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Transport model for hot positrons in layered structures

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The transport of hyperthermal, monoenergetic ($\leq$ 10-eV) positrons injected in metal bilayer structures is investigated. The transport is modeled using the Boltzmann equation and the two-flux approximation. Analyzing reported experimental data in terms of the developed model enables us to separate the different scattering channels and to estimate the mean free paths for these events. Our study is the first quantitative treatment of hot positrons, and the extracted transport parameters agree with theoretical predictions.

During the past decade there has been extensive interest in studying surface and near-surface phenomena with monoenergetic positron beams (for a recent review see Ref. 1), primarily with energies of the order of keV. Thin layered structures ($\leq$ 10 monolayers) studied by positrons have been Cu/W(110) (Ref. 2) and SiO_2/Si.[3] Recently, Gidley and Frieze[4] have demonstrated how the potential experienced by a *thermalized* positron at the interface of two materials can be used to create essentially monoenergetic positrons of very low energy (of the order of a few eV). Another possible way to adjust positron energies within a crystal in a controlled manner is to use electric fields. In this Report we present a model for the attenuation of hot positrons in a bilayer structure. It is based on solving the Boltzmann transport equation in one dimension using the two-flux formulation. The scattering probability function consists of two parts, the elastic and the inelastic channels. The model is used to interpret the recent experimental results[4] and to extract the first quantitative estimates for the inelastic and elastic mean free paths.

As electrons and positrons are in many respects alike, it is natural to follow an approach developed for hot-electron phenomena.[5,6] Hot electrons [$k_B T \ll E_{\rm kin}$ < (several tens of eV)] have recently been studied extensively, the motivation coming from applications in semiconductor devices.[7] In many cases the electron-solid interaction can be treated within the semiclassical Boltzmann equation. This allows one to study electron transport parameters and their energy dependence in different materials. On the other hand, thermalized positron motion has almost exclusively been treated by the diffusion theory (for a review see Ref. 8), neglecting any effects due to epithermal positrons.[9–11]

Figure 1 summarizes schematically the potential sensed by a positron at a solid surface covered by an overlayer. We consider positrons that have been implanted from the vacuum with an energy high enough so that the positrons are stopped well behind the overlayer and are fully thermalized before reentering the surface region by diffusion. Typical time scales for thermalization and diffusion are < 10 ps and > 100 ps, respectively.[12] Thermal positrons experience at the interface a potential difference ($\Delta\chi_+$) which is equal to the difference between the energies of the lowest positron bands in both sides of the interface. The common reference for the energy scales is obtained from the requirement that the electron Fermi energy is the same for both sides of the interface. In the case shown in Fig. 1 the positron affinity is larger in the overlayer than in the substrate and the positrons gain a kinetic energy large compared to thermal energies. The positron momentum becomes strongly directed towards the normal of the interface. These hot positrons interact with the overlayer tending to rethermalize through inelastic collisions. At low coverages a fraction of positrons reach the outer surface before any energy losses. If the positron work function is negative (as it is for many clean metal surfaces[1]), positrons are spontaneously emitted into the vacuum, and their energy can be analyzed. By following the attenuation of these essentially monoenergetic positrons as a function of layer thickness, as was demonstrated in Ref. 4, information on positron-overlayer interaction can be obtained.

For elemental metals the positron affinity differences $\Delta\chi_+$ are generally well below 10 eV.[13] In this energy re-

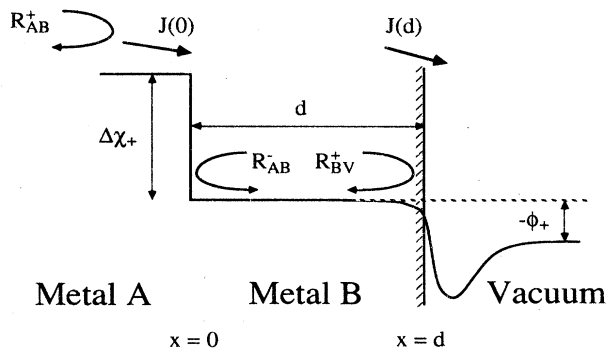

FIG. 1. Schematic picture of the potential experienced by a positron in a thin-overlayer structure.

gion there are two dominant scattering mechanisms for positrons in a homogeneous material:[12] purely inelastic electron-hole pair excitations and quasielastic acoustic-phonon scattering. Since single scattering events cannot be experimentally resolved in the latter case, we will treat it as an essentially elastic process. Other possible but less probable processes in well-characterized systems are elastic impurity scattering from foreign ions and positron trapping to vacancy-type defects. At the highest energies (~ 10 eV) also inelastic plasmon excitations may play a minor role.

In the semiclassical approach positrons are treated as pointlike particles with well-defined positions. This limits the applicable energy and thickness ranges. The positron wavelength should be smaller than both the overlayer dimensions and the mean free path between successive collisions. For these reasons the presented model is applicable to positrons with kinetic energy more than ~ 0.1 eV (wavelength ~ 35 Å). On the other hand, the standard diffusion model[1] is only applicable for length scales large compared to typical mean free paths (i.e., tens of Å) and for fully thermalized positrons.

Let us now consider the motion of a hot, initially monoenergetic positron distribution in a layer of thickness d (Fig. 1). We denote the space- and energy-dependent positron distribution function by $I(E,\mu,x)$, where E stands for kinetic energy, x for distance from the interface, and μ for the direction cosine. The initial distribution injected to the overlayer from the interface is expressed as $I(E,\mu,0)=I_0(\mu)\delta(E-\Delta\chi_+)\delta(x-0)$. In the absence of any electric fields the stationary Boltzmann equation can be written as

$$\frac{\partial I(E,\mu,x)}{\partial x}=\tfrac{1}{2}\int_{-1}^{1}d\mu'\int_0^{\infty}dE'[W(\mu'\rightarrow\mu,E'\rightarrow E)I(E',\mu',x)-W(\mu\rightarrow\mu',E\rightarrow E')I(E,\mu,x)]+J(E,\mu)\delta(x-0^+)\ , \tag{1}$$

where $W(\mu'\rightarrow\mu,E\rightarrow E')$ is the scattering probability function and $J(E,\mu)\equiv\mu I(E,\mu,0)$ is the injected distribution.

We solve the transport equation (1) using the two-flux approximation, a kind of two-state model for the positron current density. The positron current is divided into two components, one $[I_+(x)]$ moving to the direction of positive x and the other $[I_-(x)]$ to the negative direction. Scattering processes are assumed to be isotropic, which is a reasonable approximation in the considered energy range. The positron injection into the overlayer material is homogeneous and practically perpendicular to the interface ($\Delta\chi_+ \gg kT$). Also positron emission from the external surface is directed normal to the surface. It follows that the scattering probability can be written as

$$W(\mu\rightarrow\mu',E\rightarrow E')=\delta(E'-E)/\lambda_{\rm el}(E)+\delta(E'-E+\Delta E)/\lambda_{\rm inel}(E)\ , \tag{2}$$

where $\lambda_{\rm el}$ and $\lambda_{\rm inel}$ correspond to the mean free paths for elastic and inelastic processes, respectively.

We follow the transport of those positrons gaining the energy $\Delta\chi_+$ at the interface, and no incoming particles from higher energies are included. In the two-flux approximation Eq. (1) reduces to two coupled linear differential equations,

$$\begin{aligned}\frac{dI_+(x)}{dx}&=\frac{I_-(x)-I_+(x)}{\lambda_{\rm el}}-\frac{2I_+(x)}{\lambda_{\rm inel}}+4J_0\delta(x-0^+)\ ,\\ \frac{dI_-(x)}{dx}&=\frac{I_-(x)-I_+(x)}{\lambda_{\rm el}}+\frac{2I_-(x)}{\lambda_{\rm inel}}\ ,\end{aligned} \tag{3}$$

where $\lambda_{\rm el}$ and $\lambda_{\rm inel}$ are the elastic and inelastic mean free paths of positrons with the kinetic energy $E=\Delta\chi_+$.

The boundary conditions depend on possible positron reflection at the interface and at the surface. These processes include the reflection of incoming particles from the interface potential (reflection coefficient R^+_{AB}), the reflection of particles scattered at least once which are trying to escape back to the substrate (R^-_{AB}), and the reflection from the surface potential (R^+_{BV}).[14] For very thin films these coefficients may depend slightly on coverage but we assume them to be constant. The ratio between $J_+(d)$ and $J_+(0)$ can be solved taking into account the reflections, and consequently

$$\frac{J_+(d)}{J_+(0)}=\frac{4\gamma_{\rm eff}(1-R^+_{AB})}{\Delta-\Gamma_+\lambda_{\rm el}R^+_{BV}\Delta-4\gamma_{\rm eff}R^+_{BV}\exp(2\gamma_{\rm eff}d)(R^-_{AB}-\Gamma_+\lambda_{\rm el})}\ , \tag{4}$$

where the effective scattering rate per unit length is

$$\gamma_{\rm eff}\equiv[\lambda_{\rm inel}^{-1}(\lambda_{\rm inel}^{-1}+\lambda_{\rm el}^{-1})]^{1/2}\ . \tag{5}$$

Γ^+ and Γ^- are defined as

$$\Gamma_\pm\equiv\lambda_{\rm el}^{-1}+2\lambda_{\rm inel}^{-1}\pm 2\gamma_{\rm eff} \tag{6}$$

and Δ denotes the expression

$$\Delta \equiv \Gamma_+ \exp(2\gamma_{\text{eff}} d) - \Gamma_- \exp(-2\gamma_{\text{eff}} d) - \frac{2R_{\bar{A}B}^{-} \sinh(2\gamma_{\text{eff}} d)}{\lambda_{\text{el}}} . \tag{7}$$

In order to make the functional dependencies clearer we specify the reflection probabilities. R_{BV}^{+} is practically zero because only (quasi)elastically scattered hot positrons are followed, and their kinetic energy is well above any (image) potential levels at the surface.[14] R_{AB}^{+} is constant for thick films and in the first approximation can also be considered to be zero. This leaves open only the value for the reflection probability $R_{\bar{A}B}^{-}$ at the interface. The ratio $J_+(d)/J_+(0)$ reduces to

$$\frac{J_+(d)}{J_+(0)} = \frac{4\gamma_{\text{eff}}}{\Gamma_+ \exp(2\gamma_{\text{eff}} d) - \Gamma_- \exp(-2\gamma_{\text{eff}} d) - 2R_{\bar{A}B}^{-} \sinh(2\gamma_{\text{eff}} d)/\lambda_{\text{el}}} . \tag{8}$$

Gidley and Frieze have studied Cu overlayers epitaxially grown on Ni substrate. They reported data on the transmission of the elastic positrons. These data are reproduced from Ref. 4 and shown by black dots in Fig. 2. Equation (8) can now be used to model the experimental data. It has two or three free parameters, depending on how the reflection coefficient $R_{\bar{A}B}^{-}$ is treated. The solid line shown in Fig. 2 corresponds to the least-squares fit with $R_{\bar{A}B}^{-}$ equal to zero. Figure 2 shows that Eq. (8) fits the data very well, and shows in particular that two length scales (scattering channels) must be active. The two mean free paths obtained this way are $\lambda_{\text{inel}} \approx 300$ (100) Å and $\lambda_{\text{el}} \approx 20$ (5) Å, which are in agreement with theoretical values[12] for electron-hole pair excitation, and longitudinal-acoustic-phonon scattering, respectively.

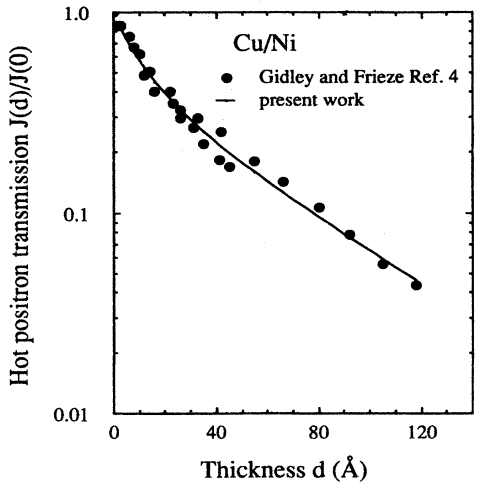

FIG. 2. Fraction of elastically transmitted positrons as a function Cu overlayer thickness on a Ni substrate. Experimental values (black dots) are reproduced from Gidley and Frieze (Ref. 4). Solid line corresponds to a fit to Eq. (8) with reflection coefficient $R_{\bar{A}B}^{-} = 0$.

These values describe the experimental data relatively well when $0 \leq R_{\bar{A}B}^{-} \leq 0.8$. For higher reflectivities λ_{el} tends to become unphysically large. Extracting reliable estimates for λ_{inel} from the experimental data is rather difficult, since the thickest overlayer is only 120 Å. In order to get more accurate estimates data at thicker coverages are required. The potential difference at the Cu/Ni interface is, according to the observations of Gidley and Frieze,[4] $\Delta\chi_+ = 0.5$ eV, which is in good agreement with the calculations of Puska *et al.*[13] For this energy the formulas from Nieminen and Oliva[12] predict mean free paths for acoustic-phonon scattering in copper $\lambda_{\text{el}} \approx 35$ Å and for electron-hole pair excitations $\lambda_{\text{inel}} \approx 350$ Å. Anyway, it should be emphasized that the nonexponential behavior can also be affected by the experimental uncertainties or by the pseudomorphic growth of the overlayers (Ref. 15).

The separation to different scattering channels enables us to recognize any new channels opening when conditions within the overlayer material are changed. An example of this is to study impurity scattering by following the signal from the elastic processes. This opens up a possibility to examine interdiffusion phenomena associated with annealing of layer structures at elevated temperatures. Recently, Kong and Lynn[16] have estimated yields of epithermal positrons in semi-infinite geometry using same kind of transport formulation. In studies of defects in heteroepitaxial structures rethermalization must also be considered.[17]

In conclusion, we have successfully used the Boltzmann equation for positrons to extract first experimental estimates for the mean free paths of different scattering channels. Hot-positron physics is rapidly developing in analogy to hot electrons. There are many interesting problems in positron solid-state physics where the present modeling and the extracted values for mean free paths are useful.

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