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Spin-exchange cross section for electron excitation of Na $3S$-$3P$ determined by a novel spectroscopic technique

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An experimental technique is described which enables determination of the partial cross sections for electron excitation of atoms as a function of changes in spin and orbital angular momentum. This method provides a good signal-to-noise ratio in the energy region near threshold, and could be used to study long- or short-lived excited states of many atomic systems. Measurements for Na $3S$-$3P$, near-threshold excitation are reported here. The results are generally in good agreement with the close-coupling calculations of D. L. Moores and D. W. Norcross [J. Phys. B **5**, 1482 (1972)] for the largest Δm_S and Δm_L components of the cross section, but not for the component with $|\Delta m_S| = 1$ and $|\Delta m_L| = 1$, indicating that the exchange interaction is larger than previously recognized.

A measurement of all observables in an electron-atom collision constitutes the so-called "complete" scattering experiment, as first enunciated by Bederson.[1] In the case of excitation of the first resonance level ($3S$-$3P$) of sodium, seven parameters, representing the magnitudes and phases of the four scattering amplitudes, must be measured as a function of electron scattering angle for each electron energy in order that the fullest possible information be obtained.

Such a "complete" experiment has not yet been performed, but several groups have measured various combinations of these parameters.[2] Here we obtain a different combination; specifically, we measure the partial (angle-integrated) cross sections for excitation of Na versus changes in m_S and m_L of the atomic electron. This is done using a novel experimental technique for several electron collision energies between 2 and 4 eV.

The principle behind the experiment is shown in Fig. 1. Atoms are initially prepared in a pure m_S', m_l' level of the $3S$ state, and then electron-excited to Zeeman-split $3P_{3/2}(m_J)$ levels. The resulting relative populations of these four levels are measured by tuning through the $3P_{3/2}(m_J)$-$5S_{1/2}(m_S'')$ transitions with a cw dye laser (designated v_2), and detecting the $4P$-$3S$, uv cascade-fluorescence signal from the $5S$ state. The $4P$ fluorescence originating from each $5S(m_S'')$ state, although isotropic, is circularly polarized. The optics do not distinguish between different circular polarizations; hence the cascade signal is proportional to the $5S(m_S'')$ population. The v_2 absorption from each $3P_{3/2}(m_J)$ to $5S(m_S'')$ transition is well resolved in a 220-G magnetic field, as seen in a representative scan shown in Fig. 2. The area under each peak is directly related to the population of the corresponding $3P_{3/2}(m_J)$ level; this proportionality depends upon the v_2 laser power, as will be discussed in more detail later. Since these $3P_{3/2}(m_J)$ populations are proportional to the $3S_{1/2}(m_S') \to 3P_{3/2}(m_J)$ excitation cross sections, the data yield the ratios of the $3S(m_S') \to 3P(m_S,m_L)$ cross sections.

The experimental arrangement is shown diagrammatically in Fig. 3. A supersonic beam of sodium atoms from a recirculating oven is collimated by a slit and state-selected into the $m_S' = \frac{1}{2}$, $m_l' = \frac{3}{2}$ state by optical pumping with a cw dye laser (designated v_1) in a $\approx$5-G magnetic field. The spins remain aligned with the z-directed magnetic field as the atoms move upward into the 220-G field of the collision region. There some of the atoms are excited to the $3P$ state first by the z-propagating electron beam, and some of these are further excited to the $5S$ state by the overlapping, counter propagating v_2 laser beam.

The 2.5-mm diam electron beam is produced by an ~1000°C oxide cathode, yielding an energy width of
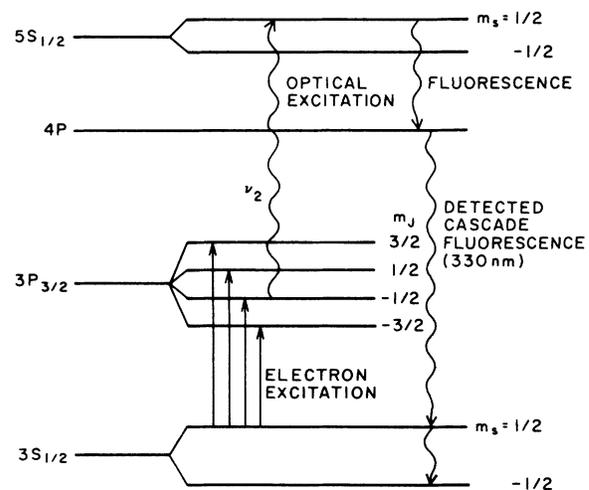

FIG. 1. Na atoms are excited from $3S_{1/2}(m_S' = \frac{1}{2})$ to the Zeeman-split m_J levels of the $3P_{3/12}$ state, and the relative populations in these levels are probed by tuning a laser to excite from $3P_{3/2}$ to $5S_{1/2}$ and observing the resultant $4P$ cascade fluorescence.
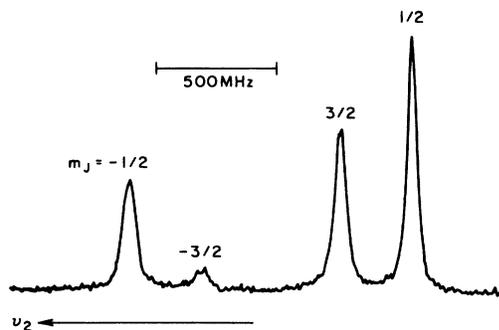
FIG. 2. Detected cascade fluorescence as a function of v_2 laser tuning through the $3P_{3/2}$-$5S_{1/2}$ transitions for a mean electron energy of 3.35 eV. Each of the four peaks is labelled by the m_J value of the $3P_{3/2}$ state being probed (see Fig. 5). The data points are separated by 4 MHz, and the integration time per point is 6 s.

~0.3 eV. The electrons are gradually drawn out by several accelerating plates to minimize transverse electric fields that could induce spiraling. The current ($\lesssim 3$ μA) is space-charge limited at the cathode and kept low enough for space-charge depression of the electron energy to be $\lesssim 0.1$ eV.

About 20% of the 330-nm $4P$ cascade fluorescence is reflected by a spherical mirror into a quartz lens, and then into a bialkali-metal photomultiplier. Glass filters block the (590 nm) $3P$ fluorescence and transmit the (330 nm) $4P$ fluorescence. The signals are collected by a data acquisition system, which in turn is interfaced to a microcomputer which processes the data.

The optical pumping is performed in a weak magnetic field using circularly-polarized v_1-laser light (~0.5 W/cm^2) tuned simultaneously to both hyperfine levels of the $3S_{1/2}$ state. This is done with first-order sidebands produced in a 886 MHz rf phase-modulated electrooptic crys-
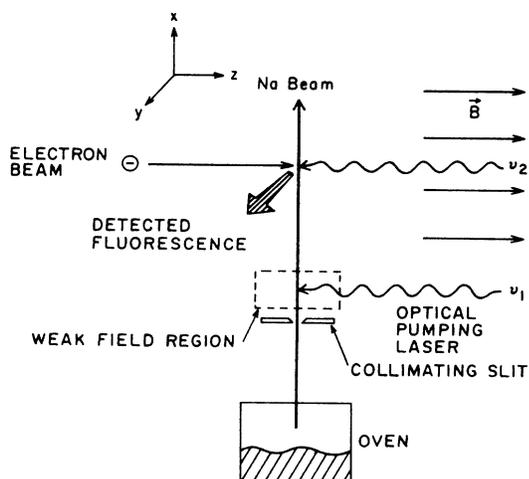

FIG. 3. Sketch of the basic experimental setup. The optical pumping of the sodium beam occurs in a ~5 G field, while the electron-atom collisions occur in a field of $B \approx 220$ G.

tal,[3] so that the sidebands are approximately 1772 MHz apart. Pumping is carried out on the $3S$-$3P_{1/2}$ transitions in order that photons from $3P_{1/2}$ fluorescence in the optical pumping region will not excite the $3P_{3/2}$ state in the interaction region. In this experiment ~96% of the population is optically pumped to $3S_{1/2}(m_S = \frac{1}{2})$.

Radiation trapping could in principle affect our cross-section results, as photons emitted from electron-excited $3P_{3/2}$ states could excite another atom in its ground state to a different $3P_{3/2}(m_J)$ level. This would mimic electron excitation of this level, and lead to erroneous partial cross sections. However, after taking into account the narrow residual Doppler widths in the beam, the frequencies and polarization properties of the photons emitted and subsequently reabsorbed along directions where the atom beam has significant extent, and the fact that the $3P_{3/2}(m_J = -\frac{3}{2})$ state cannot be excited by a dipole allowed transition from the $3S(m_S = \frac{1}{2})$ state, we conclude that radiation trapping is not significant for the Na densities and beam widths used in this experiment.

As the v_2 laser is scanned, it excites four, resolved $3P_{3/2}$-$5S_{1/2}$ Zeeman transitions as shown in Fig. 2. The area under each peak is proportional to the electron-excited population in the corresponding m_J levels of $3P_{3/2}$, but the transition strengths for $3P_{3/2}$-$5S_{1/2}$ transitions originating from $|m_J| = \frac{1}{2}$ differ from those starting from $|m_J| = \frac{3}{2}$, so the proportionality constant is also different. In the limit of low v_2 laser power (P), the probability that an atom electron-excited to $|m_J| = \frac{3}{2}$ emits a $4p$-state fluorescence photon is three times that of an atom excited to $|m_J| = \frac{1}{2}$, i.e., proportional to the ratio of transition strengths for $3P_{3/2}(|m_J|)$-$5S_{1/2}(m_S'')$ transitions. However, an optical pumping process out of the $3P_{3/2}(m_J)$ state makes this ratio approximately unity in the high-power limit for v_2 on-resonance. The full power dependence for each $|m_J|$ has been calculated, studied experimentally, and used to adjust the measured area ratios. The v_2 beam intensity is typically ~0.7 W/cm^2.

To obtain the $3S_{1/2}(m_S')$-$3P_{3/2}(m_J)$ excitation cross sections, a correction is made for the imperfect $3S$ state spin selection. The ~4% of the beam in the $3S(m_S = -\frac{1}{2})$ state has the effect of increasing the apparent populations in the $m_J = \frac{1}{2}$, $-\frac{1}{2}$, and $-\frac{3}{2}$ states by typically 1%, 2.5%, and 0.5%, respectively.

These cross sections for electron excitation directly to $3P$ at energies above the (3.2 eV) $4S$ threshold must also be corrected for the cascade contributions from $4S$. The extent to which this augments each $3P_{3/2}(m_J)$ population depends upon the cross sections for direct and exchange excitation from the $3S$ to $4S$ state. Mitroy[4] has recently calculated the $3S$-$4S$ cross sections using a four-state close-coupling calculation, and from his values, about 30% of the $3S(m_S' = \frac{1}{2}) \rightarrow 3P_{3/2}(m_J = -\frac{3}{2})$ cross section at 3.35 eV arises from $4S$ cascading. The other three $3S(m_S' = \frac{1}{2}) \rightarrow 3P_{3/2}(m_J)$ cross sections at the same energy are much larger and have contributions of less than 4% due to $4S$ cascading.

Each of the $3P_{3/2}(m_J)$ levels is a linear combination of m_S and m_L states, as shown in Fig. 4. (For clarity in this figure we show the $3P_{3/2}$ states for full $I \cdot J$ decoupling; the ~1% deviation from this at 220 G is taken into account in
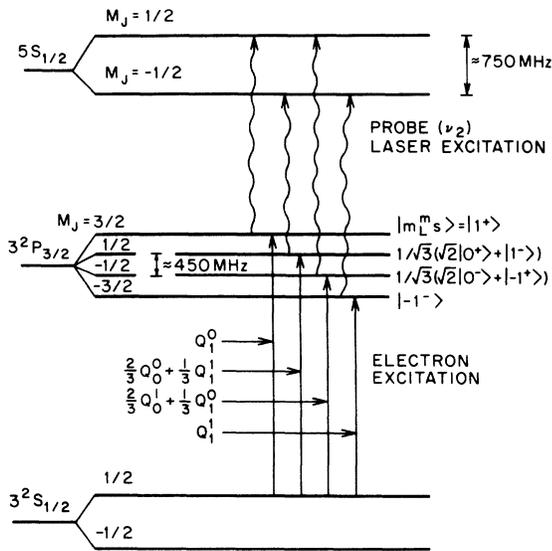
FIG. 4. Energy levels and states of Na involved in this experiment. The $3P_{3/2}(m_J)$ levels are decomposed into their m_S and m_L components, and the $Q\genfrac{|}{|}{0pt}{}{\Delta m_S}{\Delta m_L}$ cross section relevant for excitation to each m_J is indicated.

FIG. 5. Partial cross sections $Q\genfrac{|}{|}{0pt}{}{\Delta m_S}{\Delta m_L}$ obtained from this work (symbols), compared with the calculated values of Moores and Norcross (solid dots connected by line segments).

the data reduction.) The cross sections for excitation to these m_S and m_L states is given by $Q_{\Delta m_L}^{\Delta m_S}$, where $\Delta m_S = m_S - m'_S$ and $\Delta m_L = m_L$. Δm_S and Δm_L each can be 1, 0, or -1, giving nine possible such Q's. However, by symmetry, a change in the sign of both Δm_S and Δm_L will not change Q, and in the absence of an explicit spin-dependent force, a change in the sign of either Δm_S or Δm_L will not change Q. Consequently, there are only four independent Q, corresponding to $|\Delta m_S| = 1$ or 0 and $|\Delta m_L| = 1$ or 0, which we label $Q\genfrac{|}{|}{0pt}{}{\Delta m_S}{\Delta m_L}$. Cross sections for excitation to a particular m_J are given by

$$Q(3S,m'_S \rightarrow 3P_J,m_J) = \sum_{m_L,m_S} [C(J,m_J;m_L,m_S)]^2 Q\genfrac{|}{|}{0pt}{}{\Delta m_S}{\Delta m_L},$$

where $C(J,m_J;m_L,m_S)$ is the appropriate Clebsch-Gordan coefficient. Thus the $Q\genfrac{|}{|}{0pt}{}{\Delta m_S}{\Delta m_L}$ are easily obtained from the data, and the results are presented in this form.[5]

The measured partial cross sections (uncorrected for $4S$ cascade and electron spiraling effects) have been normalized so that their sum $(Q_{tot} = Q_0^0 + Q_0^1 + 2Q_1^0 + 2Q_1^1)$ agrees with the total cross-section data of several workers.[6] The resulting absolute partial cross sections are presented in Fig. 5, along with the same cross sections deduced from the four-state close-coupling calculations of Moores and Norcross.[7]

Electron spiraling can affect the observed partial cross sections, as the electron-atom collision does not occur along the quantization axis defined by the B field. For our low-energy data, this has the effect of increasing the apparent ratio of the cross sections with $|\Delta m_L| = 1$ compared to that with $\Delta m_L = 0$. By minimizing the ratio of $3P$-$3S$ fluorescence with polarization perpendicular versus parallel to the magnetic field direction, we determine the electron gun voltages that produce the minimum degree of spiraling. Voltage changes on the various accelerating
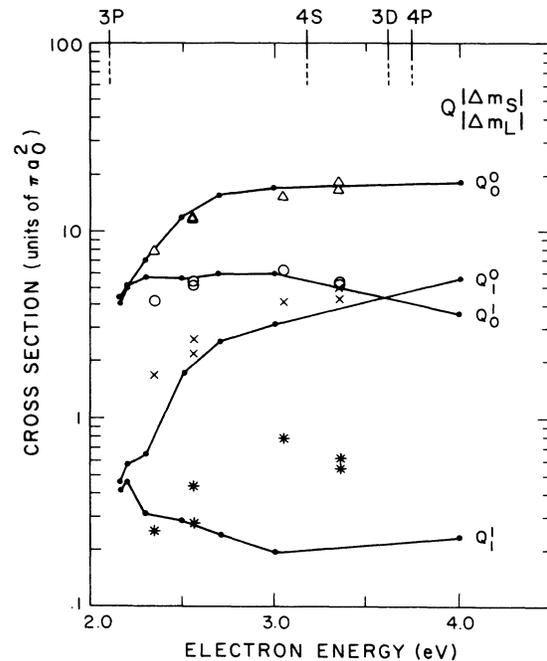
elements of $> 10\%$ from this optimum condition have no effect on the polarization ratio, indicating that a true minimum, limited only by the transverse thermal energy, has been achieved. This minimum spiraling condition occurs when the electrons are drawn out with a nearly constant electric field, as expected.

By comparing our measured ratio of $|\Delta m_L| = 1$ vs $\Delta m_L = 0$ excitation to that inferred from the polarization measurements of Enemark and Gallagher,[8] we estimate that spiraling effects are largest for the data within $\frac{1}{2}$ eV of threshold. For instance, this comparison suggests that spiraling accounts for $\sim 30\%$ of the Q_1^1 and $\sim 8\%$ of the Q_1^0 reported at 2.55 eV in Fig. 5.

The four quantities obtained in this experiment, after normalization to previous total cross-section measurements, are the angle-integrated $Q\genfrac{|}{|}{0pt}{}{\Delta m_S}{\Delta m_L}$ cross sections Q_0^0, Q_0^1, Q_1^0, and Q_1^1, as a function of energy.[9] The $|\Delta m_S| = 1$ collisions take place via electron-spin exchange rather than direct spin flip, as the spin-orbit interaction responsible for spin flip is very weak in sodium, and is negligible compared to exchange at the low energies of this experiment. In the absence of electron-spin exchange, one would obtain $Q_0^1/Q_0^0 \approx 0$ and $Q_1^1/Q_1^0 \approx 0$. In fact, we typically measured $Q_0^1/Q_0^0 \sim 0.35$ and $Q_1^1/Q_1^0 \sim 0.1$, indicating that exchange plays a significant role in the excitation process.

Our data are in good agreement with the close-coupling calculation of Moores and Norcross[7] for the largest component, Q_0^0, as well as with Q_0^1 at all but the near-threshold point. There is $\sim 30\%$ discrepancy for Q_1^0 except at this same near-threshold value. Here the ~ 0.3 eV electron-energy width may account for most of the unusu-

ally large disagreement at 2.35 eV, where the theoretical calculations indicate a rapidly increasing Q_1^0 cross section; i.e., this electron-energy spread, combined with the rapidly increasing Q_1^0, shifts the effective electron energy to a higher value. Q_1^1, on the other hand, agrees with the theory in the near-threshold region but differs by more than a factor of 2 at the higher measured energies. A detailed error analysis was not performed on our data, but it is clear from Fig. 5 that the random uncertainties are not excessively large, and we do not believe that electron spiraling, cascading, or any other systematic error is sufficient to explain these discrepancies.

In conclusion, a novel method has been used to measure the angle-integrated Δm_S and Δm_L dependences for electron excitation of the $3P$ state of sodium, at various electron energies between the $3P$ and $4P$ thresholds. We have demonstrated that this method can give an excellent signal-to-noise ratio even in the difficult threshold energy region. Significant discrepancies with the most complete

published theory for the Na $3S$-$3P$ excitation Q_1^1 cross section have been found, indicating amongst other things that the exchange interaction plays a larger role than indicated by this calculation.

It is important to note that application of this basic technique is not restricted to sodium or to short-lived excited states; in principle it can be applied to any atom to determine the partial cross sections for impact excitation. Of course, in practice one is often limited by higher-level cascades and unfavorable radiative branchings, as well as practical problems such as laser sources in the necessary spectral region and the necessity to produce, pump and polarize the atomic beam.

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[7] D. L. Moores and D. W. Norcross, J. Phys. B 5, 1482 (1972).

[8] E. A. Enemark and A. Gallagher, Phys. Rev. A 6, 192 (1972).

[9] In terms of $f_{|\Delta m_L|}(\theta)$ and $g_{|\Delta m_L|}(\theta)$, the amplitudes for $3S$-$3P$ direct and exchange excitation with $|\Delta m_L|$, the partial cross sections may be written as [see J. Kessler, Polarized Electrons, 2nd ed. (Springer-Verlag, Berlin, 1985)]:

$$Q_0^0 = \tfrac{1}{2}(k_f/k_i)\int [\,|f_0(\theta)|^2 + |f_0(\theta) - g_0(\theta)|^2]\,d\Omega\,,$$

$$Q_0^1 = \tfrac{1}{2}(k_f/k_i)\int |g_0(\theta)|^2\,d\Omega\,,$$

$$Q_1^0 = \tfrac{1}{2}(k_f/k_i)\int [\,|f_1(\theta) - g_1(\theta)|^2 + |f_1(\theta)|^2]\,d\Omega\,,$$

$$Q_1^1 = \tfrac{1}{2}(k_f/k_i)\int |g_1(\theta)|^2\,d\Omega\,,$$

where k_f and k_i are the final and initial electron momenta, respectively, and the factor of $\tfrac{1}{2}$ arises since the electron beam is initially unpolarized.