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LOW ENERGY ELECTRON-ATOM AND ELECTRON-MOLECULE SCATTERING THEORY CIRCA 1964

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E. Gerjuoy

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E. Gerjuoy

Let me begin with the remark that, to avoid continual reiteration of an awkward phrase, I shall simply say "electron scattering" where it is obvious that I am referring to electron-atom and/or electron-molecule collisions, which collisions may be elastic or inelastic. Progress in electron scattering theory has been reviewed on numerous occasions in recent years, by various authors.[1] Thus, there is not much point in trying to re-review in detail here the whole subject of electron scattering, especially since I couldn't possibly do it within the pages of this entire issue of Physics Today. Instead, I largely shall confine my attention to topics wherein what has been happening within the past two years or so seems important. By important, I mean that--to me at least--these happenings suggest modifications of apparently established points of view. Even with this very severe restriction on the subjects I intend to discuss, I really can't do justice to the available material in an article of this length. A reasonable idea of the feverish present activity in the field of electron scattering, as well as a notion of the breadth of topics falling under this general subject heading, can be obtained from thumbing through the Proceedings of the Third International Conference on the Physics of Electronic and Atomic Collisions.[1]

This article is concerned only with non-relativistic energies, i.e., with incident electron velocities $v << c$ = velocity of light. At such energies, except possibly for small relativistic and field theoretic effects, the equations governing electron scattering processes are completely known. In other words, for the purposes of this paper one can categorically assert that computing electron scattering cross sections no longer involves any questions of fundamental physical principle. Commonly, the cross section is expressed as the square of a matrix element involving the solution Ψ to Schrodinger's equation, although actually the cross section can be computed from a variety of expressions, all equally correct. The matrix element form, however, makes explicit the fact that exact knowledge of the cross section for any reaction generally cannot be expected unless there is exact knowledge of the solution Ψ itself. Unfortunately, in no actual electron scattering problem, even the simplest (namely, the scattering of electrons by atomic hydrogen), is Schrodinger's equation solvable without approximation. The theoretical uncertainties stem solely from these (as yet) unavoidable approximations.

Usually one writes

$$\Psi = \psi + \Phi \qquad (1)$$

where ψ is the so-called incoming wave, and Φ is the "outgoing" scattered wave. As will be discussed in more detail below, the main uncertainties in the theoretical cross sections arise from lack of knowledge concerning the behavior of Φ. Comparatively, ψ can be regarded as "known" and ordinarily is so regarded. Nevertheless, often ψ too is quite uncertain. In the scattering of electrons by species X, i.e., in e-X scattering, the function ψ always is a product of two factors. One factor represents the known relative motion of e and X for zero e-X interaction. Specifically, this factor represents: constant velocities v_e, v_X when X is neutral; Coulomb scattering when X carries a net charge (in which event Φ denotes the extra scattering ascribable to the fact that X is not simply a point charge). The second factor in ψ is the wave function describing the initial state of X, when e and X are at infinite separation. This wave function is expressible without approximation in useable analytic form only when X denotes a one-electron atomic system, e. g., H or He^+.

With modern computing techniques, however, quite accurate (for the purpose of approximating ψ) numerical representations of the initial state of X can be obtained for the ground states and low-lying excited states of essentially all atomic species, including their positive ions;[2] moreover, in many cases these numerical solutions are very well approximated by surprisingly uncomplicated functions.[3] For negative atomic ions, e.g., O^-, it is more difficult to obtain good wave functions, because electron attachment energies to neutral atoms tend to be much smaller than the ionization energies of neutral atoms or of positive ions. With negative ions, therefore, wave function calculations require greater numerical precision; at the same time they are more complicated, because a weakly bound electronic wave function tends to be more spread out than when the electron is tightly bound.

Computation of molecular wave functions (neutral or ionic) is more difficult still. For diatomic molecules X comprised of light atoms (in the first few rows of the periodic table), it appears possible[4] to obtain reasonably accurate numerical representations of low-lying bound state wave functions by direct solution of the Schrodinger equation describing the electrons and atomic nuclei comprising X; the same assertion may even be true for linear molecules composed of three or four light atoms. With increasing molecular weight and complexity, however, especially for non-linear molecules, accurate computation of wave functions rapidly becomes less feasible. Thus in scattering of electrons by CCl_4, for example, ψ in Eq. (1) is known only very approximately. Note that for various obvious reasons--expense, lack of interest, non-availability of machine time, etc.--it is by no means true that every wave function which can be practicably computed using presently available techniques actually has been computed.[5]

FIRST ORDER APPROXIMATIONS

With these introductory remarks out of the way, I now explain that this paper concentrates on work at low energies, because in my opinion this is the energy range where the important recent theoretical work on electron scattering from atoms and molecules has been concentrated. The term "low energies" isn't very well defined, but in practice it seems to mean less than a few hundred volts, with the main interest at energies less than a few tens of volts. "High energies" in the sense of this paper means anywhere from a few hundred to a few hundred thousand electron volts. At very high energies, > 0.2 Mev, incident electron velocities are sufficiently close to 3×10^{10} cm/sec that relativistic effects can be important. On the subject of these very high energy effects there is intensive current research, of course, because this subject (unlike non-relativistic collisions) does involve numerous as yet unsettled questions of fundamental principle. In scattering experiments, however, these still arcane effects show up mainly in those collisions wherein the incident electron comes very close to (within $\sim 10^{-13}$ cm) a single one of the electrons or atomic nuclei comprising the target atom or molecule. Thus, the detailed many-electron aspects of the target's initial state generally are inconsequential to scattering experiments probing these very high energy effects. Correspondingly, investigations of such effects usually are not regarded as belonging to the field of electron scattering by atoms and molecules.

Returning to the assertion with which I began the preceding paragraph, this present concentration of electron scattering theory on low energies stems from a twofold stimulus. In the first place, the major areas where researchers presently are making quantitative applications of detailed cross section information--for instance, laboratory discharges, the ionosphere, stellar atmospheres, or nuclear blast phenomena--typically involve energies less than a few hundred electron-volts. In fact, for practical applications, one rarely needs to know cross sections above a few tens of volts, and usually one needs accurate cross sections only at energies up to a volt, corresponding to a temperature of 12000°K.

The second stimulus for concentrating on low energies is purely theoretical. For some time a major objective of the theory has been to make better estimates of electron collision cross sections than can be obtained from Born approximation and related first order approximations, which by now are pretty much old hat. By "first order", or by "first Born approximation", or simply by "Born approximation", one usually means the approximation which--in matrix elements expressing the cross section--replaces the exact solution Ψ by its incoming part ψ. The term "related" above indicates that I am including under the heading "first order" those approximations wherein the total solution Ψ and its incoming part ψ are symmetrized more or less in accordance with the requirements of the exclusion principle, i.e., in accordance with the postulated indistinguishability of the incident and target electrons. In electron scattering, the usual rough criteria for the validity of these first order approximations typically fail when the energy decreases below just about a few hundred volts. These rough criteria are, as is well known, that the incident bombarding electron velocity should be large compared to the velocities of the bound electrons in the target system, or (very roughly equivalently) that the incident kinetic energy should be large compared to the interaction energy.

The claims of the previous paragraph, though basically correct, are too broad not to have exceptions and counterexamples. Let me mention a few. Although I said Born approximation is not expected to be good at low energies, and by now is pretty much old hat anyway, nevertheless new papers containing first order calculations of hitherto uncomputed electron collision cross sections continue to appear, with the results typically plotted all the way down to threshold energies. The reason for these papers is quite obvious. As the bee seeks the honey, so do theoretical physicists seek cross sections not yet computed in Born approximation, because Born approximation usually is comparatively easy to calculate, whereas anything better is usually much harder. Of course, for collisions with atoms the reactions studied in these present papers tend to involve highly excited initial states of the target system, or to have some other unusual feature, because all the reactions one would normally think of first--and second and third--by now already have been done.

For collisions inducing electronically excited molecular states, even the Born matrix elements are difficult to evaluate accurately, so that Born estimates of electron-molecule collisions are not yet everywhere dense in the past literature. Correspondingly, a careful Born approximation calculation of electronic excitation in the simplest molecular case, namely electrons incident on H_2^+, has only recently been given.[6] It turns out that using the exactly known H_2^+ wave functions for fixed internuclear separation, and then averaging over the vibrational wave functions, involves a fair amount of numerical integration.

My comment that first order calculations by now are pretty much old hat also must be evaluated in the light of a recent paper by Ochkur.[7] For many years it has been remarked that the so-called Born-Oppenheimer approximation, in which the exchange amplitude is estimated in first order, generally gave worse results for low energy electron scattering than if one simply ignored exchange. (This Born-Oppenheimer approximation is not to be confused with the Born-Oppenheimer approximation for calculating molecular wave functions.) It always has seemed peculiar that taking into account electron indistinguishability should yield worse results than ignoring indistinguishability, although of course one could argue that such results simply indicated how bad first order approximations really were at low energies. Ochkur's idea is as follows. All one really is entitled to claim about the first order approximation is that it is valid at high energies. Now there are several different terms in the Born-Oppenheimer exchange integral, and these have different energy dependences. In fact, some of these terms vanish so much more rapidly with increasing energy than does the leading term, that it seems likely such terms could be cancelled or anyway modified by a higher order calculation. Thus Ochkur suggests that at low energies one should calculate the exchange including only those terms which are dominant at high energies. Fig. 1 shows the results of his first order calculations for excitation of the 2^3S_1 level in Helium. In this case one has to use the Born-Oppenheimer approximation, because in first order a singlet to triplet transition can only go by electron exchange; in other words, if exchange were neglected the first order cross section would be zero. Curve 1 is Ochkur; curve 2 is experiment; curve 3 is the Born-Oppenheimer result reduced by a factor of only 20.

HIGHER APPROXIMATIONS

Of course, usually the first order calculations below a few hundred volts aren't as good as those shown in Fig. 1. Let us grant, therefore, that we do require better than first order estimates. The question is, how do we get them? The most obvious means is via the so-called Born series, wherein the exact scattering amplitude is expanded in powers of the interaction. This series can be regarded as a sum over all possible exchanges of momentum and energy between the target system and the incident or outgoing electrons. The number of such exchanges for any term in the series equals the power of the interaction in that term. The first order approximations I have been discussing retain only those terms in the Born series which are linear in the interaction. Including the terms which are quadratic in the interaction yields the so-called second Born and related approximations and similarly for higher powers. As always, I use the word "related"to mean that exclusion principle requirements are more or less being taken into account.

Now even in the case of electron-hydrogen scattering it is not practical to take into account all the second order terms in the Born series. The simplest second order approximation is to ignore all second order terms corresponding to excitation of the atom during the collision; in other words during the collision the atom is supposed to remain always in its ground state. Then, for e-H elastic scattering, the incident and outgoing electron can be thought to move in the effective field of atomic hydrogen in its ground state, that is to say in the Coulomb fields of the proton and bound electron, averaged over the ground 1s wave function. If electron exchange can be ignored, therefore, the problem has been reduced in essence to potential scattering; this second order approximation is taking into account two successive elastic scatterings of the electron by the target's effective field.

In electron scattering theory, "second Born approximation" usually designates the effectively potential scattering approximation I've just described, which also is pretty much old hat by now. Naturally, such calculations still appear in the literature--nothing ever seems to disappear from the literature--but usually second Born approximation is computed only for comparison with other approximations, not because it is taken seriously. One good reason for not taking such second Born calculations seriously at low energies is that once we've decided a single scattering will not describe the collision, why should we be content with no more than two scatterings? A rough criterion for what is meant by a scattering is a momentum transfer comparable with the initial momentum. As its energy decreases the incident electron spends more time in the vicinity of the target, and suffers correspondingly more scatterings. In fact, for Coulomb type forces it is trivial to see that the criterion "momentum transfer in the vicinity of the target must be small compared to the incident momentum" is equivalent to "interaction energy must be small compared to the incident energy."

This last argument suggests that at low energies one must sum over all possible numbers of scatterings. In other words, returning once more to the elastic scattering of electrons by atomic hydrogen, and granting that the interaction can be represented by the ground state atomic hydrogen effective field, at low energies the scattering in this potential has to be computed exactly. Note that electron exchange still is being neglected.

Calculations of the type I've just described often are termed the distorted wave approximation. Even if electron exchange really can be neglected however, it is clear that these calculations still are not very sensible at low energies, because on the one hand we have been assuming excitation of the target does not occur during the collision, while on the other hand we have recognized that there can be large energy transfers between the incident and target electrons. The next improvement, therefore, is to try to take account of this excitation, which modifies the effective field in which the incident electron moves. At long range this excitation results in the well-known polarization potential, which for a spherically symmetric ground state of an electrically neutral target is itself spherically symmetrical and proportional to the inverse fourth power of the distance. For non-spherically symmetric scatterers, the polarization potential is more complicated in form. The functional form of the polarization potential at long range always is well known, however, and often the magnitude of the polarizability has been directly measured. But there is the difficulty that the asymptotic behavior of the potential at long range does not represent the correct interaction close to the target. In fact, generally the asymptotic forms must be somewhat arbitrarily cut off to avoid divergences at the origin.

Distorted wave calculations using polarization potentials are quite popular these days, in electron-molecule as well as electron-atom collisions. For electron-atom collisions, it is becoming customary to employ polarization potentials which have a theoretical basis at all distances, not merely at long ranges, thereby avoiding the necessity for arbitrary cutoffs. A favorite and reasonable way of estimating such a potential for low energy calculations is the so-called adiabatic method. In this method, one assumes in effect that--for each position of the incident electron--the bound electron eigenfunctions can be computed as if the incident electron were at rest. Thus, the electron-atom potential obtained by the adiabatic method is analagous to the commonly employed atom-atom potentials, which at each internuclear separation are computed as if the nuclei were at rest. In this approximation one can take into account electron exchange between the incident and bound electrons, as especially Temkin[9] has shown, although of course including exchange complicates the calculations.

Approximations of this type have been employed, for example, to compute the elastic cross section for electron-Cs scattering, which has excited much theoretical interest in recent years. It is noteworthy that although there have been at least four different calculations of the effective electron-Cs potential since 1961, none of them carry out the complete adiabatic procedure I have described, because even with the vast simplification afforded by the adiabatic approximation, the problem still is very complicated.

For electron scattering, the adiabatic method is only justifiable at essentially zero incident energy, if it is justifiable at all. If the energy must be considered finite, and if electron exchange cannot be neglected at low energies--as it usually cannot--the calculations become considerably more complicated than those I have already described. Trying to do such calculations properly--and including more intermediate states than merely the ground state--inevitably leads to the horribly coupled integro-differential equations of the so-called close coupling approximation. In the close coupling approximation, the total wave function is projected essentially exactly onto some chosen set of eigenfunctions of the target system, for instance onto the 1s, 2s, and 2p atomic hydrogen functions in the case of electron-hydrogen scattering.

With the inclusion of enough intermediate states, the close-coupling approximation begins to resemble an exact rather than an approximate treatment, so that in this approximation it is perhaps finally reasonable to anticipate reliable theoretical cross sections. Of course, this relatively straightforward but very arduous close coupling approximation is not the only possible approach to good low energy calculations. For scattering by hydrogen, the most successful alternative approaches have been variational calculations--especially along lines developed by Spruch and collaborators[10]--and an ingenious alternative expansion introduced by Temkin,[11] which he calls the nonadiabatic method. However, these alternative approaches also involve arduous calculations, and indeed it should be obvious from what I've been saying that one cannot expect to get reliable theoretical cross sections at low energies without a lot of work. Unfortunately, actual electron collisions, involving many successive interactions with the target, cannot be solved in closed form, as we can solve say low energy scattering in a Coulomb field. Once it is necessary to make numerical computations starting from some reasonably mathematically tractable function of the particle coordinates, the fact that many scatterings occur implies the actual wave functions are almost certain to be very much more complicated than our starting functions, which in turn almost surely implies arduous calculations are needed to get to the exact solution.

This brings me to the subject of resonances. Resonances are a comparatively novel addition to the vocabulary of electron scattering. Although in nuclear reactions the existence of resonances has been well established since the middle 30's, the suggestion that resonances could play an important role in electron collisions seems to have been advanced seriously no earlier than 1957.[12] Since then a variety of experiments by many experimenters[13] have demonstrated the existence of resonances in numerous electron-atom and electron-molecule collisions, as well as in photoabsorption processes. On the theoretical side, the main contribution has been by Fano, who has shown quantitatively how resonances are connected with the existence and properties of autoionizing states, and who has gone on to interpret much of the experimental data on this basis.

Of especial significance to the main theme of this talk is the fact that Schulz[14] recently has reported finding a resonance in elastic electron-hydrogen scattering at an energy about half a volt below the 2s excitation threshold. This energy is very close to the energy of a resonance originally predicted theoretically,[15] on the basis of close coupling calculations. Calculations since then by a number of theorists, using the variational and non-adiabatic approaches, have confirmed and refined Burke and Schey's prediction. There still remain questions concerning the precise behavior of the phase shifts in the immediate vicinity of this resonance and of the 2s excitation threshold. However, on the whole the agreement between the different calculational approaches is so good that--in view of Schulz's recent finding--the following important conclusion seems justified. At this time, the close of 1964, the cross section for elastic scattering of electrons by atomic hydrogen--if not already being calculated essentially exactly--will be so calculated in the near future. The same conclusion holds for elastic scattering of electrons by He^+. Recent work also suggests that with these one-electron targets we soon will be computing inelastic excitation--at least to moderately low-lying states--no less accurately than elastic scattering. However, largely because of complications induced by the Coulomb long range force when two electrons can go to infinity in the presence of a positively charged nucleus, accurate theoretical predictions of H and He^+ ionization cross sections seem further away.

CONCLUDING REMARKS

This concludes my sampling of important recent progress in electron scattering theory. There is more to be said, however. It is essential to keep in mind that calculating electron-hydrogen scattering is one thing, but calculating electron-Cs scattering or electron scattering by molecular oxygen, is quite another thing. Many novel techniques for calculating cross sections have been proposed, and will be proposed, and many of these are indeed useful. But for the reasonably foreseeable future I just don't believe such techniques will make it possible to accurately calculate low energy cross sections for any moderately complicated target. I'm not quite sure how complicated my moderately complicated targets have to be, but I'll bet that atomic oxygen--let alone molecular oxygen--will be complicated enough. Please understand I am quite willing to believe we can get within a factor of three over most of the low-energy range. This we will do by playing around with various plausible but not wholly well-founded approximations--like Ochkur's, or like Gryzinski's classical (meaning non-quantum) calculations.[16] But I don't think such procedures will assuredly get within 50 per cent, nor will they reliably predict details of the cross section.

These last assertions--if really correct, not just an expression of my well-known pessimism--imply that if we want reliably accurate theoretical calculations for say electron-oxygen collisions, then for a good many years we're going to have to resign ourselves to introducing some arbitrary parameters which can be fitted to part of the measured data. With this approach, the remaining data, as well as needed unmeasurable cross sections, might then become understandable and predictable. How to carry out such a program I certainly don't know right now. But I do want to point out that this is precisely the kind of program which recently has been notably successful in predicting nuclear inelastic cross sections via optical model calculations. In nuclear physics, where the fundamental interparticle forces are not known, making use of optical potentials fitted to elastic scattering data obviously is a very reasonable thing to do. In electron scattering, however, where the fundamental forces are exactly known, introducing arbitrary parameters which must be fitted to the data seems very repugnant. I have this feeling myself: that such a procedure is a capitulation of the theory. But like it or not, knowing the fundamental forces are Coulomb does not mean we know the effective force between an electron and an atom at low energies, where during the collision the electron is undergoing many individual elastic and inelastic scatterings with the atom, and in addition is making electron exchanges with the target.

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FIGURE CAPTIONS

Fig. 1: Electron Excitation of He 2^3S_1.

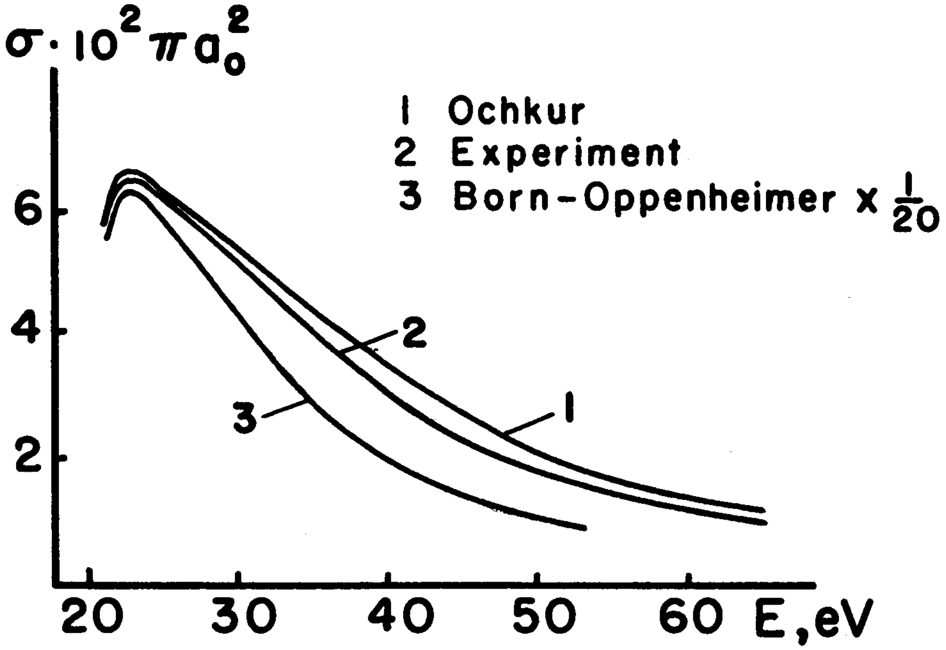
$\sigma \cdot 10^2 \pi a_0^2$
1 Ochkur
2 Experiment
3 Born-Oppenheimer x $\frac{1}{20}$
6
4
2
20
30
40
50
60
E, eV
1
2
3

Figure 1