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Inelastic Scattering of High-Energy Electrons from Atoms: The Helium Atom

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A discussion of the inelastic scattering of high-energy electrons from atomic systems using linear-response theory is presented. Following Van Hove, an expression for the differential scattering cross section within the Born approximation is derived. The relationship of this expression to the linear-response function is demonstrated and a practical procedure for its computation using the so-called random-phase approximation is given. Finally, explicit results for the excitation of the lowest singlet S and singlet P state of helium by 500-eV electrons are given.

I. INTRODUCTION

The inelastic scattering of high-energy electrons from atoms has been the subject of both experimental and theoretical interest for many years.[1] The reason for this lies in the simple fact that the differential cross section or the generalized oscillator strength gives us detailed information about the response of atomic systems to the sudden transfer of momentum from an external source.[2-4] In addition, electron impact excitation studies are useful for the discovery and elucidation of states forbidden by photon impact. Traditionally, the evaluation of the generalized oscillator strength has depended upon the knowledge of wave functions for both the ground and excited states of the system under consideration. These wave functions are usually known only crudely, and the many calculations found in the literature suggest a great sensitivity to the approximations involved.[5] Our philosophy on the calculation of these generalized oscillator strengths is unconventional in that we try to avoid the use of wave functions as much as possible. By concentrating efforts on the *direct* calculation of the matrix element needed for the oscillator strength, one can avoid the need for computing both ground- and excited-state wave functions separately and then having to integrate their product with the appropriate transition operator. This process is intended to avoid the unnecessary details associated with the computation of accurate bound-state wave functions while retaining the proper physical effects needed for quantitative accuracy of the final result. In this way the actual computational process may be considerably shortened. Below we consider just such a procedure based on the theory of the linear response. To lowest order, the theory leads to the well-known random-phase approximation (time-dependent Hartree-Fock).[6] We present calculations on the high-energy scattering of electrons from helium as an illustration of the practicality and accuracy of the procedure.

II. THEORY

The Hamiltonian for the problem of an electron incident on an atom may be written as

$$\mathcal{H}|\psi\rangle = (\mathcal{H}_A + T_e + V_{Ae})|\psi\rangle = E|\psi\rangle \quad , \tag{1}$$

where

$$\mathcal{H}_A = \frac{1}{2}\sum_{i=1}^{N} P_i^2 - \sum_{i=1}^{N}\frac{z}{r_i} + \sum_{i<j}\frac{1}{r_{ij}} \quad , \tag{2a}$$

$$T_e = \tfrac{1}{2}P_e^2 \quad , \tag{2b}$$

$$V_{Ae} = \frac{-Z}{r_e} + \sum_{i=1}^{N}\frac{1}{r_{ie}} \quad . \tag{2c}$$

Let us define the density of electrons in our atomic target as

$$\rho(\vec{r}) = \sum_{i=1}^{N}\delta(\vec{r}-\vec{r}_i) \quad . \tag{3}$$

Introducing the field operators[7] $\psi_\sigma^\dagger(\vec{r})$, $\psi_\sigma(\vec{r})$, where the subscript σ indicates a spin coordinate, we may write

$$\rho(\vec{r}) = \textstyle\sum_\sigma \psi_\sigma^\dagger(\vec{r})\psi_\sigma(\vec{r}) \tag{4}$$

and thus

$$V_{Ae} = \frac{-Z}{r_e} + \sum_{i=1}^{N}\int d\vec{x}\,\frac{\delta(\vec{x}-\vec{x}_i)}{r_{xe}}$$

$$= \frac{-Z}{r_e} + \int d\vec{x}\,\frac{\rho(\vec{x})}{r_{xe}} \quad . \tag{5}$$

In what follows we use the notation

E_A^0 = ground-state energy of atom,

E_A^n = nth-excited-state energy of atom,

$\frac{1}{2}k_0^2$ = incident electron energy,

$\frac{1}{2}k_n^2$ = final electron energy,

$q = k_n - k_0$ = momentum transfer,

$q^2 = k_n^2 + k_0^2 - 2k_0 k_0 \cos\theta$,

$\omega_n = E_A^n - E_A^0$.

The differential cross section for the excitation of the nth excited state of our system may be written in the Born approximation[1] as

$$\frac{d\sigma_{n0}}{d\Omega}=\frac{k_n}{4\pi^2 k_0}\left|\int d\vec{r}_A\,d\vec{r}_e\,e^{i\vec{q}\cdot\vec{r}_e}\left(\frac{-Z}{r_e}+\sum_{i=1}^{N}\frac{1}{r_{ie}}\right)\times\psi_n^*(\vec{r}_A)\psi_0(\vec{r}_A)\right|^2 , \tag{6}$$

where r_A stands for the collection of atomic coordinates. By virtue of the orthogonality of atomic states, we may drop the first term in the parentheses. Using (5) we have

$$\frac{d\sigma_{n0}}{d\Omega}=\frac{k_n}{4\pi^2 k_0}\left|\int d\vec{r}_e\,d\vec{x}\,e^{i(\vec{k}_n-\vec{k}_0)\cdot\vec{r}_e}\,\frac{\rho_{n0}(\vec{x})}{r_{xe}}\right|^2 , \tag{7}$$

$$\frac{d\sigma_{n0}}{d\Omega}=\frac{k_n}{4\pi^2 k_0}\left|\int d\vec{r}_e\,d\vec{x}\,\frac{e^{i\vec{q}\cdot\vec{r}_e}}{r_{xe}}\,\rho_{n0}(\vec{x})\right|^2 , \tag{8}$$

where

$$\rho_{n0}(\vec{x})=\int d\vec{r}_1\cdots d\vec{r}_N\,\psi_n^*(\vec{r}_1\cdots\vec{r}_N)\times\sum_{i=1}^{N}\delta(\vec{x}-\vec{r}_i)\psi_0(\vec{r}_1\cdots\vec{r}_N) .$$

By using

$$\int d\vec{r}_e\,\frac{e^{i\vec{q}\cdot\vec{r}_e}}{r_{xe}}=\frac{4\pi}{q^2}\,e^{i\vec{q}\cdot\vec{x}} , \tag{9}$$

we get

$$\frac{d\sigma_{n0}}{d\Omega}=\frac{4k_n}{k_0}\left|\int d\vec{x}\,e^{i\vec{q}\cdot\vec{x}}\,\rho_{n0}(\vec{x})\right|^2\frac{1}{q^4} . \tag{10}$$

In terms of the so-called generalized oscillator strength, defined as

$$f_n(q)=(2\omega_n/q^2)\left|\int d\vec{x}\,e^{i\vec{q}\cdot\vec{x}}\rho_{n0}(\vec{x})\right|^2 , \tag{11}$$

we can write

$$\frac{d\sigma_{n0}}{d\Omega}=\frac{2k_n f_n(q)}{q^2 k_0\omega_n} . \tag{12}$$

So far, we have made no reference to linear-response theory. The connection can be achieved by considering the response of an atomic system to an arbitrary time-dependent external potential (such as a fast-moving electron). As many authors[3,4,8] have shown, this response is completely determined once the quantity

$$R(\vec{r}t,\vec{r}\,'t')=i^{-1}\langle 0|\,T(\bar{\rho}(\vec{r},t)\bar{\rho}(\vec{r}\,',t'))\,|0\rangle \tag{13}$$

is known. In this expression, the average of the time-ordered product is over the ground-state wave function, and we define

$$\bar{\rho}(\vec{r},t)=\rho(\vec{r},t)-\langle 0|\rho(\vec{r},t)|0\rangle . \tag{14}$$

We now consider this quantity in detail. Explicitly, we have

$$\begin{aligned}R(\vec{r}t,\vec{r}\,'t')&=i^{-1}[\theta(t-t')\langle 0|\bar{\rho}(\vec{r},t)\bar{\rho}(\vec{r}\,',t')|0\rangle\\&\quad+\theta(t'-t)\langle 0|\bar{\rho}(\vec{r}\,'t')\bar{\rho}(\vec{r},t)|0\rangle]\\&=i^{-1}\Big[\sum_{n\neq 0}\theta(t-t')e^{-i\omega_n(t-t')}\langle 0|\rho(\vec{r})|n\rangle\langle n|\rho(\vec{r}\,')|0\rangle\\&\quad+\theta(t'-t)e^{i\omega_n(t-t')}\langle 0|\rho(\vec{r}\,')|n\rangle\langle n|\rho(\vec{r})|0\rangle\Big] .\end{aligned} \tag{15}$$

Fourier transformation yields

$$\begin{aligned}R(\vec{r},\vec{r}\,',z)&=\int d(t-t')\,e^{iz(t-t')}R(\vec{r}t,\vec{r}\,'t')\\&=\sum_{n\neq 0}\left(\frac{\langle 0|\rho(\vec{r})|n\rangle\langle n|\rho(\vec{r}\,')|0\rangle}{z-\omega_n+i\epsilon}-\frac{\langle 0|\rho(\vec{r}\,')|n\rangle\langle n|\rho(\vec{r})|0\rangle}{z+\omega_n-i\epsilon}\right) .\end{aligned} \tag{16}$$

Our reasons for considering the time-ordered rather than casual response will become evident when an equation to determine R is derived. The important fact to notice at this point is that the spectral representation of the response, Eq. (16), contains the necessary information to compute the generalized oscillator strength. That this is indeed the case follows by taking the space Fourier transform of the residue of (16) at the pole $(\omega_n-i\epsilon)$. Explicitly, we have

$$\mathrm{Res}\,\omega_n=i\langle n|\rho(\vec{r})|0\rangle\,\langle 0|\rho(\vec{r})|n\rangle \tag{17}$$

and its Fourier transform

$$\mathrm{F.T.}=i\int d\vec{r}\,d\vec{r}\,'\,e^{i\vec{q}\cdot(\vec{r}-\vec{r}\,')}\langle n|\rho(\vec{r}\,')|0\rangle\langle 0|\rho(\vec{r})|n\rangle . \tag{18}$$

Equation (18) is the generalized oscillator strength to within a known constant. In order to derive an equation for the response function which is both general and still capable of workable approximation, we make use of the functional derivative techniques of Martin and Schwinger.[9] The fundamental equation needed is

$$-i\,\frac{\delta G(1,1'^{+})}{\delta u(2)}=R(12,1'^{+}2^{+})=i^{-1}\langle 0|\,T\{[\psi^{\dagger}(1'^{+})\psi(1)]\,\bar{\rho}(2)\}\,|0\rangle . \tag{19}$$

The notation[8] used is

n = space, time, spin coordinate,

$G(n,n')$ = exact one-particle Green's function,

$u(n)$ = external time-dependent potential,

$[\psi^{\dagger}(n)\psi(n')]=\psi^{\dagger}(n)\psi(n')-\langle 0|\psi^{\dagger}(n)\psi(n')|0\rangle$.

A superscript plus on a coordinate indicates a time t' infinitesimally later than t. The one-particle Green's function obeys an equation of motion which can be written as

$$G^{-1}(1,1')=G_0^{-1}(1,1')-u(1)\delta(1-1')-\Sigma(1,1') , \tag{20}$$

where $\Sigma(1,1')$[8,10] is the exact self-energy operator. Differentiating the identity

$$\delta(1-1')=\int dx\,G(1,x)G^{-1}(x,1') \tag{21}$$

and Eq. (20) yields

$$\frac{\delta G(1,1'^{+})}{\delta u(2)} = -\int dx\,dx'\,G(1,x)\,\frac{\delta G^{-1}(x,x')}{\delta u(2)}\,G(x',1'^{+})$$
$$= G(1,2)G(2,1'^{+}) + \int dx\,dx'\,G(1,x)$$
$$\times \frac{\partial \Sigma(x,x')}{\delta u(2)}\,G(x',1'^{+}) \quad . \tag{22}$$

If we define

$$\frac{\delta\Sigma(1,2)}{\delta G(3,4)} = \Delta(12,34) \quad , \tag{23}$$

we get

$$\frac{\delta G(1,1'^{+})}{\delta u(2)} = iR(12,1'^{+}2^{+}) = G(1,2)G(2,1'^{+})$$
$$+ \int dx\,dx'\,dy\,dy'\,G(1,x)\Delta(xx',yy'^{+})$$
$$\times\; G(x',1'^{+})\,\frac{\delta G(y,y'^{+})}{\delta u(2)} \tag{24}$$

and thus

$$R(12,1'^{+}2^{+}) = -\,iG(1,2)G(2,1'^{+}) + \int dx\,dx'\,dy\,dy'$$
$$\times G(1,x)\Delta(xx',yy'^{+})G(x',1'^{+})R(y2,y'^{+}2^{+}) \;. \tag{25}$$

An exact solution of (25) is, of course, impossible, but as we now show, the lowest-order approximation leads to a well-known soluble problem, the random-phase approximation.[11] To show this, replace all single-particle Green's functions in (25) by their Hartree-Fock counterparts and let

$$\Delta(xx',yy'^{+}) \cong \frac{\delta\Sigma_{\mathrm{H.F.}}(x,x')}{\delta G_{\mathrm{H.F.}}(y,y'^{+})} = -\,i\delta(x-x')\delta(y-y')$$
$$\times V(x-y) + i\delta(x-y)\delta(x'-y')V(x-x') \quad . \tag{26}$$

Substituting into (25) yields

$$R(12,1'^{+}2^{+}) = iG(1,2)G(2,1'^{+}) - i\int dx\,dy$$
$$\times G(1,x)V(x-y)G(x,1'^{+})R(y2,y^{+}2^{+}) + i\int dx\,dy$$
$$\times G(1,x)V(x-y)G(y,1'^{+})R(x2,y^{+}2^{+}) \quad . \tag{27}$$

Setting $r=r'$, gives the necessary equation to determine $R(1,2)$. The connection between (27) and the ordinary form of the random-phase approximation (RPA) is described in detail in Ref. 10. To make the connection between the RPA and the generalized oscillator strength, it is sufficient to note that the RPA eigenvectors are actually components of $\langle n|\rho(r)|0\rangle$. In fact, a cursory examination reveals that within the RPA[11]

$$\langle n|\rho(\vec{r})|0\rangle = \sum_{k_{\mathrm{unoc}},k'_{\mathrm{occ}}} \gamma_k(\vec{r})\gamma^*_{k'}(\vec{r})X^{*n}_{kk'} + \gamma_{k'}(\vec{r})\gamma^*_k(\vec{r})Y^{*n}_{kk'} \quad , \tag{28}$$

where $X^n_{kk'}$ and $Y^n_{kk'}$ are the two components of the nth RPA eigenvector. If we consider a closed shell and specialize to singlets,

$$\langle n|\rho(\vec{r})|0\rangle = \sqrt{2}\sum_{k_{\mathrm{unoc}},k'_{\mathrm{occ}}}^{\mathrm{space}} \gamma_k(\vec{r})\gamma^*_{k'}(\vec{r})X^{*n}_{kk'} + \gamma_{k'}(\vec{r})\gamma^*_k(\vec{r})Y^{*n}_{kk'} \,. \tag{29}$$

Fourier transformation gives the needed result to compute the generalized oscillator strength. In Sec. III, we present an application to electron–helium-atom collisions.

III. NUMERICAL RESULTS

The scattering of high-energy electrons from helium is one of the most thoroughly studied problems in all of chemical physics,[12–16] and hence, an obvious choice for a test calculation. The singlet RPA equations[11] for helium may be obtained by substituting the spectral representation for the response function, Eq. (15) into Eq. (27), performing a Fourier transform, and taking matrix elements in the Hartree-Fock basis. This procedure has been performed in detail in Ref. 10 and specializes for the two-electron closed-shell system to

$$(\epsilon_{1S}-\epsilon_\alpha-\omega_n)X^n_{1S,\alpha} = \sum_{\beta_{\mathrm{unoc}}}^{\mathrm{space}} (2\langle\gamma_{1S}\gamma_\beta|\gamma_\alpha\gamma_{1S}\rangle$$
$$-\langle\gamma_{1S}\gamma_\beta|\gamma_{1S}\gamma_\alpha\rangle)X^n_{1S,\beta} + \langle\gamma_{1S}\gamma_{1S}|\gamma_\alpha\gamma_\beta\rangle Y^n_{1S,\beta} \quad ,$$
$$(\epsilon_\alpha-\epsilon_{1S}-\omega_n)Y^n_{1S,\alpha} = -\sum_{\beta_{\mathrm{unoc}}}^{\mathrm{space}} \langle\gamma_\alpha\gamma_\beta|\gamma_{1S}\gamma_{1S}\rangle X^n_{1S,\beta}$$
$$+ (2\langle\gamma_\alpha\gamma_{1S}|\gamma_{1S}\gamma_\beta\rangle - \langle\gamma_\alpha\gamma_{1S}|\gamma_\beta\gamma_{1S}\rangle)Y^n_{1S,\beta} \quad . \tag{30}$$

It proves to be convenient to rewrite these in configuration space. This is easily done once it it recognized that Eqs. (30) are merely matrix represen-

TABLE I. Generalized oscillator strength for the 2^1P excitation in He.

q^2 (a. u.)	Oscillator strength (This work)	Oscillator strength (Kim and Inokuti, Ref. 16)
10^{-6}	2.5797×10^{-2}	2.761×10^{-2}
0.04	2.4158×10^{-1}	
0.1225	2.1151×10^{-1}	
0.25	1.7328×10^{-1}	1.8596×10^{-1}
0.49	1.2124×10^{-1}	1.2852×10^{-1}
0.64	9.8057×10^{-2}	1.0408×10^{-1}
0.81	7.7805×10^{-2}	
1	6.0721×10^{-2}	6.5431×10^{-2}
1.69	2.6750×10^{-2}	
2.25	1.4843×10^{-2}	
2.56	1.0976×10^{-2}	
3.24	5.9562×10^{-3}	
4	3.2258×10^{-3}	3.4485×10^{-3}

TABLE II. Generalized oscillator strength for the 2^1S transition in He.

q^2	Oscillator strength (This work)	Oscillator strength (Kim and Inokuti Ref. 16)
10^{-6}	~0	~0
0.04	3.3588×10^{-3}	
0.1225	9.1183×10^{-3}	
0.25	1.5540×10^{-2}	1.4683×10^{-2}
0.49	2.2032×10^{-2}	2.0907×10^{-2}
0.64	2.3722×10^{-2}	2.2396×10^{-2}
0.81	2.4314×10^{-2}	2.2866×10^{-2}
1	2.3936×10^{-2}	2.2493×10^{-2}
1.69	1.9105×10^{-2}	
2.25	1.4835×10^{-2}	
2.56	1.2806×10^{-2}	
3.24	9.2686×10^{-3}	
4	6.5311×10^{-3}	5.9528×10^{-3}

TABLE IV. Comparison of the slope of the generalized oscillator strength versus q^2 for 2^1S transition in He.

Author	$\left.\frac{df\ 2^1s}{dq^2}\right\vert_{q=0}$	Comments
Altshuler (Ref. 12)	0.0366	Length form used for matrix element with hydrogenic wave functions.
	0.0557	Velocity form used for matrix element with hydrogenic wave functions.
Fox (Ref. 15)	0.072	Hartree ground-state wave function.
	0.11	Eckart ground-state wave function.
Garstang (Ref. 21)	0.13	Hartree-Fock.
Van den Bos (Ref. 22)	0.0611	Reference 22.
Kim and Inokuti (Ref. 16)	0.0836	Weiss C. I. wave functions.
This work	0.0837	Random-phase approximation.

tatives of the following pair of coupled differential equations:

$$[-\tfrac{1}{2}\nabla^2-(2/r)+\langle\gamma_{1S}|g|\gamma_{1S}\rangle-\epsilon_{1S}+\omega_n]X^n_{1S}(\vec{r})+\gamma_{1S}(\vec{r})\times(\langle\gamma_{1S}|g|X^n_{1S}\rangle+\langle\gamma_{1S}|g|Y^n_{1S}\rangle)=0\,,$$

$$[-\tfrac{1}{2}\nabla^2-(2/r)+\langle\gamma_{1S}|g|\gamma_{1S}\rangle-\epsilon_{1S}-\omega_n]Y^n_{1S}(\vec{r})+\gamma_{1S}(\vec{r})\times(\langle\gamma_{1S}|g|X^n_{1S}\rangle+\langle\gamma_{1S}|g|Y^n_{1S}\rangle)=0\,, \tag{31}$$

subject to orthogonality of X^n_{1S} and Y^n_{1S} to the $1S$ orbital. Expanding X^n_{1S} and Y^n_{1S} in the set of unoccupied Hartree-Fock orbitals and taking matrix elements immediately gives Eqs. (30). A comparison of Eqs. (31) and the time-dependent (frequency-dependent) coupled Hartree-Fock equations for helium immediately shows the two to be identical. This is not surprising since many authors have shown the mathematical equivalence of the two theories.[6] The solution to this coupled set of equations may be carried out numerically or by variational approximation. The latter procedure was chosen for the following reason: We were interested in just the lowest solutions of S and P symmetry, for which small, simple variational basis sets are easily chosen. The set we used was formed by taking the exponentials appearing in the unperturbed orbital, multiplying by various powers of r, and orthonormalizing. The resulting radial functions were multiplied by the proper spherical harmonic and used in a Rayleigh-Ritz variational procedure. In the case of an S state, the basis set was orthogonalized to the unperturbed orbital. Such sets have been used with great success by a number of authors for polarizability calculations.[17–20] The resulting non-Hermitian eigenvalue problem was solved by a program kindly provided to us by Professor Vincent McKoy of the California Institute of Technology. The integrals needed to compute the matrix elements may be performed analytically as may the Fourier transform of the density operator. The entire calculation was performed on the IBM 360-50 at the Bayside Research Center of GT&E Labora-

TABLE III. Total cross section for 2^1P and 2^1S transitions in He (a.u.).

Incident energy (eV)	Transition	Total cross section
500	2^1S	0.1545×10^{-1}
500	2^1P	0.1714

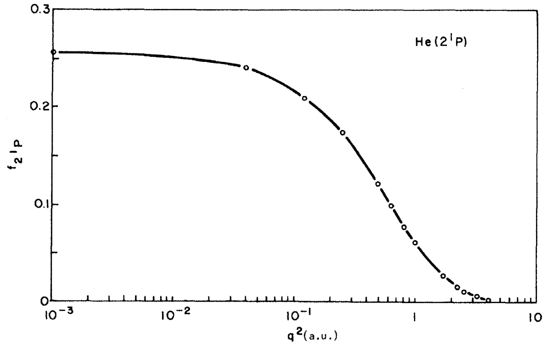

FIG. 1. Generalized oscillator strength for the 2^1P transition in He in a. u. Incident electron energy is 500 eV.

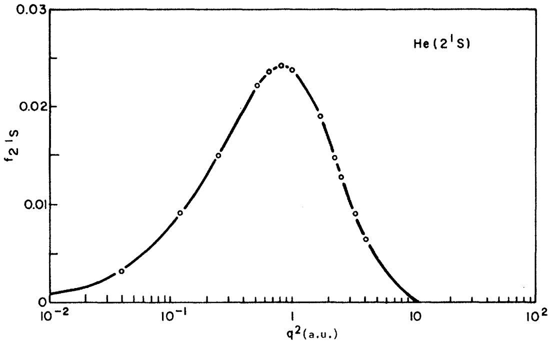

FIG. 2. Generalized oscillator strength for the 2^1S transition in He in a.u. Incident electron energy is 500 eV.

tories and took about 15 min for each state computed. In Tables I and II, we list our values of the generalized oscillator strength for the lowest singlet S and P states of helium. (See Figs. 1 and 2.) For comparison, we also list the results of Kim and Inokuti. For completeness, we also include plots of the oscillator strengths versus q^2, and in Table III a tabulation of the total cross section. As Kim and Inokuti point out, a particularly sensitive test of any oscillator strength calculation is the slope of the curve as q approaches zero for an optically forbidden transition. In Table IV, we give the results for this quantity as calculated by a number of authors using various approximate wave functions. Our results are in excellent agreement with those of Kim and Inokuti indicating the power of our approach. The important point here is not the accuracy of our result on any absolute basis, but what we get out for the time and effort put in. We anticipate applications to larger systems than could be treated by the Kim-Inokuti technique. If present results are indicative, one can expect excellent agreement with experiment.

IV. CONCLUSIONS

We have presented a general formalism to treat the inelastic scattering of high-energy electrons from atoms using linear-response theory. To lowest order, the theory leads to the well-known equations of the random-phase approximation. We have applied these equations to the excitation of the lowest singlet S and P state of helium by 500-eV electrons. Our results are in excellent agreement with the calculations of Kim and Inokuti using much more sophisticated wave functions. On the basis of these results, we anticipate further applications to helium and plan to extend the calculations to heavier systems. We are also looking into the use of response function techniques in low-energy inelastic scattering.

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