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Coherent and Incoherent X-Ray Scattering by Bound Electrons. I. Helium Isoelectronic Sequence*

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Atomic form factors and incoherent scattering functions for the helium isoelectronic sequence through $Z=10$ have been calculated with correlated ground-state wave functions. The wave functions are 120-term configuration-interaction expansions which give total ground-state energies differing from the most accurate available values by 0.02% for He I and 0.0007% for Ne IX. Comparison with published accurate calculations for helium indicates that the scattering factors, both coherent and incoherent, for the other members of the isoelectronic sequence, should be correct to at least three, and probably four, significant figures.

I. INTRODUCTION

The dependence on wave function of the cross sections for coherent and incoherent scattering of a photon by an N-electron atom or atomic ion is contained in the form factor, or atomic scattering factor,

$$F(K)=\sum_{j=1}^{N}\langle\psi_0\,|\,e^{i\vec{K}\cdot\vec{r}_j}\,|\,\psi_0\rangle, \tag{1}$$

and the incoherent scattering function

$$S(K)=N^{-1}\Big(\sum_{j,k=1}^{N}\langle\psi_0\,|\,\exp[i\vec{K}\cdot(\vec{r}_j-\vec{r}_k)]\,|\,\psi_0\rangle - |F(K)|^2\Big), \tag{2}$$

respectively. In Eqs. (1) and (2), the momentum transfer is denoted by $\hbar\vec{K}$, ψ_0 is the ground-state wave function, and $\vec{r}_j$ is the radius vector from the nucleus to the jth electron. The momentum transfer is given by $K=4\pi(\sin\frac{1}{2}\theta)/\lambda$, where λ is the wavelength of the incident photon, and θ the angle between incident and scattered photon directions. The coherent-scattering cross section is

$$d\sigma_c=I_c\,|F(K)|^2\,d\Omega, \tag{3}$$

where I_c is the Thomson cross section. The total incoherent-scattering cross section is

$$d\sigma_i=I_i N S(K)\,d\Omega, \tag{4}$$

where I_i is the Klein-Nishina cross section, which for small values of K may be replaced by the Thomson cross section.

There are now available quite accurate correlated ground-state wave functions for many low-Z atoms and it is no longer necessary to use approximations such as the Hartree-Fock or Thomas-Fermi methods in these cases. This paper describes the results of calculation of form factors and incoherent scattering functions for helium and two-electron atomic ions through $Z=10$.

II. WAVE FUNCTIONS

The two-electron ground-state wave functions used in the present work are 120-term configuration-interaction expansions

$$\psi_{1S}=\sum_{n_1=1}^{8}\sum_{n_2=1}^{n_1}\sum_{l=0}^{n_2-1}c(n_1,\,n_2,\,l)\;\Phi_{1_S}(n_1,\,n_2,\,l), \tag{5}$$

in which each configuration Φ_{1S} is the linear combination of Slater determinants

$$\Phi_{1S}=\sum_m C(l\,l\;0;\;m,\,-m)\times[\,|\,\varphi_{n_1 l m}(\vec{r}_1)\,\alpha\,\varphi_{n_2 l-m}(\vec{r}_2)\,\beta\,| + |\,\varphi_{n_2 l-m}(\vec{r}_1)\alpha\,\varphi_{n_1 l m}(\vec{r}_2)\,\beta\,|\,], \tag{6}$$

where $C(l\,l\,0;\,m,\,-m)$ is a Clebsch-Gordan coefficient, and α and β are the usual spin functions. Normalization constants which usually appear in configurations of the form given by Eq. (6) have been absorbed in the linear variational parameters $c(n_1, n_2, l)$ of Eq. (5). The single-particle functions in Eq. (6) are members of the denumerably complete set of functions[1,2]

$$\varphi_{n\,l\,m}(\rho,\theta,\phi)=\frac{[\,\Gamma(n-l)]^{1/2}}{[\,\Gamma(n+l+1)]^{3/2}}\;\rho^{l}\times e^{-\rho/2}\,L^{2l+1}_{n-l-1}\,(\rho)\,Y^{m}_{l}(\theta,\phi) \qquad (7)$$

of the variable $\vec{\rho}=2\eta\vec{r}$, where η is the single nonlinear parameter which was used in the variational calculation of the ground-state energies.[3,4] The generalized Laguerre function $L^{2l+1}_{n-l-1}(2\eta r)$ in Eq. (7) can be written in terms of a confluent hypergeometric series as

$$L^{b}_{a}(\rho)=\frac{[\Gamma(a+b+1)]^{2}}{\Gamma(a+1)\,\Gamma(b+1)}\sum_{j}\frac{\Gamma(-a+j)\,\Gamma(b+1)}{\Gamma(-a)\,\Gamma(b+1+j)}\,\frac{\rho^{j}}{j!}\,. \qquad (8)$$

Ground-state energies obtained with these functions have been found to agree with the best available values to within 0.02% for $Z=2$, with the error decreasing monotonically to 0.0007% for $Z=10$.

III. METHOD OF CALCULATION

Substitution of the wave function [Eq. (5) into Eqs. (1) and (2)] results in the form factor $F(K)$ and incoherent scattering function $S(K)$ being given as

$$F(K)=\sum_{ij}c_i f_{ij} c_j, \qquad (9)$$

and

$$2S(K)=\sum_{ij}c_i s_{ij} c_j-|F(K)|^2\,, \qquad (10)$$

where the components c_i are the $c(n_1, n_2, l)$ of Eq. (5). The c_i are independent of magnetic quantum numbers, as are the matrix elements f_{ij} and s_{ij}. However, the magnetic quantum numbers do appear in the calculation of these matrix elements. That is,

$$f_{ij}=\sum_{m,m'}\langle i\,|\,e^{i\vec{K}\cdot\vec{r}_1}+e^{i\vec{K}\cdot\vec{r}_2}\,|j\rangle \qquad (11)$$

and

$$s_{ij}=\sum_{m,m'}\langle i\,|\,e^{i\vec{K}\cdot(\vec{r}_1-\vec{r}_2)}+e^{i\vec{K}\cdot(\vec{r}_2-\vec{r}_1)}\,|j\rangle+2\langle i\,|j\rangle\,, \qquad (12)$$

where $\langle i|=\langle n_1' l' m' n_2' l'(-m')|$ and $|j\rangle=|n_1 l m n_2 l,(-m)\rangle$. The second term in Eq. (12) results from the fact that the sum in Eq. (2) includes the case $\vec{r}_j=\vec{r}_k$.

If the coordinate system is chosen so that the momentum change is in the positive z direction, the operators in Eqs. (11) and (12) may be written[5]

$$e^{iKz}=\sum_{\lambda=0}^{\infty}i^{\lambda}[4\pi(2\lambda+1)]^{1/2}\,j_{\lambda}(Kr)\,Y^{0}_{\lambda}(\theta), \qquad (13)$$

where $j_\lambda(Kr)$ is a spherical Bessel function. When Eq. (13) is substituted into Eqs. (11) and (12), the resulting expressions are linear combinations of products of integrals over angular and radial coordinates. The angular integrals contain products of three spherical harmonics, and result in sums over products of Clebsch-Gordan and Racah coefficients which may be evaluated in closed form by methods described by Edmonds[5] or Rose.[6] The results are

$$\begin{aligned}f_{ij}=\delta'_{l\,l}[&\langle n_1' l\rangle\langle n_2' l\,|\,j_0(Kr)\,|\,n_2 l\rangle\\&+\langle n_1' l\,|\,j_0(Kr)\,|\,n_1 l\rangle\langle n_2' l\,|\,n_2 l\rangle\\&+\langle n_2' l\,|\,n_1 l\rangle\langle n_1' l\,|\,j_0(Kr)\,|\,n_2 l\rangle\\&+\langle n_2' l\,|\,j_0(Kr)\,|\,n_1 l\rangle\langle n_1' l\,|\,n_2 l\rangle]\end{aligned} \qquad (14)$$

and

$$\begin{aligned}s_{ij}=2\langle i\,|\,j\rangle+2(-1)^{l-l'}&\left(\frac{2l+1}{2l'+1}\right)^{1/2}\sum_{\lambda}(2\lambda+1)\\&\times[C(l\lambda l';\,0\,0)]^{2}\,[\langle n_1' l'\,|\,j_\lambda(Kr)\,|\,n_1 l\rangle\\&\times\langle n_2' l'|\,j_\lambda(Kr)\,|\,n_2 l\rangle+\langle n_1' l'|\,j_\lambda(Kr)\,|\,n_2 l\rangle\\&\times\langle n_2' l'|\,j_\lambda(Kr)\,|\,n_1 l\rangle].\end{aligned} \qquad (15)$$

The sum over λ in Eq. (15) is limited by the parity coefficient $C(l\lambda l';\,0\,0)$ which vanishes unless $|l-l'|\leq\lambda\leq|l+l'|$, and $l+l'+\lambda$ is even. The evaluation of the radial overlap integral

$$\langle n_a l\,|\,n_b l\rangle=\int_0^{\infty}R_{n_a l}(r)\,R_{n_b l}(r)\,r^2 dr \qquad (16)$$

has been described previously.[3] The other radial integral, which is required,

$$\langle n_a l_a\,|\,j_\lambda(Kr)\,|\,n_b l_b\rangle=\int_0^{\infty}R_{n_a l_a}(r)R_{n_b l_b}(r)\,j_\lambda(Kr)\,r^2 dr \qquad (17)$$

can be expressed as a linear combination of inte-

TABLE I. Comparison of form factors $F(K)$ for He I.

$\frac{\sin\frac{1}{2}\theta}{\lambda}$ (Å^{-1})	Ka_0	Analytic Hartree-Fock (see Ref. 7)	Numerical Hartree-Fock-Slater (see Ref. 8)	Hylleraas (see Ref. 7)	Present work
0.0	0.0	2.0	2.0	2.0	2.0
0.025	0.166 243	1.989 2		1.989 06	1.989 1
0.050	0.332 485	1.957 1	1.955	1.956 82	1.956 9
0.075	0.498 728	1.905 7		1.905 00	1.905 2
0.100	0.664 971	1.837 2	1.831	1.836 20	1.836 4
0.150	0.997 456	1.662 6	1.652	1.660 85	1.661 2
0.200	1.329 94	1.460 4	1.447	1.458 16	1.458 5
0.250	1.662 43		1.241		1.252 2
0.300	1.994 91	1.060 2	1.049	1.058 42	1.058 6
0.350	2.327 40		0.879		0.886 26
0.400	2.659 88	0.738 3	0.734	0.737 85	0.737 94
0.450	2.992 37		0.613		0.613 13
0.500	3.324 85	0.508 9	0.511	0.509 48	0.509 53
0.550	3.657 34		0.428		0.424 20
0.600	3.989 81	0.352 9	0.359	0.354 04	0.354 16
0.650	4.322 31		0.303		0.296 71
0.700	4.654 80	0.248 1	0.256	0.249 36	0.249 52
0.750	4.987 28		0.217		0.210 67
0.800	5.319 77	0.177 2	0.185	0.178 51	0.178 60
0.850	5.652 25		0.158		0.152 03
0.900	5.984 74	0.128 8	0.136	0.129 96	0.129 95
0.950	6.317 22		0.117		0.111 54
1.00	6.649 71	0.095 23	0.101	0.096 185	0.096 123
1.05	6.982 19		0.088		0.083 167
1.10	7.314 68	0.071 52	0.076	0.072 308	0.072 239
1.15	7.647 17		0.066		0.062 983
1.20	7.979 65	0.054 53	0.058	0.055 162	0.055 114
1.25	8.312 14		0.051		0.048 398
1.30	8.644 62	0.042 16	0.045	0.042 662	0.042 643
1.35	8.977 11		0.040		0.037 695
1.40	9.309 59	0.033 02	0.035	0.033 416	0.033 424
1.45	9.642 08				0.029 725
1.50	9.974 56	0.026 17	0.028	0.026 483	0.026 511
1.60	10.639 5		0.022		0.021 257
1.70	11.304 5		0.018		0.017 216
1.80	11.969 5		0.015		0.014 071
1.90	12.634 4		0.012		0.011 599
2.00	13.299 4		0.010		0.009 636 3

grals over products of the spherical Bessel functions, powers of r, and exponentials. The evaluation of this integral is described in the Appendix. As shown in the Appendix, the radial integrals, and therefore the matrix elements, depend only on the ratio of the variational parameter η to the momentum transfer K. It is therefore possible to calculate values of $F(K)$ or $S(K)$ for all members of the isoelectronic sequence with a single set of matrix elements f_{ij} or s_{ij}. This results in a considerable reduction in computer time required.

IV. RESULTS AND DISCUSSION

Tables I and II compare the results of the present work for He I with the recent results of Kim and Inokuti obtained with Hylleraas-type and analytic Hartree-Fock wave functions.[7] Table I also shows the form factors obtained by Hanson *et al.* with numerical Hartree-Fock-Slater wave functions,[8] and Table II shows the incoherent scattering functions obtained by Cromer and Mann[9] with numerical Hartree-Fock wave functions. The agreement with the values obtained with Hylleraas-type wave functions is in most cases to within four significant figures. This is somewhat better than might be expected since the Hylleraas-type functions are considerably more accurate in terms of total energy than are the functions used in the present work.[10]

Numerical values for Li II through Ne IX are too voluminous to include here. However, they are available in report form.[11] The values for the higher members of the isoelectronic sequence should be at least as accurate as those for He I,

TABLE II. Comparison of incoherent scattering functions $2S(K)$ for He I.

$\frac{\sin\frac{1}{2}\theta}{\lambda}$ (Å^{-1})	Ka_0	Analytic Hartree-Fock (see Ref. 7)	Numerical Hartree-Fock (see Ref. 9)	Hylleraas (see Ref. 7)	Present work
0.0	0.0	0.0	0.0	0.0	0.0
0.025	0.166 243	0.021 63		0.020 637	0.020 589
0.050	0.332 485	0.084 80	0.085	0.080 716	0.080 542
0.075	0.498 728	0.184 3		0.175 09	0.174 75
0.100	0.664 971	0.312 3	0.312	0.296 21	0.295 75
0.150	0.997 456	0.617 8	0.618	0.584 06	0.583 52
0.200	1.329 94	0.933 6	0.934	0.881 02	0.880 56
0.250	1.662 43				1.145 7
0.300	1.994 91	1.438 0	1.438	1.362 73	1.362 4
0.350	2.327 40				1.530 4
0.400	2.659 88	1.727 4	1.727	1.656 89	1.656 6
0.450	2.992 37				1.749 7
0.500	3.324 85	1.870 5	1.871	1.817 72	1.817 5
0.550	3.657 34				1.866 7
0.600	3.989 81	1.937 7	1.938	1.902 45	1.902 3
0.650	4.322 31				1.928 1
0.700	4.654 80	1.969 2	1.969	1.946 82	1.946 7
0.750	4.987 28				1.960 3
0.800	5.319 77	1.984 3	1.984	1.970 27	1.970 2
0.850	5.652 25				1.977 5
0.900	5.984 74	1.991 7	1.992	1.982 90	1.982 9
0.950	6.317 22				1.986 9
1.00	6.649 71	1.995 5	1.995	1.989 85	1.989 9
1.05	6.982 19				1.992 24
1.10	7.314 68	1.997 4		1.993 79	1.993 96
1.15	7.647 17				1.995 28
1.20	7.979 65	1.998 5		1.996 08	1.996 29
1.25	8.312 14				1.997 07
1.30	8.644 62	1.999 1		1.997 45	1.997 67
1.35	8.977 11				1.998 15
1.40	9.309 59	1.999 5		1.998 29	1.998 52
1.45	9.642 08				1.998 81
1.50	9.974 56	1.999 7	2.000	1.998 82	1.999 039
1.60	10.639 5				1.999 367
1.70	11.304 5				1.999 576
1.80	11.969 5				1.999 713
1.90	12.634 4				1.999 802
2.00	13.299 4		2.000		1.999 862

since the relative error in the wave function decreases with increasing Z. Also, the wave functions used here have been used to calculate the expectation value $\frac{1}{2}\langle(r_1^2 + r_2^2)\rangle$, which is closely related to the atomic form factor, and the results for the higher-Z ions were considerably better than those for He I.[4]

A final result of this work is evident from Table I. It can be seen that the agreement among the analytic Hartree-Fock results and those obtained with the two types of correlated wave functions is closer than the agreement between the analytic Hartree-Fock and the Hartree-Fock-Slater results. The difference between the Hartree-Fock and Hartree-Fock-Slater wave functions is that the former includes the effects of exchange while the latter handles exchange in an approximate manner. On the other hand, neither the Hartree-Fock nor Hartree-Fock-Slater wave function includes the effect of the interelectron correlation energy. This suggests that at least for a two-electron closed-shell atom the effects of exchange are more important than correlation effects in the calculation of expectation values.

APPENDIX A: INTEGRALS CONTAINING SPHERICAL BESSEL FUNCTIONS

The integral to be evaluated is of the general form

$$I_1 = \int_0^\infty e^{-ar} j_l(br) r^m dr, \tag{A1}$$

where a and b are real and m is an integer such that $m > l$. With the change of variable $\rho = ar$ and

replacement of the spherical Bessel function by

$$j_l(br) = \left(\frac{\pi}{2br}\right)^{1/2} J_{l+1/2}(br), \tag{A2}$$

the integral I_1 becomes proportional to

$$I_2 = \int_0^\infty e^{-\rho} J_\lambda(k\rho)\, \rho^{\lambda+n} d\rho, \tag{A3}$$

where $k = b/a$, $\lambda = l + \frac{1}{2}$, and $n = m - l - 1$. There are several expressions in the literature for the integral I_2, all of which contain a hypergeometric function with nonintegral parameters.[12,13] This is an infinite series which may or may not converge rapidly. One method which has been suggested for the evaluation of I_1 is that of using the explicit form of the spherical Bessel function in terms of trigonometric functions.[14] This requires the computation of a linear combination of terms of the form

$$\cos[(n+1)\tan^{-1}(b/a)],$$

and $$\sin[(n+1)\tan^{-1}(b/a)],$$

where $n > -1$. While it is true that this results in a closed form expression for the integral, in actual practice the computation of terms such as the above may be just as time consuming as the summation of the hypergeometric series.

A convenient starting point is the expression given by Erdelyi *et al.*[12]:

$$I_2 = x^{\lambda+n+1}\Gamma(2\lambda+n+1) P_{\lambda+n}^{-\lambda}(x), \tag{A4}$$

where $x = (1+k^2)^{-1/2}$. Since k is real, $|x| \leq 1$, and the Legendre function may be expressed in terms of a hypergeometric series as

$$P_{\lambda+n}^{-\lambda}(x) = \frac{1}{\Gamma(\lambda+1)}\left(\frac{1-x}{1+x}\right)^{\lambda/2} \times F[-\lambda-n,\ \lambda+n+1,\ \lambda+1;\ \tfrac{1}{2}(1-x)]. \tag{A5}$$

If the Euler transformation is applied to the hypergeometric series, the resulting expression for the integral I_2 is

$$I_2 = x^{\lambda+n+1}\left(\frac{1-x}{1+x}\right)^{\lambda/2}\left(\frac{1+x}{2}\right)^{\lambda+n}\frac{\Gamma(2\lambda+n+1)}{\Gamma(\lambda+1)} \times F\left(-\lambda-n,\ -n,\ \lambda+1;\ \frac{x-1}{x+1}\right). \tag{A6}$$

The hypergeometric series in Eq. (A6) is now a finite sum, and can be expressed in a form which is quite amenable to rapid computation. Written out explicitly, and with the parameter λ replaced by $l + \frac{1}{2}$, it is

$$F\left(-l-n-\tfrac{1}{2},\ -n,\ l+\tfrac{3}{2};\ \frac{x-1}{x+1}\right) = \sum_{j=0}^{n}\binom{n}{j}\frac{\Gamma(n+l+\frac{3}{2})}{\Gamma(n+l+\frac{3}{2}-j)} \times \frac{\Gamma(l+\frac{3}{2})}{\Gamma(l+\frac{3}{2}+j)}\left(\frac{x-1}{x+1}\right)^j, \tag{A7}$$

where use has been made of the relationship

$$\Gamma(-z+j)/\Gamma(-z) = (-1)^j\,\Gamma(z+1)/\Gamma(z-j+1), \tag{A8}$$

which for integer z is also equal to

$$(-1)^j j!\binom{z}{j}.$$

The Γ functions with half-integer arguments in Eq. (A7) may be expressed in terms of factorials as

$$\Gamma(n+\tfrac{1}{2}) = [(2n-1)!/(n-1)!]\,\sqrt{\pi}/2^{2n-1}, \tag{A9}$$

with the result

$$F\left(-l-n-\tfrac{1}{2},\ -n,\ l+\tfrac{3}{2};\ \frac{x-1}{x+1}\right) = \sum_{j=0}^{n}\frac{\binom{n}{j}\binom{l+j}{j}\binom{2n+2l+1}{2j}}{\binom{n+l}{j}\binom{2l+2j+1}{2j}}\left(\frac{x-1}{x+1}\right)^j. \tag{A10}$$

The final expression for I_1, in terms of the integers m and l and the variable $k = b/a$, is

$$I_1 = \frac{1}{a^{m+1}}\,\frac{(2k)^l}{(1+k^2)^m}\left\{\tfrac{1}{2}\left[1+(1+k^2)^{1/2}\right]\right\}^{m-l-1}\frac{l!(m+l)!}{(2l+1)!} \times \sum_{j=0}^{m-l-1}\left[\frac{\binom{m-l-1}{j}\binom{l+j}{j}\binom{2m-1}{2j}}{\binom{m-1}{j}\binom{2l+2j+1}{2j}}\right]\left(\frac{1-(1+k^2)^{1/2}}{1+(1+k^2)^{1/2}}\right)^j. \tag{A11}$$

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Noise Calculations for Optical Parametric Oscillators*

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A general approach for describing complex quantum-mechanical systems in terms of c-number variables is discussed on the basis of a formalism developed by Lax. We apply this formalism to the particular problem of noise fluctuations in parametric oscillators. From a microscopic Hamiltonian describing light interacting with optically active atoms in a cavity with loss, we arrive at an equation for the motion in terms of c-number variables. We then solve this system for the noise properties of the signal mode near threshold and above, when the pump and idler modes may be treated adiabatically, and compare our work with the results of Graham, who treated the signal and idler symmetrically.

I. INTRODUCTION

The obvious device possibilities of a low-noise broadly tunable optical source have aroused considerable interest, both experimentally[1–3] and theoretically,[4–13] in the optical parametric oscillator. It has long been realized that in order to analyze the noise properties of such an oscillator a quantum-mechanical treatment is necessary.[4] It is also clear that a quantum treatment of a nonlinear problem is computationally cumbersome due to the noncommutability of the algebra. Consequently, formalisms which help to avoid these difficulties evolved particularly directed towards analysis of the laser.[14–19]

We have found that a formalism due to Lax,[17] which translates the quantum-mechanical problem into a more workable c-number problem, also has the added advantage of consolidating all of the necessary information into one equation. This makes the systematic analysis and solution of the parametric oscillator, and related quantum optics problems, much more straightforward.

Probably the most comprehensive and complete theoretical treatment of the quantum fluctuations of optical parametric oscillators was given in a series of three papers by Haken and Graham,[11] and by Graham.[12,13] In the first of these,[11] starting from a microscopic Hamiltonian describing light interacting with a medium of optically active atoms in a Fabry-Perot cavity, quantum Langevin equations for the optical field and atomic variables were derived. Using an iteration technique, the atomic variables were eliminated leaving nonlinear field equations. These equations were solved in operating ranges far above and far below cavity threshold. In the second paper,[12] Graham proposed an "effective Hamiltonian" which gives the quantum Langevin equations for parametrically interacting light fields in agreement with Ref. 4. The coherent state representation for boson fields was used to obtain a "classical" Fokker-Planck equation. In the final paper of the series,[13] this "classical" Fokker-Planck equation was used to write down a stochastically equivalent set of c-number Langevin equations, which Graham was able to solve, with certain approximations, in the near threshold region.

Our approach will be quite different. It avoids the complications of nonlinear quantum-mechanical Langevin equations with cross-correlated quantum-mechanical random forces.[11] From one quantum-mechanical equation, the master equation, describing the microscopic field-atomic interaction with loss, we are able to write down a c-number equation. The atomic variables are eliminated by iterating c-number Langevin equations, and a classical Fokker-Planck equation for the light fields is found directly without appealing to an effective Hamiltonian. The procedure translates the mi-