COMMENT

Comment on 'Calculations of electron angular distribution in resonant Auger decay for Na, Ba, Hg and Kr*'

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Abstract. We make a comment on the discrepancy between the numerical results for the angular anisotropy parameter α_2 for the $L_3M_1M_{4,5}$ Auger transitions of Kr, Xe, Ba, and Hg which have been obtained by Elizarov and Tupitsyn (2004 *Phys. Scr.* **70** 139) and beforehand by ourselves (Kleiman and Lohmann 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 2653). By comparing the results obtained not only for the angular anisotropy parameter α_2 but also for the dynamic spin polarization parameter ξ_2, where the latter agree considerably better, it is most likely that the discrepancies are mainly due to some of the phase differences because the parameter α_2 depends on the cosine of the phase differences whereas the parameter ξ_2 depends on the sine.

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The study of angular distribution and spin polarization of Auger electrons, which in the present comment are assumed to be ejected after creating a vacancy in an inner atomic shell by photoionization (dipole approximation), nowadays is a major part of Auger electron spectroscopy because of the complementary information it can provide on the inner atomic structure and the dynamics of the underlying process as compared with conventional investigations of Auger electron energies and intensities at a fixed angle. It is well known that the prerequisite for anisotropic angular distribution and dynamic spin polarization of Auger electrons is a photoionized atom showing an alignment [1, 2, 3]; the transferred spin polarization will be disregarded here. Applying the conventional two-step description of the Auger process [4] there are in all three quantities of interest that is the alignment parameter $\mathcal{A}_{20}$ of the primary photoionization plus the angular anisotropy parameter α_2 and the dynamic spin polarization parameter ξ_2 of the secondary Auger decay. Expressions for the angular distribution and the dynamic spin polarization of Auger electrons featuring the above parameters can be checked in the literature (e.g. [5, 6]).

About ten years ago we came across large dynamic spin polarization parameters ξ_2 for three of the four lines of the Kr $L_3M_1M_{4,5}$ Auger spectrum which has been included in the calculation of the Kr $L_3M_{4,5}M_{4,5}$ Auger spectrum [7]. To ensure that our finding was not pure chance we extended our investigation to cover all $L_3M_1M_{4,5}$ Auger spectra of closed-shell or closed-subshell atoms and found the same distinctive feature [5]. The calculations have been performed by describing the Auger emission process in the context of scattering theory (relativistic distorted wave approximation) where the Auger transition amplitudes and scattering phases have been evaluated by applying a relaxed orbital method within a multiconfiguration Dirac–Fock approach. For further reading we recommend [8, 9].

Some years later Elizarov and Tupitsyn have re-calculated the parameters α_2 and ξ_2‡ for the $L_3M_1M_{4,5}$ Auger spectra of Kr, Xe, Ba [10, 11] and Hg [10, 11, 12] employing the multiconfiguration Dirac–Fock method. Note that the chosen title of the paper [10] is misleading since the Kr, Xe, Ba and Hg Auger transitions are not resonant but normal Auger transitions. Elizarov and Tupitsyn have compared their results for the angular anisotropy parameter α_2 with ours and have come across significant discrepancies. Regardless of this finding they have published their results in different journals without elaborating on possible reasons for the discrepancies beforehand. In addition, we cannot see any reason why Elizarov and Tupitsyn have not compared their results for the dynamic spin polarization parameter ξ_2 with ours. Here, the agreement is considerably better which, in our opinion, sheds a light on what may cause the discrepancies between the two data sets obtained for the angular anisotropy parameter α_2.

Regarding the practical but missing comparison and due to a few misprints in the tables [10, 11, 12], due to an Auger spectrum of Hg consisting of five lines instead of four [10, 11, 12] and due to two columns, excluding the column headings, which have been mixed up in table I of [10]§, we have re-arranged the parameters α_2 and ξ_2 together with the Auger energies, which have been calculated by ourselves and by Elizarov and Tupitsyn, in table 1. The latter

‡ The dynamic spin polarization parameter ξ_2 was referred to as β_2 in the early publications, e.g. [8].

§ A corrected table I has been published at our special request [13].

Table 1. Comparison of Auger energies, angular anisotropy (α_2) and spin polarization (ξ_2) parameters for the $L_3M_1M_{4,5}$ Auger spectra of Kr, Xe, Ba and Hg.

	Final	Auger energy (eV)			α_2			ξ_2		
	state	a	b	c	a	b	c	a	b	c
Kr	$^1D_2$	1259.48	1274.95	1259.49	0.218	−0.081	−0.026	0.039	0.034	0.033
	$^3D_1$	1266.93	1274.19	1266.95	−0.034	−0.337	−0.279	−0.229	−0.147	−0.156
	$^3D_2$	1267.53	1273.60	1267.54	0.278	0.191	0.243	−0.188	−0.153	−0.160
	$^3D_3$	1268.29	1266.14	1268.30	0.331	0.612	0.570	0.128	0.147	0.141
Xe	$^1D_2$	2900.95	2934.63	2900.88	0.228	−0.234	−0.191	0.127	0.077	0.080
	$^3D_1$	2909.82	2929.64	2909.74	0.101	−0.422	−0.391	−0.289	−0.139	−0.147
	$^3D_2$	2918.09	2921.36	2918.02	0.342	0.584	0.638	−0.210	−0.193	−0.201
	$^3D_3$	2923.09	2912.49	2923.02	0.161	0.606	0.580	0.114	0.147	0.144
Ba	$^1D_2$	3099.05	3110.73	3098.95	0.235	−0.211	−0.211	0.133	0.076	0.076
	$^3D_1$	3107.95	3119.62	3107.85	0.147	−0.380	−0.380	−0.299	−0.152	−0.152
	$^3D_2$	3118.35	3130.03	3118.26	0.328	0.716	0.716	−0.193	−0.186	−0.186
	$^3D_3$	3123.95	3135.64	3123.87	0.134	0.553	0.553	0.109	0.140	0.140
Hg	$^1D_2$	6265.45	6392.36	6263.43	0.344	0.025	0.393	0.112	0.073	0.073
	$^3D_1$	6276.02	6379.84	6274.00	0.801	−0.080	0.034	−0.230	−0.240	−0.250
	$^3D_2$	6356.95	6298.89	6354.96	0.068	0.406	0.402	0.003	0.015	0.017
	$^3D_3$	6369.46	6288.32	6367.47	−0.162	0.133	0.112	0.020	0.057	0.053

[a] Theory: Our work [5].

[b] Theory: Elizarov and Tupitsyn [10] (calculation in the frozen-core approximation).

[c] Theory: Elizarov and Tupitsyn [10] (calculation with orbital relaxation).

have carried out one calculation in the frozen-core approximation and another calculation which accounts for orbital relaxation in the course of the Auger decay, see [10, 11, 12] for formulas and further details.

It is clear from table 1 that, for Kr, Xe and Hg, the frozen-core approximation results in an inverted energy sequence of the four Auger lines. Here, the $^1D_2$ line is positioned at the highest energy whereas the $^3D_3$ line is energetically well separated from the others. This is highly questionable especially for the medium weight Kr atom for which the Auger lines may be taken as almost true singlet and triplet lines regarding LS coupling. Thus, the $^1D_2$ line should be positioned at the lowest energy and should be energetically well separated from the $^3D_{1,2,3}$ lines which are expected to be positioned close to each other. In view of this we do not comment in detail on the results for the parameters α_2 and ξ_2 which have been obtained by Elizarov and Tupitsyn employing the frozen-core approximation. Just this, we would expect the frozen-core approximation to work well for normal Auger transitions in which huge Auger energies are involved as the outgoing Auger electron is such fast that it does not feel the potential changing. This is what Elizarov and Tupitsyn [10, 11] believe they have demonstrated for the angular anisotropy parameter α_2 though their results do not really support their opinion, see particularly the $^1D_2$ and $^3D_1$ lines of the $L_3M_1M_{4,5}$ Auger spectrum of Hg. In [12] Elizarov and Tupitsyn argue the converse. There, they have further considered the $N_3O_1O_{4,5}$ Auger transitions of Hg for which the Auger energies are of about 430 eV [6];

unfortunately they did not give the Auger energies emerging from their calculations. Their results for the parameter α_2 obtained in the frozen-core approximation and by accounting for orbital relaxation during the Auger decay are in agreement. We would like to add, that for three of the four $N_3O_1O_{4,5}$ Auger lines of Hg their angular anisotropy parameters agree quite well with ours [6], the dynamic spin polarization parameter ξ_2 has not been tabulated by Elizarov and Tupitsyn [12]. Contrary to what is said in [12] we have chosen intermediate coupling for our numerical calculation [9].

It is further not clear why for the parameters α_2 and ξ_2 of the $L_3M_1M_{4,5}$ Auger spectrum of Ba the two different calculations carried out by Elizarov and Tupitsyn [10] yield the same values, at least for up to the three digits given, although the Auger energies obtained in the frozen-core approximation and by accounting for orbital relaxation differ from each other. It is unfortunate that Elizarov and Tupitsyn do not explain how they have calculated the Auger energies.

For the remainder of the comment we will leave any data obtained in the frozen-core approximation out and focus on Elizarov and Tupitsyn's realization that there is a huge discrepancy between their [10, 11] and our [5] results for the angular anisotropy parameter α_2 for the $L_3M_1M_{4,5}$ Auger transitions of Kr, Xe, Ba, and Hg, see columns a and c of table 1. Again, these two data sets have been obtained by accounting for orbital relaxation during the Auger decay. Elizarov and Tupitsyn did not investigate this discrepancy any further [10], they only suggested that the discrepancy may have something to do with the phase differences which are 'not the same as those presented in table 2' of our work [5]. They neither give any values for their phase differences nor do they give any information on how much their phase differences deviate from ours. There are, however, reasons for the assumption that Elizarov and Tupitsyn have obtained a phase difference which is close to an odd integer multiple of $\frac{\pi}{2}$ because, as has been discussed in full in [5], such a phase difference can be seen as one of the prerequisites for a large dynamic spin polarization parameter ξ_2. It is clear from table 1 that their calculation supports our previous finding that the normal $L_3M_1M_{4,5}$ Auger transitions of various elements do have comparatively large dynamic spin polarization parameters ξ_2 [5]. The agreement between their and our results is in general much better as compared to the angular anisotropy parameter α_2.

At this point it is necessary to investigate the formulas for both α_2 and ξ_2 in more detail. According to [8, 9] the angular anisotropy parameter may be written as

$$\begin{aligned}\alpha_2 = -\sqrt{5(2J+1)} \sum_{l,j,l',j'} (-1)^{J+J_\mathrm{f}+\frac{1}{2}+l}\, \mathrm{i}^{l+l'} \sqrt{(2j+1)(2j'+1)}\, \cos(\sigma_l^j - \sigma_{l'}^{j'}) \\ \times \begin{pmatrix} j & j' & 2 \\ -\frac{1}{2} & \frac{1}{2} & 0 \end{pmatrix} \begin{Bmatrix} j & j' & 2 \\ J & J & J_\mathrm{f} \end{Bmatrix} \langle J_\mathrm{f} || \,\mathrm{V}\, || J\,(l\,\tfrac{1}{2})\,j : J_\mathrm{f}\rangle \,\langle J_\mathrm{f} || \,\mathrm{V}\, || J\,(l'\,\tfrac{1}{2})\,j' : J_\mathrm{f}\rangle \\ \times \left[\sum_{l,j} \langle J_\mathrm{f} || \,\mathrm{V}\, || J\,(l\,\tfrac{1}{2})\,j : J_\mathrm{f}\rangle^2 \right]^{-1} \end{aligned} \tag{1}$$

whereas the dynamic spin polarization parameter is as follows:

Table 2. The phase differences $\sigma_l^j - \sigma_{l'}^{j'}$ of the interfering partial waves necessary to calculate the parameters α_2 and ξ_2 for the $\mathrm{L_3M_1M_{4,5}}$ Auger transitions of Kr and Xe. Zeros have been included in order to demonstrate the maximum number of contributing phase differences.

Phase difference	Final states (Kr)				Final states (Xe)			
	$^1\mathrm{D}_2$	$^3\mathrm{D}_1$	$^3\mathrm{D}_2$	$^3\mathrm{D}_3$	$^1\mathrm{D}_2$	$^3\mathrm{D}_1$	$^3\mathrm{D}_2$	$^3\mathrm{D}_3$
$\sigma_1^{1/2} - \sigma_1^{3/2}$	0.053	0.053	0.053	–	0.114	0.114	0.114	–
$\sigma_1^{3/2} - \sigma_1^{3/2}$	0	0	0	0	0	0	0	0
$\sigma_1^{1/2} - \sigma_3^{5/2}$	2.343	2.338	2.337	–	2.270	2.268	2.266	–
$\sigma_1^{3/2} - \sigma_3^{5/2}$	2.290	2.285	2.284	2.284	2.157	2.154	2.152	2.151
$\sigma_1^{3/2} - \sigma_3^{7/2}$	2.294	–	2.288	2.288	2.169	–	2.164	2.163
$\sigma_3^{5/2} - \sigma_3^{5/2}$	0	0	0	0	0	0	0	0
$\sigma_3^{5/2} - \sigma_3^{7/2}$	0.004	–	0.004	0.004	0.012	–	0.012	0.012
$\sigma_3^{7/2} - \sigma_3^{7/2}$	0	–	0	0	0	–	0	0

$$\xi_2 = \sqrt{5(2J+1)/6} \sum_{l,j,l',j'} (-1)^{j+J+J_\mathrm{f}}\, \mathrm{i}^{l+l'} \sqrt{(2j+1)(2j'+1)}\, \sin(\sigma_l^j - \sigma_{l'}^{j'}) \times \begin{pmatrix} j & j' & 2 \\ \frac{1}{2} & \frac{1}{2} & -1 \end{pmatrix} \begin{Bmatrix} j & j' & 2 \\ J & J & J_\mathrm{f} \end{Bmatrix} \langle J_\mathrm{f} || \,\mathrm{V}\, || J\,(l\,\tfrac{1}{2})\,j : J_\mathrm{f}\rangle \,\langle J_\mathrm{f} || \,\mathrm{V}\, || J\,(l'\,\tfrac{1}{2})\,j' : J_\mathrm{f}\rangle \times \left[\sum_{l,j} \langle J_\mathrm{f} || \,\mathrm{V}\, || J\,(l\,\tfrac{1}{2})\,j : J_\mathrm{f}\rangle^2 \right]^{-1}. \quad (2)$$

Here, J and J_f denote the total angular momenta of the intermediate state and the final ionic state, respectively. l and j are the orbital and total angular momenta of the Auger electron obtained by expanding the Auger electron momentum into partial waves, that is the triangle condition $\Delta(l\,\frac{1}{2}\,j)$ applies. The accompanying scattering phases are denoted by σ_l^j. V stands for the Coulomb operator and the brackets $(\ldots)$ and $\{\ldots\}$ are the Wigner 3–j and 6–j symbols [14]. In equations (1) and (2) the sum over the two pairs of quantum numbers l, j and l', j' is limited by the triangle conditions of the 3–j and 6–j symbols as well as by parity. The parity of the intermediate state and the final ionic state is odd and even, respectively. As the Coulomb operator conserves parity, the parity of the Auger electron and thus l and l' must be odd (for further explanation see [5]). Moreover, the numerator of equations (1) and (2) is invariant to the interchange of the two pairs of quantum numbers l, j and l', j' which can be used to simplify the analysis of α_2 and ξ_2. For instance, the number of non-vanishing terms appearing in the numerator of the parameter ξ_2 can be halved.

The most important difference between the parameters α_2 and ξ_2 is their dependence on the phase difference of two interfering partial waves. The angular anisotropy parameter α_2 depends on the cosine of the phase difference whereas ξ_2 depends on the sine of the phase difference. In tables 2 and 3 we show all those phase differences which are essential for the calculation of α_2 and ξ_2. The numerical values have been taken from our original work [5]

Table 3. The phase differences $\sigma_l^j - \sigma_{l'}^{j'}$ of the interfering partial waves necessary to calculate the parameters α_2 and ξ_2 for the $L_3M_1M_{4,5}$ Auger transitions of Ba and Hg. Zeros have been included in order to demonstrate the maximum number of contributing phase differences.

Phase difference	Final states (Ba)				Final states (Hg)			
	$^1D_2$	$^3D_1$	$^3D_2$	$^3D_3$	$^1D_2$	$^3D_1$	$^3D_2$	$^3D_3$
$\sigma_1^{1/2} - \sigma_1^{3/2}$	0.123	0.123	0.123	–	0.223	0.223	0.224	–
$\sigma_1^{3/2} - \sigma_1^{3/2}$	0	0	0	0	0	0	0	0
$\sigma_1^{1/2} - \sigma_3^{5/2}$	−0.876	−0.879	−0.881	–	−0.977	−0.979	−0.990	–
$\sigma_1^{3/2} - \sigma_3^{5/2}$	−0.999	−1.001	−1.004	−1.005	−1.201	−1.202	−1.214	−1.216
$\sigma_1^{3/2} - \sigma_3^{7/2}$	−0.986	–	−0.991	−0.992	−1.171	–	−1.184	−1.186
$\sigma_3^{5/2} - \sigma_3^{5/2}$	0	0	0	0	0	0	0	0
$\sigma_3^{5/2} - \sigma_3^{7/2}$	0.013	–	0.013	0.013	0.030	–	0.030	0.030
$\sigma_3^{7/2} - \sigma_3^{7/2}$	0	–	0	0	0	–	0	0

and refer to the $L_3M_1M_{4,5}$ Auger transitions of Kr, Xe, Ba and Hg. Though the $\varepsilon h_{9/2}$ partial wave occurs in principle in equations (1) and (2) provided the Auger transition is to the final state $^3D_3$ it is not necessary to consider this partial wave because the accompanying reduced Coulomb matrix element turns out to be zero in consequence of further selection rules arising from the angular part of the direct and exchange terms [8, 9]. Tables 2 and 3 show that the phase differences between the partial waves with different orbital angular momenta l and l' are large whereas the phase differences between the partial waves with the same l are, without exception, considerably smaller. The latter can in general be expected for light and medium weigth atoms for which the spin-orbit interaction is weak. That is why the phase differences $\sigma_1^{1/2} - \sigma_1^{3/2}$ and $\sigma_3^{5/2} - \sigma_3^{7/2}$ increase when going from Kr via Xe and Ba to Hg. By the way, for Kr and Xe all the phase differences are positive and considerably larger in magnitude than those for Ba and Hg where we find negative phase differences for interfering partial waves with different orbital angular momenta.

To preclude misconceptions we remind the reader that the scattering phase σ_l^j is the sum of the Coulomb phase σ_l^{C}, which for non-relativistic as well as relativistic calculations depends only on the orbital angular momentum l of the partial waves of the Auger electron, and the asymptotic phase shift or scattering phase shift δ_l^j. When calculated relativistically, such as in our work, the latter depends on both l and j. We thus have

$$\sigma_l^j = \sigma_l^{\mathrm{C}} + \delta_l^j. \tag{3}$$

For the $L_3M_1M_{4,5}$ Auger transitions of Kr, Xe, Ba and Hg we find our Coulomb phases to be considerably smaller in magnitude than the scattering phase shifts.

To address the still open question why the numerical results obtained by Elizarov and Tupitsyn and by ourselves are such different only for the angular anisotropy parameter α_2 but not for the dynamic spin polarization parameter ξ_2, we choose the final state $^3D_3$ of the Xe $L_3M_1M_{4,5}$ Auger transitions for illustration and simplicity. It follows from table 2 that the

magnitude of ξ_2 is determined by those two terms for which the phase differences are large, that is 2.151 and 2.163‖. As the parameter ξ_2 depends on the sine of the phase differences and the sine function is symmetric about $\frac{\pi}{2}$, not only the phase differences 2.151 and 2.163, which we have obtained, but, for instance, also the phase differences 0.991 and 0.979 would yield the same value for the parameter ξ_2. We thus believe that Elizarov and Tupitsyn may have obtained phase differences such as these. They are not the same as ours [10] but result in similar values for the dynamic spin polarization parameter ξ_2. However, the parameter α_2 depends on the cosine of the phase differences and thus, using the phase differences 0.991 and 0.979 instead of 2.151 and 2.163 would cause the respective two terms to change sign. As the magnitude of the cosine of these phase differences is about 0.55, all in all six terms contribute to the numerator of equation (1). Supposing that two of the six terms are of opposite sign in Elizarov and Tupitsyn's and in our calculations this may explain why the results for the parameter α_2 (considerably) differ from each other.

In connection with the phase differences there is another aspect which, in our opinion, deserves attention. Though the multiconfiguration Dirac–Fock method has been employed by Elizarov and Tupitsyn to obtain the reduced Coulomb matrix elements as well as the continuum wavefunction [12], strangely enough the scattering phases and thus the phase differences, which enter the formulas for α_2 and ξ_2, depend only on the orbital angular momentum l but are independent of the total angular momentum j. That is they have replaced the phase differences in equations (1) and (2) simply by $\sigma_l - \sigma_{l'}$ and have not taken into accout the spin-orbit interaction in the continuum. This is undoubtedly an inconsistent alteration of the formulas originally published in [8] and later used by Elizarov and Tupitsyn for their calculations [10, 11]. By just using the phase differences $\sigma_1 - \sigma_1$, $\sigma_1 - \sigma_3$ and $\sigma_3 - \sigma_3$, of which the first and the third are equal to zero, the weighting of the terms in the numerator of equations (1) and (2) is changed. Inspecting our phase differences displayed in tables 2 and 3 it is clear that the method applied by Elizarov and Tupitsyn is, in many cases, less accurate than our calculational method. For instance, the parameter ξ_2 for the Auger transitions to the final states $^3\mathrm{D}_1$ and $^3\mathrm{D}_3$ depends on two terms in Elizarov and Tupitsyn's calculation instead of originally three terms in our calculation. In particular, considering the $^3\mathrm{D}_1$ lines of Xe, Ba and Hg, the phase difference $\sigma_1^{1/2} - \sigma_1^{3/2}$ is far from being zero. Here, the agreement among the two data sets for ξ_2 is less good. However, for the $^3\mathrm{D}_3$ lines the phase difference $\sigma_3^{5/2} - \sigma_3^{7/2}$ is quite close to zero and the agreement is much better.

In conclusion, despite the missing of almost any discussion in the work by Elizarov and Tupitsyn [10], we have tried to find out what may be the reasons for the huge discrepancies between the results for the angular anisotropy parameter α_2 obtained by Elizarov and Tupitsyn and earlier by ourselves. As Elizarov and Tupitsyn have also obtained large values for the parameter ξ_2, which, in addition, agree quite well with our results, we are more or less certain that their phase differences are close to an odd interger multiple of $\frac{\pi}{2}$, too. As discussed in [5], such phase differences are necessary (but not sufficient) for getting large dynamic spin

‖ Phase diffences of about $\frac{\pi}{2}$ are necessary but not sufficient for obtaining a large ξ_2 because one has also to take into account the reduced Coulomb matrix elements as well as the 3–j and 6–j symbols. The different terms in the numerator of equation (2) may also cancel each other, see [5] for details.

polarization parameters. When these phase differences enter the argument of a sine function, which is the case for ξ_2, it does not matter whether the phase differences are somewhat smaller or, to the same extent, larger than $\frac{\pi}{2}$. This is different for α_2 because this parameter depends on the cosine of the phase differences of two interfering partial waves. Here some of the terms in the numerator of equation (1) would change sign. Furthermore, though Elizarov and Tupitsyn perform a relativistic calculation, the incomprehensible independence of their scattering phases from the total angular momentum j in a way contributes to the discrepancies found.

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