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Coulomb "Path" Interference in Low Energy He^+ + He Collisions

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Abstract

A new interference mechanism, analogous to "classic" double-slit electron scattering, has been identified in low energy ion-atom collisions. This Coulomb "path" interference results from the existence of two trajectories, indistinguishable with respect to laboratory energy and emission angle, along which ejected autoionizing electrons may be scattered by the attractive Coulomb potential of the slowly receding spectator ion. We present a simple semi-classical model for this effect in which we account for the path dependence of the amplitude of the ejected electron following decay of the autoionizing state. Calculated model lineshapes are found to be in excellent agreement with the strong angular dependence of the interference structure observed in the He target $2s^2\,{}^1S$ autoionizing lineshape measured near 0° following 10 keV He^+ + He collisions.

In low energy ion-atom collisions, post-collision interaction (PCI) of ejected autoionization electrons with the attractive Coulomb field of the slowly receding spectator ion leads to spectral lineshapes which are broadened and shifted towards lower electron energies. In the classical description, as first proposed by Barker and Berry[1], the spectral intensity as a function of laboratory electron energy is given by

$$I_{B.B.}(E) = \frac{\lambda_n}{\Delta E^2} \cdot \exp\left(\frac{-\lambda_n}{\Delta E}\right) \qquad (1)$$

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where $\lambda_n = q\Gamma_n/v$, is a measure of the strength of PCI on a given state with decay width $\Gamma_n = 1/\tau_n$, and q is the charge of the ion moving with a velocity v. The parameter $\Delta E = \varepsilon_n - E = q/R$ is the gain in potential energy of the electron ejected when the autoionizing state, with a resonance energy ε_n, decays at an internuclear separation R. Equation (1) represents a asymmetrically broadened lineshape with a peak intensity at an energy $E_{PEAK} = \varepsilon_n - \lambda_n/2$. In this model the velocity of the ejected electrons $u_n = \sqrt{2\varepsilon_n}$ (atomic units) is assumed to be much greater than that of the receding ion, $u_n \gg v$, and thus the ejected electrons follow straight line trajectories as they emerge from a *time independent* Coulomb field. However, for those electrons with velocities of the order of the ion velocity, $u_n \approx v$, and that are emitted when the collision partners are still comparatively close together, this approximation is no longer valid. The ion's field, in this case, causes significant deflection of the electron's classical trajectory. Such Coulomb scattering results in an emission angle dependent perturbation of the lab-frame electron energy shift, which has been given by van der Straten et al.[2] as $\Delta E = q/R + \mathbf{v}\cdot\Delta\mathbf{u}_n$, where $\Delta\mathbf{u} = \mathbf{u}_n - \mathbf{v}$ is the total change in the electron's velocity vector due to the Coulomb deflection. As more recently pointed out[3], such deflections effectively compress the solid angle into which these electrons are emitted, resulting in a strong enhancement or "focusing" of intensity in the direction of the receding ion. This effect was shown to be important in low energy $He^+ + He$ collisions and a classical time-dependent model, which was presented, accurately described the emission angle, lifetime, and collision energy dependences of the observed enhancement.

The influence of the spectator ion's Coulomb field on the electron's classical trajectory is illustrated in Figure 1 for low energy $He^+ + He$ collisions. Autoionizing states (He^{**}) formed in the collision decay exponentially with a lifetime τ_n emitting an electron with a velocity u_n at a time t when the collision partners are separated by a distance $R(t) = vt + \delta$. The "unknown" quantity δ is the separation at $t = 0$ at which the autoionizing state is populated. The key new observation is that, for a localized source of autoionizing electrons and an attractive potential, there are different paths, (corresponding to slightly different emission times) by which the electrons may emerge from the collision with a given laboratory energy $E = \frac{1}{2}\upsilon^2$ and emission angle θ. Two such trajec-

tories (denoted A and B) are illustrated in Figure 1. Here u_n, υ and ϑ, θ are the electron velocity and emission angle after before and after scattering, respectively. Since paths A and B represent two indistinguishable paths to the same final electronic state, interference may occur. This Coulomb "path" interference mechanism is seen to represent an ion-atom collision analogue to "classic" double-slit electron scattering where the scattering center now serves to define as continuous set of trajectories, having a fixed emission angle θ, which are distributed along the electron energy axis by a *time-dependent* Coulomb field. A similar interference phenomenon has been previously observed in the form of "Rainbow ghosts" in near-side far-side nuclear scattering by Satchler et al.[4]

In this paper, we present evidence for this Coulomb path interference mechanism in the form of an anomalous interference structure in the Coulomb-focused PCI-broadened lineshape of the target $2s^2\ ^1S$ autoionizing state produced in 10 keV He^+ + He collisions. This feature is observed to change in character dramatically over a very narrow range of emission angles near 0°. This unusually rapid angular variation is explained semi-classically by making a simple extension of the "overlapping resonances" (OR) model of Morgenstern et al.[5] in order to include the new Coulomb path interference mechanism. Using the WKB approximation for scattering in a central field[6], we account for the path dependence of the amplitude of the ejected electron following the decay of the autoionizing state. The result of our analysis is an interference lineshape which reproduces the observed structure in the experimental $2s^2\ ^1S$ lineshape as well as its rapid variation with emission angle.

Measurements were made at the Oak Ridge National Laboratory ECR ion-source facility using a high resolution electron spectrometer and target chamber temporarily transported from Hahn-Meitner Institut, Berlin. Details of the experimental technique have been described previously[3,7]. Figure 2a shows the He target autoionization spectra, acquired in non-coincidence or "singles" mode, in the region of 30-35 eV, measured at 0°, 5°, and 10° relative to the incident beam direction for 10 keV He^+ + He collisions. The spectra shown are normalized to a constant maximum intensity at each angle to remove the strong enhancement due to Coulomb focusing[3]

which is observed near 0°. The three autoionizing states which are predominantly populated in these low energy collisions (see figure) are the $2s^2\ ^1S$, $2p^2\ ^1D$, and $2s2p\ ^1P$ states. Note the pronounced shoulder in the $2s^2\ ^1S$ lineshape which becomes equal in intensity to the main part of the peak at 5°, and has essentially disappeared in the 10° spectrum. This uncharacteristically rapid angular variation in the ^1S lineshape occurs in precisely the same angular range as that where the effects of Coulomb focusing are found to be important[3]. This similarity leads to the conclusion that, just as the intensity enhancement, the observed anomalous ^1S lineshape may have as its origin the post-collision Coulomb deflection of the ejected autoionizing electrons.

Analysis of the measured lineshapes was performed using, as a starting point, the overlapping resonances model (OR) of Morgenstern et al.[5]. The OR model was first introduced in order to explain the complicated interference structures observed in quasi-molecular electron emission in 1400 eV He^+ + He collisions observed at 180°. Because of the induced PCI broadening, the spectral lineshapes of the $2s^2\ ^1S$, $2p^2\ ^1D$, and $2s2p\ ^1P$ states overlap. As a result of the indistinguishability of the detected electrons in the region of overlap, the composite lineshape is calculated as a coherent sum of amplitudes for emission from each of the three autoionizing resonances. Our semi-classical extension of this treatment derives from the observation that the OR model takes into account the phase evolution of the autoionizing state only from the time of its crossing into the continuum to the time of its decay. The emitted electron is then assumed to "instantaneously" recede to infinity and thus produces no additional accumulation of phase. However, for He target autoionizing electrons produced in low energy He^+ + He collisions, where the electron velocity is comparable to that of the projectile ion ($u_n/v \approx 5$) and where the path through the ion's field depends strongly upon the ejection angle and the internuclear separation R(t) at the time of emission, this approximation is no longer valid. A correct description of the near-forward autoionizing spectrum must therefore include the scattering of the emitted electron by the spectator ion.

In the semi-classical approximation we write the angle dependent spectral intensity at a given electron energy as

$$I(E,\theta) = \left| \sum_{\text{paths}} \sum_{n} W_n(\vartheta,\varphi)\cdot C(\varepsilon_n,\lambda_n,E,\theta)\cdot \exp\left\{-i\left[\alpha(\varepsilon_n,v,E) + \beta(p,l)\right]\right\} \right|^2 \qquad (2)$$

where $\sum_n$ is the sum over the n autoionizing states (^1S, ^1D, and ^1P) which contribute to the observed intensity at E and θ. Coulomb scattering by the receding ion enters equation (1) through; (a) the modification of the modulus of the amplitudes $C(\varepsilon_n,\lambda_n,E,\theta)$ due to Coulomb focusing; (b) inclusion of the semiclassical scattering phases $\beta(p,l)$; and (c) the coherent sum over the different paths (A and B). The factor $W_n(\vartheta,\varphi) = \sum_{m_\ell} a_n^{m_\ell}(0)\cdot\omega_n^{m_\ell}(\vartheta,\varphi)\cdot\exp[-i\chi_n^{m_\ell}(0)]$ is the angle dependent autoionization transition amplitude for a given state which consists of contributions from different magnetic sublevels m_ℓ, where $a_n^{m_\ell}(0)$ and $\chi_n^{m_\ell}(0)$ are the initial level populations and phases, and where (ϑ,φ) is the angular coordinate of the outgoing electron prior to Coulomb scattering. Since the angular dependence of $W_n(\vartheta,\varphi)$ produces only a weak effect on the calculated lineshape for the post-scattered emission angle range $0° \le \theta \le 10°$, and the phase $\alpha(\varepsilon_n,v,E)$ given by the OR model is essentially angle independent, any significant angular variation of the calculated linshapes (apart from the intensity enhancement due to Coulomb focusing) will be solely due to the path dependent interference resulting from inclusion of the scattering phase $\beta(p,l)$.

The modulus of the amplitude $C(\varepsilon_n,\lambda_n,E,\theta)$ is derived from the square-root of the doubly-differential cross section given in reference 1 and is given by

$$C(\varepsilon_n,\lambda_n,E,\theta) = \left\{ I'_{B.B.}\cdot\frac{\varepsilon'}{\varepsilon_n}\left[\frac{E}{E'}\left(1 - \frac{E_p}{\varepsilon_n}\sin^2\vartheta'\right)\right]^{1/2}\cdot\frac{\partial\cos\vartheta'}{\partial\cos\theta'}\right\}^{1/2}, \qquad (3)$$

which consists of the Barker-Berry lineshape given in equation (1) modified by a Coulomb focusing function. The prime (') indicates those quantities evaluated in the ion frame. By making the kinematic transformations $E,\theta \rightarrow E',\theta'$ and $\varepsilon_n,\vartheta \rightarrow \varepsilon',\vartheta'$ to the the reference frame of the projectile ion, we have simplified the inherently time-dependent problem to one of electron scattering in a time-independent Coulomb field. The scattering function $\partial\cos\vartheta'/\partial\cos\theta'$ is then derived from the solution to the equation for the classical trajectory in a central field[8] which relates the emission angles in the projectile frame ϑ',θ' before and after scattering, respectively.

In the projectile frame, the phase angle $\alpha(\varepsilon_n, v, E)$ given by the OR model[5] is written as

$$\alpha(\varepsilon_n, v, E) = \frac{q}{v}\left[1 - \frac{(\Delta E')}{q}\delta + \log\left(\frac{\Delta E'}{q}\right)\right] \tag{4}$$

where $\Delta E' = \varepsilon' - E' = q/R$ is the ion-frame electron energy shift. We note that for $u_n \gg v$, or in the limit as $v \to 0$, the energy shift $\Delta E' \to \Delta E$. Also, the resulting interference structures arising from the phase differences resulting from equation (2) are frame independent.

We derive the path dependent scattering phase from a classical action integral in the central field WKB approximation[6], giving the result

$$\beta(p, r, l) = \left[\frac{q}{p_o}\ln\left|2r\left(1 + \frac{p_r}{p_o}\right) + \frac{2q}{p_o^2}\right| - l\cdot\sin^{-1}\left(\frac{q - l^2/r}{\sqrt{q^2 + l^2 p_o^2}}\right) + l\cdot\cos^{-1}\left(\frac{l}{p\cdot r}\right)\right] \tag{5}$$

which, when evaluated at the appropriate limits of the classical trajectory, yields the scattering phase $\beta(p, l)$. The term $p_r = \sqrt{p^2 - l^2/r^2}$, is the radial component of the electron's linear momentum p, given by $p = \sqrt{p_o^2 + 2q/r}$, p_o is its linear momentum at $r = \infty$, and l, its angular momentum. For convenience we have left off the primes on the various momenta and radial coordinate r since they are all ion-frame quantities.

Figure 3 shows the results of the evaluation of the OR phase $\alpha(\varepsilon_n, v, E)$ and the scattering phase $\beta(p, l)$ over the range of electron energies corresponding to the $2s^2\,{}^1S$ lineshape for an emission angle $\theta = 5°$. The slight difference in the results for the OR phases α_A and α_B (for paths A and B) are the result of the different emission angles (ϑ_A and ϑ_B) and thus ion-frame electron energies for the near and far side scattered electrons. The scattering phase for the near side path $\beta_A \to 0$ as the electron energy approaches the resonance energy $E \to \varepsilon_n = 33.28$ eV for the ${}^1S$ state, since the scattering angle $\Theta = \theta - \vartheta \to 0$ when the internuclear separation at the time of emission $R(t) \to \infty$ and therefore the Coulomb field $q/R(t) \to 0$. For the far side path, the scattering phase $\beta_B(E \to \varepsilon_n) \to \infty$ due to the fact that the emission angle $\vartheta_B(E \to \varepsilon_n) \to 0°$ for far side scattering into any angle $\theta > 0°$, which results in scattered electron passing through the origin at $r = 0$. This contrast-

ing behavior in the scattering phase for the near-side and far-side paths is the source of the rapid angular variation in the interference structure observed in the $2s^2\,{}^1S$ lineshape near the forward direction.

Figure 2b shows the result of our calculations using equation (2) evaluated at 0.1°, 5°, and 10°, convoluted with a Gaussian spectrometer resolution function with a FWHM = 0.3 eV, and normalized to a constant maximum intensity at each angle. Calculations are done at 0.1° due to the singularity which exists in the Coulomb focusing differential cross section at 0°[3]. For simplicity we assume each state consists of only a single magnetic sublevel which decays isotropically (i.e. $W_n(\vartheta,\varphi) = a_n(0)\cdot\exp[-i\chi_n(0)]/\sqrt{4\pi}$) as would be the case for an S state. We then have four parameters, consisting of the initial ${}^1S$ and ${}^1D$ level populations ($a_{1S}(0)$, and $a_{1D}(0)$) and phase angles ($\chi_{1S}(0)$, and $\chi_{1D}(0)$) relative to the ${}^1P$ amplitude ($a_{1P}(0) = 1$, $\chi_{1P}(0) = 0$), with which we fit the experimental spectrum at 0°. Holding these values fixed, we then simply vary the emission angle to obtain the calculated lineshapes at 5° and 10°. The solid curves in Figure 2b are the results of this fit ($a_{1S}(0) = 0.3$, $a_{1D}(0) = 0.4$, $\chi_{1S}(0) = 0.7$, and $\chi_{1D}(0) = 1.0$) which include Coulomb path interference. We find that this model reproduces remarkably well the observed angular dependence of the interference structure in the ${}^1S$ state. The dashed curves are these same calculations, where we have now excluded the "far-side" or path B amplitude, thus suppressing the path interference mechanism. At 0.1° this calculation is essentially identical to the solid curve since all pairs of interfering paths are nearly equivalent for emission angles very near 0°, which results in completely constructive interference. Although the path dependent phase is included in this "near-side only" calculation, the angular variation in the lineshape is negligible over this range of emission angles. Clearly, only by including *both* "near-side" and "far-side" paths in the calculation do we reproduce the rapid angular dependence of the interference structure observed in the data. For comparison we show the lineshape obtained from the OR model alone which we obtain by suppressing both the path dependence of the amplitude and Coulomb focusing in equation 1. This result is shown in Figure 2b at 0°, as the dot-dashed curve.

Finally, we calculate the Coulomb interference lineshape for the case of an isolated resonance, keeping only the term in equation (1) corresponding to the $2s^2\,{}^1S$ state. The result is shown in Figure 4 for a range of emission angles near 0°. Here we have normalized the lineshapes at each angle to the maximum intensity in the incoherent lineshape to again remove the strong Coulomb focusing enhancement found near 0° and to show the change from constructive interference near 0°, where all interfering paths are equivalent, to destructive interference which severely distorts the lineshape from its monotonic incoherent form at larger emission angles. Here, we also obtain an angular dependent lineshape similar to that which is observed when we include the contributions from the ${}^1P$ and ${}^1D$ states above. We note that there is qualitative agreement with the calculated lineshapes of Barrachina and Macek[9] obtained using a CDW approximation to the final state in the autoionizing matrix.

In summary we have provided strong evidence for the new Coulomb path interference which manifests itself in low energy ion atom collisions where post-collision deflection and focusing of ejected autoionizing electrons is found to be important. A semi-classical model was found to successfully describe the phenomenon where indistinguishable classical trajectories, along which autoionizing electrons are scattered by the attractive Coulomb potential around opposite sides of the slowly receding spectator ion, interfere to produce a strongly angular dependent structure in the coherent spectral lineshapes. We emphasize that this path interference will also manifest itself in a single isolated autoionizing state in the absence of any contributions from adjacent overlapping resonances. Observation of such isolated resonance lineshapes would provide a clear test of Coulomb path interference as well as post-collision interaction effects in these collisions.

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FIGURES

Figure 1. Effect of the spectator ion's Coulomb field on the classical trajectories of ejected target autoionizing electrons following low energy He^+ + He collisions.

Figure 2. a) He target autoionizing spectra obtained following 10 keV He^+ + He collisions. b) Calculated lineshapes using the equation (1); (solid) including Coulomb interference; (dashed) where scattering around *only* the "near-side" of the ion is considered; (dot-dashed) Calculated lineshape at 0° using "overlapping resonances" model[3] alone. (See text).

Figure 3. Calculated phases $\alpha(\varepsilon_n, v, E)$ and $\beta(p, l)$ for the $2s^2\ ^1S$ state evaluated at $\theta = 5°$ for near-side and far-side scattered trajectories (paths **A** and **B**).

Figure 4. Calculated lineshapes for the isolated $2s^2\ ^1S$ resonance at 0.1°, 2°, 4°, 6°, 8°, and 10° using the extended overlapping resonances model including Coulomb interference.

Figure 1.

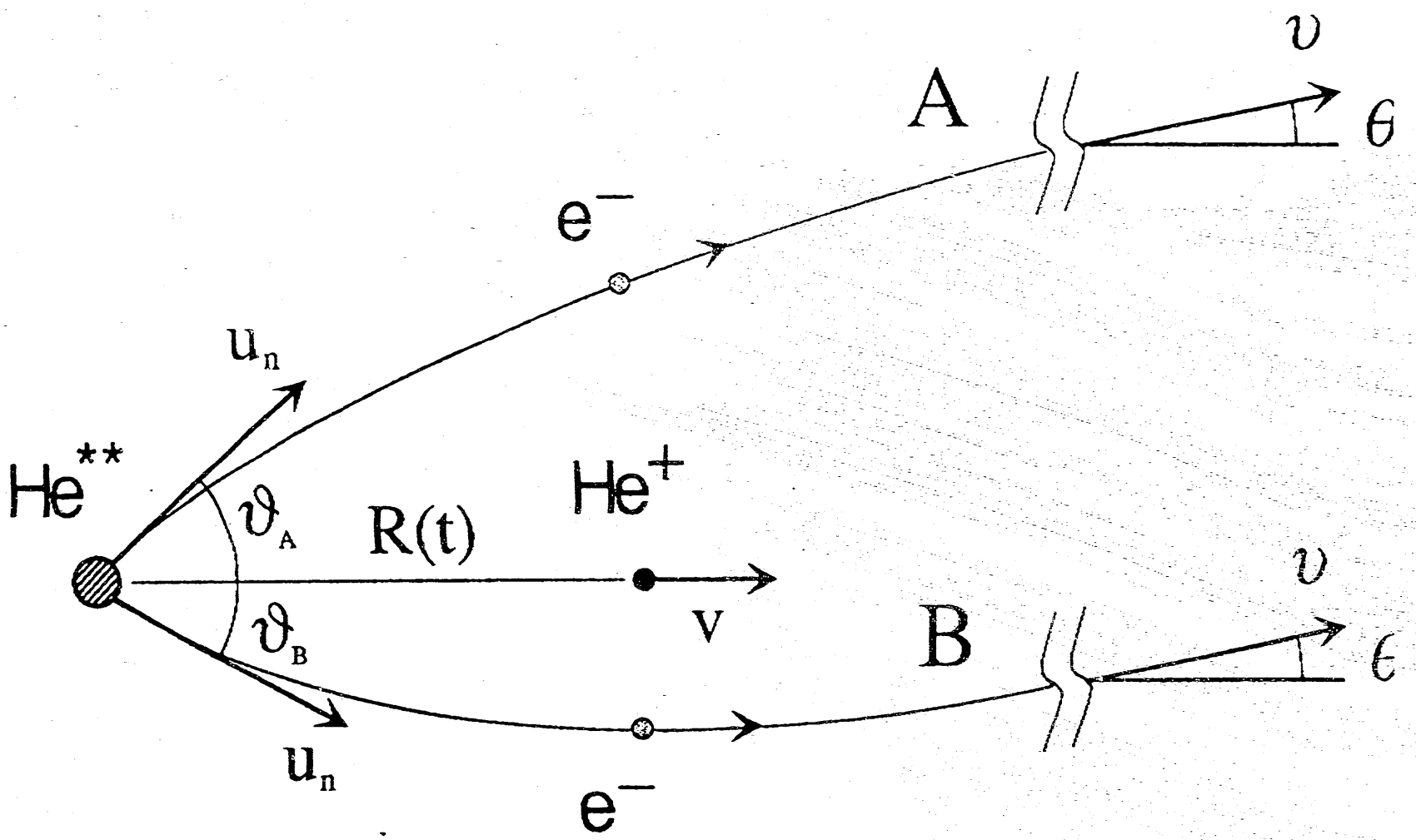


Figure 2.

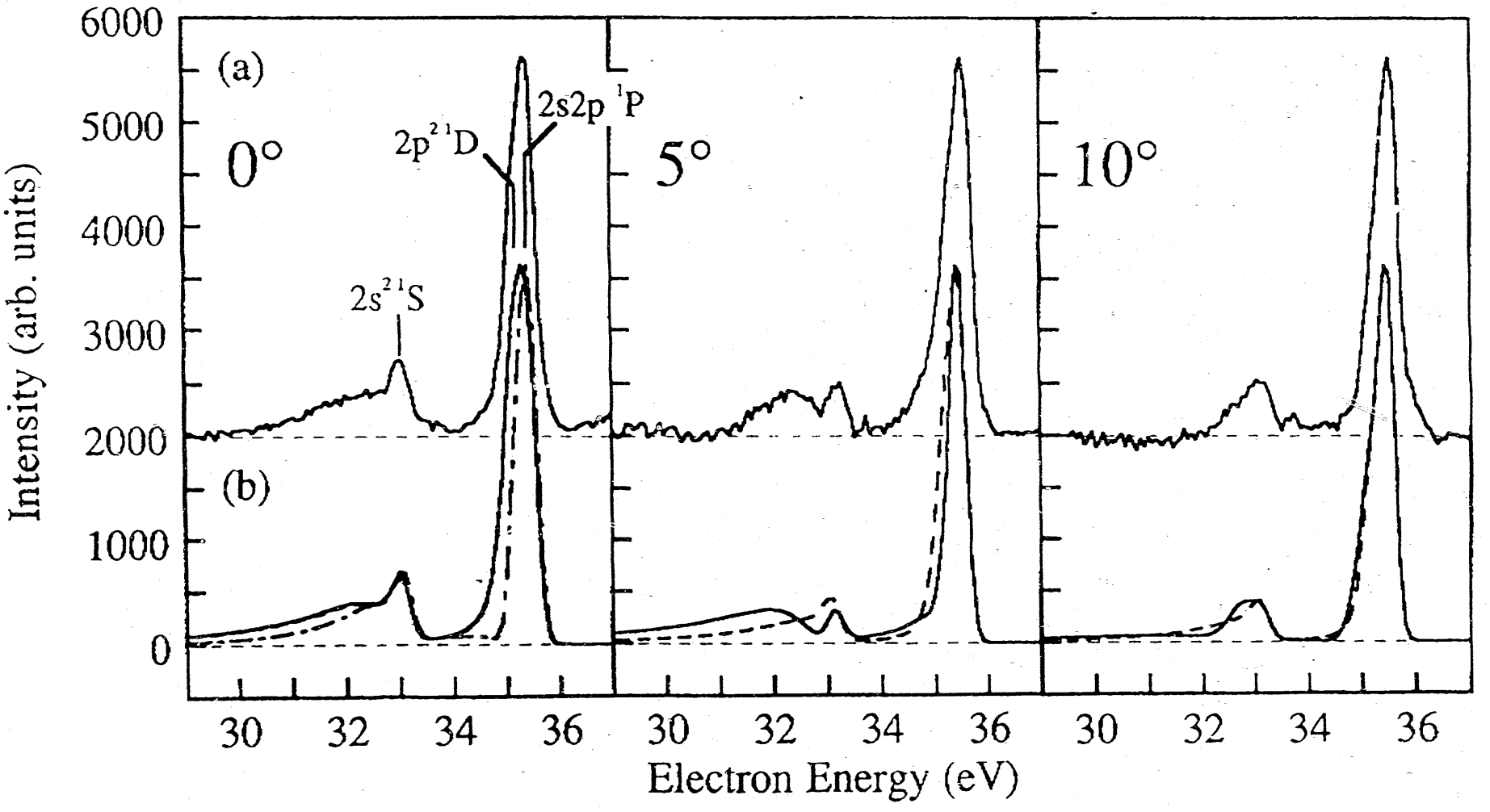

6000
5000
4000
3000
2000
1000
0
Intensity (arb. units)
(a)
(b)
0°
5°
10°
2p² ¹D
2s2p ¹P
2s² ¹S
30
32
34
36
Electron Energy (eV)

Figure 3.

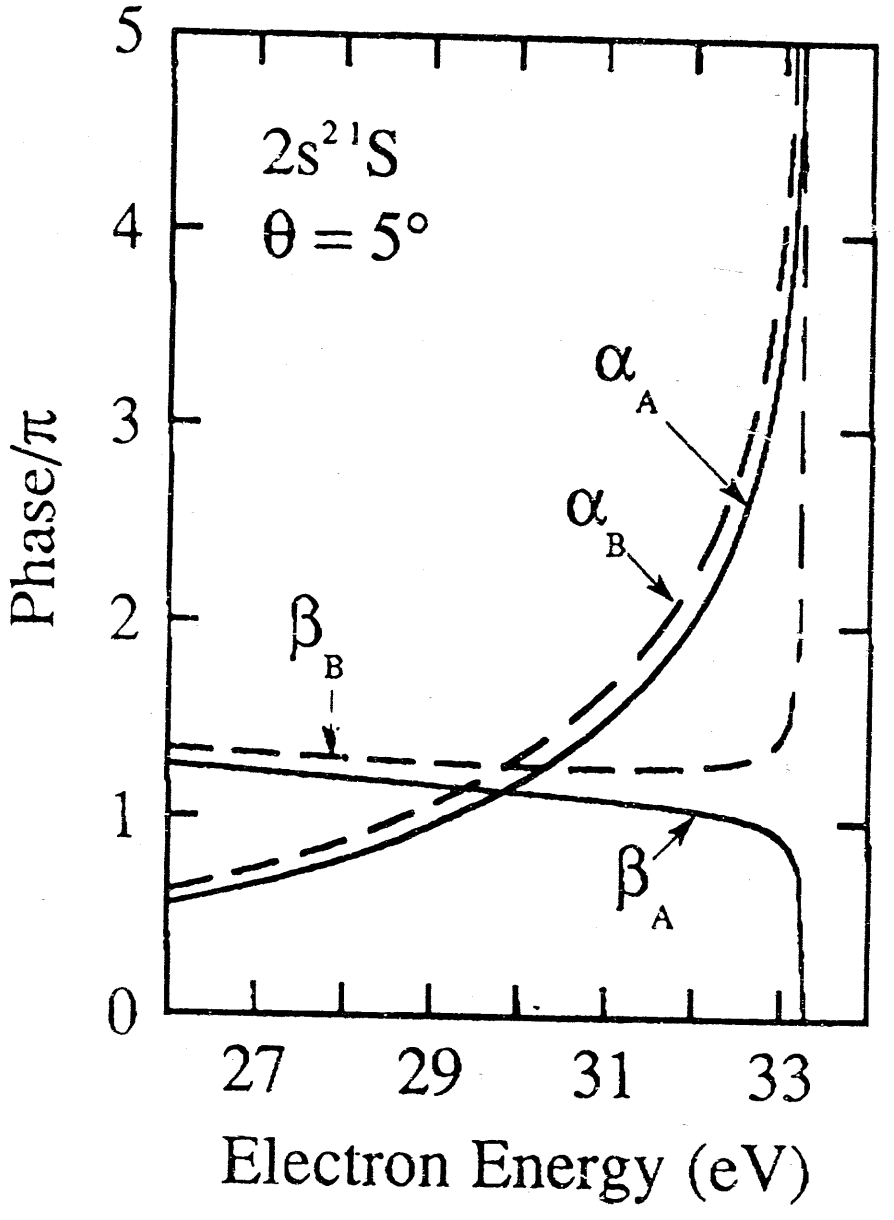
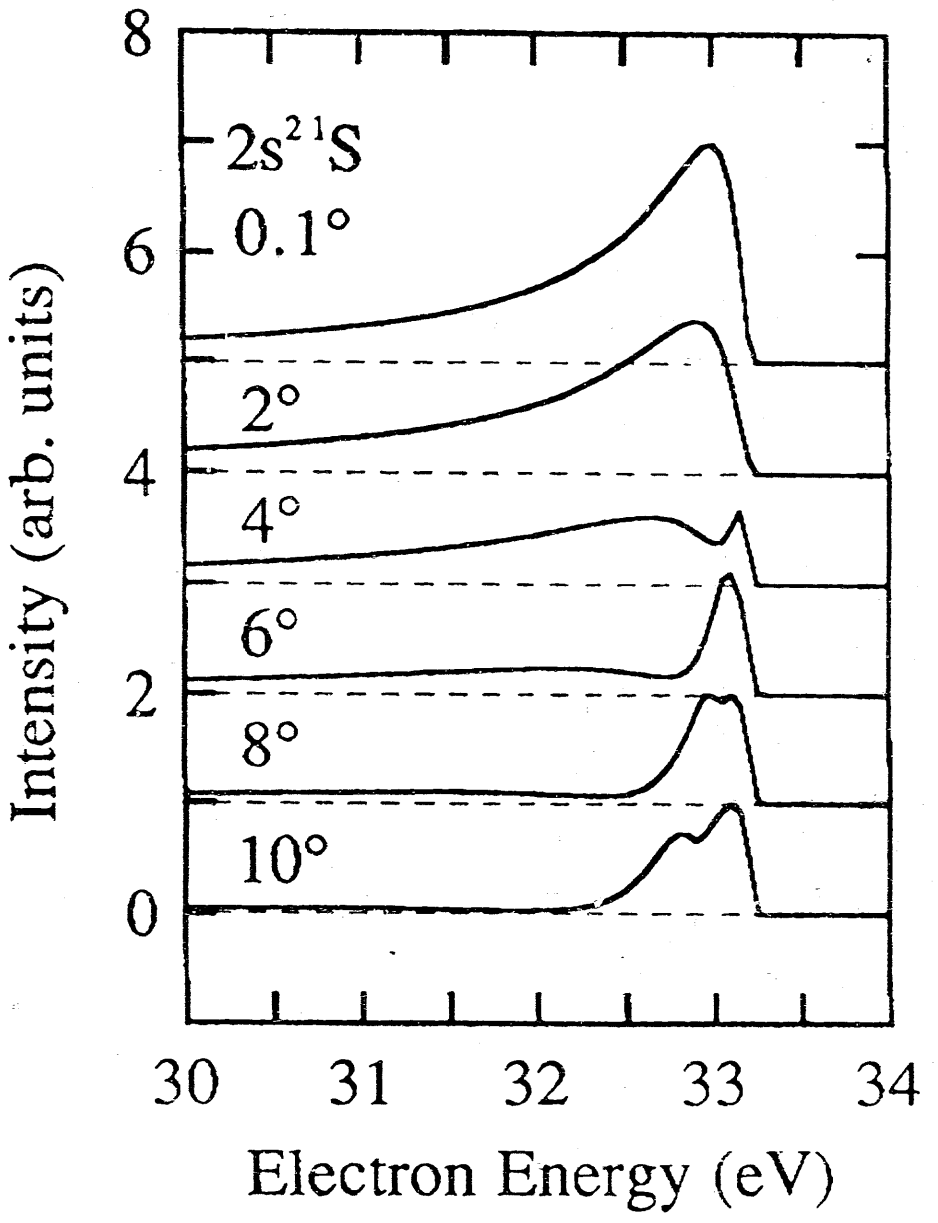

$2s^2\,{}^1S$
$\theta = 5°$
α_A
α_B
β_B
β_A
Phase/π
0
1
2
3
4
5
27
29
31
33
Electron Energy (eV)

Figure 4.


8
6
4
2
0
2s²¹S
0.1°
2°
4°
6°
8°
10°
Intensity (arb. units)
30
31
32
33
34
Electron Energy (eV)

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