# Microcontact spectroscopy of populations of two-level systems

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A study is reported of a nonlinear contribution to the current-voltage characteristics of a metal microcontact made by the scattering of electrons on low-energy excitations in the form of two-level systems. A new type of microcontact spectroscopy of populations of two-level systems is suggested: it would utilize the dependence of the occupation numbers of two-level systems on the voltage applied to a contact. A profile of a line in a microcontact spectrum—representing the dependence of $d^2I/dV^2$ on $V$—is asymmetric and consists of a central peak at a value of $eV$ equal to the excitation energy ($\lesssim 1$ meV) and of a wing in the range of energies exceeding the latter. The nonlinear component of the conductance exhibits a strong frequency dispersion in the range $f \sim 10^3$–$10^9$ Hz and this is due to relaxation of two-level excitations, which may be regarded as the distinguishing feature of this scattering mechanism.

## 1. INTRODUCTION

The progress made in microcontact spectroscopy employed in studies of phonon spectra of metals is well known.[1,2] This method is based on the following remarkable property of a microcontact: the distribution of electrons in the vicinity of a microcontact is of strongly nonequilibrium nature and the scale of nonequilibrium is determined by the applied voltage $V$ (Ref. 3). (In a sense the energy $eV$ then plays the role of the effective temperature of electrons at the contact.) Therefore, an increase of the voltage applied to a contact increases the phase volume of inelastic electron-phonon processes; as $V$ is increased, new groups of phonons of energies $\hbar\omega \leqslant eV$ begin to participate in such processes. This in turn results in a nonlinear dependence of the electron-phonon contribution to the contact resistance on the voltage $V$; the second derivative of the current-voltage characteristic $d^2I/dV^2$, known as the microcontact spectrum, provides information on the energy dependence of the electron-phonon interaction function. Obviously, this should also be true of other inelastic processes involving elementary excitations of different types, such as impurities with an internal degree of freedom.[3] Attention has been drawn recently to the possibility of using microcontact spectroscopy in studies of low-energy excitations (two-level systems) specific to disordered atomic structures.[4] A special attraction of such an experimental method is the circumstance that a microcontact can be used to select, because of its small size, a contribution of a small number or even a single two-level system.

An important feature of the interaction of two-level systems with electrons is that, in addition to the contribution of inelastic processes (when an electron transfers a two-level system to a different level), it includes also the contribution of elastic processes (when the scattering of an electron by a two-level system occurs without a change in the state of the latter). The relationship between the intensities of the corresponding processes is governed by the parameters of two-level systems, namely by the ratio $\Delta_0/E$ [$\Delta_0$ is the tunnel matrix element, $E = (\Delta_0^2 + \Delta^2)^{1/2}$ is the energy of a two-level system. $\Delta$ is the asymmetry of a two-well potential—see, for example, the review in Ref. 5] or by the relaxation time $\tau$ of two-level systems [$\tau^{-1} \propto (\Delta_0/E)^2$]. In other words, relatively rapidly relaxing two-level systems ($\Delta_0 \sim E$) participate primarily in inelastic processes, whereas slowly relaxing centers interact with electrons basically in an elastic manner. Therefore, the contribution of elastic processes to the nonlinearity of the current-voltage characteristics of a microcontact calculated in Ref. 4 and proportional to $(\Delta_0/E)^2$ does not in fact carry information on slowly relaxing centers. On the other hand, it follows from the expression for the distribution function of two-level systems, obtained using the Anderson-Halperin-Varma-Phillips model

$$P(\rho, E) = \bar{P}(1-\rho)^{-1/2}/\rho, \quad \rho = (\Delta^0/E)^2 \tag{1.1}$$

(see, for example, Ref. 5), it is the centers with the slowest relaxation that are most likely to be encountered.

It might seem that elastic processes are of no interest because two-level systems behave in these processes as ordinary impurities. However, the nontrivial circumstance is that the cross sections for the elastic scattering of electrons by two-level systems are generally different for two states, so that the contribution of elastic processes to the current across a contact depends on the occupation numbers of two-level systems[4]:

$$I_1 = -\frac{V}{R_0}\sum_j C_j(n_j^+\sigma_j^+ + n_j^-\sigma_j^-). \tag{1.2}$$

Here, $R_0$ is the resistance of a contact in the absence of two-level systems; $\sigma^+$ and $\sigma^-$ are the cross sections for the elastic scattering of electrons by such systems in the case of the upper and lower levels, respectively; $n^+$ and $n^-$ are the occupation numbers of these two levels; $C_j$ is a kinetic factor which is of the following order for two-level systems located in the region of a contact (Fig. 1):

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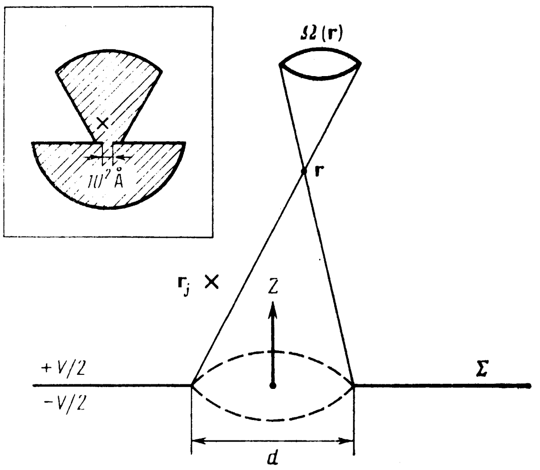

FIG. 1. Model of a microcontact in the form of a partition Σ impermeable to electrons with an aperture of diameter $d$; $r_j$ is the point at which a two-level system is located (identified by an arrow). The inset shows a realization of a contact in the needle-anvil form.

$$C_j \sim d r_j^{-4} \min(l, d). \qquad (1.3)$$

Here, $d$ is the characteristic size of the contact; $l$ is the mean free path of electrons; $r_j$ is the distance of a two-level system from the center of the contact.

For this reason two-level systems may, in particular, act as a source of low-frequency noise,[6,7] which in the case of a microcontact are analyzed in detail in Ref. 4.

In the present paper we wish to draw attention to the fact that the dependence of the contribution of elastic processes to the current on the occupation numbers of two-level systems provides an opportunity for microcontact spectroscopy of these systems which, in contrast to the traditional spectroscopy of inelastic processes considered in Ref. 4, is not limited to the case of rapidly relaxing centers. This opportunity is provided by the dependence of the occupation numbers on the voltage applied to a contact. In the case of two-level systems at a contact the electron system itself acts as a thermostat (interaction of the electron system with two-level systems characterized by $E < 5$–$10$ K is more effective than the interaction with phonons), so that the occupation numbers of two-level systems are governed by the nonequilibrium electron distribution function and, therefore, depend on $V$. This dependence results in a contribution to the nonlinearity of the current-voltage characteristic of a contact, i.e., it gives rise to a microcontact $d^2I/dV^2$ spectrum.

It follows from Eq. (1.2) that the relative change in the contact resistance (per one two-level system) at $r_j \sim d$ is

$$\Delta R/R \sim |\sigma^+ - \sigma^-|/d^2 \qquad (1.4)$$

in the ballistic regime,[1] i.e., when $l \gg d$, and

$$\Delta R/R \sim |\sigma^+ - \sigma^-| l/d^3 \qquad (1.5)$$

in the diffusion regime ($l \ll d$). When the contact diameter is $d \lesssim 10^2$ Å and we have $|\sigma^+ - \sigma^-| \sim 10^{-15} - 10^{-14}$ cm$^2$, this gives a value of $\Delta R/R$ of the order of 0.1–1% and this may be relevant in dealing with the singularities of microcontact spectra in the region of zero anomalies (low bias voltages corresponding to $eV \lesssim 1$ meV) reported in Refs. 8–10 and elsewhere and attributed to the scattering of electrons on magnetic impurities[11–13] or on hypothetical low-energy lattice excitations.[14]

Microcontact spectroscopy of the populations has another feature, namely a significant frequency dispersion of the nonlinear conductivity, manifested at low frequencies $\omega \sim 10^3$–$10^8$ sec$^{-1}$, due to slow relaxation of two-level systems. An analogous effect in the scattering of electrons by nonequilibrium phonons is discussed in Refs. 15 and 16.

## 2. NONLINEAR PART OF THE MICROCONTACT CURRENT

We shall begin with the kinetic equation for the occupation numbers of two-level systems interacting with an electron system at a microcontact[4]:

$$\frac{\partial}{\partial t}\left(\frac{n_j^+ - n_j^-}{2}\right) = -\frac{\pi}{2\hbar}\sum_{\mathbf{pp}'}\left(\frac{\Delta_{0j}}{E_j}\right)^2 |W_{\mathbf{pp}'}|^2$$
$$\times\{n_j^+[f_{p'}(1-f_p)\delta(\varepsilon_p-\varepsilon_{p'}-E_j)+f_p(1-f_{p'})\delta(\varepsilon_p-\varepsilon_{p'}+E_j)]$$
$$-n_j^-[f_p(1-f_{p'})\delta(\varepsilon_p-\varepsilon_{p'}-E_j)+f_{p'}(1-f_p)\delta(\varepsilon_p-\varepsilon_{p'}+E_j)]\}. \qquad (2.1)$$

Here, $W_{pp'} = V^+_{pp'} - V^+_{pp'}$; $V^\pm_{pp'}$ are the potentials of the interaction between two-level systems and electrons in two configurations of the systems; $f_p$ is the distribution function of electrons at the contact which in the ballistic case[3] is

$$f_p = f_0\left(\varepsilon_p + e\varphi + \frac{eV}{2}\eta_p\right), \qquad \eta_p = \begin{cases} \operatorname{sign} z, & \mathbf{v} \in \Omega(\mathbf{r}) \\ -\operatorname{sign} z, & \mathbf{v} \notin \Omega(\mathbf{r}) \end{cases} \qquad (2.2)$$

and in the diffusion case[17] is

$$f_p = {}^1/_2[f_0(\varepsilon_p + e\varphi - eV/2) + f_0(\varepsilon_p + e\varphi + eV/2)]$$
$$+ V^{-1}\varphi(\mathbf{r})[f_0(\varepsilon_p + e\varphi - eV/2) - f_0(\varepsilon_p + e\varphi + eV/2)]. \qquad (2.3)$$

Here, $f_0$ is the Fermi function and $\Omega(\mathbf{r})$ is the solid angle supported by the contact aperture at a point $\mathbf{r}$ (Fig. 1).

Equation (2.1) is readily reduced to

$$\partial N_j/\partial t + \Gamma_j N_j = B_j, \quad N_j \equiv n_j^+ = 1 - n_j^-, \qquad (2.4)$$

$$B_j = \frac{\Gamma_{0j}}{E_j}\left\{(1-q)\frac{E_j}{\exp(E_j/T)-1}\right.$$
$$\left.+\frac{q}{2}\left[\frac{E_j+eV}{\exp[(E_j+eV/T)]-1} + \frac{E_j-eV}{\exp[(E_j-eV)/T]-1}\right]\right\}, \qquad (2.5)$$
$$\Gamma_j = \frac{\Gamma_{0j}}{E_j}\left\{(1-q)E_j \operatorname{cth}\frac{E_j}{2T}\right.$$
$$\left.+\frac{q}{2}\left[(E_j+eV)\operatorname{cth}\frac{E_j+eV}{2T} + (E_j-eV)\operatorname{cth}\frac{E_j-eV}{2T}\right]\right\},$$

where

$$\Gamma_{0j} = \frac{\pi\Delta_{0j}^2}{E_j} N(0)\langle|W|^2\rangle_{FS}, \qquad (2.6)$$

$E_j$ is the excitation energy of two-level systems, $N(0)$ is the density of states of electrons on the Fermi surface, and $\langle\ldots\rangle_{FS}$ denotes averaging over the Fermi surface. The quantity $q$ is

$$q = {}^1/_2[1 - (2\varphi(\mathbf{r}_j)/V)^2]. \qquad (2.7)$$

In the case of two-level systems located at the center of the contact $[\varphi(r_j)=0]$, we have $q=1/2$ and far from a contact, we find that $q\to 0$.

The quantity $\Gamma_{0j}$ represents the characteristic relaxation frequency of two-level systems. It has been pointed out that these frequencies are very low, so that we are justified in ignoring the delay of establishment of a nonequilibrium electron distribution function of Eqs. (2.2) and (2.3) in the case when a voltage $V=V(t)$ varies with time and we can assume that $f_p$ is governed by the instantaneous voltage. This can be done simply by assuming that the frequency of a change in the potential satisfies the condition

$$\omega \ll \min(\omega_D,\ v_F/d) \tag{2.8}$$

where $\omega_D$ is the Debye frequency ($\omega_D \sim 10^{13}\ \mathrm{sec}^{-1}$) and $v_F/d$ is the reciprocal of the transit time of an electron across the accelerating voltage region (for the adopted values of the contact parameters, this last quantity is of the order of $10^{14}$ $\mathrm{sec}^{-1}$). On the other hand, the relaxation frequencies of two-level systems are known to be considerably less than $10^{13}\ \mathrm{sec}^{-1}$.

We shall first consider the static case: $V=$ const. Then, the number of two-level systems in the upper configuration is

$$N_j^0=\frac{B_j}{\Gamma_j}=\frac{1}{2}\Big\{1-\Big[E_j(1-q)\,\mathrm{cth}\,\frac{E_j}{2T}$$
$$+\frac{q}{2}\Big[(E_j+eV)\,\mathrm{cth}\,\frac{E_j+eV}{2T}+(E_j-eV)\,\mathrm{cth}\,\frac{E_j-eV}{2T}\Big]\Big]^{-1}E_j\Big\}. \tag{2.9}$$

This quantity approaches 1/2 for $eV \gg E_j$ and at $T=0$ it differs from zero only for $eV > E_j$.

The expression for the current (1.2) can be written in the form

$$I_1=-\frac{V}{R_0}\sum_j C_j[(\sigma_j^+-\sigma_j^-)N_j+\sigma_j^-]. \tag{2.10}$$

Hence, it is clear that for a given two-level system, we have

$$\left(\frac{d^2I}{dV^2}\right)_j \propto \frac{d^2}{dV^2}(VN_j),$$

i.e., the nonlinear part of the current-voltage characteristic is governed by the voltage dependence of the nonequilibrium distribution function $N_j$.

We shall represent the change in the microcontact spectrum in the form

$$\frac{1}{R}\frac{dR}{dV}=\sum_j \zeta_j S\left(\frac{eV}{E_j},\frac{T}{E_j}\right), \tag{2.11}$$
$$R=dV/dI.$$

Here,

$$\zeta_j=\frac{eC_j}{2E_j}(\sigma_j^+-\sigma_j^-)\,\mathrm{th}\,\frac{E_j}{2T}, \tag{2.12}$$

and the function $S(v,\tau)$ is described by

$$S=-\frac{d^2}{dv^2}\left[\frac{v\,\mathrm{cth}(1/2\tau)}{\psi(v,\tau)}\right],$$
$$\psi=(1-q)\,\mathrm{cth}\,\frac{1}{2\tau}+\frac{q}{2}\left[(1+v)\,\mathrm{cth}\,\frac{1+v}{2\tau}+(1-v)\,\mathrm{cth}\,\frac{1-v}{2\tau}\right]. \tag{2.13}$$

The line profile in a microcontact spectrum is shown in Fig. 2 for two-level systems located at the center of a contact ($q=0.5$) and far from it ($q=0.1$). In contrast to traditional microcontact spectroscopy,[1-3] in which the line is symmetric and has the width $\Delta_{1/2}(eV)=5.44T$, in the present case it is strongly asymmetric and has not only a peak at $eV=E_j$, but also a wing located at energies exceeding $E_j$. At $T=0$ the spectrum of two-level systems consists of a $\delta$-function contribution and a power-law term

$$S(v,\ 0)=q\delta(v-1)+2q(1-q)[1+q(v-1)]^{-3}\theta(v-1). \tag{2.14}$$

It should be noted that at all temperatures the function (2.13) is normalized by the condition

$$\int_0^\infty S(v,\tau)\,dv=1.$$

This line profile can be used as a distinguishing feature which makes it possible to separate the contribution made to a microcontact spectrum by two-level systems from the contributions of inelastic scattering by impurities with an internal structure (for example, excitation of an electron in the $f$

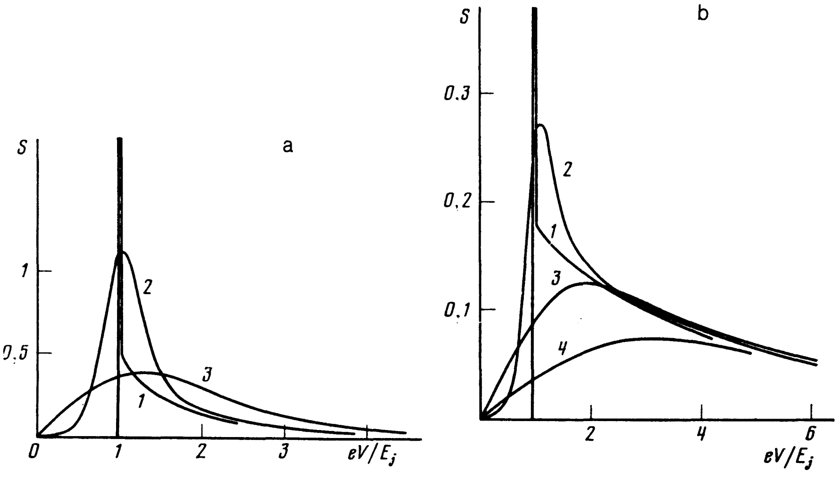

FIG. 2. Profile of a microcontact spectral line for $q=0.5$ (a) and $q=0.1$ (b). Curves 1, 2, 3, and 4 correspond to $T/E_j=0$, 0.1, 0.5, and 1.0, respectively.

shell[18]) and by paramagnetic impurities.[11]

It is interesting to note that the sign of the zeroth anomaly is, in accordance with Eq. (2.11), governed by the sign of the quantity $\sigma_j^+ - \sigma_j^-$ and, in principle, can be either positive or negative (although it is more natural to assume that the scattering by the upper level is stronger than by the lower level, i.e., that $\sigma_j^+ > \sigma_j^-$). In contrast to the scattering by impurities with an uncompensated magnetic moment (Kondo effect[11]), in which case we have a strong dependence of the intensity in the microcontact spectra on the magnetic field, when electrons are scattered by two-level systems there is no reason to expect any significant influence of an external magnetic field on the microcontact spectrum.

## 3. FREQUENCY DISPERSION OF THE CONDUCTANCE OF A MICROCONTACT

The general solution of Eq. (2.4) for an arbitrary periodic dependence $V(t) = V(t + T_0)$ is

$$N_j(t)=\exp[-\Omega_j(t)]\int_0^{T_0} dt' \tag{3.1}$$

$$\times\exp(\Omega_j(t'))$$

$$\times\left[\frac{\theta(t-t')}{1-\exp[-\Omega_j(T_0)]}+\frac{\theta(t'-t)}{\exp[\Omega_j(T_0)]-1}\right]B_j(t'),$$

$$\Omega_j(t)=\int_0^t \Gamma_j(t')\,dt',$$

where $\Gamma_j$ and $B_j$ are functions of the local value of $V(t)$.

From the experimental point of view the greatest interest lies in the response at the frequency of the second harmonic which appears on application to a contact not only of a static bias $V_0$, but also of a weak alternating signal $V_1 \cos\omega t$. This is due to the fact that it is the measurement of the second harmonic signal that provides the experimental method for the determination of microcontact spectra.[1,2] It is then naturally assumed that the frequency of the applied voltage is much less than the characteristic relaxation frequencies of the system. However, in the case of two-level systems we are dealing with objects for which the relaxation frequencies can be very low [since an exponentially wide spectrum of relaxation times follows from Eq. (1.1)]. An analytic expression for the second harmonic signal can be derived allowing for the finite nature of the relaxation times and this can be done by iteration of Eqs. (2.4) and (2.10) up to the second order in $V_1$.

The amplitude of the second harmonic of the current is proportional to $V_1^2$ and depends in a complex manner on $\omega/\Gamma_{0j}$, $eV_0/E_j$, and $T/E_j$. We shall give only the asymptotic expressions for the response at high and low frequencies. For example, if $\omega \gg \Gamma_{0j}$, we find that

$$\begin{gathered} I_2=I_2^c\cos 2\omega t+I_2^s\sin 2\omega t,\\ I_2^c=0,\\ I_2^s=\Gamma_{0j}\omega^{-1}R(eV_0/E_j,\ T/E_j), \end{gathered} \tag{3.2}$$

where

$$R(v,\ \tau)=-{}^1/_4 v\psi''+({}^1/_2 v\varphi''+2\psi')/\psi \tag{3.3}$$

(the prime denotes differentiation with respect to $v$). At low frequencies, we have

$$\begin{gathered} I_2^c=S(eV_0/E_j,\ T/E_j)\,\mathrm{th}(E_j/2T),\\ I_2^s=0. \end{gathered} \tag{3.4}$$

The function $R(v,\tau)$ is shown in Fig. 3. At $T=0$ the line profile can be represented similarly to Eq. (2.14) and it consists of a $\delta$-like peak and a power-law wing in the range $eV_0 > E_j$:

$$R(v,0)=\frac{q}{2}\delta(v-1)+\frac{2q}{1+q(v-1)}\theta(v-1). \tag{3.5}$$

The frequency dependence of the conductance of a microcontact in the case of scattering by two-level systems is analogous to the frequency dispersion of the nonlinear current-voltage characteristic of a contact due to the electron-phonon interaction considered in Refs 15 and 16. For example, at high frequencies the nonlinear conversion signal falls in accordance with the $1/\omega$ law,[15] exactly as in the expression in Eq. (3.2), whereas for arbitrary frequencies the frequency dependence of the harmonic obtained in Ref. 15 is

$$I_2 \propto \Gamma_{ph}/(\Gamma_{ph}^2+\omega^2)^{1/2}, \tag{3.6}$$

where $\Gamma_{ph}$ is the phonon-electron relaxation frequency. The difference is in the characteristic values of $\Gamma$. For phonons these frequencies are $\Gamma_{ph} \sim 10^{10}$–$10^{11}$ sec$^{-1}$, whereas for two-level systems they are several orders of magnitude lower.

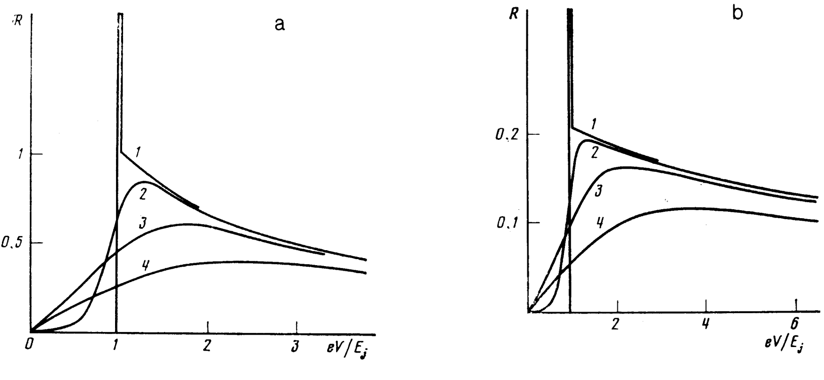

FIG. 3. Profile of a microcontact spectral line at high frequencies $\omega \gg \Gamma_{0j}$: a) $q = 0.5$; b) $q = 0.1$. Curves 1, 2, 3, and 4 correspond to $T/E_j = 0, 0.1, 0.3$, and 0.6, respectively.

The strong frequency dispersion of the nonlinear conductance appears in the case when the number of two-level systems in the vicinity of a contact is small or when one of them predominates over the others. In the case of contacts with a high degree of disorder of the lattice in the region where the metals are in contact (Fig. 1), which are characterized by large numbers of two-level systems, we find that inclusion of the expression for the distribution function (1.1) makes it possible to obtain readily the frequency dependence of the nonlinear response $I_2 \propto \ln(\Gamma_{0j}^{\max}/\omega)$, where $\Gamma_{0j}^{\max}$ is the longest of the relaxation times.

## 4. CONCLUSIONS

It is shown above that the contribution of purely elastic processes involving two-level systems to the resistance of a contact exhibits a nonlinear voltage dependence, which can be used in microcontact spectroscopy of such systems. As in the case of spectroscopy of inelastic processes,[1] the contribution of a single two-level system to a microcontact spectrum at $T \to 0$ has a $\delta$-like singularity, but there is also a wing extending to higher voltages. We shall now compare the expressions obtained with those for a microcontact spectrum of inelastic processes involving two-level systems (TLS) and calculated in Ref. 4:

$$\left(\frac{1}{R}\frac{dR}{dV}\right)_{\rm inel}^{\rm TLS} \approx \frac{1}{V}\sum_j C_j \sigma_j \zeta(eV-E_j)$$

[$\sigma_j$ is the cross section for the inelastic scattering of an electron by a two-level system given by $\sigma_j = \bar{\sigma}_j(\Delta_{0j}/E_j)^2$, where $\bar{\sigma}_j < |\sigma_j^+ - \sigma_j^-|$ and $\zeta$ is a $\delta$-like function].

We can easily see that the contribution of inelastic processes may be comparable with the contribution of elastic processes only for $\Delta_{0j}/E_j \sim 1$, i.e., in the case of rapidly relaxing systems, as already pointed out above. However, in the case of slowly relaxing two-level systems [which, as indicated by the nature of the distribution (1.1), are more probable] a microcontact spectrum is governed specifically by elastic processes. However, it should be mentioned that in our calculations we have ignored the nonelectron mechanisms of relaxation of two-level systems. If the interaction of such systems with phonons is stronger than with electrons (which may be true in the range $E_j \gtrsim 5$ K), then the contribution of elastic processes decreases on increase in the parameter $\tau_j^{\rm ph}/\tau_j^e$ ($\tau_j^e$ and $\tau_j^{\rm ph}$ are the relaxation times of two-level systems for the interaction with electrons and phonons, respectively).

Finally, we should mention that studies of the frequency dependences of microcontact spectra should make it possible to determine the characteristic relaxation times of two-level systems responsible for the singularities of the spectra. Since the minimum possible values of $\tau_j = \Gamma_{0j}^{-1}$ corresponding to $E_j \sim 1$ K amount to $10^{-8}$–$10^{-9}$ sec, it follows from the exponentially range of the scatter of $\tau_j$ that these characteristic times are very likely to exceed $10^{-6}$ sec and we can expect the frequency dispersion to be manifested at frequencies below 1 MHz. This result is important in connection with the problem of anomalies of microcontact spectra at low voltages. In our opinion, detection of the frequency dependence of such spectra at low frequencies would be a reliable indication that it is two-level systems which are responsible for the observed anomalies.

We must draw attention to the fact that the physical picture of these phenomena generally differs from that which is characteristic of microcontact spectroscopy of inelastic processes, so that we can speak of microcontact spectroscopy of a new type which can be called population spectroscopy. It is based on the dependence of the resistance of a contact on the occupation numbers of individual levels of centers, so that the singularities of the current-voltage characteristic represent the changes in these occupation numbers. The dependence $R(N)$ can then be used to record changes in the occupation numbers caused by other factors, for example, by an alternating microwave electric field (in this case the change is of resonant nature in the dependence on the field frequency).

It therefore follows that microcontact spectroscopy can provide a convenient method for investigating the distribution of two-level systems in respect of their energies and relaxation frequencies. A combined study of microcontact spectra and low-frequency noise of contacts would be interesting from the point of view of identifying the nature of the $1/f$ noise.[19]

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