

Correlation effects in ($e, 2e$) process on C_{60}

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In this work we investigate the role of electronic correlations on the electron-impact induced ionization spectrum of free fullerene clusters. The unperturbed valence states of the clusters are described within the Hartree-Fock scheme based on a jellium shell model. To account for the many-body response of the cluster to an external perturbation we utilize the random phase approximation with exchange in order to describe the interaction of the incoming projectile with the metallic cluster which introduced the effect of non-local screening of the inter-electronic interaction. Within this framework we evaluate numerically the ionization transition amplitudes and compare the results with available experimental data for a C_{60} cluster. We point out that the neglect of the collective response of the cluster leads to results at variance with experimental finding.

In this work we study the single ionization of the fullerene cluster from its (single-particle)ground state $|\varphi_{2j}\rangle$ upon the impact of an electron with wave vector \mathbf{k}_0 . In the final channel two electrons recede from the residual cluster to emerge with asymptotic momenta \mathbf{k}_1 and \mathbf{k}_2 . The transition amplitude for such a reaction is given by

$$T(\mathbf{k}_0, \mathbf{k}_1, \mathbf{k}_2, \varphi_\nu) = E_0 \langle \mathbf{k}_1 \mathbf{k}_2 | V_{12} \Pi | \varphi_{2j} \mathbf{k}_0 \rangle, \quad (1)$$

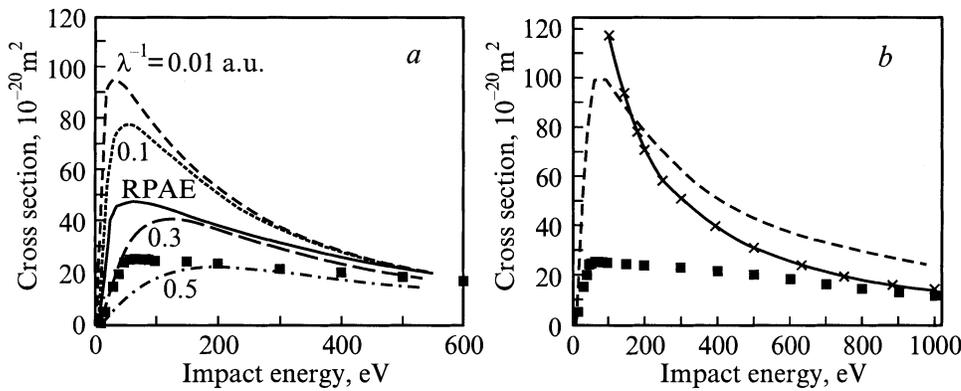
E_0 is the total kinetic energy of the two electrons and $\Pi = G_0 + G_0 V \Pi$ is the total Green operator of the projectile-cluster system with the total potential V . The interaction between the projectile and the knocked-out electron is designated by V_{12} . Here we report on the calculation of the first-order term of Eq. (1) as well as of the next terms due to the electron-hole excitations. In the presence of the external electron with momentum \mathbf{k}_0 the self-consistent cluster potential changes. Taking into account the polarization of the electronic cloud, we write the amplitude of the process as a sum of two terms. The "direct" one corresponds to the excitation of the cluster's electron labeled by index 2 from the j^{th} bound state φ_{2j} to the continuum state with the asymptotic momentum \mathbf{k}_2 .

The second "correlation" term describes correction, which appears from electron-hole excitations. Thus, in the random phase approximation with exchange (RPAE) the matrix element reads

$$T \sim \langle \mathbf{k}_1 \mathbf{k}_2 | V_{\text{eff}} | \varphi_{2j} \mathbf{k}_0 \rangle = \langle \mathbf{k}_1 \mathbf{k}_2 | V_{12} | \varphi_{2j} \mathbf{k}_0 \rangle + \sum_{\varepsilon_\alpha \leq \varepsilon_F < \varepsilon_\beta} \left(\frac{\langle \alpha \mathbf{k}_2 | V_{\text{eff}} | \varphi_{2j} \beta \rangle \langle \beta \mathbf{k}_1 | \frac{1}{r_{13}} | \mathbf{k}_0 \alpha \rangle}{\varepsilon_0 - (\varepsilon_\alpha - \varepsilon_\beta - i\delta)} - \frac{\langle \beta \mathbf{k}_2 | V_{\text{eff}} | \varphi_{2j} \alpha \rangle \langle \alpha \mathbf{k}_1 | \frac{1}{r_{13}} | \mathbf{k}_0 \beta \rangle}{\varepsilon_0 + (\varepsilon_\alpha - \varepsilon_\beta - i\delta)} \right), \quad (2)$$

V_{eff} being an effective interelectron interaction, $1/r_{13}$ — bare Coulomb between impact and intermediate electrons, α, β are the intermediate electron's and hole's states with the energies $\varepsilon_\alpha, \varepsilon_\beta, \varepsilon_0$ and ε_F are the initial energy of the projectile and the Fermi energy respectively. The different terms in the sum describe electron-hole excitation and deexcitation.

The differential electron-impact ionization cross-section is to be summed over ionization events from all the initial



Integrated electron impact ionization cross section of C_{60} as a function of the projectile energy: (a) present calculations in Tomas-Fermi model (dashed curves) with different values of the inverse screening length and in the random phase approximation with exchange (solid curve), symbols: experimental cross section for production of C_{60}^+ ions [4]; (b) solid line: calculation in the plane wave Born approximation for the jellium-shell model [5], dashed line: result of semi-empirical model calculation [6], symbols: experimental cross section for production of C_{60}^+ ions [4].

bound states φ_{2j} accessible within the energy conservation, i.e.

$$\frac{d^6\sigma}{d^3\mathbf{k}_1 d^3\mathbf{k}_2} = \frac{(2\pi)^4}{|k_0|} \sum_j |T(\mathbf{k}_0, \mathbf{k}_1, \mathbf{k}_2, \varphi_{2j})|^2 \times \delta(\varepsilon_0 - (\varepsilon_j^{\text{ion}} + k_1^2/2 + k_2^2/2)). \quad (3)$$

The quantum states of the fullerene cluster are constructed within the Hartree-Fock approximation and within the spherical jellium model. The potential of C_{60} , which is a superposition of atomic potentials, is replaced by a potential of a fullerene shell. The latter is formed by delocalized valence electrons of carbon atoms and modelled by a potential well: $V(r) = V_0$ within the region $R - \Delta < r < R + \Delta$, and $V = 0$ elsewhere. Here $R \approx 6.7a_0$ is the radius of the fullerene, the thickness of the shell is $2\Delta \approx 2a_0$, a_0 being the Bohr radius. The height of the well was chosen such that the experimental value of the electron affinity of C_{60} and the number of valence electrons are correctly reproduced.

Numerical calculation of the Hartree-Fock states is based on the non-local variable phase approach [1–3]. This method allows to find the eigen-functions and eigen-values through the scattering phase, which is built for the set of cut-off at different distances potentials, and through the poles of the scattering amplitude in the complex plane of the wavevector of the particle. Such a choice of numerical realization of the self-consistent procedure allows to accelerate its convergency significantly.

To get a qualitative insight into the role of screening for the shape of the cross section we compare our RPAE calculations with the calculations in intuitively transparent approximation: due to the presence of a large number of mobile electrons on the surface of the fullerene the interaction of the incident electron with the cluster electrons is screened. A simple model to account for this effect is the Thomas-Fermi approach which yields a local screening of the electron-electron interaction described by the one-parameter Yukawa potential: $V(r_{12}) = \exp(-\lambda r_{12}/r_{12})$, where λ is the inverse screening length. The results of our calculations are presented in Figure, *a*, where the total cross section of the process $\sigma(\varepsilon_0)$ is given as a function of the impact energy. Dashed curves correspond to different values of the inverse screening length, solid curve is the RPAE result.

Despite their conceptual simplicity, these models of electronic correlation yield encouraging results. We compare them with available experimental data [4] together with the theoretical calculation of work [5,6], where the bare Coulomb interaction was taken into account and the (e , $2e$) total cross section was calculated in the plane wave Born approximation (see Figure, *b*). When contrasted with calculations that neglect screening effect, the account of it results in a flattening of the total ionization cross-section at lower energies. This effect can be traced back to the fact that for the unscreened Coulomb interaction the main contribution to the cross sections originates from far collisions, whereas in case of screening there is a cut-off impact parameter beyond which the contribution of collisions to the cross-section diminishes.

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