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ON THE THEORY OF ANOMALOUS TUNNELING DUE TO PARAMAGNETIC IMPURITIES

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Summary

Recently some anomalies have been observed in the characteristics of tunnel diodes at zero bias. Anderson and Suhl have called attention to the resonance scattering of electrons on paramagnetic impurities in the oxide layer. The tunnel current is calculated by summing up the contributions of single resonant scatterings. We have accepted the expression of the resonant scattering amplitude calculated by Abrikosov. The effective density of states is determined appearing in the formula of tunnel current.

The final results are:

The effect of paramagnetic impurities is the decrease of density of states in all of the cases. The actual appearance of the effect has a great variety strongly depending on the parameter values:

1, for ferromagnetic coupling:
resistivity minimum at zero bias /relative amplitude: 0-0,2/

2, for antiferromagnetic coupling:

$2/_a$ giant resistivity maximum at zero bias /relative amplitude 0-1oo/

$2/_b$ local resistivity minimum /relative amplitude 0-0,2/ superimposed on a background curve with resistivity maximum /type $2/_a$/.

The investigation of tunneling characteristics may be a powerful method to check the resonant scattering amplitude for paramagnetic impurities.

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Recently dynamical resistance maxima and minima at zero bias have been observed in some metal - metal oxide-metal[1,2] and semiconductor[3,4] tunnel junctions. Anderson[5] and Suhl[6] suggested that these anomalies are due to paramagnetic impurities in the oxide layer. The scattering of electrons on the impurities in the insulator shows the Kondo anomaly[7] which is the result of resonance scattering[8,9]. This scattering has a great influence on the electron wave function in the barrier and therefore on the tunneling current, too. The tunneling current was calculated by Appelbaum[10] and Zawadowski[11] treating the scattering in the third-order of the perturbation theory. These approximations can explain only small effects and the cutoff energy chosen to fit the experimental data is much smaller /10 meV/ than the usual cutoff energy i.e. the Fermi energy /1-10 eV/. In some actual cases the relative amplitude of the resistance maximum is about 5-50[2,4]. This shows that the scattering on the impurities has to be taken into account in a more appropriate way, avoiding perturbation theory.

In the theory given here the effective density of states appearing in the expression of current is always smaller than the unrenormalized one and it would have quite different character depending on the coupling and energy. Therefore it might result in great resistance maximum or small conductance maximum as well.

We use the Hamiltonian proposed by Kondo[7]

$$H = -\frac{J}{N}\vec{S}\,\psi^{+}(\vec{R})\,\vec{\sigma}\,\psi(\vec{R}) \qquad (1)$$

where $\vec{S}$ is the spin operator of the paramagnetic impurity and $\psi^{+}(\vec{R})\vec{\sigma}\psi(\vec{R})$ is the spin density of conduction electrons at the position of the impurity.

One of the authors has elaborated a particular theory[12] of tunneling between superconductors to take into account the motion of electrons in the barrier, too. This theory may be applied here. The proposed approach starts with the so-called "left and right side problems". The Green's functions of the left, right and original problems /G_l, G_r, G/ are determined by the potentials V_l, V_r and V given by Fig.1. and by the complete mass operator due to the scattering on paramagnetic impurities. The Green's functions are calculated in a self-consistent way. The tunneling current may be calculated using the one-particle Green's functions of the particular problems. The corresponding diagrams are as follows

$$E_0 = E_c \exp\left(\frac{N}{7\rho\chi^2}\right) \tag{5}$$

where E_c is the cutoff energy and χ is the amplitude of the wave function at the position of the impurity. In a selfconsistent calculation ρ must be replaced by ρ_{eff} /see later/.

At finite temperatures a homogeneous function of E and T might stand instead of $\log \frac{E_0}{|E|}$ and sign E is replaced by 1-2n/E/[15].

If the space variables of the Green's functions are taken in the barrier at the same point, the spectral density function of the Green's function[16] is

$$\rho_{eff}(E) = \rho\, Z(E) \tag{6}$$

where

$$Z(E) = \frac{\mathrm{Im}\, G(E)}{\pi\rho(2n(E)-1)} = \frac{1}{1 + \mathrm{Im}\, G^{(0)}(E)\, \mathrm{Im}\,\Sigma(E)} \tag{7}$$

Z/E/ depends only on the wave vector laying in the plane of the barrier $\vec{K}$ and the energy variable K. In the calculation of the tunneling current it may be taken at K=0.

The well known formula for the tunneling current is as follows

$$I(V) = C\int_{-\infty}^{\infty} \rho_{eff,l}(E)\,\rho_{eff,r}(E+eV)\left[n(E+eV) - n(E)\right] dE \tag{8}$$

where C is a constant, V is the applied voltage and ρ_{eff}/E/ is defined in /6/. According to this the effect of impurities may be taken into account as a formal renormalization of the density of states.

A straightforward calculation gives the following form for Z/E/

$$Z(E) = \frac{\left(\log\frac{E_0}{|E|}\right)^2 + \kappa^2}{\left(\log\frac{E_0}{|E|}\right)^2 + \kappa^2 + a^2} \tag{9}$$

where $a^2 = S(S+1)\frac{\pi^2}{4}\frac{N_i}{N_s}$ /N_i and N_s are the number of impurities and the total number of atoms on the surface of the barrier, respectively./

The effective coupling constant of the scattering on the impurity $\frac{\mathcal{J}\varrho\chi^2}{N}$ is strongly dependent on the position of the impurity. The amplitude of the wave function sharply decreases in the barrier and therefore only impurities on the surface give considerable contribution[17]. A rough estimation gives that $a^2 \sim 10 - 20$, $\frac{\mathcal{J}\varrho}{N} \sim 0{,}05$[7] and $E_c \sim 1 - 10$ eV.

The current is determined by the renormalization factor Z/E/ /see equations /6/, /8/ / and therefore we are going to discuss the behaviour of Z/E/ at different values of the parameters.

1/ In the case of ferromagnetic coupling /$J > 0$/ $E_o \gg E_c \gg T$ /$k_B=1$/. The function Z/E/ is schematically plotted against E in Fig.2/a/. In the interval $0<E<E_c$ Z/E/ is a decreasing function and at $E=E_c$ it is

$$Z(E_c) = 1/\left[1+\left(a\frac{\mathcal{J}\varrho\chi^2}{N}\right)^2\right]$$

In the interesting region of the energy $Z>0{,}8$. In this case we have a conductance maximum which is not larger than 25% /see Fig.3/a/ /.

2/ In the case of antiferromagnetic coupling /$J<0$/ $E_c \gg E_o, T$. The function Z/E/ is plotted in Fig.2/b/. This curve has a minimum at $E=E_o$, where $Z(E_o) = \frac{\kappa^2}{a^2+\kappa^2} \sim 0{,}1$. This deep minimum in the density of states causes a maximum in the resistivity. The position and character of this maximum is very sensitive on the value of E_o, T and the applied voltage /V/.

2.a/ If $E_o \ll T$, the peak in Z/E/ at zero energy may be neglected and the function Z/E/ might be replaced by the dotted line in Fig. 2/b/. In this case the resistivity maximum appears at zero bias. The relative amplitude of this maximum is proportional to $Z^{-2}/E_o/$ or $Z^{-1}/E_o/$ depending whether the paramagnetic impurities are on both surfaces of the barrier or only on one side of it. In the first case this amplitude may reach the value about 100./ Fig.3/b//.

2.b/ If $E_o \geq T$, at very small value of the bias the peak at zero energy has to be considered. This maximum in Z/E/ causes a local maximum of the conductance at zero bias. At larger values of the voltage the minimum at E_o in Z/E/ gives a maximum in the resistivity /Fig.3/c//. This means that the maximum in the conductance at zero bias is superimposed on a background /dotted lines in Fig.3 /d// which has a minimum there. This background is determined by the modified Z/E/ function which is represented by the dotted line in Fig. 2/b/. The maximum depends on the temperature while the background is only slightly dependent on it.

It is worth mentioning that conductance maximum at zero bias can occur for ferro- and antiferromagnetic coupling as well /cases 1. and 2.b/. But resistivity maximum can occur only in the case of antiferromagnetic coupling /2.a./ In the case of conductance maximum the ferro- and antiferromagnetic cases cannot be distinguished on the basis of the characteristics at small bias, there is however, a great difference in the characteristics at large bias.

The measurement by Rowell and Shen[2] on Cr-I- /Ag, Pb/ shows a resistance maximum with a relative amplitude about 5o. This great value can be understood if we assume that there is an antiferromagnetic coupling between the conduction electrons and impurities with $E_0 \sim 0,1-0,2$ meV. If $E_c \sim 1 - 1o$ eV, we get a reasonable value for the coupling constant, $\frac{3\rho\chi^2}{N} \sim 0,1$.

The explanation of the occurrence of conductance maxima in a series of diodas [1,2] is much more dubious. Only the investigation of the behaviour of the background curve may give possibilities to distinguish between the two cases. It seems to us not very unreasonable to interpret the Ta-I-Ag and Ta-I-Al curves as the result of antiferromagnetic coupling with relatively large value of E_0 /$E_0 \sim 3-6$ meV/. The value of $\frac{3\rho\chi^2}{N}$ is roughly in the same range as before.

Similar phenomena have been observed investigating semiconductor tunnel junctions[3,4]. It is possible that similar effect may occur in the junction region. In these cases there are also two different groups of measurements with conductance and resistance maxima, respectively. The resistance maxima may have a very large relative amplitude /about 5-25/, on the other hand the conductance maxima are only about 1o%, similarly as in the above discussed cases.

Mention must be made that in this consideration we have made use only the main features of Z/E/, in this way that of the imaginary part of the self energy. With the aid of this type of measurements the imaginary part of the self energy /the life time/ may be experimentally investigated in that region of energy which is not available by the simple resistivity measurements on bulky dilute magnetic alloys. We think that measurements on junctions composed of metal-metal oxide - about one atomic layer of paramagnetic impurities - metal would be very interesting to compare their results with the present theory.

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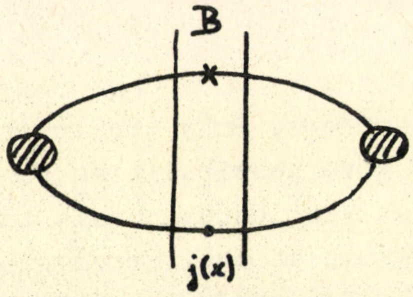
Appendix

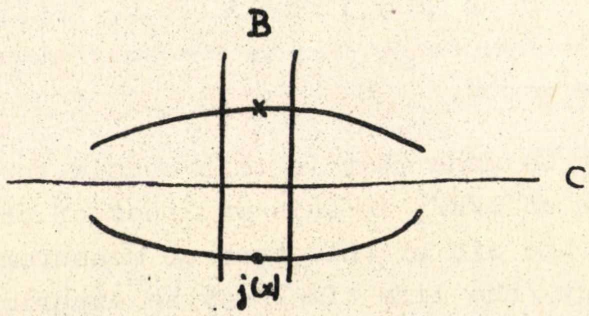
Remarks on Appelbaum's calculation of the anomalous current

The general diagram for the tunneling current density is /11/



/I/

In some cases the total current may be calculated from the transition matrix elements corresponding to the tunneling current by using the "golden rule". It is a possible way of calculation if the previous diagram /I/ can be cut into two parts, which correspond to single particle tunneling through the barrier. It may be represented symbolically by the diagram:



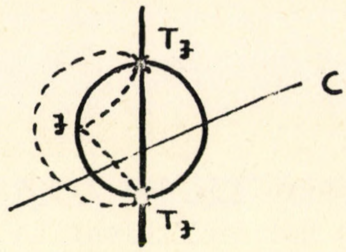
/II/

where line C is the cutting line.

The product of the matrix elements of these single particle tunnelings appears in the "golden rule formula".

This procedure can be applied to non-interacting electron gases without any further considerations. In the case of superconductivity this method does not work, because it gives only the one-particle current and fails to account for the Josephson current. In the second case we have to go back to e.g. the calculation of the original diagram /I/.

Appelbaum has calculated the anomalous current due to paramagnetic impurities. The diagram calculated by him is in Abrikosov's notation:



/III/

where T_J stands for the spin dependent tunneling coupling and J for the simple interaction with spin. He has calculated the contribution of this diagram by the application of the "golden rule". If this diagram is cut into two parts by line C, then the two particular scattering cannot be interpreted as single electron scatterings through the barrier, because there are spin lines, too. The electron and the fictitious spin particles corresponding to the lines cut by C are not necessarily on the energy shell and this fact makes the application of the "golden rule" very ambiguous.

We have calculated in our paper the tunneling current in the absence of external magnetic field and we have got similar current expression as Appelbaum[10].

On the other hand we have calculated the tunneling current in the presence of magnetic field /to be published/ and our results show disagreement with the results derived by Appelbaum[10]. E.g. in the third order of perturbation theory in the conductance we have got terms proportional to $\langle M^3 \rangle$ /where M is the magnetization of the localized spin/, too.

We may conclude that the application of the "golden rule" to the calculation of the tunneling current is very ambiguous in the case of interacting electron gas. In some cases it gives wrong results: e.g.1, Josephson current, 2, anomalous current due to paramagnetic impurities in external magnetic field. In the latter one the magnetic splitting of spin energies probably makes the energy variables of lines cut by line C much more important than in the field-free case and that may be the reason why the difference between the results calculated by different methods occurs only in the case of external magnetic field.

R E F E R E N C E S

1/ F.F.G.Wyatt, Phys.Rev.Letters 13, 401 /1964/
The experimental data are not sufficient to determine the mathematical formula for the temperature dependence of $\Delta G(0)$ unambiguously.

2/ J.M.Rowell and L.Y.Shen, Phys.Rev.Letters 17, 15 /1966/.

3/ R.A.Logan and J.M.Rowell, Phys.Rev.Letters 13, 404 /1964/.

4/ B.M.Vul, E.J.Zavaritsky and N.V.Zavaritsky, Sol.State Phys. /Russ./ 8, 888 /1966/.
According to N.V.Zavaritsky's private communication they have found resistance maximum about 1000-2600% in PbTa and PbSe semiconductor tunnel junctions.

5/ P.W.Anderson, Phys.Rev. Letters 17, 95 /1966/.

6/ H.Suhl, Lecture at the 1966 International School of Physics "Enrico Fermi", Varenna, Italy.

7/ J.Kondo, Progr. Theoret, Phys. /Kyoto/ 32, 37 /1964/.

8/ A.A.Abrikosov, Physics 2, 5 /1965/.

9/ H.Suhl, Phys.Rev. 138, A515 /1965/, 141, 483 /1966/.

10/ J.Appelbaum, Phys.Rev.Letters 17, 91 /1966/. He supposed that only the spin dependent terms of the tunneling Hamiltonian are important in calculating the current and therefore he has got a wrong dependence on the coupling constant / $\frac{\Delta G}{G_0} \sim J$ instead of J^3 in the third order of the perturbation theory/. Using the tunneling Hamiltonian method it would be difficult to sum up all the diagrams considered by us. This calculation was done by the "golden rule" method. The applicability of this method is discussed in the Appendix.

11/ A.Zawadowski, Proceedings of International Conference on the Tenth Low Temperature Physics, Moscow, /1966/ /to be published and KFKI preprint 10/1966/.

12/ A.Zawadowski, to be published, KFKI preprint 10/1966.

13/ J.Bardeen, Phys.Rev. Letters 6, 57 /1961/, 9, 147 /1962/.

14/ K.Yosida and A.Okiji, Progr.Theoret, Phys. /Kyoto/ 34, 505 /1965/.

15/ The homogeneous dependence of $\mathrm{Im}\,\Sigma$ on E and T may be responsible for the similarity of the voltage dependence or R/V/ and the temperature dependence of R/O/ in some region of the variables.[2]

16/ V.L.Bonch-Bruevich and S.V.Tyablikov, The Green Function Method in Statistical Mechanics, /North-Holland Publishing Company, Amsterdam, /1962/.

17/ A detailed discussion of the validity of this approximation will be published later.

Figure captions.

Fig.1. The potential of the right /a/, left /b/ and the original problem with the barrier B /c/.

Fig.2. Schematic plot of the renormalization factor against the energy in the case of ferro- /a/ and antiferromagnetic /b/ coupling. In Fig. 2/b/ the dotted lines represent a good approximation of Z/E/ if $E_0 \ll T$ /the background curve/.

Fig.3. Schematic plot of voltage dependence of conductance or resistance. /a/ The conductance for ferromagnetic coupling. /b/ Resistance for antiferromagnetic coupling if $T \gg E_0$. /c/ - /d/ Resistance and conductance for antiferromagnetic coupling if $T \leq E_0$. The dotted lines represent the background curve.

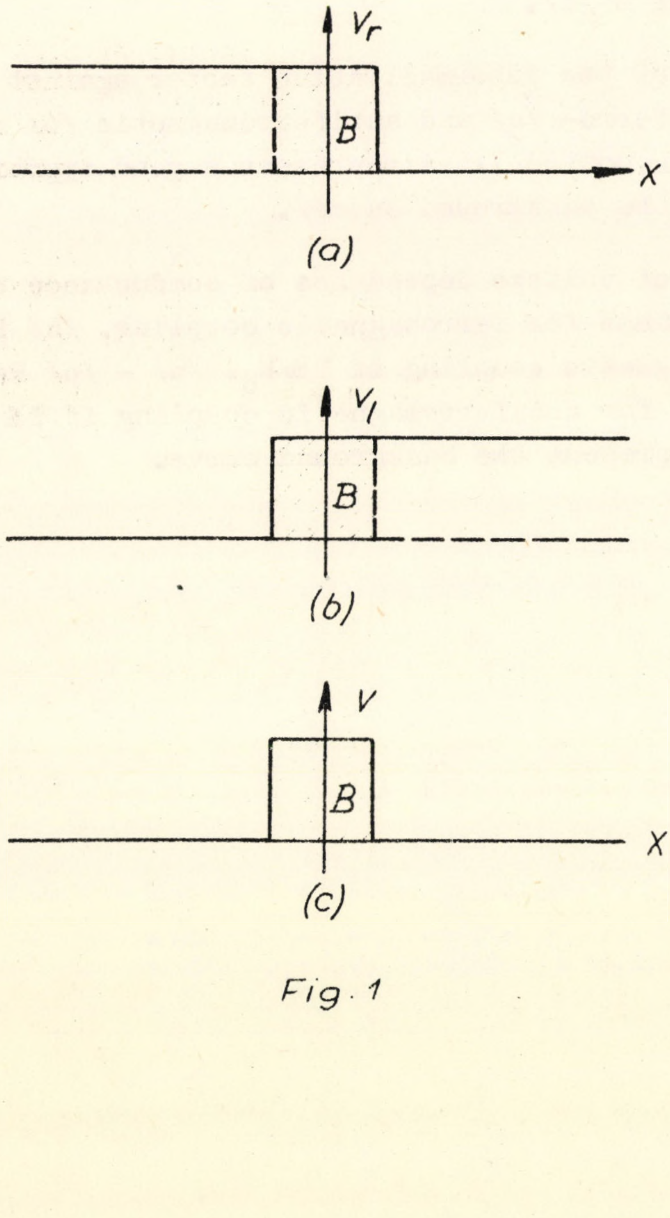
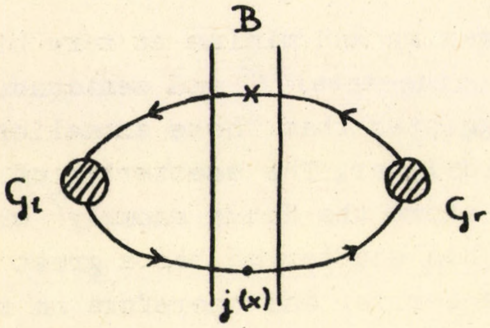
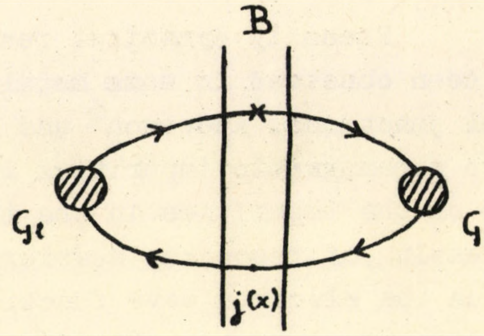


Fig. 1



and



where the cross in the barrier /B/ denotes the current coupling of the Green's functions, ontroduced by Bardeen[13] and the dot stands for the current operator.

The corresponding formula is

$$j(x) = (C \to R)\, e \left\{ \int_S d\vec{f}_{y,\beta} \int dy_0 \times \lim_{x \to x'} \left(\frac{\vec{\nabla}_x - \overleftarrow{\nabla}_{x'}}{2m} \right)_\alpha G_l(x,y) \left(\frac{\vec{\nabla}_y - \overleftarrow{\nabla}_y}{2m} \right)_\beta G_r(y,x') - r \leftrightarrow l \right\} \qquad (2)$$

where S denotes an arbitrary surface in the barrier. The expression of the current may be regarded as a response function of the coupling of the two different Green's functions and the operation /C→R/ denotes the replacement of the causal response function in this formula by the retarded one.

The Green's functions may be written as

$$G_\alpha = G_\alpha^{(0)} \frac{1}{1 - \Sigma_\alpha G_\alpha^{(0)}} \qquad (\alpha = l, r) \qquad (3)$$

where Σ_α is the self energy due to scattering on paramagnetic impurities. In our approximation the interaction between the impurities and the interference effects are neglected. We use the self energy obtained by Abrikosov[8] summing up a very wide class of diagrams in order to describe the resonance scattering.

$$\mathrm{Im}\, \Sigma(E) = -\frac{1}{2} \pi \rho\, S(S+1)\, \mathrm{sign}\, E \, \frac{1}{\left(\log \frac{E_0}{|E|}\right)^2 + \kappa^2} \qquad (4)$$

where ρ is the density of states at the Fermi energy, κ is a number of order unity as estimated by Yosida and Okiji[14].

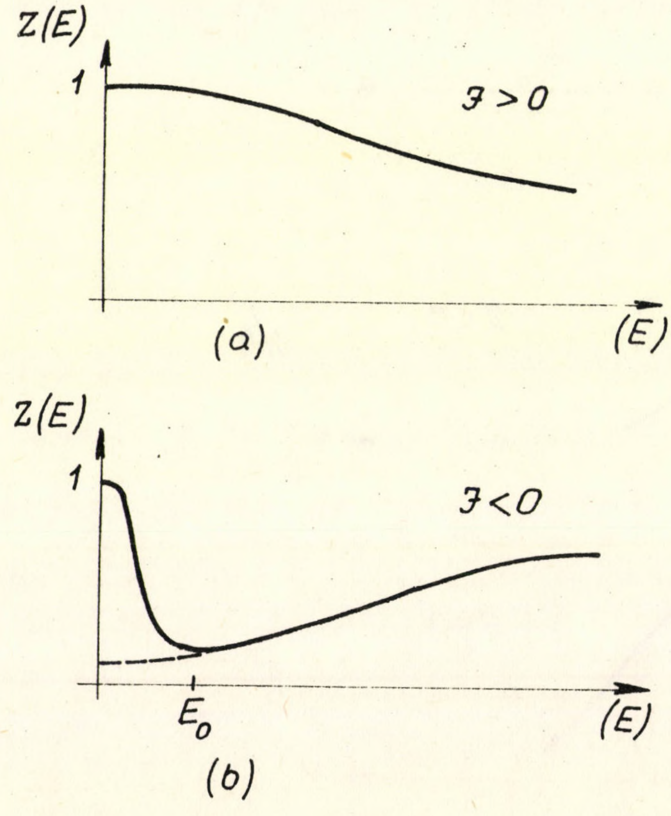


Fig. 2

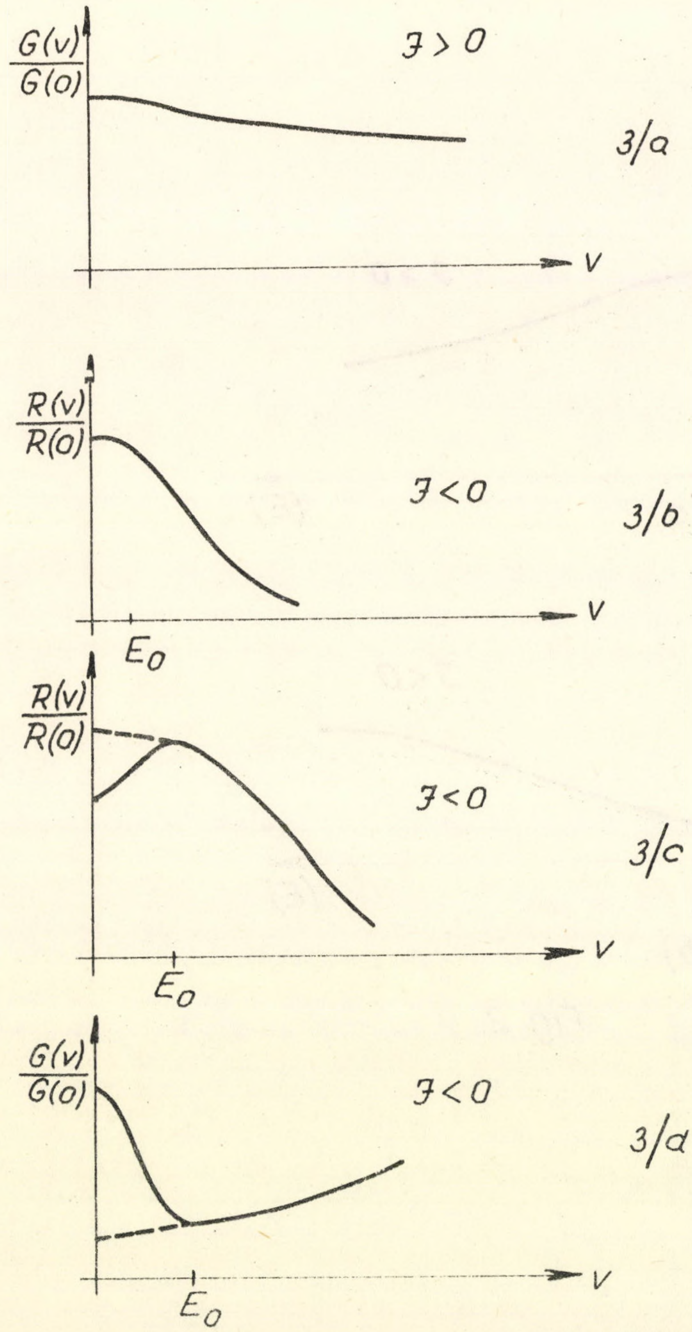


Fig 3