Spectroscopy of electron-phonon interaction in point contacts with a barrier layer

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It is shown that the spectrum of electron-phonon interaction in metallic point contacts with an extremely small inelastic relaxation length, $\lambda_\epsilon < d$, can be analyzed if the transmission D of the barrier layer in the point contact is low: if it satisfies the relation $Dd < l_i$ (l_i is the elastic scattering length of the electron in the metal and d is the contact size).

Metallic point contacts with the narrow-part dimension $d \lesssim 10^2$–10^3 Å make it possible to study the energy dependence of the time required for inelastic scattering of electrons by elementary excitations in metals.[1,2] The point-contact spectrum $S(V) = [(1/R)(dR/dV)]$ [$R(V)$ is the contact resistance] usually consists of three composite elements (Fig. 1a): the spectral part A proportional to the electron-phonon (or electron-exciton, electron-magnon, etc.) interaction function $g(\omega)$ (Ref. 3), the background B—a smoothly varying part of the $S(V)$ curve beyond the boundary of the elementary-excitation spectrum (we will be dealing here with phonons), which stems from the rescattering of electrons by nonequilibrium phonons which are reabsorbed near the contact,[4] and a recurrence of the phonon spectrum C at the multiple and combination frequencies.[5,6] But some materials, in particular, the d metals, which usually form on their surfaces thin, strong, insulating oxide films, frequently reveal other types of spectra[2] (Fig. 1b) which are characterized by a lower total intensity, the absence of a background, and more pronounced two-phonon structural features.

Figure 1 shows two experimental tantalum point-contact spectra obtained for various Cu–Ta heterojunctions with approximately equal resistances at the same temperature and the same voltage of the second harmonic of the modulating signal along the ordinate scale. However, since at zero bias voltage, $V_1(0)$, the modulating voltages differ by a factor of 2.4, the amplitude of the point-contact spectrum (b) $S(V) = 2\sqrt{2}V_2(V)/V_1^2(0)$, reduced to the same value of $V_1(0)$, decreases by a factor of $(2.4)^2 = 5.7$. Spectrum (a) is, according to Ref. 7, typical of pure contacts of small dimensions ($d < l_i$). "Anomalous" spectrum (b), on the other hand, in addition to being a low-intensity spectrum, is characterized, as was already noted, by a considerably larger two-phonon peak in the absence of a background. Such a combination contradicts the theory of point-contact spectroscopy for contacts with a direct conductivity.

The behavior described above can be explained on the basis of the formation of a barrier layer at the boundary at which the solid metallic electrodes come in contact. The corresponding structures can be viewed as point-tunnel junctions which behave, if

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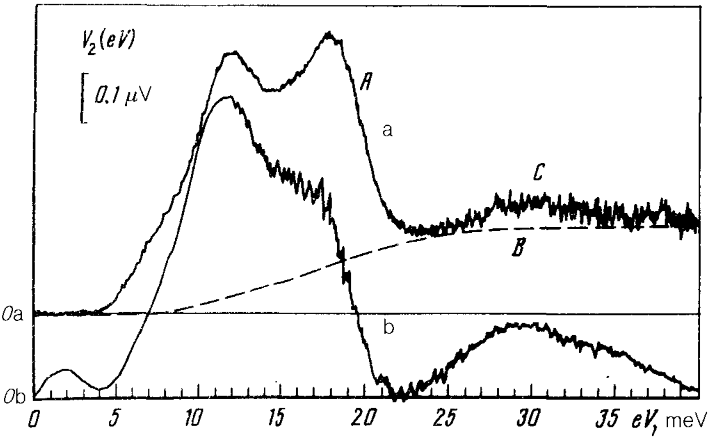

FIG. 1. Second harmonic of the modulating signal vs the bias voltage for the Cu–Ta heterojunctions, $T = 4.5$ K. The resistances of the contacts and the modulating voltages at $V = 0$ are (a) 85 Ω, 0.569 mV and (b) 120 Ω, 1.358 mV.

the relation

$$dD < l_i \tag{1}$$

is satisfied (D is the transmission level of the barrier associated with the insulating layer, and l_i is the path length of electrons in the scattering by impurities), as structures which differ from tunnel structures (planar structures which do not concentrate the current near the barrier layer) and from point-contact structures (which have a direct conductivity and which exhibit a nonlinear behavior because of the concentration of the field in a small region $d < \lambda_\epsilon$, where λ_ϵ is the length of the inelastic relaxation of electrons, $\lambda_\epsilon = \sqrt{l_i l_{ep}}$). Planar tunnel junctions also exhibit a nonlinear behavior[8] because of the scattering of electrons by the barrier-layer excitations. Such contributions to $S(V)$, which are small, have, however, the opposite sign with respect to the point-contact spectrum. The relatively small value of these contributions depends on the satisfaction of the condition (λ_B is the electron wavelength)

$$D > \frac{\lambda_B}{\min\{d, \lambda_\epsilon\}}, \tag{2}$$

which determines, jointly with inequality (1), the transmission interval, for which the dominant mechanism is the mechanism responsible for the nonlinearity of the current-voltage characteristic, which we are considering here.

The nonlinear behavior of the I–V characteristic of the junction stems from the multiple scattering of electrons in the metal near the barrier layer, i.e., it is an effect on the order of D^2, whereas the elastic current is proportional to the first power of the transmission. As applied to the planar tunnel junctions, the condition under which spectrum (1) is detected must be replaced by the relation $LD < l_i$ (L is the size of the sample), which would make the effect in question negligible.

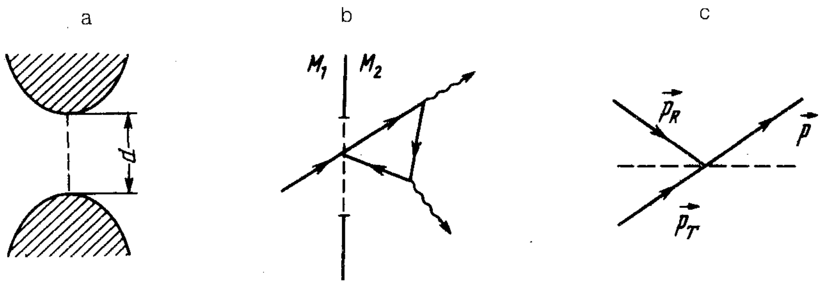

FIG. 2. (a) Theoretical model of the point contact; (b) schematic diagram of the inelastic processes in the electrodes; (c) electron momenta before the passage of the electron through the barrier in the transit regime ($\mathbf{p}_T$) and in the elastic-reflection regime ($\mathbf{p}_R$); and (c) after the passage of the electron through the barrier ($\mathbf{p}$). The dashed line represents the barrier layer.

Condition (1) implies that the tunnel resistance of the barrier, R_t, is large compared with the resistance of the region in which the current flows through the point contact (Fig. 2a), allowing us to ignore the electric field in the metal. The electron-phonon relaxation in the metal is analyzed on the basis of the Boltzmann kinetic equation for the electron distribution

$$\mathbf{v}\frac{\partial f_{\mathbf{p}}}{\partial \mathbf{r}} - I_i\{f_{\mathbf{p}}\} = I_{ep}\{f_{\mathbf{p}}\}, \tag{3}$$

where I_i and I_{ep} are the integrals of the collision of electrons with impurities and phonons.

The boundary conditions corresponding to the scattering of electrons by the insulating point-contact boundary and to the time at which equilibrium is reached in the bulk of the conducting edges should be augmented with the condition under which electrons are injected through the tunnel barrier (the dashed line in Fig. 2b). This condition can be written in the form

$$f_{\mathbf{p}}(\epsilon_{\mathbf{p}}) = Df_{\mathbf{p}_T}(\epsilon_{\mathbf{p}} + eV) + (1 - D)f_{\mathbf{p}_R}(\epsilon_{\mathbf{p}}). \tag{4}$$

Here $\mathbf{p}$ is the momentum of the tunneled electron, and $\mathbf{p}_R$ is the momentum of the reflected electron (Fig. 2c). Using the explicit form of Eq. (3), we can identify the contribution to the point-contact current I which corresponds to the inelastic scattering of electrons in the contact[9]

$$I = I^{(0)} - \frac{2e}{(2\pi\hbar)^3}\int d^3r\int d^3p\,\alpha_{-\mathbf{p}}(\mathbf{r})I_{ep}\{f'_{\mathbf{p}}(\mathbf{r})\}, \tag{5}$$

where $I^{(0)}$ is the elastic current; the second term in (5) describes the return current produced as a result of the interaction of electrons with phonons at the contact edges and $\alpha_{\mathbf{p}}(\mathbf{r})$ is the probability that the nonequilibrium electron originating at the contact edge will reach the point ($\mathbf{r}$, $\mathbf{p}$). This probability satisfies Eq. (3) (at $I_{ep} = 0$) and boundary condition (4). If condition (1) is satisfied, integral I_{ep} in (3) and (5) can be linearized with respect to the nonequilibrium correction to the electron distribution. Calculation of the second derivative of the current from the voltage in this case leads to the point-contact spectrum which at $T \ll \omega_D$ can be written in the form

$$\frac{1}{R}\frac{dR}{dV} = \frac{8}{3}\frac{ed}{\hbar v_F} D\left\{ g_{pc}(eV)\frac{\lambda_\epsilon(eV)}{d+\lambda_\epsilon(eV)} + C(eV)\right\} \tag{6}$$

The quantity $g_{pc}(\omega)$ in (6) is the point-contact function of the electron-phonon interaction[3] with the form factor which is normalized to unity. The factor of $g_{pc}(eV)$ in the braces indicates that at $\lambda_\epsilon < d$ the effective phonon-generation volume decreases. The function $C(eV)$ describes the energy relaxation of electrons near the contact [when $\lambda_\epsilon \gg d C(eV) = 0$)]. This function also indicates that the principal spectrum is repeated at comparable intensities at the multiple and combination phonon frequencies. If, for example, a sharp peak in the phonon state density is present at the frequency ω_0, we find

$$C(eV) = \sum_{n=2}^{\infty} A_n \delta(eV - n\hbar\omega_0); \quad A_n \approx \Lambda\omega_0 \frac{\lambda_\epsilon}{d} \quad \text{for } \lambda_\epsilon < d, \tag{7}$$

where Λ is the electron-phonon coupling constant. The recurrence of the peaks in the point-contact spectrum is accompanied by their broadening. If, for example, the width of the main peak is Δ when $eV = \omega_0$, then its nth repetition will have a width $n\Delta$.

The applicability of relation (6) *does not require* that the inequality $d < \lambda_\epsilon$ be satisfied. This inequality, which is characteristic of point-contact spectroscopy without a barrier layer, does not allow the use of point-contact spectroscopy to study exotic materials (metals with a fluctuating valence and with heavy fermions, oxide superconductors) for which $l_i \approx 10$ Å. The use of tunnel point contacts therefore raises the possibility of studying electron-relaxation-time spectra with less rigorous constraints imposed on the contact diameter [relation (1)]. We note accordingly an effect, reported in Ref. 10, involving an anomalously slight ($\sim 30\%$) change in the point-contact resistance of compounds, whose valence varies over the bias-voltage range $0 < eV < 30$ meV. This effect occurs regardless of the strong temperature dependence of the resistance of these materials $R(T = 30 \text{ meV})/R(0) \approx 100$. Such an unusual behavior and the structural features which were detected in the point-contact spectrum, which mirror the spectrum of elastic[10,11] and inelastic (electron-phonon)[11] relaxation times, can be explained on the basis of our study as the result of the effect produced by the barrier layer at the point at which the metals come in contact in the heterojunction.

In conclusion we would like again to call attention to the fact that the operating regime of the point contact which we have analyzed here lies between the standard point-contact spectroscopy and the inelastic tunneling effect. The distinctive features of this regime are the negative sign of the nonlinear correction to the point-contact current, low background, and the strong many-phonon peaks in the point-contact spectrum. We should point out that in the case of inelastic tunneling the spectrum has an opposite (positive) sign and is characterized by a low many-phonon repetition rate proportional to $(\lambda_B / l_{ep})^n$.

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