Transferred orbital angular momentum in the excitation of ^{138}Ba$(\ldots 6s6p\ 6\,^1P_1)$ by electron impact

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We have determined the $L_\perp$ coherence parameter for electron-impact excitation of the $(\ldots 6s6p\ 6\,^1P_1)$ state in ^{138}Ba by measuring the superelastic scattering of electrons from laser-excited ^{138}Ba. Three electron-impact energies were studied and the scattering-angle coverage in each case was sufficient to allow the observation of pronounced structure in the angular behavior of this parameter. We also present a detailed comparison of our results with available theory.

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I. INTRODUCTION

The fuller characterization of electron-atom collisions by the determination of electron-impact coherence parameters (EICP's) has been a subject of considerable interest in recent years. In addition to the differential cross section, measured EICP's provide a sensitive test of theoretical approaches to the scattering problem since they are defined in terms of relative phases and moduli of the appropriate scattering amplitudes. Experimental techniques for the measurement of EICP's involve the selection of an ensemble of collision events in which the momenta of the incident and scattered electrons as well as the state of the excited atom are well defined. This selectivity can be achieved by detecting the scattered electron and fluorescence photon (from the collisionally excited state) in time coincidence (electron-photon coincidence experiments) or by scattering electrons superelastically from an excited state that has been prepared by laser excitation (superelastic-scattering experiments). In the present discussion we concentrate on experiments that involve unpolarized electron beams and spin-insensitive detection so that measured EICP's are spin-averaged quantities.

A comprehensive review of measurements made using these two techniques and comparison with available theory has been given by Andersen, Gallagher, and Hertel [1] for work carried out before 1987. In the case of a $J_0=0$ ground state and a $J=1$ excited state (i.e., He, Hg, the alkaline-earth-metals, and the heavy rare gases) these authors also defined a set of frame-independent EICP's with an appealing physical significance regarding the shape of the electronic charge cloud excited by an inelastic collision. These are P_l^+ (the charge cloud anisotropy or charge cloud linear polarization), γ (the charge cloud alignment angle), $L_\perp$ (the transferred orbital angular momentum), and ρ_{00} (the "height parameter"). The + superscript indicates that P_l^+ describes the component of the charge cloud with positive reflection symmetry with respect to the scattering plane.

In general, the charge cloud may have mixed reflection symmetry due to the presence of spin-dependent terms in the interaction Hamiltonian. The deviation of ρ_{00} from zero indicates the extent to which such spin-dependent terms are significant. When $\rho_{00}=0$ the scattering can be described purely in terms of LS coupling (He, for example). In this case the two EICP's P_l^+ and $L_\perp$ are related by $(P_l^+)^2+(L_\perp)^2=(P^+)^2=1$, where P^+ is called the degree of polarization.

Since the review by Andersen, Gallagher, and Hertel [1] substantial progress has been made in the measurement and calculation of EICP's for alkaline-earth-metal atomic targets. Because they are essentially two-electron systems, such atoms offer a natural extension to studies carried out on the helium atom which is, by far, the most extensively investigated collision target.

Experimental determination of EICP's for alkaline-earth-metal-atom targets has been accomplished using both electron-photon coincidence and superelastic-scattering techniques. The coincidence method has been employed by Brunger *et al.* [2], Zohny *et al.* [3], and Hamdy *et al.* [4] to measure EICP's for the Mg $3\,^1P_1$, Ca $4\,^1P_1$, and Sr $5\,^1P_1$ excitations, respectively. Further measurements on Ca and Sr were reported by Hamdy *et al.* [5] and Beyer *et al.* [6]. Superelastic-scattering experiments have been used to measure EICP's for the Ca $4\,^1P_1$ excitation (Law and Teubner [7]) and the Ba $6\,^1P_1$ excitation (Zetner, Li, and Trajmar [8,9]).

Theoretical approaches which have successfully described EICP for the He $2\,^1P_1$ excitation face additional challenges in describing the corresponding excitations in the alkaline-earth metals. With increasing atomic number Z, the electronic structure becomes more complicated and the description of target states generally involves substantial configuration mixing. Also, electron scattering by heavy atoms is characterized by non-negligible contributions from partial waves of high angular momentum which must therefore be retained in partial-wave expansions of the scattering states. Furthermore, theoretical treatment of scattering by high-Z atoms is expected to require a relativistic approach since the interaction Hamiltonian may be spin dependent. In spite of these difficulties, calculations of EICP's for alkaline-earth atoms have recently been reported for Mg by Meneses, Pagan, and Machado [10], Clark, Csanak, and Abdallah [11], and Mitroy and McCarthy [12], for Sr by Beyer

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et al. [6], for Ba by Clark *et al.* [13], for Mg and Ba by Fabrikant [14], and for Ca, Sr, and Ba by Srivastava *et al.* [15].

In this paper we report on measurements of the $L_\perp$ coherence parameter carried out using superelastic scattering from laser-excited ^{138}Ba(...6*s*6*p* $6\,^1P_1$). The present results supplement and extend our previous measurements [8,9] of the charge-cloud alignment parameters P_l^+ and γ. In Sec. II we give a brief description of the experimental apparatus and an outline of the theory required to extract EICP's from the measured scattering signal. In Sec. III we present the data and we compare our measured $L_\perp$ values with those calculated by Clark *et al.* [13,16] in the unitarized distorted-wave approximation (UDWA), with those calculated by Srivastava *et al.* [15,17] in a fully relativistic distorted-wave (RDW) theory, and with those calculated by Fabrikant [14] using the close-coupling (CC) approach. Concluding remarks are made in Sec. IV.

II. EXPERIMENTAL PROCEDURE

Figure 1 shows a schematic diagram of our experimental arrangement. The electron gun (impact-energy resolution full width at half maximum of 0.6 eV) is rotatable with respect to a fixed, hemispherical electron detector. The laser beam is incident upon the target Ba vapor beam from below the scattering plane and strikes the scattering plane perpendicularly. The polarization state of the laser beam at the target is determined by a retardation plate with a phase retardation of δ whose fast axis makes an angle β with respect to the forward-scattering direction. Rotation of a right-handed screw in the direction of decreasing β defines the laser incidence direction. The Glan-Taylor prism is employed to ensure that pure linearly polarized laser light is incident upon the retardation plate. The angle α gives the angular displacement of the Glan-Taylor prism transmission axis with respect to the forward-scattering direction.

The theory which relates superelastic-scattering intensity to the EICP's has been described earlier [9,18] and stems from the work of Macek and Hertel [19] who showed that

$$I^s = \bar{C}\,\mathrm{Tr}[\hat{\rho}\hat{\tau}]\ , \tag{1}$$

where I^s is the superelastic scattering intensity: $\bar{C}$ is a constant consisting of multiplicative factors such as detection solid angle, detection efficiency, laser-excited atom population, incident electron flux, and the differential superelastic scattering cross section; $\hat{\tau}$ is the density operator of the laser-excited atomic population; $\hat{\rho}$ is the density operator of the excited-state atomic population produced by the (time-inverse) inelastic collision; and Tr[ ] is the "trace." For the arrangement shown in Fig. 1, the superelastic-scattering intensity can be expressed as

$$I^s(\beta-\alpha)=\bar{C}'\left[1+\frac{P_l^+}{2}(1+\cos\delta)\cos(2\alpha-2\gamma)+\frac{P_l^+}{2}(1-\cos\delta)\cos(4\beta-2\gamma-2\alpha)+L_\perp\sin\delta\,\sin(2\beta-2\alpha)\right], \tag{2}$$

where $\bar{C}'$ is a constant proportional to $\bar{C}$ in Eq. (1) and $I^s(\beta-\alpha)$ indicates explicitly the functional dependence of I^s on $\beta-\alpha$. Examination of Eq. (2) reveals that a perfect half-wave retardation plate ($\delta=\pi$) will allow the straightforward extraction of P_l^+ and γ through a measurement of the depth of modulation and phase of I^s as a function of $\beta-\alpha$. In this case the laser light is linearly polarized at the target. A perfect quarter-wave retardation plate ($\delta=\pi/2$) gives circularly polarized light at the target and allows the extraction of $L_\perp$ through the measurement of I^s for $\beta-\alpha=\pm\pi/4$.

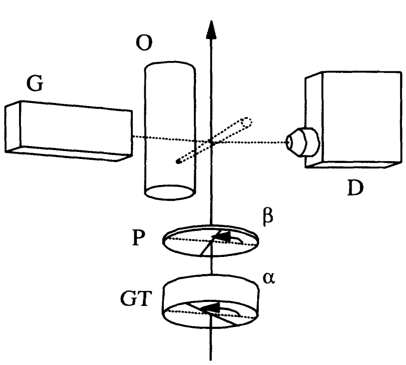

FIG. 1. Schematic diagram of the experimental arrangement. The electron gun (G), scattered-electron detector (D), metal-vapor beam source (O), retardation plate (P), and Glan-Taylor prism (GT) are indicated. Angles α and β are defined in the text.

In the present experiments we employed a multiple-order quartz retardation plate which was specified as $\lambda/4$ at $\lambda=555$ nm. Carrying out diagnostic measurements of the type described by Wedding, Mikosza, and Williams [20] we found that $\cos\delta=-0.374(\pm0.02)$ for the $\lambda/4$ plate at the Ba resonance wavelength $\lambda=553.5$ nm.

Deviation from the "perfect" phase retardation (namely, $\cos\delta=0$ for the $\lambda/4$ plate) forced us to use a more complicated scheme to extract $L_\perp$ from Eq. (2). We measured $I^s(\beta-\alpha)$ for $\beta-\alpha=\pm\pi/4,0,\pi/2$ and formed the ratio

$$R=\frac{I^s\left(+\frac{\pi}{4}\right)-I^s(0)\cos\delta}{I^s\left(-\frac{\pi}{4}\right)-O^s(0)\cos\delta}$$

from which we get $L_\perp$ by the relation

$$L_\perp = \frac{R-1}{R+1}\left[\frac{1-\cos\delta}{1+\cos\delta}\right]^{1/2}.$$

The measurements of $I^s(\pi/2)$ is redundant with the $I^s(0)$ measurement [i.e., $I^s(0)$ should be equal to $I^s(\pi/2)$] but allows us to determine whether rotation of the $\lambda/4$ plate introduces any steering of the laser beam away from the target region.

Electron-impact energy was calibrated against the known position of the He $2\,^2S$ resonance in the elastic-scattering channel. We calibrated the scattering angle by calculating $(1-L_\perp^2)^{1/2}$ and aligning the observed minimum with the corresponding minimum previously measured in P_l^+ [9]. This calibration was estimated to be accurate to within $\pm 1°$. The sign of $L_\perp$ was normalized to theory.

Experimental conditions can sometimes lead to a significant reduction in the sensitivity of the superelastic-scattering signal to the polarization state of the laser beam. This depolarizing influence can arise from the well-known process of radiation trapping and from the presence of a scattering volume of finite spatial extent. With regard to superelastic-scattering experiments the former problem has been discussed by Hertel and Stoll [21] and Fischer and Hertel [22], while the latter effect has been discussed in detail in our previous work [18]. Based on calculations such as those made by Hertel and Stoll [21], we predict that the effect of radiation trapping in our measurements is small. A test of this prediction was made by measuring $L_\perp$ as a function of Ba target beam density. We estimate that, in our experiments, carried out with a target atom density of, approximately, 7×10^{10} cm^{-3}, depolarization by radiation trapping was less than 5%. The effect of the finite scattering volume is discussed below, when we make a comparison of our data with theory.

III. RESULTS AND DISCUSSION

The $L_\perp$ coherence parameter is an interesting probe of the electron-atom collision dynamics. It gives the expectation value of the orbital angular momentum (measured in the perpendicular direction to the scattering plane) transferred by an inelastic collision. Several "universal" features of the behavior of this parameter have been observed for electron-impact excitation of simple systems such as hydrogen and helium. These are discussed by Lin *et al.* [23].

Typically, for these systems, $L_\perp$ will vary monotonically with scattering angle in such a way that it increases from zero (in the forward-scattering direction) to a maximum, then decreases to change sign and reach a minimum, returning to zero in the backward-scattering direction. The vanishing of $L_\perp$ in the forward- and backward-scattering directions can be understood classically by noting that the projectile electron exerts no torque on the target atom in these two situations.

The relatively simple behavior of $L_\perp$ prompted the development of a semiclassical model [24] which linked the sign of this parameter to the attractive (+ sign) or repulsive (− sign) nature of the collisional interaction. According to this model, electrons suffering glancing collisions experience a net attractive force while backward-scattering electrons penetrate deep into the atomic charge cloud and experience a repulsion. Madison, Csanak, and Cartwright [25] subsequently demonstrated the inadequacies of this model by reproducing the observed $L_\perp$ behavior using the distorted-wave Born approximation and a purely attractive interaction potential. The importance of this work was twofold. It showed that the introduction of distortion into the entrance and exit scattering channels in a first-order theory was sufficient to yield reasonably quantitative agreement with observations of $L_\perp$. In the absence of such distortion one obtains the first Born approximation which fails completely by giving $L_\perp=0$ for all scattering angles. Second, by performing a partial-wave expansion of the entrance and exit scattering channels and introducing distortion selectively into the various partial waves, these authors concluded that the angular behavior of $L_\perp$ arises from quantum-mechanical interference among the distorted partial waves and does not lend itself to a simple semiclassical interpretation.

In the excitation of He $2\,^1P_1$ by 80-eV electrons, distorted partial waves up to $l=2$ were shown to make the dominant contribution. For the Ba $6\,^1P_1$ excitation measured in the present work, we would expect significant contributions from higher-order partial waves with the resultant quantum-mechanical interference giving rise to a more highly structured angular behavior of $L_\perp$. Figures 2–4, in which we compare our present measurements with available theory, illustrate that this is indeed the case. The angular variation of $L_\perp$ exhibits pronounced structure and is substantially different from that occurring in H and He excitations. We do, however, note a resemblance to $L_\perp$ behavior predicted and observed in Ca [3,5,15], Sr [4,5,6,15], and, in some cases, the heavier rare

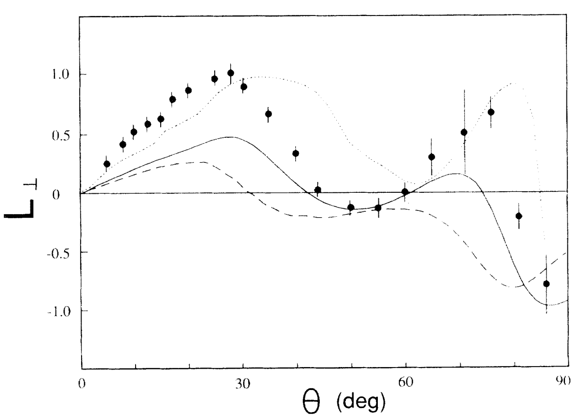

FIG. 2. Comparison of the measured $L_\perp$ coherence parameter (circles with error bars) with UDWA calculations of Clark *et al.* [13,16] (dashed curve), RDW calculations of Srivastava *et al.* [15,17] (solid line), and CC calculations of Fabrikant [14] (dotted line). The measurements were obtained in superelastic scattering experiments involving incident electrons with kinetic energy $E_0^s=17.76$ eV, while the calculations describe an inelastic collision involving incident electrons with kinetic energy $E_0=20.0$ eV.

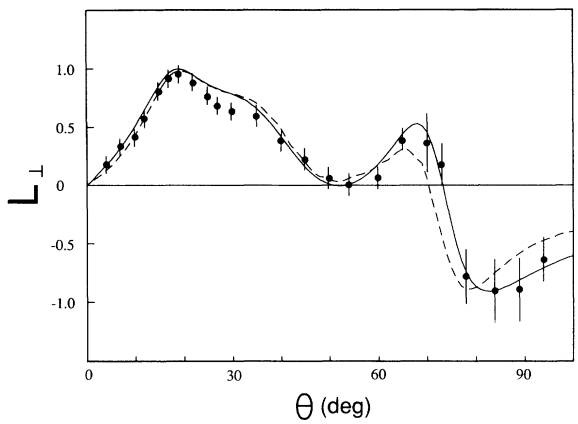

FIG. 3. Same as Fig. 2 except $E_0^s=34.5$ eV and $E_0=36.67$ eV. No CC calculations were available in this case.

gases [1,26–28].

In Figs. 2–4 the $L_\perp$ parameters extracted from our superelastic-scattering experiments with incident electron energy E_0^s are compared to theoretical parameters calculated for the time-inverse inelastic collision involving incident electrons with energy $E_0=E_0^s+\Delta E$, where ΔE is the excitation energy of the Ba(...$6s6p\ 6\,^1P_1$) state ($\Delta E=2.24$ eV). Superelastic and inelastic collisions are related by time-reversal symmetry of the scattering amplitudes. EICP's extracted from a superelastic-scattering measurement are the same as EICP's extracted from an electron-photon coincidence experiment involving the time-inverse inelastic collision unless the collisional interaction is significantly spin dependent in which case the nature of the spin average in the two experiments is difficult. Spin-dependent forces can give rise to partial spin polarization of an inelastically scattered electron beam which is initially unpolarized. The time inverse of such an inelastic process corresponds to superelastic scattering in which the incident electrons are partially polarized. Since our superelastic-scattering measurements were carried out with unpolarized incident beams we sacrifice some rigor in making the comparisons shown in Figs. 2–4, but we note that RDW theory predicts spin polarizations in elastic scattering of less than 10% over the range of impact energies and scattering angles studied.

In making a detailed comparison with theory it was necessary to determine to what extent the presence of a finite-sized interaction volume influenced the measurement. This was investigated through the use of a modeling calculation identical to that discussed in our previous report on the charge-cloud alignment parameters [8,9] but applied to the $L_\perp$ parameter. The calculation indicated that the measurement of $L_\perp$ was negligibly affected by the interaction volume size even at near-forward-scattering angles. It is therefore legitimate to directly compare experimental and theoretical results over the whole range of scattering angles studied.

At impact energy $E_0=20$ eV quantitative agreement between experiment and theory is poor, although all three theories reproduce, to some extent, qualitative aspects of the angular behavior of $L_\perp$. The failure of the first-order perturbative theories (UDWA and RDW) is not surprising at this low impact energy. The CC approach should be more successful in this regime but the two-state, nonexchange, nonrelativistic treatment adopted by Fabrikant [14] appears to be inadequate.

Comparison of the first-order perturbative theories with experiment at $E_0=36.7$ eV reveals excellent quantitative agreement out to 92° scattering angle. An interesting situation occurs, however, at $E_0=50$ eV in which a discrepancy between theory and experiment develops beyond the first maximum in $L_\perp$ and worsens with increasing scattering angle to the point where both UDWA and RDW theories fail to predict an apparent sign change in $L_\perp$ near 60° scattering angle.

In Figs. 5–7 the present $L_\perp$ measurements are combined with our earlier P_l^+ measurements to give P^+, the degree of polarization, using $P^+=[(P_l^+)^2+(L_\perp)^2]^{1/2}$. If $P^+=1$, then the excitation is said to be fully coherent and spin dependence in the collision is negligible. Over

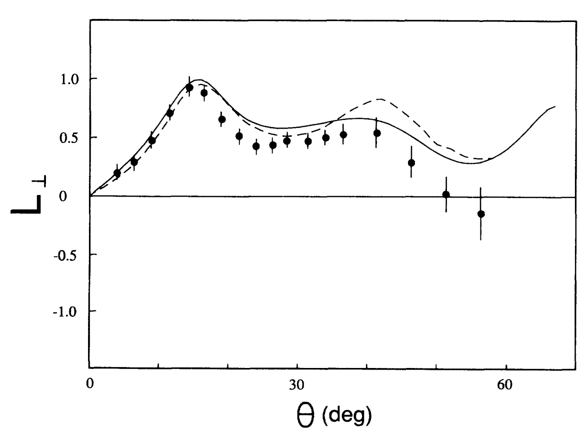

FIG. 4. Same as Fig. 3 except $E_0^s=47.76$ eV and $E_0=50.0$ eV.

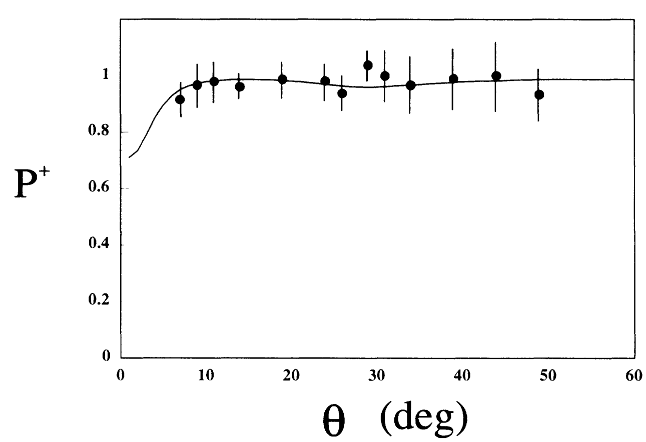

FIG. 5. The degree of polarization P^+ for $E_0=20$ eV. Measured values (circles with error bars) were obtained by combining the present measured $L_\perp$ values with earlier measurements of P_l^+ [9]. The solid line represents a calculation which models the effect of a finite interaction volume using the RDW EICP's of Srivastava *et al.* [15,17].

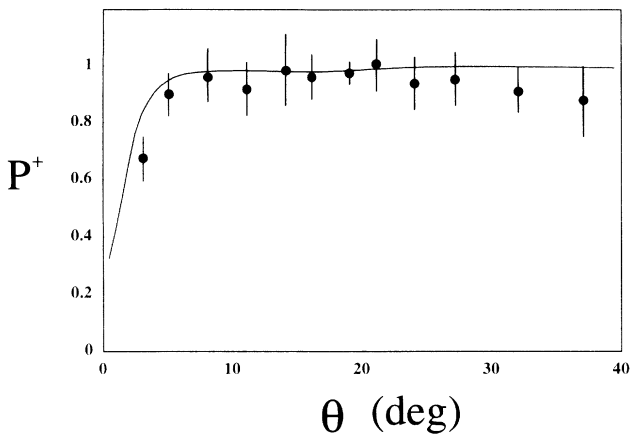

FIG. 6. Same as Fig. 5 except $E_0 = 36.7$ eV.

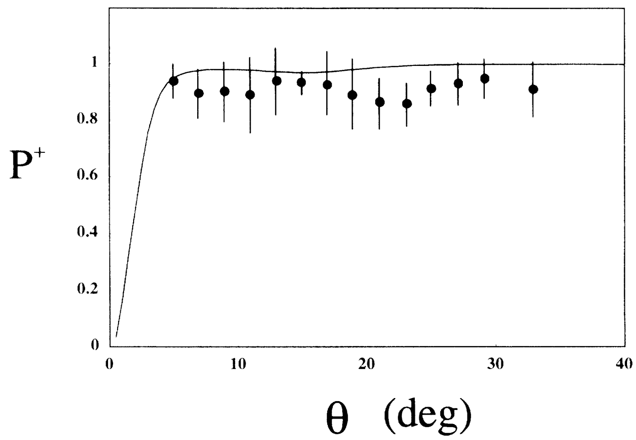

FIG. 7. Same as Fig. 5 except $E_0 = 50$ eV.

the range of impact energies and scattering angles presented in the figures, it is clear that $P^+ = 1$ to within experimental error limits. We note that large error bars in the case of $E_0 = 50$ eV arise partly due to calibration uncertainty in the scattering angle. At this impact energy both P_l^+ and $L_\perp$ vary rapidly with angle and small uncertainties can give large variations in P^+. The fully coherent nature of the Ba $6\,{}^1P_1$ excitation is predicted by both UDWA and RDW theories. We point out, however, that the RDW theory does give a nonzero spin-polarization function but the manifestation of spin effects in the EICP's is predicted to be small. The solid line in Figs. 5–7 results from a calculation which models the effect of the finite interaction volume on P^+ using RDW EICP's.

IV. CONCLUSIONS

We have measured the $L_\perp$ coherence parameter for excitation of the Ba(...$6s6p\,{}^1P_1$) state using the superelastic-scattering technique. The angular behavior of this parameter shows pronounced structure which exhibits some similarities to predicted and observed behavior in the heavy rare gases and the other alkaline earth metals (namely, Ca and Sr). Comparison with two first-order perturbative theories reveals some qualitative agreement at impact energy $E_0 = 20$ eV, excellent quantitative agreement at impact energy $E_0 = 36.7$ eV, and quantitative agreement at $E_0 = 50$ eV for small scattering angles. Results of a close-coupling calculation at $E_0 = 20$ eV give some qualitative aspects of the observed $L_\perp$ angular behavior.

We have also combined the present results with earlier measurements of P_l^+ in order to calculate P^+, the degree of polarization. Observation that $P^+ = 1$ over the range of impact energies and scattering angles studied indicates that the Ba $6\,{}^1P_1$ excitation is fully coherent and the role of spin dependent forces is small in this regime. This is in agreement with the predictions of available first-order perturbative theories.

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