aration of the carbon foils. We would also like to take this opportunity to express our deep appreciation to Dr. Eliel Skurnik, to the Weizmann Institute team of accelerator technicians, and to the National Electrostatics Corporation technicians who worked here, for their skill and devotion in assembling the Pelletron and making it available for experimentation in good order and in a very short time. This work was supported by the Israel Commission for Basic Research.

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Coherent-State Multipole Moments: Source of Important Scattering Information

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Coherent states are completely parametrized in terms of multipole moments and their time derivatives. For hydrogen atoms excited by electron impact, our calculations show that these parameters provide new information concerning the final-state interactions of the hydrogen atom and the projectile electron. These large final-state-interaction effects make first-order perturbation calculations inappropriate. We present calculations which properly incorporate these dynamical effects. Experiments which determine these parameters are discussed.

We discuss an intuitively accessible parametrization for the density matrix of a manifold of coherently excited states in terms of average values of the familiar electric and magnetic multipole operators taken together with their time-derivative operators. This multipole-moment and multipole-moment time-derivative picture can be used to provide physical insight into coherently excited states produced by *any* process.

We use the multipole-moment picutre to guide us in pilot calculations of mixed-L coherence multipoles produced in atomic hydrogen by electron impact. We find that certain of the coherence multipoles are extremely sensitive to the final-state interaction between scattered and bound electrons. As a result, first-order perturbation scattering models are totally incapable of predicting these multipoles, even at very high energies. After comparing our calculations with available experimental results,[1] we suggest further experiments to fully determine coherence multipoles.

A general coherently excited state can be represented by a Hermitian density matrix, ρ, where

$$\rho = \sum_{\alpha JM,\alpha' J' M'} \rho_{\alpha JM,\alpha' J' M'},$$

$$\rho_{\alpha JM,\alpha' J' M'} = |\alpha JM\rangle\langle\alpha JM|\rho|\alpha' J' M'\rangle\langle\alpha' J' M'|. \quad (1)$$

When excitation amplitudes, $A_{\alpha JM}$, describe the excitation process, the density matrix elements are given by

$$\langle\alpha JM|\rho|\alpha' J' M'\rangle = A_{\alpha JM} A_{\alpha' J' M'}^{*}. \quad (2)$$

The off-diagonal terms incorporate the relative phases of the excitation amplitudes. The diagonal elements are proportional to cross sections. Only the diagonal density matrix elements are easy to interpret physically.

Fano and Macek[2] introduced average values of tensor operators, J_{kq}, constructed from the angular momentum operator J, thereby providing an understandable description of the diagonal-block density matrix elements, $\rho_{\alpha J,\alpha J}$, involved in dipole radiation. No such easily understood description has yet been provided for the off-diagonal blocks ($\rho_{\alpha J\alpha' J'}$ with $\alpha J \neq \alpha' J'$). We naturally begin by using the familiar electric and magnetic multipole operators. Whereas average values of these operators provide a complete specification of the diagonal density matrix blocks, there are only enough operators to specify half of the off-diagonal density matrix elements. An *additional* set of operators is needed, similar to the multipole operators except with opposite time-reversal behavior. We choose to use the time-derivative operators of the electric and magnetic multipole operators. The operators are defined

as

$$Q_{kq}{}^{E}=\left(\frac{4\pi}{2k+1}\right)^{1/2} r^{k} Y_{kq}, \quad \dot{Q}_{kq}{}^{E}=i[H, Q_{kq}{}^{E}],$$
$$Q_{kq}{}^{B}=\nabla Q_{kq}{}^{E}\cdot\left(\frac{\vec{L}}{k+1}+\vec{S}\right), \quad \dot{Q}_{kq}{}^{B}=i[H, Q_{kq}{}^{B}]. \tag{3}$$

Our convention for spherical harmonics is that Y_{k0} are real.

Average values of these operators completely parametrize the density matrix, and provide additional insight into the physics contained in the off-diagonal elements. The details of the description will be discussed elsewhere. Here we only mention a few features. The operator average values are given by the familiar expression $\langle Q_{kq}\rangle = \mathrm{tr}[\rho Q_{kq}]$. If for ρ we substitute the sum of the density matrix components given in Eq. (1), we obtain the average value $\langle Q_{kq}\rangle$ as a sum of terms. We consider each of these terms to be an operator average value in its own right. Specifically,

$$\langle Q_{kq}\rangle_{\alpha JM,\alpha' J'M'}=\frac{1}{1+\delta_{\alpha JM,\alpha' J'M'}}\mathrm{tr}[(\rho_{\alpha JM,\alpha' J'M'}+\rho_{\alpha' J'M',\alpha JM})Q_{kq}]. \tag{4}$$

The two off-diagonal density matrix blocks, $\rho_{\alpha JM,\alpha' J'M'}$ and $\rho_{\alpha' J'M',\alpha JM}$ are included together because the Hermiticity condition determines one in terms of the other. The Kronecker δ in the denominator is included so the diagonal elements are normalized correctly. Nonzero average values can occur only when k, J, and J' satisfy the triangle inequality, when $M+M'+q=0$, and when the parity $(-1)^{L+L'}$ is equal to the parity of the operator. Either $\langle Q_{kq}{}^{E}\rangle_{\alpha LJM,\alpha' L'J'M'}$ or $\langle Q_{kq}{}^{B}\rangle_{\alpha LJM,\alpha' L'J'M'}$ will therefore vanish for all choices of αLJ and $\alpha' L' J'$. Figure 1 represents the density matrix for coherently excited states of principal quantum number $n=3$. Instead of density matrix elements $\langle LM|\rho|L'M'\rangle$ we have filled the density matrix with the operator average values needed to equivalently specify the density matrix.

For the case of electron-impact excitation of atomic hydrogen we now show that the (mixed-L) coherence multiple moments contain new information concerning the collision dynamics. In this Letter we consider only the case in which the scattered electron is not detected. The collision symmetry for excitation of $n=2$ states then permits only five nonzero excitation density matrix elements; three cross sections[3]; and one complex, off-diagonal term, $\sigma_{sp0}=\int\langle 00|\rho|10\rangle d\Omega$ for electrons incident along the z axis. In terms of operator average values[4] (in atomic units), we find that

$$\langle z\rangle_{sp}=-\langle Q_{10}{}^{E}\rangle_{sp}=-6\,\mathrm{Re}\sigma_{sp0}=-6\sigma_{sp0},$$
$$\langle \dot{z}\rangle_{sp}=\langle \dot{Q}_{10}{}^{E}\rangle_{sp}=A\sigma_{sp0}{}^{I}, \quad A=6.3\times10^{-6}, \tag{5}$$

where z is the distance from proton to electron.

The multipole picture leads us to consider the real and imaginary parts of σ_{sp0} rather than its magnitude and phase. Considering the effect of the incident electron upon a charge distribution of the bound electron, we can anticipate the properties of the multipole moments. We expect that both the electric dipole moment $\langle z\rangle_{sp}$ and its time derivative $\langle \dot{z}\rangle_{sp}$ are negative since most of the electrons are forward scattered and repel the atomic electron. Similarly, we expect that the quadrupole moment $\langle Q_{20}{}^{E}\rangle$, and its time derivative (for $n\geq 3$), are positive.

We now investigate the predictions of various scattering models for these moments in order to learn more about the dynamics of the collision. We begin by considering first-order perturbation theories. Since the plane-wave Born approximation (PWBA) is invariant under PT transformations (parity, time reversal) and is axially symmetric about the momentum-transfer direction, $\langle Q_{kq}{}^{E}\rangle_{LL'}$ and $\langle \dot{Q}_{kq}{}^{B}\rangle_{LL'}$ are identically zero for $L+L'$ odd, while $\langle Q_{kq}{}^{B}\rangle_{LL'}$ and $\langle \dot{Q}_{kq}{}^{E}\rangle_{LL'}$ are identically zero for $L+L'$ even. The distorted-wave

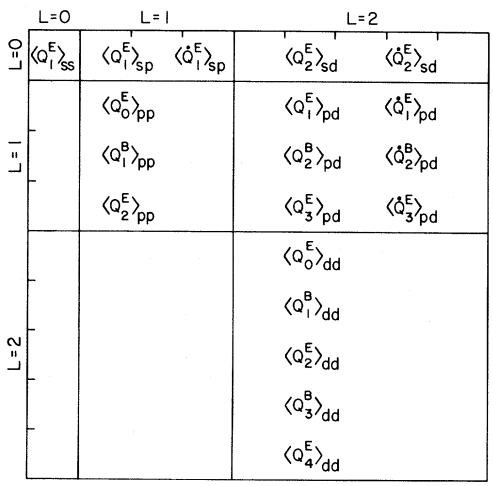

FIG. 1. Representation of the density matrix for coherently excited $n=3$ states using the operator average values needed to specify these states.

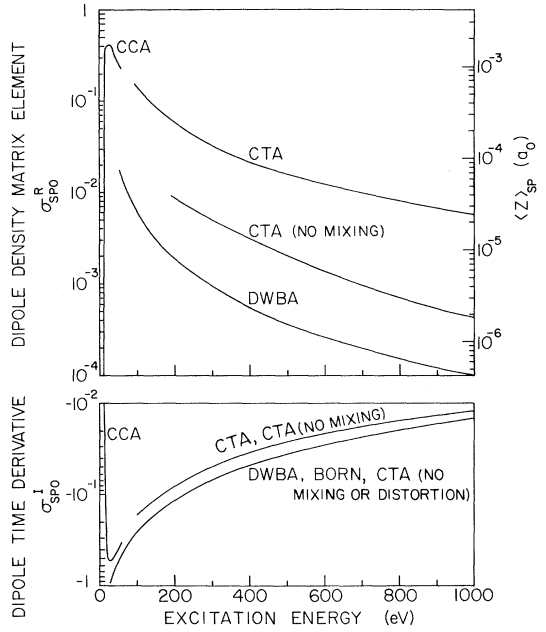

FIG. 2. Comparison of calculated $n = 2$ coherence parameters, $\sigma_{sp0}{}^R$ and $\sigma_{sp0}{}^I$, which are normalized as cross sections in atomic units.

Born approximation (DWBA) replaces the PWBA plane waves with scattering functions generated in the weak attractive potential of the ground-state hydrogen atom. The scattering-wave functions, however, deviate from plane waves only to first order in the scattering phase shifts, all of which are small.[5] The operator average values calculated in the DWBA thus possess the PWBA properties mentioned above to a very good approximation (see Fig. 2). For the $n = 2$ case we are considering in detail, the PWBA predicts $\langle z \rangle_{sp} = 0$. The DWBA predicts a small $\langle z \rangle_{sp}$.

The long-range nature of the Coulomb force, however, enables the departing electron to strongly mix the nearly degenerate excited states, thereby destroying the PT symmetry of the Born approximations and making perturbation calculations unsuitable. Therefore we turn to a close-coupling approximation (CCA) to incorporate this effect. We use published $1s$-$2s$-$2p$ R matrices[6] to calculate the low-energy values of σ_{sp0} shown in Fig. 2. Far from being zero, $\sigma_{sp0}{}^R$ is larger in magnitude than $\sigma_{sp0}{}^I$. Both $\sigma_{sp0}{}^R$ and $\sigma_{sp0}{}^I$ decrease rapidly as threshold is approached from above, because an angular momentum barrier enforces spherical symmetry. Since for low energies the couplings to higher excited bound and continuum states are small, the CCA values of σ_{sp0} should be quite accurate.

At higher energies, where close-coupling R matrices are not immediately available to us, we use instead a classical-trajectory approximation[5] (CTA) which we find extrapolates smoothly to the low-energy CCA values of σ_{sp0} and the cross sections. Transitions are produced in a quantum-mechanical hydrogen atom by the time-dependent potential of an electron following a classical trajectory in the attractive potential of a ground-state hydrogen atom. While cross sections and $\sigma_{sp0}{}^I$ calculated in the $1s$-$2s$-$2p$-$3s$-$3p$-$3d$ CTA are similar to the PWBA and DWBA values (thereby bolstering our confidence in this approximation), again we find that $\sigma_{sp0}{}^R$ is large (comparable in magnitude to $\sigma_{sp0}{}^I$), even above kilovolt energies. Repeating the CTA calculation setting the terms in the Hamiltonian which are able to mix the excited states equal to zero, we find that neither $\sigma_{sp0}{}^I$ or the cross sections are much affected, but $\sigma_{sp0}{}^R$ is orders of magnitude closer to the Born and DWBA values (Fig. 2). We conclude that the dipole moment, $\sigma_{sp0}{}^R$, is very sensitive to the final-state mixing brought about by the scattered electron as it leaves the hydrogen atom. If, in addition, we set the diagonal coupling terms of the interaction Hamiltonian equal to zero, we obtain values much closer to those from perturbation theory. The dipole moment $\sigma_{sp0}{}^R$ becomes very small, even smaller than the DWBA value. The dipole-moment time derivative $\sigma_{sp0}{}^I$ approaches the PWBA and DWBA values (Fig. 2). This indicates that the mixed-L coherence multipole σ_{sp0} is also sensitive to the distortions of the target bound states by the projectile electron. Since both final-state mixing and target distortion are large effects, the Born approximations and similar perturbation results are inappropriate for the calculation of hydrogen coherence.

Calculation of the coherent excitation of $n = 3$ produces the same qualitative results. Three dipole moments, ($\sigma_{sp0}{}^R$, $\sigma_{pd0}{}^R$, $\sigma_{pd1}{}^R$), and three dipole-moment time derivatives ($\sigma_{sp0}{}^I$, $\sigma_{pd0}{}^I$, and $\sigma_{pd1}{}^I$) are involved. Two new types of coherence terms are also present, an electric quadrupole moment $\sigma_{sd0}{}^R$, and its time derivative, $\sigma_{sd0}{}^I$. Whereas the Born approximation predicts that the dipole moments and $\sigma_{sd0}{}^I$ be identically zero for all energies, the CTA produces real and imaginary parts of comparable magnitude for all four complex coherence parameters. Glauber[7] and $1s$-$3s$-$3p$-$3d$ CCA, neglecting d couplings,[8] at 200 eV give similar features, presumably be-

cause some final-state-interaction effects are included.

At present, experimental results are available only for coherent excitation of hydrogen $n=3$ states by electron impact. For incident electron energies of 200 and 500 eV, Mahan and Smith[1] measured the total Balmer radiation emitted by the coherently excited state as a function of an electric field along the incident electron axis. The electric field is important because mixed-parity coherence parameters can only be detected when an electric field is used to mix the opposite-parity excited states. Krotkov first calculated this total intensity curve using PWBA,[9] but neglecting the σ_{sd0} terms. He found a large disagreement between theory and experiment.

Unfortunately, six cross sections and the eight coherence terms listed previously are involved in this intensity curve. To break this problem into more manageable parts, we replot the experimental data, along with the predictions of scattering models we have used, in two parts[5,10] (Fig. 3). The symmetric curve, $I(E)+I(-E)$, is linear in the even-parity cross sections and σ_{sd0}. The antisymmetric curve $I(E)-I(-E)$ is linear in the odd-parity dipole moments ($\sigma_{sd0}{}^{I}$, $\sigma_{pd0}{}^{I}$, $\sigma_{pd1}{}^{I}$), but still depends on the even-parity terms for overall normalization. The asymmetry produced by the electric-dipole time derivatives is sharply peaked at 20 V/cm and dies off by about 40 V/cm. Asymmetry due to the dipole moment terms (zero in PWBA) increases rapidly from 40 V/cm out to at least 100 V/cm. The concavity of the experimental difference curve therefore suggests that the dipole moments are comparable in magnitude to the dipole-moment time derivatives, as predicted by our calculations. Obviously, experimental points for larger electric fields are needed.

Careful choice of external electric and magnetic fields and the analysis of the polarization of the Balmer α radiation would allow measurement of the individual dipoles and the dipole time derivatives. Our calculations predict that the multipole coherence parameters are responsible for rich structure in the Stokes parameters[11] as a function of electric field applied parallel and perpendicular to the incident electron axis. The $n=2$ coherence parameters, $\sigma_{sp0}{}^{R}$ and $\sigma_{sp0}{}^{I}$ may also be measured. Our calculations suggest that the asymmetry is much larger for Lyman α photons detected in coincidence with forward-scattered electrons.

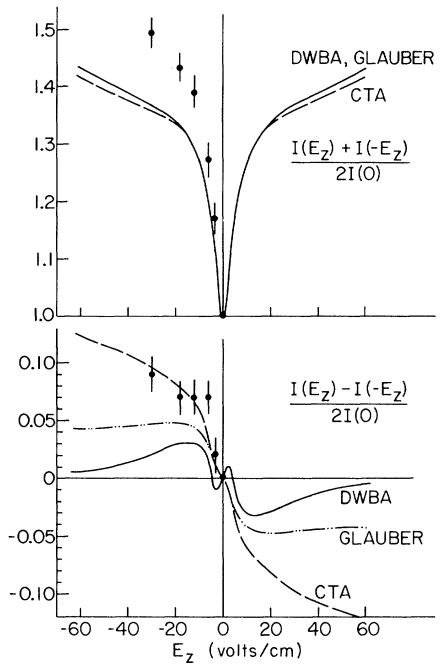

FIG. 3. Symmetric and antisymmetric H_α intensity vs applied external electric field for 200-eV electron impact energy. Data are taken from Ref. 2.

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