# PHYSICAL REVIEW A

## GENERAL PHYSICS

## Magnetic behavior of icosahedral $C_{60}$

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We have identified a contribution to the ring current magnetic susceptibility of icosahedral $C_{60}$ which diverges paramagnetically as the relative strengths of the two inequivalent bonds of this molecule are varied. This divergence is invoked to explain the near cancellation of the diamagnetism generally expected of aromatic hydrocarbons.

## I. INTRODUCTION

Kroto and co-workers[1,2] have recently shown that 60 carbon atoms are capable of forming a very stable molecule. They point out that the uniqueness of the number 60 may be explained when one considers the possibilities for forming closed surfaces from fragments of graphite. The smallest such structure having nearly planar $sp^2$ bonding is the truncated icosahedron (Fig. 1) with exactly 60 carbon atoms. The stability of this structure and its aromatic character are now also supported by a number of theoretical studies.[3-9]

The semimetallic nature of the $\pi$ electrons in graphite would lead one to expect a large diamagnetic response for $C_{60}$. Indeed, Kroto and co-workers[1] predicted that substantial ring currents would be generated in $C_{60}$ and that the NMR chemical shift for a centrally located atom would be large. Contrary then to these classical expectations are the results of a calculation using the quantum-mechanical London theory[10] which show that the $\pi$-electron ring current magnetic susceptibility of $C_{60}$ is anomalously small—perhaps only one-fifth that of benzene and of the wrong sign.[11] The purpose of this letter is to explain this somewhat surprising result in terms of a near cancellation by strong Van Vleck paramagnetism.[12]

The unique icosahedral structure of $C_{60}$ has not yet been demonstrated. Although no nonspherical candidates with enhanced stability at mass number 60 have been identified, this possibility remains open. Another possibility is that the spherical molecule occurs in a number of different structures, related by rearrangements of five- and six-membered rings.[13] NMR chemical shift measurements are sensitive probes of structure and, we believe, would discriminate even between the different spherical structures.

The London theory[10] of the magnetic response of aromatic hydrocarbons consists in replacing each element $\beta_{rs}$ in the $\pi$-orbital bonding matrix by $\beta_{rs} \exp(i\theta_{rs})$, where $\theta_{rs}$ is the line integral of the vector potential along a straight-line path joining the atomic orbitals $r$ and $s$. The perturbations of first and second order in the magnetic field are given, respectively, by the pure imaginary matrix $(\Delta_1)_{rs} = i\beta_{rs}\theta_{rs}$ and the real matrix $(\Delta_2)_{rs} = -\frac{1}{2}\beta_{rs}\theta_{rs}^2$. If the unperturbed ground state is nondegenerate the expectation value of $\Delta_1$ vanishes and the magnetic response consists of the two terms,

$$\delta E_{\text{mag}} = \sum_{j(\le n)} \langle j | \Delta_2 | j \rangle + \sum_{j(\le n)} \sum_{k(>n)} \frac{|\langle k | \Delta_1 | j \rangle|^2}{E_j - E_k}.$$

(1)

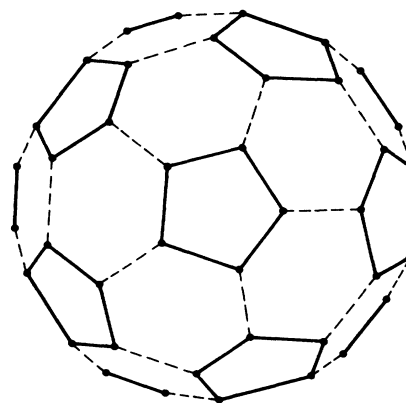The indices $j$ and $k$ refer to molecular orbitals, the first $n$



FIG. 1. Structure of icosahedral $C_{60}$, emphasizing the decomposition into 12 five-membered rings. The dashed and solid bonds have, respectively, resonance integrals $k\beta$ and $\beta$.

      

of which are occupied in the ground state. These terms represent the separation of the magnetic susceptibility into diamagnetic and paramagnetic parts; this separation being dependent on the gauge choice used in the definition of $\theta_{rs}$. Thus there is no fundamental distinction between the terms involving (i) a ground-state expectation value and (ii) a sum over excited states. The second, strictly negative term is known as Van Vleck paramagnetism.[12] In spite of the gauge arbitrariness of this separation, the diamagnetic and paramagnetic terms behave very differently in one special situation. Suppose we modify the $\pi$-electron Hamiltonian by introducing a continuous variation of the matrix elements $\beta_{rs}(k)$, where $k$ is a parameter. Now, if $k = 1$ corresponds to the original molecule, it is possible that for the fictitious molecule at $k = 0$ the susceptibility actually diverges. This divergence distinguishes between the two terms in (1); provided the $\beta_{rs}(k)$ remain finite, the diamagnetic term is clearly finite whereas the paramagnetic term can diverge in the event that the energy levels $E_n$ and $E_{n+1}$ cross (at $k = 0$). The generic crossing of two levels in the presence of a magnetic field $H$ is described (to lowest order in $H$) by the Hamiltonian matrix

$$\begin{bmatrix} -k & iH \\ -iH & k \end{bmatrix} ,$$

where the gap (in the absence of $H$) is assumed to vary linearly with $k$. From the energy of the lowest (i.e., occupied) level,

$$E = -(k^2 + H^2)^{1/2} \sim -|k| - \frac{1}{2}\frac{H^2}{|k|} + \cdots ,$$

we see that the susceptibility diverges paramagnetically as $k \rightarrow 0$. Moreover, this divergence is associated with the appearance of a permanent magnetic moment at the special point $k = 0$.

Our strategy for explaining the cancellation of diamagnetism in $C_{60}$ is to exhibit a divergent paramagnetic susceptibility of the kind described above. The modification of the molecule is illustrated in Fig. 1. The 60 carbon atoms in the structure may be divided into 12 sets where each set forms a five-membered ring (5MR). The $\pi$-orbital matrix elements $\beta_{rs}$ are now of two types: between carbon atoms on different 5MR's ($\beta_1$), and between carbon atoms on the same 5MR ($\beta_2$). The former can be varied relative to the latter using the parametrization $\beta_1 = k\beta$, $\beta_2 = \beta$; $0 \leq k, \beta = \text{const} > 0$.

It is easy to check that $k = 0$ is a special point where the (modified) $C_{60}$ molecule has a permanent magnetic moment. For this case $C_{60}$ consists of 12 disconnected 5MR's, each supporting five states with rotational quantum numbers $m = 0, \pm 1, \pm 2$ about the pentagon axis. The 60 molecular orbitals thus collapse onto three highly degenerate sets of orbitals with energies $E_m = -2\beta \cos(2\pi m/5)$. Of the 30 orbitals needed to accommodate 60 electrons, 12 can have the lowest energy with $m = 0$. That leaves 18 electron pairs to be distributed among the $|m| = 1$ orbitals of which there are 24. Each of the 12 pairs of $|m| = 1$ orbitals undergo a Zeeman splitting proportional to the magnetic flux en-

closed by the 5MR. Consequently, the sum of the energies of the 18 lowest $|m| = 1$ orbitals also varies linearly with magnetic field.

This analysis is to some extent incomplete. A more detailed treatment in Sec. III shows that in fact eight of the $|m| = 1$ orbitals coalesce much faster than the remaining 16 as $k \rightarrow 0$. These consist of two degenerate sets of three and five orbitals which separately transform as irreducible representations of the icosahedral group. These orbitals are also the highest occupied and lowest unoccupied orbitals of $C_{60}$. Their energy difference $\Delta E$ is the energy gap of the molecule and varies as $k^2$ for small $k$. Thus the magnetic response of $C_{60}$ is strongly influenced by a $k^{-2}$ paramagnetic divergence at $k = 0$.

Our results for small $k$, discussed in Secs. III and IV, may be compared with the results of Ref. 11 where the full range of $k$ was treated. Details of the latter calculation are given in the Appendix.

## II. CHOICE OF UNITS

Our basic unit of length is the carbon-carbon bond length $a = 1.4$ Å; for energy, the resonance integral $\beta$ ($= \beta_2$). A unit magnetic field is $\hbar c/ea^2 (= 3.35 \times 10^8$ G) and represents a single flux quantum passing through the area $a^2$. The unit of magnetic susceptibility is therefore $\beta(e/\hbar c)^2 a^4$ with a positive sign indicating paramagnetism. Traditionally the $\pi$-electron magnetic susceptibility $\chi_\pi$ of planar aromatic hydrocarbons has been expressed in terms of the square of the area enclosed by the rings of the molecule rather than $a^4$. To aid in comparing our results with other work, we note that in our units the London theory gives $\chi_\pi = -\frac{3}{2}$ for benzene.

## III. ORBITALS AND ENERGY LEVELS FOR SMALL $k$

The appropriate basis functions for the $k \rightarrow 0$ limit of $C_{60}$ have the form

$$u_m^\alpha = \frac{1}{\sqrt{5}} \sum_{n=1}^{5} e^{i2\pi mn/5} \Phi_n^\alpha(\mathbf{r}) , \qquad (2)$$

where $\Phi_1^\alpha, \ldots, \Phi_5^\alpha$ is the sequence of atomic orbitals around a 5MR with label $\alpha$ ($\alpha = 1, \ldots, 12$). By neglecting matrix elements between basis functions with different $|m|$, the $\pi$-electron Hamiltonian reduces to $\mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2$. $\mathcal{H}_{|m|}$ has diagonal elements $E_m = -2\cos(2\pi m/5)$ and off-diagonal elements of the form

$$-(k/5)e^{-i2\pi mn_\alpha/5} e^{i2\pi m'n_\beta/5} , \qquad (3)$$

where $|m| = |m'|$ and $n_\alpha, n_\beta$ refer to the particular pair of atomic orbitals in the expansion (2) where adjacent 5MR's $\alpha$ and $\beta$ are joined by a bond.

$\mathcal{H}_0$ is a simple tight-binding Hamiltonian on the icosahedron; the spectrum is given in Table I. While $\mathcal{H}_1$ and $\mathcal{H}_2$ also describe particle hopping among vertices of the icosahedron, these are complicated by the presence of the complex phases (3). That these cannot be eliminated by suitably rotating the 24 basis functions $u_m^\alpha$ about their fivefold axis is demonstrated by the fact that

TABLE I. The spectra of $\mathcal{H}_0$, $\mathcal{H}_1$, and $\mathcal{H}_2$ in units of $k\beta$.

| $\mathcal{H}_0$ | | $\mathcal{H}_1$ | | $\mathcal{H}_2$ | |
|---|---|---|---|---|---|
| $T_{2u}$ | $1/\sqrt{5}$ | $T_{1g}$ | $1$ | $T_{2g}$ | $1$ |
| $H_g$ | $1/5$ | $T_{1u}+H_u$ | $1/\sqrt{5}$ | $G_u$ | $2/\sqrt{5}$ |
| $T_{1u}$ | $-1/\sqrt{5}$ | $G_g$ | $0$ | $G_g$ | $0$ |
| $A_g$ | $-1$ | $H_g$ | $-3/5$ | $T_{2u}+H_u$ | $-1/\sqrt{5}$ |
| | | $G_u$ | $-2/\sqrt{5}$ | $H_g$ | $-3/5$ |

the product of phases taken around a closed path need not be trivial. For example, consider the subspace of 12 basis functions having $m = +1$ where the sign is defined consistently with respect to the outward normal of each 5MR. For three such mutually adjacent states forming a triangle on the icosahedron, the product of phases on a closed path that encloses the triangle once is $\exp(\pm i 4\pi/5)$ (the sign depending on the sense of the path). This situation may be described without recourse to a specific choice of basis function ("gauge") with the statement that the $m = +1$ sector of $\mathcal{H}_1$ corresponds to the usual London theory applied to an icosahedron with a magnetic monopole at its center. Since the spherical area enclosed by the triangular path (above) is $4\pi/20$, the appropriate magnetic charges are (in units $\hbar c/e$) $g = \pm(4\pi/5)(20/4\pi) = \pm 4$ for $m = \pm 1$, and $g = \pm 2$ for $m = \pm 2$. In the actual Hamiltonian $\mathcal{H}_1$ ($\mathcal{H}_2$) the basis states with $m = +1(2)$ have nonvanishing matrix elements with the $m = -1(2)$ states, so that this monopole description is incomplete (i.e., the particle charge $m$ may change sign). The spectra of $\mathcal{H}_1$ and $\mathcal{H}_2$ are given in Table I; the degeneracy of their ground states is characteristic of the "frustration" induced by the monopoles.

When the combined spectra of $\mathcal{H}_0$, $\mathcal{H}_1$, and $\mathcal{H}_2$ are juxtaposed with the spectrum of $C_{60}$ ($k = 1$), it is clear that the $\pi$-molecular orbitals of $C_{60}$ may be separated according to their (average) value of $|m|$ (see Fig. 2). Al-

though we have not explicitly verified this, it is reasonably likely that there are no level crossings for $0 < k \le 1$. Notable, however, is the relatively rapid convergence of the $T_{1u}, H_u$ and $T_{2u}, H_u$ levels of the $|m| = 1$ and $|m| = 2$ sectors, respectively. These are accidentally degenerate at the level of the reduced Hamiltonians $\mathcal{H}_1$ and $\mathcal{H}_2$, so that their splitting is due to $|m|$ changing elements of the $C_{60}$ Hamiltonian. Consequently, the magnitude of these splittings varies as $k^2$ for $k \to 0$. Significant in this discussion is the splitting of the $T_{1u}$ and $H_u$ levels for $|m| = 1$, since this is also the energy gap $\Delta E$ of $C_{60}$. Numerically, we find $\Delta E \sim 0.937 k^2$ as $k \to 0$. In the next section we relate the vanishing of this gap to the paramagnetic divergence of $C_{60}$ in the $k \to 0$ limit.

## IV. MAGNETIC BEHAVIOR FOR SMALL $k$

In the basis of orbitals $u_m^\alpha$, a uniform magnetic field $\mathbf{H}$ gives rise to the diagonal perturbation

$$(\mathcal{H}_{mag})_{\alpha m, \alpha m} = 2\sin(2\pi m/5)(A/5)\mathbf{e}_\alpha \cdot \mathbf{H},$$

where $\mathbf{e}_\alpha$ is the outward normal to the 5MR $\alpha$ and $A/5 = 0.344$ is one-fifth the area enclosed by a 5MR. As in the usual atomic orbital basis there are also off-diagonal elements involving $\mathbf{H}$. However, these do not contribute to the linear (Zeeman) splitting of levels being
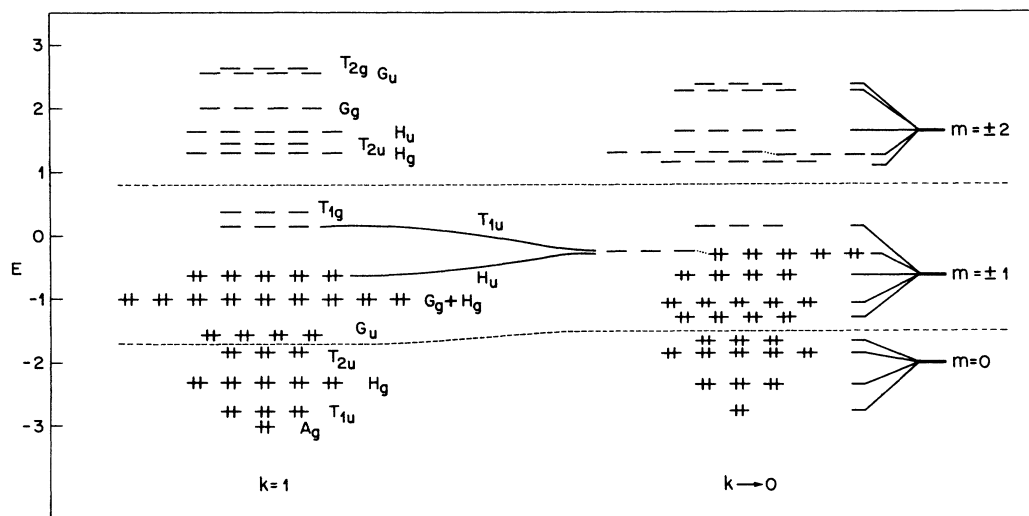


FIG. 2. Energy-level diagram of $C_{60}$ for $k = 1$ and the limit $k \to 0$. In this range there is apparently no level crossing and the orbitals are easily identified with the orbitals of definite $|m|$ at $k = 0$. The diagram on the right shows the splittings linear in $k$ (drawn to scale for $k = \frac{3}{4}$). To linear order in $k$, the $T_{1u}$ and $H_u$ levels of the $|m| = 1$ sector are accidentally degenerate. The rapid convergence of these levels as a function of $k$ leads to the paramagnetic divergence in $C_{60}$.

considered here. The linear energy change with $H = |\mathbf{H}|$ for an $n$-fold degenerate subspace of orbitals $j$ is found by diagonalizing the $n \times n$ matrix $\langle j' | \mathcal{H}_{\mathrm{mag}} | j \rangle$. For the $H_u$ and $T_{1u}$ levels on opposite sides of the energy gap, the corresponding magnetic eigenvalues (for $k \to 0$) are, respectively, $0, \pm h/6, \pm h/3$ and $0, \pm h/2$, where $h = 2\sin(2\pi/5)(A/5)H$. As expect-

ed, the net linear magnetic response of the lower-energy, fully occupied $H_u$ levels vanishes. This situation changes dramatically when the $H_u$ and $T_{1u}$ levels become degenerate at $k = 0$. In the basis of orbitals which diagonalize $\mathcal{H}_{\mathrm{mag}}$ in the $H_u$ and $T_{1u}$ subspaces, the total Hamiltonian for the eight-dimensional space has the form[14] (neglecting an additive constant)

$$
\begin{bmatrix}
-h/3 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -h/6 & 0 & 0 & 0 & \sqrt{3/20}h & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & h/\sqrt{5} & 0 \\
0 & 0 & 0 & h/6 & 0 & 0 & 0 & \sqrt{3/20}h \\
0 & 0 & 0 & h/3 & 0 & 0 & 0 & 0 \\
0 & \sqrt{3/20}h & 0 & 0 & 0 & \Delta E - h/2 & 0 & 0 \\
0 & 0 & h/\sqrt{5} & 0 & 0 & 0 & \Delta E & 0 \\
0 & 0 & 0 & \sqrt{3/20}h & 0 & 0 & 0 & \Delta E + h/2
\end{bmatrix}
. \tag{4}
$$

The presence of off-diagonal elements indicates a change in the pattern of Zeeman splittings when $\Delta E = 0$. The eigenvalues in that case are $\pm h/3$, $\pm h/\sqrt{5}$, $\frac{1}{3}(\pm 1 \pm 4/\sqrt{10})h$, and lead to a linear energy change with $H$ for the five lowest levels, $\delta E_{\mathrm{mag}} \sim 2.01H$. In physical units this translates into an effective orbital moment of 1.2 Bohr magnetons (when both spin states are included and $\beta = 2.4$ eV). In the limit $\Delta E \to 0$ this linear behavior emerges from a paramagnetic divergence of the susceptibility analogous to the example given in Sec. I. On summing the five lowest eigenvalues of (4) for small $h$, one obtains $\delta E_{\mathrm{mag}} \sim -\frac{1}{2}(h^2/\Delta E)$. Substituting the definition of $h$ and taking into account the two spin states, the susceptibility takes the form $\chi_\pi \sim 0.857/\Delta E \sim 0.915 k^{-2}$. This behavior, as well as the linear energy change with $H$ for $H \gg \Delta E$ is also seen by direct diagonalization of the full $C_{60}$ Hamiltonian for decreasing values of $k$ (see Fig. 3).

Figure 4 shows the behavior of $\chi_\pi$ and $\Delta E$ as a function of $k$ for $k$ considerably outside the asymptotic region $k \to 0$. From the smooth, monotonic rise in $\chi_\pi$ with accompanying decrease in $\Delta E$ for $k \lesssim 1$ one would conclude that the influence of the paramagnetic singularity at $k = 0$ extends well into the region of physical interest. That the change of sign in $\chi_\pi$ occurs at a value of $k$ very close to 1 should probably be regarded as an accident.

## V. CONCLUSION

Useful information concerning the energy levels and magnetic response of icosahedral $C_{60}$ can be gained by considering the effect of varying the two inequivalent bonds of the molecule (even to the point of decomposing the structure into 12 weakly coupled 5MR's). In particular, the small ring current magnetic susceptibility can be understood as arising from a near cancellation between the usual diagmagnetism one expects in aromatic hydrocarbons with a paramagnetic contribution that

diverges as a function of this bond variation. The mechanism of this divergence involves the vanishing of the energy gap and formation of an orbital magnetic moment at the singular point.
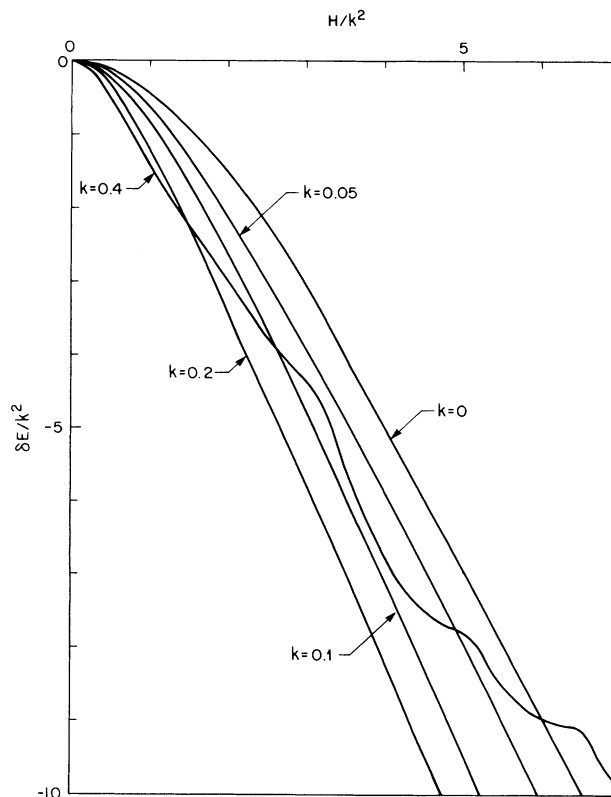


FIG. 3. Magnetic behavior of $C_{60}$ in the limit of small $k$. $H$ and $\delta E = E(H) - E(0)$ are given in dimensionless units (defined in the text). The approach to the quadratic behavior (near the origin) of the $k = 0$ curve implies a divergent susceptibility since the two axes are rescaled by $k^2$. Linear behavior for larger $H$ indicates the presence of a magnetic moment.

## APPENDIX: APPLICATION OF THE LONDON THEORY TO ICOSAHEDRAL $C_{60}$

Although London[10] originally derived his treatment for planar $\pi$ systems where $\sigma$-$\pi$ separability is clear, recent work[9,15] has shown that the distinction (orthogonality) between $\sigma$ and $\pi$ electrons persists in many nonplanar conjugated organic molecules, including $C_{60}$. The magnetic vector potential seen by the $\pi$ electrons in $C_{60}$ may be written as

$$\mathbf{A}(\mathbf{r}) = \tfrac{1}{2}H\hat{\mathbf{z}} \times \mathbf{r} \pm \mu \frac{\hat{\mathbf{z}} \times \mathbf{r}}{|\mathbf{r}|^3} \ .$$

The first term represents a uniform magnetic field in the direction of the unit vector $\hat{\mathbf{z}}$, while the second term describes a nuclear spin (quantized along $\hat{\mathbf{z}}$) with moment $\mu$ located at the center of the $C_{60}$ cage. Since the excursions of the $\pi$ electrons away from a spherical shell are small, we may assume $|\mathbf{r}|$ is constant. Consequently, the presence of the nuclear spin is merely to replace $H$ by

$$H_{\text{eff}} = H \pm \frac{2\mu}{|\mathbf{r}|^3} \ .$$

As originally derived by London,[10] the Hamiltonian matrix elements in the atomic $\pi$-orbital basis are given by

$$\mathcal{H}_{rs} = -\beta_{rs} e^{i\theta_{rs}} \ , \tag{5}$$

where the phase angles,

$$\theta_{rs} = \tfrac{1}{2}(x_r y_s - x_s y_r)\frac{e}{\hbar c}H_{\text{eff}} \ ,$$

are now expressed explicitly in terms of the atomic coordinates $(x_r, y_r, z_r)$. At this point, rather than treating (5) perturbatively for small $\theta_{rs}$, we simply calculate its eigenvalues numerically for various values of $H_{\text{eff}}$. [This avoids the gauge-dependent separation (1).]

The $\pi$-electron energy is given by the sum of the 30 lowest eigenvalues of (5) and behaves as

$$\delta E = E(H_{\text{eff}}) - E(0) = -\tfrac{1}{2}\mathcal{X}_\pi H_{\text{eff}}^2$$

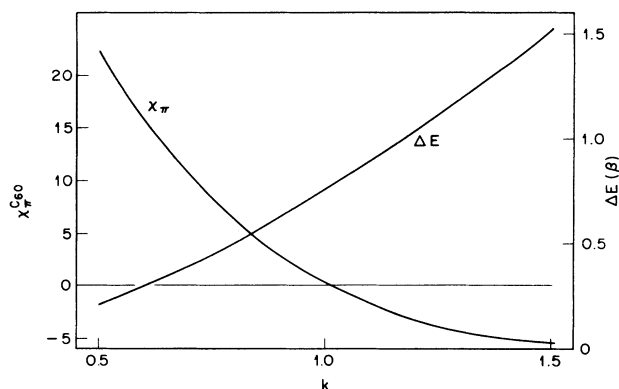for small $H_{\text{eff}}$. Numerically, the susceptibility $\mathcal{X}_\pi$ can be



FIG. 4. Susceptibility and energy gap of $C_{60}$ as functions of $k$. Although the range of $k$ is considerably outside the asymptotic region $k \rightarrow 0$, the divergent behavior is already noticeable. (In our units $\mathcal{X}_\pi$ for benzene is $-1.5$.)

extracted for values of the dimensionless magnetic field $H_{\text{eff}}(ea^2/\hbar c)$ as large as $10^{-3}$. With double precision arithmetic on a computer, accurate values of $\mathcal{X}_\pi$ were obtained by evaluating $-2\delta E/H_{\text{eff}}^2$ for smaller and smaller values of $H_{\text{eff}}$. The method was tested on simple aromatics, such as benzene, with satisfactory results. In retrospect, we find it surprising that this simple-minded approach has not been used before.

The NMR chemical shift for the central atom is found by calculating the change in total magnetic energy when the nuclear moment changes from $+\mu$ to $-\mu$. (We neglect the interference from the NMR ion's own atomic orbitals.) Thus

$$E_{\text{tot}}(+\mu) = -\tfrac{1}{2}\mathcal{X}_\pi \left[ H + \frac{2\mu}{|\mathbf{r}|^3} \right]^2 - \mu H \ ,$$

$$E_{\text{tot}}(-\mu) = -\tfrac{1}{2}\mathcal{X}_\pi \left[ H - \frac{2\mu}{|\mathbf{r}|^3} \right]^2 + \mu H \ ,$$

and

$$E_{\text{tot}}(+\mu) - E_{\text{tot}}(-\mu) \cong -2\mu H \left[ 1 + \frac{2\mathcal{X}_\pi}{|\mathbf{r}|^3} \right] \ ,$$

giving a chemical shift, due to ring currents,

$$\delta_{\text{RC}} = \frac{2\mathcal{X}_\pi}{|\mathbf{r}|^3} \ .$$

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