Spin Dependence in Superelastic Electron Scattering from Na($3P$)

J. J. McClelland, M. H. Kelley, and R. J. Celotta
Radiation Physics Division, National Bureau of Standards, Gaithersburg, Maryland 20899
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Measurements are presented of spin asymmetries for superelastic scattering of 10-eV spin-polarized electrons from the excited Na($3P_{3/2}$) state created by linearly polarized laser optical pumping. Asymmetries as large as 16% are observed in scattering from a state which is not spin polarized. Results are shown both as a function of scattering angle with fixed laser polarization direction, and as a function of the laser polarization direction at a fixed scattering angle.

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In the past, observations of superelastic electron scattering from Na($3P$)[1] and Ba($6s6p\,^1P_1$)[2] have demonstrated a powerful method for the investigation of electron-atom collisions. These experiments have shown that the use of state selection for the target atoms allows new details of the collision process to be extracted. In particular, one can study the important role played by the target angular momentum in determining the outcome of the scattering process. By using different polarizations of the laser light for preparation of the initial atomic excited state, various mixtures of the target angular momentum states are created.[3] The interaction between these angular momentum states and the orbital angular momentum of the scattered electron leads to rather large effects in the cross section, such as a significant left-right asymmetry in the scattering intensity when circularly polarized excitation is used.[4]

In this Letter we present first results from the next generation of this type of experiment, the scattering of spin-polarized electrons from state-selected atoms. By studying the spin dependence of the superelastic cross section from an alkali-metal atom, the possibilities are opened for the investigation of not only the interaction of orbital angular momenta, but also the roles played by exchange and spin-orbit coupling in this interaction.

Elementary considerations of exchange and spin-orbit coupling[5] lead one to certain conclusions about when a spin-dependent effect should be observable, and how it should behave. For example, one might think that for the exchange interaction to cause a difference between the cross sections for spin "up" and spin "down" incident electrons, it would be necessary to have a spin-polarized target. Furthermore, one would expect any such exchange effect to be symmetric in scattering angle. On the other hand, the spin-orbit interaction should give a spin asymmetry which does not require a spin-polarized target and is antisymmetric in scattering angle. The asymmetry, though, should become negligible for low incident electron energy, small scattering angle, and small target Z.

As the measurements presented in this Letter show, these statements are in general not true for inelastic or superelastic scattering processes. Our observations show a spin-dependent effect which has behavior suggesting continuum spin-orbit coupling, as in Mott scattering,[5] despite the fact that the incident energy (10 eV), target $Z(11)$, and scattering angle (0°–35°) are all small. In fact, as will be discussed below, the cause of the spin dependence is the exchange interaction, although it requires the simultaneous presence of two effects: the correlation between orbital angular momentum and spin in an L-S coupled atom, and the interaction of the orbital angular momenta of the scattered and target electrons. Hanne[6,7] has discussed some aspects of this process and has labeled it the "fine-structure effect."

The experimental results presented here were obtained with a recently developed apparatus for measuring spin-dependent cross sections from polarized alkali-metal atoms.[8] This apparatus will be described fully in a forthcoming publication. A schematic of the experimental geometry is shown in Fig. 1. Polarized electrons are produced[9] by photoemission from a negative electron affinity GaAs photocathode and transported through electron optics to the scattering center, where they form a 2-mm diameter, 10-eV, 1-μA beam with an energy spread of typically 0.1 eV. This beam intersects an atomic sodium beam of density 10^{10} atoms/cm^3 and diameter 4 mm produced by an effusive oven. The scattering region is illuminated with light from a frequency-stabilized, single-frequency ring dye laser locked to the Na $2S_{1/2}(F=2) \rightarrow 3P_{3/2}(F=3)$ transition, creating a significant population of sodium atoms in the first excited state. The laser light is linearly polarized, and is incident perpendicular to the scattering plane from the $+z$ direction. Superelastically scattered electrons, which have gained the 2.1-eV Na excitation energy, are detected with a channel electron multiplier mounted on a rotatable turntable. The detector includes a retarding-field analyzer with resolution (approximately 0.5 eV) sufficient to reject all elastically or inelastically scattered

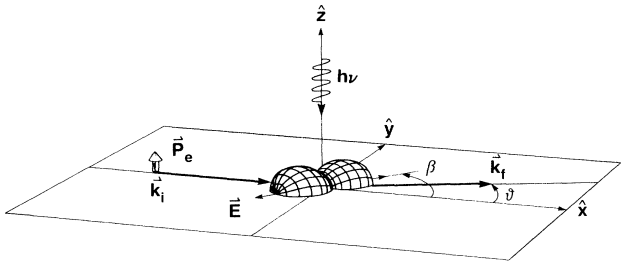

FIG. 1. Schematic of the scattering geometry, showing a representation of the charge density of the prepared $3P$ state. Electrons with spin polarization $\mathbf{P}_e$ perpendicular to the scattering plane are incident with momentum $\mathbf{k}_i$, and scatter into an angle θ with momentum $\mathbf{k}_f$. The initial atomic state is prepared with linearly polarized laser light incident perpendicular to the scattering plane. The electric vector $\mathbf{E}$ of the light makes an angle β with the incident electron direction.

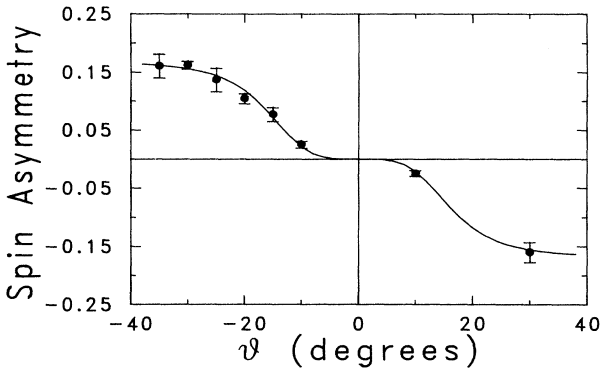

FIG. 2. Spin asymmetry $(N_\downarrow - N_\uparrow)/(N_\downarrow + N_\uparrow)$ in superelastic scattering as a function of scattering angle θ. The laser is linearly polarized parallel to the incident electron direction ($\beta = 0$). Positive scattering angle corresponds to scattering to the left. An antisymmetric curve is drawn to guide the eye.

electrons. The electron spin is modulated at 100 Hz by changing the helicity of the circularly polarized photoemission light with a Pockels cell driven by a high-voltage bipolar square wave. Electrons are counted separately for each spin orientation, and after averaging over ten measurements of 200 seconds each, the spin asymmetry is calculated from $P_e^{-1}(N_\downarrow - N_\uparrow)/(N_\downarrow + N_\uparrow)$, P_e being the magnitude of the incident electron polarization (26%) and N being the number of electrons detected when the incident electrons have spin up ($\uparrow$) or down ($\downarrow$) with respect to the laboratory frame. Typical superelastic counting rates range from 300 Hz at small angles to 20 Hz at large angles. One-standard-deviation error bars are derived from the reproducibility of these measurements and agree very well with error bars predicted by counting statistics. Not shown is an overall systematic uncertainty in the vertical scale of $\pm 6\%$ (1σ) of the asymmetry value, resulting from the uncertainty in electron polarization measurement.[10]

Figure 2 shows the measured angular dependence of the spin asymmetry for positive and negative scattering angles, where positive corresponds to scattering to the left. These data seem to violate one's intuition concerning the nature of the spin effects which should arise as a result of the exchange interaction in scattering from sodium. An atom excited by linearly polarized light can have no net orientation or spin polarization, since equal amounts of positive and negative magnetic sublevels in the excited state are created. Hence the expectation would be that there be no spin asymmetry in the cross section. The experiment shows quite the opposite. In fact, the observed spin asymmetry is antisymmetric in scattering angle, which might lead one to ascribe the asymmetry not to the exchange interaction, but to the spin-orbit interaction.

Next we show, in Fig. 3, the effect on the spin asymmetry of changing the laser polarization direction with respect to the incident electron direction. The resulting variation in the asymmetry is somewhat surprising when one considers what effect a rotation of the electric vector has on the excited atomic wave function. In geometric terms, this corresponds to causing the dumbbell-shaped orbital representing the excited state to rotate in the scattering plane. Quantum mechanically this is described by a change in the relative quantum phases, but no change in the relative amplitudes, of the magnetic sublevels of the excited state (the quantization axis is the $+z$ direction, which is antiparallel to the incident laser direction). With either description it is difficult to visualize how the spin asymmetry is sensitive to such changes in the excited-state wave function.

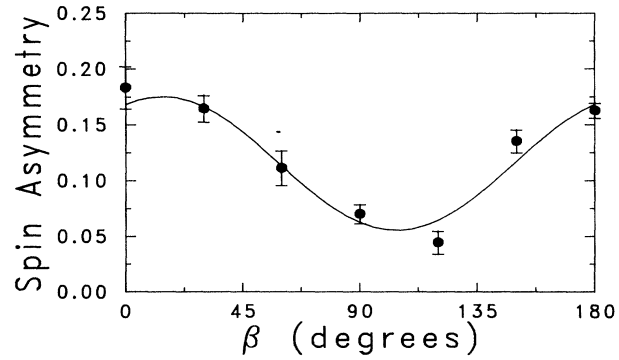

FIG. 3. Spin asymmetry $(N_\downarrow - N_\uparrow)/(N_\downarrow + N_\uparrow)$ in superelastic scattering vs laser polarization angle β. The scattering angle is $-30°$. $\beta = 0$ corresponds to the incident electron direction. The curve is a least-squares fit of the function $A + B\cos(2\beta + C)$. The parameters A, B, and C have the values 0.115 ± 0.006, 0.060 ± 0.008, and $-28° \pm 9°$.

As suggested above, the apparent paradoxes presented by the data can be resolved when one considers not only the interaction of the incident spin with the spin of the target or with its own orbital angular momentum, but also the interaction of the two orbital angular momenta when one of these (that of the atom) is coupled with its corresponding spin. Theoretically, this process can be analyzed in terms of complex scattering amplitudes and the density-matrix formalism,[6,11] provided proper assumptions are made when performing the angular momentum algebra. Reserving for a future publication a discussion of the mathematical details as they apply to our experiment, we concentrate at present on a physical explanation of the phenomena.

While consideration of the effects of hyperfine coupling in the atom is important for the details of the analysis, it is sufficient for present purposes to assume that the excited state is adequately described by a $J=\frac{3}{2}$ angular momentum wave function. In this case, transition probabilities for linearly polarized light lead to the conclusion that the excited state consists of $\frac{3}{8}$ each of the $M_J=\pm\frac{3}{2}$ and $\frac{1}{8}$ each of the $M_J=\pm\frac{1}{2}$ sublevels (recall the quantization direction is antiparallel to the incident light direction, perpendicular to the scattering plane). This distribution of levels indicates that superelastic scattering should be dominated by deexcitation of $M_J=\pm\frac{3}{2}$ states, which consist of either an $M_L=+1$ state coupled with an $M_S=+\frac{1}{2}$ spin wave function, or an $M_L=-1$ coupled with an $M_S=-\frac{1}{2}$.

It is well documented[12-14] that for an attractive scattering potential, the superelastic cross section for positive angles (scattering to the "left") is dominated by deexcitations of the $M_L=+1$ excited state, while for negative angles the bulk of the scattering is from the $M_L=-1$ state. This is what leads to the left-right–intensity asymmetry in superelastic scattering of unpolarized electrons from atoms excited with circularly polarized light. This being the case, we can say that when our spin-polarized electrons are scattered to the left they interact mostly with the $M_L=+1$ atoms, the majority of which are in the $M_J=+\frac{3}{2}$ state. There is, of course, some $M_L=+1$ in $M_J=+\frac{1}{2}$ as well, but that contribution to the scattering is less. Thus, the choice of scattering angle in effect selects a spin-polarized subset of the excited atoms. The exchange interaction acting on this subset leads to the observed spin asymmetry in the superelastic scattering, and explains why the effect is antisymmetric in scattering angle: Scattering to the left (right) interacts mostly with $M_L=+1$ (-1), which is correlated with $M_S=+\frac{1}{2}$ $(-\frac{1}{2})$. Hence the spin asymmetry changes sign in going from positive to negative angles.

In the discussion up to now, the emphasis has been on the fact that the scattering to one side is dominated by deexcitation of only positive M_J levels. There is, of course, some contribution from negative M_J levels as well, although exactly how much is a function of the incident electron energy amd scattering angle. The important point is that the contribution to the scattering amplitude from $M_J=+\frac{3}{2}$ is coherent with the contribution from $M_J=-\frac{1}{2}$, since the linearly polarized excitation light creates a coherent superposition of these two states. Thus the scattering cross section will have an interference term reflecting the phase correlation between the two states $M_J=-\frac{1}{2}$ and $+\frac{3}{2}$. The spin asymmetry will have a similar interference term because $M_J=-\frac{1}{2}$ is mostly composed of $M_S=-\frac{1}{2}$, while $M_J=+\frac{3}{2}$ has $M_S=+\frac{1}{2}$ as its spin component. Now we note that when the electric vector of the linearly polarized light is rotated by an angle β, an M_J-state amplitude is multiplied by the factor $\exp(iM_J\beta)$. Hence the phase difference between $M_J=-\frac{1}{2}$ and $+\frac{3}{2}$ is changed by 2β, causing the interference term to vary as a function of β with a period of π. This interference is the origin of the angular dependence of the curve shown in Fig. 3. Such a measurement can give previously inaccessible information about the relative phases of the scattering amplitudes.

We have presented the first experimental results for superelastic scattering of polarized electrons from sodium. The presence of a large spin-polarization effect when the target is unpolarized has been demonstrated, a situation which is counterintuitive if one considers the coupling of the spins of the incident and target electrons into singlet and triplet channels as the sole source of a spin asymmetry. Although the left-right antisymmetry of the effect suggests the spin-orbit interaction, this interaction in fact has little to do with the effect.

This type of measurement shows that when state selection in electron-atom collisions is extended to include the spin of the incoming electron, new and interesting effects surface. Not only are more stringent tests made available for *ab initio* calculations of the scattering process, but also a more carefully considered analysis of the interaction becomes necessary. It is hoped that in the future more work in this area will result in a further enhancement of our basic understanding of the collision process.

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