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Spin-Polarized Electrons from Autoionizing Transitions in Thallium

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The spin polarization of photoelectrons emitted by thallium atoms exposed to circularly polarized light has been measured between 1400 and 1700 Å. The polarization curve shows a pronounced structure which is determined by the autoionization resonances. The results are explained on the basis of Fano's resonance theory.

Recent theoretical and experimental investigations have shown that spin-polarized electrons can be obtained by photoionization of alkali atoms with circularly polarized light (Fano effect[1]). This conspicuous effect of spin-orbit coupling prompted strong interest in the question of whether other elements also yield polarized photoelectrons. Several theoretical approaches to this problem have been made recently, but so far no experimental results have been obtained.

It is the purpose of this Letter to present and to explain experimental results showing that high spin polarization can result from autoionization transitions. Although thallium has been chosen as a specific example, the polarization phenomenon described here exists also for autoionization resonances in other elements.

Figure 1 shows the transitions from the ground state $6^2P_{1/2}$ of thallium[2] to the continuum which are induced by circularly polarized σ^+ light according to the selection rules $\Delta m_j = +1$, $\Delta l = \pm 1$, and $\Delta j = 0, \pm 1$. As we start from unpolarized thallium atoms, the states $\epsilon^2S_{1/2}$, $\epsilon^2D_{3/2}$ $(m_j = \frac{1}{2})$, and $\epsilon^2D_{3/2}$ $(m_j = \frac{3}{2})$ are reached. The transition probabilities are proportional to $\frac{4}{9}R_S^2$, $\frac{2}{9}R_D^2$, and $\frac{2}{3}R_D^2$, respectively, as the simple calcula-

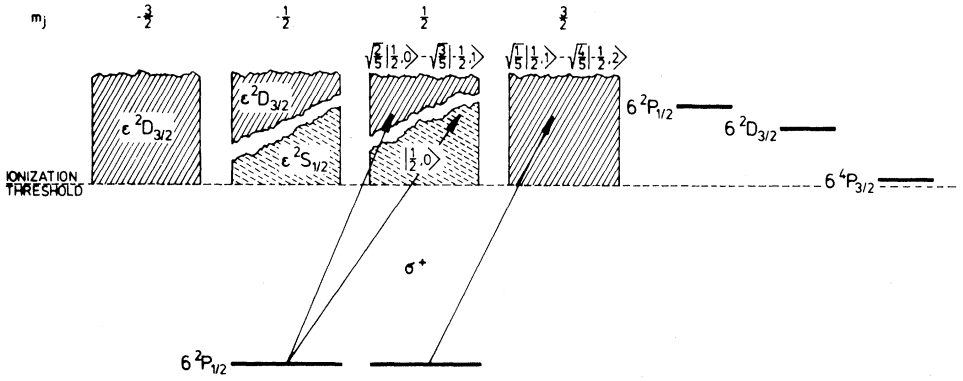

FIG. 1. Autoionization states and continuum states reached from the ground state $6^2P_{1/2}$ of thallium with circularly polarized σ^+ light.

tion of the matrix elements of the dipole operator $x+iy$ shows. R_S and R_D are the radial parts of the matrix elements for transitions to the states $\epsilon^2S_{1/2}$ and $\epsilon^2D_{3/2}$.

The spin polarization of the photoelectrons in a specific final state follows immediately from the coupling coefficients of the wave functions $|m_s, m_l\rangle$ given in Fig. 1. For example, we find for the state $\epsilon^2D_{3/2}$ ($m_j = \frac{3}{5}$) the polarization

$$P = (\tfrac{1}{5} - \tfrac{4}{5})/(\tfrac{1}{5} + \tfrac{4}{5}) = -0.6 .$$

Superposition of the spin polarizations of the final states weighted with the corresponding transition probabilities yields the resulting polarization of the photoelectrons. The superposition can be made incoherently, since we are interested in the polarization of all the photoelectrons independent of their direction of emission, so that coherent terms disappear as a result of the orthogonality of the spherical harmonics. With the transition probabilities given above one easily finds that the two final $\epsilon^2D_{3/2}$ states together contribute a polarization of -0.5, whereas the $\epsilon^2S_{1/2}$ state has a polarization of $+1$. Because of the different signs of these values the resulting polarization

$$P = (1\times Q_S - 0.5\times Q_D)/(Q_S + Q_D) \qquad (1)$$

would normally not be very large, the exact value depending on the ratio of the cross sections for transitions to the S and D states, Q_S/Q_D, which depends on the wavelength λ. Since in general Q_D is rather large compared with Q_S, negative polarization would be the rule.[3]

So far we did not take into account, however, that the polarization of the photoelectrons is considerably modified as a result of the fact that the final states can be reached also via autoionization. The autoionizing states resulting from excitation of a $6s$ electron are also shown in Fig. 1. They give rise to a resonance behavior of the photoionization cross section Q at the corresponding wavelengths as shown in Fig. 2 which is based on the results of Marr and Heppinstall[4] and Berkowitz and Chupka.[5] A general expression for the energy dependence of the cross section Q has been derived by Fano[6] by a theoretical description of the interaction of the discrete autoionizing states with the continuum states. In applying these results to our specific case one must take into account that the autoionizing states interact only with the continuum states of the same quantum numbers J and M_J[7]; i.e., $^2P_{1/2}$ decays into the $\epsilon^2S_{1/2}$ continuum and $^2D_{3/2}$ and $^4P_{3/2}$ into $\epsilon^2D_{3/2}$.

One then obtains for the energy dependence of the cross section Q

$$Q = Q_S \frac{(1+q_0/\epsilon_0)^2}{1+\epsilon_0^{-2}} + Q_D \frac{(1+q_1/\epsilon_1+q_2/\epsilon_2)^2}{1+\epsilon_1^{-2}+\epsilon_2^{-2}}$$
$$= Q_S' + Q_D', \qquad (2)$$

where Q_S and Q_D are the cross sections for transitions into the S and D continuum states for the case discussed before, where autoionization does not play a role. In the energy parameter

$$\epsilon_i = (E - E_i)/\tfrac{1}{2}\Gamma_i , \qquad (3)$$

the "resonance energy" E_i determines the position and Γ_i the width of a resonance. By $i = 0, 1, 2$ we denote the resonances at 1490, 1610, and 2007 Å, i.e., 67137, 62112, and 49826 cm^{-1}, respectively. The shape of a resonance depends on the profile index q_i.

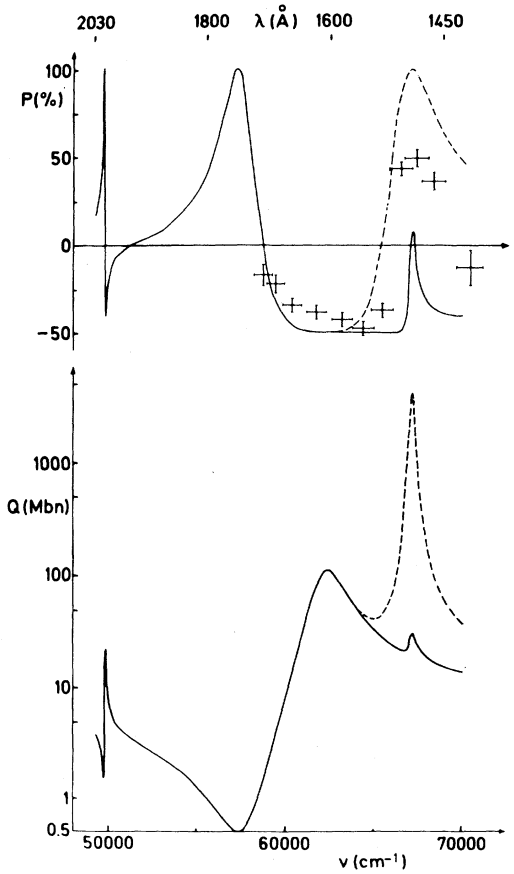

FIG. 2. Lower part: photo cross section of thallium according to Ref. 4 (full curve) and Ref. 5 (broken curve). Upper part: spin polarization of photoelectrons; experimental results and values calculated according to Eq. (4) with use of the cross sections from the lower part.

Instead of Eq. (1) we now obtain for the spin polarization

$$P = (1 \times Q_S' - 0.5 \times Q_D')/(Q_S' + Q_D'), \quad (4)$$

showing that the polarization curve strongly reflects the resonance structure of the cross section and thus is strongly influenced by the autoionizing transitions.

This is illustrated by Fig. 2 which gives the polarization calculated from Eq. (4) with use of the parameters ϵ_i and q_i which follow from the experimental cross sections.[4,5] Whenever interference between direct and autoionizing transitions causes a dominance of one of the cross sections Q_S' or Q_D', the polarization tends to +100% or −50%, respectively. As an example we point out that Q_D' disappears because of destructive interferences at approximately 49 730 and 57 239 cm^{-1}, so that Q_S' makes the only contribution to P; in these cases the polarization should be exactly 100%. On the other hand Q_S' dominates near 67 137 cm^{-1} because of constructive interference between direct and indirect photoionizations. A tendency of the polarization towards 100% is then to be expected. The minima of P (exactly −50% at about 61 336 cm^{-1} and not far from −50% near 49 845 and 62 360 cm^{-1}) are explained vice versa.

Using the results discussed above as a guide, we have performed an experiment whose setup is shown in Fig. 3. The uv light is produced by means of a H_2 discharge lamp (Hinteregger lamp) and passes through a Seya-Namioka vacuum uv monochromator (McPherson Model 235, 0.5 m) which was calibrated with Cs and Hg resonance lines. The unpolarized light emerging from the

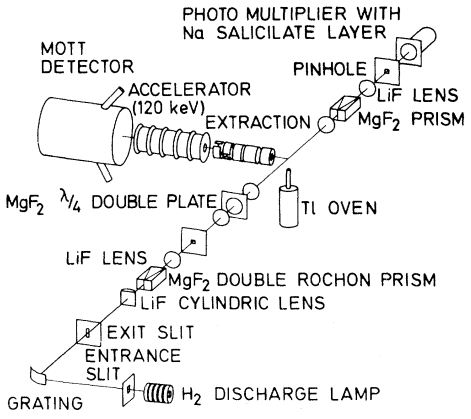

FIG. 3. Schematic diagram of the apparatus.

1-mm exit slit is linearly polarized by a MgF_2 double Rochon prism. The transformation into circular polarization is made with two MgF_2 plates with an angular difference of 90° between their axes and a thickness difference such that the optical path difference for the two linear polarization states is $\frac{1}{4}\lambda$ at $\lambda = 1500$ Å. The circular polarization varied between 93% and 60% in the wavelength range used.

The circularly polarized light is focused onto a thallium-vapor beam emerging from an oven which was heated to 1100°C by a pulsed current (duty cycle 1%) through a bifilar coil. By careful design of the extraction system it was made certain that only the electrons resulting from photoionization of the thallium beam were detected. The electrons were extracted independently of their direction of emission. The first element of the extraction system was a quadrupole lens not shown in Fig. 3 with an extraction field of 50 V/cm in the region of the atomic beam.

After extraction the electrons were accelerated to 120 keV for analysis of their polarization in a Mott detector [(scattered intensity)/(incident intensity) = 1.5×10^{-3}, Sherman function $S = 0.26 \pm 0.1$, gold foil 180 $\mu g/cm^2$]. In order to eliminate apparatus-related asymmetries of the Mott detector the sign of the light polarization and thus the sign of the electron polarization was changed every minute. It was made certain that the results did not depend on the conditions of the Tl target (pressure, space charge, light intensity, etc.).

The analyzer for the light polarization has a construction analogous to that of the polarizer. The positions of the optical elements in analyzer and polarizer can be varied in order to compensate for changes of the lens properties with varying wavelength. For detection of the uv light a sodium salicylate layer in combination with an interference filter for the fluorescence light and a photomultiplier is used. Measurements of light polarization and electron polarization were made simultaneously.

The experimental results are shown in Fig. 2. The vertical error bars are the rms sum of the errors of the light polarization and the Sherman function and of the single statistical error. The horizontal error bar is the resolution of the monochromator (31 Å half-width of triangle). In the wavelength range from 1400 to 1700 Å covered by the experiment, the agreement between our calculated results which have already been

communicated during the construction of the apparatus[8] and the experimental data is satisfactory. The differences might be due to uncertainties of the experimental resonance cross sections or to the possible existence of a $6^2S_{1/2}$ autoionization transition[9] near 1300 Å. According to the measurement the value $P = -50\%$ [where $Q_S' = 0$, Eq. (4)] is at the wave number $64\,400 \pm 600$ cm^{-1}. Since from Eq. (2) $\epsilon_0 = -q_0$ for $Q_S' = 0$, Eq. (3) together with the measurement yields $\Gamma_0 q_0 = 5500 \pm 1200$ cm^{-1} for the $6^2P_{1/2}$ autoionization transition.

We have shown that polarized electrons can be obtained from autoionizing transitions and that the polarization can be utilized for the analysis of the autoionization resonances. It is worth noting that the intensity distribution $I(\lambda)$ of the ionizing light (depending on the light source and the absorption coefficients of the optical devices) does not have to be known for the measurement of the wavelength-dependent $P(\lambda)$, since the electron polarization is obtained from the ratio of the electron intensities in the two counters of the Mott detector.

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Measurements of Photon Polarization and Angular Correlations for He^+ - He Collisions Coincidence Technique Using an Ion-Photon

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The polarization of photons from the transition $3^3P \rightarrow 2^3S$ and the angular distribution of radiation from the decay $2^1P \rightarrow 1^1S$ emitted in He^+ +He collisions at an incident energy of 150 eV and an ion scattering angle of 13.5° were measured by detecting the scattered ions and photons in delayed coincidence. From these measurements we conclude that the 3^3P excitation takes place through a Σ-Σ radial coupling, and the 2^1P excitation predominantly through a Σ-Π rotational coupling, probably at large internuclear separation.

The interpretation of excitation processes in ion-atom collisions at moderate energies (a few hundred eV) is based on a molecular description of the colliding-particle system. In most cases, the primary excitation mechanism consists in an interaction between ground and excited molecular states, which occurs at a rather small internuclear distance (a few bohrs). In general, many coherent scattering amplitudes, corresponding to several output channels, are produced at the same time by the collision and these amplitudes can interfere at large or infinite internuclear distances. Thus, in spite of the fact that the primary excitation mechanisms are now generally well understood, transition probabilities—or inelastic differential cross sections—for excitation to *one* specific atomic level are often difficult to determine because of these "secondary" interactions occurring at large distances. For the particular case of the excitation of a ^1P state of helium, in a He^+-He collision, for a specific scattering angle (the ion is assumed to remain in the ^1S state), different and coherent amplitudes $a_m(\theta, \Phi)$ are obtained for the different magnetic sublevels $m = -1, 0, +1$, with the symmetry relation $a_1 = -a_{-1}$ (see Macek and Jaecks[1]). As the collision time at moderate energies (10^{-15} sec) is much shorter than the lifetime of the excited