the characteristic flourescent lines $K_\alpha$ and $K_\beta$ of the scatterer and Pb, as well as the peak of elastic scattering at 159.00 keV. For each metal four independent spectra were obtained with statistics of $2\cdot 10^5$ pulses in the peak for 180 eV width of the channel of the analyzer.

In Fig. 3 the spectra for Ho and Tm are given as an example. The threshold singularities can be seen on the profile of the Compton scattering for $\Delta E = I_K$ and at a distance of $\sim$ 10 keV from them the first observed resonances, which are shown in the inset in Fig. 3 magnified in scale. The intensity of the resonance peaks relative to the integral intensity of the Compton spectrum amounts to $\sim 10^{-4}$. The modulation depth of the spectrum in the resonance region exceeds three standard deviations. The $K$-threshold energies $E_{\rm thr}$ and the resonances $E_{\rm res}$ are given in Table I.

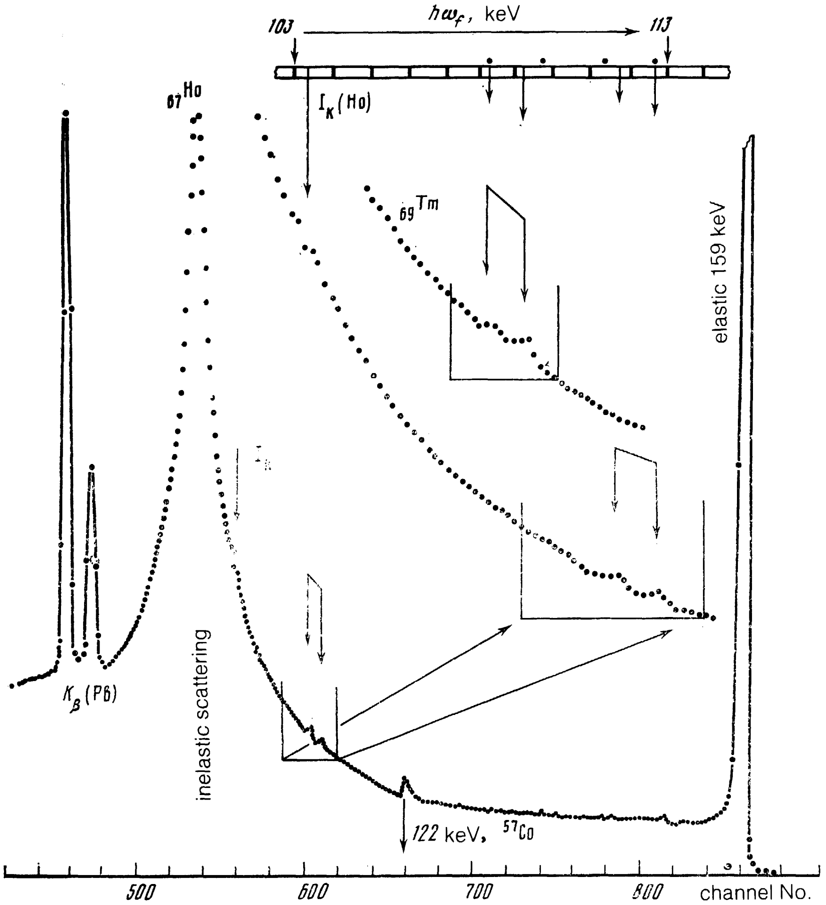


FIG. 3. The spectrum of inelastic scattering of $\gamma$ quanta with 159 keV of energy on Ho and fragments of the spectra for Ho and Tm (in the inset).

TABLE I. Energy of $K$-thresholds $E_{thr}$ and resonances $E_{res}$, keV.

| Sample | $_{64}$Gd | $_{65}$Tb | $_{67}$Ho | $_{69}$Tm |
|---|---|---|---|---|
| $E_{thr}$ | 108.5 | 107.0 | 103.4 | – * |
| $E_{res}$ | { 117.0<br>{ – * | 115.7<br>114.6 | 112.4<br>111.4 | 109.5<br>108.2 |

*Insufficient statistics and resolution.

### 3. THEORY

Under the conditions of the experiment the radiation wavelength considerably exceeds the Bohr radius. Therefore in the description of the interaction of the atom with the electromagnetic field the dipole approximation is sufficient. The differential cross section (in atomic units) for the scattering of the photon $\omega_i$ by the atom in the initial (ground) state $|i\rangle$ with the production of a vacancy in the $q$-shell has the following form:[7]

$$\frac{d^2\sigma}{d\Omega_f\, d\omega_f}=\alpha^2\int d\mathbf{k}_f\omega_i\omega_f^3|A_{if}|^2\delta\left(E_i+\omega_i-E_{q^{-1}}-\omega_f-\frac{k_f^2}{2}\right), \tag{1}$$

$$A_{if}=\langle f(q^{-1},\ \mathbf{k}_f)|\mathbf{de}_fG(E_i+\omega_i)\mathbf{de}_i|i\rangle$$
$$+\langle f(q^{-1},\ \mathbf{k}_f)|\mathbf{de}_iG(E_i-\omega_f)\mathbf{de}_f|i\rangle. \tag{2}$$

Here $\mathbf{d}$ is the dipole moment operator, $\mathbf{e}_i$, $\mathbf{e}_f$ are the polarization vectors of the incident and scattered photons, $E_i$, $E_{q^{-1}}$ are the energies of the atom (ion) in the initial and final states, $\mathbf{k}_f$ is the momentum of the Compton electron,

$$G(\varepsilon)=\sum_\mu\frac{|\Psi_\mu\rangle\langle\Psi_\mu|}{\varepsilon-E_\mu} \tag{3}$$

is the full Green function of the electronic system, $\Psi_i$, $\Psi_f$, and $\Psi_\mu$ are the eigenfunctions of the Schrödinger equation with the interaction between electrons fully taken into account.

Usually in the discussion of Compton scattering on bound atomic electrons[1,8] the exact wave functions $\Psi_i$, $\Psi_f$, and $\Psi_\mu$ are appproximated by the single-determinant wave functions constructed, for example, from Hartree-Fock orbitals. As a result $A_{if}$ reduces to the amplitude for Compton scattering from the electron in the state $|q\rangle$:

$$A_{if}=\sum_n\left\{\frac{\langle\mathbf{k}_f|\mathbf{de}_f|n\rangle\langle n|\mathbf{de}_i|q\rangle}{\varepsilon_q+\omega_i-\varepsilon_n}+\frac{\langle\mathbf{k}_f|\mathbf{de}_i|n\rangle\langle n|\mathbf{de}_f|q\rangle}{\varepsilon_q-\omega_f-\varepsilon_n}\right\}.$$

Here the summation is over a complete set of orbitals, including continuum states. The expression (4) has no singularities for energy transfers of the order of differences of ionization potentials of inner shells. Consequently, one cannot confine oneself in the problem under consideration to the single-electron approximation for $\Psi_{i,f}$ and $G(\varepsilon)$, and it is necessary to take into account higher order terms in the correlation interaction $\lambda_{corr}$ $V_{corr}\equiv HH-{}_{HF}$.

$\Psi_i$, $\Psi_f$, and $G(\varepsilon)$ in Eq. (2) may be expressed as follows:

$$\Psi_{i,f}=\Psi_{i,f}^{HF}+\Delta\Psi_{i,f},\qquad G(\varepsilon)=G^{HF}(\varepsilon)+\Delta G(\varepsilon), \tag{5}$$

where $\Delta\Psi_{i,f}$ and $\Delta G(\varepsilon)$ are higher order terms in the effective constant of the correlation interaction $\lambda_{corr}$. An analysis shows that substitution into Eq. (2) of the more precice values $\Delta\Psi_{i,f}$ and keeping $G^{HF}(\varepsilon)$ leaves $A_{if}$ a smooth function of $\omega_f$ for the $\Delta E$ under consideration. We therefore confine ourselves in the following in Eq. (2) to the Hartree-Fock approximation for the wave functions of the initial and final $\Psi_i^{HF}$ and $\Psi_j^{HF}$, while in $G(\varepsilon)$ we shall keep just the lowest order terms in $\lambda_{corr}$ needed for the explanation of the resonance structure of $A_{if}$. The most intense ($\sim\lambda^0_{corr}$) are single-electron radiative transitions. Since the contributions to $A_{if}$ of excitations from all shells other than the $K$-shell contain no resonances in the region of energy transfers $\Delta E$ of interest, we shall include them in the nonresonance part $A_{if}^{nres}$ of the amplitude and consider in more detail the contribution of $A_{if}$ of the virtual states in $G(\varepsilon)$, corresponding to the excitation of the $K$-electron into unoccupied states $npj$ of the discrete and continuous spectrum ($A_{if}^{res}$). It is of great importance for the description of the process that the state of the atom $|K^{-1}\rangle$ with a vacancy in the $K$-shell is unstable to, decay via the correlation interaction, which gives rise to the emission of an Auger electron produces two vacancies in higher-lying states. The "correct" many-configuration wave functions of the frame, which take into account the strong mixing of the state $|K^{-1}\rangle$ with "decaying" two-hole states, has the following form[9]:

$$|\varkappa,E\rangle=\left(\frac{2}{\pi\Gamma_K}\right)^{1/2}\left\{\frac{1}{[1+x^2(E)]^{1/2}}|K^{-1}\rangle+\frac{\pi x(E)}{[1+x^2(E)]^{1/2}}\sum_{\mu\nu}V_{\mu\nu}(\varkappa,E_{\mu\nu})|\mu^{-1};\ \nu^{-1};\ \varkappa,E_{\mu\nu}\rangle\right\}. \tag{6}$$

Here $x(E)=(E-E_{K^{-1}})/(\Gamma_K/2)$ is the resonance detuning, referred to the halfwidth of the $K$-vacancy,

$$V_{\mu\nu}(\varkappa,E_{\mu\nu})=\langle\mu^{-1};\ \nu^{-1};\ \varkappa,E_{\mu\nu}|V_{corr}|K^{-1}\rangle,$$

$|\boldsymbol{\kappa},E_{\mu\nu}\rangle$ is the wave function of the Auger electron with energy $E_{\mu\nu}=E-E_{\mu^{-1}\nu^{-1}}$ and unit vector $\boldsymbol{\kappa}$ in the direction of the momentum, and the summation over $\mu$ and $\nu$ is over all channels of the Auger-decay of the state $|K^{-1}\rangle$. The functions, Eq. (6), are orthonormalized accurate to leading terms in the correlation interaction $V_{corr}$ between electrons. Ignoring the interaction of the frame with the electron in the state $npj$, responsible for effects of "finite-state interaction" type,[10] we obtain for the part of the Green function corresponding to all excitations that contribute to $A_{if}^{res}$

$$G^{res}(\varepsilon)=\sum_{nj}\int dE\int d\varkappa\frac{|\varkappa,E;\ npj\rangle\langle\varkappa,E;\ npj|}{\varepsilon-E-E_{npj}}. \tag{7}$$

Here $|\boldsymbol{\kappa},E;\ npj\rangle$ are the wave functions with the corresponding filled atomic orbitals, and the summation over $n$ includes integration over the states of the continuous spectrum. Since $d$ is a sum of single-electron operators we find after substitution of Eq. (7) into Eq. (2)

$$\langle \varkappa, E; npj|\mathbf{de}|i\rangle=(2/\pi\Gamma_K)^{1/2}[1+x^2(E)]^{-1/2}\langle npj|\mathbf{de}|1s_{1/2}\rangle. \quad (8)$$

Only the second term in the expression (6) makes a contribution to the matrix element for the transition to the final state, and the orthogonality condition for the orbitals leads to the requirement that one of the vacancies in $|\varkappa,E;npj\rangle$, say $\mu^{-1}$, should coincide with the vacancy in the final state $q^{-1}$:

$$\langle f(q^{-1},\mathbf{k}_f)|\mathbf{de}|\varkappa,E;npj\rangle =\left(\frac{2}{\pi\Gamma_K}\right)^{1/2}\frac{\pi x(E)}{[1+x^2(E)]^{1/2}}\sum_\nu V_{q\nu}(\varkappa,E-E_{q^{-1}\nu^{-1}}) \times\langle\nu|\mathbf{de}|npj\rangle\delta\left(E-E_{q^{-1}\nu^{-1}}-\frac{k_f^2}{2}\right)\delta\left(\varkappa-\frac{\mathbf{k}_f}{k_f}\right). \quad (9)$$

After substitution of Eqs. (7)–(9) into Eq. (2) we obtain for the resonance part of the scattering amplitude

$$A_{if}^{res}=\frac{2}{\Gamma_K}\sum_\nu\frac{x(E_{q^{-1}\nu^{-1}}+k_f^2/2)}{1+x^2(E_{q^{-1}\nu^{-1}}+k_f^2/2)}V_{q\nu}\left(\frac{\mathbf{k}_f}{k_f},\frac{k_f^2}{2}\right) \times\sum_{nj}\left\{\frac{\langle\nu|\mathbf{de}_f|npj\rangle\langle npj|\mathbf{de}_i|1s_{1/2}\rangle}{E_i+\omega_i-E_{q^{-1}\nu^{-1}}-k_f^2/2-E_{npj}} +\frac{\langle\nu|\mathbf{de}_i|npj\rangle\langle npj|\mathbf{de}_f|1s_{1/2}\rangle}{E_i-\omega_f-E_{q^{-1}\nu^{-1}}-k_f^2/2-E_{npj}}\right\}. \quad (10)$$

The factor

$$\frac{x(E_{q^{-1}\nu^{-1}}+k_f^2/2)}{1+x^2(E_{q^{-1}\nu^{-1}}+k_f^2/2)}$$

in Eq. (10) grows rapidly for

$$k_f^2/2\approx E_{Kq\nu}=k_{Kq\nu}^2/2\equiv E_{K^{-1}}-E_{q^{-1}\nu^{-1}},$$

i.e., for energies of the Compton electron coinciding with energies of the $Kq\nu$-Auger electron. With the energy conservation law in Compton scattering [compare with Eq. (1)] taken into account the resonance condition takes on the form

$$\Delta E=\omega_i-\omega_f=E_{K^{-1}}-E_i+E_{q^{-1}}-E_{q^{-1}\nu^{-1}}=E_{Kq\nu}-I_q,$$

and, consequently, the location of the resonances corresponds to the structure of the Auger spectrum. Upon ignoring of the insignificant effect of the additional vacancy in the $q$-shell on the ionization potential of the $\nu$-shell we obtain $\Delta E=I_K-I_\nu$. The remaining entries in Eq. (10) vary weakly over the width of the resonance structure, and one may set $k_f$ in them equal to the corresponding resonance value $k_{Kq\nu}$. As a result we have, with the definition of $x(E)$ taken into account,

$$A_{if}^{res}=\sum_\nu\frac{\omega_i-\omega_f+I_\nu-I_K}{(\omega_i-\omega_f+I_\nu-I_K)^2+\Gamma_K^2/4}V_{q\nu}\left(\frac{\mathbf{k}_f}{k_f},E_{Kq\nu}\right)A_{1s,\nu}(\omega_i), \quad (11)$$

where $A_{1s,\nu}(\omega_i)$ is the amplitude for combination scattering $1s\rightarrow\nu$.

In the neighborhood of an individual resonance the dependence of $A_{ij}^{res}$ on $\omega_f$ has the form shown schematically in Fig. 4, and the cross section for Compton scattering is proportional to $|A_{if}^{nres}+A_{if}^{res}|^2$, where $A_{if}^{nres}$ is the background amplitude. For an analyzer channel width much larger than the width of the resonance $\Gamma_K$, as is the case in our experiment, the interference term in the cross section vanishes as a result of averaging. For the process of $\nu=L$ an estimate of the ratio of the integral intensities of the resonance peaks and the background Compton scattering gives the value

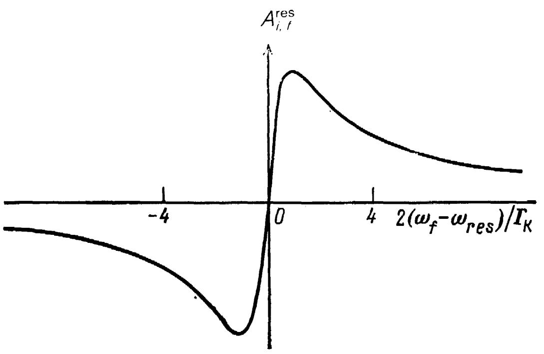


FIG. 4. Dependence of $A_{i,f}^{res}$ (in arbitrary units) on $\omega_f$ in the vicinity of an individual resonance.

$$\delta=\gamma\frac{(d\sigma/d\Omega_f)_{1s\rightarrow2s}}{(d\sigma/d\Omega_f)_{nres}}\sim10^{-3}-10^{-4},$$

which agrees with the experimental value $\sim 10^{-4}$. Here $\gamma$ stands for the partial contribution of all $KLq$-Auger processes to the full width of the $K$-level, while $(d\sigma/d\Omega_f)_{1s\rightarrow2s}$ and $(d\sigma/d\Omega_f)_{nres}$ are the cross sections for combination $1s\rightarrow2s$ and background Compton scattering respectively.

Two peaks separated by $\sim 1$ keV can be seen in the spectrum in Fig. 3. A detailed investigation shows that the appearance of the two peaks is caused by mixing due to the interelectron interaction of two-hole configurations, containing $2s$-vacancies, and the splitting between them is approximately equal to the difference of energies $E_{L_1}-E_{L_{2,3}}$, in the one-particle representation.

## 4. DISCUSSION

Resonances that do not coincide with thresholds and characterisitc excitation lines of the target atom were observed in the spectra of energy losses in inelastic scattering of photons. We assume that these resonances are dynamical in nature and are due to the effect on the scattering of relaxation processes in many-particle systems. The diagram in Fig. 1, which characterizes this effect on Compton scattering of photons on inner atomic shells, describes the totality of the sequential in time processes of photoionization, Auger relaxation, and radiative capture; the diagram is irreducible and connected in the particle-hole representation. The discrete nature of the relaxation energy explains the resonance character with respect to energy transfer of the mechanism under consideration.

In principle, the relaxation mechanism for the formation of resonances in the continuous spectrum of energy losses is common for all inelastic scattering processes on many-particle systems. However, experimentally it is most convenient to observe these resonances in the region of a homogeneous background. Measurement of Compton scattering on inner atomic shells for sufficiently high statistic provides such an opportunity.

Increasing the resolving power of the setup to sizes smaller than the width of the resonance would permit the observation of the form of the resonance line, caused by interference between the resonance and background components of the amplitude, and would additionally confirm the mechanism for resonance formation under consideration.


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