Electron Bulk Perturbation Induced by Radioactive Nuclei

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A strong perturbation of the conduction electrons accompanying the radioactive decay of nuclei is discussed. It is demonstrated that this effect depends strongly on recombination phenomena. A resonant behavior of the excitation process as a function of temperature is predicted.

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Recently some interest has been shown in atomic-nuclear phenomena. It is well known that the use of unstable nuclei as a probe provides a powerful tool to study local properties of materials. We consider in this Letter the spatial dependence and gross properties of condensed matter electron bulk excitation caused by a decay process. The method discussed here is quite general, and it can be applied to study other time dependent phenomena connected with the valence electron excitations, such as photoabsorption and stopping power [1]. However, as we show below, probing the electronic structure of matter by radioactive decay is particularly sensitive to the correlation effects in a local region. For instance, it can be employed in order to understand the role of different elements in superconducting ceramics. At the same time, the use of unstable nuclei can have some advantages, for example, with respect to the implantation problem, since the following radioactive transition can give direct information about the position of implanted ions and solid structure. Furthermore, the time dependent electric field of a decaying radioactive element can considerably modify the local electronic properties. For example, the decay can modify the critical temperature for the transition from amorphous to the crystalline phase or for the defect annealing process [2], similar to what is observed in experiments with laser or ion beams [3]. Especially in the cases of internal conversion or K capture, the radiative and Auger deexcitations of internal shell vacancies give ions with a large charge $Z_0 \approx 10-20$ [4] ($e = m_e = \hbar = 1$). These ions can yield also a strong electron emission from the solids [5].

The time dependence of the ion charge at radioactive processes can be approximated as

$$Z(t) = Z_0 \times \begin{cases} \exp(t/\tau_v) & \text{for } t < 0, \\ \exp(-t/\tau_{\rm rec}) & \text{for } t > 0, \end{cases} \tag{1}$$

where τ_v is the time for the Auger cascade, while $\tau_{\rm rec}$ is the recombination time.

The dynamics of the valence electrons in this *external field* can be considered within the semiclassical kinetic theory [6]. Then for the time dependent phase-space distribution function of the electrons $f(\vec{r}, \vec{p}; t)$ we have

$$\frac{\partial f}{\partial t} + \{H, f\} = \mathrm{St}[f], \qquad H = H_0 + Z(t) V_c(|\vec{R} - \vec{r}\,|), \tag{2}$$

where $\vec{R}$ is the coordinate of the ion, $V_c(r) = \exp(-r/r_D)/r$ is the screened Coulomb potential, r_D is the Debye radius, and $H_0(\vec{r}, \vec{p}) = U(\vec{r}) + p^2/2$ is the unperturbed Hamiltonian with a self-consistent mean field $U(\vec{r})$. $\mathrm{St}[f]$ is the collisional term which simulates the correlation effects in semiclassical electron dynamics. This approximation corresponds to the Thomas-Fermi model ($\hbar \to 0$) for the electron gas (see [7–10]), with respect to the Hartree self-consistent mean field equations in quantum mechanics.

We use perturbation theory (pt) for our estimations and separate the actual distribution function of the electrons into stationary and time dependent parts, $f(\vec{r}, \vec{p}; t) = f_0(\vec{r}, \vec{p}) + \delta f(\vec{r}, \vec{p}; t)$. Here $f_0(\vec{r}, \vec{p}\,) \equiv f_0(H_0(\vec{r}, \vec{p}\,))$ is the Thomas-Fermi ($\hbar \to 0$) unperturbed distribution function, which in the zero temperature limit is given by $f_0(H_0) = (2\pi)^{-3} 2\theta(\epsilon_F - H_0)$, where $\theta(x)$ is the theta function, and ϵ_F is the Fermi energy of the valence electron gas. Neglecting correlation effects ($\mathrm{St}[f]$), which are reasonably small for a strongly degenerate electron gas in the low temperature limit because of Pauli blocking (zero sound regime [6,8,11]), we obtain, with linear accuracy, the following expression for the distant dependent excitation strength (see, for example, [12]):

$$P(r, \epsilon) = (2\pi)^{-1} \int dt\, d\vec{p}\ \delta f(\vec{r}, \vec{p}; t) e^{-i\epsilon t} \approx Z_0^2 \frac{2\epsilon(\tau_{\rm rec}^{-1} + \tau_v^{-1})^2}{(\tau_{\rm rec}^{-2} + \epsilon^2)(\tau_v^{-2} + \epsilon^2)} S(r, \epsilon), \tag{3}$$

where r is the distance from the radioactive nucleus, and

$$S(r, \epsilon) = (2\pi)^{-1} \int dt\, C(r, \epsilon_F, t) \exp(-i\epsilon t), \tag{4}$$

$$C(r, E, t) = (2\pi)^{-3} \int d\vec{p}\ \delta[E - H_0(\vec{r}, \vec{p}\,)] \times V_c(|\vec{R} - \vec{r}\,|) V_c(|\vec{R} - \vec{r}\,(t)|). \tag{5}$$

Here $\vec{r}(t)$ is a point in coordinate space that belongs to a classical trajectory governed by the Hamiltonian $H_0(\vec{r}, \vec{p}\,)$, with initial conditions $\{\vec{r}, \vec{p}\,\}$. We have used the following expression for the Fourier transform (Z_ϵ) of the time dependent charge (1):

$$Z_\epsilon = Z_0 \left[(\tau_v + i\epsilon)^{-1} + (\tau_{\rm rec} - i\epsilon)^{-1} \right]. \tag{6}$$

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For metals we can use a free electron approximation neglecting the gradients of the mean field $U(\vec{r})$. Then, since $\vec{r}\,(t) = \vec{r} + \vec{p}\,t$, we obtain the distant dependent strength function $S_c(r, \epsilon)$ in the following form:

$$S_c = \frac{r_D}{(\pi r)^2} F(\zeta, \xi), \quad \text{with } \zeta = r/r_D, \quad \xi = \epsilon/\epsilon_0,$$

$$F(\zeta, \xi) = \int_0^\infty dx \cos(\xi x) \times \frac{\exp(-|\zeta - x|) - \exp(-|\zeta + x|)}{x}, \quad (7)$$

where $\epsilon_0 = v_F/r_D = \sqrt{3}\epsilon_p$, ϵ_p is the plasma frequency, and $v_F = (2\epsilon_F)^{1/2}$. Figure 1 shows the function $F(\zeta, \xi)$. We see that the considered process gives a strong perturbation of the valence electrons close to the radioactive nucleus volume with linear size $\Delta R \approx 8r_D$.

To estimate the power of this effect we consider the energy moment,

$$\langle \epsilon^k \rangle = \int d\vec{r}\, d\epsilon\, \epsilon^k P(r, \epsilon) = \left(\frac{Z_0 r_D}{\pi a_B}\right)^2 \times 2\epsilon_0^{k+2}(\tau_{\rm rec}^{-1} + \tau_v^{-1})^2 I_k, \quad (8)$$

$$I_k = \int \frac{dx\, x^{k+1}}{[x^2 + (\epsilon_0\tau_{\rm rec})^{-2}][x^2 + (\epsilon_0\tau_v)^{-2}](x^2 + 1)},$$

that gives the total number of excited electrons ($k = 0$, $N \equiv \langle \epsilon^0 \rangle$), mean excitation energy ($k = 1$), etc. The first three moments are written in the following form:

$$N_0 \equiv \langle \epsilon^0 \rangle = 2\left(\frac{Z_0 r_D}{\pi a_B}\right)^2 \frac{\tau_v + \tau_{\rm rec}}{\tau_v - \tau_{\rm rec}} \times \left(\frac{\ln(\epsilon_0\tau_v)}{1 - (\epsilon_0\tau_v)^{-2}} - \frac{\ln(\epsilon_0\tau_{\rm rec})}{1 - (\epsilon_0\tau_{\rm rec})^{-2}}\right), \quad (9)$$

$$\langle \epsilon^1 \rangle = \left(\frac{Z_0 r_D}{a_B}\right)^2 \frac{\epsilon_0^2}{\pi} \frac{\tau_v + \tau_{\rm rec}}{(1 + \epsilon_0\tau_v)(1 + \epsilon_0\tau_{\rm rec})}, \quad (10)$$

$$\langle \epsilon^2 \rangle = 2\left(\frac{Z_0 r_D}{\pi a_B}\right)^2 \epsilon_0^2 \frac{\tau_v + \tau_{\rm rec}}{\tau_v - \tau_{\rm rec}} \times \left(\frac{\ln(\epsilon_0\tau_v)}{(\epsilon_0\tau_v)^2 - 1} - \frac{\ln(\epsilon_0\tau_{\rm rec})}{(\epsilon_0\tau_{\rm rec})^2 - 1}\right). \quad (11)$$

Since for realistic cases the relations

$$\epsilon_p\tau_{\rm rec} \gg 1 \quad \text{and} \quad \epsilon_p\tau_v \sim 1$$

are fulfilled, the energy dependence of the excitation spectrum is determined by the second term on the right hand side of Eq. (3). We evaluate the values $\langle \epsilon^k \rangle$ (9), (10), and (11) in a simple way as

$$\langle \epsilon^k \rangle = \left(\frac{Z_0 r_D}{\pi a_B}\right)^2 \times \begin{cases} 2\ln(\epsilon_0\tau_{\rm rec}) \\ \pi\epsilon_0 \\ 2\epsilon_0^2 \ln(\epsilon_0\tau_{\rm rec}) \end{cases} \sim Z_0^2 \times \begin{cases} 1 & \text{for } k = 0, \\ 0.1\epsilon_p & \text{for } k = 1, \\ 3\epsilon_p^2 & \text{for } k = 2. \end{cases} \quad (12)$$

Thus we see that this effect is very large and can modify considerably the material properties locally.

To estimate the accuracy of pt with respect to this problem we consider numerical simulations of the semiclassical electron dynamics in the external field $V(t)$ [see Eq. (2)], using the test-particles (TP) method [9,10]. The case of the infinite matter is simulated using the periodic boundary conditions for the considered volume. In our calculations this volume corresponds to 64 atoms of Al with total number of conduction electrons $A_e = 192$. In Fig. 2 we compare the results of the numerical simulations and Eq. (10) for Al. We see good quantitative

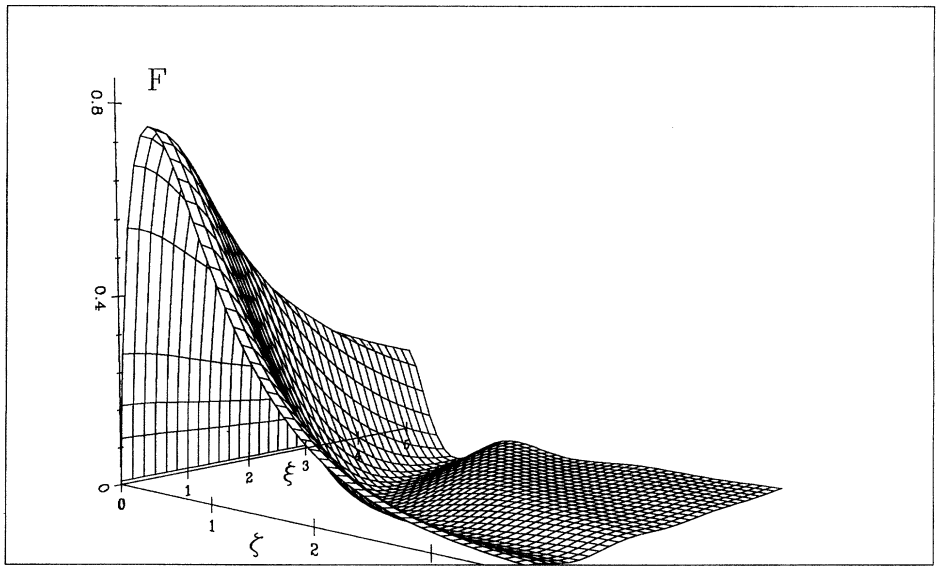

FIG. 1. The function $F(\zeta, \xi)$ versus excitation energy and distance from a radioactive nucleus.

agreement for the wide region of parameter Z_0 up to value $Z_0^m \approx \epsilon_0 \tau_{\rm rec} \gg 1$.

It is not surprising that pt works rather well for the description of the considered process. The application of pt is generally restricted by the condition

$$\delta\epsilon_e/\epsilon_F \ll 1\,, \tag{13}$$

where $\delta\epsilon_e$ is the energy that is absorbed by an electron from the external field. This value ($\delta\epsilon_e$) can be estimated as

$$\delta\epsilon_e \approx \frac{\partial\langle V\rangle}{\partial t}\delta t_{\rm int}\,, \tag{14}$$

where $\delta t_{\rm int}$ is the interaction time being in the order of magnitude of $\delta t_{\rm int} \approx \epsilon_0^{-1}$. The potential of the external field ($\langle V\rangle$) averaged over the spatial electron density (ρ) is estimated as $\langle V\rangle \approx 4\pi\rho r_D^2 Z_0 \exp(-t/\tau)$. Using Eqs. (13) and (14) we obtain the relation $Z_0 \ll \epsilon_0\tau$.

At finite temperature (T) of the system, correlation effects, reflecting the inexactness of the Hartree method and associated with the deviation of the true interaction from the self-consistent (averaged) one (U), become important. The correlation as well as thermodynamic fluctuation effects can be included in this semiclassical independent-particle picture of fermionic dynamics in a phenomenological way using the Langevin equation of motion (see [5,11,13]),

$$\dot{\vec{p}}\,(t) = -\vec{\nabla}_r h'(\vec{r},\vec{p}\,) + \vec{f}(t)\,, \tag{15}$$

where $\vec{f}(t)$ is a Gaussian random force associated with the mean field fluctuations. This additional force always arises whenever we deal with a reduced description of a system, and it simulates the coupling of the degrees of freedom, which are not explicitly considered. Neglecting *memory effects* we assume that the equilibrium correlation function of $\vec{f}(t)$ is a delta function,

$$\langle \vec{f}(t)\vec{f}(t')\rangle = 2D_f\delta(t-t')\,. \tag{16}$$

The fluctuation properties of the mean field are related to the dissipation properties through the fluctuation-dissipation theorem. In the case of small fluctuations around equilibrium we can use the well-known (see [11,13]) result $D_f \approx \eta\hbar\omega_0\coth(\hbar\omega_0/2T)$, where ω_0 is the lattice phonon frequency and η is the friction coefficient, which can be practically estimated from the conductivity.

Since the Langevin equation of motion is not integrable we define the probability $\rho(\vec{r}_1,\vec{r};t)$ of finding an electron with initial conditions $\{\vec{r},\vec{p}\,\}$ at the point $\vec{r}_1$ at the time t. Then the correlation function $C(r,E,t)$ can be written as

$$C(r,E,t) = \int d\vec{r}_1\, d\vec{p}\;\delta(E - H_0(\vec{r},\vec{p}\,)) \times (2\pi)^{-3}\rho(\vec{r}_1,\vec{r};t)V_c(|\,\vec{R}-\vec{r}\,|)V_c(|\,\vec{R}-\vec{r}_1\,|)\,. \tag{17}$$

In the case of white noise random forces, e.g., (16), the probability $\rho(\vec{r}_1,\vec{r};t)$ is given, for infinite matter, by the Gaussian distribution function (see [14]),

$$\rho(\vec{r}_1,\vec{r};t) \approx \exp\{-[\vec{r}_1 - \vec{r}\,(t)]^2/D_r t\}/(\pi D_r t)^{3/2}, \tag{18}$$

with spatial diffusion coefficient $D_r = \eta^{-2}D_f$. Thus using Eqs. (17) and (18) we can write for the temperature dependent strength function [see Eq. (4)] integrated over the distance from the radioactive nucleus the following equation:

$$S_\gamma(\epsilon) \approx \frac{(r_D)^2}{(\pi a_B)^2}[1 + (\gamma\epsilon/\epsilon_0)^2]^{-1/2} \times \frac{a^3 + \gamma/4 + (a+1)\{a[\gamma(\gamma-1)/2 + a] - 1\}}{(1-\gamma/4)^2 + (a^2-1)(2a/\gamma)^2}\,, \tag{19}$$

where $a^2 = 0.5\{1 + [1 + (\gamma\epsilon/\epsilon_0)^2]^{1/2}\}$. The parameter $\gamma = D_r/v_F r_D$ indicates the ratio of correlation and interaction lengths. The zero temperature limit in absence of zero point lattice vibrations ($\hbar \to 0$) can be obtained from the last equation putting $\gamma \to 0$ (see [5]). Figure 3 shows the excitation strength $P_\gamma(\epsilon)$ versus excitation energy and temperature for some realistic cases. We see that at small temperature the excitation spectrum in the low energy region is very sensitive to the recombination processes. At the same time $P_\gamma(\epsilon)$ is practically independent from the time τ_v for the Auger cascade.

Since, for realistic cases, the main contribution to the excitation spectrum comes from the energy interval $\epsilon \sim \tau_{\rm rec}^{-1} \ll \epsilon_p$ [see Eq. (3) and Fig. 3], we can estimate the moments of energy at finite temperature as

$$\langle\epsilon^k\rangle_\gamma \approx \langle\epsilon^k\rangle_0 \frac{1 - 3\gamma/4 + \gamma^2/2}{(1-\gamma/4)^2 + (\epsilon_0\tau_{\rm rec})^{-2}}\,, \tag{20}$$

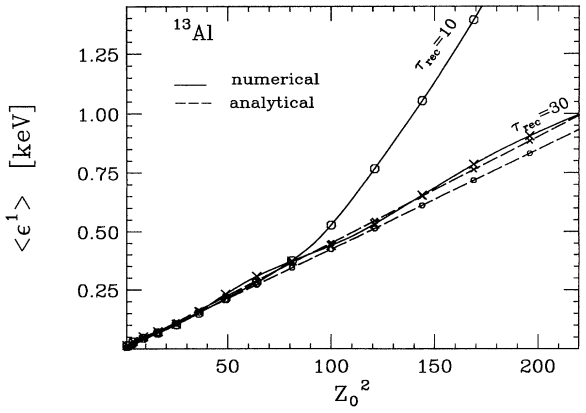

FIG. 2. The mean excitation energy ($\langle\epsilon^1\rangle$) in Al versus parameter Z_0. The results of numerical simulations (see text) are given for $\tau_{\rm rec} = 10$ a.u. (○) and $\tau_{\rm rec} = 30$ a.u. (×), while −− is an analytical estimation [see Eq. (10)].

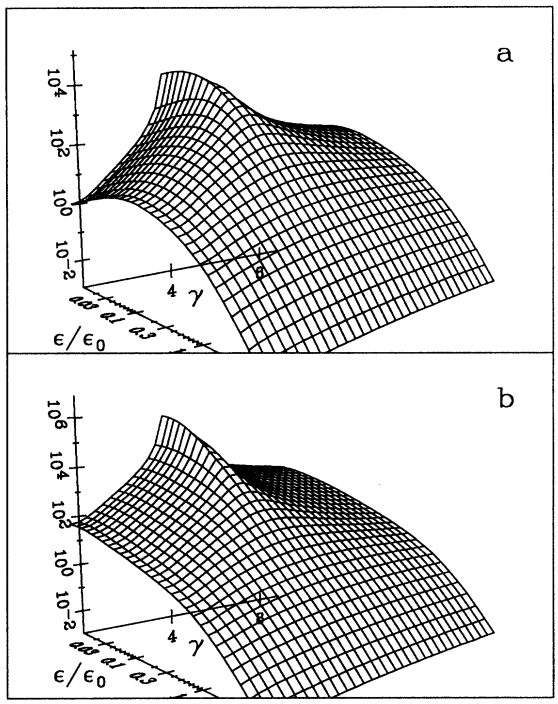

FIG. 3. The function $\frac{\epsilon_0(\pi a_B)^2}{2(Z_0 r_D)^2} P_\gamma(\epsilon)$ versus excitation energy (ϵ) and temperature ($\gamma = D_r/v_F r_D$) for cases $\epsilon_0\tau_v = 0.3$ and $\epsilon_0\tau_{\rm rec} = 10$ (a), 100 (b).

where $\langle\epsilon^k\rangle_0$ are corresponding values in the zero temperature limit given by Eqs. (9), (10), and (12).

From Eq. (20) we can see that the electron bulk excitation process has a resonant behavior, with a resonance at a temperature $T_r \approx 4 v_F r_D \eta$ (Fig. 4). The correlation length

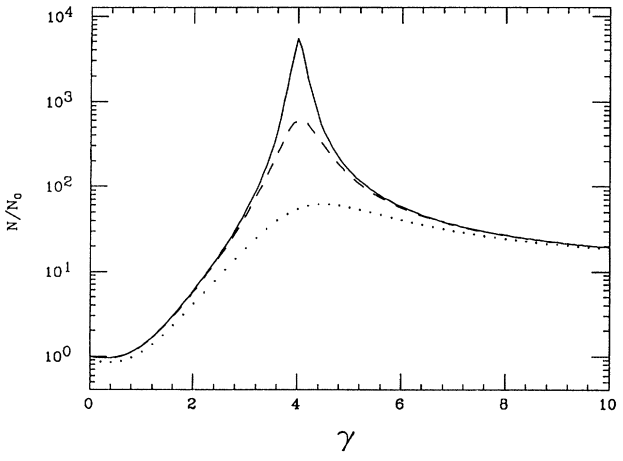

FIG. 4. Temperature ($\gamma = D_r/v_F r_D$) dependence of the total number of excited electrons for $\epsilon_0\tau_{\rm rec} = 30$ (full line), 10 (dashed), and 3 (dotted).

in medium becomes comparable with the interaction length at this condition. The total number of excited electrons is very large at this temperature: $N_{\rm res} \approx 6N_0(\epsilon_0\tau_{\rm rec})^2 \sim 10^4 - 10^6$ per radiative transition.

An experimental study of the process considered here would be very interesting. Especially, experiments where coincidences between conversion and low energy electrons are measured could give information, for example, about the emission depth. On the other hand, the resonant behavior of the excitation process predicted here can give some *curious surprises,* such as an anomalous temperature and concentration dependence of the radioactivity rates (see [15]).

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