

High-frequency rectification in UPt_3 point contacts

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Rectification experiments have been performed on UPt_3 - UPt_3 and on Cu-UPt_3 point contacts, using far-infrared radiation of 562 GHz. The voltage dependence of the rectification signal differs significantly from the second derivative d^2V/dI^2 , which is also measured for these point contacts, which demonstrates that the point contacts are not in the ballistic regime. The results are explained with a heating model.

I. INTRODUCTION

During the past few years point-contact spectroscopy has been used in the study of various valence-fluctuation and heavy-fermion compounds. In most of these experiments large nonlinearities are observed in the current-voltage characteristics and their derivatives.¹⁻⁴ Explanations of these effects are given in terms of the scattering of the conduction electrons with the intermediate valence configurations,¹ with the existence of a hybridization gap in the electron density of states,² and in terms of a resonance in the density of states.^{3,4} For these spectroscopic explanations the applied contact voltage is supposed to be a direct measure of the relevant energy of the electrons, thus allowing energy-resolved spectroscopy of the electron scattering or of the density of states of the electrons.

In the different case of pure metals, metallic point contacts have been applied most successfully for a detailed study of the energy dependence of the electron-phonon interaction by measuring the nonlinearity of the current-voltage characteristics.^{5,6} In these experiments the electron mean free path is larger than the contact dimension, yielding a ballistic transport of the electrons through the contact. The applied voltage defines the energy scale for the scattering processes of the electrons, determining the voltage-dependent nonlinearity of the point-contact characteristic.

However, for the point-contact experiments in compounds with strong electron scattering as mentioned above, another explanation of the data has been given in terms of local heating in the contact area, which takes place for point contacts in the so-called dirty regime,⁷⁻⁹ i.e., where the electron mean free path is small compared with the contact dimension. For a dirty contact the Joule heat is released in the contact area, which results in nonlinear characteristics due simply to the temperature dependence of the contact resistance.

In an attempt to clarify these points further, we investigated the high-frequency rectification of UPt_3 point contacts. From the optical frequency used in these measurements, an estimation can be made of the time scale of the processes which determine the shape of the current-voltage characteristic and its derivatives. These relaxation times are expected to be quite different for a ballistic

and a dirty contact. For the ballistic model the relevant time scale should be given by the scattering of the electrons, for the heating model by the thermal relaxation of the contact area.

II. POINT-CONTACT EXPERIMENTS ON UPt_3

Point-contact spectroscopy in metals can be performed in three different regimes. In the ballistic regime, the inelastic mean free path l_i and the elastic mean free path l_e of the electrons are larger than the contact radius a , i.e., $l_i, l_e \gg a$. In this regime spectroscopic information is obtained about the energy dependence of the scattering of the conduction electrons with elementary excitations in the metal. This information is directly determined from a metallic point contact at low temperatures by measuring the second derivative d^2I/dV^2 of the current I with respect to the voltage V through the relation⁶

$$\frac{d^2I}{dV^2}(eV) \propto a^3 S(eV), \quad (1)$$

where $S(eV)$ is the spectral function for the interaction to be studied. For instance, in the case of the electron-phonon interaction, this spectral function $S(eV)$ equals $\alpha^2 F(eV)$, the Eliashberg function for the electron-phonon interaction.^{5,6}

In analogy to a tunnel experiment, it is often^{2,4} stated that with point-contact experiments one should be able to observe the energy dependence of the electron density of states directly, using the integral $I(eV) \propto \int_0^{eV} N(\epsilon) d\epsilon$ for the current. Hence the differential conductance dI/dV should be proportional to the density of states. However, for a point contact in the ballistic regime this proportionality does not hold. The ballistic electron current through a constriction, i.e., the current in zeroth order without any collisions, is given for a spherical Fermi distribution by⁶

$$I^{(0)} \propto \int k^2 v_k dk = \int_{E_F - eV/2}^{E_F + eV/2} k^2 v_k \left(\frac{\partial k}{\partial E} \right) dE, \quad (2)$$

where v_k is the velocity of an electron with wave vector k . Writing $v_k \propto \partial E / \partial k$ we can see that the electron density of states $N(eV) \propto \partial k / \partial E$ drops out so that there

remains only

$$I^{(0)} \propto \int k^2 dE. \quad (3)$$

Hence the density of states cannot be observed directly by measuring the differential conductance dI/dV . Only nonparabolicity and anisotropy can make the electronic band structure enter the problem.

The second point-contact regime is the diffusive regime where the elastic scattering is strong, i.e., where $l_e \ll a$, but where the inelastic diffusion length $\Lambda_i = (l_i l_e)^{1/2}$ is still large compared with the contact diameter, i.e., $\Lambda_i > a$. In this regime, spectroscopic information can still be obtained; however, the intensities of the spectra are reduced, since in Eq. (1) an extra factor l_e/a has to be added.¹⁰ This factor is due to the reduced "efficiency volume" $a^2 l_e$ where the crucial scattering processes take place. Note that in the ballistic and diffusive regimes the inelastic lengths l_i and Λ_i have to be taken at an electron energy eV with respect to the Fermi level for an applied voltage V over the contact, whose values differ from the low temperature values at $V=0$.

The thermal point-contact regime, in which $l_i, l_e \ll a$, no longer allows energy-resolved spectroscopy. Due to the small electron mean free path, the electrons are not injected ballistically and the Joule heat is released in the contact area itself. The electron and phonon systems are in equilibrium and a complete thermalization takes place. With the assumption that the electrical and thermal currents follow equal paths, one can calculate the maximum temperature at the center of the contact at a certain applied voltage. This maximum temperature is independent of the specific contact geometry and given by¹¹

$$T_{\max}^2 = T_{\text{bath}}^2 + V^2/4L. \quad (4)$$

Here T_{bath} is the bath temperature and L is the Lorenz number.

In this thermal regime the resistance of the point contact is given by the so-called Maxwell value

$$R_M = \rho/2a, \quad (5)$$

where a circular contact is assumed with contact radius a . In this equation ρ is the bulk resistivity of the metal under study. A combination of the expressions (4) and (5) indicates that in this regime a measurement of the point-contact resistance as a function of bias voltage should correspond to a measurement of the bulk resistivity ρ as a function of temperature.

In point-contact measurements on the heavy-fermion system UPT₃, large nonlinearities were observed in the current-voltage characteristics. In an attempt to explain the experimental findings in terms of the heating model, a clear resemblance was found between the voltage-dependent contact resistance $dV/dI(V)$ and the temperature-dependent bulk resistivity $\rho(T)$, but the relative change of $dV/dI(V)$ was roughly a factor of 5 smaller than the relative change of $\rho(T)$ between 4 and 100 K.⁹ To account for the local resistivity in the contact area, a comparison of $dV/dI(V)$ of the contact was made with the measured point-contact resistance as a function of temperature. Such a comparison gives a better quantita-

tive agreement. The heating model fails to explain the measured temperature-dependent contact resistance from the bulk-resistivity data in an exact quantitative way using Eq. (5). It is possible that the local (temperature-dependent) resistivity in the point-contact case deviates from the bulk (influence of pressure, deformation). This discrepancy between the two dependences $\rho(T)$ and $dV/dI(T)$ is often observed in point-contact experiments with high-resistivity materials.

III. POINT CONTACTS IN HIGH-FREQUENCY ELECTROMAGNETIC FIELDS

A. Ballistic regime

Because of the nonlinear current-voltage characteristics of metallic point contacts, these can be used as rectifiers for high-frequency electromagnetic (hf EM) radiation. Since the contact resistance is in most cases much smaller than the vacuum impedance, such a hf-radiation field is then used as a hf-current source. The whisker from which the point contact is made serves as an antenna to couple in the radiation.

When an ac current $i_0 \cos(\omega_{\text{rad}} t)$ is induced in a point contact with hf EM radiation, and if this radiation is chopped, it is easily shown that the measured ac voltage at this chopper frequency equals

$$V_{\text{sig}} = \frac{i_0^2}{8} \frac{d^2 V}{dI^2} \bigg|_{I_0} = \frac{i_0^2}{8} R_D \frac{dR_D}{dV} \bigg|_{I_0}. \quad (6)$$

Here I_0 is the applied dc current through the contact and $R_D = dV/dI$ is the dynamical resistance of the point contact. Thus this technique enables another method for measuring the second derivative $d^2 V/dI^2$. For instance, at far-infrared (FIR) frequencies, rectification measurements on Cu point contacts reveal the phonon structure as can also be measured with a traditional low-frequency modulation technique.¹² In order to measure the second derivative $d^2 V/dI^2$ according to the rectification technique described by Eq. (6), it is necessary that the characteristic time τ for the processes of the observed nonlinearities is short compared with the time scale imposed by the radiation. In other words, the voltage over the point contact must follow the induced ac current modulation. For the mentioned rectification experiments on Cu this characteristic time τ is the electron-phonon scattering time, $\tau_{\text{el-ph}}$, which is typically 10^{-14} – 10^{-13} s for Cu at electron energies corresponding to the Debye phonon frequencies. With FIR radiation with a frequency ω_{rad} of approximately 500 GHz, one finds $\omega_{\text{rad}} \tau_{\text{el-ph}} \simeq 10^{-2} \ll 1$. This explains why the electron-phonon spectra are observed in the rectification experiments on Cu.

B. Thermal regime

For the case that a point contact is not in the ballistic, but is in the thermal regime, one has to derive another expression for the detected signal. In this case the characteristic time τ is the thermal relaxation time τ_{thermal} for the flow of heat out of the contact area and is given by the quotient of the total heat capacity C of the

contact and the conduction of heat Λ out of the contact area, i.e., $\tau_{\text{thermal}} \sim C/\Lambda \simeq cd^3/(\lambda d^2/d) = cd^2/\lambda$, where d is the diameter of the contact, c the specific heat, and λ the thermal conductivity of UPt_3 . Using a typical value for the contact diameter $d \sim 100$ nm and experimental values for c and λ ,¹³ one finds $\tau_{\text{thermal}} \sim 10^{-9}$ s. With FIR radiation of approximately 500 GHz one finds $\omega_{\text{rad}}\tau_{\text{thermal}} \simeq 10^3 \gg 1$. Therefore, since the heating in the contact area is a slow process compared to the FIR frequency, the ac current modulation will not follow the nonlinear dc current-voltage characteristics, and no rectification will be observed. However, the induced hf current can increase the local temperature in the contact area, causing a detectable change in the voltage across the constriction. The voltage over the contact will be $V(I_0)|_{\text{no rad}} = V(I_0)|_T = I_0 R_S(I_0)|_T$ in the absence of radiation, and $V(I_0)|_{\text{with rad}} = V(I_0)|_{T+\Delta T} = I_0 R_S(I_0)|_{T+\Delta T}$ in the presence of radiation. Here $R_S = V(I)/I$ is the static resistance, depending on the temperature T . In a rectification experiment the detected voltage then will be

$$V_{\text{det}} = V(I_0)|_{\text{with rad}} - V(I_0)|_{\text{no rad}} \simeq I_0 \frac{dR_S}{dT} \Delta T, \quad (7)$$

when the temperature difference ΔT is small. In the presence of a hf current $i_0 \cos(\omega_{\text{rad}} t)$ across the contact, we have to take the effective value $\langle V + i_0 R_S \cos \omega_{\text{rad}} t \rangle_{\text{eff}}$ for the voltage V in Eq. (4). Using this expression for the effective voltage in Eq. (4), the detected voltage in Eq. (7) yields

$$V_{\text{det}} = \frac{\sqrt{2} i_0^2}{8} R_S \frac{dR_S}{dV}. \quad (8)$$

Note that the specific voltage dependence of the contact temperature can deviate from Eq. (4), but still the result of the last expression is found. Comparing Eqs. (6) and (8) for the measured signals in both regimes, the difference is the replacement of the dynamical resistance in Eq. (6) by the static resistance in Eq. (8). In the traditional way of measuring point-contact spectra, i.e., with a low-frequency current modulation and detection of the second derivative at the double frequency, the same signal is measured as Eq. (6). The expected difference between the signals for the ballistic and the thermal regime enables one to distinguish experimentally between both regimes.

The expression for the detected voltage in the thermal regime is derived with the assumption that $\omega_{\text{rad}}\tau_{\text{thermal}} > 1$, where ω_{rad} is the applied radiation frequency and τ_{thermal} the characteristic time for the thermal relaxation. When a radiation frequency is used which is low enough, i.e., $\omega_{\text{rad}}\tau_{\text{thermal}} < 1$ again, one detects a signal conforming to Eq. (6). For Ni-Ni point contacts which are in the thermal regime at high bias voltages, this effect has been observed by Balkashin *et al.*¹⁴ They measured the signal at a few frequencies where $\omega_{\text{rad}}\tau_{\text{thermal}} \sim 1$. A considerable change in the shape of the signal was observed, which can be ascribed to the effects mentioned above.

IV. EXPERIMENTAL DETAILS

Rectification experiments were performed on UPt_3 - UPt_3 and on Cu- UPt_3 point contacts. In the latter case a Cu whisker was pressed on a piece of bulk polycrystalline UPt_3 . The UPt_3 - UPt_3 point contacts were formed by pressing two small pieces of bulk polycrystalline UPt_3 together. However, the sample from which the whisker was made had a rather small diameter (~ 0.2 mm) in order to facilitate the coupling of the radiation to the point contact. The point contacts were made at helium temperatures by means of a differential screw mechanism. The obtained contact resistances varied from 0.3 to 8.5 Ω . Using $\rho(2 \text{ K}) = 3 \times 10^{-8} \Omega \text{ m}$ in the interpolation formula $R = 4\rho l/3\pi a^2 + \rho/2a$ for the contact resistance between clean and dirty limit, we obtain values from 70 to 8 nm for the contact radius a . From the free-electron value $\rho l = 10^{-15} \Omega \text{ m}^2$ we obtain $l = 33$ nm at 2 K. The contacts are in the intermediate regime at low temperatures and zero voltage. With an applied voltage, however, the strong electron-electron scattering will diminish the electron mean free path $l(eV)$ and the contact is brought out of the ballistic limit for inelastic scattering. Note in this context that the electrical resistivity of bulk UPt_3 changes nearly by a factor of 10 between 1 and 5 K.

The radiation from an optically pumped FIR laser was focused onto the contact by means of a light cone which was mounted at the end of an oversized waveguide. Most of the measurements were performed at a radiation frequency of 562 GHz (i.e., $533.7 \mu\text{m}$). However, also some measurements were performed at laser frequencies 2523 GHz (i.e., $118.8 \mu\text{m}$) and 639 GHz (i.e., $469 \mu\text{m}$). The measurements were performed at temperatures between 1.5 and 4.2 K.

V. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows both the measured second derivative d^2V/dI^2 (upper curve) and the laser-detected signal (lower curve) for a UPt_3 - UPt_3 point contact as a function of applied voltage V . The second derivative d^2V/dI^2 was measured with a standard low-frequency modulation technique. The wiggly structure at voltages above roughly 10 mV in this spectrum is reproducible for this contact but differs from contact to contact. It can be explained in terms of local boiling of the helium at crystallite boundaries near the contact, which is known to occur particularly below the λ point of helium in point-contact experiments with shorted tunnel junctions.¹⁵ The occurrence of the boiling phenomena influences the heat transfer from the contact region, giving rise to changes in the contact resistance, from which follows the observed structure in the current-voltage characteristics. From Fig. 1 it is clear that the signal measured with the rectification method is broader than the measured second derivative. One can conclude that the rectified signal is not the expected signal for a point contact in the ballistic regime with a sufficiently fast response [$\omega_{\text{rad}}\tau < 1$, Eq. (6)]. The expected signal for the thermal regime with a slow response was also calculated using the measured dynamic resistance dV/dI of the contact in Eq. (8). The calculat-

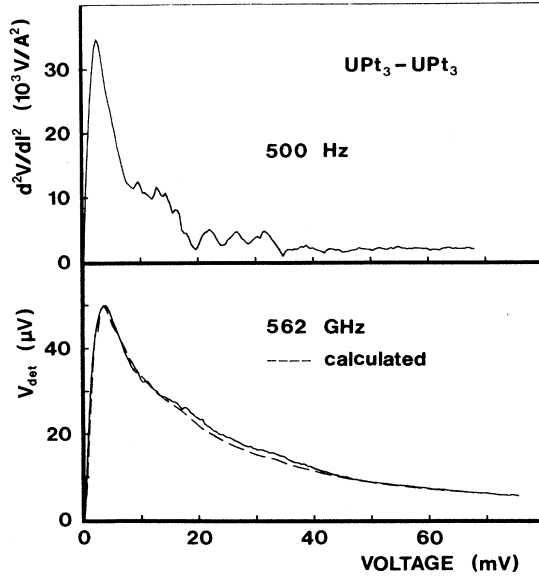


FIG. 1. Measured d^2V/dI^2 spectrum (upper curve) and the detected rectification signal V_{det} (lower curve) for a UPt_3 - UPt_3 point contact with resistance $R(V=0)=1.3 \Omega$ at bath temperature $T_{\text{bath}}=2 \text{ K}$. The applied radiation frequency was 562 GHz. The dashed curve represents the expected rectification signal in the model of local heating, calculated with expression (8).

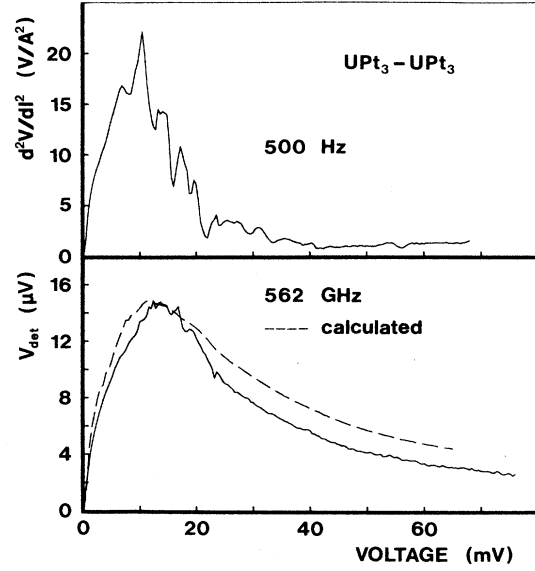


FIG. 2. Measured d^2V/dI^2 spectrum (upper curve) and the detected rectification signal V_{det} (lower curve) for a UPt_3 - UPt_3 point contact with resistance $R(V=0)=0.44 \Omega$ at bath temperature $T_{\text{bath}}=2 \text{ K}$. The applied radiation frequency was 562 GHz. The dashed curve represents the expected rectification signal in the model of local heating, calculated with expression (8).

ed curve is represented by the dashed line in Fig. 1. There is a good agreement between the measured and calculated curves. For another contact (Fig. 2) the measured second derivative d^2V/dI^2 already is much broader than the one in Fig. 1. The observed difference between point-contact spectra with UPt_3 is probably due to local-pressure effects in the contact area.⁹ For the experimental data in Fig. 2 the rectification signal also differs from the measured d^2V/dI^2 signal. The dashed curve again represents the signal as calculated from Eq. (8). In order to optimize the coupling of the electromagnetic radiation with the point contact, we also measured contacts between a Cu whisker (antenna) and UPt_3 with an experimental result shown in Fig. 3. The structure observed in Figs. 1 and 2 due to the local boiling of helium is absent in this case since the measurement was performed at 4.2 K. Also here there is a good agreement between the rectification signal and the calculation in the thermal model with a slow response.

Measurements on UPt_3 - UPt_3 point contacts were also performed at radiation frequencies 639 and 2523 GHz. In all measurements the results were found to be the same as those at 562 GHz. The photon-assisted-tunneling (PAT) effect yields a broadening of the rectification spectra for $\hbar\omega_{\text{rad}} \sim e\Delta V$, where ΔV is the characteristic voltage range for the changing resistance.¹⁶ Even the frequency of 2523 GHz (10.5 meV) is not large enough to explain the broadening in laser-detected signals between 20 and 60 mV via the PAT effect. Also, the intensity of the radiation was changed during some measurements. The shape of the signal turned out to be independent of the applied intensity.

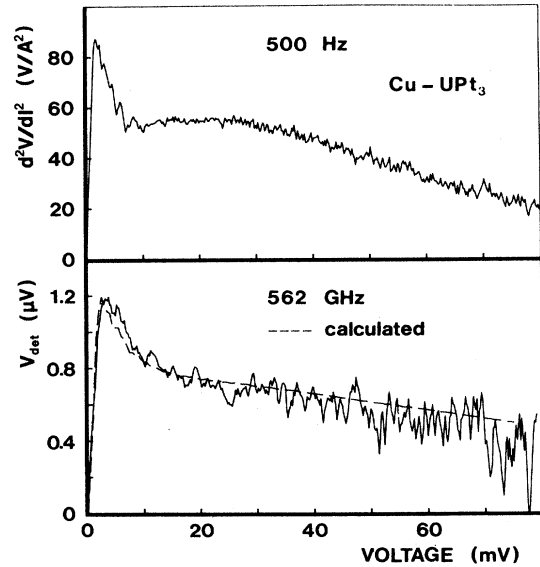


FIG. 3. Measured d^2V/dI^2 spectrum (upper curve) and the detected rectification signal V_{det} (lower curve) for a Cu- UPt_3 point contact with resistance $R(V=0)=1.3 \Omega$ at bath temperature $T_{\text{bath}}=4.2 \text{ K}$. The applied radiation frequency was 562 GHz. The dashed curve represents the expected rectification signal in the model of local heating, calculated with expression (8).

From the agreement between the measured and the calculated signals one may conclude that the point contacts are in a regime where $\omega_{\text{rad}}\tau > 1$. For UPt_3 we can make an estimation for the scattering time τ_{scatt} of the conduction electrons. At 2 K the value of $\tau_{\text{scatt}} = l/v_F^*$ will be 5×10^{-12} s. Here we used the value of the specific resistivity at low temperatures ($\rho = 3 \mu\Omega \text{ cm}$) with a free-electron expression $\rho l = 10^{-15} \Omega \text{ m}^2$, together with the renormalized Fermi velocity ($v_F^* = 7 \times 10^3$ m/s) for this heavy-fermion system.¹⁷ Thus $\omega_{\text{rad}}\tau_{\text{scatt}} \sim 1$ in this case. Therefore it is not clear what kind of rectification signal will be measured at low temperatures and zero voltage. At higher temperatures and at higher voltages, the electron mean free path will decrease strongly and the Fermi velocity may obtain a normal higher value. The scattering time τ_{scatt} of the electrons will also decrease, i.e., $\tau_{\text{scatt}} < 5 \times 10^{-12}$ s in this case. Therefore $\omega_{\text{rad}}\tau_{\text{scatt}} < 1$ for a higher temperature or higher voltage. Thus in this case, a rectification signal according to Eq. (6) can be expected. We want to emphasize that this is only the case when electronic processes determine the current-voltage characteristics of the point contact. From the experiments ($\omega_{\text{rad}}\tau > 1$), we can conclude that electronic processes do not determine the nonlinear characteristics, but simple heating effects have to be considered.

One could imagine that the heating which is observed is not local heating by an induced high-frequency ac current but simply by absorption of radiation in the bulk sample, and therefore heating of the whole sample. For this case we can also estimate the essential time scale of the heating process. The thermal conduction is now determined by the Kapitza resistance to the bath. A very rough estimate gives $\tau_{\text{thermal}} \sim 10^{-3}$ s for this case. However, a prominent effect of the chopper frequency is not observed. If heating of the whole sample would take place with a constant temperature change independent of the applied voltage, Eq. (7) would yield a signal approxi-

mately linear in bias voltage at sufficiently large voltages. These effects are not observed either.

VI. CONCLUSIONS

The rectification measurements reported in this paper demonstrate that the measured large nonlinearities in the current-voltage characteristics of UPt_3 point contacts can be described using a heating model. Because of the small electron mean free path, an applied voltage leads to local heating in the contact area. This local heating involves a thermal relaxation which is much slower than the time scale imposed by the applied radiation, hence leading to rectification signals which differ from the second derivative d^2V/dI^2 at low frequencies. In a spectroscopic interpretation⁴ of point-contact data in UPt_3 , the applied voltage defines the characteristic energy for the nonequilibrium distribution of the electrons. In such a ballistic model, the fast scattering of the electrons would allow a detection of FIR radiation corresponding to the function d^2V/dI^2 .

Other metals with narrow-band phenomena (mixed-valence systems, heavy fermions) are good candidates for such rectification experiments. This type of experiment would yield important information about the time scale of the processes which determine the observed non-Ohmic behavior of point contacts in these systems.

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