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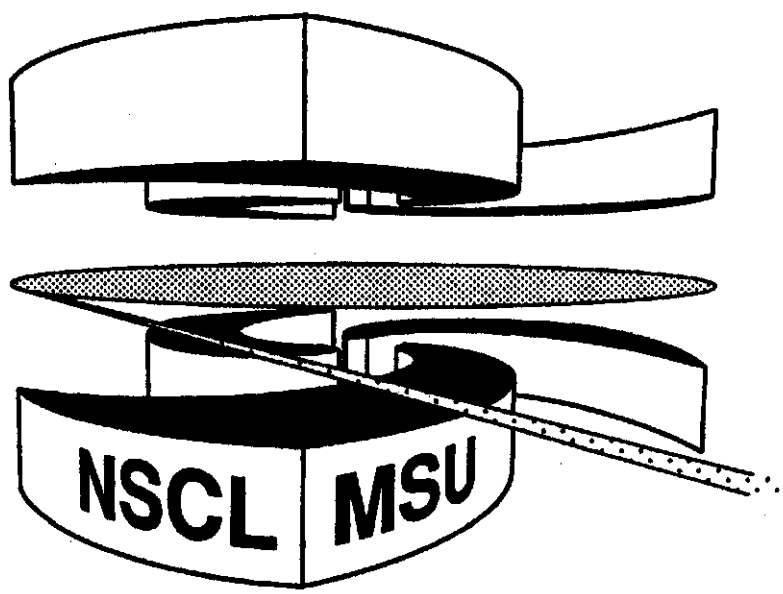

MSUCL-965

JANUARY 1995

Funny Hill in a "Warm Bath":

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 behaviour of the excitation process as . function of temperature is predicted.

PACS: 23.40.Nx; 79.70.+q; 79.20.Fv

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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, like photo-absorption and stopping power [1]. However, as we show below the probing of 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 super-conducting 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], similarly 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 de-excitations of internal shell vacancies give ions with a large charge $Z_0 \approx 10 \div 20$ [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 the radioactive processes can be approximated as:

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

where τ_v is the time for the Auger cascade, while τ_{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:

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

where $\vec{R}$ is the coordinate of the ion, $V_c(r) = \frac{exp(-r/r_D)}{r}$ is the screened Coulomb potential corresponding to the interaction of two charges in an electron gas, r_D is the Debye radius, and $H_0(\vec{r},\vec{p}) = U(\vec{r}) + \frac{p^2}{2}$ is the unperturbed Hamiltonian with a self-consistent meanfield $U(\vec{r})$. $St[f]$ 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 meanfield equations in quantum mechanics.

We use the perturbation theory (p.t.) for our estimations and separate the actual distribution function of the electrons into the 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 the correlation effects ($St[f]$), 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]):

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

where r is the distance from the radioactive nucleus, and

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

with

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

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):

$$Z_\epsilon = Z_0((\tau_v + i\epsilon)^{-1} + (\tau_{rec} - i\epsilon)^{-1}) \qquad (6)$$

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

$$S_c = \frac{\tau_D}{(\pi r)^2} F(\zeta,\xi)\ ;\ \text{with}\ \ \zeta = r/r_D\ ,\ \xi = \epsilon/\epsilon_0 \qquad (7)$$
$$\text{and} \qquad F(\zeta,\xi) = \int_0^\infty dx\ \cos(\xi x)$$
$$\frac{exp\{-\,|\,\zeta - x\,|\} - exp\{-\,|\,\zeta + x\,|\}}{x}\ ,$$

where $\epsilon_0 = v_F/r_D = \sqrt{3}\ \epsilon_p$, ϵ_p is the plasma frequency and $v_F = (2\epsilon_F)^{1/2}$. Fig.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 $\Delta R \approx 8 r_D$.

To estimate the power of this effect we consider the energy moments:

$$< \epsilon^k >= \int d\vec{r}\, d\epsilon\, \epsilon^k\, P(r,\epsilon) = \left(\frac{Z_0 r_D}{\pi a_B}\right)^2 * \quad (8)$$
$$2\epsilon_0{}^{k+2}\,(\tau_{rec}{}^{-1}+\tau_v{}^{-1})^2\;\; I_k\;,$$
$$I_k \;=\; \int \frac{dx\; x^{k+1}}{(x^2+(\epsilon_0\tau_{rec})^{-2})(x^2+(\epsilon_0\tau_v)^{-2})(x^2+1)}$$

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

$$N_0 \equiv < \epsilon^0 > \;=\; 2\left(\frac{Z_0 r_D}{\pi a_B}\right)^2 \frac{\tau_v+\tau_{rec}}{\tau_v-\tau_{rec}} \quad (9)$$
$$\left(\frac{\ln(\epsilon_0\tau_v)}{1-(\epsilon_0\tau_v)^{-2}} - \frac{\ln(\epsilon_0\tau_{rec})}{1-(\epsilon_0\tau_{rec})^{-2}}\right)$$

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

$$< \epsilon^2 > = \; 2\left(\frac{Z_0 r_D}{\pi a_B}\right)^2 {\epsilon_0}^2 \frac{\tau_v+\tau_{rec}}{\tau_v-\tau_{rec}} \quad (11)$$
$$\left(\frac{\ln(\epsilon_0\tau_v)}{(\epsilon_0\tau_v)^2-1} - \frac{\ln(\epsilon_0\tau_{rec})}{(\epsilon_0\tau_{rec})^2-1}\right)$$

Since for realistic cases the following relations:

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

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

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

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

To estimate the accuracy of the p.t. with respect to this problem we consider the numerical simulations of the semiclassical electron dynamics in the external field $V(t)$ (see eq. (2)). Using the test-particles (TP) method [9,10] we write the distribution function $f(\vec{r},\vec{p};\,t)$ as:

$$f(\vec{r},\vec{p};\,t) = \frac{1}{N}\sum_{i=1}^{M} \delta(\vec{r}-\vec{r}_i(t))\delta(\vec{p}-\vec{p}_i(t)) \quad (13)$$

$$\text{with}\;\; \delta\vec{r}_i = \delta t\;\vec{p}_i/m\;;\; \delta\vec{p}_i = -\delta t\;\vec{\nabla}_r(U+V(t)); \quad (14)$$
$$i=1,....,M=NA_e$$

where A_e is a total number of electrons and N is an integer large enough to ensure numerical convergency and associated with a number of TP per electron. The free electron approximation corresponds to the relation: $U=0$, in eq. (14). While 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 the good quantitative agreement for the wide region of the parameter Z_0 up to the value: $Z_0^m \approx (\epsilon_0\tau_{rec}) \gg 1$.

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

$$\frac{\delta\epsilon_e}{\epsilon_F} \ll 1 \quad (15)$$

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:

$$\delta\epsilon_e \approx \frac{\partial < V >}{\partial t}\;\delta t_{int} \quad (16)$$

where δt_{int} is the interaction time being in the order of magnitude of: $\delta t_{int} \approx \epsilon_0{}^{-1}$. The potential of the external field ($< V >$) averaged over the spatial electron density (ρ) is estimated as: $< V > \approx 4\pi\rho r_D^2\, Z_0 exp\{-t/\tau\}$. Using the eq.s (15,16) we obtain the following 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. It is the electron-phonon interaction that plays a dominant role in the kinetic phenomena for the electron gas in semiconductors and metals [11]. Furthermore we can omit the electron influence on the lattice vibrations which is always of the negligible order of magnitude and consider the phonons as an "external bath" that generates the fluctuations in the electron evolution. Therefore 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 [11,13]):

$$\dot{\vec{p}}(t) = -\vec{\nabla}_r h'(\vec{r},\vec{p}) + \vec{f}(t) \quad (17)$$

where $\vec{f}(t)$ is a Gaussian random force associated with the meanfield 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 the *memory effects* we assume, that the equilibrium correlation function of $\vec{f}(t)$ is a delta function:

$$< \vec{f}(t)\vec{f}(t') >= 2D_f\;\delta(t-t') \quad (18)$$

The fluctuation properties of the meanfield 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: $D_f \approx \eta\hbar\omega_0 cth(\frac{\hbar\omega_0}{2T})$, 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:

$$C(r, E, t) = \int d\vec{r}_1 d\vec{p}\, \delta(E - H_0(\vec{r}, \vec{p})) \qquad (19)$$

$$(2\pi)^{-3} \rho(\vec{r}_1, \vec{r}; t)\; V_c(|\vec{R} - \vec{r}|)\; V_c(|\vec{R} - \vec{r}_1|)\; .$$

In the case of white noised random forces (18) the probability $\rho(\vec{r}_1, \vec{r}; t)$ is given, for infinite matter, by the Gaussian distribution function (see [14]):

$$\rho(\vec{r}_1, \vec{r}; t) \approx \frac{exp(-(\vec{r}_1 - \vec{r}(t))^2/D_r t)}{(\pi D_r t)^{3/2}} \qquad (20)$$

with spatial diffusion coefficient: $D_r = \eta^{-2} D_f$. Thus using eq.s (19) and (20) we can write for the temperature dependent strength function (see eq. (4)) integrated over the distance from the radioactive nucleus the following equation:

$$S_\gamma(\epsilon) \approx \frac{(r_D)^2}{(\pi a_B)^2} [1 + (\gamma\epsilon/\epsilon_0)^2]^{-1/2} \qquad (21)$$

$$\frac{a^3 + \gamma/4 + (a+1)\{a(\gamma(\gamma-1)/2 + a) - 1\}}{[(1-\gamma/4)^2 + (a^2-1)(2a/\gamma)^2]}\; ,$$

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]). Fig. 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 is coming from the energy interval $\epsilon \sim {\tau_{rec}}^{-1} \ll \epsilon_p$ (see Eq.(3) and fig.3), we can estimate the moments of energy at finite temperature as:

$$< \epsilon^k >_\gamma \approx < \epsilon^k >_0 \frac{1 - 3\gamma/4 + \gamma^2/2}{(1-\gamma/4)^2 + (\epsilon_0 \tau_{rec})^{-2}} \qquad (22)$$

Where $< \epsilon^k >_0$ is corresponding values in the zero temperature limit given by the eq.s (9,10) and (12).

From this equation we can see that the electron bulk excitation process has a resonant behaviour, with a resonance at a temperature $T_r \approx 4 v_F r_D \eta$ (fig.4). The correlation length in medium becomes comparable with interaction length at this conditions. The total number of excited electrons is very large at this temperature: $N_{res} \approx 6 N_0 (\epsilon_0 \tau_{rec})^2 \sim 10^4 \div 10^6$ per radiative transition.

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

Acknowledgments. Authors are indebted to Prof. G.Weyer for numerous constructive discussions. The hospitality of the INFN-LNS Catania and financial support from INFN post-doctoral fellowship program are gratefully acknowledged by one of us (V.N.K.). A.B. thanks the theory group at the NSCL-MSU for the warm hospitality.

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Figure captions

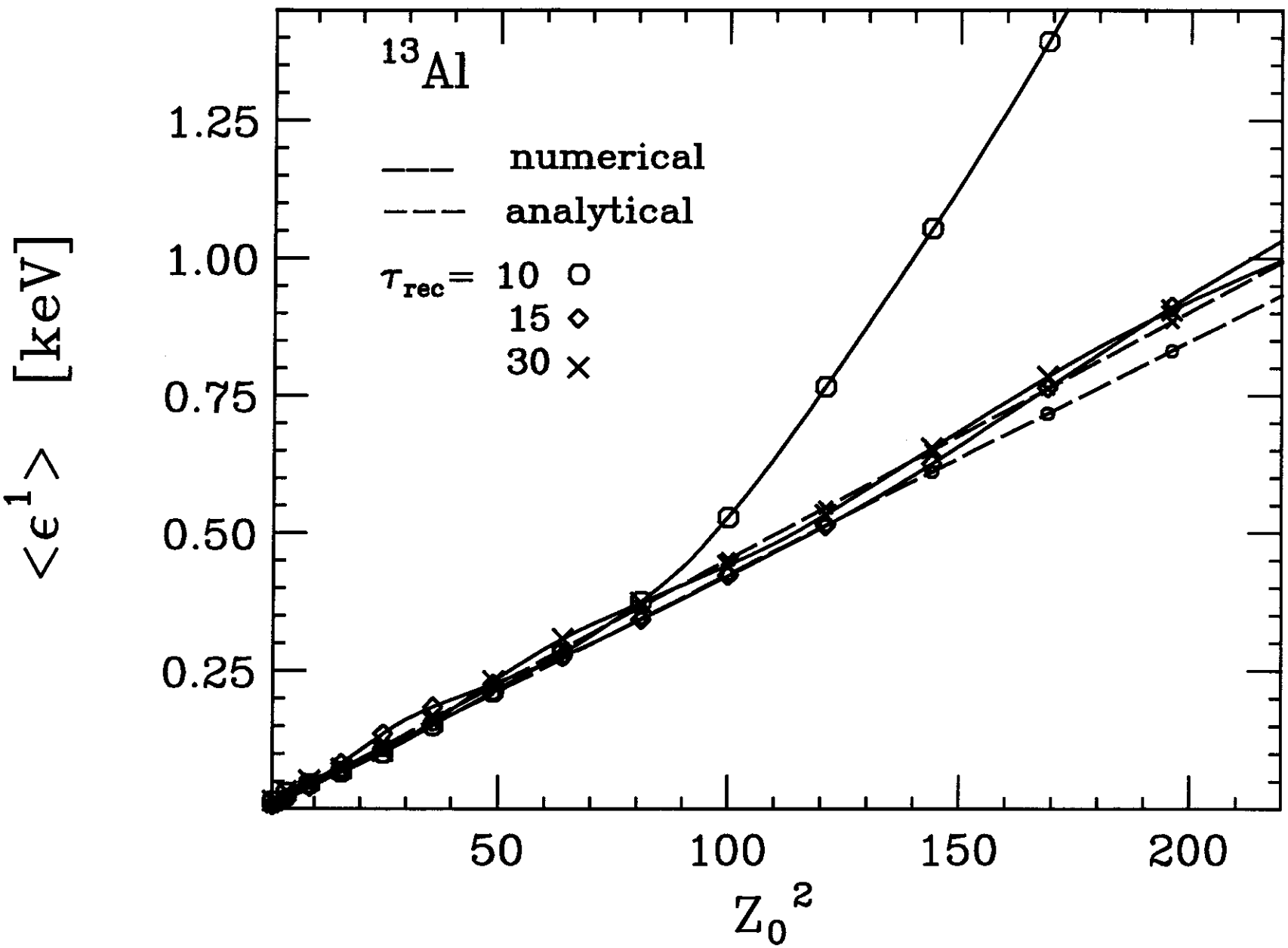
Fig. 1. The function $F(\zeta, \xi)$ *versus* excitation energy and distance from a radioactive nucleus.

Fig. 2. The mean excitation energy ($< \epsilon^1 >$) in *Al versus* parameter Z_0. The results of numerical simulations (see text) are given for: $\tau_{rec} = 10$ a.u. (o) and $\tau_{rec} = 30$ a.u. (x), 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 $\epsilon_0 \tau_{rec} = 10$ (a), 100 (b).

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

0
0.4
0.8
F
0
1
2
3
ζ
0
1
2


^{13}Al
numerical
analytical
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15
30
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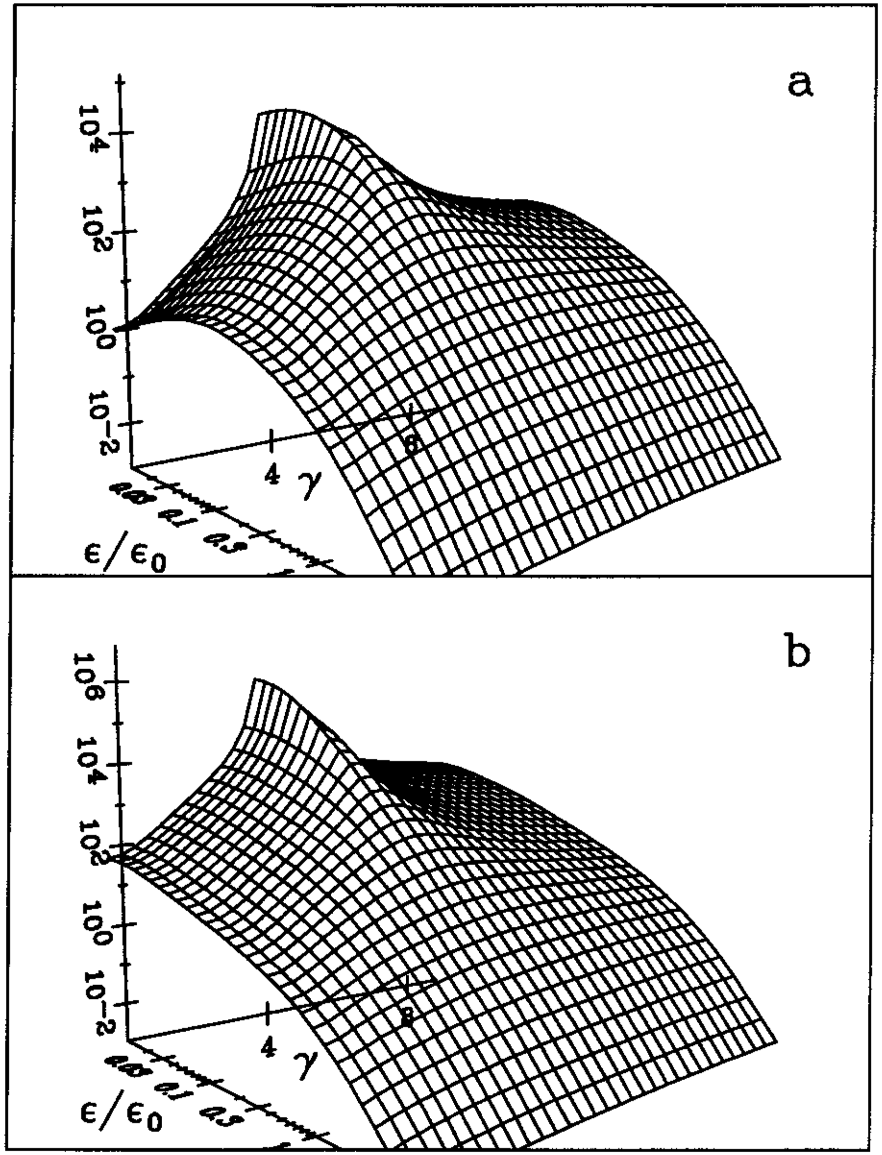
1.00
0.75
0.50
0.25
$\langle\epsilon^1\rangle$ [keV]
50
100
150
200
Z_0^2


a
10⁴
10²
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γ
ε/ε₀
b
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γ
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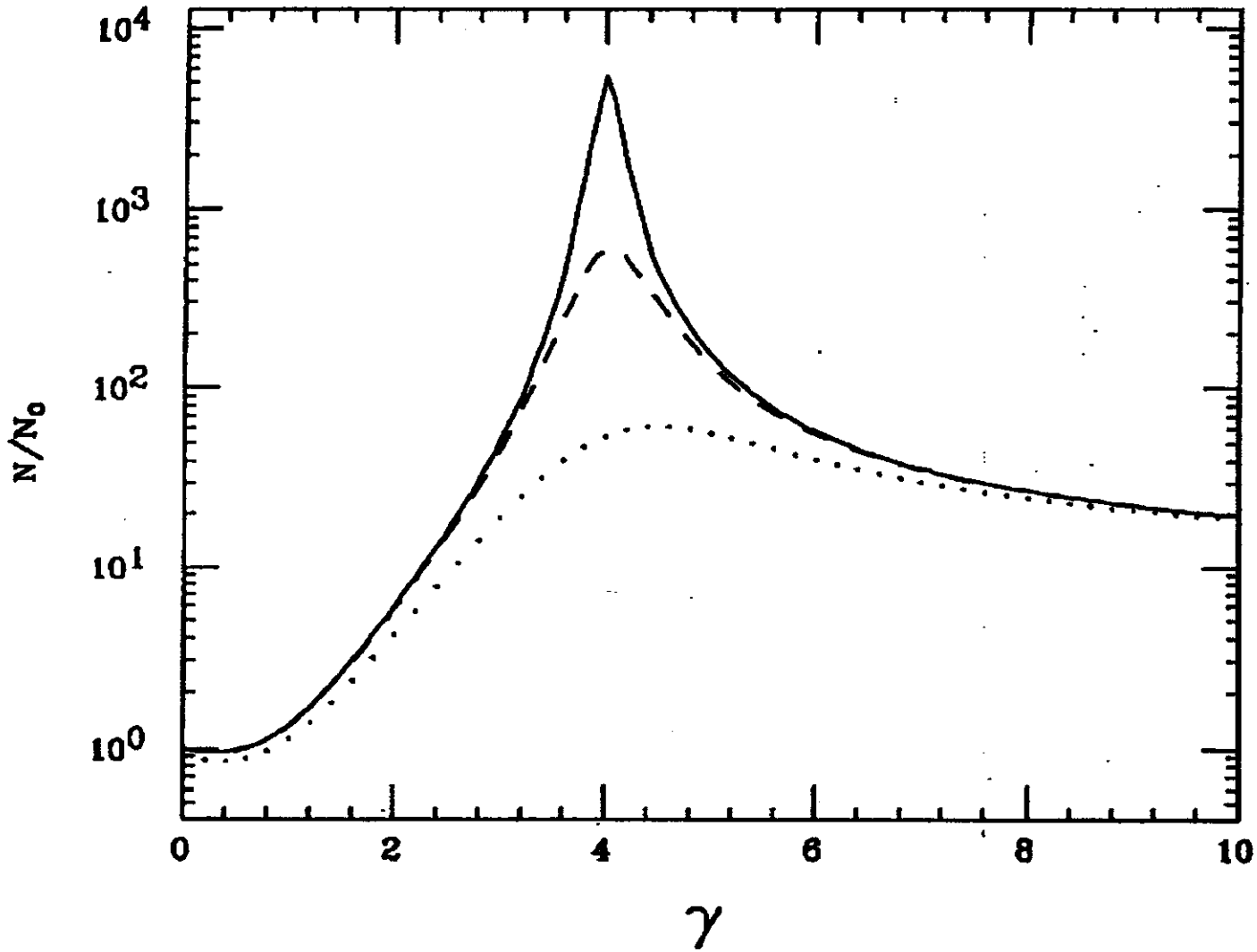
10^4
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10^0
N/N₀
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2
4
6
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