

# Interference of electrons during the $E3$ conversion transition ( $\hbar\omega \approx 76.8$ eV) of the uranium isomer $^{235m}\text{U}$ in a silver lattice

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The uranium isomer  $^{235m}\text{U}$  in a silver lattice is used as an example for a first theoretical analysis of internal conversion accompanied by an interference (rescattering) of the conversion electrons by the nearest neighborhood of the isomer. It is found that these effects play a decisive role for essentially any configuration of the atoms of the medium around the isomer. The greatest effect is found in the case of a close packing (with 12 nearest atoms) corresponding to the geometry of an fcc lattice. The interference makes the conversion probability exceedingly sensitive to geometric factors—the arrangement of the atoms around the isomer and the distances between these atoms—and also to the binding energies of the electron orbitals in which the conversion occurs. As a result, the conversion rate may be either sharply higher (+ 47%) or sharply lower (– 21%) than for an isolated isomer. It is concluded that these effects must be taken into account in interpreting experimental data obtained from electron-conversion spectroscopy of the isomer  $^{235m}\text{U}$ . The possible suppression of the conversion decay of  $^{235m}\text{U}$  in silver for which there are experimental indications might be a consequence of an interference of conversion electrons at the Ag atoms nearest the isomer.

## 1. INTRODUCTION

An internal conversion occurs in the uranium isomer  $^{235m}\text{U}$  through a nuclear  $E3$  transition with a uniquely low energy  $\hbar\omega \approx 76.8$  eV and a fairly long decay half-life  $T_{1/2} \approx 26$  min. Since the conversion process is highly localized in space, information on the local electron density near the isomer nucleus can be extracted from corresponding experiments. This is the essence of conversion-electron spectroscopy. This method has been used widely to study the electronic structure of the valence region of certain chemical compounds of uranium.<sup>1</sup>

However, in the interpretation of the experimental data in some published studies<sup>1–4</sup> by  $^{235m}\text{U}$  conversion-electron spectroscopy, it has been assumed that the conversion electrons do not undergo a further scattering. In other words, the interference of electrons at the atoms of the medium containing the isomer has been completely ignored. For the isomer  $^{235m}\text{U}$  the allowed kinetic energies of the conversion electrons are roughly in the range 25–72 eV, which corresponds to wavelengths  $\lambda \approx 4.7$ – $2.7$  a.u. Since  $\lambda$  turns out to be comparable in magnitude to the interatomic distances, one might expect interference effects to have a substantial influence on the probability for the conversion transition.

In this paper we examine the effect of an interference of conversion electrons on transitions from the  $6p_{1/2}$  and  $6p_{3/2}$  atomic shells of uranium, using as examples systems consisting of the uranium isomer  $^{235m}\text{U}$  in various clusters of silver atoms. Silver was selected for the following two reasons.

1. Because of the selectivity of the  $E3$  conversion transition of the isomer  $^{235m}\text{U}$  in terms of the angular momentum of the initial electron, this transition occurs primarily in the uranium  $6p$  orbitals.<sup>2</sup> In order to separate an interference effect from effects of the chemical surroundings, we would like a situation in which there are no orbitals of the atoms of the medium near the binding energy of the uranium  $6p$  shells ( $\varepsilon_{6p_{1/2}} \approx 24$  eV and  $\varepsilon_{6p_{3/2}} \approx 34$  eV; Ref. 3) with which these

shells might interact (hybridize, split, etc.). This favorable situation prevails in silver.<sup>5</sup>

2. Several experiments were recently carried out to measure (indirectly) the decay half-life of a uranium isomer in the interior of a layer of silver.<sup>6</sup> The results found in several cases were significantly different from the results found in measurements of the  $^{235m}\text{U}$  decay rate on the surface of metals (including silver),<sup>7</sup> for which the relative changes in the decay half-life of the isomer,  $\Delta T_{1/2}/T_{1/2}$ , were less than 5%. These results led Kol'tsov *et al.*<sup>6</sup> to suggest that the  $^{235m}\text{U}$  decay half-life increases by a factor of several units in silver. It was shown theoretically in Ref. 5 that hybridization of the electron shells of uranium surrounded by silver atoms could not lead to any substantial slowing of the conversion process. While remaining in the one-electron model, we need to examine the last of the possibilities which were cited, but not studied, in Ref. 5 for explaining the experimental data of Ref. 6. A pronounced slowing of the  $^{235m}\text{U}$  decay in silver might be caused by a decrease in the conversion-electron wave density near the isomer because of its scattering by the surrounding atoms.

In Sec. 2 we construct the wave function for the continuous spectrum in a muffin-tin (MT) potential. In Sec. 3 we write expressions for the conversion factors which incorporate the scattering of conversion electrons. In Sec. 4 we describe the model which we are using, and we analyze cases of the scattering of an electron by one atom. In Secs. 5 and 6, we present the results, discuss them, and draw some conclusions. The necessary definitions and theorems are gathered in the Appendix.

## 2. CONTINUUM WAVE FUNCTION

An electron emitted in a conversion process is scattered by the atoms around the isomer. The electron has a positive kinetic energy and is thus in a state of the continuum. Finding a correct description of such a state in a multicenter po-

tential of a real substance is a rather complex problem. We restrict the discussion below to a very simple model: a finite cluster with an MT potential. This cluster consists of a U atom surrounded by one or several coordination spheres of silver atoms. Inside the MT spheres, the potential is spherically symmetric, while outside these spheres (and, correspondingly, at infinity) the potential is constant and equal to that potential at the boundaries of the MT spheres which is smallest in absolute value. We denote this constant potential by  $V_{MT}$ . We specify the particular atom in a cluster by the index  $\alpha$ , and we specify its position by a corresponding vector  $\mathbf{R}_\alpha$ .

Since uranium is a heavy element, relativistic effects are important for it. We accordingly assume that both the initial electrons and the final ones are described by bispinor wave functions and that we need to solve the Dirac equation in order to find these functions. The results of the calculations, which are presented in Sec. 4, verify that a relative description of the electrons is necessary.

Precisely the same problem, of constructing a continuum wave function, arises in a description of photoabsorption by electrons of a medium at  $\gamma$ -ray energies close to a threshold. Theories for these effects (EXAFS, XANES, etc.) have been developed well. However, several important features distinguish the conversion transition in the isomer  $^{235m}\text{U}$  from photoabsorption. (1) The conversion process is spatially localized. (2) The transition is of multipolarity  $E3$ . (3) A relativistic description of the electrons is necessary.

For this reason, and also for reference, we will write out all the equations required for calculating conversion probabilities in cases in which the conversion electrons undergo an interference.

To find the continuum wave function, we use a relativistic analog of the scattered-wave method.<sup>8</sup> A similar analysis was carried out in Ref. 9 for bound states (the wave functions of the discrete spectrum) in a cluster.

In the potential which we are considering here, the solution of the Dirac equation for a continuum state ( $\varepsilon > mc^2$ ) can be written out explicitly. For a wave function whose asymptotic behavior far from the cluster can be described as "a plane wave plus an incoming spherical wave," we have the following expansion in bispinors:

$$\Psi_{\mathbf{k}\lambda}^{(-)}(\mathbf{r}) = \begin{cases} (2\varepsilon')^{-1/2} u_{\mathbf{k}\lambda} e^{i\mathbf{k}\mathbf{r}} + i \sum_{\alpha jlm} A_{\alpha jlm}^{(j)} \psi_{\mathbf{k}jlm}^{(j)}(\mathbf{r}_\alpha), \\ \text{for } \mathbf{r} \text{ outside the MT spheres,} \\ \sum_{jlm} A_{\alpha jlm} \psi_{\alpha jlm}(\varepsilon, \mathbf{r}_\alpha), \\ \text{for } \mathbf{r} \text{ inside MT sphere } \alpha. \end{cases}$$

Here  $\mathbf{r}_\alpha = \mathbf{r} - \mathbf{R}_\alpha$ ,  $\mathbf{k} = \mathbf{p}/\hbar$  is the wave vector,  $k = (\hbar c)^{-1}(\varepsilon'^2 - m^2 c^4)^{1/2}$ , the shifted energy is  $\varepsilon' = \varepsilon - V_{MT}$ , and  $\lambda$  specifies the polarization of the electron at infinity.

Inside sphere  $\alpha$ , the solution of the Dirac equation (in the standard representation<sup>10</sup>) with the angular quantum numbers  $j$ ,  $l$ , and  $m$  is

$$\psi_{\alpha jlm}(\varepsilon, \mathbf{r}_\alpha) = \frac{1}{r_\alpha} \begin{pmatrix} f_{jl}(\varepsilon, r_\alpha) \Omega_{jlm}(\mathbf{n}_{r_\alpha}) \\ i^{l+1-l'} g_{j'l'}(\varepsilon, r_\alpha) \Omega_{j'l'm}(\mathbf{n}_{r_\alpha}) \end{pmatrix},$$

$$\mathbf{n}_{r_\alpha} = \mathbf{r}_\alpha / r_\alpha.$$

We normalize this function in such a way that the following condition holds at the boundaries of the MT spheres ( $a_\alpha$  is the radius of the corresponding sphere):

$$\psi_{\alpha jlm}(\varepsilon, \mathbf{r}_\alpha) \Big|_{r_\alpha=a_\alpha} = \cos \delta_{\alpha j l} \psi_{\mathbf{k}jlm}^{(j)}(\mathbf{r}_\alpha) \Big|_{r_\alpha=a_\alpha} - \sin \delta_{\alpha j l} \psi_{\mathbf{k}jlm}^{(N)}(\mathbf{r}_\alpha) \Big|_{r_\alpha=a_\alpha}. \quad (2.2)$$

The phase shifts  $\delta_{\alpha j l}$  introduced in this manner correspond to the scattering of an electron by an individual MT sphere, namely, the sphere  $\alpha$ . The bispinors  $\psi_{\mathbf{k}jlm}^{(j)}$ ,  $\psi_{\mathbf{k}jlm}^{(N)}$ , and  $\psi_{\mathbf{k}jlm}^{(H^{(2)})}$  are defined in the Appendix [see (A1)].

The expansion coefficients  $A_{\alpha jlm}(\mathbf{k}, \lambda)$  and  $A_{\alpha jlm}^{(H^{(2)})}(\mathbf{k}, \lambda)$  are found from the condition that the bispinor wave function in (2.1) is continuous at the boundaries of the MT spheres. Using the plane-wave expansion

$$u_{\mathbf{k}\lambda} e^{i\mathbf{k}\mathbf{r}} = \begin{pmatrix} (\varepsilon' + mc^2)^{1/2} w_\lambda \\ (\varepsilon' - mc^2)^{1/2} (\mathbf{n}_\mathbf{k} \boldsymbol{\sigma}) w_\lambda \end{pmatrix} e^{i\mathbf{k}\mathbf{r}} = \sum_{jlm} A_{\alpha jlm}^{(0)} \psi_{\mathbf{k}jlm}^{(j)}(\mathbf{r}_\alpha),$$

$$A_{\alpha jlm}^{(0)}(\mathbf{k}, \lambda) = e^{i\mathbf{k}\mathbf{R}_\alpha} \frac{2\pi}{k} i^l (\Omega_{jlm}^+(\mathbf{n}_\mathbf{k}) w_\lambda), \quad \mathbf{n}_\mathbf{k} = \frac{\mathbf{k}}{k}$$

and the theorem for combining spherical bispinors [see (A4b) in the Appendix], we find a system of linear algebraic equations with a nonvanishing right side:

$$\sum_{n_2} S_{n_1 n_2}^{-1} A_{n_2} = A_{n_1}^{(0)}, \quad (2.3a)$$

$$A_{n_1}^{(H^{(2)})} + \sin \delta_n A_{n_1} = 0, \quad (2.3b)$$

For brevity we have introduced the generalized index  $n \equiv \{\alpha, j, l, m\}$ . We will also use this index for the phase shifts  $\delta_{\alpha j l}$  to simplify the equations, but with the understanding that the phase shifts are independent of the angular momentum projection  $m$  in the case of a spherically symmetric potential.

The inverse scattering matrix  $\mathbf{S}^{-1}$  in (2.3a) is defined in terms of the structure matrix  $\mathbf{D}^{(H^{(2)})}$  [see (A3d) in the Appendix] and the phase shifts for the scattering by the individual atoms:

$$S_{n_1 n_2}^{-1} = \delta_{n_1 n_2} \cos \delta_{n_2} + i D_{n_1 n_2}^{(H^{(2)})} \sin \delta_{n_2}, \quad (2.4)$$

where  $\delta_{n_1 n_2}$  is the Kronecker delta.

The problem of finding the continuum wave function in a multicenter MT potential has thus been reduced to one of solving the linear, inhomogeneous system of equations in (2.3). The problem has actually been reduced to one of calculating the scattering matrix  $\mathbf{S}$  from its inverse, Eq. (2.4).

We can work from the amplitudes  $\mathbf{A}$  to construct a one-electron density matrix, which we define as follows:

$$\rho_{n_1 n_2}(\varepsilon) \equiv \sum_{\lambda} \int d\omega_{\mathbf{k}} A_{n_1}(\mathbf{k}, \lambda) A_{n_2}^*(\mathbf{k}, \lambda). \quad (2.5)$$

Various physical quantities, in particular, the conversion

probability (Sec. 3), are expressed in terms of this matrix. Let us assume that we have found a solution of (2.3a) by inverting the matrix  $\mathbf{S}^{-1}$ :  $\mathbf{A} = \mathbf{S}\mathbf{A}^{(0)}$ . Substituting it into (2.5), we then find, in matrix form,

$$\rho = \mathbf{S}\rho^{(0)}\mathbf{S}^+. \quad (2.6)$$

Here  $\rho^{(0)}$  is the corresponding matrix for the free electron:

$$\rho_{n_1 n_2}^{(0)}(\varepsilon) = \sum_{\lambda} \int d\omega_k A_{n_1}^{(0)}(\mathbf{k}, \lambda) A_{n_2}^{(0)*}(\mathbf{k}, \lambda) = \left(\frac{2\pi}{k}\right)^2 D_{n_1 n_2}^{(j)}$$

The structure matrix  $\mathbf{D}^{(j)}$  is derived in the Appendix [see (A3a)].

### 3. ELECTRON CONVERSION FACTORS

Let us consider the internal-conversion process for an isomer in a medium in which an electric  $L$ -pole ( $EL$ ) transition of the isomer nucleus occurs, with the subsequent ejection of an electron from a state of the discrete spectrum into the continuum. We assume that the initial and final states of the nucleus are characterized by definite values of the energy  $E$ , the spin  $I$ , and the spin projection  $M$ . We assume that the discrete states of the electron are atomic states with a main quantum number  $n$ , a total angular momentum  $j$ , an angular-momentum projection  $m$ , and a parity  $(-1)^l$ . Such a state can be described by the bispinor<sup>10</sup>

$$\psi_{njlm}(\mathbf{r}) = \frac{1}{r} \begin{pmatrix} f_{njl}(r)\Omega_{jlm}(\mathbf{n}_r) \\ i^{l+1-l'} g_{njl'}(r)\Omega_{j'l'm}(\mathbf{n}_r) \end{pmatrix},$$

$$\int_0^{\infty} dr (f_{njl}^2 + g_{njl'}^2) = 1.$$

We use an index  $n$  to specify quantities which refer to the nucleus, while  $e$  specifies quantities which refer to the electrons. We denote the initial and final states of the system by indices 1 and 2; the energy of the conversion transition is then  $\hbar\omega = E_1 - E_2$ .

The probability for a conversion process of this sort (for  $L \neq 0$ ), averaged over the initial states and summed over all the final states of the nucleus and the electron, is given by<sup>3,4</sup>

$$w = \frac{e^4 m}{\hbar^3} \left(\frac{R_{(n)}}{a_B}\right)^{2L} \frac{1}{2I_1+1} |\langle I_2 \| EL \| I_1 \rangle_{(n)}|^2 w_{(e)}, \quad (3.1)$$

where  $R_{(n)}$  is the radius of the nucleus, and  $a_B$  is the first Bohr radius. The dimensionless reduced nuclear matrix elements  $\langle I_2 \| EL \| I_1 \rangle_{(n)}$  are given by

$$R_{(n)}^L \langle I_2 \| EL \| I_1 \rangle_{(n)} (-1)^{I_2-M_2} \begin{pmatrix} I_2 & L & I_1 \\ -M_2 & M & M_1 \end{pmatrix}$$

$$= \langle E_2 I_2 M_2 | \left( \int d^3r r^L Y_{LM}(\mathbf{n}_r) \hat{\rho}_{(n)}(\mathbf{r}) \right) | E_1 I_1 M_1 \rangle \quad (3.2)$$

They do not depend on the electronic structure. All the information on this structure is incorporated in the factor  $w_{(e)}$ , which is called the "electron conversion factor":<sup>3</sup>

$$w_{(e)} = \frac{8\pi^2}{m^2 c^2 k a_B^2} \frac{1}{(2j_1+1)(2L+1)}$$

$$\times \left[ \frac{(k^{(0)} a_B)^{L+1}}{(2L+1)!!} \right]^2 \sum_{j_2 l_2} N_{j_2 l_2} |\langle j_2 l_2 \| EL \| j_1 l_1 \rangle_{(e)}|^2, \quad (3.3)$$

where  $k^{(0)} = \omega/c$ . The conversion factors in Eq. (3.3) have

been defined in such a way that they are dimensionless. The operator  $\hat{\rho}_{(n)}(\mathbf{r})$  in Eq. (3.2) represents the charge density of the nucleus.

As in (3.1), a factorization of the expression for the conversion probability is possible under the condition  $\omega R_{(n)}/c \ll 1$ . This condition clearly holds for the soft transition with  $\hbar\omega \approx 76.8$  eV in <sup>235m</sup>U. In deriving Eq. (3.1) we also assumed that the electron does not penetrate into the nucleus. The reduced  $EL$  electron matrix elements in Eq. (3.3) are nonzero only if element  $l_2 + L + l_1$  is even; they are given by

$$\langle j_2 l_2 \| EL \| j_1 l_1 \rangle_{(e)} = \int_0^{R_{MT}} dr h_L^{(1)}(k^{(0)}r)$$

$$\times [f_{j_2 l_2}(r) f_{n j_1 l_1}(r) + g_{j_2 l_2'}(r) g_{n j_1 l_1'}(r)]$$

$$\times \langle j_2 l_2 \| L \| j_1 l_1 \rangle - \left(\frac{2L+1}{L}\right)^{1/2} \int_0^{R_{MT}} dr h_{L-1}^{(1)}(k^{(0)}r)$$

$$\times [i^{l_2+l_1-l_1'} f_{j_2 l_2}(r) g_{n j_1 l_1'}(r)$$

$$\times \langle j_2 l_2 \| LL-1 \| j_1 l_1 \rangle + i^{l_2-l_1-l_1'} g_{j_2 l_2'}(r) f_{n j_1 l_1}(r)$$

$$\times \langle j_2 l_2' \| LL-1 \| j_1 l_1 \rangle]. \quad (3.4)$$

The reduced matrix elements  $\langle j_2 l_2 \| L \| j_1 l_1 \rangle$  and  $\langle j_2 l_2 \| LL-1 \| j_1 l_1 \rangle$  in (3.4) which arise upon an integration over the angular variables are derived in the Appendix [see (A2a) and (A2b)]. The normalization of the continuum wave function is determined by Eq. (2.2) and corresponds to the asymptotic behavior

$$f_{jl}(r \rightarrow \infty) = 2(\varepsilon + mc^2)^{-1/2} \sin(kr - \pi l/2 + \delta_{jl}).$$

Since the  $E3$  conversion transition is spatially localized and occurs in a region with a size smaller than the first Bohr radius,<sup>2</sup> it is a good approximation to use simply the MT sphere of the isomer atom in the calculation of the electron matrix elements  $\langle j_2 l_2 \| EL \| j_1 l_1 \rangle_{(e)}$ ; this is the approach which we have taken in (3.4).

Expression (3.3) for the electron conversion factors differs from the corresponding expression for the case of an isolated atom only in the appearance of a factor  $N_{j_2 l_2}$ , which is proportional to the probability for the conversion electron to be near the isomer nucleus (more precisely, inside its MT sphere), in a state with quantum numbers  $j_2$  and  $l_2$ . For the isolated atom we would have  $N_{j_2 l_2} = 1$ . When we take the scattering of the conversion electron by the atoms of the medium into account, we find that  $N_{j_2 l_2}$  deviates from one. In our model, of a finite cluster with an MT potential, the quantity  $N_{j_2 l_2}$  is expressed in a simple way in terms of the density matrix  $\rho$  introduced above [see Eq. (2.5)]. Specifically, this quantity is essentially the average of its diagonal part for the sphere  $\alpha$  which corresponds to the isomer atom:

$$N_{j_2 l_2} = \left(\frac{k}{2\pi}\right)^2 \frac{1}{2j_2+1} \sum_{m_2=-j_2}^{j_2} \rho_{\alpha j_2 l_2 m_2, \alpha j_2 l_2 m_2} | \alpha - \text{isomer atom} \rangle. \quad (3.5)$$

### 4. SCATTERING PHASE SHIFTS

We turn now to a study of the "uranium-in-silver" system. We carry out the calculations in two steps. First, we use

the ASA-LMTO method to calculate a self-consistent MT potential in the model of an isolated impurity:<sup>11</sup> a uranium atom in an infinite, periodic, fcc lattice of Ag with a lattice constant  $a = 7.7217$  a.u. The radius of the MT sphere of Ag is determined (as in the ASA approach) by equating the volume of the MT sphere to the volume of the Wigner-Seitz cell. This radius is found to be  $R_{Ag} = 3.0176$  a.u. In this model we first calculate the electronic structure and the potential of an ideal silver crystal. We next replace a Ag atom by a U atom at one lattice site. The U atom has the normal electronic configuration of valence electrons,  $5f^3 6d^1 3/2 7s^2 1/2$ . We impose self-consistency under the assumption that the uranium does not affect the surrounding Ag atoms. The potential of the silver thus remains the same for all atoms. The radius of the MT sphere of uranium,  $R_U = 3.2324$  a.u., is chosen in accordance with the same rule as for Ag, but we use the intrinsic uranium lattice in the  $\gamma$  phase. The electrons of the  $6d^1 3/2, 7s^2 1/2$  valence shells of uranium and those  $4d^4 3/2, 4d^6 5/2, 5s^1 1/2$  of silver are collectivized. The other shells are assumed to be "frozen," and their electrons do not participate in the imposition of self-consistency. This comment also applies to the  $6p$  shells of U, which are treated as atomic. This approximation is completely justified, since it was shown in Ref. 5 that the  $6p$  shells of a U atom change only slightly in silver.

All that was the first step. In the second step, the potentials found for the U and Ag atoms are used to calculate continuum wave functions and electron conversion factors for finite clusters.

Before we derive the continuum wave functions for a cluster, let us analyze the scattering of electrons by individual U and Ag atoms. We use the MT potentials of these atoms which we calculated in the first step.

Figure 1a (for U) and Fig. 1b (for Ag) show the phase shifts  $\delta_{jl}$  and the partial scattering cross sections  $\sigma_l$  in the region of kinetic energies of the conversion electrons in which we are interested. The phase shifts are calculated in accordance with definition (2.2). In this case we have

$$\sigma_l = \left(\frac{2\pi}{k}\right)^2 \sum_j (2j+1) \sin^2 \delta_{jl},$$

where the angular momentum  $j$  takes on two values for non-zero  $l$ :  $j = l \pm 1/2$ . For  $l = 0$ , we have  $j = 1/2$ .

We should first point out that there are large differences between the phase shifts  $\delta_{p_{1/2}}$  and  $\delta_{p_{3/2}}$  ( $\sim 20$  eV) and also between  $\delta_{d_{3/2}}$  and  $\delta_{d_{5/2}}$  ( $\sim 10$  eV) in the case of uranium. These differences between the phase shifts with identical values of  $l$  are due entirely to relativistic effects. The conversion electrons of the  $^{235m}\text{U}$  uranium isomer must therefore be

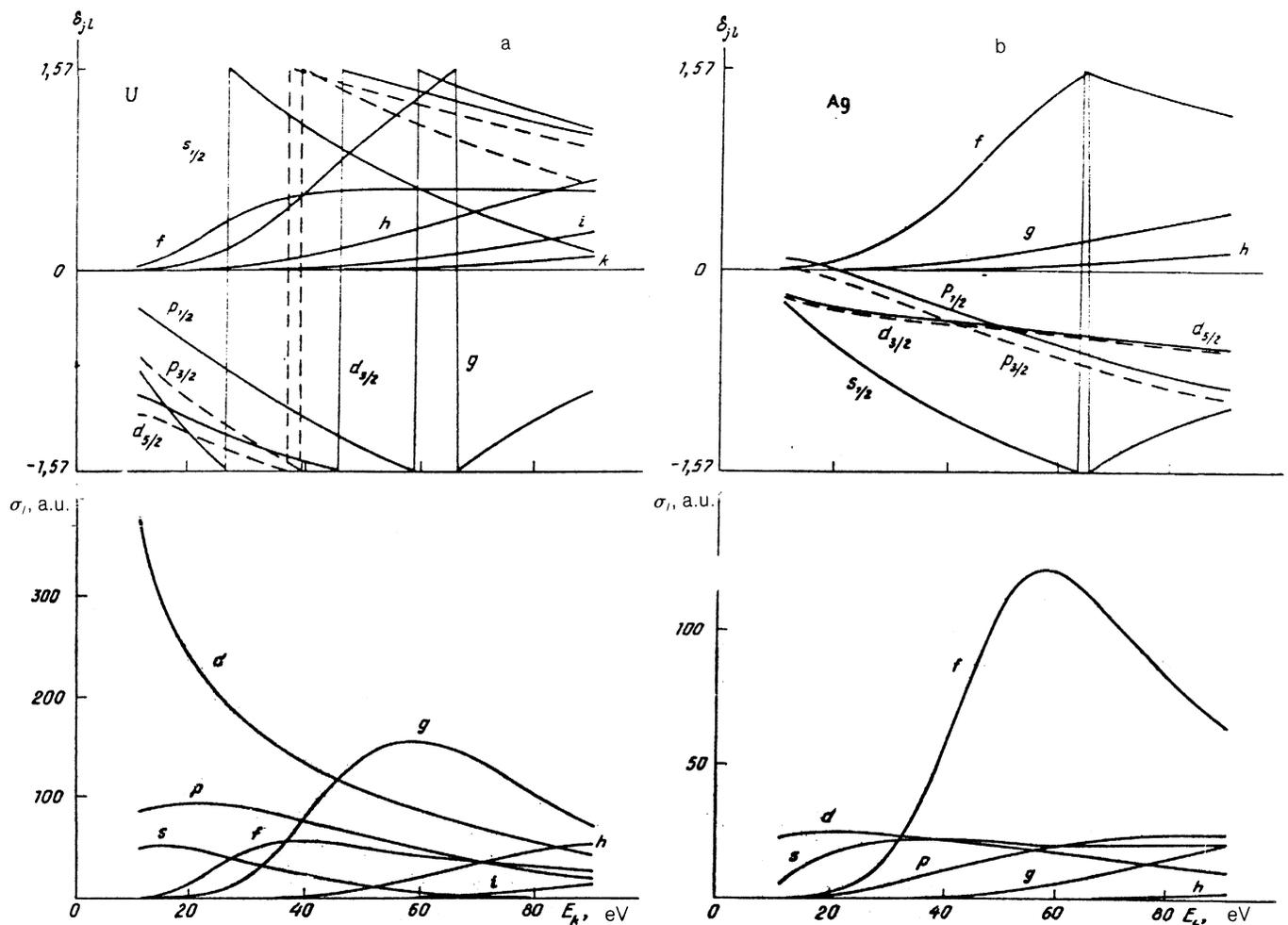


FIG. 1. The phase shifts  $\delta_{jl}$  and the partial cross sections  $\sigma_l$  for scattering for (a) uranium and (b) silver versus the kinetic energy of the electron,  $E_k = \varepsilon - mc^2 - V_{MT}$ . The phase shifts are corrected to the interval  $(-\pi/2, \pi/2)$ .

TABLE I. The ratio  $\sigma_l / [\pi(R_0/2)^2]$  for angular momenta  $l \leq 7$  for the scattering of an electron by the potentials of U and Ag.

$l$	$6p_{1/2}$ ( $E_k = 51.70$ eV)		$6p_{3/2}$ ( $E_k = 63.34$ eV)	
	U	Ag	U	Ag
0 (s)	0,47	0,85	0,21	0,72
1 (p)	2,51	0,71	1,83	0,89
2 (d)	4,35	0,87	3,35	0,86
3 (f)	2,05	4,88	1,67	5,05
4 (g)	6,17	0,12	6,46	0,30
5 (h)	0,45	$4,4 \cdot 10^{-3}$	1,09	$1,6 \cdot 10^{-2}$
6 (i)	$2,3 \cdot 10^{-2}$	$1,1 \cdot 10^{-4}$	$9,7 \cdot 10^{-2}$	$6,3 \cdot 10^{-4}$
7 (k)	$8,0 \cdot 10^{-4}$	$1,9 \cdot 10^{-6}$	$5,4 \cdot 10^{-3}$	$1,6 \cdot 10^{-5}$

Note. The electrons are ejected from the  $6p_{1/2}$  shell ( $\varepsilon_b \approx 33.94$  eV) and the  $6p_{3/2}$  shell  $\varepsilon_b \approx 22.30$  of uranium with kinetic energies  $E_k = \hbar\omega - \varepsilon_b - V_{MT}$ . The energy of the nuclear transition is  $\hbar\omega = 76.8$  eV.

dealt with relativistically, even at low kinetic energies,  $E_k \ll mc^2$ . For Ag, the relativistic effects are much weaker.

The scattering by the individual atoms in the crystal lattice is conveniently characterized by the ratios  $\sigma_l / [\pi(R_0/2)^2]$  where  $R_0 = a/2^{1/2}$  is the interatomic distance ( $R_0 = 5.4601$  a.u.), and  $\pi(R_0/2)^2$  might be called the "area of an atom" in the lattice. Using the partial scattering cross sections in Fig. 1, a and b, we obtain Table I, where these ratios are written for the kinetic energies of the conversion electrons ejected from the  $6p_{1/2}$  and  $6p_{3/2}$  shells of uranium at  $\hbar\omega = 76.8$  eV.

It can be seen from Fig. 1 and also Table I that the scattering is extremely important (in the energy range of interest here) for angular momenta up to  $l_{\max} = 5$  for U and  $l_{\max} = 4$  for Ag. Since the partial cross sections for angular momenta  $\leq l_{\max}$  are comparable in magnitude to the "area of an atom" in the lattice, the rescattered waves cannot be treated as small perturbations in the main wave. For this reason, we cannot use the method which is ordinarily used to describe EXAFS. That method deals with the scattering of the electrons by the atoms of the medium as a sequence of single scattering, double scattering, etc. In the case at hand, multiple scattering must be taken into account comprehensively. Mathematically, this means that we have to exactly invert the inverse scattering matrix in Eq. (2.4). However, that matrix is always of infinite size (in terms of the angular momenta  $l, m$ ), even in a bounded cluster, and no explicit expression can be written for it in the general case. Nevertheless, as we see from Table I, the scattering weakens rapidly with increasing  $l$  (at a fixed energy). In real calculations it is thus assumed that at  $l > l_{\max}$  the phase shifts are zero  $\delta_{jl} = 0$ , and the matrix is "cut off" in this manner. In our calculations we used  $l_{\max} = 7$  for U and  $l_{\max} = 5$  for Ag.

## 5. RESULTS AND DISCUSSION

Experimentally, the local surroundings of a U atom in silver are not known. They could apparently vary in terms of both the number of neighboring atoms and the distances between them. To model this situation, we should look at various configurations of the silver around a U atom. The chemical shifts of the uranium  $6p$  levels, in particular, will vary with the local surroundings. There is also the possibility that the fourfold-degenerate  $6p_{3/2}$  level will be split into two sublevels by the low-symmetry electrostatic field of the medi-

um. To some extent, energy shifts of this sort can be simulated by varying the nuclear transition energy  $\hbar\omega$ . All these factors should have a strong influence on the wave function of the conversion electrons and thus on the conversion probability.

The formalism described in the preceding sections of this paper has been used to calculate  $w_{6p_{1/2}}$  and  $w_{6p_{3/2}}$ , the conversion factors per electron for transitions from the  $6p_{1/2}$  and  $6p_{3/2}$  atomic shells, respectively, of uranium in silver and also their partial sum:

$$w_{6p} = \frac{n_{6p_{1/2}} w_{6p_{1/2}} + n_{6p_{3/2}} w_{6p_{3/2}}}{n_{6p_{1/2}} + n_{6p_{3/2}}}.$$

Here  $n_{6p_{1/2}} = 2$  and  $n_{6p_{3/2}} = 4$  are the numbers of electrons in the corresponding subshells. The total conversion probability in the isomer  $^{235m}\text{U}$  is determined essentially completely by the factor  $w_{6p}$  (Ref. 5).

Two calculations were carried out to investigate the effect of the nearest neighborhood on the conversion probability.

In the first calculation, we determined the effect of various configurations of Ag atoms around the uranium isomer. For this purpose we calculated the electron conversion factors in the following symmetric clusters:  $\text{UAg}_4$  (a tetrahedron),  $\text{UAg}_6$  (an octahedron),  $\text{UAg}_8$  (a cube), and  $\text{UAg}_{12}$  (a cuboctahedron). The arrangement of atoms in the latter cluster ( $\text{UAg}_{12}$ ) corresponds to the geometry of an fcc lattice. This calculation was carried out at a fixed distance  $R_{\text{U-Ag}} = R_0$ , where  $R_0$  is the distance between the atoms in the silver crystal. The energy of the nuclear transition,  $\hbar\omega$ , was varied.

In the second calculation we studied the effect of a variation in the distance between the atoms. In this case the electron conversion factors were calculated for the experimental transition energy  $\hbar\omega_0 = 76.8$  eV for the  $\text{UAg}_{12}$  cluster with distances  $R_{\text{U-Ag}}$  ranging from  $0.75R_0$  to  $1.25R_0$ .

The results of these calculations are shown in Figs. 2 and 3. Figure 4 shows conversion factors for  $\text{UAg}_{12}$  for three distances  $R_{\text{U-Ag}}$  as a function of the transition energy  $\hbar\omega$ . The labels on the curves specify the conversion factors: 1— $w_{6p_{1/2}}$ ; 2— $w_{6p_{3/2}}$ ; 3— $w_{6p}$ . The dotted lines show the corresponding factors for the isolated U atom. The same potentials for U and Ag were used in all the calculations. These

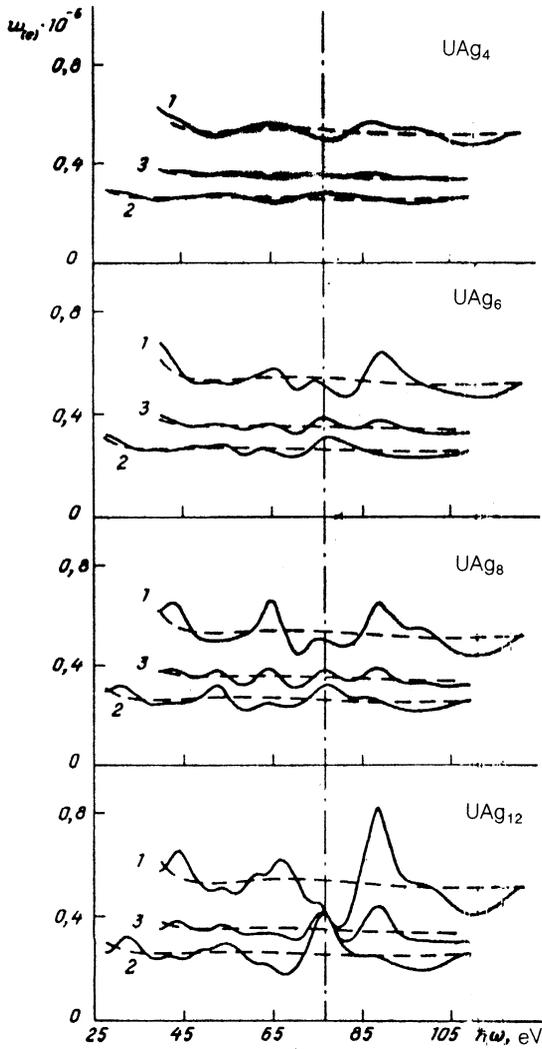


FIG. 2. Electron conversion factors  $w_{6p_{1/2}}$  (1),  $w_{6p_{3/2}}$  (2), and  $w_{6p}$  (3) versus the energy of the conversion transition,  $\hbar\omega$ , for  $\text{UAg}_x$  clusters, where  $x = 4, 6, 8$ , and  $12$ . The dotted lines show the corresponding conversion factors for the isolated U atom.

potentials were calculated by the method described in Sec. 4 for the normal state of the fcc lattice of silver.

We wish to stress that, since we treated the  $6p$  shells of U as atomic, the conversion probabilities for the uranium-silver system differ from those in the case of isolated U only by virtue of the change in the wave function of the conversion electron. This approach allows us to single out the interference effects in their pure form.

It can be seen from Fig. 2 that the interference has only a minor effect in the case of the tetrahedral cluster with four silver atoms. When the number of neighboring atoms is large, however, interference effects become extremely important. They are seen most clearly in the case of the last cluster, which corresponds to the arrangement of atoms in an fcc lattice. Analysis of the data in Figs. 2–4 shows that the total conversion probability and, especially, the probabilities for conversion at the individual shells of U are extremely sensitive to both (on the one hand) the arrangement of Ag atoms around the U and the distances between these atoms and (on the other) the energies of the U shells in which the conversion occurs.

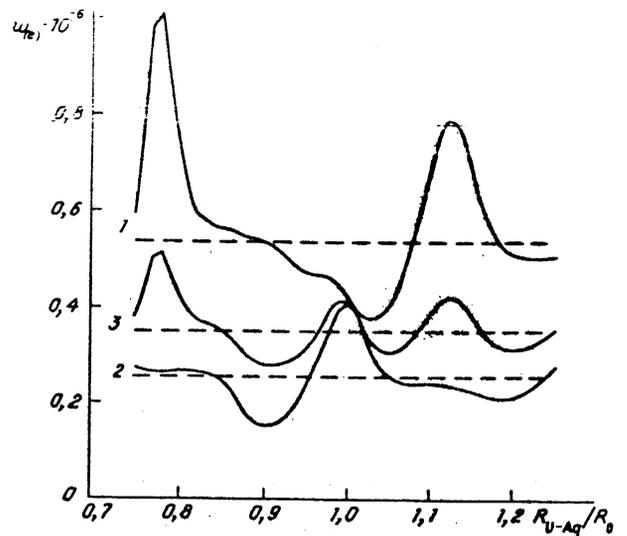


FIG. 3. The electron conversion factors  $w_{6p_{1/2}}$ ,  $w_{6p_{3/2}}$ , and  $w_{6p}$  for a  $\text{UAg}_{12}$  cluster versus the distance between atoms,  $R_{\text{U-Ag}}/R_0$ . The notation is the same as in Fig. 2.

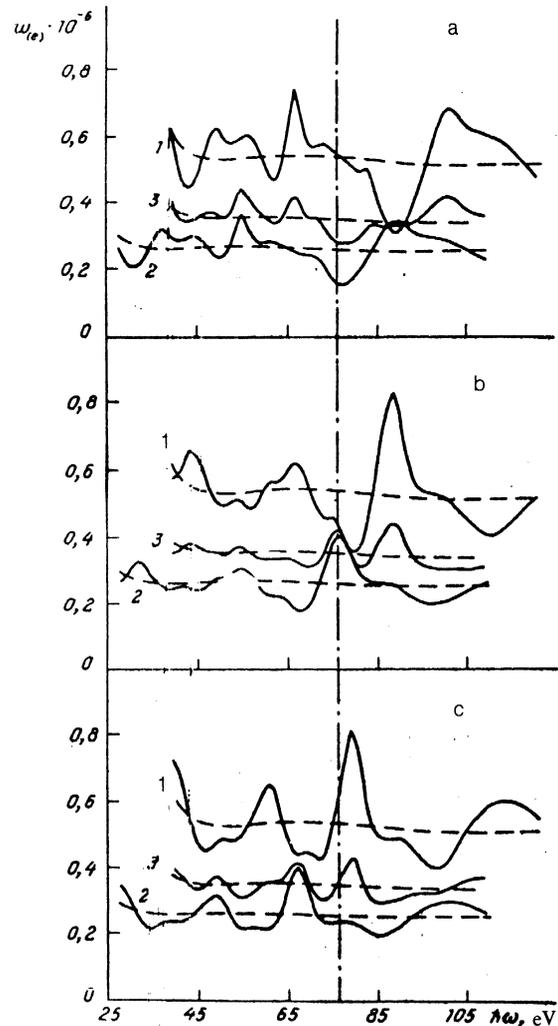


FIG. 4. The electron conversion factors  $w_{6p_{1/2}}$ ,  $w_{6p_{3/2}}$ , and  $w_{6p}$  for a  $\text{UAg}_{12}$  cluster as a function of the energy of the conversion transition,  $\hbar\omega$ , for various distances between the atoms. a— $R_{\text{U-Ag}} = 0.9R_0$ ; b— $R_0$ ; c— $1.1R_0$ . The notation is the same as in Fig. 2.

TABLE II. Nonzero reduced electron matrix elements (atomic units) for an  $E3$  conversion transition from the  $6p_{1/2}$  and  $6p_{3/2}$  shells of U.

$l_2 j_2$	$l_1 j_1$	$\langle j_2 l_2    E3    j_1 l_1 \rangle_{(e)}$
$d_{5/2}$	$p_{1/2}$	$5,342 \cdot 10^5$
$g_{7/2}$	$p_{3/2}$	$4,255 \cdot 10^{-3}$
$d_{3/2}$	$p_{3/2}$	$1,575 \cdot 10^5$
$d_{5/2}$	$p_{1/2}$	$0,973 \cdot 10^5$
$g_{7/2}$	$p_{1/2}$	$0,062 \cdot 10^{-2}$
$g_{9/2}$	$p_{3/2}$	$1,185 \cdot 10^{-3}$

Note. The energies of the  $6p$  shells of U and the energy of the nuclear transition are the same as in Table I.

We would like to call attention to one aspect of the  $E3$  conversion electron for initial electrons with an angular momentum  $l = 1$  (the  $p$  shell). Table II shows the nonzero reduced electron matrix elements in Eq. (3.4) for transitions from the  $6p_{1/2}$ - and  $6p_{3/2}$ -shells of U. It follows from this table that the ejection of bound  $p$  electrons into a continuum state in the course of the  $E3$  conversion transition essentially occurs only with an angular momentum  $l = 2$  with respect to the isomer atom. Interference effects are thus important only under conditions such that there is a strong scattering of the conversion electrons, which leads to a significant deformation of their  $d$  waves near the isomer atom.

It can be seen from Figs. 2–4 that the spectra for  $w_{6p_{1/2}}$  and  $w_{6p_{3/2}}$  are very similar in shape, with only a relative shift equal to the difference between the binding energies of the  $6p$  shells of uranium. The slight difference is explained on the basis that (as can be seen from Table II) a  $p_{3/2} \rightarrow d_{3/2}$  transition is possible (it does not occur in the case of the  $6p_{1/2}$  shell) and makes an additional contribution to  $w_{6p_{3/2}}$ .

Since the reduced electron matrix elements in Eq. (3.4) remain essentially constant over the energy range under consideration here, and since they are naturally independent of the distance between atoms, according to Table II, the spectra for  $w_{6p_{1/2}}$  in Figs. 2–4 correspond completely to the behavior of the density of the  $d_{3/2}$  wave of a conversion electron near an isomer atom. The spectra for  $w_{6p_{3/2}}$ , on the other hand, contain a mixture of  $d_{3/2}$  and  $d_{5/2}$  waves.

The resultant spectrum, 3, in Figs. 2–4 was found by adding spectra 1 and 2 with appropriate weights. Because of the oscillatory structure of the spectra for  $w_{6p_{1/2}}$  and  $w_{6p_{3/2}}$ , the resultant spectrum for  $w_{6p}$  could in principle be anything between two extreme cases. First, the maxima of one spectrum might fall on the maxima of the other (and the minima on the minima), and we would see a maximum manifestation of the interference. Second, the maxima of one spectrum might fall on minima of the other, and the interference would be at its weakest. The result might be either a slowing or an acceleration of the conversion decay of  $^{235m}\text{U}$  in silver in comparison with the isolated isomer. In our model, we have an “intermediate” situation, in which the minima of one spectrum basically fall on maxima of the other, so the changes cancel out to some extent. As a result, the change in the total conversion probability due to the interference is much weaker than for transitions from the individual shells of U. It follows from our calculations that interference ef-

fects should be seen particularly clearly in the spectra of the conversion electrons. As can be seen from Figs. 2–4, the intensities of the transitions from the  $6p_{1/2}$  and  $6p_{3/2}$  shells of uranium would be greatly different from those for the isolated isomer, depending on the number of nearest atoms around the isomer, the arrangement of these atoms, and the distances between them.

The crudeness of our theoretical model, on the one hand, and the uncertainty on the experimental side, on the other, rule out any accurate predictions of the conversion probabilities. Nevertheless, our model can be used for a qualitative study of how an interference of conversion electrons affects the conversion probability, and we can estimate the scale of this effect. Working from the results in Figs. 2–4, we can find the maximum changes in the conversion probability for a  $\text{UAg}_{12}$  cluster as the corresponding parameters are varied. We find the magnitude of the changes for a transition from shell  $n$  from the ratio  $\Delta_n = (w_n - w_n^{(0)})/w_n^{(0)}$ , where  $w_n^{(0)}$  corresponds to the isolated atom. We then find

$$\Delta_{6p_{1/2}} = \begin{pmatrix} +89 \\ -41 \end{pmatrix} \%, \quad \Delta_{6p_{3/2}} = \begin{pmatrix} +60 \\ -44 \end{pmatrix} \%, \quad \Delta_{6p} = \begin{pmatrix} +47 \\ -21 \end{pmatrix} \%.$$

In the experiments<sup>6</sup> on indirect measurements of the decay rate of  $^{235m}\text{U}$  deep in the interior of a silver layer, which we mentioned back in the introduction, the activity of a portion of material prepared by a definite procedure was studied. Kol'tsov and Rimskii-Korsakov<sup>6</sup> interpreted their data as indicating a suppression of the  $^{235m}\text{U}$  isomer transition under these conditions, but they did not explain the nature of this suppression.

The results of the present study suggest that a situation such that the  $d$ -wave density of conversion electrons is greatly reduced near the isomer because of an interference of these electrons at the silver atoms around the isomer might prevail in the uranium-in-silver system. As we mentioned above, a decrease in the  $d$ -wave density of conversion electrons clearly leads to a decrease in the conversion probability.

Experimental conversion spectra of the  $^{235m}\text{U}$  isomer in silver could apparently shed some light on the situation. We wish to stress that the calculated spectra of conversion electrons should be averaged over the various configurations of the surroundings of U in the silver for an appropriate comparison with the experimental data.

Significant effects of an interference of electrons in the course of a conversion decay of the  $^{235m}\text{U}$  isomer, similar to the effects which we have been discussing here for the uranium-in-silver system, could be expected when  $^{235m}\text{U}$  is inserted in the lattices of other elements (not necessarily metals). As a criterion for selecting these elements one might use the requirement that the scattering of the conversion electrons by the atoms around the isomer lead to a significant change in the density of their  $d$  waves with respect to the isomer.

## 6. CONCLUSION

The basic conclusion which follows from the results of the present study is that the interference (rescattering) of conversion electrons with the atoms around the isomer must be taken into account in order to find a satisfactory description of the soft ( $\hbar\omega \approx 76.8$  eV)  $E3$  conversion transition in the  $^{235m}\text{U}$  isomer in a silver lattice. Interference effects are amplified as the number of atoms in the first coordination

sphere increases. The interference effects turn out to be at their greatest in the case of the dense packing corresponding to the geometry of an fcc lattice. The probability for conversion in the individual shells of the isomer atom and also the total probability for the conversion transition are exceedingly sensitive to geometric factors—the number and arrangement of the atoms of the medium around the isomer and the distances between these atoms—and also to the binding energies of the atomic orbitals in which the conversion occurs. As a result, the rate of the conversion decay of the isomer may either decrease or increase. The scale of the changes in the resultant conversion probability in the  $6p$  shells of U turns out to be on the order of ( $\pm 47\%$ ) in comparison with the isolated uranium isomer according to our model. The magnitude of the interference effect (for transitions from atomic  $p$  shells) turns out to depend directly on the  $d$ -wave density of conversion electrons near the isomer atom.

We have examined only the simplest, high-symmetry clusters, with a single coordination sphere of silver atoms. The actual surroundings of a U atom in silver may instead turn out to be extremely arbitrary, with a much lower symmetry. On the other hand, since multiple scattering (Sec. 4) is important in this system, it is generally necessary to consider the second, third, etc., coordination spheres. Because of all these factors, there might be significant changes in the quantitative conclusions. Nevertheless, the qualitative results of the present paper would remain the same, in our opinion.

I am particularly indebted to D. P. Grechukhin for constant support, stimulating discussions, and critical comments and to M. M. Vsevolodov for interest in this study and critical comments. I also thank A. A. Soldatov, A. D. Panov, A. V. Lomonosov, and D. V. Grebennikov for useful discussions.

Finally, I thank G. D. Samolyuk for furnishing the program for calculating the self-consistent potential in the model of an isolated impurity.

## APPENDIX

### 1. Spherical bispinors

We assume that the index  $Z$  takes on one of the values  $J$ ,  $N$ ,  $H^{(1)}$ ,  $H^{(2)}$ , corresponding to the spherical Bessel functions  $z_l = j_l$ ,  $n_l$ ,  $h_l^{(1)}$ ,  $h_l^{(2)}$ . The spherical bispinor  $\psi_{klm}^{(Z)}(\mathbf{r})$  for the functions  $z_l$  and the energy  $\varepsilon = [(\hbar kc)^2 + (mc^2)^2]^{1/2}$  is then given by

$$\psi_{klm}^{(Z)}(\mathbf{r}) = \frac{2k}{(2\varepsilon)^{1/2}} \begin{pmatrix} (\varepsilon + mc^2)^{1/2} z_l(kr) \Omega_{jlm}(\mathbf{n}_r) \\ -(\varepsilon - mc^2)^{1/2} z_{l'}(kr) \Omega_{j'l'm}(\mathbf{n}_r) \end{pmatrix}, \quad \mathbf{n}_r = \frac{\mathbf{r}}{r}, \quad (\text{A1})$$

where  $l' = 2j - l$ . For  $z_l = j_l$ , the wave function  $\psi_{klm}^{(J)}$  describes a free relativistic electron with an energy  $\varepsilon$  in a state with definite values of the angular momentum  $j$ , the angular-momentum projection  $m$ , and the parity, specified by  $l$  (Ref. 10). These states are orthonormalized by

$$\int d^3r \psi_{k_1 j_1 l_1 m_1}^{(J)+}(\mathbf{r}) \psi_{k_2 j_2 l_2 m_2}^{(J)}(\mathbf{r}) = 2\pi \delta(k_1 - k_2) \delta_{j_1 l_1 m_1, j_2 l_2 m_2}.$$

### 2. Reduced matrix elements

The reduced matrix elements are given by the Wigner-Eckart theorem

$$\begin{aligned} & \int d\mathbf{O}_n \Omega_{j_1 l_1 m_1}^+(\mathbf{n}) Y_{LM}(\mathbf{n}) \Omega_{j_2 l_2 m_2}(\mathbf{n}) \\ &= (-1)^{j_1 - m_1} \begin{pmatrix} j_1 & L & j_2 \\ -m_1 & M & m_2 \end{pmatrix} \langle j_1 l_1 \| L \| j_2 l_2 \rangle, \\ \langle j_1 l_1 \| L \| j_2 l_2 \rangle &= (-1)^{L+j_2+1/2} i^{-l_1+L+l_2} \\ & \times \left[ \frac{(2j_1+1)(2l_1+1)(2L+1)(2j_2+1)(2l_2+1)}{4\pi} \right]^{1/2} \\ & \times \begin{pmatrix} l_1 & L & l_2 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} j_1 & L & j_2 \\ l_2 & 1/2 & l_1 \end{Bmatrix}. \end{aligned} \quad (\text{A2a})$$

$$\begin{aligned} & \int d\mathbf{O}_n \Omega_{j_1 l_1 m_1}^+(\mathbf{n}) (Y_{JLM}(\mathbf{n}) \sigma) \Omega_{j_2 l_2 m_2}(\mathbf{n}) \\ &= i (-1)^{j_1 - m_1} \begin{pmatrix} j_1 & J & j_2 \\ -m_1 & M & m_2 \end{pmatrix} \langle j_1 l_1 \| JL \| j_2 l_2 \rangle, \\ \langle j_1 l_1 \| JL \| j_2 l_2 \rangle &= (-1)^{l_1+j_2+1/2} i^{-l_1+L+l_2} \\ & \times \left[ \frac{6(2j_1+1)(2l_1+1)(2J+1)(2L+1)(2j_2+1)(2l_2+1)}{4\pi} \right]^{1/2} \\ & \times \begin{pmatrix} l_1 & L & l_2 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} J & L & 1 \\ j_1 & l_1 & 1/2 \\ j_2 & l_2 & 1/2 \end{Bmatrix}. \end{aligned} \quad (\text{A2b})$$

Definitions of the spherical function  $Y_{LM}$ , the spherical spinor  $\Omega_{jlm}$ , and the spherical vector  $Y_{JLM}$  are given in Ref. 10.

### 3. Structure matrices

We introduce the definitions

$$\begin{aligned} D_{\alpha_1 j_1 l_1 m_1, \alpha_2 j_2 l_2 m_2}^{(J)} &= D_{j_1 l_1 m_1, j_2 l_2 m_2}^{(J)}(\mathbf{R}_{\alpha_1 \alpha_2}) \\ &= 4\pi \sum_{LM} i^{l_1+L-l_2} j_L(kR_{\alpha_1 \alpha_2}) Y_{LM}^*(\mathbf{n}_{\alpha_1 \alpha_2}) \\ & \times (-1)^{j_1 - m_1} \begin{pmatrix} j_1 & L & j_2 \\ -m_1 & M & m_2 \end{pmatrix} \langle j_1 l_1 \| L \| j_2 l_2 \rangle, \end{aligned} \quad (\text{A3a})$$

$$\begin{aligned} D_{\alpha_1 j_1 l_1 m_1, \alpha_2 j_2 l_2 m_2}^{(N)} &= D_{j_1 l_1 m_1, j_2 l_2 m_2}^{(N)}(\mathbf{R}_{\alpha_1 \alpha_2}) \\ &= (1 - \delta_{\alpha_1 \alpha_2}) 4\pi \sum_{LM} i^{l_1+L-l_2} h_L(kR_{\alpha_1 \alpha_2}) Y_{LM}^*(\mathbf{n}_{\alpha_1 \alpha_2}) \\ & \times (-1)^{j_1 - m_1} \begin{pmatrix} j_1 & L & j_2 \\ -m_1 & M & m_2 \end{pmatrix} \langle j_1 l_1 \| L \| j_2 l_2 \rangle, \end{aligned} \quad (\text{A3b})$$

$$\mathbf{D}^{(H^{(1)})} = \mathbf{D}^{(J)} + i \mathbf{D}^{(N)}, \quad (\text{A3c})$$

$$\mathbf{D}^{(H^{(2)})} = \mathbf{D}^{(J)} - i \mathbf{D}^{(N)}. \quad (\text{A3d})$$

Here  $\mathbf{R}_{\alpha_1 \alpha_2} = \mathbf{R}_{\alpha_1} - \mathbf{R}_{\alpha_2}$  and  $\mathbf{n}_{\alpha_1 \alpha_2} = \mathbf{R}_{\alpha_1 \alpha_2} / |\mathbf{R}_{\alpha_1 \alpha_2}|$ .

We note one property of the structure matrices in the case  $\alpha_1 = \alpha_2 = \alpha$ :

$$\begin{aligned} D_{\alpha_1 j_1 l_1 m_1, \alpha_2 j_2 l_2 m_2}^{(J)} &= D_{\alpha_1 j_1 l_1 m_1, \alpha_2 j_2 l_2 m_2}^{(H^{(1)})} = D_{\alpha_1 j_1 l_1 m_1, \alpha_2 j_2 l_2 m_2}^{(H^{(2)})} = \delta_{j_1 l_1 m_1, j_2 l_2 m_2}, \\ D_{\alpha_1 j_1 l_1 m_1, \alpha_2 j_2 l_2 m_2}^{(N)} &= 0 \end{aligned}$$

for any  $\alpha$ th muffin-tin sphere.

#### 4. Theorem for combining spherical bispinors

The theorem for combining spherical harmonics is well known (Refs. 8, for example). A corresponding theorem can be found for the spherical bispinors in Eq. (A1) (see also Ref. 9):

$$\begin{aligned} \Psi_{hj_1l_1m_1}^{(Z)}(\mathbf{r}_1 + \mathbf{r}_2) &= \sum_{j_2l_2m_2} D_{j_2l_2m_2, j_1l_1m_1}^{(J)}(\mathbf{r}_<) \Psi_{hj_2l_2m_2}^{(Z)}(\mathbf{r}_>) \\ &= \sum_{j_2l_2m_2} D_{j_2l_2m_2, j_1l_1m_1}^{(Z)}(\mathbf{r}_>) \Psi_{hj_2l_2m_2}^{(J)}(\mathbf{r}_<). \end{aligned} \quad (\text{A4a})$$

The expansion matrices are the same as structure matrices Eq. (A3).

We can use this theorem to decompose spherical bispinors specified with respect to different centers in terms of each other:

$$\Psi_{hj_1l_1m_1}^{(Z)}(\mathbf{r}_{\alpha_1}) = \sum_{j_2l_2m_2} D_{\alpha_2 j_2 l_2 m_2, \alpha_1 j_1 l_1 m_1}^{(Z)} \Psi_{hj_2 l_2 m_2}^{(J)}(\mathbf{r}_{\alpha_2}), \quad (\text{A4b})$$

where  $\mathbf{r}_{\alpha_1} = \mathbf{r} - \mathbf{R}_{\alpha_1} = \mathbf{r}_{\alpha_2} + \mathbf{R}_{\alpha_2} - \mathbf{R}_{\alpha_1}$ , and  $\mathbf{r}_{\alpha_2} = \mathbf{r} - \mathbf{R}_{\alpha_2}$ , with  $|\mathbf{r}_{\alpha_2}| < |\mathbf{R}_{\alpha_2} - \mathbf{R}_{\alpha_1}|$ .

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