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Abstract

In oxide coating metals, localized states exist, which strongly hybridize with conduction electrons forming interface states (IS). This hybridization yields several qualitative and quantitative new effects typical for oxide coated metals. The Hamiltonians for these new effects describe the:
- enhanced coupling of IS to phonons,
- strengthened and weakened superconducting interaction of IS,
- additional - resonant - tunnel channels via IS.

1. Introduction

Localized electron states in oxides adjacent (< 5 nm) to a metal hybridize with the conduction electrons forming interface states (IS). IS have been proposed as cause for:
- rf and phonon interface losses;[1]
- strengthened and weakened superconducting interaction;[2]
- tunnel anomalies in the normal and superconducting states.[3,4]

As shown by these proposals, the electron-phonon interaction (EPI) of IS and their observation by tunneling is of primary importance for the understanding of metal oxide interfaces. In this paper we analyze the electron-phonon interaction (EPI) and non-phonon contribution in the presence of localized states and resonant tunneling (RT) via such states. This includes the formulation of the Hamiltonian describing EPI (Sec. 2), appropriate for the enhanced EPI of IS. The superconducting state is affected also by non-phonon mechanisms caused by the presence of localized states as discussed in Sec. 3. In Sec. 4 a specific tunnel Hamiltonian is presented able to describe RT including EPI. Inelastic processes are discussed in Sec. 5.

2. Effect of Hybridization on Electron-Phonon Coupling

Inelastic tunneling as well as superconducting interaction is affected by the enhanced strength of electron-phonon interaction (EPI) in interface states (IS). According to Ref. 3, the hybridization of localized electrons $\phi_\ell(r, R_\ell)$ and conduction electrons of the metal ($x \leq 0$) decaying ($\sim\exp(-\kappa x)$) into the oxide yield as IS:

$$\psi_E = a(E)\,\phi_\ell + \int dE'\,b(E')\psi(E')$$

with:

$$a(E) = \frac{c}{\sqrt{\pi}}\,(E-\epsilon_\ell + \delta\epsilon_\ell + i\Delta_\ell(E))^{-1}; \quad |c| = 1 \qquad (1)$$

and $\Delta_\ell(E,x) = \Delta_\ell(E,0)\exp(-2\kappa x)$ with the decay constant $\kappa = \sqrt{2m(E_c-E_F)}/\hbar$, where E_c is the lower edge of the conduction band in the oxide and E_F is the Fermi energy. $\delta\epsilon_\ell$ and Δ_ℓ are the shift and the decay width of the localized state due to the hybridization with conduction electrons.

The effective constant describing the electron-phonon coupling can be noticeably affected by the formation of interface states. In order to study this problem, we use the general expression for λ, obtained in the adiabatic theory[5]

$$\lambda = \frac{1}{2p_F^2}\int_0^{k_1} q\,dq\,\frac{\zeta u_s^2 q^2 \gamma_s(q)}{\omega_s^2(q)}; \qquad (2)$$

with the phonon momentum q, $k_1 = \min(2p_F, q_D)$ depending on Fermi momentum p_F and the Debye momentum q_D and with $\omega_s(q)$ being the adiabatic phonon frequency. The summation in Eq. (3) is taken over the different phonon branches, u_s is the sound velocity, $\gamma_s(q)$ is a slowly varying function of q and ζ is the Frohlich parameter

$$\zeta = |H'|^2\, mp_f/2\pi\hbar^4\, \omega_s(q)$$

where H' is the electron phonon matrix element (see below Eq. (5)). Hence the coupling constant λ (Eq. (2)) can be written in the form

$$\lambda = \zeta < \sum_s^3 u_s^2 q^2 \gamma_s/\omega_s^2(q) >_{aver} . \qquad (3)$$

Equations (2), (3) are analogous to the McMillan representation[6]

$$\lambda \sim \nu|H'|^2 <\omega^{-2}>. \qquad (4)$$

The presence of IS affects all the factors. The partial localization enhances the matrix element H'. Indeed, according to the BO approximation H' can be written in the form[7]

$$H'_{m\nu',n\nu} = \sum_k \frac{\hbar^2}{2M_k}\int \psi_m^*(\vec{r}, \vec{R})\, \phi_{m\nu'}(\vec{R})\, \frac{\partial\psi_n(\vec{r}, \vec{R})}{\partial\vec{R}_k}\,\frac{\partial\phi_{n\nu}(\vec{R})}{\partial\vec{R}_k}\, d\vec{r}\, d\vec{R}\ . \qquad (5)$$

where the summation is over the ions k. This expression is equivalent to the usual representation of EPI, which contains the change of the potential of the electron - ion interaction.[8]

In the representation of EPI in Eq. (5) it is obvious that delocalized states, which are characterized by a weak dependence of ψ_n on R, yield a small EPI: $H' \sim \partial\psi_n/\partial R_k \approx 0$. I.e., EPI for IS at E_F will be proportional to the localized part (Eq. 1). This increase of EPI with localization saturates and then decreases, because for $\Delta_\ell < \hbar\omega_{ph}$ and $\Delta_\ell < \Delta u^*$ nonadiabatic processes, and hence H' (Eq. 5), diminish (Δu^* is the correlation energy,[3,4]; usually $\Delta u^* \sim$ 10–50 meV).

A rough estimate for EPI can be obtained by the following consideration: Eqs. (1) and (5) yield $H' \sim a/x_\ell$, where x_ℓ is the distance of the localized state to the metal. If the concentration of the localized states[4] up to a distance $x_\ell \sim 0.5$ nm is about $n_\ell \sim 10^{20}/cm^3$, this can enhance the EPI up to 100% compared with EPI of the bulk metal, e.g. Al.

Expression (4) contains also the density of states. The increase of λ can be caused by the size quantization due to the granular structure of the interface region. For example, the tunnel junction Al/Al oxide/Pb prepared in ultrahigh vacuum[9] is characterized by the formation of Al oxide crystals. As is known, the size quantization leads to an increase of ν.[10] In addition, the phonon frequency can be affected by the stresses between metal and oxide.

3. Non-Phonon Interaction

Beside the above discussed interaction strengthened in IS as compared to bulk electrons, virtual transitions of localized electrons "ℓ"[11,12] (see Fig. 1) caused by interactions with delocalized – conduction – electrons "c" result in an additional effective attraction of "c" electrons.

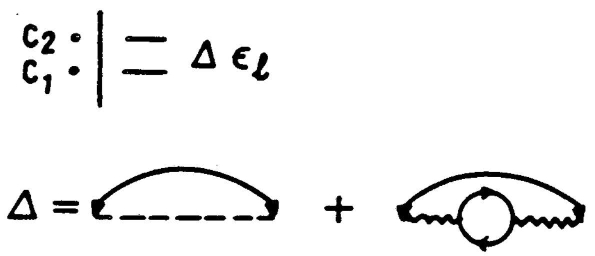

Fig. 1. Non-phonon interaction: a) C_1, C_2 are the conduction electrons, $\Delta\epsilon_\ell$ corresponds to the localized states; b) self-energy part describing the pairing; second term is due to the non-phonon interactions.

The superconducting order parameter contains an additional term and can be written in the form

$$\Delta_\lambda = \sum_{\lambda_1,\omega_n'} \Gamma_{\lambda_1,\lambda} F_{\lambda_1,\omega_n'} \quad , \tag{6}$$

where $\omega_n' = (2n'+1)\pi kT$ and $F_{\lambda_1,\omega_n'} = \Delta_{\lambda_1}(\omega_n'^2+\xi_{\lambda_1}^2+\Delta_{\lambda_1}^2)^{-1}$ is the abnormal Green's function. $\Gamma_{\lambda_1\lambda} = g^{cp}_{\lambda_1\lambda} + \Gamma^{c\ell}_{\lambda_1\lambda}$ is the total vertex, which contains the usual term $g^{cp}_{\lambda_1\lambda}$ describing EPI and an additional term

$$\Gamma^{c\ell}_{\lambda_1\lambda} = \sum_{\lambda',\lambda''} V^c_{\lambda_1\lambda'} V^c_{\lambda''\lambda} \Pi_{\lambda'\lambda''} \quad , \tag{7}$$

where $V^{c\ell}_{\lambda\lambda'}$ is the matrix element of the Coulomb interaction between the conduction and the localized electrons; $\Pi_{\lambda'\lambda''} = (n_{\lambda'}-n_{\lambda''})/(\epsilon^\ell_{\lambda'}-\epsilon^\ell_{\lambda''} + \hbar\omega)$; ϵ^ℓ_λ are the energy levels of the localized electrons and $\hbar\omega$ is the transferred energy. The constant $g^{c\ell}$ describing the non-phonon mechanism depends strongly on the energy spacing $\Delta\epsilon^\ell$; for $\Delta\epsilon^\ell \gg \Omega_D$ (= Debay frequency) the method described in Ref. 12 yields $g^{c\ell} = c_\ell V/\Delta\epsilon_\ell$, where c_ℓ is the normalized concentration of localized states ($c_\ell \sim n_L/n$) and $V = (e^2/q^2)^2\, mp_F^4\, I$ holds with $I = \int \exp(i\vec{q}\vec{r}).\ \chi_\lambda(\vec{r})\ \chi_{\lambda'}(\vec{r}) = (p_F r_o)^2$ ($q \sim p_F$ is the transferred momentum, χ_λ is the localized wave function extended over a radius r_o). The increase of T_c due to this additional coupling can be estimated for $g^{c\ell} \ll g^{EPI}$ on the basis of the step method.[12] One obtains $T_c = T_{co}\exp(g^{c\ell}/g^{EPI})$. For values $\Delta\epsilon^\ell \sim 0.5$ – 1 eV and $c_\ell \sim 10^{-2} - 10^{-3}$ one can obtain $\Delta T_c/T_{co} \approx$ 1/10. The above results can be qualitatively applied to partly localized states, namely IS housing more than 1 electron. Thus, the non-phonon interaction due to the presence of IS, results in an additional contribution to the Cooper pairing, as worked out above, and compensates partly for the degradation due to pair weakening discussed in Ref. 2.

Note that the existence of the localized states and their hybridization with the conduction electrons is similar to the situation in heavy fermion systems like UBe_{13} (see, e.g.[13,14]).

The question arises: How one might separate the contribution of the usual EPI and the non-phonon mechanism. According to Ref. 15, one can use two methods of the reconstruction of the function $g(\Omega) = \alpha^2(\Omega)\, F(\Omega)$. One of them is the usual method based on the inversion of the Eliashberg equation.[16] As a second method, the point-contact spectroscopy[17] or the measurements of the electronic heat capacity can be used. If the superconducting state is caused by EPI, both methods give the same result. However, if a non-phonon mechanism makes a noticeable contribution, there will be a discrepancy between these two pictures.

4. Resonant Tunnel Hamiltonian

IS and their enhanced EPI can be detected most directly by tunneling through the strong increase of the tunnel current at the resonant energy ϵ_ℓ of IS, as discussed in Ref. 3. In order to discuss the change of resonant tunneling (RT) by inelastic effects or, e.g., by finite temperature, the treatment based on the stationary Schrödinger equation is not well suited. These processes are more easily described by the method of the tunnel Hamiltonian:[18]

$$\hat{H}_T = \sum_{i,f} T_{fi} a_f^+ a_i \tag{8}$$

where "i" and "f" denote the initial and the final states. As will be shown below, RT yields for H_T:

$$\hat{H}_T = \hat{H}_T^D + \hat{H}_T^R \text{ or } T_{fi} = T_{fi}^D + T_{fi}^R . \tag{9}$$

Here T_{fi}^D describes the direct tunnel channel[15] and

$$T_{fi}^R = \frac{T_{f\ell}\, T_{\ell i}}{E-\epsilon_\ell+\delta\epsilon_\ell+i\Delta_\ell} \tag{10}$$

holds for RT, where "ℓ" corresponds to the localized state; $\delta\epsilon_\ell$ abd Δ_ℓ are the shift and the decay width of the state due to the hybridization with the adjacent conduction electrons – see Eq. (1) or Ref. 3. Consider a barrier containing a localized state ℓ sketched in Fig. 2. Following Ref. 18, we introduce the wave function $\psi_i(r)$, which is the solution of the Schrodinger equation in the region $x < a$ and which is matched to an exponentially decreasing function in the barrier region $x > a$ (Fig. 2). The function ψ_i is thus a correct solution of the Schrodinger equation for $x < a$ and almost everywhere in the barrier. This ψ_i is not a correct solution for $x > d$ and in the small region x_{bc} housing the localized state. The function ψ_f, being a solution for $x > d$, can be written in a similar way. In addition, let us introduce the wave function ψ_ℓ of the localized state, which decreases exponentially for $x < b$ and $x > c$ - see Fig. 2.

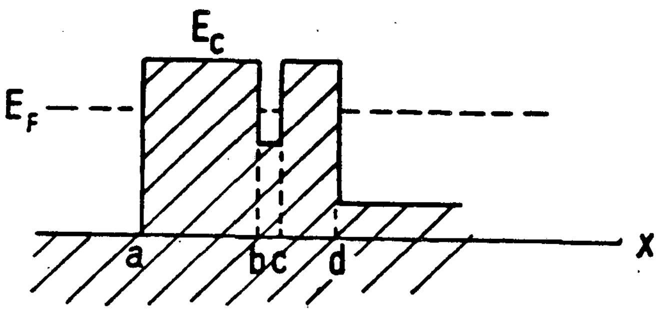

Fig. 2. Resonant tunneling; sketch of the barrier.

Assuming the initial electron ψ_i to decay into ψ_e and ψ_f, one can ask for the total solution of the Schrödinger equation $i\partial\psi/\partial t = \hat{H}\psi$ in the form:

$$\psi(t) = c_i(t)\ \psi_i\ e^{-iE_i t/\hbar} + c_\ell(t)\ \ \psi_\ell e^{-iE_\ell t/\hbar} + c_f(t)\ \psi_f\ e^{-iE_f t/\hbar} \quad . \tag{11}$$

Substituting Eq. (11) into the Schrodinger equation, we arrive after some manipulations at the following equations:

$$i\dot{c}_k = \sum_{n=i,\ell} c_n A_{kn} \exp(-(E_n - E_k)t/\hbar);\ (k = \ell, f)\ , \tag{12}$$

where the matrix element A_{kn} are given by:

$$A_{kn} = \int \psi_k^* \ (\hat{H} - E_n)\ \psi_n\ d^3\vec{r}. \tag{13}$$

These Eqs. (11-13) assume $c_f << c_i$ - see also Refs. 18 or 19. Based on Eqs. (12) and (13) we obtain:

$$i\ c_f(t) = T_{fi} \exp(-i\ (E_i - E_f)t/\hbar)\ , \tag{12'}$$

with

$$T_{fi} = T_{fi}^D + T_{fi}^R$$

$$T_{fi}^D = A_{fi}\ , \tag{14}$$

$$T_{fi}^R = A_{f\ell}\ A_{\ell i}/(\bar{E}_i - E_\ell + i\Delta_\ell)\ ,$$

with $\bar{E}_\ell = E_\ell + \delta\epsilon_\ell$; $\delta\epsilon = \mathrm{Re}A_{ff}$; $\Delta = \mathrm{Im}\ A_{ff}$ where $\delta\epsilon_\ell$ and Δ_ℓ describe the shift and the broadening of the level E_ℓ due to the hybridization - see Eq. (1). The first term in Eq. (14) describes the direct tunneling as obtained by J. Bardeen[18] - see also Ref. 19. The second term describes tunneling via the localized state being resonant at $E_i \approx \bar{E}_\ell$.[3,20] The total tunneling probability is given by Eq. (12') and according to the Golden rule we obtain:

$$dW_{i\to f} = \frac{2\pi}{\hbar}\ |T_{fi}|^2\ \delta(E_i - E_f)\ d\nu\ .$$

The resonant term makes a noticeable contribution to tunneling for $E_i \approx \bar{E}_\ell$ (Eq. (14)). The resonant contribution T_R depends on the barrier thickness d roughly like $\exp(-\kappa d)$,[3,4] i.e. more slowly than the direct tunnel contribution $T^D \sim \exp(-2\kappa d)$. Hence T^R will dominate with increasing d over T^D, as discussed in Ref. 3. For $T^R \sim T^D$ the interference between both will give rise to interference patterns space or energy wise.

In summary, the total tunnel Hamiltonian

$$\hat{H} = \sum_{i,f} T_{fi}\ a_f^+\ a_i$$

is given by:

$$\hat{H}_T = \hat{H}_T^D + \hat{H}_T^R:$$

$$\hat{H}_T^D = \sum_{i,f} T_{if}^D\ a_f^{+} a_i; \quad \hat{H}_T^R = \sum_{i,f} T_{if}^R\ a_f^{+} a_i. \tag{15}$$

It is worth noticing that RT is not bound by the same selection rules as direct tunneling.

In the paper[22] the method allowing to separate the direct and resonant channels, has been proposed. According to the analysis[22] RT dominates if $eU < 50$ meV.

5. Inelastic Processes and Tunneling

Tunneling through a barrier is accompanied by different inelastic processes, which enhance the tunnel current by opening new decay channels. Excitation of vibrations of molecules, phonons are examples for such a process. This is the foundation of the well known method of vibrational molecular spectroscopy.[23] In the presence of inelastic processes the total tunnel Hamiltonian (Eq. (9)) contains an additional term H^i.

$$H_T^i = H_T^{Di} + H_T^{Ri} \tag{16}$$

with

$$T^{Di} = T^D + \sum_\Omega T_\Omega^D; \quad T^{Ri} = T^R + \sum_\Omega T_\Omega^R \tag{17}$$

and $\Delta\epsilon = E_i - E_F$ is the transferred energy. The matrix element T^D depends on the overlap of the wave function ψ_i, ψ_ℓ and ψ_f and the interaction with the sub-system. An explicit expression for T^D has been obtained in Ref. 24 for the radiation of phonons and in Ref. 25 for vibrational excitations of molecules.

The tunnel current contains then the elastic terms proportional to $(T_T^D + T_T^R)^2$, the inelastic terms $\propto (\Sigma\ T_\Omega^D + \Sigma\ T_\Omega^R)^2$ and the interference terms $\propto (T_T^D + T_T^R)\ (\Sigma\ T_\Omega^D + \Sigma\ T_\Omega^R)$. According to Ref. 24, the last term gives the largest contribution. For tunnel barriers showing large resonant tunneling, the enhanced EPI of quasilocalized states - see Sec. 2 - enhances the interface phonon signal in the tunnel characteristic by modifying, e.g., the phonon spectrum deduced from the usual tunneling. RT results in an appearance of the additional channel, namely: for $\Delta x > 0.5$ nm the localized states become charged during the time

$\tau_\ell(x) = \hbar/2\pi(\Delta_\ell(x))$ (see Eq. (1)). Thus during this time $\tau \lesssim 10^{-12}$sec the electrons and the localized states attract each other and they are able to radiate phonons. These inelastic processes may be related to the measured phonon distribution in amorphous Si, Ge barriers.[26]

Summary

In this paper interaction mechanisms of interface states, caused by the hybridization of localized states with conduction electrons in oxides adjacent to metals, have been discussed. The main results can be summarized as follows:

- The presence of interface states results in an increase of the electron phonon coupling due to partial localization at the surface. This increase affects the superconducting properties and enhances the phonon structure in tunnel current.
- The presence of the interface states results in an appearance of a nonphonon interaction mechanism, which influences the superconducting interaction.
- Interface states cause resonant tunneling. For this resonant tunneling a tunnel Hamiltonian has been developed in the representation of second quantization.
- Inelastic processes are noticeably affected by the resonant tunneling.

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