Intermediate-coupling phenomena in electron-impact excitation of Hg atoms

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Abstract. In my talk the influence of the intermediate-coupling scheme of Hg on various electron scattering observables such as spin exchange cross sections, spin up-down asymmetries or spin-resolved circular polarization is discussed with special emphasis to recent experimental results from our group and recent theoretical data.

1. Introduction: intermediate coupling in Hg atoms

The study of spin-resolved electron-atom collisions reveals various scattering mechanisms that are masked when only spin-averaged observables are measured. The most significant spin effects in low-energy scattering generally result from electron exchange and the spin-orbit interaction, both in the target alone and in the projectile-target interaction.

As a benchmark target in atomic physics, Hg has been investigated with respect to intermediate-coupling phenomena in electron collisions since more than 40 years. The theoretical description of electron-impact excitation of heavy atoms such as Hg has managed to model more and more detailed structures of experimental data. However, along with this progress the complexity of the mathematical and numerical methods has increased as well. McConnell and Moiseiwitsch [1], used a Born-Ochkur approximation neglecting spin-orbit terms for the continuum electron in the interaction potential, but applied the intermediate-coupling scheme of the 6s6p states of Hg by

\[ \Psi(6s6p^3P_0) = \Psi^0(6s6p^3P_0) \]
\[ \Psi(6s6p^3P_1) = \alpha \Psi^0(6s6p^3P_1) + \beta \Psi^0(6s6p^3P_1) \]
\[ \Psi(6s6p^3P_2) = \Psi^0(6s6p^3P_2) \]
\[ \Psi(6s6p^1P_1) = \alpha \Psi^0(6s6p^1P_1) - \beta \Psi^0(6s6p^3P_1) \]

(1)

where \( \alpha = 0.987 \) and \( \beta = -0.171 \). The excitation energies are 4.71 eV, 4.89 eV, 5.46 eV and 6.70 eV for the states labelled with \(^3P_0\), \(^3P_1\), \(^3P_2\) and \(^1P_1\), respectively.

With this scheme the Born-Ochkur approximation lead to an astonishing quantitative agreement with the experimental data of the linear light polarization of the 6s6p \( \rightarrow \) 6s\(^2\) transitions excited by electron impact [2]. Since the Born-Ochkur calculations from the late 1960\(^{th}\) more sophisticated approaches such as the relativistic distorted-wave Born approximation (RDWBA) or relativistic close-coupling approaches were developed and have improved the description of electron-Hg scattering considerably [3-8].
2. Direct observation of exchange

It is interesting to note that the nature of a spin-exchange singlet-triplet transition was not quite clear in the 1960s. In their famous textbook “Electronic and Ionic Impact Phenomena” Massey and Burhop wrote [9]: “The multiplicity of the state may therefore change by ±2, but this may occur only if the spin direction of the outgoing electron is opposite to that of the incident electron – electron exchange must have taken place in the collision, for we are assuming that spin-orbit interaction is negligible, so the spin of the incident electron cannot be reversed by the impact. Exchange can also occur between electrons with the same spin but in that case it will not be associated with a change of the multiplicity.” It is easy to see that this statement is wrong for a singlet-triplet transition by electron impact as can be seen from the following basic reactions which are obtained if spin conservation is assumed [10]:

\[
e^\uparrow + \text{Hg}(S=0, M_s = 0) \rightarrow \text{Hg}^+(S = 1, M_s = 1) + e(\downarrow) : -\sqrt{2}g
\]

\[
e^\uparrow + \text{Hg}(S=0, M_s = 0) \rightarrow \text{Hg}^+(S = 1, M_s = 0) + e(\uparrow) : g
\]

(2)

The second reaction is a singlet-triplet transition with a change of the multiplicity but does not change the spin of the continuum electron. Only a phase change is required. As a result of a formal analysis of electron-impact excitation of triplet states [10,11], using the indicated amplitudes of the reactions (2), the spin polarization

\[
P = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}
\]

of the incident electrons will be changed to \(P' = P/3\) after the collision for a pure singlet-triplet excitation. Here \(N_\uparrow\) and \(N_\downarrow\) are the numbers of electrons with spin up and down with respect to a given quantization axis. It is true, however, that a singlet-singlet transition with electron exchange cannot result in a spin change at all, i. e. we will then have \(P' = P\).

The cross section calculations of McConnell and Moiseiwitsch [1] using the intermediate coupling scheme (1) stimulated us to perform an experiment where spin exchange effects are directly observed by measuring \(P'\) and \(P\). The idea was that one would observe \(P' = -P/3\) if only the triplet parts of the wave functions contribute to the excitation of the triplet \(6s6p\) levels, and \(P' = P\) if the excitation is caused only by the singlet part in the wave function of the \(6s6p\) \(^1P_1\) state (1). We chose 4.9 eV as a mean excitation energy for the \(6s6p\) \(^3P\) excited states. The contribution of the singlet part to the excitation is expected to become more and more significant with increasing scattering energy, because the dominant excitation via exchange effects (triplet part) diminishes simultaneously. A scheme of the experiment and results [10] which confirm the anticipation about the energy behaviour of spin exchange are shown in Fig. 1.

![Fig. 1 Scheme of the experiment (left) and results (right) for electron-impact excitation of the \(6s6p\) \(^3P\) orbitals in Hg. Polarized electrons produced by Mott scattering from Hg atoms are fired onto another beam of Hg atoms and the polarizations \(P'\) (for scattering in the forward direction) and \(P\) are measured by means of a Mott detector.](image)

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Spin-resolved fine-structure excitation

Another type of experiments are so-called $S_A$ measurements where the observable $S_A$ is given by the differential cross sections $\sigma(\uparrow)$ and $\sigma(\downarrow)$ for spin-up and spin-down electrons, respectively:

$$S_A = \frac{\sigma(\uparrow) - \sigma(\downarrow)}{\sigma(\uparrow) + \sigma(\downarrow)}. \quad (4)$$

This spin-up-down asymmetry is interesting with respect to the fine-structure coupling in Hg [11] which causes an effect illustrated in Fig. 2. It is well known that in electron scattering the electrostatic Coulomb interaction leads to an orbital orientation $L_\perp$, i.e. a nonzero component of the expectation value of the orbital angular momentum operator perpendicular to the scattering plane (cf Fig. 2 left). Excitation of the 6p orbital in Hg may produce such an orbital orientation. The spin-orbit-coupling within the target will thus produce angular momentum configurations as illustrated in Fig. 2 (right) with an orbital angular momentum vector oriented perpendicular to the scattering plane.

![Fig. 2 Orbital orientation (left part): the differential cross section (in electron scattering to the left) for exciting a p orbital with positive orientation (left orbital) may be considerably different from that with negative orientation (right orbital) because of the different Coulomb interactions when continuum and target electrons move “parallel” or “antiparallel”. Right part: the spin-orbit interaction within the target will couple the spin of a triplet state (for LS coupling with $L = 1$ and $S = 1$) with the oriented vector $L$ to resulting fine-structure states with $J = 0$, $J = 1$ and $J = 2$.](image)

If we use polarized electrons we can see from Fig. 2 (right part) that we must “offer” mainly spin-down electrons to excite a $3^3P_0$ state and preferentially spin-up or spin-down electrons to excite a $3^3P_1$ or a $3^3P_2$ state, respectively. A simple angular momentum coupling procedure [11] yields indeed the following relation between the spin-up-down asymmetries for the different fine-structure states:

$$S_A(3^3P_0) = S_A(3^3P_1)/2 = -S_A(3^3P_2)/2 \quad (5)$$

In Hg we have a strong violation of LS coupling for the 6s6p $3^3P_1$ state (1) and, therefore, we cannot expect the relation $S_A(3^3P_1) = -S_A(3^3P_2)$ to be valid. However, because of the coupling scheme shown in Eq. 1, one can show that sign $s_n S_A(6^3P_1) = -s_n S_A(6^1P_1)$ if the continuum spin-orbit interaction is almost negligible and exchange is the dominant spin effect [12]. Experimental and theoretical result confirm this model assumption. In Fig. 3 are shown experimental and theoretical results of the asymmetry functions $S_A$ for the 6s6p $3^3P_1$, 6s6p $3^3P_2$ and 6s6p $1^3P_1$ states at 20 eV. Clearly the relation $S_A(3^3P_1) = -S_A(3^3P_2)$ is violated, however, the results as well as the calculation (though not agreeing well with the experimental data) confirm the relation sign $S_A(6^3P_1) = -s_n S_A(6^1P_1)$. This indicates that exchange in conjunction with the angular momentum coupling scheme is the dominant spin effect.

![Fig. 3 Spin-up-down asymmetry functions $S_A$ for excitation of the 6s6p $3^3P_1$, 6s6p $3^3P_2$ and 6s6p $1^3P_1$ states (from left to right) in Hg at 20 eV. The results (dots) are taken from Borgmann et al [12] where the DWBA calculations (solid lines) are presented in Ref. [4].](image)
4. Study of circular polarization

Another method to study electron-impact excitation processes and the role of intermediate coupling are measurements of the circular polarization of light emitted from Hg after excitation with polarized electrons. The scheme of such experiments is shown in Fig 4. Without electron detection we can measure the so-called integrated circular polarization of light emitted in the direction of the initial transverse electron polarization vector. In that case a nonzero circular polarization is mainly caused by electron exchange in conjunction with the spin-orbit coupling within the target. In so-called electron-photon coincidence studies the scattered electrons are detected in coincidence with the photons. In that case a scattering plane may be defined by the scattered electrons, and a circular polarization for photons emitted perpendicular to the scattering plane may be electron polarization dependent, but is already caused by the Coulomb interaction alone (orbital angular momentum orientation by electron impact, see Fig. 5).

![Fig. 4](image)

**Fig. 4** Left: Scheme of an experiment where a target is excited by electron impact with polarized electrons and the circular polarization of photons emitted in the direction of the electron polarization is determined. In electron-photon-coincidence studies presented below, the electron polarization vector is perpendicular to the scattering plane. Right: Measured (dots) and calculated (lines) integrated circular polarization for the 185 nm line (6s6p $^3P_1 \rightarrow 6s6p ^1S_0$, top) and the 254 nm line (6s6p $^3P_1 \rightarrow 6s6p ^1S_0$, bottom) after excitation with polarized electrons [8,13,14].

Observable exchange processes are expected to contribute to electron-impact excitation of the 6s6p states of Hg with total angular momentum $J = 1$ because of the intermediate-coupling nature (1) of these states. They have excitation energies of 4.89 eV (6s6p $^3P_1$) and 6.7 eV (6s6p $^1P_1$) above the 6s$^2$ $^1S_0$ ground state. Fig . 4 shows the results of the integrated circular polarization for the transitions 6s6p $^1P_1 \rightarrow 6s6p ^1S_0$ (185 nm) and 6s6p $^3P_1 \rightarrow 6s6p ^1S_0$ (254 nm) after excitation with polarized electrons. The experimental and theoretical results are from references [13,14] which we obtained recently. In the near-threshold regime, where the circular polarization is dramatically affected in the vicinity of resonances, i.e., temporary negative-ion states of the collision system, a fully relativistic 36-state Dirac
B-spline R-matrix (DBSR) approach with non-orthogonal orbital sets [8] provides very satisfactory agreement with the experimental data and represents a substantial improvement over an earlier 5-state semi-relativistic Breit-Pauli model (not shown here). At higher energies, the experimental data are in qualitative agreement with the expected reduction of exchange effects with increasing energy. Whereas the calculation and the experimental results for the 185 nm line show excellent agreement, there is a significant quantitative disagreement for the 254 nm line at energies above 12 eV where cascade effects are expected. And, unfortunately, cascade effects seem particularly strong for the spin-dependent light polarizations of the 254nm line, thereby making a straight-forward study of exchange effects by integrated circular polarization measurements virtually impossible. Note that the general sign of the circular polarization is different for the two lines. In the case of the 254 nm line the triplet part of the wave function dominates the sign, whereas for the 185 nm line interference between the singlet and the triplet part dominates which causes the sign change.

Finally, we present results of the angle-differential circular polarization of the $6s6p \, ^3P_1 \rightarrow 6s6p \, ^1S_0$ (254 nm) transition at a scattering energy of 25 eV from recent electron-photon coincidence studies, where cascading is eliminated. Clearly, the circular polarization is strongly electron polarization dependent with the astonishing result that even positive values at small scattering angles are possible (for spin-down electrons). This seems to contradict existing propensity rules, but it was shown [15] that this is an interference effect caused by the triplet-singlet mixture of the triplet state. The experimental results confirm the predicted positive circular polarization for scattering of spin-down electrons into small angles. The extraction of the orbital part of the orientation from the theoretical results, however, yields a consistently negative sign in both calculations, again confirming the validity of the propensity rules.

The results illustrate also that present calculations (which are the models mentioned above) are still a difficult task to describe specific observables such as $P_3$ for electron scattering from complex targets.

**Fig. 5** Angle-differential, spin-resolved circular polarization (Stokes parameter $P_3$) of the 254 nm line ($6s6p \, ^3P_1 \rightarrow 6s6p \, ^1S_0$ transition) at a scattering energy of 25 eV. The experimental results (\(\uparrow\) spin-up, \(\downarrow\) spin-down, \(\circ\) unpolarized electrons) are from Jüttemann [14]. DSBR theories: solid line DSBR-90, dashed line DSBR-36 [16]; RDWBA theories: solid line (MCGS), dashed line (SCGS) [17]:

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