of the field is correlated with the energy loss of the source even though the field and source do not interact dynamically when the field energy is measured.

*This research was sponsored by U. S. Air Force Project RAND and by the U. S. Army Research Office (Durham).

[1]I. R. Senitzky, Phys. Rev. Letters 15, 233 (1965).

[2]Senitzky uses quotation marks around the word detector because the interaction under consideration involves all systems coupled to the radiation field other than the source.

[3]The Hanbury Brown–Twiss experiment, the quantum mechanical nature of laser radiation, and the validity of semiclassical coherence theory (references 6-9 of Senitzky's paper).

[4]L. I. Schiff, Quantum Mechanics (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed., Chap. X.

[5]Senitzky's claim that certain commutators vanish contradicts this view, and can easily be shown to be incorrect by explicit calculation.

PERTURBATION INDUCED IN ELASTIC SCATTERING BY CROSSING OF MOLECULAR STATES*

F. T. Smith, D. C. Lorents, W. Aberth, and R. P. Marchi

Stanford Research Institute, Menlo Park, California

(Received 28 September 1965)

Differential ion-atom scattering is becoming a sensitive tool for the study of interactions in diatomic systems. For ease of both experimental and theoretical study, the system $He^+ + He$ excels. Its differential elastic scattering spectra combine sufficient structure to be interesting with sufficient simplicity to be intelligible. The most pronounced feature is a regular oscillation representing interference between scattering in even (g) and odd (u) electronic states of the ion He_2^+. As a result of a good deal of experimental[1,2] and theoretical[3,4] study, this interference is rather well understood. At larger angles it is supplemented, in the case of identical nuclei, by a secondary interference pattern arising from the nuclear symmetry,[2,4,5] while at small angles and energies ($E\theta \lesssim 250$ eV deg) one observes the expected rainbow structure arising from the attractive portion of the odd (u) potential.[2,4]

From a rather early stage in our study of the simple oscillations, we had noted that their regularity was marred by reproducible perturbations at certain points, and one of us suggested that some of these irregularities might be due to the crossing (or "pseudocrossing") of two molecular electronic curves of the same symmetry.[6] We now believe we have identified one of these features with a particular crossing, and we wish to point out how such a perturbation can be used to estimate both the location of such a crossing and the magnitude of the interaction energy.

The feature in question is clearly shown in Fig. 1 in the region near $\theta_{lab} = 17°$ on the curve representing the scattering spectrum of $He^+ + He$ at $E_{lab} = 100$ eV: The peak at 17° is notably taller than the envelop of its neighbors, and the next peak on the left, at 14°, is distorted in shape and lowered; the valleys at 13.5° and 19° are raised above the smooth curve through their neighbors, while the one between them, at 16°, is approximately normal. Figure 1 also shows examples of similar patterns of rises

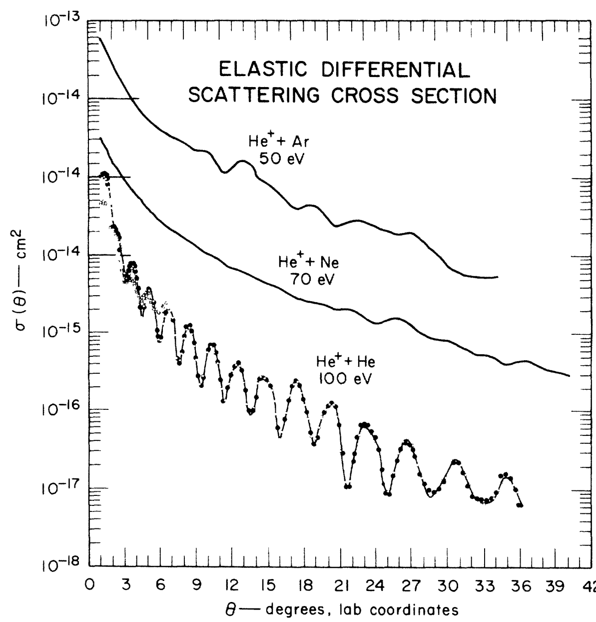

FIG. 1. Perturbation effects in differential elastic-scattering patterns. For the $He^+ + He$ system the actual experimental points are shown and the data are taken from reference 2. The $He^+ + Ar$ and $He^+ + Ne$ data are from unpublished work of Aberth and Lorents.

and dips that appear in the asymmetric scattering systems $He^+ + Ne$ and $He^+ + Ar$, where they are free of the distracting interference peaks.

If this sort of feature is indeed due to a curve crossing, the interaction responsible for it will be felt most strongly in a fairly narrow region of width Δr about the internuclear distance r_x where the crossing point is located. Furthermore, the interaction will affect the scattering most severely in those cases where the particles are moving most slowly through this interaction region, i.e., in those cases where the classical turning point r_0 is close to the crossing point r_x. At the energies of these experiments the turning point r_0 is closely related to the impact parameter b, and thus to the reduced scattering angle[7] (a quantity which depends mainly on b and hardly at all on E):

$$\tau = E\theta = \tau(b, E) = \tau_0(b) + E^{-1}\tau^1(b) + \cdots. \quad (1)$$

Any scattering feature associated with a particular turning point will therefore show up at about the same reduced angle τ when the energy is changed—and also when the masses are changed by isotopic substitution.

In all three cases, $He^+ + He$, Ne, and Ar, the features in question move in the same way as the energy is changed, so that they are found at substantially constant τ. In Table I we list the values of τ associated with the most prominent exalted maximum in the system $^4He^+ + {}^4He$ at five energies, and in $^4He^+ + {}^3He$ at two energies. In all these cases the structure is generally similar to the one described above for $E_{lab}{}^{4,4} = 100$ eV. Several of the examples can be dimly discerned by careful study of Fig. 6 of reference 2. The fact that this feature occurs at substantially identical reduced angles τ independently of energy and isotopic species is consistent with the hypothesis that it arises from some localized feature of one of the two potentials, $V_g(r)$ or $V_u(r)$, centered about some point r_x.

Table I. Location of the curve crossing: Reduced angle and even (g) turning point for the perturbation at several energies.

	E (eV)	$E\theta$ (10^3 eV deg)	R_0 (Å)
$^4He^+ + {}^4He$	200	1.52	0.91
	150	1.50	0.91
	100	1.64	0.89
	50	1.74	0.87
	35	1.90	0.89
$^4He^+ + {}^3He$	85.7	1.65	0.88
	42.9	1.80	0.85

It seems clear from data in some of the examples, such as Fig. 1, that the scattering perturbation affects both the upper (+) and lower (−) envelopes of the interference pattern in parallel. These envelopes are connected with the g and u cross sections by the formulas[4,6]

$$\sigma_+(E,\theta) = \sigma_g(E,\theta) + \sigma_u(E,\theta),$$
$$\sigma_-(E,\theta) = \sigma_g(E,\theta) - \sigma_u(E,\theta), \quad (2)$$

since $\sigma_g(E,\theta) > \sigma_u(E,\theta)$ for all angles greater than the rainbow angle $(\theta_u)_r$. Since we observe that σ_+ and σ_- rise and fall in parallel at the perturbation, the effect must occur in σ_g and not in σ_u (see Fig. 2). This is fortunate for the crossing hypothesis, since there are crossings in $V_g(r)$ but not on $V_u(r)$, as may be seen in Lichten's potential curves.[3]

Naturally, each potential V_g and V_u has separate turning points appropriate to each pair of variables (θ, E) or (τ, E). From previous calculations[4] we know the function

$$r_{0,g}(\tau, E) \cong r_{0,g}{}^0(\tau) + E^{-1} r_{0,g}{}^1(\tau) + \cdots, \quad (3)$$

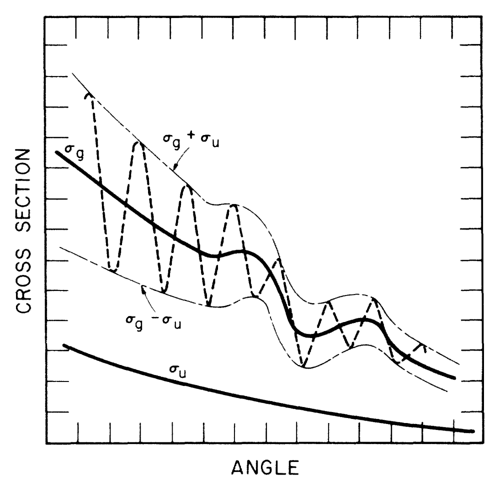

FIG. 2. Hypothetical example of a perturbed elastic-scattering pattern. σ_u and σ_g are the differential cross sections from ungerade and gerade potentials. The oscillating dashed line represents the scattering resulting from the interference of the gerade and ungerade states [see reference 4]. Since the perturbation lies in the σ_g curve, the undulations in the upper and lower envelopes are in phase. If the perturbation were in the σ_u curve, the undulations in the envelopes would be 180° out of phase.

from which we have computed the values of r_0 given in Table I. These values lie between 0.85 and 0.91 Å. These should probably be increased by a few hundredths of an Å to take account of the fact that τ_x should not be located at the major maximum but perhaps rather at the minimum to its left. We conclude that the crossing occurs at a location $r_x^{\rm g}$ close to 1.0 Å. Since there is no indication of any other feature of this type at larger $r_x^{\rm g}$ or smaller τ_x [other than the well-understood rainbow structure at $(\tau_{\rm u})_r \approx 250$ eV deg], we conclude that this represents the outermost crossing among the g states. In the curve sketched by Lichten,[3] this crossing occurs at about 1.1 Å.

Since the interference fringes march past τ_x as the energy E is shifted, they can be used as a probe to explore the detailed shape of the perturbation $\Delta\sigma_{\rm g}(\tau, E)$ that is superposed on the smooth background $\sigma_{\rm g}(\tau, E)$ due to the diabatic potential $V_{\rm g}(r)$. We may expect the shape to be characterized by a width $\Delta\tau_x$ and a height $\lambda_x = \Delta\sigma_{\max}/\sigma_+(\tau_x)$. A reliable evaluation of these parameters must await a further series of experiments at close energy intervals, but they can be roughly estimated from the spectra we already have. The fact that one and only one heightened maximum is always seen, and that a raised minimum is usually associated with it, implies that the width $\Delta\tau_x$ is not far different from the wavelength of the interference pattern. From this we conclude roughly that $\Delta\tau_x \approx 300 \pm 100$ eV deg. The height parameter is found directly from the spectra to be $\lambda_x \approx 0.3$.

The characteristic width Δr_x of the interaction region where the potentials are strongly perturbed by the crossing can be determined directly from $\Delta\tau_x$ by using

$$\Delta r_x = (\partial r_0^{\rm g}/\partial\tau)_x \Delta\tau_x. \tag{4}$$

From the relationship (3) we know the value $(\partial r_0^{\rm g}/\partial\tau) \cong 2\times10^{-4}$ Å/eV deg, from which we find $\Delta r_x \approx 0.06 \pm 0.02$ Å. From this interaction width we can estimate the interaction energy $H_{12}^{\rm g}(r_x)$, by taking note of the fact that a natural measure of the interaction region is the quantity

$$R = H_{12}(r_x) \Big/ \left|\frac{\partial V_1}{\partial r} - \frac{\partial V_2}{\partial r}\right|_{r_x}. \tag{5}$$

From Lichten's curves we obtain

$$\left|\frac{\partial V_1}{\partial r} - \frac{\partial V_2}{\partial r}\right| \cong 35 \text{ eV/Å},$$

from which, if we make the natural identification $R = \Delta r_x$, we find

$$H_{12}^{\rm g}(r_x) \approx 2.0 \pm 0.7 \text{ eV}.$$

This appears to be quite a reasonable result: Lichten[3] suggested a value in the neighborhood of 3 eV.

Corresponding to the reduced variable τ, the reduced perturbation cross section is properly taken as

$$\Delta s_{\rm g}(\tau, E) = E^{-1} \sin\theta \Delta\sigma_{\rm g}(\theta, E). \tag{6}$$

From λ_x and the average behavior of the upper envelope cross section $\sigma_+(\tau)$, we can deduce the value $\Delta s_{\rm g}(\max) \approx 2.5\times10^{-3}$ Å^2/eV-deg. It is natural to attempt to measure the area under one of the wings of $\Delta s_{\rm g}(\tau)$, which roughly we may do as follows:

$$\Delta q_{\rm g} = \int_{\tau_x}^{\infty} \Delta s_{\rm g}(\tau) d\tau = \beta \Delta s_{\rm g}(\max) \Delta\tau_x. \tag{7}$$

Thus we find $\Delta q_{\rm g} \approx \beta\times0.8$ Å^2. The quantity $2\pi\Delta q_{\rm g}$ can be considered as a total cross section for a displacement of the elastic scattering from smaller to larger angles near τ_x due to the perturbation associated with the crossing of potentials. As such, we may naturally write it in the form

$$\Delta q_{\rm g} = b_x^{\rm g} \Delta b_x \cong r_x^{\rm g} \Delta r_x \cong 0.06 \text{ Å}^2, \tag{8}$$

from which we can estimate the constant β of (7): $\beta \approx 0.075$. This is a reasonable number, which provides a consistency test for the interpretation.

The underlying interference peaks appear to be perturbed mainly in their amplitude and scarcely at all in their location by the crossing. The peaks are known to be described as an interference between two scattering amplitudes

$$f_j(E, \theta) = \sigma_j^{1/2}(E, \theta) \exp[iA_j(E, \theta)/\hbar], \quad j = (\text{g}, \text{u}), \tag{9}$$

where A_j is the classical action which can be put in the reduced form

$$a(\tau, E) = (E/2\mu)^{1/2} A(E, \theta) = a_0(\tau) + E^{-1} a_1(\tau) + \cdots. \tag{10}$$

Connected with this are the impact parameter,

$$b(\tau,E)=\partial a/\partial\tau \;(\tau \text{ in eV rad}), \tag{11}$$

and the reduced cross section

$$s(\tau,E)=\tfrac{1}{2}\partial b^2/\partial\tau = E^{-1}\sin\theta\sigma(\theta,E). \tag{12}$$

Specializing to the perturbation functions Δa_g, Δb_g, Δs_g, and integrating we find

$$\Delta a_g(\tau) = -\int_\tau^\infty \Delta b_g(\tau')d\tau' = \frac{1}{b_x{}^g}\int_\tau^\infty d\tau'\int_{\tau'}^\infty d\tau''\Delta s_g(\tau'',E), \tag{13}$$

and the maximum value of this function we can write as

$$\Delta a_g(\max) = \beta\gamma\Delta s_g(\max)(\Delta\tau)^2/b_x{}^g. \tag{14}$$

The maximum shift in phase can therefore be evaluated; since the lowest energy at which the perturbation is seen is 50 eV, we have

$$\Delta A_g(\max)/\hbar = (2\mu/E)^{1/2}\Delta a_g(\max)/\hbar \approx 0.22\gamma(50/E)^{1/2} \;(E \text{ in eV}). \tag{15}$$

Since γ is expected to be less than 1, $\Delta A_g/\hbar$ is so small that the peaks cannot be expected to show a very large shift in location. This is entirely consistent with our observations.

It is striking that Everhart's analysis of his data for $He^+ + He$ scattering,[3] particularly his Fig. 6(b), shows a pronounced effect of the perturbation in question. In that figure there is a peak centered at about 2000 eV deg, with a width of a few hundred eV deg, which undoubtedly corresponds to the structure we have been discussing. The height of that peak is a measure of $\Delta A_g/\hbar$ – the value, ≈0.4, is somewhat greater than (15) would suggest, but not wildly out of line in view of the crudeness of the estimates involved here.

The curves for asymmetric scattering (He^+ + Ne, Ar) in Fig. 1 show clearly that the first perturbation structure is followed by other features of the same sort at larger angles; the system $He^+ + He$ appears to show similar behavior. This is to be expected since a succession of excited g potential curves cross the basic diabatic $V_g(r)$ at fairly close intervals inside the initial $r_x{}^g$. Lichten shows these crossings in $He_2{}^+$ lying in the region from 1.1 Å to about 0.6 Å, at which point a doubly ionized state disssociating to $2He^+(1S)$ is encountered. This may correspond to a region in τ roughly 2000 eV deg in width, extending outward from the initial τ_x. This gives enough space for the first one or two crossing structures to be isolated from each other, but beyond that they may merge in a more complicated way; the details must wait until further experimental and theoretical work is done.

We believe this type of feature will turn out to be quite common in differential scattering. It therefore deserves a name, and we recommend that it be called a perturbation in the elastic-scattering pattern. Our reason for recommending this rather pallid term is to emphasize the similarity between the behavior in question and the perturbations of molecular spectroscopy which are also caused by the interaction of two molecular electronic states.

It need hardly be pointed out that these elastic perturbations are intimately connected with specific inelastic-scattering processes. A search of the inelastic differential-scattering spectrum for the companion processes will surely be interesting.

The curve-crossing perturbation represents a coupling of electronic states induced primarily by the radial motion of the nuclei. It should be clearly distinguished from the rotation-inversion coupling that induces a phase-shift of π in the odd (u) scattering amplitude in the limit of high energy and small impact parameter.[8] The latter coupling is responsible for the change in slopes seen in Everhart's Fig. 5.[3] The transition region in that case occurs near $E\theta \approx 10^4$ eV deg or $b_u \approx 0.25$ Å – the nuclei pass by mostly inside the electron cloud – and near the velocity $v = 6\times10^7$ cm/sec; this corresponds to a maximum angular velocity of the nuclear axis $\omega_{max} \approx 0.5\omega_0$, where ω_0 is the angular velocity of an electron in the ground state of H, so that the condition for optimum nuclear-electronic rotational coupling is fulfilled.

---

*Work supported in part by the Defense Atomic Support Agency through the U. S. Army Research Office (Durham), and by the National Aeronautics and Space Administration.

[1]F. P. Ziemba and E. Everhart, Phys. Rev. Letters 2, 299 (1959); G. J. Lockwood, H. F. Helbig, and E. Everhart, Phys. Rev. 132, 2078 (1963).

[2]D. C. Lorents and W. Aberth, Phys. Rev. 139, A1017 (1965).

[3]F. P. Ziemba and A. Russek, Phys. Rev. 115, 922 (1959); W. L. Lichten, Phys. Rev. 131, 229 (1963);

E. Everhart, Phys. Rev. 132, 2083 (1963).
[4]R. P. Marchi and F. T. Smith, Phys. Rev. 139, A1025 (1965).
[5]W. Aberth, D. C. Lorents, R. P. Marchi, and F. T. Smith, Phys. Rev. Letters 14, 776 (1965).
[6]F. T. Smith, Bull. Am. Phys. Soc. 9, 411 (1963).
[7]See Everhart, reference 3; and F. T. Smith, J. Chem. Phys. 42, 2419 (1965).
[8]D. R. Bates and D. A. Williams, Proc. Phys. Soc. (London) 83, 425 (1964); F. J. Smith, Proc. Phys. Soc. (London) 84, 889 (1964); Yu. N. Demkov and Yu. E. Murakhver, in Proceedings of the Fourth International Conference on Physics of Electronic and Atomic Collisions, Quebec, August 1965 (to be published).

POSITRON LIFETIMES IN LOW-TEMPERATURE HELIUM GAS*

L. O. Roellig and T. M. Kelly†

Department of Physics, Wayne State University, Detroit, Michigan
(Received 24 September 1965)

A variety of new phenomena have recently been observed in positron annihilation in dense noble gases.[1-3] One of the most interesting is the observance of a nonexponential decay in the annihilation spectrum. The spectrum is characterized by a shoulder or plateau followed by an exponential decay. The shoulder has been attributed to a velocity-dependent positron annihilation rate for free positrons with energies below the Ore gap. This paper presents the results of a series of positron lifetime measurements in helium gas at 4.2°K. A shoulder was observed in the lifetime spectrum which exhibited an exponential decay, in contrast to the results reported in argon, followed by a hump and then another exponential decay. The shape of the annihilation spectrum reported here is the basis for a re-examination of the decay process of positrons in helium.

The helium gas was contained in a brass bucket ($1\frac{1}{4}$ in. in diameter and $1\frac{1}{2}$ in. in height) which was located in the tail section of a helium Dewar. In order to increase the number of positrons stopping in the gas, the positrons from a Na^{22} source were moderated by a 2-mil brass foil. The moderating foil caused 13% of the positrons emitted by the source to be stopped in the gas. The annihilating γ rays were detected by Pilot B plastic scintillators which were mounted on 56AVP photomultiplier tubes. A Simms-type time-to-amplitude converter with a range of 300 nsec and a resolving time of 3.5 nsec (full width at half-maximum) was used in the lifetime measurements.[4]

Typical delayed coincidence spectra at various helium-gas pressures are shown in Fig. 1. Note that the curves are characterized by a prompt lifetime (τ_1) due mainly to annihilations in the bucket walls, a shoulder of width T, a lifetime on top of the shoulder (τ_2), which is followed by a hump, and then a lifetime τ_3. At pressures less than $\frac{3}{4}$ of an atmosphere the shoulder region was obscured by the background, and hence only a single lifetime was observed. The values of τ_2, τ_3, and T for helium gas at various pressures between one-half an atmo-

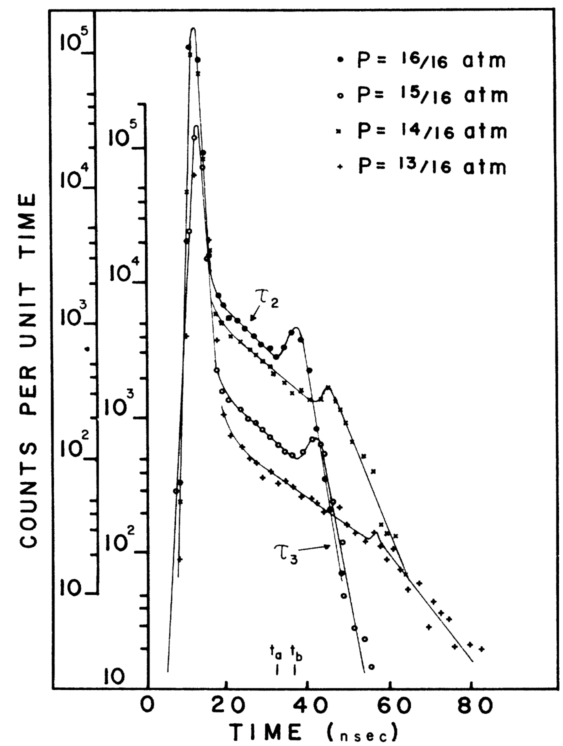

FIG. 1. Positron annihilation spectra in helium gas at 4.2°K. Random background and the long lifetime have been subtracted. The lifetimes τ_2 and τ_3 and the times t_a and t_b are indicated only for the data taken at $P \simeq 16/16$ atm. The ordinate on the left (right) corresponds to data at 16/16 and 14/16 (15/16 and 13/16) atm.