Coincident Photon Coherence Analysis of the Mercury 6^1S_0-6^3P_1-6^1S_0 Excitation/Deexcitation Process in Terms of Spin-Orbit Effects

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The electron-photon delayed-coincidence technique was used to measure the coincident photon radiation for the $6^1S_0 \rightarrow 6^3P_1 \rightarrow 6^1S_0$ process of mercury by electron impact. This process, which is found to be partially coherent, has been analyzed with a new parametrization which takes into account spin-orbit coupling.

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There has been a growing interest in the delayed-coincidence technique since its introduction to the field of atomic physics nearly a decade ago. Many such investigations have been performed and a recent review of electron-photon angular correlation in atomic physics by Blum and Kleinpoppen[1] has covered these developments. Standage and Kleinpoppen[2] conducted an experiment to investigate the coherent nature of the 1^1S-3^1P excitation process in helium. They reported on linear and circular polarization measurements of 3^1P-2^1S (501.6-nm) photons detected in delayed coincidence with electrons inelastically scattered after the excitation of the 3^1P state. Zehnle *et al.*[3] used this method for the vector polarization analysis of the K 4^2P-4^2S photons for K-He collisions. Anderson *et al.*[4] performed measurements of the Stokes parameters for the Mg 3^2P-3^2S photons in a coincidence experiment involving Mg^+-He, -Ne, and -Ar collisions.

In this Letter we report how the delayed-coincidence method has been applied in a crossed-beam apparatus to measure the electron-photon coincidence rates for the $6^1S_0 \rightarrow 6^3P_1 \rightarrow 6^1S_0$ excitation/deexcitation process in mercury by low-energy electron impact (~5 to 7 eV). The results of this experiment show for the first time the influence of spin-orbit effects in the analysis of electron-photon angular correlations. Accordingly the experiment reported here extends the technique of angular correlations into a new area of research in impact excitation of atoms. The electrons scattered with an energy loss of 4.89 eV after the excitation of the 6^3P_1 state were detected in delayed coincidence with $6^3P_1 \rightarrow 6^1S_0$ (253.7-nm) photons. The inelastically scattered electrons were detected in (and thus defined) the scattering plane while the subsequent decay photons were observed perpendicular to the scattering plane. A set of Stokes parameters was obtained from the polarization measurements of the coincident photon radiation. With use of these Stokes parameters, the coherence parameter μ and the polarization correlation parameters were determined.

The experimental arrangement was similar to that of Standage and Kleinpoppen.[2] The atom beam was produced by effusing mercury through a heated capillary source of 1 mm diameter positioned 4 mm away from the interaction region. The mercury beam, directed vertically downwards, was collected in the liquid-nitrogen cold trap of the diffusion pump. The atom source was operated under conditions such that the background pressure in the chamber with the mercury beam on was normally 1×10^{-6} Torr compared to 3×10^{-7} Torr with the beam off. The atom density in the interaction region was estimated to be 7×10^{18} atoms/m^3. This operating condition ensured that pressure-dependent effects, such as resonance trapping, were absent. The onset of such effects was found by performing measurements at different target pressures.

An electron beam from a gun-lens system was focused onto the interaction region. The electrons scattered in a particular direction entered a cone of 1 mm diameter with an acceptance solid angle of 3.5×10^{-3} sr. These electrons after energy selection through a 127° cylindrical analyzer were detected by a channel electron multiplier (Mullard Model 318-BL). The overall electron resolution of the system was 400 meV, which was sufficient to resolve the 6^3P_1 state from the 6^1P_1 state (Fig. 1). Moreover, since the timing circuitry was set for a lifetime of 120 ns for the 6^3P_1 state (King and Adams[5]), no significant contribution is to be expected from the decay of the metastable $6^3P_{0,2}$ states which have lifetimes of 5.6 and 6.8 s, respectively (Krause *et al.*[6]). The resolution time of the apparatus was ~5 ns.

The photon radiation, after being collected by a quartz lens, passed through a rotatable polarizer (Glan-Taylor) and was focused on to a photomultiplier tube (EMI Model 6256S) by another quartz lens. This arrangement was used for the linear

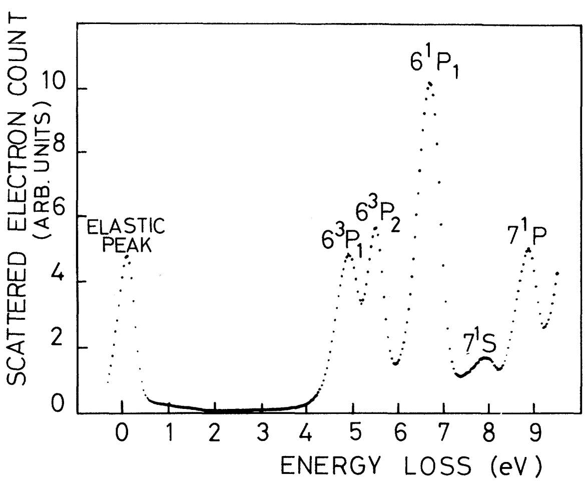

FIG. 1. Energy-loss spectrum of electrons scattered from mercury at 10.5 eV incident electron energy and 70° scattering angle.

polarization analysis. A $\lambda/4$ quartz retardation plate (Melles Griot Model 02-WRQ-005) was placed in front of the linear polarizer when taking measurements for the circular polarization. An isotope cell (Zaidi *et al.*[7]), which absorbs the radiation from the odd mercury isotopes, was positioned in front of the photomultiplier in order to eliminate the effect of hyperfine structure in the observed mercury radiation. No interference filter was used since, for the results presented here, the energy of the bombarding electrons was not high enough to excite the 6^1P_1 and higher states.

The Stokes parameters are defined as follows:

$$P_0 = I(0°) + I(90°), \quad P_1 = I(0°) - I(90°),$$
$$P_2 = I(45°) - I(135°), \quad P_3 = I(\mathrm{RHC}) - I(\mathrm{LHC}). \qquad (1)$$

$I(\alpha)$ is the intensity of light, linearly polarized at an angle α with respect to the electron beam direction. I(RHC) and I(LHC) are, respectively, the right- and left-handed circularly polarized intensities of the photon radiation. When P_0 is normalized to unity, P_1 and P_2 are then linear polarizations measured with reference to the incident electron beam direction and at 45° to this direction, respectively, and P_3 is the circular polarization. The coherence correlation factor (Born and Wolf[8]) is then related to the normalized Stokes parameters P_1, P_2, and P_3 as follows:

$$\mu = |\mu| e^{i\beta} = (P_2 + iP_3)/(1 - P_1^2)^{1/2}, \qquad (2)$$

where $|\mu|$ is the degree of coherence between two orthogonal linearly polarized components of the radiation, parallel and perpendicular to the incident electron beam, and β is their effective phase difference.

A useful quantity which characterizes the coherence of the emitted light is the "vector polarization" $\vec{P}$. The three normalized Stokes parameters (P_1, P_2, P_3) form the components of this three-dimensional vector whose magnitude is the "degree of polarization" (Born and Wolf[8]), given by

$$|\vec{P}| = (P_1^2 + P_2^2 + P_3^2)^{1/2}. \qquad (3)$$

The experiment involved the accumulation of electron-photon coincidence counts for various positions of the polarizer angle α at fixed incident electron energies and scattering angles. The total number of coincidences was normalized to the total number of scattered electrons collected over the duration of each run. In this way the effect of variations in the electron beam current, the target density and efficiency of the electron detector was eliminated. The statistical uncer-

TABLE I. Coincident-photon parameters for the $6^1S_0 \rightarrow 6^3P_1 \rightarrow 6^1S_0$ excitation/deexcitation process in mercury. Quoted experimental uncertainties represent 1 standard deviation.

Incident electron energy	Scattering angle	Normalized Stokes parameters P_1	P_2	P_3	$\|\vec{P}\|$	$\|\mu\|$	β (rad)
5.5 eV[a]	50°	0.31 ± 0.05	− 0.14 ± 0.06	0.40 ± 0.05	0.40 ± 0.05	0.27 ± 0.05	0.59 ± 0.13
5.5 eV[b]	50°	0.42 ± 0.08	− 0.34 ± 0.08	− 0.37 ± 0.07	0.65 ± 0.08	0.55 ± 0.08	0.83 ± 0.15
5.5 eV[b]	70°	0.13 ± 0.08	− 0.52 ± 0.08	− 0.27 ± 0.04	0.60 ± 0.07	0.59 ± 0.08	0.48 ± 0.08
6.5 eV[b]	50°	0.26 ± 0.09	− 0.39 ± 0.09	− 0.44 ± 0.04	0.64 ± 0.07	0.61 ± 0.07	0.85 ± 0.12
6.5 eV[b]	70°	0.13 ± 0.06	− 0.45 ± 0.09	− 0.42 ± 0.07	0.63 ± 0.08	0.62 ± 0.09	0.75 ± 0.13

[a]Results taken without the isotope cell and so the hyperfine effect was present.
[b]Results taken using the isotope cell so as to eliminate the hyperfine effect.

tainty of the coincidence counts was calculated assuming Poisson statistics were applicable. The data so obtained were fitted to a cosine function of period π in order to have a consistent set of radiation intensities and to reduce the error in the Stokes parameters. The radiation intensities I ($\alpha = 0°, 45°, 90°, 135°$) were determined from the fitted values of the data. After introducing the $\lambda/4$ plate, similar measurements were performed and I(RHC) and I(LHC) were determined for $\alpha = 45°$ and $135°$, respectively.

The experimental results are given in Table I. The coherence parameters $|\mu|$ and β and the degree of polarization are also tabulated here as functions of incident electron energy and scattering angle. The effect of hyperfine structure can clearly be seen in the coherence parameters and in the degree of polarization. In comparison with the $1^1S \to 3^1P \to 2^1S$ process in helium (Standage and Kleinpoppen[2]) which is completely coherent, that is $|\vec{P}| = |\mu| = 1$, the present experiment shows that the $6^1S_0 \to 6^3P_1 \to 6^1S_0$ excitation/deexcitation process in mercury is not completely coherent even when the effect of hyperfine structure has been eliminated.

It is interesting to note from our results that for a given scattering angle, the degrees of coherence are equal for incident energies of 5.5 and 6.5 eV. Since, unlike at 6.5 eV, the excitation at 5.5 eV occurs via a negative-ion state (Zaidi *et al.*[7]), this equality may be due to either a fortuitous choice of scattering angle, or to the excitation process being coherent and the incoherency arising during the period of relaxation of the excited state. This latter reason is consistent with the negative-ion state decaying very fast ($\sim 10^{-14}$ s) to the 6^3P_1 state. However, it may be seen that the normalized Stokes parameters do show significant changes at these energies.

It has been shown by da Paixão *et al.*[9] that as a consequence of spin-orbit coupling, the radiation emitted from an excited state will not be completely polarized. These authors proposed a new parametrization in the analysis of angular correlation data which takes into account the spin-orbit interaction. According to their suggestions the magnetic-sublevel excitation cross section can be written as $\sigma_{m_j} = \langle a(m_j)a(m_j)\rangle$, where $\langle\cdots\rangle$ indicates an average over the spin of the incident electron and a sum over the spin of the scattered electron. The parameters which can be defined for a complete representation of the angular correlation data from the $^3P_1$ excitation are

$$\lambda = \sigma_0/(\sigma_0 + 2\sigma_1), \quad \cos\Delta = |\langle a(0)a(1)\rangle|/(\sigma_0\sigma_1)^{1/2},$$

$$\langle a(0)a(1)\rangle = |\langle a(0)a(1)\rangle| e^{i\bar{\chi}},$$

$$\cos\epsilon = -\langle a(-1)a(1)\rangle/\sigma_1.$$

These parameters λ, Δ, $\bar{\chi}$, and ϵ can be related to the Stokes parameters and the degree of coherence as follows:

$$P_1 = \frac{(3\lambda - 1) - (1-\lambda)\cos\epsilon}{(1+\cos\epsilon) + (1-\cos\epsilon)\lambda}, \tag{4}$$

$$P_2 = \frac{-4|\lambda(1-\lambda)|^{1/2}\cos\Delta\cos\bar{\chi}}{(1+\cos\epsilon) + (1-\cos\epsilon)\lambda}, \tag{5}$$

$$P_3 = \frac{-4|\lambda(1-\lambda)|^{1/2}\cos\Delta\sin\bar{\chi}}{(1+\cos\epsilon) + (1-\cos\epsilon)\lambda}, \tag{6}$$

$$\mu = \frac{\sqrt{2}\cos\Delta}{(1+\cos\epsilon)^{1/2}}. \tag{7}$$

By using the results of Table I and these formulae one directly obtains values for $\bar{\chi}$ ($=\beta$) from Eqs. (5) and (6). However, values for the remaining quantities are limited to intervals since we have only a set of three independent equations for four parameters. These intervals for λ and Δ, as given in Table II, follow from the possible values for $\lambda (0 \leq \lambda \leq 1)$, $\Delta (0 \leq \Delta \leq \pi/2)$, and $\epsilon (0 \leq \epsilon \leq \pi)$. In order to determine fully these quantities a further experiment is required where the photon is detected out of the scattering plane and with the photon azimuthal angle different from $\pi/2$. The connection between the χ value without spin-orbit interaction and the spin-orbit phase $\bar{\chi}$ is given by the relation $\cos\chi = \cos\bar{\chi}\cos\Delta$. Since Δ is found to lie between 0.87 and $\pi/2$, this demonstrates the unambiguous detection of the spin-orbit effects (which would vanish for $\Delta = 0$) in the process under investigation.

TABLE II. Values of $\bar{\chi}$, Δ, and λ ($0 \leq \Delta \leq \pi/2$, $0 \leq \lambda \leq 1$) for the 6^1S_0-6^3P_1-6^1S_0 excitation/deexcitation of mercury. The hyperfine effect has been eliminated.

Incident energy	Scattering angle	$\bar{\chi}$ ($=\beta$) (rad)	Δ (rad)	λ
5.5 eV	50°	0.83 ± 0.15	>0.84	<0.71
5.5 eV	70°	0.48 ± 0.08	>0.94	<0.56
6.5 eV	50°	0.85 ± 0.12	>0.91	<0.63
6.5 eV	70°	0.75 ± 0.13	>0.91	<0.56

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Optical Measurement of Free-Electron Polarization

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The spin polarization of photoelectrons emitted by an activated gallium arsenide photocathode excited with circularly polarized light has been measured by an optical method: A crossed-beam experiment has been performed in which polarized electrons transfer spin angular momentum to zinc atoms in inelastic exchange collisions. The polarized atoms emit circularly polarized light in the direction of the spin transfer. The degree of circular polarization is directly related to the electron polarization.

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We have performed the first experiment where spin polarization of free electrons is detected by an optical method. The method used was discussed theoretically a few years ago.[1-3] Polarized electrons can transfer spin angular momentum to atoms by inelastic exchange collisions. In such a collision, the electrons polarize the atoms and the information about the polarization of the electron beam is contained in the emitted light. This light, observed in the direction of the spin transfer, is circularly polarized. For two-electron atoms, when there is no orbital angular momentum transferred, the degree of light polarization is very simply related to the polarization of the incident electrons. The Pauli principle plays a dominant role in the polarization of these atoms; this is to be distinguished from situations where atomic polarization arises because of magnetic interaction.

In our experiment, excitation from the $ns^2\,{}^1S_0$ ground state to the triplet $ns(n+1)s\ {}^3S_1$ states has been considered. For a collision where opposite-spin electrons are exchanged, a spin up (down) electron transfers a unit of angular momentum to the atom and the substate $|{}^3S;1-1\rangle$ ($|{}^3S;1+1\rangle$) cannot be populated. Therefore with polarized electrons the population of the ${}^3S_1$ state is not equally distributed among the magnetic sublevels. More precisely if n_+ (n_-) represents the number of electrons with spin parallel (antiparallel) to the quantization axis Z, the polarization of the electron beam being $P_e=(n_+-n_-)/(n_++n_-)$, the relative populations $N(M)$ of excited atoms in the $|{}^3S;1M\rangle$ substates are

$$N(M=0)=\tfrac{1}{3},\quad N(M=\pm 1)=\tfrac{1}{3}(1\pm P_e).$$

The light polarization from the transition to the $nsnp\,{}^3P_J$ states which is observed is therefore connected to the population $N(M)$. In fact, light emission involves orbital variables and any optical effect related to the spin appears only through spin-orbit interaction. It is therefore essential to have the fine structure well resolved in order to observe separately the circular polarization of the three lines ($J=0,1,2$). The total emitted light intensity $I=I_++I_-$, where I_+ (I_-) is the intensity of the light with σ_+ (σ_-) polarization, is proportional to $2J+1$. The polarization $P=(I_+-I_-)/(I_+$