PHYSICAL REVIEW A VOLUME 38, NUMBER 12 DECEMBER 15, 1988

Atomic scattering factor and spin-polarization calculations

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(Received 18 July 1988)

A new set of atomic scattering factors derived from relativistic Hartree-Fock wave functions has been calculated for publication in the new edition of the *International Tables for Crystallography,* 3rd ed. (Reidel, Dordrecht, in press). Presented here is a discussion of the numerical stability of the results and a discussion of the spin-polarization function, $S(\theta,\phi)$, including a comparison of the new calculations with experimental data. After the calculated $S(\theta,\phi)$ functions are normalized to the Coulombic values, the difference between theory and experiment leads to some interesting conclusions.

I. INTRODUCTION

Electron-spin polarization has become a frequently used quantity in the analysis of atomic and molecular systems, surfaces,[1] and β emitters.[2] Since the invention of GaAs sources, polarized electron beams have become available which allow experiments to probe the spin-dependent properties of many targets. The transverse polarization relative to the scattering plane of the incident and scattered electrons is analyzed by Mott scattering. While other analyzers are becoming available which are more compact and easier to build, the well-studied Mott system remains the standard by which all new systems are compared. In spite of the central importance of this analyzer, there still remains some discrepancy between experimental and theoretical results.[3] However, the theory of scattering from central potentials at high incident energies is so well understood that, given a large computer and a good atomic potential function, one should be able to compute the polarization (or Sherman) function with sufficient precision to be of valuable assistance to experimentalists in this area. New calculations employing a relativistic Hartree-Fock potential for Au are presented here with an analysis of how the cross section and spin polarization are effected by the matching radius, integration step size, and convergence of the partial-wave sum.

II. THE POLARIZATION FUNCTION

For an unpolarized incident electron beam, the polarization of the scattered electron is described by the Sherman function,[4]

$$S=\frac{i(fg^*-gf^*)}{|f|^2+|g|^2}=\frac{2|f||g|\sin(\eta_g-\eta_f)}{|f|^2+|g|^2}\ , \tag{1}$$

where f is the direct scattering amplitude, g is the spin-flip scattering amplitude, and η_f and η_g are the phases of the scattering amplitudes when they are given in polar form, $f=|f|e^{i\eta_f}$. The cross section of a double scattering experiment is given by[5]

$$\frac{d\sigma(\theta_1,\theta_2,\phi)}{d\Omega}=\frac{d\sigma(\theta_1)}{d\Omega}\frac{d\sigma(\theta_2)}{d\Omega}[1-\delta(\theta_1,\theta_2)\cos\phi]\ , \tag{2}$$

where θ_1 and θ_2 are the scattering angles for the first and second scattering processes, ϕ is the azimuthal angle between the two processes, and $\delta(\theta_1,\theta_2)$ is the product of the Sherman functions of the two scattering processes; i.e., $\delta(\theta_1,\theta_2)=S_1(\theta_1)S_2(\theta_2)$.

To measure the Sherman function of an atom one needs to perform a double scattering experiment. The first scattering process is used to produce an electron beam with polarization $P=S_1(\theta_1)$, where the polarization of the beam is defined as[1]

$$P=\frac{I_\uparrow-I_\downarrow}{I_\uparrow+I_\downarrow}\ , \tag{3}$$

where $I_\uparrow$ and $I_\downarrow$ are the currents of the electrons in the spin-up and spin-down states, respectively, relative to a plane which defines P. The second scattering process is used to measure the Sherman function of the target, S. In most experiments two detectors are placed at the same scattering angle θ_2, but at different azimuthal angles, ϕ_R and ϕ_L An asymmetry can then be recorded in the two detectors. For an experiment having the right detector at $\phi_R=0$ and the left at $\phi_L=\pi$, the ratio of the count rates according to Eq. (2) will be given by

$$\frac{R_L}{R_R}=\frac{d\sigma(\theta_1,\theta_2,\phi_L)/d\Omega}{d\sigma(\theta_1,\theta_2,\phi_R)/d\Omega}\Delta=\frac{1-\delta(\theta_1,\theta_2)\cos\phi_L}{1-\delta(\theta_1,\theta_2)\cos\phi_R}\Delta$$
$$=\left[\frac{1+SP}{1-SP}\right]\Delta\ , \tag{4}$$

where Δ is a constant inserted to account for the instrumental asymmetry.[6] If it is possible to reverse the polarization P of the beam formed from the first scattering process, a second measurement can be performed to determine Δ. Thus, with the beam reversed,

$$\frac{R'_L}{R'_R}=\left[\frac{1-SP}{1+SP}\right]\Delta\ . \tag{5}$$

Solving for S one obtains

$$S=\frac{1}{P}\left[\frac{X-1}{X+1}\right], \tag{6}$$

where

$$X=\left[\frac{R_L}{R_R}\frac{R_R'}{R_L'}\right]^{1/2}.$$

Unfortunately, the reversal of the polarization introduces new problems which are not eliminated with the double measurement. Furthermore, there are several issues which are not addressed in this sequence of measurements. A detailed critique of the experimental approach is presented in Ref. 3. There, a series of arguments is given why it is reasonable to question the data and why certain issues and problems should be addressed in future experiments. A similar set of questions can be raised in regard to the theoretical values, such as why the results of Holzwarth and Meister[7] disagree with those of Bühring.[8] Both use partial-wave techniques and both claim little sensitivity to the inner-shell screening electrons. Therefore a new set of scattering factors was calculated and the results were tested for all possible numerical issues such as stability, step size, and convergence.

III. ABOUT THE CALCULATIONS

The direct scattering amplitudes f for each atom are presented in the *International Tables for Crystallography*[9] in the range of $s\,[=(4\pi/\lambda)\sin(\theta/2)]$ from 0 to 60 Å^{-1} in steps of 1 Å^{-1} at energies of 10, 40, 60, and 90 keV. Though not presented in the tables, the spin-flip scattering amplitudes g have also been calculated and are available upon request. The scattering factors for each atom were calculated by the partial-wave summation method[4,9] with partial-wave phase shifts calculated from relativistic Hartree-Fock potentials[10] using the computer program A SCAT by Yates.[11] For each calculation the phase shifts were calculated down to a value of $\sim 10^{-7}$ rad to assure convergence of the partial-wave sum. About 200–300 partial waves were used for light atoms at low energies, but up to 900 partial waves were used for heavier atoms at high energies (e.g., Au at 90 keV). As a further test of convergence, the coefficient reduction method of Bühring[8] was used to calculate the partial-wave sum. Identical results were obtained for large angles ($>45°$) using only 30 partial waves with five coefficient reductions.

As is described elsewhere,[4] in order to integrate the Dirac equation, the logarithmic derivative of the solution is matched to the field free solution at a radius far from the atom, where the atomic potential is considered negligible. This *matching radius* was chosen to be 10 Å for all of the calculations presented in the tables. Though varying the matching radius from 5 Å to 10 Å causes little variation in the results (less than 0.1%), it proportionally alters the number of partial waves required for convergence (from 900 partial waves with a 10 Å matching radius to 450 with 5 Å for Au at 90 keV).

The calculations were more sensitive to the choice of integration step size. For the tables, an integration step size of 0.0010 Å was used for the partial-wave phase-shift calculations. However, because of the steepness of the potential function near the nucleus, it is necessary to perform the integration with a considerably smaller step size. It was found that all step-size dependence could be eliminated if a step size of 0.00005 Å was used for the first 0.005 Å of the integration for partial waves with $l<5$.

IV. RESULTS OF THE CALCULATION AND COMPARISON WITH EXPERIMENT

After the new cross sections were calculated for Hg using the present potential and computer code, they agreed with the results of Kessler and Weichert[12] within the quoted uncertainties as well as with previously calculated results.[8] Since the experimental data are normalized to the theoretical cross sections of a hydrocarbon, the cross sections were recalculated and the matching procedures were found to be justified. In order to get a coarse estimate of the importance of the effect of exchange of atomic electrons with the incident electron, the free-electron gas approximation with its $\rho^{1/3}$ potential was employed. The calculated cross sections increased slightly, but remained within the experimental error limits. A similar effect occurred in the polarization results.

The topology of the spin-polarization function as a function of angle as shown in Fig. 1(a) can be explained by noting that $|f(s)| \gg |g(s)|$ for all values of s. Equation (1) then reduces to

$$S\approx\frac{2|g|}{|f|}\sin(\eta_g-\eta_f)\,. \tag{7}$$

For small values of s, the function $|g|/|f|$ is fairly smooth, therefore the $\sin(\eta_g-\eta_f)$ factor governs the form of S. Figure 1(a) shows $\eta_g-\eta_f$ plotted along with S for comparison. As can be seen in Fig. 1(b), a maximum in $|g|/|f|$ is responsible for the large extremum in S at $s\sim 180$ Å^{-1}, though S is negative at this angle due to the sign of the $\sin(\eta_g-\eta_f)$ term. The spin-flip amplitude $|g|$ must go to 0 (and therefore so does S) at $\theta=0$ and 180° since, in the partial-wave expansion, g is a sum of associated Legendre polynomials in $\cos\theta$, all of which are zero at these angles. The minimum of S stays in a fairly constant position for all atoms and energies, changing monotonically from 115° for H to 117° for Au. Figure 2 shows the spin polarization at its extremum as a function of atomic number for 40 keV electrons.

Figure 3 depicts various experimental and theoretical values of the Sherman function of Au at 120° as a function of energy. The experimental results of Gray *et al.*[4] are renormalized to the value of our results at 120 keV where the calculated polarization has a value of -0.404. The discrepancy at lower energies between this experiment and our calculations could be due to elastic multiple scattering, not accounted for in the experiment. At lower energies, deviations are expected not only because the multiple-scattering probability increases, but also because the momentum acceptance cone of the detectors is larger, and consequently the count rate from multiply scattered electrons is larger.[13]

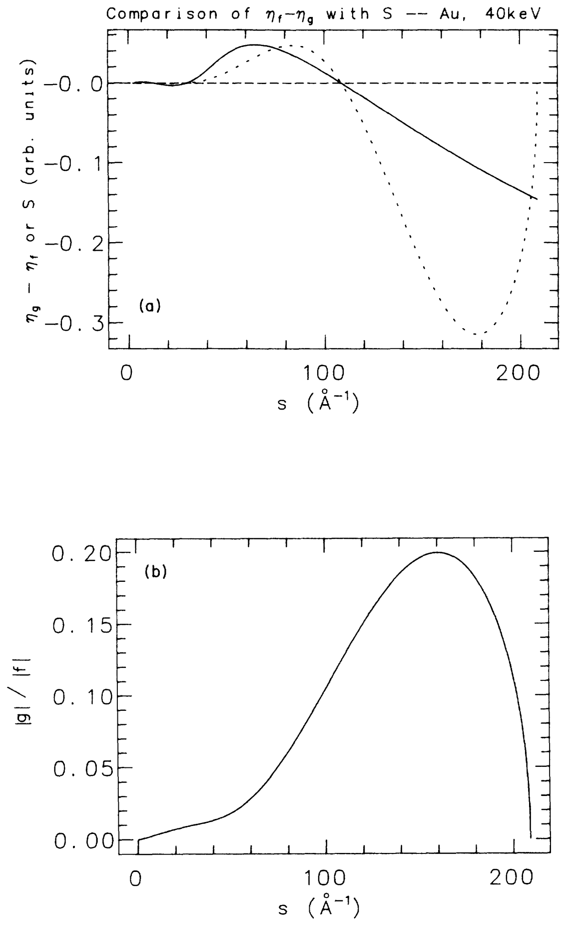

FIG. 1. (a) Dashed line, the Sherman function S as a function of s (Å^{-1}) for $E=40$ keV and $Z=79$ (Au). Solid line, the phase difference, $\eta_g-\eta_f$, a a function of s (Å^{-1}). The function $\eta_g-\eta_f$ is scaled down by a factor of 10 for comparison with S. (b) $|g|/|f|$ plotted as a function of s (Å^{-1}) for $E=40$ keV and $Z=79$ (Au).

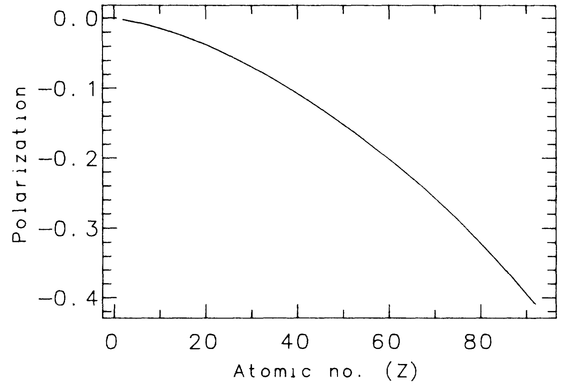

FIG. 2. Maximum value of the Sherman function as a function of atomic number Z for $E=40$ keV.

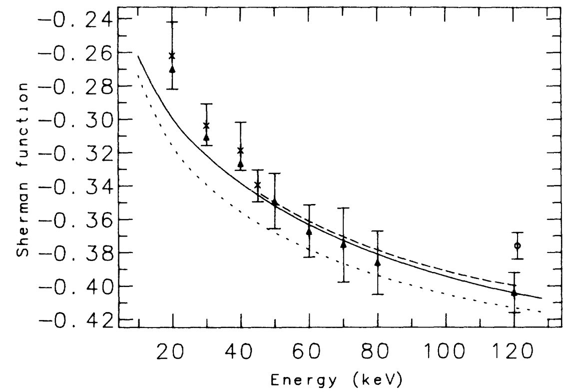

FIG. 3. The Sherman polarization function of Au at 120° as a function of energy. Solid line, present theory. Short-dashed line, calculation for a pure Coulomb potential using Sherman's method (Ref. 5). Long-dashed line, the calculations of Holzwarth and Meister. Δ, data from the cylindrical Mott analyzer of Ref. 6; ×, data from the spherical Mott analyzer of Ref. 6; ○, the data point of van Klinken, Ref. 14.

Also shown in the figure is the absolute data point of -0.376 ± 0.008 measured by van Klinken[14] at 121 keV obtained by double scattering. This value is quite low, probably due to the energy resolution and calibration problems inherent in such experiments.[3] Though incorporation of energy analyzers into Mott detectors was a vast improvement, it is still necessary to consider elastic multiple scattering in the experiment of Gray *et al.*[4] in order to expect agreement between experiment and theory.

The new calculations approach the analytic values for a pure Coulomb field[5] at higher energies faster than those calculated by Holzwarth and Meister,[7] but the two theories agree within the experimental error limits. It is difficult to pinpoint the reasons for the deviation of these two theories from each other, but it is clear that the computer technology of 1964 did not allow the high partial-wave and fine step-size analyses now available; similar shortcomings have to be expected in the potential functions of those days. However, it is worth mentioning that we have verified that the use of the potential for $Z=80$ multiplied by 79/80 as the potential for $Z=79$ by Holzwarth and Meister does not cause a significant error in the polarization (less than 0.04% for scattering at 120°). It is hard to speculate further as to why their calculations differ from the new ones, but we are certain that we have accounted for all errors due to the choice of the integration step size to better than 0.5%.

The spin polarization produced by high-energy scattering has two limits. For light Z and small scattering angle $S(\theta,\phi)$ is zero. For high Z, high energy, and large scattering angle, the polarization is given by LS coupling in the pure Coulombic field. In Fig. 3, the high-energy limit of the bare nuclear field is shown. The difference between this curve and the other theoretical curves is due to screening by the s electrons of the atom. In Fig. 4, the difference between all of the curves and the Coulomb

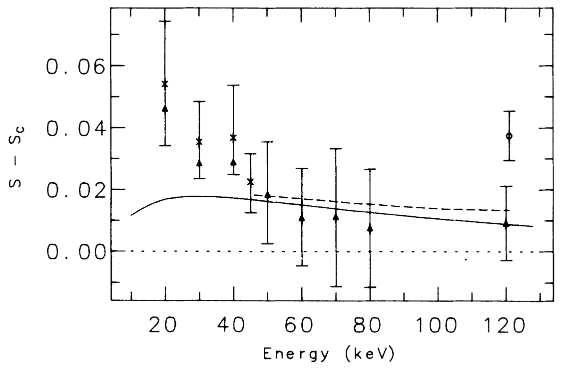

FIG. 4. The same legend as Fig. 3 except that the Coulomb potential results have been subtracted from all of the data points to point out how small the effect of adding the shielding electrons to the calculation is compared to the experimental uncertainty.

curve is shown. As expected, the difference function decreases toward zero for higher energies since screening contributes less to the scattering potential. The comparison with the experimental results of van Klinken[14] is particularly interesting. If the experimental result is correct, then the atomic wave function which describes the charge density in the atom must be severely in error. This is of significance since the electron charge density has its only maximum at the nuclear position. A modification to the extent needed to bring experiment and theory into agreement is far outside the range of uncertainty of Hartree-Fock theory. Another possibility rests with the breakdown of potential theory at small impact parameters. This would also be a very unexpected conclusion. As noted before, several possible arguments have been given which raise questions about the experiment. However, the Mott analyzer used in these measurements has confined for several elements and energies that the spin polarization of electrons from radioactive β decay is $-v/c$, thus confirming to better than 1% the electroweak V-A theory and the two component neutrino description.

The description of $\Delta S(\theta,\phi)$ of Fig. 4 shows that there is little hope of modifying any of the present theories to reach agreement with the van Klinken value. However, if in the future the experimental values and the theory ever agree, significant inferences can be drawn about the weak-interaction description.

ACKNOWLEDGMENTS

This work was supported by the R. A. Welch Foundation and by the National Science Foundation under Grant No. CHE-83-07174. Thanks go to L. G. Gray, F. B. Dunning, and G. K. Walters of Rice University for their valuable comments and suggestions.

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