‘C_{60} spin-charging’ with an eye on a quantum computer

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Abstract
A question whether there exists an interaction between the spins of the endohedral atom A@C_{60} and the properties of the confining shell which might affect the alignment of, or manipulation by, the spins for building a register for a quantum computer is discussed. It is argued that an effect, termed the ‘C_{60} spin-charging’ effect, can occur in endohedral atoms and would affect the operation of a quantum register. The effect is exemplified by choosing the 3d (Cr and Mn) and 4d (Mo and Tc) transition metal atoms as well as a rare-earth Eu atom as the case study. A class of high-spin atoms which are less suitable for building a quantum register is, thus, identified.

Keywords: endohedral fullerenes, spin-polarized Hartree–Fock approximation, square well model of C_{60}, register for a quantum computer

The use of a non-zero spin atom confined by C_{60} (referred to as the endohedral A@C_{60} atom) as the building block of the register for a quantum computer was proposed by Harneit [1]. Obviously, the higher the spin of the atom, the better. Confined atoms then must be atoms with one or more multielectron semi-filled subshells in their configuration whose electron spins are aligned. The study by Harneit [1] focused on the use of a semi-filled shell N(2p^3)@C_{60} atom. The general idea for building the register for a quantum computer depends on the freedom to align the spin of the encapsulated atom, on the ability of the C_{60} confining cage to screen the spins from the influence of unwanted decohering fields and on the ability to write (read) to/from an assembly of confined atoms held together as an array.

It is, therefore, interesting to explore whether, in fact, the freedom to align the spins of encapsulated atoms exists independently of the properties of a confining shell and whether external fields are able to perturb this alignment. The latter question has already been addressed theoretically by Connerade and Solovyov [2] and Amusia and Baltenkov [3] who studied the properties of a spherical C_{60} cage and showed under what conditions the C_{60} screening of an external field remains effective. The former of the two questions is addressed in the present paper by accounting for an effect termed the ‘C_{60} spin-charging effect’.

The C_{60} spin-charging effect was recently uncovered as a by-product by Dolmatov et al [4] in the study of e^- + A@C_{60} electron elastic scattering. The quintessence of the effect is that both the valence electron spin-up P_{\ell\uparrow}(r) and spin-down P_{\ell\downarrow}(r) functions of a high spin encapsulated atom A, such as an atom with one or more multielectron semi-filled subshells in its configuration, may be drawn noticeably, but very differently, into the region of the C_{60} wall. The valence n\ell\uparrow- and n\ell\downarrow-orbitals are drawn into the wall of the C_{60} cage because the C_{60} cage is attractive. Furthermore, they are drawn differently because they originally differ from each other in a free atom in view of a different impact of exchange interaction on spin-up and spin-down electrons in semi-filled shell atoms, as is detailed below in the paper. This results in loading the C_{60} cage with electron density of a preferred spin orientation, i.e., in a spin-dependent drain of the electron density from the atom to the cage. Naturally, the effect is accompanied by the loss of some electron spin-density localized on the confined atom A itself. Clearly, the phenomenon is potentially important for the proposed realization of an A@C_{60} register for a quantum computer. It is the ultimate aim of the present paper to delineate the spin-charging effect more precisely for this purpose. To meet this goal, the 3d-, 4d- and 4f-semi-filled shell Cr(…3d^4s^1, ^7S), Mn(…3d^4s^2, ^5S), Mo
Encapsulated inside C60 are chosen for the completeness of the case study.

In the following, we briefly outline the methodology to calculate the C60 spin-charging effect in an endohedral semifilled shell atom, A@C60.

A convenient way to account for the structure of a semifilled shell atom is provided by the spin-polarized Hartree–Fock approximation (SPHF) developed by Slater [6]. SPHF accounts for the fact that spins of all electrons in a semifilled subshell of the atom (e.g., in the 3d5 subshell of Mn) are co-directed, in accordance with Hund’s rule, say, all pointing upward. This results in splitting of a closed nℓ2(2ℓ+1) subshell in the atom into two semifilled subshells of opposite spin orientations, nℓ2+1↑ and nℓ2+1↓. This is in view of the presence of exchange interaction between nℓ↑ electrons with only spin-up electrons in the original semifilled subshell of the atom (like the 3d↑ subshell in the Mn atom) but absence of such for nℓ↓ electrons. Thus, the SPHF configurations of Cr, Mn, Mo, Tc, and Eu are as follows: Cr(...3p13p↑13d↑ 4s↑, ↑), Mn(...3p↑3p↑3d↑4s↑4p↑, ↓), Mo(...4p↑4p↑4d↑5s↑, ↓), Tc(...4p↑4p↑4d↑5s↑5p↑, ↓) and Eu(...4d↑4d↑4f↑4↑6s↑6s↑6d↑6s↑, ↓). SPHF equations for the ground state of a semifilled shell atom differ from ordinary HF equations for closed shell atoms by accounting for exchange interaction only between electrons with the same spin orientation (↑, ↑ or ↓, ↓) [6, 7]. To model a A@C60 atom, we account for the presence of the C60 cage by adding a rectangular (in the radial coordinate r) potential well U_{C60}(r) of a finite width Δ, depth U₀ and inner radius r₀ to the HF (SPHF) equations [5], as in many of other studies, see, e.g., [4, 8–11] and references therein:

\[ U_{C60}(r) = \begin{cases} -U_0, & \text{if } n_0 \leq r \leq n_0 + \Delta, \\ 0, & \text{otherwise}. \end{cases} \] (1)

In the literature, some inconsistency is present in choosing the magnitudes of Δ, U₀ and r₀ of the C60 phenomenological potential (1), cp., e.g., [4, 5, 8–11] with each other. In the present paper, following [10], we choose Δ = 2.9102 au (which is twice the radius of the carbon, n₀ = 5.262 au = R_e – 1/2Δ (R_e = 6.7173 au being the radius of the C60 skeleton) and U₀ = 7.0725 eV (which was found by matching the electron affinity EA = −2.65 eV of C60 with the assumption that the orbital momentum of the 2.65 eV-state is ℓ = 1) [10]. This choice is most consistent with observations. Calculated P_{n↓}(r) and P_{n↑}(r) functions of valence spin-up and spin-down electrons of the Cr, Mn, Mo, Tc and Eu atoms, both free and encapsulated inside C60, are depicted in figure 1.

Note how the encapsulation of the chosen atoms inside the C60 cage draws their outer P_{n↓}(r) and, respectively, P_{n↑}(r) orbital functions into the region of the C60 wall. This implies a significant transfer of electron density from the encapsulated atom to the cage, but, more importantly in the context of the present paper, a transfer of electron spin-density from the atom to the cage. The transfer makes the cage become ‘spin-charged’. The C60 cage becomes spin-charged even for the spin-neutral 4f2 and 5s2 subshells of endohedral Mn and Tc, respectively. This is because of the stronger drain of the valence ns↓ than ns↑ electron density from the atom to the cage. Interestingly enough, the spin-dependent drain of the valence electron density does not emerge in Eu@C60 where both the P_{n↓}(r) and P_{n↑}(r) orbital functions are drawn into the C60 cage equally strongly, in contrast to the outer P_{n↓}(r) and P_{n↑}(r) orbital functions of the endohedral Mn and Tc atoms. This is because the semifilled 4f↑ subshell of Eu lies much deeper relative to its 6s↑ and 6s↓ subshells than the semifilled nd↑ subshell of Mn (n = 3) or Tc (n = 4) relative their valence (n + 1)s↑ and (n + 1)s↓ subshells. For this reason, the exchange interaction between the 4f↑ and 6s↑ electrons in Eu is negligibly small. Hence, there is practically no difference between the P_{n↓}(r) and P_{n↑}(r) functions both in free and encapsulated Eu. As a result, the Eu atom retains its electron spin-density intact upon confinement inside C60—which could prove important for an eventual application.

Note, relativistic effects cannot alter results of our predictions, because (a) they are negligible for valence ns-electrons (ℓ = 0) in the atoms under discussion and (b) cannot cause or affect the spin-splitting of a valence ns2-subshell in view of the absence of spin–orbit interaction for ns-states.

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Figure 1. Calculated P_{n↑}(r) and P_{n↓}(r) radial functions (in atomic units, au) of the valence subshells of the Cr@C60, Mn@C60, Mo@C60, Tc@C60 and Eu@C60 atoms versus those of the free atoms, as marked. Vertical dotted lines locate the position of the C60 wall, 5.262 ≤ r ≤ 8.17 au.
In conclusion, the authors believe that the C\textsubscript{60} spin-charging effect we have described will affect the manipulation of spins in the corresponding A@C\textsubscript{60} systems and that it must inhibit, or at least render more complex, the operation of a quantum register. The present paper thus brings to light a class of high-spin atoms which are less suitable for building a quantum register, namely those which are subject to a strong electron spin-density drain from the atom to the C\textsubscript{60} cage.

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