MODULATION TEMPERATURE SPECTROSCOPY OF ELEMENTARY EXCITATIONS IN FERROMAGNETICS USING MICROCONTACTS

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The article considers the possibilities of the method of microcontact spectroscopy in relation to the relationship between the elastic and inelastic electron mean free paths l_i and l_e and the contact diameter d. Microcontact spectra of Ni, Fe, and Ni_xCu_{1-x} alloys over a wide range of energies are given and discussed.

INTRODUCTION

Investigation of the nonlinear properties of the volt-ampere characteristics (VAC) of metallic microcontacts can yield direct information on the phonon spectrum of the metal and the characteristics of electron-phonon interaction (EPI). This method of recovering the phonon spectrum has come to be known as microcontact spectroscopy [1-3]. It was noted in [1] that similar information can be theoretically obtained regarding any Bose branches of the spectrum of elementary excitations that interact with electrons, including magnons in ferromagnetic or antiferromagnetic metals. Whereas there have been a large number of studies of phonon spectra (see the bibliography in [3]), the investigation of electron-magnon interaction is only just beginning [4, 5].

The possibilities for spectroscopy of quasi-particle excitations using microcontacts are governed by the relationship between the elastic and inelastic electron mean free paths l_i and l_e and the contact diameter d. Figure 1 shows a schematic representation of a microcontact (MC).

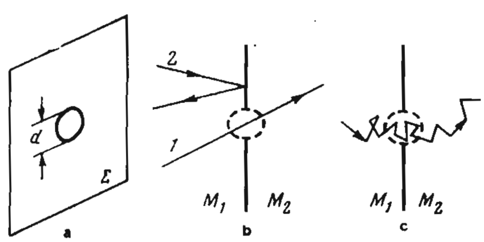

Fig. 1. Diagram of microcontact (a) and characteristic electron transit trajectories through opening in ballistic (b) and diffusion (c) modes; Σ is an infinitely thin screen with an opening.

The free path l_i is determined by interaction between electrons and impurities, and is usually less than the mean free path l_e corresponding to interaction between an electron and Bose excitations. The path l_e depends on the electron energy E associated with the voltage V applied to the microcontact (E = eV). Substantial variation in l_e and hence in the contact resistance R, begins when eV attains the characteristic excitation energy. In accordance with the theory developed in [2], the second derivative of the VAC of the

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microcontact is given by the function

$$G(\omega)=\frac{1}{(2\pi)^6}\int\frac{dS_p}{V_\perp}\int\frac{dS_{p'}}{V_\perp'}\sum_s|M_{q,s}|^2\delta(\omega-\omega_{q,s})K(\mathbf{p},\mathbf{p}')\Big/\int\frac{dS_p}{V_\perp}, \tag{1}$$

which has the meaning of the transport function of EPI (or other electron/quasi-particle interaction) and differs from the ordinary EPI function (for $\varepsilon(\mathbf{p})=\varepsilon_F$):

$$g(\omega)=\frac{1}{(2\pi)^3}\int\frac{dS_p}{V_\perp}\int\frac{dS_{p'}}{V_\perp'}\sum_s|M_{q,s}|^2\delta(\omega-\omega_{q,s})\Big/\int\frac{dS_p}{V} \tag{2}$$

by virtue of the form-factor $K(\mathbf{p},\mathbf{p}')$, which takes account of the microcontact geometry [ ]. The quantity $\omega_{q,s}$ in (1) and (2) is the Bose excitation dispersion law; $M_{q,s}$ is the matrix element of interaction between the corresponding excitations and electrons. Integration is performed over the Fermi surface of the metal.

Depending on the relationship between l_i, l_e and d, we will distinguish three limiting cases, which differ in terms of the nature of the trajectories of electron motion in the contact region.

1. **Ballistic mode.** The mean free paths l_i and l_e are large as compared to the contact diameter d,

$$l_i\gg d,\ l_e\gg d, \tag{3}$$

i.e., the Knudsen limit for the electron gas is realized. Condition (3) can be achieved relatively easily in experiments.

2. **Diffusion mode.** The elastic relaxation length is small as compared to the contact diameter d, whereas the latter does not exceed the electron energy scattering length, which is given for $l_i\ll l_e$ by the expression $\lambda=\sqrt{l_e l_i}$:

$$l_i\ll d\ll\lambda. \tag{4}$$

This situation obtains for disordered systems: amorphous or polycrystalline films, strongly deformed metals.

In cases 1 and 2 no energy dissipation occurs in the contact region, and heating effects play no major role.

3. **Thermal mode.** The elastic and inelastic relaxation lengths are small as compared to the contact diameter:

$$l\ll d,\ l_e\ll d. \tag{5}$$

In this case the excitation gas in the contact region becomes strongly nonequilibrium in nature, this corresponding to a change in the effective contraction temperature. This mode is easy to implement in practice if interaction between electrons and elementary excitations is strong.

For ferromagnetic metals cases 1 and 2 are achieved at low Curie temperatures, whereas case 3 evidently corresponds to ferromagnetics with large T_C values. In the ballistic and diffusion modes, the second derivative of the VAC of microcontacts makes it possible to determine the function $G(\omega)$ (with its value of K in each of the cases), whereas in the "thermal" mode the VAC depends in a more complicated fashion on $G(\omega)$. The first situation corresponds to the case of microcontact spectroscopy as it is traditionally understood [3], whereas the second makes it possible to achieve a kind of "modulation temperature spectroscopy" of elementary excitations in metals.

We investigated the MC spectra of iron and nickel, as well as of Ni_xCu_{1-x} alloys, over a wide range of energies (up to 0.3 eV). This made it possible to detect features associated with violation of magnetic order upon heating of the contact region above the Curie temperature, whose shape is governed by the nature of ρ(T) - the part of the resistance resulting from scattering by magnons, while the position on the energy axis is related in simple fashion to T_C. A theory of MC spectroscopy in the "thermal" mode is developed for this case.

EXPERIMENT AND RESULTS

The results to follow were obtained using clamp contacts of the "needle-anvil" type. The advantages of this system over film structures are discussed in detail in [3]. We employed monocrystalline Ni and Fe with a purity of 99.99-99.998%. Specimens of Ni_xCu_{1-x} with specified concentration were obtained by alloying the initial components in a high vacuum with subsequent prolonged annealing in order to make them homogeneous.

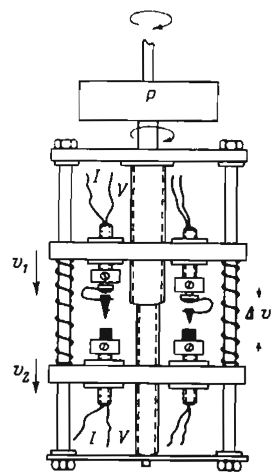

Fig. 2

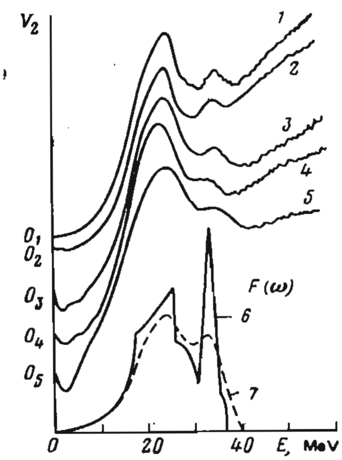

Fig. 3

Fig. 2. Device for creating point contacts of "needle-anvil" type; P is a reducer, $\Delta v = v_1 - v_2$.

Fig. 3. Microcontact spectra of nickel for specimens with differing resistance: 1) R = 2.5; 2) 7; 3) 10.5; 4) 9; 5) 10 ohms. In addition, 6 and 7 represent the phonon state density F(ω) as a function of energy, computed in [7] and obtained on the basis of neutron scattering in [8].

Electrodes of the requisite geometry were manufactured using a spark-cutting machine, after which they were treated by finer and finer grades of emery paper, and then electrically polished and etched. The radius of the resultant "needle" was a few microns. The electrodes were mounted in a special device, similar to that shown in Fig. 2, by means of which they were brought together until formation of a contact with the necessary resistance directly in liquid helium. The second derivative of the VAC was measured at the temperature of 1.5-4.2°K using the standard modulation procedure. The voltage of the second harmonic of the modulating signal V_2 was proportional to d^2V/dI^2 in this case.

The use of monocrystalline specimens of iron and nickel of high purity meant that the relationship $l_\varepsilon \geq d$ was satisfied in the region of phonon energies, and made it possible to obtain good EPI spectra in these metals. Figure 3 shows the MC spectra of Ni, as well as the phonon state density, as computed in (7) and obtained from inelastic neutron scattering data in [8].

The experimentally observed spreading of the MC spectrum as compared to the theoretical relationship F(ω) greatly exceeds the broadening resulting only from the effect of temperature and the modulating voltage. This makes it necessary to introduce "structural" broadening as well, related to structural differences in individual contact regions and to the effect of pressure and impurities. It was noted in [2] that experiments yield information from EPI that is averaged over a small contact region $\Omega \sim d^3$, and it is this region that frequently corresponds to the contaminated surface layer of the metal. The successive degradation of the spectra in Fig. 3 (curves 1-5) corresponds to increasing influence of these factors. Increasing the contact dimensions extends the effective phonon generation volume to a region of purer metal, but l_i comes to be on the order of or less than d, causing an increase in the background on the experimental curves (obviously related to heating of the contact).

Similar results were obtained in investigating the MC spectra of iron (Fig. 4). The theoretical relationship F(ω) shown in the figure [9] (curve 8), and the relationship obtained in no-model experiments involving slow neutron scattering [10] (curve 9), are in good agreement with our results.

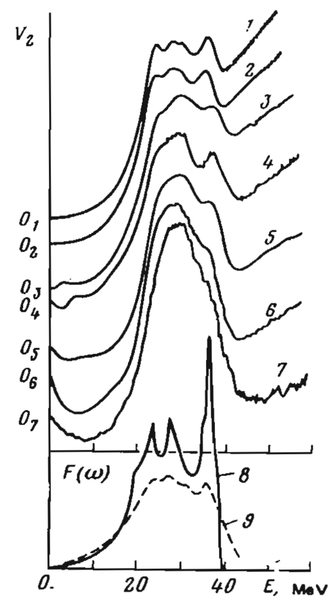

Fig. 4. Microcontact spectra of iron for specimens with differing resistance: 1) R = 3.8; 2) 4.7; 3) 3.6; 4) 4.3; 5) 7; 6) 5.5; 7) 50 ohms. In addition, 8 and 9 represent the phonon state density F(ω) as a function of energy, computed in [9] and obtained on the basis of neutron scattering in [10].

A feature of the MC spectra of iron and nickel is the high background level beyond the boundary of the phonon spectrum; unlike nonmagnetic metals, this background does not emerge onto some constant value but increases markedly as the energy increases. Figure 5 shows the MC spectra of Ni and Fe in the region of high energies (as compared to phonon energies). These spectra display anomalies at energies on the order of exchange energies, which cannot be accounted for by electron-phonon interaction.

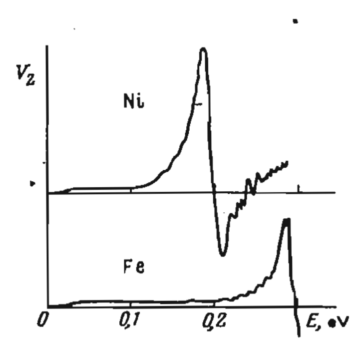

Fig. 5. Microcontact spectra of Ni and Fe in the range of energies of order kT_C.

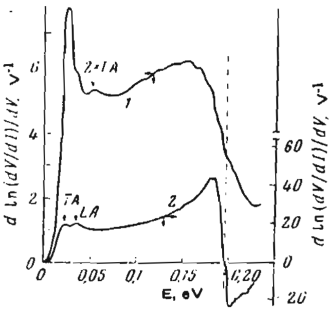

Fig. 6

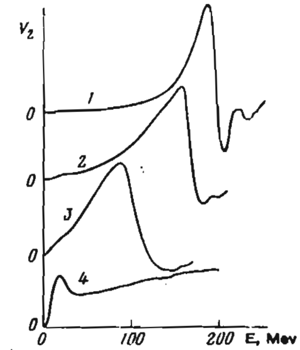

Fig. 7

Fig. 6. Logarithmic derivative of differential resistance of voltage for nickel contacts: 1) R_0 = 7.6 ohms; γ = 0.66; 2) R_0 = 1.8 ohms, γ = 0.91.

Fig. 7. Microcontact spectra of Ni_xCu_{1-x} alloys with differing concentrations of Ni: 1) x = 1; 2) 0.85; 3) 0.61; 4) 0.2.

The ratio of the voltages at which these anomalies occur corresponds roughly to the ratio of the Curie temperatures for iron and nickel, so that it is possible to interpret them as violation of magnetic order when the contact region is heated above T_C by transport current, under the assumption that for large energies the current conditions change from Knudsen conditions ($l_i \gg d$) to Maxwellian ones ($l_i \ll d$).

Moreover, as shown by Holm [11], under Maxwellian conditions the "softening" temperature may be attained, this amounting to 770-790°K for Ni and Fe. Therefore, beginning at voltages exceeding 0.22 eV, as a rule, irreversible shorting of the contacts occurs, so that measurements in the anomaly region of iron become extremely complicated. Accordingly, our primary experimental results were obtained for Ni.

Both the magnitude and shape of the feature observed in the spectrum correlate with the background in the region of phonon energies. The greater the coefficient γ characterizing the relative magnitude of the background [3], the greater the amplitude of the anomaly. As γ decreases the feature spreads out, and the second derivative no longer changes sign. This can be well illustrated by the relationship between the logarithmic derivative and the voltage for two nickel microcontacts with different R_0 and γ (Fig. 6). The logarithmic derivative

$$d \ln (dV/dI)/dV = (d^2V/dI^2)(dI/dV)^2 \qquad (6)$$

can be readily obtained if a constant modulation-signal level is maintained in the process of recording the spectrum.

We also made measurements using Ni_xCu_{1-x} alloy, for which the T_C depends on the concentration of the components. In particular, the alloy becomes nonmagnetic at a Ni content less than 47 at%. Figure 7 shows the results of measurements. It can be seen that as the Ni concentration decreases (i.e., as T_C drops) the anomaly shifts into the region of lower energies. Its relatively large spreading is obviously due to inadequate homogeneity of the alloy near the surface of the specimen. For the microcontact spectrum of the nonmagnetic alloy $Ni_{0.21}Cu_{0.79}$ we observe only features associated with the EPI spectrum, and in the region of higher energies, as expected, a weakly increasing background.

THEORY OF THERMAL MODE OF MC SPECTROSCOPY

Let us consider the "thermal" mode of contact in greater detail. We will assume that the nonequilibrium of the quasi-particle gas can be characterized by the local temperature T(r). The latter is determined by joint solution of the continuity equation for the electric current

$$\operatorname{div} \mathbf{j}=0 \tag{7}$$

and the continuity equation for the entropy, i.e., the thermal balance condition:

$$-\operatorname{div} \mathbf{q}+\mathbf{j}\mathbf{E}=0. \tag{8}$$

The electric current density **j** and thermal flux density **q** are, generally speaking, nonlinear functions of the field strength $\mathbf{E}=-\nabla\Phi$ and the temperature gradient ∇T, and also the local temperature of the medium $T(\mathbf{r})$. If the condition $l_i \ll l_e$ is satisfied, then the nonlinear parts of these expressions are small:

$$\mathbf{j}=-\sigma_i \nabla\varphi+\mathbf{j}', \quad \mathbf{q}=-\varkappa_i \nabla T+\mathbf{q}', \tag{9}$$

and the elastic kinetic coefficients are related by the Wideman-Frantz expression:

$$\frac{\varkappa_i}{\sigma_i T}=L, \tag{10}$$

where $L=\frac{\pi^2}{3}\frac{k^2}{e^2}$ is the Lorentz number.

In this approximation we can readily determine the distribution of the temperature and the electric field:

$$T^2(\mathbf{r})=T_{ed}^2+\frac{1}{L}\left[\frac{V^2}{4}-\Phi^2(\mathbf{r})\right], \tag{11}$$

$$\Phi(\mathbf{r})=\left(\frac{2}{\pi}\operatorname{arctg} e^{v}-\frac{1}{2}\right)V, \tag{12}$$

where T_{ed} is the temperature of the edges; v is the coordinate of an oblate spheroid of revolution [12]. In accordance with (11), the equipotential surfaces are ellipsoids of revolution (v-ellipsoids).

Distributions (11) and (12) can be simplified by considering, instead of the exact solutions for the opening, a "spherical spread-out" geometry [11], namely a radially symmetrical temperature distribution near the contact.

Assuming T_{ed} to be zero, we obtain the following simple expression for the contraction temperature (v = 0):

$$kT=eV/\beta, \tag{13}$$

where the numerical factor

$$\beta=2\pi/\sqrt{3}=3{,}63. \tag{14}$$

Thus, the heating temperature is directly related to the voltage applied to the contact. Since the characteristic establishment times for thermal equilibrium ($t_0\sim d^2/\chi$, χ is the thermal conductivity coefficient) are extremely small because of the small contact diameter ($t_0\sim 10^{-9}-10^{-10}$ sec), we have the possibility of virtually instantaneous conversion of temperature to electrical voltage. Measurement of voltage is technically a vastly simpler problem than measurement of temperature. Moreover, by modulating V by a small harmonic additional component, we obtain the possibility of automatically measuring the derivatives of various quantities with respect to temperature, which contain information on the energy distribution of quasi-particle states. This method of temperature modulation can be called "modulation temperature spectroscopy" of elementary excitations.

Using (11) and (12), we can obtain the following expression [4] for the current at the contact as a function of the voltage on its edges:

$$I(V)=Vd\int_0^1 \frac{dx}{\rho[T(V)\sqrt{1-x^2}]}, \tag{15}$$

where T(V) is given by (13); $\rho(T)$ is the temperature-dependent resistivity of the metal. For a ferromagnetic with T_C, it is convenient to write (13) in the form

$$T(V)=\frac{T_C}{V_C}V, \tag{16}$$

in which case the quantity

$$eV_C=3{,}63kT_C \tag{17}$$

yields the position of the feature of the VAC corresponding to the resistance anomaly at the Curie point on the voltage axis.

For the feature in the microcontact spectra of nickel, the ratio $eV_C/\kappa T_C$, as averaged over data for 25 specimens, amounts to 3.709, or very close to the computed value. For iron the ratio amounts to only 3.1, but, as already noted, for energies $\geqslant$0,22 eV we were able to obtain only individual measurements.

For Ni_xCu_{1-x} alloys the value of eV_C/kT_C is 4,37 ($Ni_{0,85}Cu_{0,15}$); 4,29 ($Ni_{0,73}Cu_{0,27}$); 6,36 ($Ni_{0,61}Cu_{0,39}$). This spread can be accounted for by a number of factors: insufficient heat treatment of the alloys; presence of ferromagnetic clusters [13], and hence differences between T_C and the calculated value; and deviations from the Wideman-Frantz law, particularly for low T_C.

Expression (15) can be regarded as an integral equation in the unknown function $\rho(T)$. A solution of this equation can be obtained by employing the Abel transformation;* in this case we obtain an equation for determining $\rho(T)$ on the basis of the known VAC:

$$\frac{V}{\rho(T)}=-\frac{2}{\pi d}\frac{d}{dV}\int_0^V I(\tau)\,d(\sqrt{V^2-\tau^2}), \tag{18}$$

where it is taken into account that $I(0)=0$.

We reconstructed the dependence of $d\rho/dT$ on T from microcontact characteristics. The results for $Ni_{0,85}Cu_{0,15}$ alloy are shown in Fig. 8.

Integral expression (15) makes it possible to obtain the explicit form of the nonlinear contribution to the VAC for the case in which it is small, i.e., $l_e \gg l_i$. Under the assumption that Mathiessen's rule is valid, the temperature-dependent part of the resistance is given by the formula

$$\rho_e(T)=\frac{4\pi m}{ne^2}\int_0^\infty G_{tr}(\omega)\Psi\left(\frac{\omega}{T}\right)d\omega, \tag{19}$$

$$\Psi(x)=\frac{x}{2\,\mathrm{sh}^2(x/2)}, \tag{20}$$

where G_{tr} has the structural factor $K=(\mathbf{p}-\mathbf{p}')^2/p_F^2$ in (1).

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*The authors are grateful V. M. Kirzhner, who called their attention to this fact.

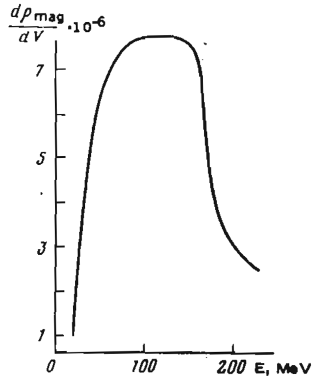

Fig. 8

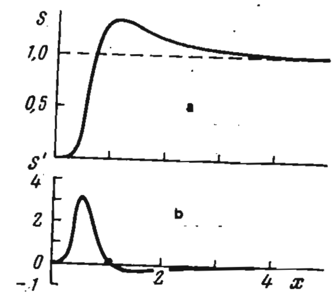

Fig. 9

Fig. 8. Temperature dependence of magnon part of resistance of $Ni_{0.85}Cu_{0.15}$ alloy. Temperature is converted to voltage in accordance with (13).

Fig. 9. Graphs of the functions S(x) and S'(x).

Substituting (19) into (15), we obtain the following expression for the second derivative of the VAC:

$$-\frac{d^2I}{dV^2}=c\int_0^\infty G_{tr}(\omega)S\left(\frac{eV}{\omega}\right)\frac{d\omega}{\omega}, \tag{21}$$

where c is a constant that is not of importance to us, while

$$S(x)=\frac{\beta^2}{2\pi}\frac{d^2}{dx^2}\int_0^{\pi/2}\frac{d\varphi}{\mathrm{sh}^2(\beta/2x\sin\varphi)}. \tag{22}$$

Function S(x) and its derivative S'(x) are shown in Fig. 9. S(x) has a diffuse maximum near the point x = 1. Therefore, despite the fact that the anomaly corresponding to T_C, in accordance with (17) occurs at a voltage $\sim 3{,}62T_c$, the features corresponding to magnon-spectrum frequencies ω_m, will occur for voltages $eV\approx\hbar\omega_m$, although they will be strongly diffuse.

Function S(x) has the following evident meaning: it indicates the form of the feature in the MC spectrum for a delta-function distribution of the excitation frequencies $G(\omega)\sim\delta(\omega-\omega_0)$. Given this distribution, the MC spectrum has the form shown in Fig. 9a, if we set $x=eV/\omega_0$. It is interesting to note that the third derivative d^3I/dV^3 of the VAC, which can be fairly simply recorded experimentally, will be approximately proportional to the transport function of electron-phonon or electron-magnon interaction, shifted into the low-frequency region (by roughly a factor of two) and made diffuse as a result of convolution with the function $S'\left(\frac{eV}{\omega}\right)$ (Fig. 9b):

$$\frac{d^3I}{dV^3}=-\mathrm{const}\int_0^{\omega_m}G_{tr}(\omega)S'\left(\frac{eV}{\omega}\right)\frac{d\omega}{\omega^2}. \tag{23}$$

CONCLUSIONS

Thus, we have shown that the volt-ampere characteritics of large-diameter contacts, which can readily be obtained experimentally, can also provide a source of useful information regarding electron-magnon or electron-phonon interaction. Since the contact diameter is much greater than the characteristic electron mean free paths, the result is independent of the contact geometry and can theoretically be obtained from a uniform specimen in which there is a current-created overheating region, and at a sufficient distance from it the metal temperature is maintained constant and fairly low. In practice, however, it is not easy to create such a situation. It can be realized most naturally and simply using microcontacts. Moreover, it is precisely the smallness of the hot region of the metal that makes possible fairly high-frequency temperature modulation in the contact region, and hence experimental implementation of modulation temperature spectroscopy of magnons and other Bose quasi-particle excitations in metals.

REFERENCES

1. I. K. Yanson, Zh. eksperim. i teor. fiz., vol. 66, p. 1035, 1974.
2. I. O. Kulik, A. N. Omel'yanchuk and R. I. Shekhter, Fiz. nizkikh temperatur, vol. 3, p. 1543, 1977; Solid State Communs., vol. 23, p. 301, 1977.
3. I. K. Yanson and I. O. Kulik, Proc. LT-15, Phys. 39, Colloque. C6, p. 1564, 1978; I. K. Yanson and I. O. Kulik, preprint, Physicotechnical Institute of Low Temperatures, AS UkrSSR, no. 24, Khar'kov, 1978.
4. B. I. Verkin, I. K. Yanson and I. O. Kulik et al., Solid State Communs., vol. 30, p. 215, 1979.
5. H. P. Leppin and D. K. Wohlleben, Preprint Physikalisches Institut der Universitat zu Koln, October, 1978.
6. I. O. Kulik and I. K. Yanson, Fiz. nizkikh temperatur, vol. 4, p. 1267, 1978.
7. R. J. Birgeman, J. Cardes, G. Dolling and A. D. B. Woods, Phys. Rev., vol. 136, p. 1359, 1965.
8. B. I. Gorbachev, P. G. Ivanitskii, V. T. Korotenko and M. V. Pasechnik, Ukr. fiz. zh., vol. 18, p. 1528, 1973.
9. J. Bergsma, C. van Dijk and D. Tocchetti, Phys. Letts., vol. 24A, p. 270, 1967.
10. B. I. Gorbachev, P. G. Ivanitskii, V. T. Korotenko and M. V. Pasechnik, Ukr. fiz. zh., vol. 18, p. 1528, 1973.
11. R. Holm, Electrical Contacts [Russian translation], Izd-vo inostr. lit., Moscow, 1961.
12. E. Madelung, The Mathematical Apparatus of Physics [Russian translation], Moscow, p. 278, 1961.
13. C. J. Tranchita and H. Claus, Solid State Communs., vol. 27, p. 583, 1978.

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