

## Relaxation of two-level fluctuators in point contacts

O. P. Balkashin

*B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Science of Ukraine, 47 Lenin Avenue, 310164 Kharkov, Ukraine*

R. J. P. Keijsers and H. van Kempen

*Research Institute for Materials, University of Nijmegen, Toernooiveld 1, NL-6525 ED Nijmegen, The Netherlands*

Yu. A. Kolesnichenko

*B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Science of Ukraine, 47 Lenin Avenue, 310164 Kharkov, Ukraine*

O. I. Shklyarevskii

*B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Science of Ukraine, 47 Lenin Avenue, 310164 Kharkov, Ukraine*

*and Research Institute for Materials, University of Nijmegen, Toernooiveld 1, NL-6525 ED Nijmegen, The Netherlands*

(Received 29 December 1997)

The contributions by three different mechanisms of interactions between electrons and two-level fluctuators to the low-energy singularity present in point-contact spectra of metallic glasses have been studied by measuring rf response signals at 600 MHz and 60 GHz, and the low-frequency response at 1.85 kHz. The resulting curves indicate that a nonmagnetic Kondo-like interaction is the most important contribution, but, depending on the exact shape of the background signal due to electron-phonon and electron-electron interactions, elastic scattering on highly asymmetric two-level fluctuators may also be quite important. [S0163-1829(98)01927-4]

### I. INTRODUCTION

The elucidation of the origin of low-energy singularities, the so-called “zero-bias anomalies” (ZBA), is one of the most long-standing problems in point-contact (PC) spectroscopy of conductors.<sup>1</sup> In some special cases (e.g., dilute magnetic alloys)<sup>2,3</sup> the ordinary or one-channel Kondo effect is responsible for the observed maximum in the differential resistance of PC’s at zero-bias voltage. Nowadays it is acknowledged that in the majority of cases these effects originate from interactions of electrons with lattice defects that switch between two nearly equivalent positions (two-level fluctuators or TLF’s). In normal metals PC’s, nonequilibrium defects were created during the PC fabrication process, and room-temperature “annealing” of nanoconstrictions<sup>4</sup> or break junctions<sup>5</sup> brings about a dramatic decrease or even a complete disappearance of the ZBA. In amorphous conducting materials or metallic glasses (MG’s) the structural defects are quenched in, and the high density of TLF’s determines their anomalous low-temperature properties.<sup>6</sup> This makes this class of materials a model object for investigations of electron-TLF interactions employing the PC spectroscopy technique.

There are two different mechanisms that can result in the observed nonlinearity of the  $I$ - $V$  curves. The first one was predicted by Vladar and Zawadowskii<sup>7</sup> (VZ) and stems from a nonFermi liquid behavior of the electrons due to their coupling with TLF’s. The second is determined by a specific nonequilibrium distribution of the electrons in the vicinity of the contact that depends on the applied bias  $V_b$ . Relaxation of these nonequilibrium electrons on TLF’s and a variation

of the occupation numbers of the latter as a result of electron-TLF scattering cause a nonlinear behavior of the current through the contact as a function of voltage that was described by Kozub and Kulik<sup>8,9</sup> (KK). There are some experimental data<sup>4,10</sup> that strongly support the two-channel scattering model,<sup>7</sup> whereas in our previous papers, we failed to discriminate unambiguously between the VZ and KK theory on the basis of spectral features<sup>11</sup> or modulation of electron-TLF scattering by slowly moving defects<sup>12</sup> alone. (See also the discussion in Refs. 13 and 14.)

It should be noted that although the asymmetric TLF’s, accountable for the KK mechanism, and the nearly symmetric TLF’s in the VZ model are frequently referred to as “fast” ones, the difference in their relaxation times ( $\approx 10^{-5}$ – $10^{-6}$  and  $\approx 10^{-11}$  s, respectively) reaches 5–6 orders of magnitude. From the short theoretical analysis presented in the next section it becomes evident that yet another possibility to separate different contributions to the zero-bias anomaly is an investigation of the nonsteady-state conductivity of PC’s in the frequency range where  $\omega\tau_{\text{TLF}} \sim 1$ . This, in principle, gives the possibility to study the relaxation kinetics and to determine characteristic relaxation times of various scattering processes, as was shown before.<sup>15–17</sup>

Here, we report experimental observations of response signals for metallic glass PC’s in rf electromagnetic fields at irradiation frequencies  $\omega_1/2\pi = 6 \times 10^8$  Hz and  $\omega_2/2\pi = 6 \times 10^{10}$  Hz. The results will be compared to the low-frequency response, after which some conclusions on the most important contributions are presented. But first, a short theoretical description of the different electron-TLF interac-

tion mechanisms that affect the current through a PC will be given.

## II. THEORY

The addition to the point-contact current due to the presence of TLF's can be written as a sum

$$\Delta I = \Delta I_1 + \Delta I_2 + \Delta I_3, \quad (1)$$

where the first addend  $\Delta I_1$ , related to the nonFermi-liquid behavior of the electrons described by VZ,<sup>7</sup> is the result of "elastic" scattering on individual defects in the contact area,<sup>18</sup>

$$\Delta I_1 = \frac{V}{R_0} \frac{\sigma_1}{S_c} \sum_j \mathbf{M}(\mathbf{r}_j) \quad (2)$$

with  $R_0$  the contact resistance in the absence of TLF's and  $S_c$  the contact area.  $\mathbf{M}(\mathbf{r}_j)$  is a geometrical factor that depends on the individual TLF positions,<sup>19</sup>

$$\mathbf{M}(\mathbf{r}_j) = 4 \int \frac{d\Omega_{\mathbf{p}}}{4\pi} \int \frac{d\Omega_{\mathbf{p}'}}{4\pi} \alpha_{\mathbf{p}}(\mathbf{r}_j) [\alpha_{-\mathbf{p}'}(\mathbf{r}_j) - \alpha_{-\mathbf{p}}(\mathbf{r}_j)]. \quad (3)$$

$\alpha_{\mathbf{p}}(\mathbf{r})$  is the probability for an electron with momentum  $\mathbf{p}$  to reach the point  $\mathbf{r}$  starting from one of the electrodes of the contact. For  $eV \ll \epsilon_F$  the function  $\alpha_{\mathbf{p}}(\mathbf{r})$  satisfies a field-independent kinetic equation as well as the boundary condition corresponding to the requirement of zero-current flow across the metal surface.<sup>20</sup> The effective scattering cross section  $\sigma_1$  for electrons on TLF's can be represented by matrix elements of the electron-TLF coupling. Its dependence on the energy  $eV$  of the incident electrons and on temperature is determined by renormalization effects that are essential only for nearly symmetric double-well potentials<sup>7</sup> for which the energy splitting between the two minima (asymmetry energy)  $\Delta_j$  is much smaller than the tunneling energy (tunnel-matrix element)  $\Delta_{0j}$ . The tunneling rate between the two minima of the  $j$ th TLF can mostly be taken proportional to  $(\Delta_{0j}/E_j)^2$ ,<sup>8,9</sup> with the excitation energy  $E_j = (\Delta_j^2 + \Delta_{0j}^2)^{1/2}$ . It must be emphasized that the inequality  $\Delta_j \ll \Delta_{0j}$  therefore generally corresponds to quickly relaxing TLF's.

Inelastic scattering of electrons on these TLF's results in a second addend to the current, which is given by<sup>8</sup>

$$\Delta I_2 = \frac{1}{eR_0} \frac{\sigma_2}{S_c} \sum_j \mathbf{M}(\mathbf{r}_j) \left[ 2eV \Theta(E_j - eV) + \frac{2E_j^2}{E_j + q(eV - E_j)} \Theta(eV - E_j) \right] \quad (4)$$

at  $T=0$ , with<sup>19</sup>

$$q = \frac{1}{2} \left[ 1 - \left( 1 - 2 \int \frac{d\Omega_{\mathbf{p}}}{4\pi} \alpha_{\mathbf{p}}(\mathbf{r}_j) \right)^2 \right]$$

and  $\sigma_2$  the effective inelastic scattering cross section for electrons on quickly relaxing TLF's.

For elastic scattering on highly asymmetric TLF's ( $\Delta_j \gg \Delta_{0j}$ ), it is important to take the bias dependence of the occupation numbers for each level into consideration. This

bias dependence arises as a result of a modification of the TLF state by inelastic interactions and leads to a third current addend or "spectral" term<sup>9</sup> at  $T=0$

$$\begin{aligned} \Delta I_3 &= \frac{V}{2R_0} \frac{1}{S_c} \sum_j \mathbf{M}(\mathbf{r}_j) [\sigma_j^+ (1 - N_j) + \sigma_j^- N_j] \\ &= \frac{V}{2R_0} \sum_j \mathbf{M}(\mathbf{r}_j) \frac{\sigma_j^+ - \sigma_j^-}{2S_c} \left[ \Theta(E_j - eV) \right. \\ &\quad \left. + \frac{E}{E + q(eV - E)} \Theta(eV - E_j) \right], \end{aligned} \quad (5)$$

where  $\sigma_j^+$  and  $\sigma_j^-$  are the scattering cross sections for the upper and lower levels, and  $N_j$  is the occupation number for the lower level.

The contributions of  $\Delta I_1$ ,  $\Delta I_2$ , and  $\Delta I_3$  to the PC spectrum are different. The first one describes a negative Kondo-like anomaly in the second derivative of the  $I$ - $V$  curve. The second is analogous to the inelastic scattering of electrons on phonons and results in an increase of the contact resistance. This contribution to the anomalous behavior therefore has a positive sign with respect to the electron-phonon interaction spectrum. The sign of  $\Delta I_3$  depends on the sign of the difference between the effective scattering cross sections  $\sigma_j^\pm$ . We emphasize that for the negative ZBA observed in our experiments, the effective scattering cross section of TLF's in the upper state,  $\sigma_j^+$ , is less than  $\sigma_j^-$ . In this case the *increase* of the bias voltage results in an increase of the occupation number for electrons in the upper state and a *decrease* of current backflow. When the energy-distribution function for TLF's in the PC has a maximum at  $E_j = E_0$ , then, in accordance with Eqs. (4) and (5), a singularity at  $eV = E_0$  in the PC spectrum appears.

The  $\omega$  ranges at which the frequency dependence for each contribution manifests itself differs considerably. At relatively low frequencies the dependence  $d^2V/dI^2$  is determined by  $\Delta I_3$ . According to KK,<sup>9</sup> the amplitude of the second derivative of the  $I$ - $V$  curve for TLF's with a relaxation frequency  $\Gamma_j < \omega$  at  $T=0$  is proportional to

$$\begin{aligned} \Delta I_3^{(2)} &\sim \frac{\sigma^+ - \sigma^-}{S_c} \sum_j \mathbf{M}(\mathbf{r}_j) \frac{\Gamma_j}{\omega} \left[ \frac{qeV}{2} \delta(eV - E_j) \right. \\ &\quad \left. + \frac{2qE_j}{E_j + q(eV - E_j)} \Theta(eV - E_j) \right]. \end{aligned} \quad (6)$$

Because of the factor  $\Gamma/\omega$  the intensity of the  $\Delta I_3$  term in the rf response signal must drop considerably already in the MHz range.<sup>9</sup>

## III. EXPERIMENT

For the rf experiments we used conventional pressure-type PC's described elsewhere (see, e.g., Ref. 21 and references therein). The PC's were produced by bringing the edges of two MG strips together by means of differential screws while being directly immersed in liquid helium. Before mounting into the cryostat, the electrodes were cleaned by etching in a solution of nitric and hydrochloric acids ( $\text{HNO}_3:\text{HCl}:\text{H}_2\text{O} = 1:1:5$ ).

All measurements were done at  $T=1.6$  K. In a single cycle both the second harmonic of the low-frequency (1.85 kHz) modulation current and the rectification signal for rf irradiation (using a 100% 2.43-kHz low-frequency amplitude modulation) were registered using a conventional lock-in technique. The amplitude of the modulation signal at audio frequency was kept as low as possible (0.5–0.6 mV) to minimize smearing of the spectrum.

The time-averaged  $I$ - $V$  curve of a PC under rf irradiation of frequency  $\omega$  may be expressed in the following form:<sup>22,23</sup>

$$\overline{I(V)} = \sum_{n=-\infty}^{\infty} J_n^2\left(\frac{ev_1}{\hbar\omega}\right) I_0\left(V_0 + n \frac{\hbar\omega}{e}\right), \quad (7)$$

where  $V = V_0 + \cos \omega t$ , the  $J_n$  are Bessel functions of order  $n$ ,  $v_1$  is the ac voltage amplitude in the contact determined by the rf field, and  $I_0(V_0)$  is the nonperturbed  $I$ - $V$  curve for dc current.

This expression can be transformed into the standard form for a classic detector in the low-frequency limit ( $\hbar\omega \ll ev_1$ ),<sup>24,25</sup>

$$\overline{I(V)} = \frac{\omega}{\pi} \int_0^{\pi/\omega} I_0(V_0 + v_1 \cos \omega t) dt. \quad (8)$$

In the low-signal limit ( $v_1 \ll V_0$ ) the current response under irradiation (the difference between the perturbed and nonperturbed  $I$ - $V$  curves) can readily be obtained from Eq. (8),

$$\delta I(V_0) \equiv \overline{I(V)} - I_0(V_0) = \frac{v_1^2}{4} \frac{d^2 I_0(V)}{dV^2} \quad (9)$$

and is proportional to the second derivative of the  $I$ - $V$  curve. Therefore the rf response in effect leads to *exactly the same function as the low-frequency point-contact curve*. This allows a comparison between the two measurements. It must be mentioned that in the experiments we measure the voltage response (a small addition to the voltage due to the rf irradiation), which is related to the current response by  $|\delta V| = |\delta I| (dV/dI)$ .

In the rf experiments the typical resistance of the PC's was about one order of magnitude smaller than the free-space wave impedance,  $\rho = 120\pi\Omega \approx 377\Omega$ . We therefore used current sources for both the rf and the low-frequency measurements.

The electromagnetic field was delivered to the PC using a standard  $10 \times 23$  mm cross-section X-band wave guide with a smooth transition to a  $2 \times 23$  mm cross section, or using a coaxial cable with a 75- $\Omega$  resistor in a coupling loop close to one of the electrodes. The electrodes were positioned in a hole through the wave guide (see Fig. 1) in such a way that the PC was at its center. By moving the short-circuit plunger one can change the structure of the rf field near the PC. Its optimal position can be found by maximizing the output signal. We restricted the frequency range to 60 GHz to prevent a transition to the quantum-detection regime. The energy of a photon is then  $\hbar\omega \approx 0.25$  meV, which is considerably less than the spectral width of the observed singularity.

During the measurements the rf power level, which is controlled by a rf diode, was kept constant. The intensity of the rf irradiation was adjusted to the minimal level that provided a detection signal amplitude of about 1  $\mu$ V.

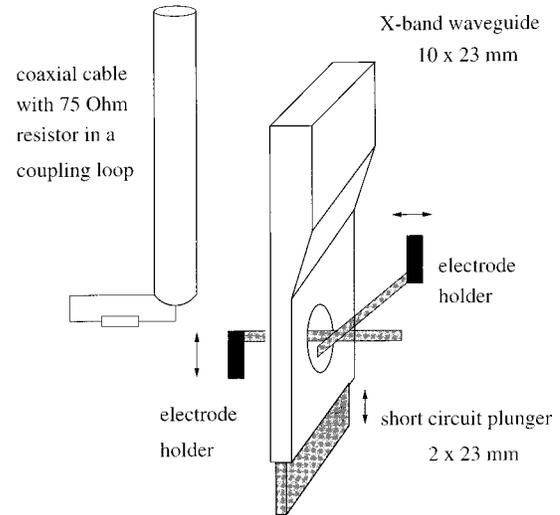


FIG. 1. Experimental setup for rf response signal measurements in point contacts.

In a part of the experiments the response signal was measured for a few levels of rf irradiation to verify the linear dependence between the amplitude of the detected signal and the applied power.

#### IV. RESULTS AND DISCUSSION

Typical  $d^2V/dI^2(V)$  dependencies and rf response signals for the iron-based  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Fe}_{78}\text{Mo}_2\text{B}_{20}$  (known also as MG 2605 and MG 2605A) and nickel-based  $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$  (MG 2826A) metallic glasses are presented in Fig. 2. The second derivative of the  $I$ - $V$  curves shows a sharp minimum at a bias voltage  $V_b \approx 1$  mV due to the electron-TLF interaction, accompanied by a transition to a smooth negative background at  $V_b \geq 10$  mV. There, the differential resistance of the contact decreases proportional to  $V_b$  or  $V_b^{0.5}$ , which can be explained by an interaction between conduction and weakly localized electrons.<sup>6</sup> In the intermediate region a rather pronounced maximum sometimes occurs at  $V_b \sim 5$  mV for  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Fe}_{78}\text{Mo}_2\text{B}_{20}$ . This can be understood as a result of a superposition of electron-TLF, electron-phonon, and electron-electron interactions.

The main problem in rf measurements is the calibration of the response signal with respect to the second derivative signal. For normal metal PC's this can easily be done by fitting the intensities of the low-energy electron-phonon interaction maxima.<sup>16</sup> Here, the fact that in MG's the electron-electron interaction at elevated bias voltages is the dominant-scattering mechanism is very important, because one can expect that the corresponding scattering time is very short and the second derivative and rf signal amplitudes for different  $\omega$  must therefore be practically the same. A calibration can then be made by fitting the background signals at  $V_b \geq 15 - 20$  mV.

Experimental proof for this suggestion is presented in Fig. 3, where the  $d^2V/dI^2(V)$  dependence and the rf response signals are plotted for a  $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$  PC, which was obtained as a result of a spontaneous electrical breakdown of a more high-Ohmic junction. Evidently, during this process, which includes local heating (or even melting) of material

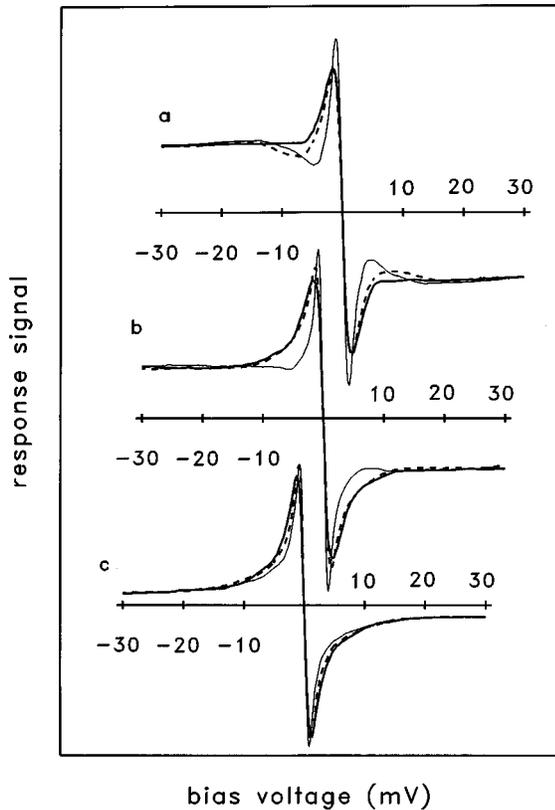


FIG. 2. rf response signals at 60 GHz (thick solid line) and 0.6 GHz (dashed line) and  $d^2V/dI^2(V)$  dependence (thin solid line) for (a) a 26- $\Omega$  Fe<sub>80</sub>B<sub>20</sub> point contact, (b) a 8- $\Omega$  Fe<sub>78</sub>Mo<sub>2</sub>B<sub>20</sub> point contact, and (c) a 15- $\Omega$  Fe<sub>32</sub>Ni<sub>36</sub>Cr<sub>14</sub>P<sub>12</sub>B<sub>6</sub> point contact.

within the contact area, the degree of disorder decreases and phonon-electron reabsorption processes become more important at sufficiently high  $V_b$ , explaining the change to a positive sign for  $d^2V/dI^2(V)$ . An estimation of the phonon-electron relaxation time gives  $\omega\tau_{\text{ph-e}} \approx 1$  at  $\omega/2\pi = 6-8$  GHz and therefore  $\omega_1 \ll \omega_{\text{ph-e}} \ll \omega_2$ , in full accordance with the fact that the response signal at 0.6 GHz follows the behavior of the second derivative, whereas for the 60-GHz curve the background remains negative. It must be noted, however, that for the nickel-based MG with a relatively small background (less than 10% of the zero-bias anomaly amplitude) the proposed calibration procedure is

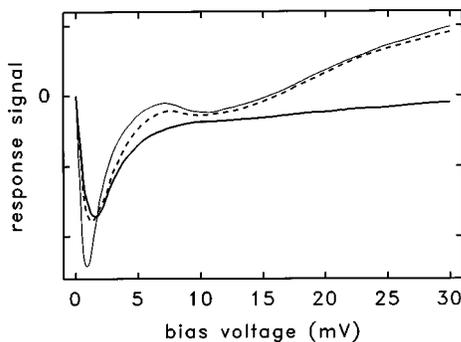


FIG. 3. rf response signals at 60 GHz (thick solid line) and 0.6 GHz (dashed line) and  $d^2V/dI^2(V)$  dependence (thin solid line) for a short circuited (partly recrystallized) 22- $\Omega$  Fe<sub>32</sub>Ni<sub>36</sub>Cr<sub>14</sub>P<sub>12</sub>B<sub>6</sub> point contact.

not very accurate and may result in a 10–20 % error in the intensity.

The zero-bias anomaly in the rf response signals (Fig. 2) is somewhat smeared and has a reduced amplitude compared to the low-energy singularity in  $d^2V/dI^2$ . This effect is already clearly visible at 0.6 GHz and becomes larger at 60 GHz for a majority of the contacts. It should be stressed that the shape of the rf curve is completely different from that of the second derivative smeared by temperature or a large (up to 2–3 mV) modulation voltage. This indicates that the rf power dissipation in the contacts is moderate enough and does not cause overheating. The size of the effect varies slightly from material to material and from contact to contact. Those variations are most likely due to a different nature of the structural defects responsible for the TLF formation, and a different spectral distribution of relaxation times. However, the amount of TLF's contributing to the nonlinear behavior of the contact conductivity ranges from a few hundreds to a few thousands for PC's with a resistance of 10–30  $\Omega$  according to estimations based on a universally adopted figure for the TLF density of  $10^{-4}-10^{-5}$  per atom. This excludes a large variation in distribution functions for TLF's.

The estimated minimal relaxation time in the MG's under investigation<sup>4,6</sup> ranges from  $10^{-10}$  to  $10^{-11}$  s. The situation where the relaxation frequency  $\Gamma_{\text{TLF}} \ll \omega_1$  therefore corresponds to the suppression of the signal from relatively slowly (in the MHz range) relaxing TLF's. A further reduction of the zero-bias anomaly amplitude at  $\omega_2$  is determined mainly by an additional decrease of the contribution from these TLF's, because faster relaxing ones (GHz range) are nearly symmetric (with the same effective scattering cross section for both levels) and do not contribute much in  $\Delta I_3^{(2)}$ . Note that the  $\Delta I_2^{(2)}$  has the positive sign.

At first glance, the relatively high amplitude of the low-bias singularities in the rf response signal up to 60 GHz seems to indicate that the main mechanism behind the zero-bias anomaly is the nonmagnetic Kondo resonance.<sup>7</sup> It furthermore suggests that the spectral density of almost symmetric, quickly relaxing TLF's in metallic glasses must be surprisingly high (or, otherwise, that the VZ model is not so sensitive to the  $\Delta \ll \Delta_0$  condition). It should be noted that the original VZ model does not anticipate any frequency dependence, but it certainly must exist at sufficiently high  $\omega$ . The term  $\Delta I_3^{(2)}$  that arises due to the elastic scattering of electrons is determined by relatively "slow" TLF's with relaxation times of  $10^{-5}-10^{-8}$  s, and seems to be noticeably smaller in our case. However, the relative magnitude of the contribution due to the nonmagnetic Kondo effect may be overestimated here when a considerable background is present in the spectrum.

To clarify this issue,  $d^2V/dI^2(V)$  was integrated to obtain the bias dependence of the differential resistance  $R_d(V)$  of a contact. By applying the same procedure to the rf response signals, quantities are obtained that are formally proportional to the differential resistances of the contact measured at  $\omega_1$  and  $\omega_2$ . Figure 4 shows a set of such curves corresponding to the data plotted in Fig. 2. An increase of the differential resistance in rf measurements can be interpreted as a suppression of the spectral contribution. It occurs approximately

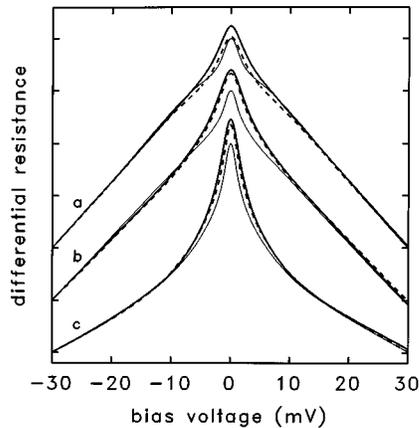


FIG. 4. Differential resistance  $dV/dI(V)$  (thin-solid line) and integrated rf response signals at 60 GHz (thick solid line) and 0.6 GHz (dashed line) for the contacts presented in Fig. 2(a)–2(c).

within the same  $V_b$  limits of  $\pm 7$ – $10$  mV where a modulation of the differential resistance by slowly moving defects was observed.<sup>12</sup> For the iron-based MG 2605 the situation is somewhat complicated, but for the other materials the relative increase of the “differential resistance” as a function of frequency in the range  $-10 < V_b < 10$  mV is nearly the same for  $\omega_1$  and  $\omega_2$  and amounts to 15–20%. This means that the KK part of the signal is still rather substantial at low frequency. The uncertainty in the calibration and in the determination of the contribution to the background signal at low biases by electron-electron and electron-phonon scattering processes makes more accurate estimations very difficult. In fact, if we assume a linear background starting from  $V=0$  and subtract this background, the KK and VZ contributions are found to be of approximately the same magnitude. This indicates that the KK contribution might certainly be present.

## V. CONCLUSION

Above, it was shown that the contributions to the zero-bias anomaly in point-contact spectra of metallic glasses by different electron-TLF interaction mechanisms can be separated by measuring rf response signals. These measurements indicate that the dominant mechanism in a certain class of metallic glasses is the two-channel Kondo scattering proposed by VZ,<sup>7</sup> and that the elastic scattering (KK) model<sup>9</sup> is of less significance. The exact shape of the background signal due to electron-phonon and electron-electron interactions is unfortunately unknown. Depending on the chosen background, the KK contribution may be of the same magnitude as the VZ contribution, and may therefore still be quite important.

We wish to stress that the results presented here should not be generally applied to different classes of metallic glasses, where the TLF’s present may be of a different nature and the relative importance of the different electron-TLF interactions are most likely not the same as for the metallic glasses studied here. This is corroborated by a study of the low-energy singularity in spectra measured at simple metal-point contacts.<sup>11</sup> The sign of the peak was different for crystalline bulk samples and PC’s of thin films of low crystalline quality, indicating that different types of TLF’s may be under study, and different interaction mechanisms may be of importance.

## ACKNOWLEDGMENTS

Part of this work was supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO). O.I.S. wishes to acknowledge the NWO for a visitor’s grant.

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