Problems associated with the measurement of coherence parameters: Superelastic electron scattering by laser-excited ^{138}Ba(...6s6p $^1P_1$) atoms

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Measurements of superelastic scattering of electrons by laser-excited ^{138}Ba(. . .6s6p $^1P_1$) atoms were carried out. An asymmetry observed by us and previously by Register *et al.* [Phys. Rev. A **28**, 151 (1983)] has been explained using a model of scattering from a target with finite dimensions. This model employed coherence parameters which we calculated in the distorted-wave approximation. The results indicated that the interpretation of coherence experiments in terms of scattering from a pointlike target can lead to serious errors in the deduction of coherence parameters at low scattering angles.

Electron-photon coincidence[1] experiments and superelastic scattering studies from laser-excited atoms[2] represent an active new field in electron-atom collision physics. These studies go beyond the conventional differential-cross-section (DCS) measurements in that they yield information about the complex scattering amplitudes by measuring quantities that will be referred to here as electron-impact coherence parameters (EICP). The existing experimental and theoretical data are well summarized in the recent comprehensive review of Andersen *et al.*[3]

In the interpretation of electron-atom, beam-beam scattering experiments (including the measurement of EICP) it has always been assumed, so far, that the scattering signal originates from a pointlike scattering source and is associated with a reasonably well-defined geometry and impact energy. While this assumption is acceptable in most DCS measurements, we show here that it can lead to serious errors in EICP measurements. Utilizing detailed modelings, based on our theoretically calculated EICP, and experimental checks, we explicitly demonstrate this point on superelastic scattering of electrons by ^{138}Ba($^1P_1$) atoms prepared by "in-plane," linearly polarized laser pumping as described by Register *et al.*[4] We will show that geometrical effects, associated with the finite scattering volume, drastically influence the superelastic scattering signal and that the EICP deduced from these experiments can be subject to large errors at small scattering angles. Particularly, conclusions concerning the importance of spin-orbit-coupling effects cannot be readily drawn. The exact nature and the magnitude of the extended scattering volume effect on the superelastic signal depends on the particular experimental arrangement and on the behavior of the EICP themselves.

We calculated the EICP in the distorted-wave approximation (DWA) using multiconfiguration ground- and excited-state wave functions. The basis orbitals were obtained numerically from Hartree-Fock calculations. The details of these calculations and results for a number of impact energies will be given elsewhere.[5] The calculations show that the EICP associated with spin-orbit coupling, ϵ and Δ, satisfy $\cos\epsilon = \cos\Delta = 1$ to within the numerical accuracy of the calculation at all impact energies (E_0) and scattering angles (θ_e) considered here. Therefore, we can treat the scattering process in the *LS* coupling scheme. For $E_0 = 7.24$- and 32.24-eV impact energies the calculated λ and χ parameters are given in Table I.

Our modeling calculation is based on the theory of Macek and Hertel[6] which describes the superelastic-scattering intensity measured at impact energy E_0^s in terms of the time-inverse, inelastic-scattering process taking place at impact energy E_0 [$E_0^s = E_0 - 2.24$ eV for ^{138}Ba($^1P_1$)]. The modeling is carried out by representing the extended scattering volume as an array of discrete scattering centers and applying the theoretical formalism, which incorporates our calculated EICP, to each center. To this end, a laboratory frame is fixed to the apparatus such that a nominal scattering plane is defined by the electron gun and detector axes. A nominal superelastic-scattering angle θ_e^{NS} is determined by the angle between these two axes. The laser-beam polar, azimuthal, and polarization angles, as measured in this frame, are denoted by θ_ν, ϕ_ν, and ψ_ν.

The overall superelastic signal (I_Σ^s) is the weighted average of the contributions (I_j^s) from scattering centers located at position vectors $\mathbf{r}_j$ in the laboratory frame and theoretically can be given as

$$I_\Sigma^s(\psi_\nu) = \sum_j a_j I_j^s(\psi_\nu) \sim 1 + \eta_\Sigma \cos[2\psi_\nu + 2(\alpha_{\mathrm{tot}})_\Sigma] \ . \quad (1)$$

The summation over j was applied to the expression for the superelastic-scattering intensity obtained from the Macek and Hertel theory for a $J=0$ to $J=1$ transition[7] and the coefficients a_j are weighting factors normalized so that $\sum_j a_j = 1$. Equation (1) defines an overall modulation depth η_Σ and overall total modulation phase shift $(\alpha_{\mathrm{tot}})_\Sigma$ which are functions of the EICP and the scatter-

TABLE I. Coherence parameters for electron-impact excitation of the ^{138}Ba$(\ldots 6s6p\,^1P_1)$ level at $E_0=7.24$ and 32.24 eV.

	Calculated				Unfolded		
θ_e	7.24 eV		32.24 eV		32.24 eV		
(deg)	λ	$\cos\tilde{\chi}$	λ	$\cos\tilde{\chi}$	λ	$\cos\chi$	$\cos\epsilon$
0.0	1.00	1.00	1.00	1.00			
0.5	1.00	1.00	0.93	1.00	0.70	0.29	0.15
1.0	0.99	1.00	0.80	1.00	0.62	0.40	0.45
2.0	0.94	1.00	0.47	1.00	0.44	0.66	0.71
4.0	0.83	1.00	0.17	0.99	0.19	0.88	0.91
6.0	0.67	1.00	0.06	0.92	0.08	0.83	0.96
8.0	0.51	1.00	0.04	0.66	0.04	0.66	0.98
10.0	0.37	1.00	0.04	0.26	0.04	0.26	0.99
15.0	0.10	0.98	0.16	−0.23	0.14	−0.21	0.99
20.0	0.01	0.70	0.39	−0.05	0.33	0.02	1.00
30.0	0.25	−0.94	0.64	0.40			
50.0	0.70	−0.01	0.65	1.00			
70.0	0.52	−0.48	0.09	0.99			
90.0	0.72	−0.87	0.55	0.80			

ing geometry and completely specify the dependence of the superelastic signal on the laser-beam polarization.

The results of modeling calculations at $E_0=30$ eV are shown in Figs. 1(a)–1(d). Triangular symbols in these figures represent the results of experimental measurements which are described below. The finite scattering volume was, for these particular calculations, approximated by five single-point scattering centers distributed uniformly along a line perpendicular to the nominal scattering plane. Asymmetry was introduced by displacing the center of gravity of this five-point array above or below the nominal scattering plane. This crude model was found to give the essential elements of the behavior of η_Σ and $(\alpha_{\text{tot}})_\Sigma$ when compared with more sophisticated versions such as an array of 45 points distributed in a cylinder. In fact, the crude model gives surprisingly good agreement with experiment. One crucial point must be made concerning these results. It can be shown that, in the *LS* coupling case, for $\phi_v=0°$ and for a pointlike scattering event occurring at the laboratory-frame origin, $(\alpha_{\text{tot}})_\Sigma=0$ and $\eta_\Sigma=1$ for all scattering angles. As the figures show, this is definitely not the case when the single-point scatterer is replaced by a more realistic extended volume of scatterers. Dramatic features in the behavior of η_Σ and $(\alpha_{\text{tot}})_\Sigma$ are exhibited which indicate that the interpretation of experimental data within the framework of a single pointlike scattering is inadequate. In the present case, one would be led to conclude that significant spin-orbit coupling is present when, in fact, EICP which are appropriate for an *LS*-coupled system have been incorporated into the model. The calculations presented here are a subset of a thorough modeling investigation into the behavior of η_Σ and $(\alpha_{\text{tot}})_\Sigma$. This investigation shows that $(\alpha_{\text{tot}})_\Sigma=0$ for an extended scattering volume which is symmetrically located with respect to the nominal scattering plane. The asymmetry problem observed by Register *et al.*[4] and by us in the present work should therefore, in principle, be surmountable by precisely controlling the scattering geometry (as elaborated upon below). The modeling has revealed, however, that the geometry-induced variation in η_Σ will persist for both symmetrically and asymmetrically located scattering volumes.

One expects that the geometrical effect would be largest at near-zero scattering angles and would become negligible at high scattering angles where the individual collision frames become nearly identical with the laboratory frame. It can be seen from the figures, however, that the largest distortion effects in η_Σ and $(\alpha_{\text{tot}})_\Sigma$ materialize at $\theta_e^{\text{NS}}=9°$ for $E_0^s=30$ eV in the ^{138}Ba$(^1P_1)$ case. This is the result of the particular behavior of these EICP. For EICP appropriate to $E_0^s=5$ eV, for example, the large effects appear at around 20° scattering angle. This behavior can be expressed in physical terms as suggested by McConkey.[8] The scattering angles at which the dramatic variation in $(\alpha_{\text{tot}})_\Sigma$ or η_Σ occur are those corresponding to particular alignment angles of the excited-state charge cloud prepared by the inelastic scattering process. Specifically, we have verified that when the laser beam views this charge cloud "end on" (i.e., the classical dipole produced by the inelastic collision is oscillating along an axis parallel to the laser-beam incident vector), the superelastic signal becomes extremely sensitive to geometrical effects.

Clearly, the influence of an extended scattering volume manifests itself dramatically in the observed polarization modulation of the superelastic signal. Conventionally, under the assumption of pointlike scattering, the EICP are unfolded from this signal by measuring modulation depths for three different laser geometries. To assess the effect of an extended scattering volume on the measurement of EICP, we used the modulation depth η_Σ given by our modeling calculation [Figs. 1(b) and 1(d)] as "fictitious" experimental data and unfolded the EICP from this fictitious data under the (incorrect) assumption of pointlike scattering. In Table I, the extracted EICP are compared with the theoretical EICP used as input to the modeling code. Below $\theta_e = 8°$, clear discrepancies exist (especially for $\cos\chi$ and $\cos\epsilon$) which become increasingly severe as θ_e approaches zero. It is important to note that, although the geometry-related influence on the modulation depth is dramatic at $\theta_c = 9°$ (for $E_0^s = 30$ eV), no such drastic effect appears in the unfolded EICP. The EICP extracted by analyzing η_Σ in terms of a pointlike scattering process become meaningless for θ_e approaching zero, as intuitively expected.

In order to verify the predictions of the modeling calculations, we designed and carried out specific experiments. The critical question was the following. If geometrical effects are responsible for $(\alpha_{tot})_\Sigma \neq 0$ and $\eta_\Sigma \neq 1$, why were Register *et al.*[4] (and also we at the beginning of our more recent experiments) unable to modify the scattering geometry and cause a change in $(\alpha_{tot})_\Sigma$? In particular, deliberate displacements of the electron beam or laser beam from their aligned positions had no effect on the phase-shift behavior. We surmised that our detector can effectively see only scattering events occurring very close to the axis of the view cone. The size and exact location of the effective scattering volume depends on the tuning of the detector optics but we estimated that, in our experiments, a typical dimension of about 0.05 cm is obtained at the interaction region. Since the laser-beam diameter and electron-beam diameter are, in this case, much larger (0.3 cm) than the size of the effective scattering volume, it can be understood that, by displacing the laser or electron beam, we simply lose scattering intensity (as observed) but cause no significant change in the character of the scattering geometry. To prove this point, we focused down the laser-beam size in the scattering region to about 0.05-cm diameter and modified our detector by introducing a set of electrostatic deflector plates between the two view-cone-defining apertures. The smaller laser-beam size allowed us to move the volume of excited-state scatterers with respect to the nominal scattering plane

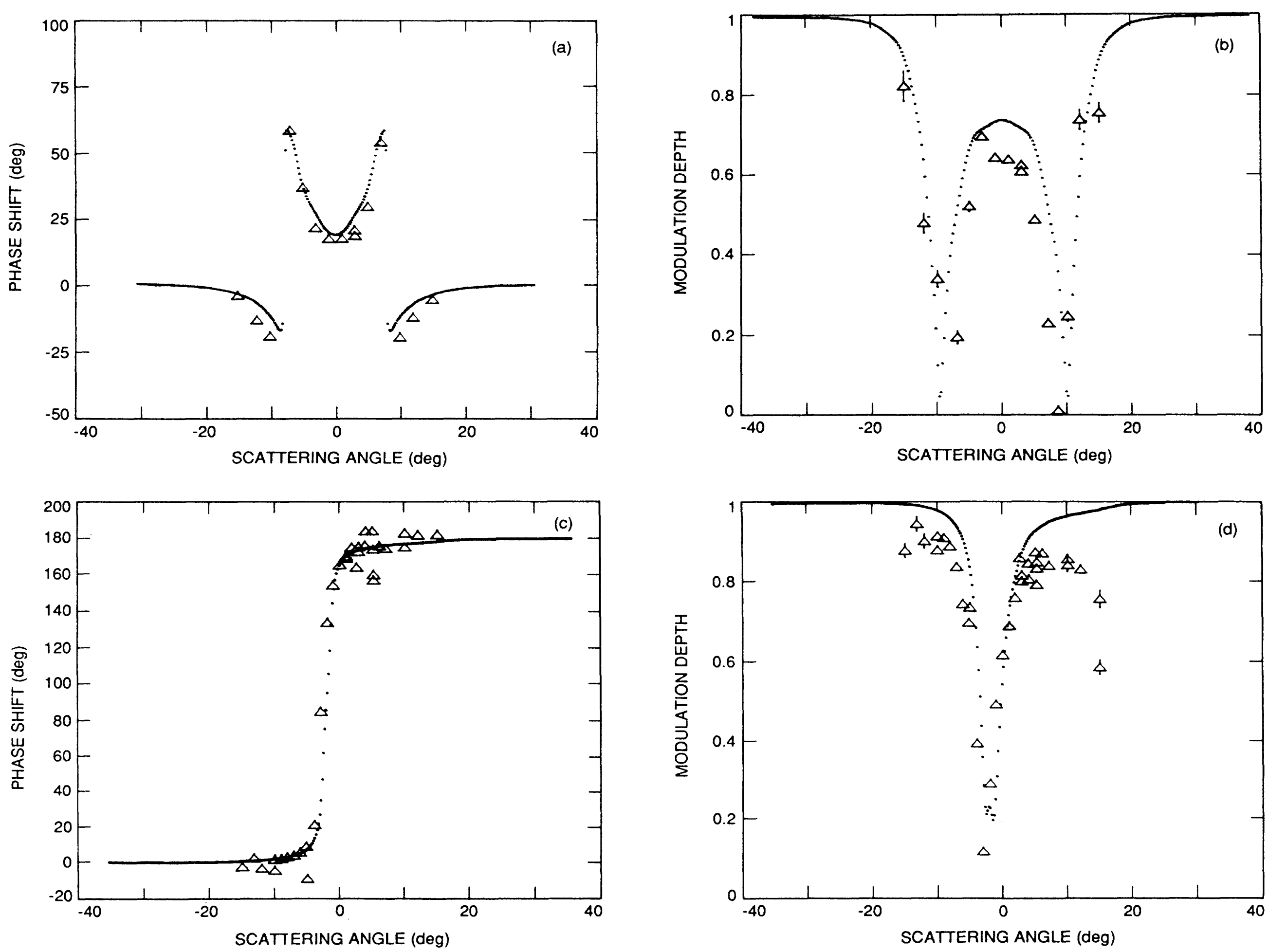

FIG. 1. (a) Overall total modulation phase shift at $E_0^s = 30$ eV, laser geometry $\theta_v = 90°$, $\phi_v = 0°$. The scattering volume is represented by five points distributed evenly on a 0.05-cm line perpendicular to the nominal scattering plane. The center of gravity of this array is offset by +0.04 cm from this plane. (b) Same as (a) except for the overall modulation depth (dimensionless). (c) Same as (a) except for $\theta_v = 45°$ and offset of 0.02 cm. (d) Same as (c) except for overall modulation depth.

and the deflector plates enabled us to force the detector to look at this displaced volume. One more important remark has to be made. The investigation of $(\alpha_{tot})_\Sigma$ by Register *et al.*[4] was carried out at 30- and 100-eV impact energies for a laser-beam configuration corresponding to $\theta_v=45°$, $\phi_v=0°$. The modeling calculations show that, for this configuration, a sharp 180° change in $(\alpha_{tot})_\Sigma$, around zero nominal scattering angle, occurs which depends only slightly on the scattering geometry. This configuration was, therefore, unfavorable for the purpose of investigating, experimentally, the dependence of the phase shift on a misaligned geometry. We, therefore, carried out our investigation under the more favorable conditions of $E_0^s=5$ eV and $\theta_v=90°$ ($\phi_v=0°$). The results were very convincing. A small (fraction of a mm) movement of the laser (and retuning of the detector) caused large changes in the phase shift. Further confirmation of our hypothesis came from comparing the results of well-controlled experimental measurements with model calculations. $I_\Sigma^s(\psi_v)$ modulation measurements were carried out at 5-, 10-, 30-, and 100-eV impact energies over a range of nominal superelastic scattering angles. The phase shift and modulation depth as a function of θ_e^{NS} were compared to the results of modeling calculations. An example is shown for $E_0^s=30$ eV with laser configurations $\phi_v=0°$, $\theta_v=45°$ and 90° in Figs. 1(a)–1(d). The agreement between measurements and calculated results is excellent. Model calculations indicate that a slight asymmetry persists in these experiments despite our efforts to preserve a precise optical alignment of the electron gun, detector, and target beam.

In conclusion, we found that the unavoidable fact that the effective scattering volume in any electron-atom, beam-beam experiment is finite may seriously influence the superelastic scattering intensity and consequently have a detrimental effect on the determination of the EICP near 0° scattering angle. In particular, conclusions about spin-orbit coupling effects based on these experiments must be made with caution. We explicitly proved this for the case of superelastic scattering of electrons by ^{138}Ba($^1P_1$) atoms prepared by a linearly polarized laser beam which is located in the nominal scattering plane. In light of these investigations, the asymmetry observed by Register *et al.*[4] can now be understood. The phase-shift behavior observed by them is identical to that shown in Figs. 1(a) and 1(c). (At $\theta_v=90°$, Register *et al.*[4] shifted the upper part of their curve by $-180°$ and plotted the absolute value of α.) It is clear that similar effects may be present in other superelastic and electron-photon coincidence experiments and the interpretation of these measurements with the assumption of a single pointlike scatterer (which was always made in the past) has to be reexamined. Such efforts are in progress in our laboratory.

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