Spin-polarized electrons upon nondipole photodetachment of fullerene anions

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Synopsis Spin-polarization of electrons ejected from fullerene anions C_N^- in the geometry where only nondipole photodetachment effects matter is studied at photon energies of only few tens of eV. The degree of electron spin-polarization is found to be on the order of 5 to 10%, at certain energies. This is remarkable, given that this happens at low photon energies.

To the best of the authors' knowledge, little, if any, is known about spin-polarized photoelectron fluxes from fullerene anions C_N^- , in general, not to mention the impact of a nondipole part of the electron-photon interaction on spin-polarization of outgoing photoelectrons, in particular. C_N^- s possess remarkable qualities relating to their huge polarizability, large sizes (they are, in essence, giant atoms with a loosely bound outer electron) and large empty inner spaces in their structures. This makes it a matter of significant interest to gain insight into the impact of these features on spin-polarization of photoelectrons from C_N^- s. In the present work, we focus on the study of spin-polarization of electrons ejected from $C_{\scriptscriptstyle N}^-s$ in the geometry where only nondipole photodetachment effects matter.

To meet the goal, we utilize Cherepkov's theory [1]. Following [1], we consider two photoionization geometries where only electric dipole(*E*1)quadrupole(*E*2) interference [2] matters. One geometry, resulting in *longitudinal* spin-polarization of photoelectrons, is when the photon momentum **k**, photoelectron momentum **p** and spin **s** are such that $\mathbf{p} \perp \mathbf{k}$ and $\mathbf{s} \parallel \mathbf{p}$. The degree of longitudinal spinpolarization P_{lon}^{\pm} from a single-electron *n*p subshell is given by $P_{\text{lon}}^{\pm} = \mp \frac{2\delta}{4+\beta}$ [1]. Here, the signs \pm stand for the left/right circularly polarized light, β is the dipole photoelectron angular-asymmetry parameter and δ is a *E*1-*E*2 nondipole angular-asymmetry parameter [2]. In the other geometry, resulting in *transverse* spin-polarization P_{tr} , $\mathbf{p} \perp \mathbf{k}$, $\mathbf{s} \perp \mathbf{k}$ and $\mathbf{p} \perp \mathbf{s}$ [1].

To address photodetachment of a fullerene anion C_N^- , we combine Cherepkov's theory with modelling of a C_N cage by a spherical attractive potential $U_0(r)$ of certain depth U_0 , inner radius r_0 and thickness Δ [3] (and references therein). A fullerene anion C_N^- is, then, due to the binding of an external electron by this potential into a $n\ell$ -state. To account for the impact of the polarization potential V_{pol} of a fullerene-remainder on the motion of a released electron, V_{pol} is approximated by a static dipole polarization potential: $V_{\text{pol}}(r) \approx -\alpha/[2(r^2 + b^2)^2]$ [4]. Here, α is the static polarizability of C_N and *b* is a parameter of the order of r_0 . Thus, a released from C_N^- electron moves in the field of an effective potential $U_{\text{eff}}(r) = U_{\text{c}}(r) + V_{\text{pol}}(r)$. Finally, we consider C_N^- s with progressively increasing sizes (N = 60, 240 and 540), assume that the attached electron is a 2p-electron and calculate both P_{lon} and P_{tr} .

For illustration, calculated degree of longitudinal spin-polarization P_{lon} of the photoelectron flux from $C_{60}^{-}(2p)$ is depicted in Fig.1. The details and strength of the impact of both nondipole effects and fullerene polarizability on P_{lon} are evident.



Figure 1. The degree of longitudinal spin-polarization P_{lon} of the photoelectron flux from $C_{60}^{-}(2p)$, due to *E*1-*E*2 interference upon photodetachment by light of the left circular polarization, calculated with (solid line) and without (dashed line) account for fullerene polarizability α . Parameters: $\alpha \approx 850$, $U_0 \approx -0.26$, $r_0 \approx 5.26$ and $\Delta \approx 2.91$ a.u. [4].

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