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# SEMIPHENOMENOLOGICAL MODEL FOR THE RESONANCES AND CHARGE NEUTRALITY IN DILUTE MAGNETIC ALLOYS

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ABSTRACT

The optical, impurity resistivity and charge oscillation amplitude data for Cu- and Al-based alloys are analyzed on the basis of a semiphenomenological model of the resonances, in which a narrow resonance of Abrikosov--Suhl type formed at the Fermi level is superimposed on the broad background reflecting the d-level structure. The experimental data fit well the assumption that the amplitude of the narrow resonance is determined by the charge neutrality limit calculated using the Friedel sum rule, and in both alloy systems the d-level structure is more or less split.

РЕЗЮМЕ

Был проведен анализ результатов оптических измерений, а также значений сопротивления примеси и амплитуды осцилляции заряда, измеренных в сплавах на медной и алюминиевой базе, при помощи полуфеноменологической модели резонанса, в которой учитываются узкий резонанс типа Абрикозова-Сула и структура уровня "d". Экспериментальные данные подтверждают предположение, что амплитуда узкого резонанса определяется предельной нейтральностью заряда, высчитанной по правилу суммирования Фриделя, а также что структура уровня "d" более или менее расщеплена.

KIVONAT

A réz és aluminium alapu ötvözetekben végzett optikai, ellenállás és töltésoszcillációs amplitudó méréseket analizáltuk a rezonanciák fél-fenomenologikus modelljével, melyben az Abrikozov-Suhl tipusu keskeny rezonancia és a d-nivó szerkezete van figyelembe véve. A kisérleti adatok jól egyeznek azzal a feltevéssel, hogy a keskeny rezonancia amplitudóját a töltéssemlegességi limit és a Friedel összegszabály határozza meg, valamint hogy a d-nivó szerkezete többé-kevésbé felhasadt.

During the last ten years, great experimental [1] and theoretical [2] efforts have been concentrated on the problem of resonances induced by transition metal impurities in nonmagnetic metallic hosts. Two different kinds of resonances appear: first, the broad resonance /b.r./, consisting of atomic d-levels essentially broadened due to interaction with the conduction electrons [3] and, secondly, the narrow resonance /n.r./ of Abrikosov-Suhl [4] type formed symmetrically at the Fermi level. In the Hartree-Fock /HF/ approximation of the Anderson model the b.r. has a single /double/ Lorentzian form in the non-magnetic /magnetic/ limit. In the theoretical treatments of the n.r. there occur typical logarithmic terms known as the Kondo anomaly. The aims of the present letter are (i) to present systematically the experimental data providing information on these resonances; (ii) to suggest a simple semiphenomenological model in which both resonances are of importance; and finally (iii) to analyze these experimental data in the light of the model.

(i) Optical and photoemission measurements give direct information on the structure of the b.r. Experiments on AgMn and CuMn alloys [5,6] reveal a double-peaked structure at room temperature, showing that these alloys are in the magnetic limit, with /in the usual notation of the Anderson model/ $U \cong 5$ eV and $\Delta \cong 0.5$ eV. In CuNi [7] the resonance has a single Lorentzian form corresponding to the nonmagnetic limit, with $\Delta \cong 0.4$ eV. No equivalent experiments have been performed yet for Al-based alloys so we can only estimate $U$ and $\Delta$. Being an intraatomic property, $U$ cannot be very different in this case from the value measured in AgMn, while the width of the resonance should be about twice as large as in noble metal hosts, due to the larger density of states of Al. With this estimate $\Delta \cong 1.0$ eV and $U/\pi\Delta \sim 1.5$, placing AlMn, for example, in the magnetic limit [1].

The impurity resistivities $\Delta R$ measured at room temperature in noble metals [1], as well as preliminary high temperature measurements on Al-based alloys [8], show a double-peaked distribution versus the impurity atomic number, $N$ which suggests the development of a double-peaked b.r. in the middle of the series. The impurity resistivities at zero temperature /i.e. well below the characteristic /Kondo/ temperature $T_K$/ for $Al^9$-, $Cu^{10}$-, and $Au^1$-based alloys are shown in Fig. 1/a/. The behaviour of $\Delta R$ versus the atomic impurity number shows a remarkable similarity for the different hosts and can be fitted adequately by the expression

$$\Delta R = R_o 5 \sin^2 \frac{N\pi}{10} \qquad /1/$$

where $R_o$ is a constant characterizing the host and the values of $N$ are given in Fig. 1.

The temperature dependence of the impurity resistivity which exhibits a $\Delta R(T) = \Delta R(O) \left[1 - \left(T/\Theta_\rho\right)\right]^2$ form at $T << T_K$ |9|,|11| reflects both the energy and temperature dependence of the scattering amplitude $t^T/\omega/$ for temperature $T$ and energy $\omega$. The standard Sommerfeld expansion of the conductivity gives

$$R(T) \sim \mathrm{Im}\, t^T(\varepsilon_F) + \frac{\pi^2 k_B^2 T^2}{\sigma} \frac{\partial^2 \mathrm{Im}\, t(\omega)}{\partial \omega^2}\Big|_{\omega=\varepsilon_F} + \ldots \quad . \qquad /2/$$

The experimental findings have been analyzed [9] on the assumption that below $T_K$ a narrow resonance is formed at the Fermi level, and

$$t(\omega) = \frac{-1}{\pi \rho_o} \frac{\Gamma}{\omega + i\Gamma} \qquad /3/$$

where the width $\Gamma$ is taken to be temperature independent. Using Eqs./2/ and /3/, $\Gamma = \frac{\pi}{\sqrt{3}} \Theta_\rho$, where $\Theta_\rho = 22^oK$ for CuFe [9] and $530^oK$ for AlMn [11]. This concept seems to describe other transport properties as well [9], although the temperature dependence of $t(\omega)$ cannot be ruled out.

The charge perturbation around the impurities likewise reflects both the energy and temperature dependence of $t(\omega)$, but in this case the two contributions can be separated [13]. The perturbation is given by

$$\Delta\rho(r) = \frac{(2\ell+1)m^{*2}}{(2\pi r)^2} \mathrm{Im} \int n_F(\omega)\, t(\omega) \exp\left[i\left(2kr - \ell\pi\right)\right] d\omega \qquad /4/$$

where $m^*$ the effective mass $n_F(\omega)$ is the Fermi function and the momentum $k_\omega$ belongs to energy $\omega$. The energy dependence of the scattering amplitude is manifested in the preasymptotic behaviour of the charge perturbation [14]. Within a coherence length given by $\xi_\Gamma = \frac{v_F}{2\Gamma}$ /where $v_F$ the Fermi velocity/, in the case of a scattering amplitude taking the form of Eq./3/ the amplitude of the charge perturbation is reduced compared with the prediction of the asymptotic form given by [13]

$$\Delta\rho(r) = \frac{(2\ell+1)}{2\pi^2 r^3} \mathrm{Re}\left[t^T(\varepsilon_F) \exp\left(2ik_F r\right)\right] \xi/\mathrm{sh}\xi \qquad /5/$$

where $\xi = \frac{2\pi r}{\beta k_F}$ at temperature T. Expressing the scattering in terms of phase shift $\eta_2(\omega)$, in the asymptotic region at $T = O^oK$ we get

$\Delta\rho(r) = \frac{-\alpha}{2\pi^2 r^3}\cos(2k_F r + \varphi)$ , with $\alpha = 5\ \sin\eta_2(\varepsilon_F)$ and $\varphi = \eta_2(\varepsilon_F)$.

The oscillation amplitudes around 3d-transition metal impurities in Al can be evaluated from the first order quadrupole wipe-out numbers [15] by the method described in [16]. Fig. 1/b/ shows the $n\ T = 0$ values and the corresponding oscillation amplitudes [17] together with the first order wipe-out numbers measured at $T = 420^{\circ}K$. The high-temperature behaviour of $n$ shows a double-peaked distribution, which resembles the situation occurring in Cu-based alloys[18] and therefore suggests the onset of development of a double-peaked b.r. The low-temperature values have been fitted to $\alpha = 5\ \sin\frac{N\pi}{10}$ . The agreement is good for Ti,V and Fe impurities, while for Mn and Cr the measured values are somewhat smaller than the computed ones. By comparing the difference between the measured and computed $\alpha(T=0)$ values for AlMn with the theory of the preasymptotic behaviour, one can estimate the width of the scattering amplitude to be $\Gamma \sim 0.5$ eV, assuming the form of Eq./3/ [14].

Turning to the temperature dependence of the charge perturbation, it can be shown [15] that the distance-dependent reduction of $\Delta\rho(r)$ represented by $\xi(sh\xi)^{-1}$ in Eq./5/ can be neglected, and so $\alpha(T)$ reflects the temperature dependence of $t(\varepsilon_F)$. The experimental results for AlMn can be at low temperatures represented by the formula $\alpha(T) = \alpha(0)\left[1-(T/\Theta_\alpha)^2\right]$, with $\Theta_\alpha = 860^{\circ}K$ [15]. By comparing this temperature dependence with that of the impurity resistivity [11] given by Eq./2/, the temperature dependence of $t(\varepsilon_F)$ can be eliminated, and we get $\Gamma \sim 0.14$ eV for a Lorentzian resonance [15]. This estimation is in good agreement with the values derived from the specific heat, superconducting transition temperature and dHvA effect [19].

The discrepancy between the two different estimations of the width of the resonance, both of them based on the assumption of a Lorentzian form, strongly suggests that the resonance in fact has a sharper top and longer tails than a Lorentzian one. The behaviour at the top of the resonance is sampled by the temperature dependence of $\alpha$ and $\Delta R$. From the theory of the preasymptotic behaviour [14] it follows that the charge oscillation amplitude at a distance $R_c$ should be sensitive only to such structures in the scattering amplitude which occur in an energy range not much smaller than $\Delta_c = \frac{V_F}{2R_c}$ /i.e. $\xi_{\Delta_c} \sim R_c$/. In the present case $R_c = 20\text{Å}$, corresponding to the measured first order wipe-out numbers, and so $\Delta_c \sim 1.0$ eV.
The relatively large value of $\alpha(T=0)$, and hence the large $\Gamma$ derived from it on the assumption of a single Lorentzian resonance shows that the main part of the scattering amplitude is distributed in energy regions larger than $\Delta_c$.

(ii) Working in the framework of the Anderson model [3] , we have developed a semiphenomenological model, in which the scattering amplitude $t(\omega)$ of the conduction electrons scattered by the impurity with angular momentum $\ell = 2$ and the density of states of the d-level $\rho_d(\omega)$ are connected by $\mathrm{Im}\{t(\omega)\} = V_{kd}^2 \rho_d(\omega)$. The following assumptions are made:

(I) The density of the impurity d-level $\rho_d(\omega)$ exhibits the structure of d-levels on a wide energy scale /b.r./. It shows two well separated peaks, in noble metal hosts, as given by the HF treatment in the magnetic limit [3], whereas for the Al host, it displays two less separated peaks, which may be regarded as a HF result strongly perturbed by correlations.

(II) At zero temperature the charge localized at the impurity is completely screened. Thus the Friedel sum rule holds and, furthermore, the dominating phase shifts correspond to d-like symmetry. According to the susceptibility data, the ground state is nonmagnetic /spherically symmetrical/, as in the Kondo [20] and spin fluctuation models [21], so that all of the phase shifts for the dominating channel $\ell = 2$ taken at the Fermi level are the same at $T = 0^{\circ}K$. It immediately follows that

$$\eta_2(\varepsilon_F) = \frac{N\pi}{10} \quad . \qquad /8/$$

This has been regarded as a consequence of the HF treatment in the non-magnetic case [22] but in fact it holds generally for a spherically symmetrical ground state. Considering the value of the scattering amplitude with this $\mathrm{Im}t(\varepsilon_F) = \frac{\sin^2\eta_2(\varepsilon_F)}{\pi\rho_o}$ , here called the "charge neutrality limit" /this can not exceed the "unitarity limit", although in the case of $N = 5$ the two limits are the same/.

(III) At low temperature a n.r. is formed around the Fermi level, which can be smeared out by a temperature of the order of $T_K$, in contrary to the b.r. The width of the n.r. can be related to $T_K$. The energy /temperature/ dependence far from the Fermi level /above $T_K$/ is determined by logarithmic terms. However, at the top of this level it probably be described by power terms, apparently in consequence of the infrared cathastrophe [23] suggested by Anderson [24]. The $\rho_d(\omega)$ function is schematically plotted in Fig. 2a and 2b for noble metal and Al hosts, respectively.

The main point of our model is that as the temperature is decreased, the amplitude of the n.r. superimposed on the b.r. increases to reach the charge neutrality limit at zero temperature; the b.r., in contrast, is only slightly deformed. The reasoning behind this is that, except in the non-magnetic Lorentzian case of the Anderson model, it is hard to believe a

lowering of temperature can lead to drastic deformation of the b.r. instead of formation of a n.r., because this would imply a reordering of the localized level corresponding to an energy change determined by the basic parameters U and Δ of the Anderson model. The formation of a n.r. requires much less energy.

(iii) In the light of the above assumptions we may go on to discuss briefly the experiments mentioned in part (i).

The available optical [5] and photoemission [6] measurements provide direct information on the b.r., but not on the n.r.

The impurity resistivity at zero temperature is fitted by Eq./1/, which follows directly from assumption (II). Small deviations may be due to the neglection of non-resonant phase shifts. The temperature dependence can be qualitatively interpreted on the basis of the behaviour of the n.r. described under (III). It should be pointed out, however, that in noble metal based alloys most of the n.r. are extremely narrow compared to the b.r. and therefore the temperature dependence is mainly determined by the structure of the n.r. The Al-based alloys behave somewhat differently, because here the difference in the width of the n.r. and that of the b.r. is not so great, so that the formation of a n.r. must be followed by some deformation of the b.r. /see Fig.2b/; the temperature dependence is therefore much less independent of the structure of the b.r. At temperatures $T > T_K$ only the b.r. remains, thus at high temperatures one gets information only about the b.r., as was suggested by Friedel [25]. The difference between the high-temperature and low-temperature data for Cu- and Au-based alloys seem to be in fairly good agreement when calculated from the HF solution [3] and the charge neutrality on the assumption that the position of the Fermi level is determined by the occupation numbers. It may be mentioned that in certain cases /CuCo, for example/ the amplitude of the n.r. may be negative [26].

Comparison of the charge perturbation and resistivity data for Al-based alloys confirms that the temperature and energy dependence is similar for low temperatures and energies and hence provides direct evidence for the existence of the n.r. Moreover, the existence of the b.r. is evident from the preasymptotic behaviour discussed in (i). At zero temperature, according to the charge perturbation theory there are two limiting cases: if the n.r. is much broader than the characteristic energy $\Delta_c$, the asymptotic form given by Eq./6/ should be valid and $\alpha = 5\sin\frac{N\pi}{10}$, while in the opposite case only the b.r. plays an important role, due to the large coherence length connected with the small Γ. The experimental results given in Fig. 1b show that AlMn /and probably AlCr/ represents an intermediate case and can be explained

by the structure shown in Fig.2b. A detailed analysis will be given elsewhere.

It should be emphasized that the picture presented here of resonance formation in dilute alloys is a semiempirical one based on, and containing, all the well-confirmed conclusions of different theories. In this picture the concept of the neutrality limit - which is missed by many theories through the neglection of long-range Coulomb forces - is crucial. In contrast to previous interpretations, the present analysis of the experimental data suggests that in Al-based alloys the d-level structure is slightly split.

The main features of this picture are based on an analysis of the behaviour of the charge perturbation around the impurities in Al, which allows the energy and temperature dependence of the scattering amplitude to be evaluated separately. Similar experiments on Cu-based alloys would complete the experimental confirmation of the suggested scheme.

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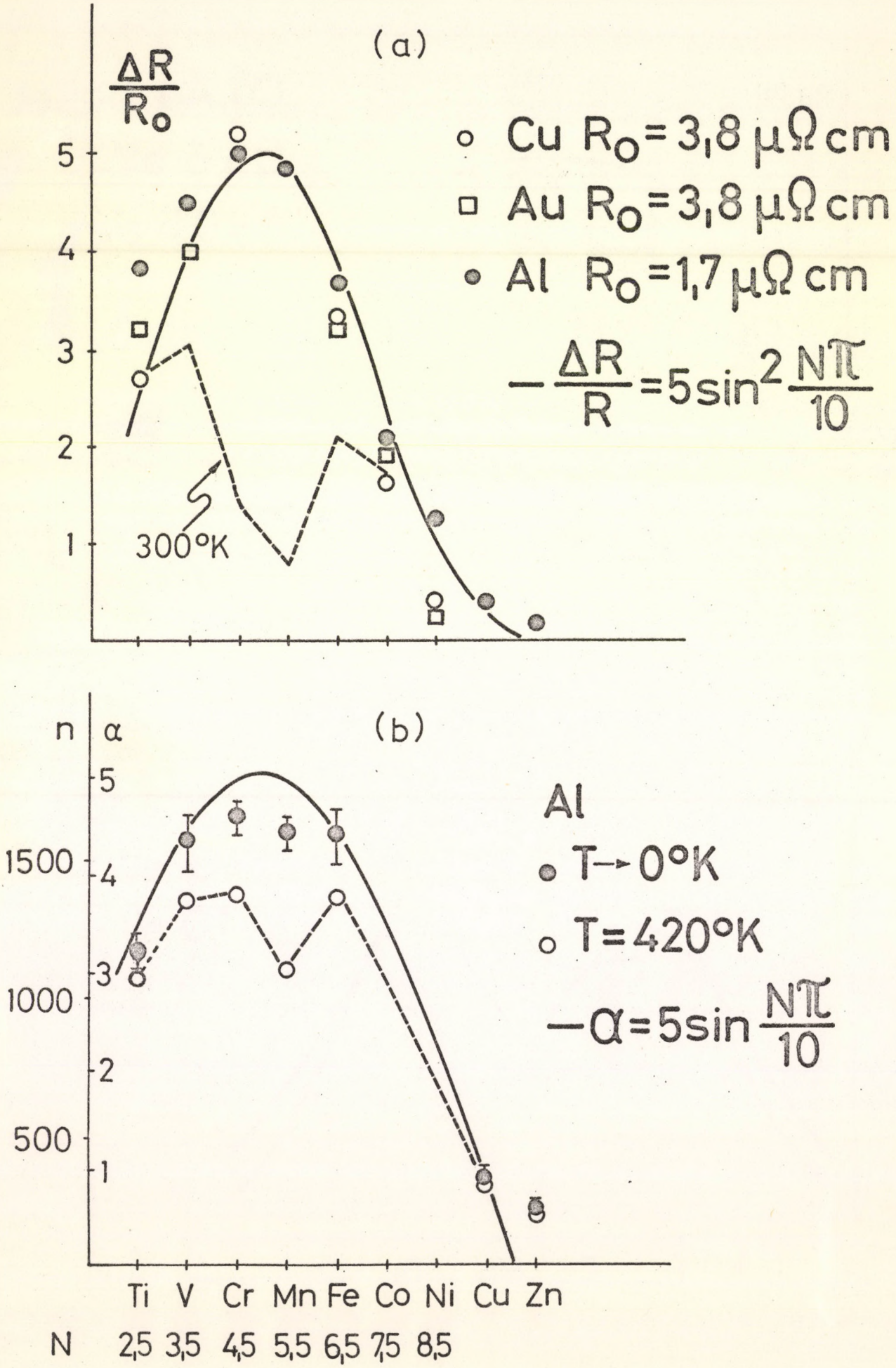
FIGURE CATIONS

Fig.1a The impurity resistivities $\Delta R$ extrapolated to $T = 0^{o}K$ for 3d-transition metal impurities in Cu,Au and Al. The $R_o$ values are taken from Ref's 1 and 10. The dotted line refers to the room-temperature values measured in Cu- and Au-bashed alloys.

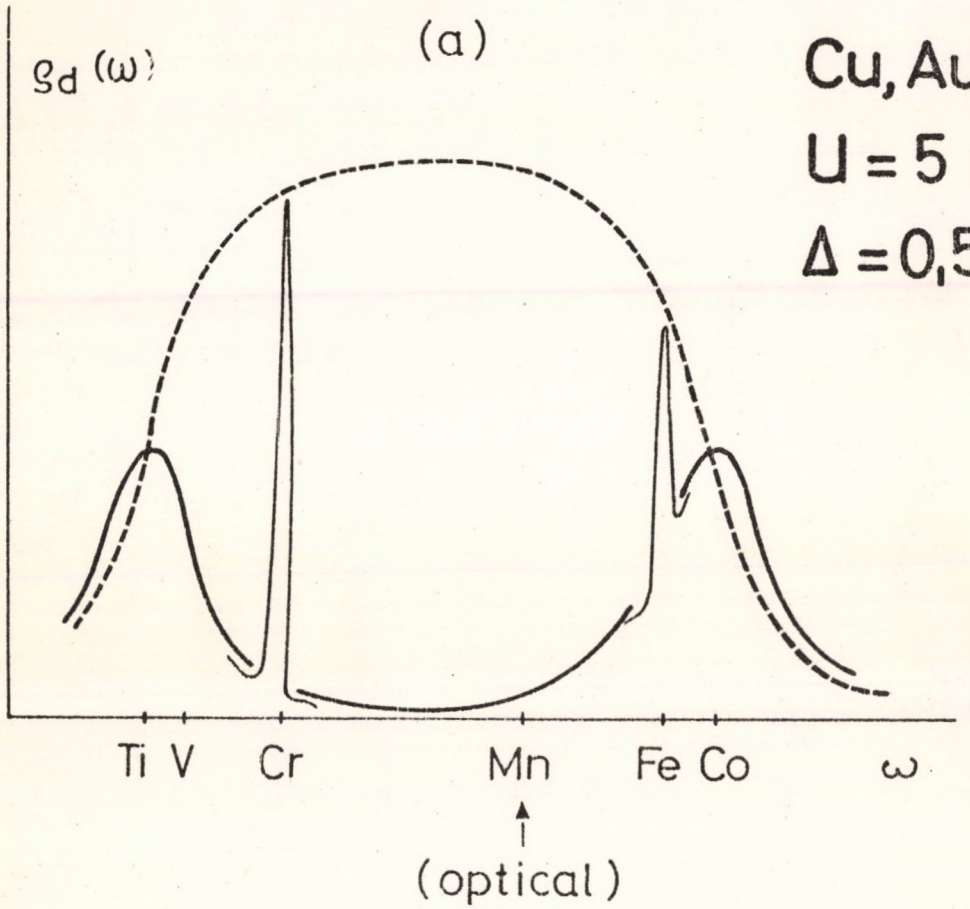
1b First order wipe-out numbers and charge oscillation amplitudes for 3d-transition metal impurities in Al.

Fig.2a The density of states for 3d-transition metal impurities in noble metal hosts. The positions of the resonances with respect to the Fermi level are derived from the occupation numbers N, and in the case of Mn impurities from the optical experiments. The n.r. is shown in the case of Cr and Fe.

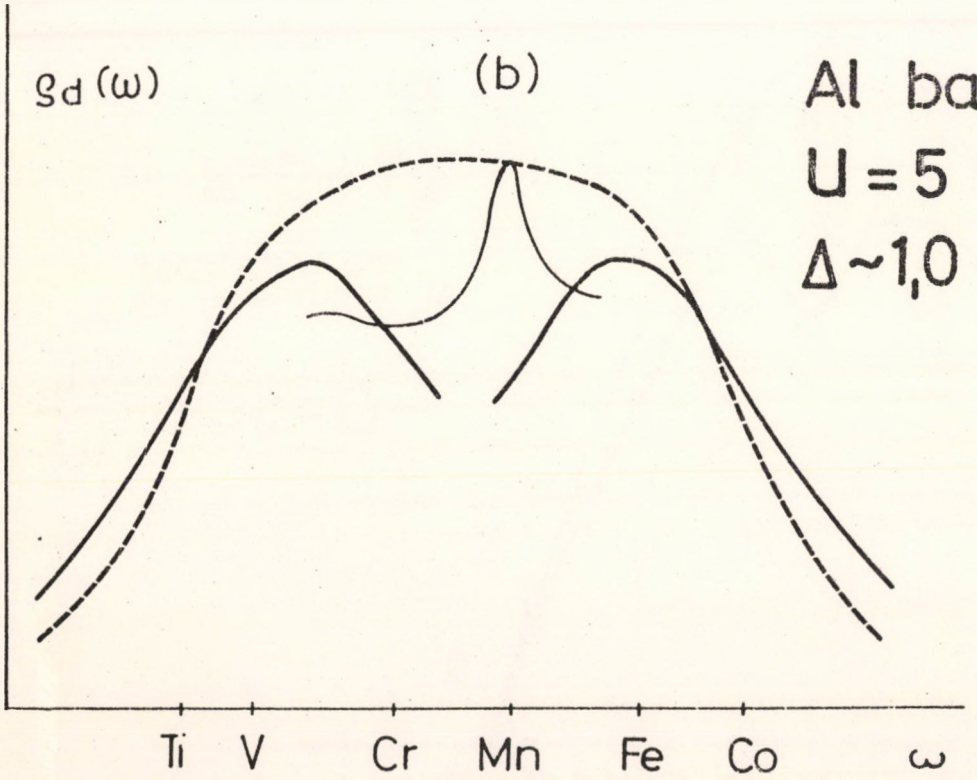
2b Schematic plot of the density of states for 3d-transition metal impurities in Al. The n.r. is shown in the case of Mn.


(a)
ΔR/R₀
5
4
3
2
1
300°K
○ Cu R₀ = 3,8 μΩ cm
□ Au R₀ = 3,8 μΩ cm
● Al R₀ = 1,7 μΩ cm
−ΔR/R = 5 sin² NΠ/10
(b)
n α
1500
1000
500
5
4
3
2
1
Al
● T → 0°K
○ T = 420°K
−α = 5 sin NΠ/10
Ti V Cr Mn Fe Co Ni Cu Zn
N 2,5 3,5 4,5 5,5 6,5 7,5 8,5

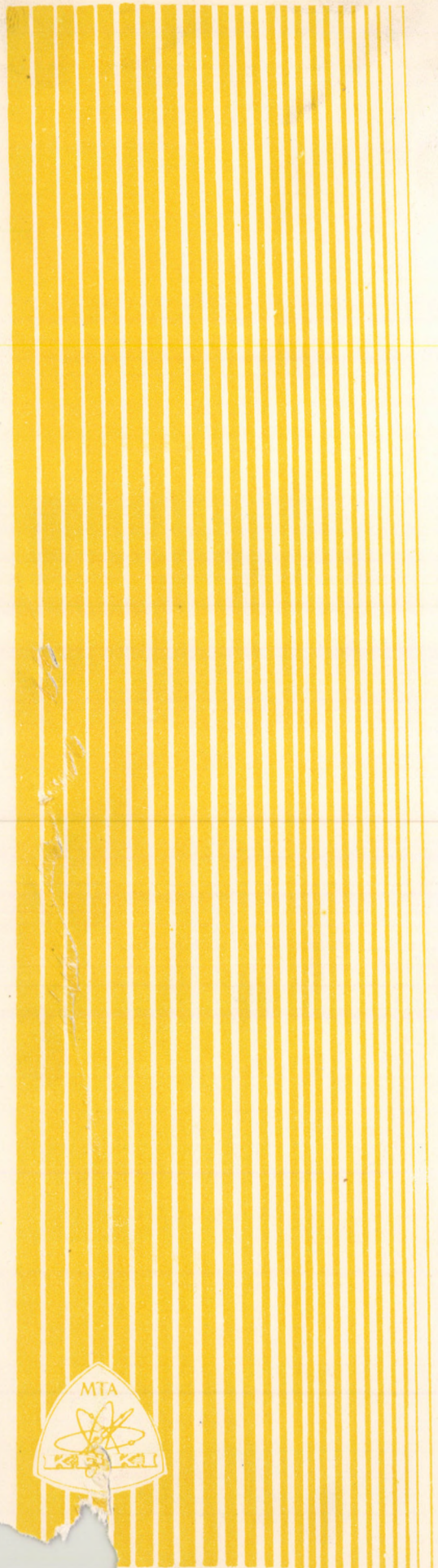
61.963




Cu, Au based
U = 5 eV
Δ = 0,5 eV




Al based
U = 5 eV
Δ ~ 1,0 eV



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