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RADIATIVE ELECTRON CAPTURE FROM A METAL SURFACE

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Radiative electron capture (REC) from a metal conduction band to a 1s vacancy of a H-like ion is investigated in a semi-classical approximation. The ion is assumed to move on a classical trajectory which is idealized by a sequence of two straight-line-trajectory segments at a grazing angle to the surface plane. The REC amplitude is calculated in lowest order perturbation theory and the spectrum of the emitted light is given in terms of the incident ion velocity components parallel and perpendicular to the surface. The close relation of the photon spectrum to the structure of the Galilei-shifted conduction band indicates a new possibility for obtaining information about the surface density of states.

1. Introduction

The process of radiative capture of electrons in ion collisions with atoms and during ion penetration of solid targets is well-studied both experimentally and theoretically. A typical example for the observation of REC X-rays is given by $Ne^{10+} \rightarrow He$, Ne collisions at 140 MeV that have been investigated experimentally by Kienle et al. [1]. REC X-rays arising from the penetration of polycrystalline targets by highly stripped heavy ions have first been observed by Schnopper et al. [2]. Appleton et al. [3] measured photon spectra for 17 to 40 MeV oxygen ions and single crystal Ag and Si targets under channeling conditions in comparison with random crystal orientations, where the ions encounter the lattice cores as they would in an amorphous medium. For 27.78 MeV O^{7+} on Ag single crystals, the AgL characteristic X-ray production for channeled ions was found to be reduced to about 3% of the "random-orientation" value. This small so-called minimum fraction corresponds to ions which are not channeled and approach the target cores close enough to excite characteristic X-rays. The channeling effect therefore drastically reduces close encounters. As a channel can be thought to be surrounded by "surfaces", a similar situation occurs in ion–surface scattering at grazing incidence angles. Indeed, in calculations performed within the Lindhard continues string model [4] in connection with a Thomas–Fermi interatomic potential we obtain distances of closest approach for high energetic ion–surface collisions at grazing incidence of the same order of magnitude as the minimum distance for channeled ions given by Appleton et al. Following this analogy, REC in ion–surface collisions is regarded as an interesting process for obtaining information about the surface-electronic structure of a solid target. The information obtained by the emitted X-ray-spectrum is closely related to the properties of delocalized metal electrons, which, at large impact parameters, become more important than localized target-core electrons. The emitted light can therefore be related to the band structure of the solid target, i.e. to the band-density of states. The purpose of this paper is to elucidate this interdependence of band structure and emitted X-ray spectrum and to explain the distinguishable influence of the motion of the projectile parallel and perpendicular to the surface, i.e. of its large parallel velocity component $v_{\parallel}$ and its small normal component $v_{\perp}$.

Theoretically, REC in ion–atoms collisions has been investigated by several authors [5–8] within a nonrelativistic description, a first-order treatment of the radiation field and the dipole approximation. The theories differ with respect to the approximations applied to the electronic wave function in the entrance and exit channel. Briggs and Dettmann [5b] developed a unified theory of photon emission for both slow and fast collisions, leading to REC and molecular orbital emission in the limiting cases of low and high (but still nonrelativistic) projectile velocities, respectively. In this paper we apply the impact parameter description [5] well established in the theory of ion–atom collisions to ion–surface collisions even though, strictly speaking, an "impact parameter" is not well defined in our case. However, there is a close formal correspondence between the impact parameter used in the case of atomic targets and the distance of closest approach in surface scattering. The main differences between energetic collisions with isolated atoms and surfaces (including delocalized electrons) are: (1) a more complicated solid target electronic structure, which we describe in an effective one electron model using jellium wave functions, (2) an additional

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incoherent summation over the initial band states in our context hardly distinguishable by experimental methods and (3) the absence of an incoherent integration over impact parameter-selected straightline-trajectories due to the broken-straight line description of the projectile motion in front of the surface.

Since nonradiative capture of electrons from surfaces under grazing incidence conditions has been much studied in recent years [9,10], the REC process under the same conditions indicates an alternative method of indirectly measuring the momentum distribution of electronic surface states.

We proceed as follows. In section 2.1, the scattering amplitude corresponding to capture of an electron out of a particular state of the metal conduction band is given. In section 2.2, the incoherent integration over the band is performed. Section 3 contains numerical results and our conclusions. As usual, atomic units are used unless otherwise stated. Vectors will frequently be split into components parallel and perpendicular to the surface according to $\boldsymbol{r} = (\boldsymbol{r}_\parallel, r_\perp)$.

2. Theory

2.1. The transition amplitude

We describe the REC process nonrelativistically and in first order with respect to the electron-radiation field and with unperturbed wave functions. As was pointed out earlier [5a,8b] the nonrelativistic Born approximation leads to the correct high velocity limit only when applied in the frame of the moving projectile. We therefore transform the target wave function in the projectile frame and relate electronic energies to the projectile ionization limit.

In dipole approximation the amplitude for emission of a photon of energy ω, momentum $\boldsymbol{p}$ and linear polarization $\hat{\boldsymbol{e}}_\lambda$ is [5,6]

$$f_\lambda(\boldsymbol{k}, \omega) = \mathrm{i}\left(\frac{2\pi}{V_\mathrm{F}\omega}\right)^{1/2} \hat{\boldsymbol{e}}_\lambda \cdot \int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{\mathrm{i}\omega t - \eta|t|} \langle \phi_\mathrm{P}(t) | \boldsymbol{p} | \psi_k(t) \rangle, \tag{1}$$

where V_F is the quantization volume of the radiation field. The converge parameter $\eta \to 0^+$ expresses the boundary conditions for $|t| \to \infty$. In jellium approximation [11] and after transformation of the initial and final state wave functions, ψ_k and ϕ_P respectively, in momentum space, eq. (1) can be written as

$$f_\lambda(\boldsymbol{k}, \omega) = -(2\pi V V_\mathrm{F}\omega)^{-1/2} v_\perp^{-1} \int \mathrm{d}q_\perp \tilde{\phi}_\mathrm{P}^*(\boldsymbol{Q}) \tilde{u}_{k_\perp}(q_\perp) \mathrm{e}^{\mathrm{i}q_\perp b} \times \hat{\boldsymbol{e}}_\lambda \cdot \boldsymbol{Q} \left[\frac{1}{q_\perp - v_\perp^{-1}\Delta + \mathrm{i}\eta} + \frac{1}{q_\perp + v_\perp^{-1}\Delta + \mathrm{i}\eta} \right], \tag{2}$$

with

$$\boldsymbol{Q} \equiv (\boldsymbol{k}'_\parallel, q_\perp - v_\perp), \qquad \boldsymbol{k}'_\parallel \equiv \boldsymbol{k}_\parallel - \boldsymbol{v}_\parallel,$$

$$\Delta \equiv \omega + \epsilon_\mathrm{P} - \tfrac{1}{2}\left(k_\parallel'^2 + k_\perp^2 + v_\perp^2\right) + V_0.$$

The factor of the jellium wave function perpendicular to the surface in momentum representation is $\tilde{u}_{k_\perp}$. The momentum of the jellium electron, the height of the potential step at the surface, the energy of the final bound state, the metal-quantization volume and the distance of the closest approach of the projectile nucleus to the surface are denoted by k, V_0, ϵ_P, V and b, respectively. The two terms in brackets in the integrand of eq. (2) are the incoming and outgoing part contribution of the broken straightline-trajectory. Note that in contrast to ion–atom collisions, where an analogous treatment of eq. (1) leads to a factor $\delta(\Delta)$, in the case of ion–surface scattering the REC spectrum is folded with the initial and final state momentum distributions. This difference is due to differences in the trajectories. The condition $\Delta = 0$ found for atomic targets is a consequence of the straightline line projectile motion inherent in the impact parameter method. In the limiting case $v_\perp \to 0$ the principal 'value parts in eq. (2) cancel, whereas the remaining parts, proportional to $\delta(\Delta)$, add coherently, leading back to the more familiar result of a straight-line trajectory. For grazing incidence ion–surface collisions eq. (2) is still sharply peaked at $\Delta \approx 0$. The REC peak produced by capture of a particular band electron has a (small) finite width, which increases with increasing $v_\perp$. Therefore, with respect to capture out of the whole band (see section 2.2), the intra-band structure is the more clearly seen in the REC spectrum, the smaller $v_\perp$.

Eq. (2) is the first order amplitude in jellium approximation. For hydrogenic final states the $q_\perp$ integration can be performed by contour integration. Even for capture into a hydrogenic ground state the analytical result is rather lengthy and will not be shown here. However, we mention that, regarded as a function of b, it has the form

$$f_\lambda = a_1 \mathrm{e}^{-Fb} + a_2 \mathrm{e}^{-\gamma b}, \qquad F^2 \equiv Z_\mathrm{P}^2 + k_\parallel'^2, \tag{3}$$

where a_1 and a_2 are polynomials of first order in b and γ is the decay constant of the exponentially decaying part of the jellium wave function outside the surface. The first term in f_λ reflects the exponential decay of the final state wave function (Z_P is the projectile nucleus charge) and the matching condition $\boldsymbol{k}_\parallel = \boldsymbol{v}_\parallel$ for the parallel motion of the jellium electron and the projectile.

2.2. The transition probability

We integrate incoherently over the occupied jellium states representing the conduction band. At zero temperature the probability for emission of a photon with

energy ω in the interval $[\omega, \omega + d\omega]$ into the solid angle $d\Omega$ is

$$\frac{dP_\lambda}{d\omega\, d\Omega}(\omega) = \rho \frac{dN}{d\omega\, d\Omega} \int_{k \le k_F} d^3k\, |f_\lambda|^2, \quad (4)$$

where $\rho = V/(4\pi^3)$ is the jellium-density of states including a factor 2 for the two spin orientations. The number dN of photons emitted in $d\Omega$ with energies in $[\omega, \omega + d\omega]$ is given by [12] $dN/d\omega d\Omega = V_F \omega^2/(2\pi c)^3$.

After transformation to the $v_\parallel$-shifted momentum sphere the integration over the polar angle ϕ' of $k'_\parallel$ can be performed analytically [13], leading to a final expression for eq. (4) containing two integrations (over $k'_\parallel$ and $k_\perp$), which have been performed numerically.

3. Numerical results and conclusions

At grazing incidence ion–surface collisions, the dependence of the capture process on the parallel projectile motion is simply described by a Galilei-transformation, i.e. by the $v_\parallel$-shift of the Fermi-sphere. This results in increasing energies of the emitted REC photons and an increasing width of the total emission spectrum as $v_\parallel$ increases. In table 1, we show the upper and lower limit of the emission spectrum for different $v_\parallel$ for protons colliding with tungsten.

The projectile motion perpendicular to the surface influences the width of each REC-peak originating from the capture of a particular band electron (eq. (2) and subsequent discussion). Since the width of these peaks increases with increasing $v_\perp$, the bottom, the Fermi level and a possible internal structure of the conduction band are, in principle, the most distinctly seen in the total emission spectrum for very small $v_\perp$. In fig. 1, the REC peaks due to capture of jellium electrons with $k_\perp = 0.1$ and $k'_\parallel = 8.2$ are shown for different $v_\perp$ during O^{8+} impact on tungsten. The emission direction is perpendicular to the surface, where the radiation is linearly polarized along $v_\parallel$ to more than 99%. The distance of closest approach $b(v_\perp)$ has been determined on the basis of a Thomas–Fermi interatomic potential and the continuum string model [4] (see table 2). The curves in fig. 1 have been obtained by plotting eq. (4)

Table 1

Limits of the $v_\parallel$-shifted conduction band as seen in the REC spectrum

$v_\parallel$	ω_{min}	ω_{max}
0	0.1	0.3
k_F	0.1	1.1
1	0.2	1.5
2	1.0	3.7
5	9.4	16.3

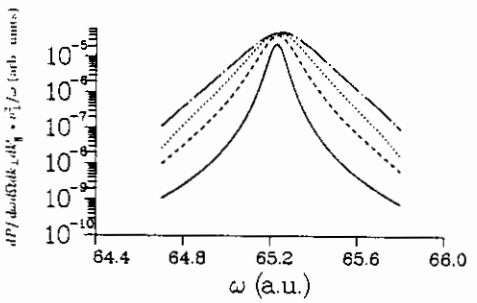

Fig. 1. REC spectrum for emission perpendicular to the surface due to capture of jellium electrons with $k_\perp = 0.1$ and $k'_\parallel = 8.2$ in the ground state of O^{8+} colliding with a tungsten surface at $v_\parallel = 8$. The perpendicular velocity is: ——— $v_\perp = 0.05$, - - - - - $v_\perp = 0.1$, ····· $v_\perp = 0.15$, — · — $v_\perp = 0.2$.

Table 2

Distance of closest approach for $O^{8+} + W$ calculated in the continuum string model [4] using a Thomas–Fermi interatomic potential.

$v_\perp$	b
0.05	0.49
0.1	0.18
0.15	0.06
0.2	0.016

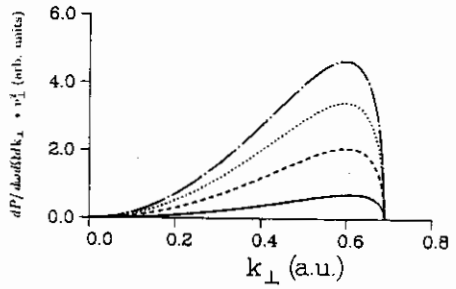

Fig. 2. Collision system and parameters as in fig. 1, but integrated over $k'_\parallel$. REC capture probability for photons emitted with $\omega = 64$ a.u. as a function of the perpendicular momentum component of the jellium electrons.

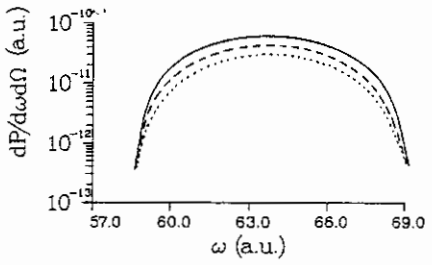

Fig. 3. Collision system and parameters as in fig. 1. Total emission spectrum according to eq. (4). The perpendicular velocity is: ——— $v_\perp = 0.05$, - - - - - $v_\perp = 0.1$, ····· $v_\perp = 0.15$.

before performing the final $k_\perp$ and $k'_\parallel$ integration; they include the integration over ϕ'. With increasing $v_\perp$ the peaks become broader and slightly Galilei-shifted towards higher emission energies.

The curves in fig. 2 show the integrand of the final integration over $k_\perp$ as a function of $k_\perp$ for different values of $v_\perp$ and for the same collision system and parameters as fig. 1. The emission energy is $\omega = 64$ a.u.. The range of $k_\perp$ values covers the tungsten conduction band ($k_F = 0.69$). For increasing $k_\perp$ the emission probability first increases due to the increasing transmission of the jellium wave functions through the potential barrier situated at the surface. Close to k_F it decreases to zero according to the more and more limited range of possible $k_\parallel$ vectors ($k_\parallel^2 + k_\perp^2 \leq k_F^2$).

The doubly differential emission probability (differential in energy and emission angle) is shown in fig. 3 for three values of $v_\perp$, polarization along $v_\parallel$ and the same system as in figs. 1 and 2. The width of the spectrum corresponds to the width of the shifted conduction band. Due to the perpendicular projectile motion, there is no sudden cutoff corresponding to the limits of the shifted band. Because of the constant density of jellium states used in eq. (4), the spectrum in fig. 3 does not show any internal structure.

The emission probability decreases for increasing $v_\perp$ in agreement with a decreasing collision time. However, this feature is partly balanced by a decreasing distance of closest approach $b(v_\perp)$.

In an improved description of the conduction band a k-dependent density of states could be used. Such a more realistic model-density of states would appear as a weight-factor inside the integral in eq. (4). Its influence on the emission probability is expected to be more important than corrections induced by the use of more sophisticated (distorted) initial-state wave functions, since the more detailed information contained in those wave functions is, to a certain amount, averaged out in the calculation of the transition amplitude. Correspondingly, structures in measured spectra could be used to check calculated densities of states.

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Erratum

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In fig.3 the differential probability for photon emission appeared with an incorrect magnitude. The corrected figure is shown below. We now conclude that the emission probability is large enough for experimental investigation e.g. for $v_\perp = 0.1$ a.u. it reaches a peak of $2.5 \cdot 10^{-6}$ a.u. at $\omega = 63$ a.u..

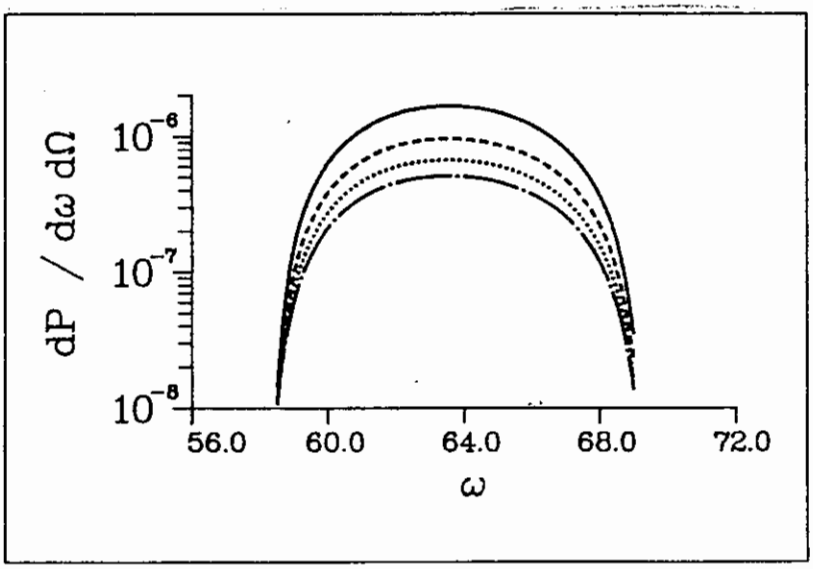

Fig.3.: REC spectrum for emission perpendicular to the surface for capture in the ground state of O^{8+} colliding with a tungsten surface at $v_\parallel = 8$ a.u. . The perpendicular velocity is _____ $v_\perp = 0.05$; - - - - - $= 0.1$; . . . . . $= 0.15$; _ . _ . _ . _ $= 0.2$ (in a.u.).