Spin-resolved measurements of the $^{12}$Ar$^{+}$ resonantly excited $L_{2,3}M_{2,3}$ Auger decay have been performed. The low resolution Auger spectrum, which due to cancellation between different multiplet components should exhibit virtually zero dynamic spin polarization, reveals an unexpected nonvanishing polarization effect. Calculations within a relativistic distorted wave approximation explain this effect as configuration-interaction (CI) induced. The CI generates experimentally unresolved fine structure components with low and high total angular momentum, giving rise to asymmetric cases where the high $J/\ell$ part of certain multiplets is suppressed by internal selection rules for diagram lines. In this case, only the low $J$ components survive with no partner for spin-polarization cancellation.

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meters containing solely the dynamics of the primary excitation, and angular distribution and spin polarization parameters describing the dynamics of the resonant Auger decay.

In our case, measuring the DSP after photoexcitation with a linearly polarized photon beam yields the advantage of a maximum alignment \( A_{30} = -\sqrt{2} [16] \) in the polarization reference system, and the component of the spin polarization vector may be written as (see [14])

\[
P_{\text{dyn}}(\theta, \phi) = \left\{ \frac{1}{\sqrt{2} + \alpha_2} P_2(\cos \theta) - \frac{1}{2} \cos(2\phi) \sin^2 \theta \right\},
\]

where \( P_2(\cos \theta) \) denotes the second Legendre polynomial and \( \theta \) is the angle between the synchrotron beam axis and the direction of the Auger emission while \( \phi \) gives the angle between the reaction plane and the oscillation plane of the electric field vector, which coincides with the synchrotron storage ring plane. \( \xi_2 \) and \( \alpha_2 \) are the dynamic spin polarization and angular anisotropy parameters, respectively. The spin-polarization parameter \( \xi_2 \) has not been uniquely defined in the literature, e.g., [6,8–10]. We use here the expression given by Lohmann [8], which is based on a somewhat different separation between the spin polarization parameter and the angular part. An expression for \( \alpha_2 \) is given in [17]. These parameters consist of coherent sums over products of partial wave Auger transition amplitudes multiplied by their corresponding phase shift differences and related Clebsch-Gordan and Wigner coefficients.

Our experiment has been performed in the emission reference system under the fixed geometry \( \theta_{\text{exp}} = 90^\circ \) and \( \phi_{\text{exp}} = 135^\circ \). Inserting \( \theta_{\text{exp}}, \phi_{\text{exp}} \) into Eq. (1) we obtain the DSP as

\[
P_{\text{dyn}}(\theta_{\text{exp}}, \phi_{\text{exp}}) = (6\xi_2)(2\sqrt{2} - \alpha_2).
\]

Similarly, for a fully circularly polarized photon beam, the TSP can be related to the same component of the spin polarization vector which yields

\[
P_{\text{trans}}(\theta) = (\sqrt{3} \xi_1 \sin \theta)(\sqrt{2} + \alpha_2 P_2(\cos \theta)),
\]

where \( \xi_1 \) refers to the transferred spin-polarization parameter [5]. The TSP becomes independent of the azimuthal angle \( \phi \) because the combined photonic and target system, where the latter has been assumed as unpolarized, is axially symmetric with respect to the synchrotron beam axis.

Thus, the TSP can be measured under the same solid angle as the DSP, i.e., \( \theta_{\text{exp}} = 90^\circ \), while \( \phi_{\text{exp}} \) becomes redundant, which yields

\[
P_{\text{trans}}(\theta_{\text{exp}}) = (2\sqrt{3} \xi_1)(2\sqrt{2} - \alpha_2).
\]

Our numerical data have been obtained employing a relativistic distorted wave approximation (RDWA). Here, the bound state wave functions of the excited intermediate and the ionized final state of the atom are constructed using the multiconfigurational Dirac-Fock (MCDF) computer code of Grant et al. [18]. Intermediate coupling has been taken into account where the mixing coefficients have been calculated applying the average level calculation mode (see [18]). The calculation of the Auger transition matrix elements has been done applying a relaxed orbital method. Thus, the bound electron wave functions of the intermediate state are calculated in the field of the excited atom, whereas the bound electron wave functions of the final state are calculated in the field of the singly ionized atom.

The atomic state function (ASF) of the intermediate excited and the singly ionized final state have been constructed as linear combinations of configuration state functions (CSF)

\[
\left| \psi_{r}(\alpha) \right| = \sum_{r=1}^{n_c} c_{r}(\alpha) \left| \gamma_{r}(\alpha) \right|.
\]

The configuration states \( \left| \gamma_{r}(\alpha) \right| \) are constructed from antisymmetrized products of Dirac orbitals, which are eigenstates of the total (one-electron) angular momentum and parity. The label \( \gamma_r \) distinguishes the occupation of the different subshells and angular coupling schemes (see Grant [19] for further details). \( n_c \) is the number of CSF included in the expansion and \( c_r(\alpha) \), \( r=1, \ldots, n_c \), are the configuration mixing coefficients for the state \( \alpha \).

The intermediate excited state has been generated as a linear combination from the five possible \( jj \)-coupled \( \text{Ar}^+ (2p_{1/2,3/2}3s_{1/2})J_{\text{tot}}=1 \) and \( \text{Ar}^+ (2p_{1/2,3/2}3d_{3/2,5/2})J_{\text{tot}}=1 \) CSF. We find the \( \text{Ar}^+ (2p_{3/2}3s_{1/2})J_{\text{tot}}=1 \) and \( \text{Ar}^+ (2p_{1/2}3s_{1/2})J_{\text{tot}}=1 \) ASF as almost pure states.

Two calculations have been performed for the final ionic state. In calculation (a) a basis set of eight CSF has been used to generate the final state ASF from the possible linear combinations of the \( \text{Ar}^+ (3p_{1/2,3/2}4s_{1/2})J_{\text{tot}}=1 \) \( jj \)-coupled states (8 CSF-CI). Since this approach has not been able to reveal all lines of the spectrum [20,21] our calculation (b) accounts for all \( jj \)-coupled \( \text{Ar}^+ (3p_{1/2,3/2}3d_{3/2,5/2}) \) CSF, too. Thus, a basis of 36 CSF has been used to generate the final state ASF (36 CSF-CI).

Eventually, the continuum wave function of the Auger electron is evaluated by solving the Dirac equation with an intermediate coupling potential where electron exchange with the continuum has been taken into account. The intermediate coupling potential is constructed from the mixed CSF of the final ionic state. Thereby we take into account, that the ejected electron moves within the field of the residual ion. With this, the Auger transition matrix elements are obtained for calculating the relevant angular anisotropy and spin polarization parameters, respectively. Note, that both are not functions of the transition matrix elements, only but explicitly depend on the scattering phases. Further information may be found in [5,7].

We have measured the spin-resolved electron spectra of the \( \text{Ar} 2p_{3/2} \rightarrow 4s_{1/2} \) and \( \text{Ar} 2p_{1/2} \rightarrow 4s_{1/2} \) autoionization resonances. The experiment has been performed at the ALS operating in two-bunch mode, utilizing the elliptically polarizing undulator (EPU) at Beamline 4.0.2 [22]. The EPU was set to deliver 100% circularly or linearly polarized light at all used photon energies. Electron energy analysis has been performed using a time-of-flight (TOF) spectrometer, collecting electrons emitted at 45° with respect to the storage ring plane [27] in the direction perpendicular to the photon propagation. A spherical Mott polarimeter of the Rice type, operated at 25 kV, mounted at the end of the TOF has been used to carry out the electron spin polarization analysis [23–25]. The geometry of the experiment has been chosen to measure the
polarization of the spin components of the electrons along the photon propagation direction [26]. The instrumental asymmetries of the Mott polarimeter have been eliminated by reversing the helicity in the case of circularly polarized light and switching the polarization from horizontal to vertical in the case of linearly polarized light. The transferred and dynamic electron spin polarizations, corresponding to $p_{\text{trans}}(\theta_{\text{exp}}=90^\circ)$ and $p_{\text{dyn}}(\theta_{\text{exp}}=90^\circ, \phi_{\text{exp}}=135^\circ)$, respectively, can be calculated from the four measured intensities $I_1^r$, $I_2^r$, $I_1^s$, and $I_2^s$ as follows:

$$p_{\text{trans, dyn}}(\theta_{\text{exp}}, \phi_{\text{exp}}) = \frac{1}{S_{\text{eff}}} \sqrt{\frac{I_1^r I_2^s - I_1^s I_2^r}{I_1^r I_2^r + I_1^s I_2^s}} \ (6)$$

The lower index denotes the multichannel plates (MCP) of the Mott detector and the upper index stands for the helicity, respectively the horizontal or vertical polarization of the light. $S_{\text{eff}}$ describes the analyzing power of the polarimeter (the effective Sherman function), which has been determined to be $S_{\text{eff}}=0.13(3)$. Measurement of the DSP and TSP has been achieved arranging MCP$_1$ and MCP$_2$ perpendicular to the reaction plane using linearly and circularly polarized light for the DSP and TSP, respectively. A more detailed description of the experiment and the analysis is given by Snell et al. [26].

Using Eqs. (2) and (4) we have been able to plot the spin-up and spin-down partial intensities of the spectrum. Our results are shown in Figs. 1(a) and 1(b) for the spin-resolved spectrum for the TSP and in Figs. 1(c) and 1(d) for the spin-resolved DSP spectrum of the $L_{2,3}M_{2,3}M_{2,3}$ Auger decay for the excited intermediate argon $(2p_{1/2}^{-1}4s_{1/2})_{j=1}$ and $(2p_{3/2}^{-1}4s_{3/2})_{j=1}$ states, respectively, along with the results of the extended 36 CSF calculation.

The spectrum has been generated assuming a Lorentz profile with a full width at half maximum (FWHM) = 0.1 eV for the Auger lines folded with the appropriate Gaussian line shape. The partial intensities of the 36 CSF-CI represent the calculated spin polarizations, as shown in Table I, but normalized to the experimental total intensities and shifted by an energy offset. A bar diagram underneath of the partial intensities shows the integral values of the spin polarization for energy offset. A bar diagram underneath of the partial intensities shows the integral values of the spin polarization for energy offset. A bar diagram underneath of the partial intensities shows the integral values of the spin polarization for energy offset.

![FIG. 1. (Color) Spin-resolved spectrum for the TSP [(a) and (b)] and DSP [(c) and (d)] for excitation with circularly and linearly polarized light, respectively. Full curves: 36 CSF-CI. Circles with errorbars: experimental data. Blue: partial intensities for spin up ($\Gamma'$). Red: partial intensities for spin down ($\Gamma$). The degree of DSP and TSP for the unresolved group of lines is shown in the chart underneath. Open diamonds: 8 CSF-CI. Open squares: 36 CSF-CI. Filled circles: experimental data. The dashed lines in all figures separate spectra taken with different resolutions and, hence, retardation voltage.](image-url)

**TABLE I.** The calculated degree of dynamic, $p_{\text{dyn}}(\theta_{\text{exp}}, \phi_{\text{exp}})$, and transferred, $p_{\text{trans}}(\theta_{\text{exp}})$, spin polarization is given. Note, that transitions to fine structure terms with $J=7/2$ are suppressed due to $J$-dependent selection rules. (a) Our tentative assignment of the observed and calculated peaks (36 CSF) of the unresolved LSJ fine structure terms. (b) Peak numbers are as assigned in the experimental spectrum.

<table>
<thead>
<tr>
<th>Final states No.</th>
<th>$2p_{1/2} \rightarrow 4s$</th>
<th>$2p_{3/2} \rightarrow 4s$</th>
<th>Spin polarization %</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(3p^2[3p]4s)^2P_{1/2,5/2}$</td>
<td>1a</td>
<td>-0.30</td>
<td>-42.51</td>
</tr>
<tr>
<td>$(3p^2[3p]3d)^2D_{1/2,3/2}$</td>
<td>1b</td>
<td>-0.06</td>
<td>-2.57</td>
</tr>
<tr>
<td>$(3p^2[3p]3d)^2P_{1/2,3/2}$</td>
<td>2</td>
<td>4.42</td>
<td>69.50</td>
</tr>
<tr>
<td>$(3p^2[3d]4s)^2D_{3/2,5/2}$</td>
<td>3</td>
<td>10.14</td>
<td>10.14</td>
</tr>
<tr>
<td>$(3p^2[3d]3d)^2P_{3/2,5/2}$</td>
<td>4</td>
<td>79.56</td>
<td>79.56</td>
</tr>
</tbody>
</table>
roduces the experimental data already very well, particularly for the $3p_{3/2}$ excitation—the corresponding results for the DSP differ qualitatively. In this case, the eight CSF-CI yield a vanishing spin polarization over the whole spectrum. Reasonable agreement with the experiment, showing nonvanishing spin polarization for some lines (2, 3), can be achieved only by employing the extended 36 CSF-CI. Inspecting more closely the different transitions contributing to the spin-polarized part of the spectrum reveals that the CI with 36 CSF, in contrast to the eight CSF-CI, generates unresolved fine structure components with low and high total angular momentum as shown in Table I. The 36 CSF-CI produces $J$ components that correspond in a tentative $LSJ$ scheme to $2L_J$ states with $L \geq 3$ and $J, J' \geq 5/2, 7/2$. These virtual fine structure components however, require non-diagram lines in the form of Auger satellites in order to populate the high $J$ part. Stressing the picture of Auger decay as a two-electron transition, the nondiagram lines are normally an order of magnitude lower than the corresponding diagram lines, similar to the situation in photoionization concerning main and satellite lines. Consequently, such a situation of fine structure multiplets with a mixed diagram-nondiagram order of magnitude lower than the corresponding diagram two-electron transition, the nondiagram lines are normally an order of magnitude lower than the corresponding diagonal two-electron transition. This effect is due to the quenching of the cancellation between different multiplet components as a result of their asymmetric population by internal selection rules for diagram lines.

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[27] This refers to $\phi_{\text{exp}} = 135^\circ$ in our chosen coordinate frame.