Sodium excitation by spin-polarized electrons: A reanalysis of existing experiments

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We generalize the existing description of collision-induced atomic $S \leftrightarrow P$ transitions by spin-polarized electrons on spin-polarized target atoms. Analysis of existing experimental and theoretical data on the $3s \leftrightarrow 3p$ transitions in Na determines how closely the "perfect scattering experiment" has been achieved to date. Using a newly developed inversion technique, we demonstrate to what extent the various scattering amplitudes can be extracted from present experiments, and how state-of-the-art theory can be used to remove the remaining ambiguities.

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Almost 25 years ago Bederson [1–3] coined and discussed the term "perfect scattering experiment." By this he meant the experimental determination of a complete set of quantities fully characterizing the set of quantum-mechanical scattering amplitudes, using elastic [1] and inelastic [2] electron–alkali-metal-atom collisions as prototypes. He later discussed in detail $S \to P$ excitation with spin analysis of the electron and/or atom before and/or after the collision [3]. While significant progress has been made both experimentally and theoretically, the "perfect scattering experiment" for this class of simple collision events has not yet been fully realized.

In this report, however, we demonstrate that the goal has actually been reached to a much larger extent than is generally assumed; in fact, *present experimental setups*, together with an improved *coordination* of efforts allow for a complete determination of all the relevant scattering amplitudes, except for a few remaining ambiguities termed "ghost solutions" below. We present a newly developed inversion technique to reanalyze and discuss the existing experimental data for e-Na ($3s \leftrightarrow 3p$) scattering, the most thoroughly investigated system to date. In a final step, using also state-of-the-art theory, this technique allows us to identify unambiguously a complete set of parameters from an inversion of experimental data.

For light alkali-metal atoms, $S \to P$ excitation may be fully described by four complex scattering amplitudes, two for triplet (t) and two for singlet (s) scattering. This assumes that explicitly spin-dependent forces, such as the spin-orbit interaction between projectile and target, may be neglected. If we choose a coordinate system (x^n, y^n, z^n) fixed by $\vec{k}_{\rm in} \parallel x^n$ and quantization axis $z^n \parallel \vec{k}_{\rm in} \times \vec{k}_{\rm out}$ perpendicular to the scattering plane, parity conservation implies that only the (complex) amplitudes $f^{s,t}_{+1}$ and $f^{s,t}_{-1}$ (the subscript denotes the orbital magnetic quantum number) are nonzero. Neglecting an overall phase, we see that seven independent parameters are needed for each scattering angle θ to characterize the amplitudes completely. Besides the *absolute* differential cross section corresponding to unpolarized particles, six dimensionless parameters may be defined, three to characterize the relative lengths of the four amplitude vectors, and three to define their relative phase angles.

Following the formalism developed for unpolarized collision partners in Ref. [4], we can choose these parameters as follows. We begin, for each total spin channel individually, with the angular momentum transfer

$$L_\perp^{s,t} = \frac{|f^{s,t}_{+1}|^2 - |f^{s,t}_{-1}|^2}{|f^{s,t}_{+1}|^2 + |f^{s,t}_{-1}|^2} \tag{1}$$

and the alignment angle $\gamma^{s,t}$ of the major symmetry axis of the charge cloud in the scattering plane. This is related to the phase difference $\delta^{s,t} \equiv \arg[f^{s,t}_{+1} - f^{s,t}_{-1}]$ through

$$\delta^{s,t} = -2\,\gamma^{s,t} + \pi. \tag{2}$$

These facts may be expressed compactly through the density matrix formalism. For unpolarized beams, the density matrix becomes the weighted sum of two matrices, ρ^s and ρ^t, i.e.,

$$\begin{aligned}\rho_u &= Q\,\frac{1}{2}\begin{pmatrix} 1+L_\perp & 0 & -P_\ell\, e^{-2i\gamma} \\ 0 & 0 & 0 \\ -P_\ell\, e^{2i\gamma} & 0 & 1-L_\perp \end{pmatrix} \\ &= \frac{1}{3r+1}\,Q\,\frac{1}{2}\begin{pmatrix} 1+L^s_\perp & 0 & -P^s_\ell\, e^{-2i\gamma^s} \\ 0 & 0 & 0 \\ -P^s_\ell\, e^{2i\gamma^s} & 0 & 1-L^s_\perp \end{pmatrix} \\ &\quad + \frac{3r}{3r+1}\,Q\,\frac{1}{2}\begin{pmatrix} 1+L^t_\perp & 0 & -P^t_\ell\, e^{-2i\gamma^t} \\ 0 & 0 & 0 \\ -P^t_\ell\, e^{2i\gamma^t} & 0 & 1-L^t_\perp \end{pmatrix}\end{aligned} \tag{3}$$

where $r = Q^t/Q^s$ is the ratio of the differential cross sections for triplet and singlet scattering, $P^{s,t}_\ell$ is the magnitude of the linear light polarization, and $Q = \frac{1}{4}Q^s + \frac{3}{4}Q^t$ is the differential cross section.

The above parameters can be measured either through electron-photon coincidence techniques or by superelastic scattering (deexcitation) from laser excited atoms [4]. In the following we will assume that all corrections to account for fine- and hyperfine-structure depolarization effects have been applied, i.e., the parameters corresponding to the nascent charge cloud have been recovered.

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Before we discuss individual experiments, it seems appropriate to list a possible set of parameters that (i) allow for a complete description of the scattering process; (ii) are accessible in "noncomplete" experiments; (iii) can be interpreted in simple physical pictures; and (iv) are a natural generalization of the parameters used for unpolarized beams.

Following [4,5], we suggest the following complete set:

$$Q, L_\perp^s, L_\perp^t, \gamma^s, \gamma^t, r, \Delta^{+1}. \tag{4}$$

It should be noted that the singlet-triplet phase angles $\Delta^{+1} \equiv \arg[f_{+1}^t - f_{+1}^s]$ and $\Delta^{-1} \equiv \arg[f_{-1}^t - f_{-1}^s]$ are not independent, but are related through

$$\Delta^{+1} - \Delta^{-1} = \delta^t - \delta^s = 2\,(\gamma^s - \gamma^t). \tag{5}$$

Over the past few years, McClelland and co-workers [6–9] have determined the angular momentum transfers $L_\perp^s$ and $L_\perp^t$ for the two spin channels individually, as well as the ratio r of triplet and singlet contributions to the differential cross section. This was achieved by scattering spin-polarized electrons [11] superelastically from spin-polarized sodium atoms which were produced by pumping with circularly polarized laser light.

Note that this experiment does not allow for a unique determination of the alignment angles γ^s and γ^t. This can be seen by comparing the off-diagonal elements of Eq. (3) which show that

$$P_\ell\, e^{2i\gamma} = \frac{1}{3r+1}\, P_\ell^s\, e^{2i\gamma^s} + \frac{3r}{3r+1} P_\ell^t\, e^{2i\gamma^t}, \tag{6}$$

where P_ℓ and γ are the linear polarization and the alignment angle for unpolarized beams, respectively. Note that, in general,

$$P_\ell^2 \le 1 - L_\perp^2, \tag{7}$$

while $P_\ell^{s,t} = \sqrt{1 - \left(L_\perp^{s,t}\right)^2}$ for the individual spin channels.

As illustrated in Fig. 1, the complex Eq. (6) corresponds to the addition of two vectors $\mathbf{P}_\ell^s$ and $\mathbf{P}_\ell^t$, multiplied by weighting factors $1/(3r+1)$ and $3r/(3r+1)$, respectively, to form the resulting vector $\mathbf{P}_\ell$. Hence, elementary geometry can be applied to obtain the *two* pairs of solutions $(\gamma^s, \gamma^t)_{\text{true}}$ (the true solution) and $(\gamma^s, \gamma^t)_{\text{ghost}}$ (the other possibility):

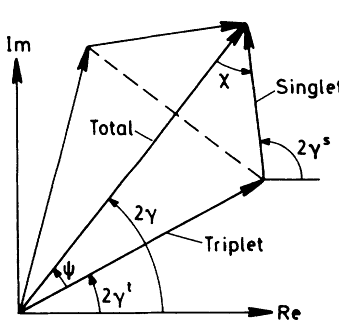

FIG. 1. Vector diagram corresponding to Eq. (6).

$$\gamma^s = \gamma \pm \psi/2, \tag{8}$$

$$\gamma^t = \gamma \mp \chi/2. \tag{9}$$

Hence, provided experimental data are available for the set of parameters $(P_\ell, \gamma, L_\perp^s, L_\perp^t, r)$ at a given collision energy and scattering angle, the two sets of possible angles (γ^s, γ^t) can be determined. However, this experiment contains *no information* about the missing phase angle Δ^{+1} relating singlet and triplet amplitudes.

Another recent experiment involving spin-polarized electrons in electron-sodium excitation was performed by Hegemann, Oberste-Vorth, Vogts, and Hanne [12] who measured the ratio $T \equiv P'/P$ of the final electron spin polarization P' and the initial spin polarization P after excitation of unpolarized sodium atoms by spin-polarized electrons. The interesting and, at least for inelastic collisions, so far neglected point associated with this experiment is the fact that information about relative phases between singlet and triplet amplitudes can be extracted, i.e., this experiment can complement the results of Kelley, McClelland, Lorentz, Scholten, and Celotta. This can be seen when the T parameter is expressed as

$$T = \frac{1}{3r+1}\Big[2r + \sqrt{r\,(1+L_\perp^s)\,(1+L_\perp^t)}\cos\Delta^{+1} + \sqrt{r\,(1-L_\perp^s)\,(1-L_\perp^t)}\cos\Delta^{-1}\Big]. \tag{10}$$

This equation reduces to $T = 2(r + \sqrt{r}\cos\Delta)/(3r+1)$ in the case of elastic S $\to$ S scattering by setting $L_\perp^{s,t} = 0$ and $\Delta^{+1} = \Delta^{-1} = \Delta$. This result (with different notations) was first derived as Eq. (4.40) of Kessler's book [11] and later by McClelland *et al.* [10].

The nonlinear Eq. (10) for the unknown phase angles Δ^{+1} and Δ^{-1} can be used in connection with Eq. (5) to narrow down the possible values of these parameters to four pairs, indicating that the "complete experiment" could almost be achieved with present experimental setups — except for a few ambiguities that can, for the most part, be resolved with state-of-the-art theory, as we now demonstrate.

To illustrate our points, we recall that it is possible to determine two sets of angles (γ^s, γ^t), provided experimental data are available for the set of parameters $(P_\ell, \gamma, L_\perp^s, L_\perp^t, r)$ at a given collision energy and scattering angle. While data for all these parameters have indeed been measured for electron-sodium (de-)excitation, the energy and scattering angle combinations investigated by Kelley, McClelland, Lorentz, Scholten, and Celotta [9] and Teubner and Scholten [13], unfortunately, do not overlap at all. Consequently, we demonstrate the feasibility of the approach by replacing the missing experimental data with the theoretical results from Bray's calculation [14]. This seems justified in light of the excellent agreement between theory and experiment for P_ℓ and γ at both 12.1 eV and at 22.1 eV total collision energy, as well as the very good agreement of the above theory with other measured parameters over the energy range from 1 eV to 40 eV [15].

The results of this inversion are shown in Fig. 2 for both angles γ^s and γ^t. The error bars on the experimental points were obtained by changing the theoretical results for the set of input parameters $(L_\perp^s, L_\perp^t, r)$ by a very small amount and looking at the effect on the inverted theoretical results for the two pairs of (γ^s, γ^t). This gives the partial derivatives of γ^s and γ^t with respect to these three input values and allows for the calculation of approximate error bars. As demonstrated in Fig. 2, both the true and the ghost solutions after inverting the experimental and the theoretical data are in very good agreement with each other. Hence, it seems entirely justified to select the "true" experimental data as those that follow the "true" theoretical solution.

A more difficult procedure is required to obtain the phase difference between singlet and triplet amplitudes by combining the results of the experiments by Kelley, McClelland, Lorentz, Scholten, and Celotta with those for the T parameter measured by Hegemann, Oberste-Vorth, Vogts, and Hanne [12]. Because of the different collision energies investigated by the various groups, we demonstrate the principle by using theoretical data for the contraction parameter $T = P'/P$ at 4.1 eV total collision energy where data for $(L_\perp^s, L_\perp^t, r)$ from Kelley, McClelland, Lorentz, Scholten, and Celotta are available [8,9]. Again, the very good agreement between theory and experiment for the T parameter at the energies measured experimentally (4.0 eV and 12.1 eV) gives us confidence in the results presented below.

The main idea of our inversion procedure is the following: Solutions of the nonlinear Eq. (10) for Δ^{+1} and Δ^{-1}, subject to their difference fixed through Eq. (5) can be found by searching for crossings between the lines determined by

$$A \cos\Delta^{+1} + B \cos\Delta^{-1} = C, \tag{11}$$

where the constants A, B, and C are evaluated from Eq. (10), and the lines defined by Eq. (5). Due to the ambiguity in the sign of the cosine arguments, as well as the ambiguity in the proper pair of (γ^s, γ^t), one will usually find four solutions, only one of which, however, is the correct one. Exploring further the symmetry of the problem [16], we have written a computer code to perform this inversion.

The technique was applied using experimental data for the set of parameters $(L_\perp^s, L_\perp^t, r)$ [8,9] to first construct experimental pairs of (γ^s, γ^t) and the corresponding difference lines. The results for Δ^{+1} and Δ^{-1} as a function of the scattering angle for a total collision energy of 4.1 eV are shown in Fig. 3. For simplicity, only one ghost solution is shown in these graphs, but it can be seen how the theoretical results help once more to distinguish, in most cases unambiguously, between the various possibilities that one would get from an inversion of experimental data alone.

In conclusion, we have demonstrated how results from several experimental groups can be combined to de-

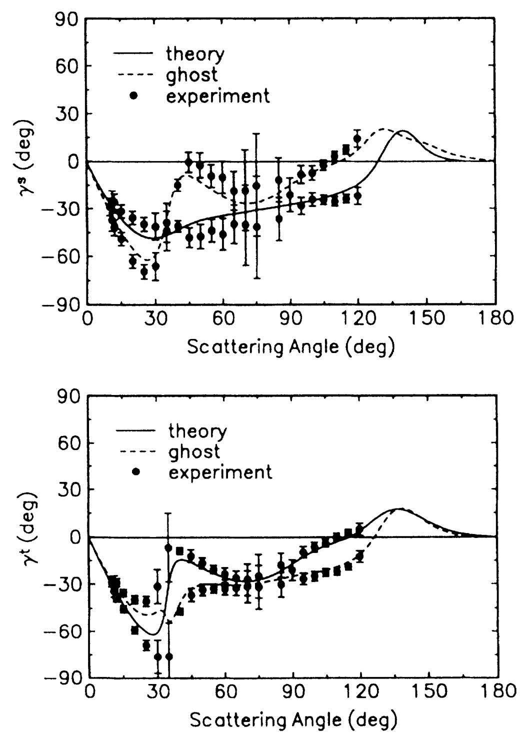

FIG. 2. Alignment angles for singlet and triplet channels calculated from the NIST (Ref. [9]) data for $(L_\perp^s,\ L_\perp^s,\ r)$ and theoretical results for P_ℓ and γ from scattering amplitudes of Bray (Ref. [14]) for electron impact excitation of the $(3p)^2P$ state of sodium at an incident electron energy of 4.1 eV; •, two sets of inverted experimental data as well as true (———) and ghost (— — —) theoretical solutions.

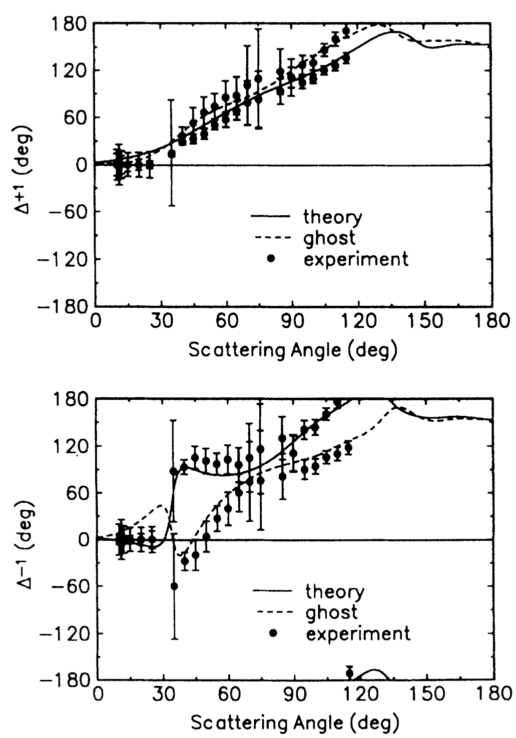

FIG. 3. Singlet-triplet phase angles Δ^{+1} and Δ^{-1} calculated from the NIST data (Ref. [9]) for $(L_\perp^s,\ L_\perp^s,\ r)$, the corresponding angles (γ^s, γ^t) and theoretical results for $T = P'/P$ from scattering amplitudes of Bray (Ref. [14]) for electron impact excitation of the $(3p)^2P$ state of sodium at an incident electron energy of 4.1 eV; •, two sets of inverted experimental data as well as the true (———) and one ghost (— — —) theoretical solutions.

termine the full set of seven independent parameters $(Q, L_\perp^s, L_\perp^t, \gamma^s, \gamma^t, r, \Delta^{+1})$, except for a "ghost pair" of alignment angles (γ^s, γ^t) and, consequently, three ghost pairs of singlet-triplet angles $(\Delta^{+1}, \Delta^{-1})_{\mathrm{ghost}}$. Furthermore, the use of state-of-the-art theory enabled an unambiguous identification of the true solutions, see Figs. 2 and 3.

Additional experiments to remove the remaining ambiguities will require an extension of the presently available setups, such as in-plane pumping in the experiment by Kelley, McClelland, Lorentz, Scholten, and Celotta, the determination of "generalized STU parameters" [17] for excitation of individual fine-structure levels, or the measurement of an electron polarization component after scattering in a direction normal to the plane spanned by initially orthogonal electron and atomic spin polarizations. Details will be outlined in a separate publication [16].

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