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# Vacancy decay in endohedral atoms

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It is demonstrated that the fullerene shell dramatically affects the radiative and Auger vacancy decay of an endohedral atom $A@C_{60}$. The collectivized electrons of the $C_{60}$ shell add new possibilities for radiative and nonradiative decays similar to that in ordinary atoms where the vacancies in the initial and final state almost always belong to different subshells. It is shown that the smallness of the atomic shell radii as compared to that of the fullerene shell provides an opportunity to derive the simple formulas for the probabilities of the electron transitions. It is shown that the radiative and Auger (or Koster-Kronig) widths of the vacancy decay due to electron transition in the atom $A$ in $A@C_{60}$ acquire an additional factor that can be expressed via the polarizability of the $C_{60}$ at transition energy. It is demonstrated that due to an opening of the nonradiative decay channel for vacancies in subvalent subshells the decay probability increases by five to six orders of magnitude.

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## I. INTRODUCTION

It has already been more than 30 years since it was predicted that intrashell and intershell interactions in atoms could modify considerably the probability of the radiative and Auger decay of vacancies in atoms [1]. It was demonstrated that due to this interaction the decay probability could increase or decrease, up to complete disappearance of the decay photon or electron line. A new phenomenon was predicted, namely radiative self-locking of atomic shells, the essence of which is complete suppression of the radiative decay of some atomic vacancies due to interelectron interaction [1,2].

Since then, new multiatomic and hence multielectronic structures, such as metallic clusters and fullerenes, became available and started to be objects of intensive investigations. There the electron correlation effects in all processes, including vacancy decays, could be much stronger than those in isolated multielectron atoms.

Recently in a number of papers (see, e.g., Refs. [3–5] and references therein) it was demonstrated that in endohedral systems $A@C_{60}$, in which an atom $A$ is located inside the "empty" inner part of the $C_{60}$, the photoionization characteristics of the atom $A$ are impressively modified by the $C_{60}$ shell. In these papers the fullerene shell is considered as a source of some static spherically symmetrical potential playing the role of a resonator for the photoelectron wave. The electronic structure of this shell is not taken into consideration. This approach is parametrically justified for the deep subshells of the encapsulated $A$ atom with the ionization potential significantly exceeding the energy of electronic excitations of the fullerene shell itself. For such frequencies of ionizing radiation the fullerene is practically transparent for incident photons and the electronic structure of $C_{60}$ plays no role in the $A$ atom ionization. The situation becomes quite different when the photon energy is close to the characteristic excitation energy of electrons of the fullerene $C_{60}$ itself. A remarkable example here is the Xe 5*s* subshell photoionization in the molecule $Xe@C_{60}$. The ionization potential of this subshell is close to energy of the giant resonance of the fullerene electronic system. As shown in papers [6–8] this results in the radical modification of the 5*s* subshell photoionization cross section near the threshold.

The aim of this paper is to show that the $C_{60}$ fullerene electron shell drastically modifies also the decay probability of a vacancy in the atom $A$ in $A@C_{60}$. This modification is a result of a number of effects, including the opening of new decay channels of a vacancy and the profound modification of those channels that exist in the isolated atom $A$. Namely, the decay can proceed due to the opening of new channels responsible for the participation of $C_{60}$ electrons. This is similar to the situation in ordinary molecules [9] and clusters [10], where a vacancy in one atom can decay due to transition in another.

It is natural to distinguish decays of vacancies in endohedral atoms by the degree of participation of the $C_{60}$ electrons. Namely, the vacancy in the endohedral atom can be occupied by an outer electron either from the same atom or from the collectivized electrons of the $C_{60}$, forming what could be called *atomic* and *fullerene* decay, respectively. If atomic vacancy is deep enough, it can be closed by one of the noncollectivized inner electrons of carbon atoms that form the $C_{60}$, presenting *molecular* decay.

If the decay results in the emission of a photon, the $C_{60}$ shell can intercept it, and then emit a photon with the same energy. As a result, the vacancy's decay in the encapsulated $A$ atom occurs through two channels. The first one is the recombination radiation of a photon in the isolated $A$ atom. The second one is the two-step process running through the electron excitation of the fullerene shell. The amplitudes of these channels interfere similarly in isolated atoms [1] where the decay probability can be altered dramatically from strong enhancement until almost complete suppression. The same situation can happen in the Auger decay: the interaction between electrons, inner and outer, can be modified due to virtual excitations of $C_{60}$ electrons, thus substituting the pure Coulomb interelectron interaction $V$ by a transferred energy $\omega$ dependent effective one $\Gamma(\omega)$ (see, e.g., Ref. [11]). The difference between $V$ and $\Gamma(\omega)$ is determined by the so-called intrashell or intershell effects.

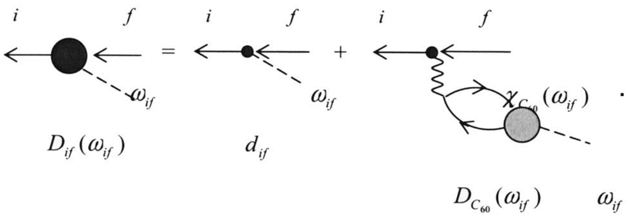
The physical nature of these so-called intershell effects in the vacancy decay processes has the following meaning. A many-electron atomic subshell is polarized by the emission of the decay photon. Therefore a time-dependent dipole moment is induced. Under the action of this dipole moment a neighboring atomic subshell is ionized. The inclusion of this effect in isolated atoms was performed in Ref. [1] in the frame of the so-called random-phase approximation exchange (RPAE) [11], which is very convenient to describe this effect.

The direct application of RPAE to $A@C_{60}$ is an extremely complex task. In fact this is a hard task even for isolated atoms, as it has already been demonstrated in Ref. [1]. However in $A@C_{60}$ we have an important simplifying factor: the radius of the $C_{60}$ shell significantly exceeds that of an encapsulated atom, i.e., the electronic shells of the $A$ atom and fullerene are spatially separated. This makes it possible to consider the electronic subsystems of the fullerene shell and atom as practically independent of each other, and consider the Coulomb interaction between electrons of these two subsystems as small. The situation is quite different in the case of atomic shells. Their electronic densities are significantly overlapped, so the Coulomb interaction between electrons is necessary to take into account in all orders of the perturbation theory, which results in the RPAE equations.

The great difference between atomic and fullerene radii, as shown below, permits deriving the formulas that are valid even beyond the RPAE frame, presenting expressions not for all but in principal for experimentally distinguishable decay channels of vacancies in the $A$ atoms encapsulated in $A@C_{60}$ via experimentally measurable dipole polarizability $\alpha_{C_{60}}(\omega)$ and radiative decay probability of isolated atom $A$. The present paper is devoted to deriving the formulas connecting the probabilities of radiative and nonradiative decays of the vacancies in the endohedral and free atoms. The method developed can be applied to other objects where many other atoms with collectivized electrons surround a given atom or an ion.

## II. ESSENTIAL FORMULAS—RADIATIVE DECAY

The amplitude of the radiative decay process can be presented by the following diagrams [1,11]:



(1)

Here the line with an arrow to the left represents a vacancy in the atom $A$ either in the initial $i$ or in the final state $f$. The dashed line represents a photon; the dark circle stands for the vacancy decay $i\rightarrow f$ amplitude $D_{if}\ (\omega_{if})$ and $\omega_{if}$ is the energy of the emitted photon. The vertically oriented wavy line denotes the potential of the Coulomb interaction between an electron in the atom $A$ and in $C_{60}$. The loop $\chi_{C_{60}}(\omega_{if})$ represents the electron excitation of $C_{60}$. The gray circle is the amplitude $D_{C_{60}}(\omega_{if})$ of the photon $\omega_{if}$ emission. The small dark dot represents the intra-atomic corrections to the amplitude of real or virtual photon emission.

The first of these diagrams corresponds to the dipole recombination photon emission from the isolated atom. The amplitude of this process is linear in the electric field of the light wave. The second diagram defines the process of the second order of the perturbation theory. Here we deal with two types of perturbation, namely in the electric field of the light wave and the usual Coulomb interaction of atomic electrons and electrons of $C_{60}$. The first of these perturbations is always small in the processes of recombination radiation. The second one, i.e., the interaction between the atomic and $C_{60}$ electrons, can also be considered as small because the radius $R$ of $C_{60}$ is much greater than the radius $\rho$ of the atom $A$ and the thickness $t_{C_{60}}$ of the fullerene $C_{60}$. Therefore, it is sufficient to take into account this Coulomb interaction in the first (lowest) order. So, the addition of each extra interaction wave line leads to a small factor $\rho/R\ll 1$, in fact to $(\rho/R)^3 \ll 1$, in the corresponding contribution to the amplitude and so it can be neglected. Note that due to the same reason we can neglect an exchange between atomic and $C_{60}$ electrons.

Representing the Coulomb interaction $V=|\mathbf{r}_a-\mathbf{r}_{C_{60}}|^{-1}$[1] between an atomic electron with a coordinate $\mathbf{r}_a$ and a fullerene shell electron with a coordinate $\mathbf{r}_{C_{60}}$ by the first term that leads to a nonvanishing contribution to $D_{if}(\omega_{if})$, one substitutes $V$ by

$$V\approx \mathbf{r}_a\cdot\mathbf{r}_{C_{60}}/R^3. \tag{2}$$

It is taken into account in Eq. (2) that $r_a\ll r_{C_{60}}\approx R$. The relation $r_{C_{60}}\approx R$ holds since the integration over $|\mathbf{r}_{C_{60}}|$ takes place within the thickness $t\geqslant\Delta r_{C_{60}}$ of the $C_{60}$ shell with $t \ll R$. The atomic electron wave functions are functions of $\mathbf{r}_a$, while the wave functions of $C_{60}$ electrons depend on $\mathbf{r}_{C_{60}}$ only. In this case, as it follows from Eq. (2), the Coulomb matrix element becomes a factorable function: $\langle i,j|V|f,k\rangle \approx\langle i|\mathbf{r}_a|f\rangle\langle j|\mathbf{r}_{C_{60}}|k\rangle/R^3$, i.e., it is transformed into the product of dipole matrix elements corresponding to the dipole excitations of the $A$ atom and $C_{60}$ shell. Here $(j,k)$ is an electron-vacancy pair that belongs to the $C_{60}$ shell. The amplitude of radiative decay $D_{if}\ (\omega_{if})$ in the operator form is defined by the following expression:

$$\hat{D}(\omega_{if})=\hat{d}+\hat{d}\hat{\chi}_{C_{60}}(\omega_{if})\hat{D}(\omega_{if})/R^3. \tag{3}$$

Here the "loop" $\hat{\chi}_{C_{60}}(\omega_{if})$ or the propagator of $C_{60}$ electron excitation, i.e., electron-vacancy pair creation, is given by the relation $\hat{\chi}_{C_{60}}=1/(\omega-\hat{H}_{ev})-1/(\omega+\hat{H}_{ev})$ [11] where $\hat{H}_{ev}$ is the electron-vacancy pair Hartree-Fock Hamiltonian of $C_{60}$.

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[1] The atomic system of units: $e=m=\hbar=1$ is used throughout this paper.

This loop corresponds to the summation and integration over all intermediate electronic states of the fullerene shell as it is usually done in the perturbation theory.

Then one has to use the definition of the dipole polarizability $\alpha(\omega)$ of a multielectron system (see, e.g., Ref. [11])

$$\alpha(\omega) \equiv -\left[2\sum_{evexit}\omega_{ev}D_{ev}(\omega)(\omega^2-\omega_{ev}^2)^{-1}d_{ev}\right]. \qquad (4)$$

In Eq. (4) we employ an alternative definition of $\alpha(\omega)$. Indeed, usually $\alpha(\omega)$ is defined as

$$\alpha(\omega) \equiv -\left[2\sum_{evexit,o}\omega_{ev}|D_{ev}(\omega_{ev})|^2(\omega^2-\omega_{ev}^2)^{-1}\right], \qquad (5)$$

but it can be easily demonstrated that this definition and that in Eq. (4) are identical [11]. Then we obtain the following relation for the radiative decay amplitude:

$$D_{if}(\omega_{if}) = d_{if}\left[1-\frac{\alpha_{C_{60}}(\omega_{if})}{R^3}\right], \qquad (6)$$

which leads to a remarkably simple formula for the radiative width

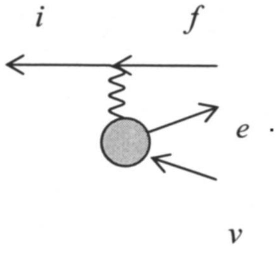
$$\Gamma^{C_{60}}_{\gamma,if} = \Gamma^{A}_{\gamma,if}\left|1-\frac{\alpha_{C_{60}}(\omega_{if})}{R^3}\right|^2. \qquad (7)$$

Note that a factor $G_{C_{60}}(\omega)=|1-\alpha_{C_{60}}(\omega)/R^3|^2$ was recently introduced in Refs. [6–8] while discussing the role of the $C_{60}$ electron shell upon photoionization of the endohedral atom $A@C_{60}$. A similar factor in pure classical derivation was obtained in Ref. [12].

## III. AUGER DECAY

In this section we will start with discussing the Auger decay that become possible in $A@C_{60}$ while it is energetically forbidden in the isolated atoms $A$. We will concentrate only on dipole transitions $i \rightarrow f$, because only for them the radiative decay channel in the $A$ atom is open.

The diagram that describes this process is depicted in the following way:



(8)

Here $(e,\nu)$ is the electron-vacancy pair that belongs to $C_{60}$. As in the previous section, we can consider the process in the lowest order in interaction between atomic and $C_{60}$ electrons, neglect exchange and use Eq. (2) for the interelectron interaction represented by the wavy line. The probability of the Auger decay is given by the square modulus of the Coulomb matrix element $\langle i,\nu|V|f,e\rangle \approx \langle i|\mathbf{r}_a|f\rangle\langle\nu|\mathbf{r}_{C_{60}}|e\rangle/R^3$ summed over all possible final states. Then one has for the Auger-decay width $\Gamma^{C_{60}}_{A,if}$ for the vacancy $i \rightarrow f$ transition in $A@C_{60}$:

$$\Gamma^{C_{60}}_{A,if} = r_{if}^2\frac{1}{R^6}\sum_e|\langle\nu|\mathbf{r}_{C_{60}}|e\rangle|^2. \qquad (9)$$

Here $r_{if}$ is the radial matrix element between states of the atom $A$—$i$ and $f$. The expression (9) can be conveniently transformed into a relation that connects only directly measurable characteristics of the isolated atom $A$ and $C_{60}$;

$$\Gamma^{C_{60}}_{A,if} = \Gamma^{A}_{\gamma,if}\frac{3}{8\pi}\left(\frac{c}{\omega_{if}}\right)^4\frac{\sigma_{C_{60}}(\omega_{if})}{R^6} = \Gamma^{A}_{\gamma,if}\frac{3}{2}\left(\frac{c}{\omega_{if}}\right)^3\frac{\mathrm{Im}\,\alpha_{C_{60}}(\omega_{if})}{R^6}, \qquad (10)$$

where $\sigma_{C_{60}}(\omega_{if})$ is the total photoabsorption cross section of $C_{60}$. If one is interested to have the Auger width that corresponds to one-electron ionization channel, $\sigma_{C_{60}}(\omega_{if})$ has to be substituted in Eq. (10) by $\sigma^{+}_{C_{60}}(\omega_{if})$—the one-electron photoionization cross section that can be found in Refs. [13–15].

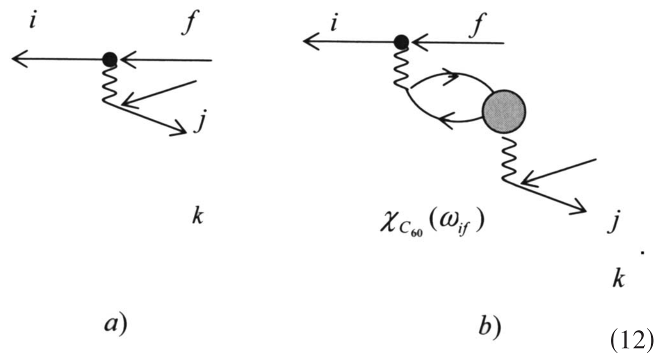
Note that if $i$ and $f$ vacancies belong to different subshells of the same shell, the transition presented by Eq. (8) is called the Koester-Kronig transition. Since $\mathrm{Im}\,\alpha_{C_{60}}(\omega)$ rapidly decreases with $\omega$ growth, $\Gamma^{C_{60}}_{A,if}$ is considerable for $\omega \leqslant 50$ eV, i.e., for Koester-Kronig transitions only.

An important characteristic of the vacancy decay is the so-called fluorescence yield $J_R$ that is the ratio of the radiative $\Gamma_{rad}$ and the total $\Gamma_{tot}=\Gamma_A+\Gamma_\gamma$ widths. Dividing Eq. (7) by Eq. (10) only, as in our case $\Gamma^{C_{60}}_{\gamma,if} \ll \Gamma^{C60}_{A,if}$, one has

$$J_R(\omega_{if}) = \frac{8\pi}{3}\left(\frac{\omega_{if}}{c}\right)^4\frac{R^6}{\sigma_{C_{60}}(\omega_{if})}\left|1-\frac{\alpha_{C_{60}}(\omega_{if})}{R^3}\right|^2 = \frac{2}{3}\left(\frac{\omega_{if}}{c}\right)^3\frac{R^6}{\mathrm{Im}\,\alpha_{C_{60}}(\omega_{if})}\left|1-\frac{\alpha_{C_{60}}(\omega_{if})}{R^3}\right|^2. \qquad (11)$$

If the $i \rightarrow f$ transition is not dipole, but monopole $l=0$ or quadrupole $l=2$, the Auger width is given by an expression similar to Eq. (9): $\Gamma^{C_{60}}_{Aif(l)} \sim |(r^2)_{if}|^2\,\mathrm{Im}\,\alpha^{l}_{C_{60}}(\omega)/R^{10}$. This width is smaller than the dipole decay width Eq. (9) because it contains an additional factor $\rho^2/R^2 \ll 1$. Since nothing is known about nondipole polarizabilities $\alpha^{l}_{C_{60}}(\omega)$ of the fullerene $C_{60}$, we cannot present numerical results for the widths of such transitions.

Let us turn now to the Auger decay of those vacancies not forbidden also in the isolated $A$ atom. In this case the process has two pathways—direct decay with emission of atomic outer electron and indirect one that leads to the same atomic electron emission via excitation of the $C_{60}$ electrons



(12)

Here $k$ and $j$ are the electron-vacancy excitations of the $A$ atom that are formed in Auger decay. The dark dot includes all virtual intra-atomic excitations.

In this paper we concentrate only on dipole transitions $i \to f$, since we know well the dipole excitation spectrum from experiment, namely the giant resonance. The two diagrams of Eq. (12) present two pathways that lead to the Auger decay. They can interfere leading to both enhancement and suppression of the decay probability. Obviously, the contribution of Eq. (12) is particularly important when $\omega_{if}$ is close to the giant resonance frequency. It means that important $C_{60}$ influence can be expected, as it was mentioned above while discussing Eq. (10), for Koester-Kronig (super Koester-Kronig) transitions, in which the vacancy $f$ belongs to the same shell as $i$.

Analytically, the amplitude $A_{i\to fkj}$ of Eq. (11) can be presented by using Eq. (2) in the following form:

$$A_{i\to fkj}=A^{A}_{i\to fk}-r^{A}_{if}\frac{\alpha_{C_{60}}(\omega_{if})}{R^{6}}r^{A}_{jk}, \tag{13}$$

where the upper index $A$ denotes the respective values that correspond to the isolated $A$ atom. The relation (13) leads to the following equation for the Auger width of $A@C_{60}$:

$$\Gamma^{C_{60}}_{A,i\to fkj}=\Gamma^{A}_{A,i\to fkj}\left|1-\frac{1}{2\pi}\sqrt{\frac{3}{2}}\frac{c^{2}}{\omega_{if}}\frac{\alpha_{C_{60}}(\omega_{if})}{R^{6}}\times\left(\frac{\Gamma^{A}_{\gamma,if}}{\Gamma^{A}_{A,i\to fkj}}\right)^{1/2}[\sigma^{A}_{j}(\varepsilon_{k})]^{1/2}\right|^{2}. \tag{14}$$

Here $\sigma^{A}_{j}(\varepsilon_{k})$ is the photoionization cross section of the atomic subshell $j$ with emission of a photoelectron with the energy $\varepsilon_{k}$. Note that the factor in brackets is by the order of magnitude close to one. This is seen from Eq. (13), where the first expression can be estimated as $A^{A}_{i\to fk}\sim r^{A}_{if}r^{A}_{jk}/\rho^{3}$ so that the second term in Eq. (13) gives a correction of the order $\rho^{3}\alpha_{C_{60}}(\omega_{if})/R^{6}\approx\rho^{3}/R^{3}\ll 1$.

## IV. DETERMINATION OF POLARIZABILITY

The dipole dynamical polarizability of $C_{60}$ defining, as shown above, the role of the fullerene shell in the processes of the vacancy decay in the encapsulated $A$ atom can be found from the experimental data [13–15] on photon absorption by the $C_{60}$ empty cage. Such an approach allows us to avoid considering the electronic structure of the fullerene shell. Any attempt to take into account this structure is inevitably accompanied by radical simplifications, the correctness of which is very difficult to estimate. The method to calculate $\alpha_{C_{60}}(\omega)$ from the experimental data was proposed in Ref. [16]. First, by using the relation $\mathrm{Im}\,\alpha_{C_{60}}(\omega)=c\sigma_{C_{60}}(\omega)/4\pi\omega$ between the photoabsorption cross section and the imaginary part of the polarizability, the $\mathrm{Im}\,\alpha_{C_{60}}(\omega)$ is calculated and then the $\mathrm{Re}\,\alpha_{C_{60}}(\omega)$ is found with the help of the dispersion relation.

The $\omega$ dependence of the photoabsorption cross section $\sigma_{C_{60}}(\omega)$ of $C_{60}$ exhibits a powerful maximum at the giant resonance energy that is of about 20 eV. This energy is close to the decay energy of a number of radiative and nonradiative atomic transitions. Therefore, the significant effect of electronic excitations of the $C_{60}$ shell on the probability of the captured atom decays can be expected. Although the experiments [13–15] provide no direct absolute values of $\sigma_{C_{60}}(\omega)$, it can be reliably estimated by using the different normalization procedures based on the sum rule $(c/2\pi^{2})\int\sigma_{C_{60}}(\omega)d\omega=N$. Here $N$ is the number of collectivized electrons. This method was used to find the polarizability of $C_{60}$ in Refs. [6,16], where $N$ was considered to be $N=240$, i.e., 4 electrons collectivized from each $C$ atom. Using the photoabsorption data considered in Ref. [14] as most reliable, we obtained $N_{eff}\approx 250$. This is sufficiently close to the value accepted in Refs. [6,16]. Note that since the one-electron photoionization cross section of the positive ion $C^{+}_{60}$ measured and calculated in Ref. [14] is much smaller than the cross section of the neutral fullerene it cannot be used to define the dynamical dipole polarizability of the neutral $C_{60}$. The dispersion relation

$$\mathrm{Re}\,\alpha_{C_{60}}(\omega)=\frac{c}{2\pi^{2}}\int_{I_{60}}^{\infty}\frac{\sigma_{C_{60}}(\omega')d\omega'}{\omega'^{2}-\omega^{2}} \tag{15}$$

connects the real part of the polarizability $\mathrm{Re}\,\alpha_{C_{60}}(\omega)$ with the photoionization cross section. In Eq. (15) $I_{60}$ is the $C_{60}$ ionization potential. The calculation results of the real and imaginary parts of the dynamic polarizability of the fullerene shell $C_{60}$ are given in Fig. 1. The experimental photoabsorption cross section for $C_{60}$ taken from Ref. [14] is presented in the inset of this figure. As seen from this figure, the cross section is small at the threshold (which also means relatively low intensity of discrete excitations) and is dominated by a huge giant resonance well above the threshold. Therefore, in Eq. (15) a small contribution of discrete excitations of the $C_{60}$ collectivized electrons are neglected.

The frequency dependence of the imaginary part, as it should be, is similar to the frequency dependence of $\sigma_{C_{60}}(\omega)$. A small peak in the cross section for photon energy ~5 eV is transformed into a significant maximum, which is explained by a small value of photon energy as compared to energy of the giant resonance ~22 eV. The real part of the polarizability obtained as a result of integration according to formula (15) behaves more systematically and with the raise of radiation frequency $\omega$, as it should be, decreases as $\mathrm{Re}\,\alpha_{C_{60}}(\omega)\sim -N_{eff}/\omega^{2}$. The calculation results of the dynamical polar-

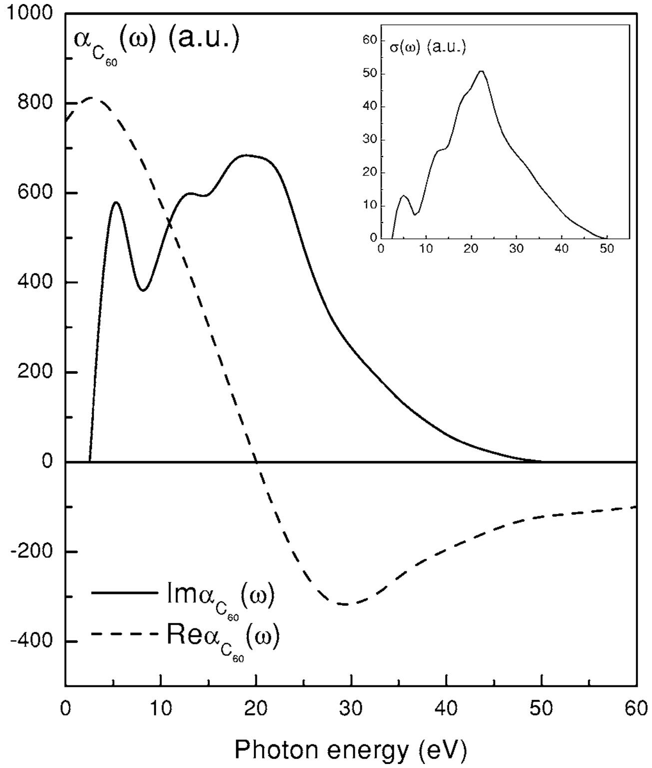


FIG. 1. Real and imaginary parts of the dynamical dipole polarizability of $C_{60}$.

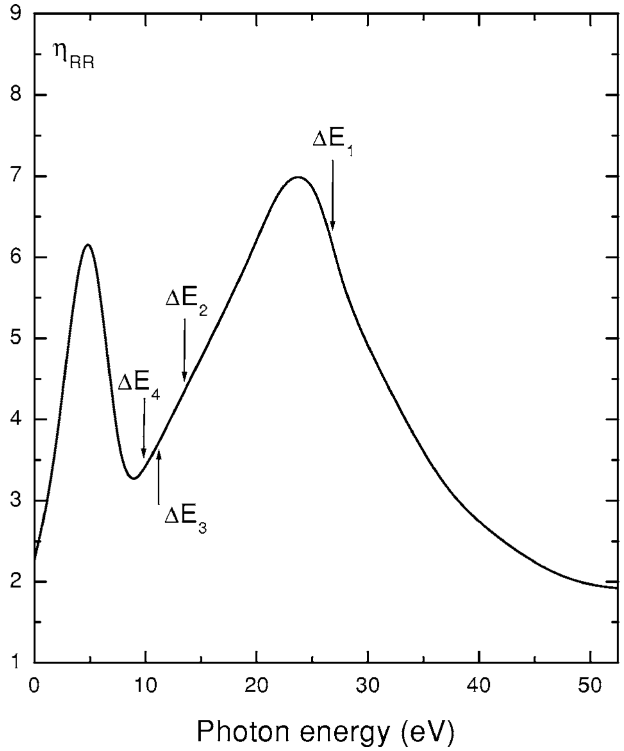


FIG. 2. The ratio $\eta_{RR}(\omega)$ of the radiative widths for the same transition in endohedral and free atom as a function of photon energy. $\Delta E_1$=26.85 eV corresponds to the $2s$-$2p$ transition in Ne; $\Delta E_2$=13.50 eV is the $3s$-$3p$ transition in Ar or $4s$-$4p$ in Kr; $\Delta E_3$=11.20 eV and $\Delta E_4$=9.90 eV are the $5s$-$5p_{3/2}$ and $5s$-$5p_{1/2}$ transitions in Xe, respectively. All transition energies are given in Ref. [18].

izability will be used further to calculate the ratio of widths of the radiative and nonradiative decays of vacancies in the endohedral atoms.

### V. ROLE OF AUGER-ELECTRON REFLECTION

Until now we considered the effect of the fullerene shell on the probabilities of the radiative and nonradiative transitions in the endohedral atoms. However, the $C_{60}$ shell can affect the outgoing Auger electrons from the $A$ atom in the same way as it affects photoelectrons [4,5,17]. This can be taken into account similarly to how it was done in these papers. In the Auger decay of a vacancy inside the atom $A$ of $A$@$C_{60}$ strong modification of its probability can come from reflection and refraction of the Auger-electron wave.

In order to take into account these processes for the Auger electron's wave, we use here, just as in Refs. [4,5,17] a $\delta$-bubble model that represents the static $C_{60}$ potential as $U(r)=-V_0\delta(r-R_c)$, with $V_0$ chosen in such a way as to reproduce the binding energy of the $C_{60}^-$ negative ion. In the frame of this model potential, its influence upon the photoionization amplitude is presented by a factor $F_A(\varepsilon)$, the oscillatory behavior of which is connected with the reflection of the Auger electron with energy $\varepsilon$ by the $C_{60}$ shell. The details of calculation of $F_A(\varepsilon)$ can be found in the abovementioned papers. In short, this function is expressed via the regular and irregular at $r=0$ photoelectron wave functions.

Entirely, the following relation gives the Auger-decay amplitude $A_{i\to fk}^{C_{60}}$ for an atom inside the $C_{60}$ shell:

$$A_{i\to fk}^{C_{60}} \approx F_A(\varepsilon_k)A_{i\to fk}. \tag{16}$$

Using this amplitude, one has for the Auger width the following relation:

$$\Gamma_{A,i\to fk}^{C_{60}} \approx |F_A(\varepsilon_k)|^2\Gamma_{A,i\to fk}^{A}. \tag{17}$$

The amplitude factor $|F_A(\varepsilon)|^2$ as a function of kinetic photoelectron energy for different encapsulated atoms was calculated in Refs. [4,5,17]. This is a rapid variation function. The shape and positions of oscillations in $|F_A(\varepsilon)|^2$ are very sensitive to the particular $A$ atom encapsulated and to the magnitude of the fullerene shell radius $R$ [17].

### VI. RESULTS OF CALCULATING RADIATIVE AND AUGER WIDTHS

The calculation results, according to formula (7), of a ratio between the width of the radiative vacancy decay in the $A$ atom inside the fullerene shell and the width of the same decay in the free $A$ atom: $\eta_{RR}(\omega)=\Gamma_{\gamma,if}^{C_{60}}/\Gamma_{\gamma,if}^{A}$ is given in Fig. 2. The frequency dependence $\eta_{RR}(\omega)$ is a curve with two pronounced maxima, for ~5 and ~25 eV.

The curve presented in Fig. 2 differs significantly from that obtained within the assumption that the fullerene shell can be considered as an ideally conducting sphere [12]. In the static limit, i.e., for zero radiation frequencies, the func-

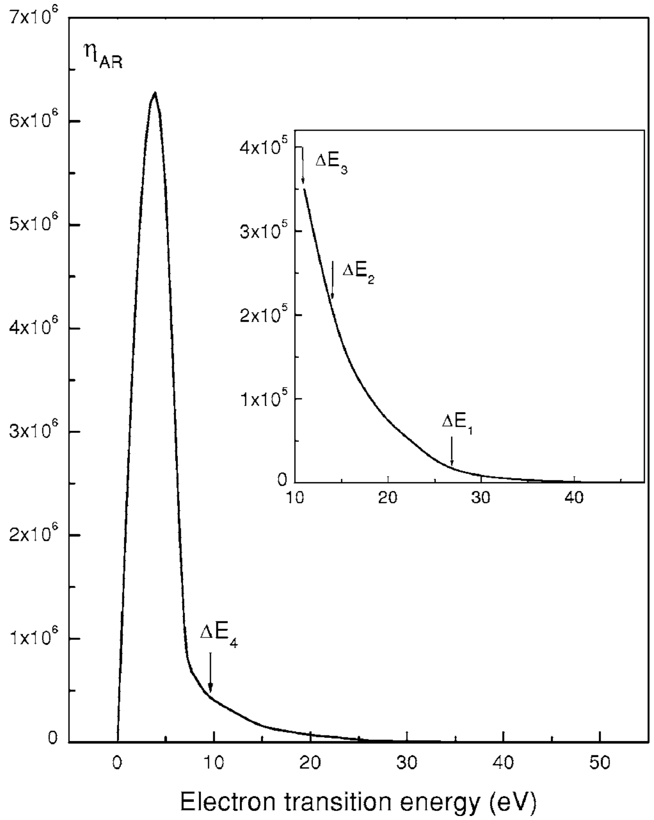


FIG. 3. The ratio of Auger and radiative widths $\eta_{AR}(\omega)$ as a function of transition energy. Arrows have the same meaning as in Fig. 2.

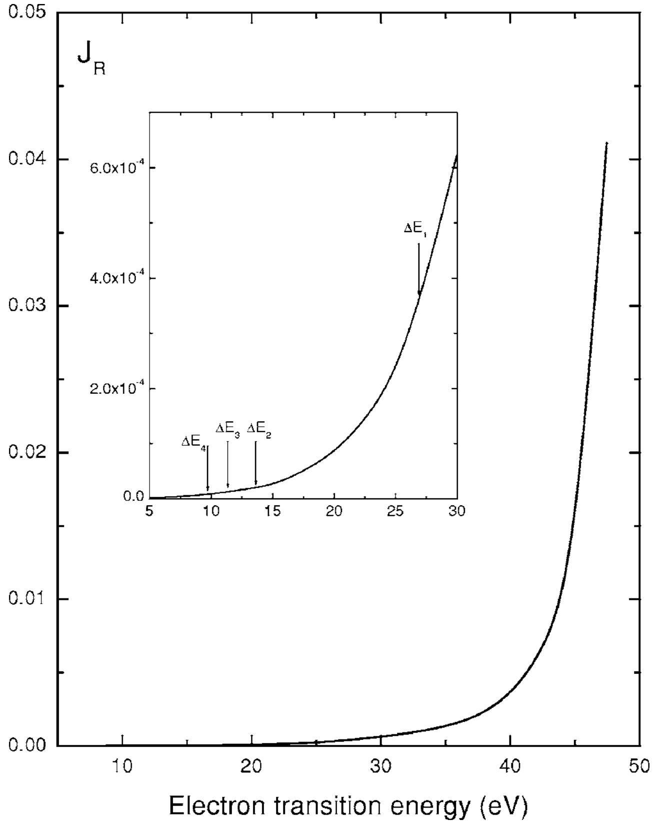


FIG. 4. The fluorescence yield $J_R(\omega)$ as a function of transition energy.

tion is $\eta_{RR}(0)\approx 2.26$ while for the ideally conducting sphere this function has to be equal to zero. When the frequency $\omega$ increases, the function $\eta_{RR}(\omega)\to 1$. The curve maxima reach values of about 6–7. According to Fig. 2, over the range of photon energies 0–50 eV the radiative widths of the vacancy decays in the endohedral and free atoms are significantly different and strongly depend on recombination photon energy.

Calculated with formula (10), the ratio of the Auger-decay width of the vacancy in the endohedral atom to the radiative width of the free atom $\eta_{AR}(\omega)=\Gamma^{C_{60}}_{A,if}/\Gamma^{A}_{\gamma,if}$ is given in Fig. 3. At the threshold of the $C_{60}$ ionization this ratio vanishes and then rapidly reaches its maximal value $\eta_{AR}(5\text{ eV})\approx 6.3\times 10^6$. With an increase of transition frequency $\eta_{AR}(\omega)$ rapidly decreases. The function $\eta_{AR}(\omega)$ for transition energy within the range of $10\leqslant\omega\leqslant 50$ eV is given in the insert of this figure. Thus, intershell interaction in the endohedral systems $A@C_{60}$ radically (by several orders of magnitude) increases the probability of the vacancy decay. This result is in agreement with the corresponding value for the $3s\to 3p$ transition in Ne@$C_{60}$ obtained in Refs. [18,19].

Calculated with formula (11), the fluorescence yield $J_R(\omega)$ as a function of electron transition energy is given in Fig. 4. This function goes to infinity at the threshold since the imaginary part of polarizability at this point is equal to zero and then rapidly increases with the raise in electron transition energy. In Fig. 4 we omitted the rapidly changing part of the curve near the threshold where the experimental data on the photoabsorption cross section cannot be considered to be sufficiently reliable. As in the previous case, the ratio of the widths is extremely sensitive to transition energy and the fluorescence yield changes by several orders within the range of transition energy $10\leqslant\omega\leqslant 50$ eV.

## VII. CONCLUSIONS

It was demonstrated in this paper that the dynamic reaction of the $C_{60}$ collectivized electrons radically changes the probabilities of the radiative and nonradiative vacancy decays in the endohedral atoms. Existence of a great number of the collectivized electrons in the fullerene shell and their participation in the processes of vacancy decays in the encapsulated atoms changes by several orders of magnitude the widths of these processes.

The small ratio of the atomic shell radius to that of $C_{60}$ shell provides an opportunity to obtain remarkably simple formulas for the probabilities of the electron transitions. This small parameter makes it possible to consider the electronic subsystems of the inner atom and $C_{60}$ shell as weakly interacting and to describe the effect of the spherical shell upon the electronic process in the $A$ atom by several universal formulas.

(1) This is the function $F_A(\varepsilon)$ [see Eq. (16)] built on the basis of the experimental data on energy of electron affinity to the empty fullerene shell and on radius of this $C_{60}$-shell [5,17]. By this function the shape of electronic spectra of

photoionization of an endohedral atom and the Auger-vacancy decay in this atom are defined.

(2) This is the function $\eta_{RR}(\omega)$ built on the basis of the experimental data on photoionization of the $C_{60}$ shell and making it possible to sequentially consider a dynamic reaction of the collectivized electrons both to the processes of endohedral atom photoionization and to those of radiative recombination.

(3) And finally, this is the function $\eta_{AR}(\omega)$ describing the nonradiative collapse of the endohedral atom vacancy with simultaneous ionization of the fullerene shell. The remarkable moment here is that all considered processes are parametrically correctly described with no consideration of any details of the electronic structure of the $C_{60}$ shell.

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