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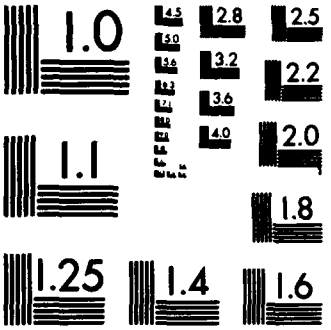

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Dr. Kristl Hathaway
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Annual Letter Report

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The main achievement in the period May 1,1986 through April 30, 1987 is the development of computer codes for electronic structure calculations for systems without any rotational symmetry. At present, these codes are only tested for atomic systems, but exactly the same technique will be used for solids. This will then enable us to calculate the effects of spin-orbit coupling on the electronic structure of solids. In the next paragraph I will describe the numerical techniques which are used. After that detailed results of the investigation of the carbon atom are given.

When one applies a magnetic field to a solid all rotational symmetry is broken, unless the field points along a high symmetry direction of the crystal. Therefore the charge density and the potential appearing in electronic structure calculations only show translational symmetry. This immediately brings forward the question how to represent these quantities numerically. Since the effects of magnetism are usually smaller than the crystal field terms in the potential and the charge density, these two contributions have to be treated separately. In our computer programs for bulk solids the crystal field effects are treated in a standard way by representing them in terms of symmetry adapted spherical harmonics inside the muffin tin spheres and plane waves in the interstitial. The changes due to magnetic effects are represented on a numerical grid. We choose our numerical points equally spaced along the primitive translation vectors, because this will give the best precision for integrating periodic functions. Recall that a periodic function, which is not strongly peaked, in one dimension is best integrated using a trapezoidal rule.

We start from a calculation without magnetic fields and obtain self-consistent energy eigenvalues and wave functions. These serve as a basis for a next calculation in which we include all external fields. The matrix elements of the external fields are calculated by transforming the wave functions to the numerical grid used for representing the potential. The relative error in the numerical calculation of these matrix elements is not small, but since the elements themselves are small we only introduce small absolute errors in this procedure. After calculating the matrix elements the Hamiltonian is diagonalized and new values of the wave functions are obtained. This procedure is then continued until self-consistency is reached. After that we calculate the total energy and other quantities. It is very important that one only has to calculate the changes in all quantities compared to the reference calculation, in order to minimize the numerical errors.

We have tested this procedure in atomic calculations where our reference always was a spherical, paramagnetic calculation. In this case we solve the full Dirac equation and make the following extension to the local density approximation. At every point in space the charge density and spin density are known for the last iteration. From this we construct locally a majority and a minority density and calculate the exchange-correlation potentials using the von Barth-Hedin form of this potential as parametrized by Janak [1]. These potentials are then written as a Coulomb term and a magnetic term. For the direction of the magnetic field we then take the local spin direction. Finally, we assume that this magnetic field couples to the spin only.

The total energy obtained for Carbon in this procedure can be compared directly with Hartree-Fock results [2] by assuming that the total energy of our reference calculation is equal to the value obtained by spherically averaging over all configurations in the Hartree-Fock calculations (Figure 1). The restricted and spin-polarized Hartee-Fock results are almost identical for the one determinant result. In an earlier calculation, von Barth [3] obtained the total energy of Carbon for broken symmetry, but his work was not self-consistent. His result was close to the spin-polarized spherical result [4]. Since in the local density approximation one also includes correlation, one might expect that a self-consistent calculation with broken symmetry reduces the total energy even more. This was indeed found in our calculations, but the real value of the total energy, as obtained in a configuration interaction calculation, is still lower. The main error is due to the contribution of l=2 states, which are important in the exact result. In local density, these states are pushed away from the nucleus by the centrifugal forces in the Hamiltonian and the exchange-correlation potential drops too rapidly to counteract these forces and to bring some l=2 wave functions closer to the nucleus.

Next, we investigated the total energy and the induced dipole moment of Carbon in an electric field. Our calculations show that the Carbon atom wants to align itself with its orbital angular momentum parallel to the field (Figure 2). This is a direct result from the iterations to self-consistency, which show that any other alignment of the angular momentum gives a higher total energy and a new charge density which is closer to the preferred alignment. In the aligned situation we then evaluate the polarizability of the Carbon atom. The number we find is 1.2 Angstrom^3, which is smaller than the experimental number of 1.76 Angstrom^3. The polarizability is clearly anisotropic, but we can only estimate the values for directions perpendicular to the angular momentum because of the alignment of the atom. We estimate that the perpendicular value is around 0.5 Angstrom^3.

The cause of this discrepancy is exactly the same as that for the total energy, and also the relative size of the deviation is similar. In both cases the error is about a factor of 1/3. The electric field gradient couples the p states to the s states and also to d-like states. The latter type of states are too far extended from the nucleus in the local density approximation and hence do not give a large enough contribution to both the total energy and the polarizability. Note that this situation is different in the case of closed shell atoms, where the local density approximation gives much better results[5]. This shows that the average extend of the d-like states in the local density approximation is well represented, but that in cases where the spherical symmetry is broken the splitting of the d-like states is not well represented. In carbon the d-like states which are lowered in energy due to the lower symmetry are not pulled in far enough towards the nucleus.

At present we are investigating the effect of diverging magnetic fields on a carbon atom. Here we simulate the conditions as seen in the Stern-Gerlach experiments. Our preliminary results show that the carbon atom develops a dipole moment in this case. Since there is no counter force due to the magnetic field on the nucleus (as there is in an external electric field) this means that there is a net force on the atom. More quantitative results are on their way.

References.

[1] U. von Barth and L. Hedin, J. Phys. C 5, 1629 (1972); J. F. Janak, Solid State Commun. 25, 53 (1978).

[2] J. V. Mallow and P. S. Bagus, J. Quan. Spectrosc. Radiat. Transfer 16, 409 (1976); P. S. Bagus and C. M. Mosser, J. Phys. B 2, 1214 (1969).

[3] U. von Barth, Phys. Rev. A 20, 1693 (1979).

[4] M. Weinert, E. Wimmer, and A. J. Freeman, Phys. Rev. B 26, 4571 (1982).

[5] G. D. Mahan, Phys. Rev. A 22, 1780 (1980); A. Zangwill and P. Soven, Phys. Rev. A 21, 1561 (1980).

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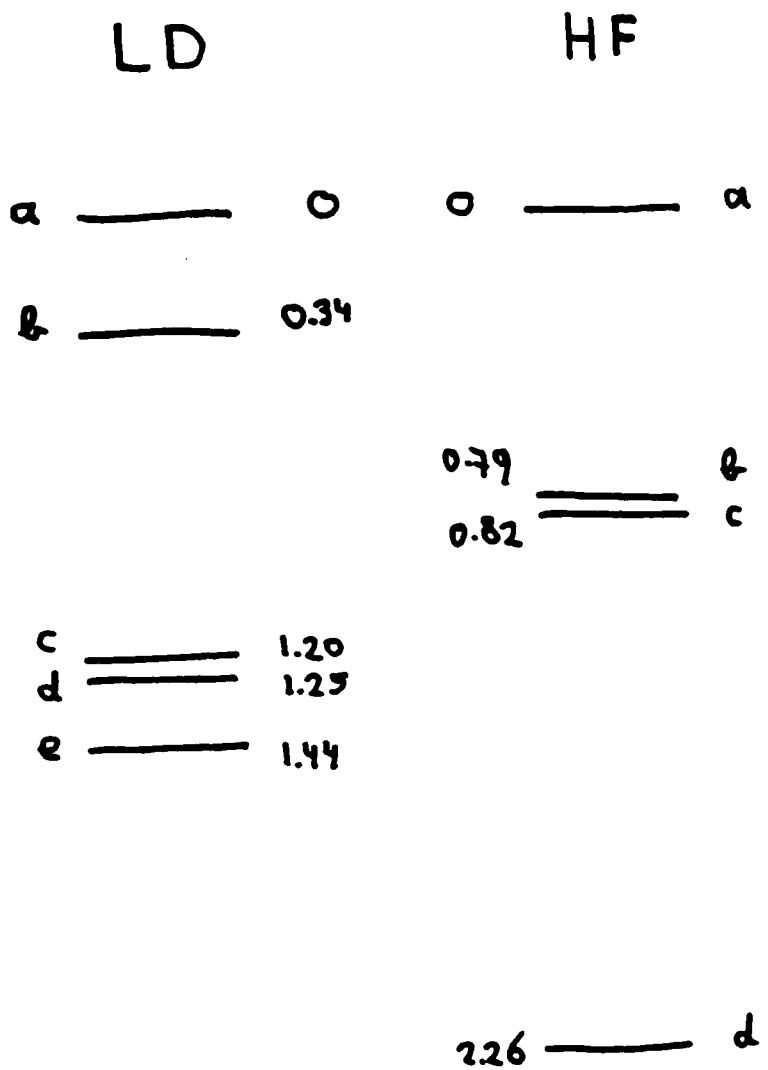

Figure 1.

Energy splittings (eV) of the carbon atom in local density (LD) and Hartree-Fock (HF). In LD a) indicates the energy of a non-spin-polarized, spherical calculation and this has the same energy as a) in HF, which is the result from an spherical average of configurations. LD.b is a non-spin-polarized, non-spherical result, LD.c is spin-polarized, non-spherical after one iteration, LD.d is spin-polarized, spherical, and LD.e is spin-polarized, non-spherical, self-consistent. HF.b is obtained in a spin-restricted calculation, while HF.c is a spin-polarized calculation. HF.d finally is the result of a multi-configuration calculation and represents the true ground state energy.

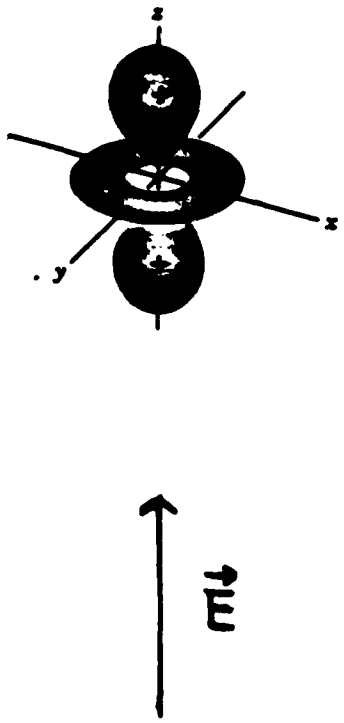

Figure 2.

Orientation of the carbon atom in an external electric field along the z-axis. The region labeled - indicates an excess of electrons as compared to the spherical average, while the region marked with + denotes less electrons than the spherical average (and hence a positive charge).

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