QUANTUM INTERFERENCE ON DEFECTS – THE INFLUENCE OF HYDROGEN ON THE MAGNETORESISTANCE OF THE $(Zr_2Ni)_{1-x}H_x$ SYSTEM

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1 INTRODUCTION

Electronic properties of metals have traditionaly been observed using models of a more or less idealized crystal lattice in which the electron wave function behaves like a Bloch wave. The wave is scattered by the defects and impurities. This idealised model is based on the concept of "weak disorder" and it yields Boltzmann transport equation for quasiparticles. At low temperatures the model predicts the resistivity of such a system in the form

$$\rho(T) = \rho_0 + \alpha T^n \qquad (1)$$

where ρ_0 is the residual resistivity due to electron scattering on impurities while α is positive and n is of the order of 2 or more.

Unfortunately even ordinary alloys do not always behave according to this model. In fact, real materials are often in a state of disorder that is not, even in principle, "weak". The coefficient α can sometimes assume negative values and the coefficient n can be significantly different from 2.

An increase in disorder implies smaller values of the electron mean-free-path ℓ. When this mean-free-path becomes of the order of the Fermi wave length λ_F the value of α will tend to zero. Classically viewed, however the coefficient α should not become negative.

In the past decade or so, new models of disorder in systems have been put forward most of them based on the concept of localisation first described by Anderson (1958). This concept stipulates that in conditions where $\lambda_F \sim \ell$ electron eigen states are no longer extended but are exponentially localized (Fig. 1):

$$|\Psi(r)| \sim \exp|r - r_0|/\xi \qquad (2)$$

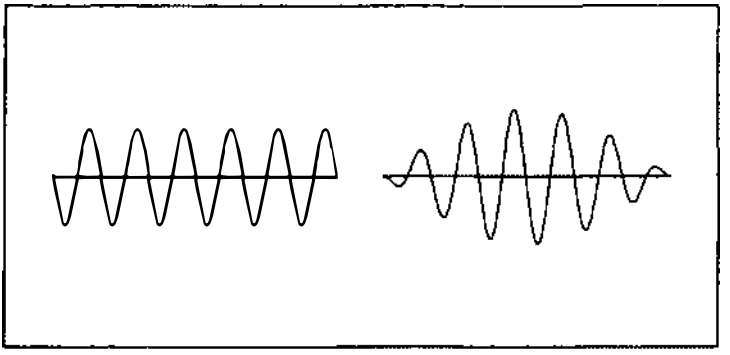

FIG. 1.

The parameter ξ is termed the localisation length. In Anderson's Model the localisation is the result of "randomisation" of potential which occurs as a result of disorder in the atomic matrix. As can be easily visualised (Fig. 2) such random potential will result in localisation as evidenced by the smearing of the electron band edges. On both sides of the band there appear the mobility edges ϵ_c' and ϵ_c. Electrons beyond the mobility edges are localised because there are no final states available for them. The extended states are situated between ϵ_c and ϵ_c' and if ϵ_F falls within this region the system is still "metallic". For cases where $\epsilon_F < \epsilon_c$ the only conduction mechanism is that of "hopping" for all temperatures $T > 0$. At $T = 0$ there is no conduction.

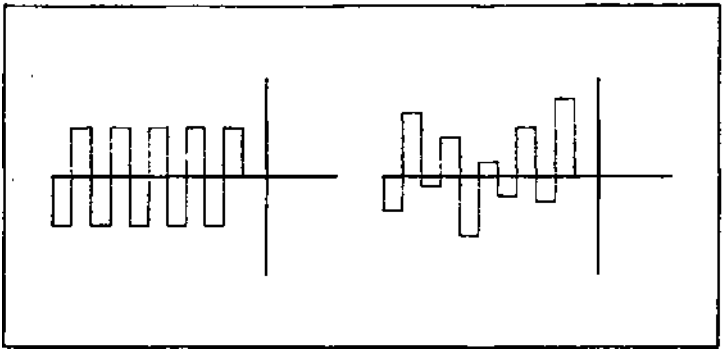

FIG. 2.

2. A PERTURBATIONAL MODEL OF LOCALISATION

The lowest-order Perturbation-theory does not predict any localisation in non-interacting electron gas undergoing weak scattering.

On the basis of higher-order Perturbation theory Langer and Neal (1966) have evaluated the crossed diagram (Fig. 3) and found that it contributes to conductivity with a term proportional to $n_i^3 \ln n_i$, where n_i is the defect concentration. Using such diagrams Anderson et al have evaluated the amplitude for such a process as

$$W(p, p') = \frac{d(n_i V^2)^2}{(p+p')^2 v_F^2 \tau^2} \qquad (3)$$

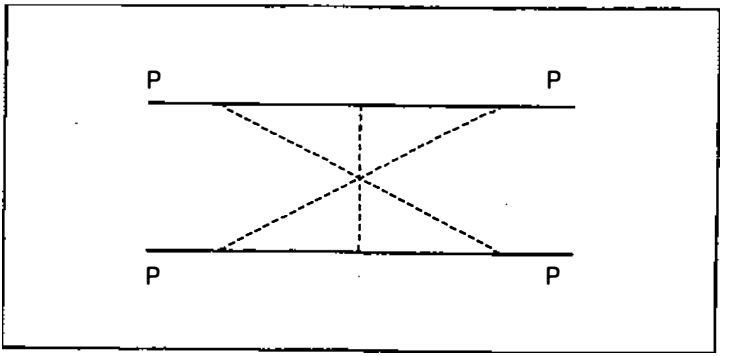

FIG. 3.

where V is the scattering potential and τ is the electron relaxation time. The amplitude diverges for p = p' and the physical meaning of this result can best be visualized as back scattering of self-crossing trajectories (Fig. 4). The electron can undergo two equivalent sequences of quasi elastic scattering and return to the point of origin in the same eigenstate of energy. The two amplitudes corresponding to two equivalent sequences, are the same and they interfere constructively giving a kind of "echo" in the back scattering of electrons.

On the basis of scaling theory we have the following expressions for the conductivity of 3, 2 and 1 dimensional systems.

$$\sigma_{3D}(L) = \sigma_0 - \frac{e^2}{h\pi^3}\left(\frac{1}{\ell} - \frac{1}{L}\right) \tag{4}$$

$$\sigma_{2D}(L) = \sigma_0 - \frac{e^2}{h\pi^2}\ln\left(\frac{L}{\ell}\right) \tag{5}$$

$$\sigma_{1D}(L) = \sigma_0 - \frac{e^2}{h\pi}(L - \ell) \tag{6}$$

Compared with Ohm's Law ($\sigma(L) = \sigma_0$) we observe that the systems in question behave very differently. For 1D and 2D systems the perturbation theory does not anticipate any conductivity at large values of L. For such cases the localisation legths are: $\xi^{1D} = \pi\ell$ and $\xi^{2D} = \ell \exp(\pi k_F \ell/2)$. In the three dimensional case the theory gives finite conductivity at all scales.

All these results pertain to the case of T = 0 and for the values of L that are finite but small. In reality experiments are carried out at, say, liquid helium temperatures and with samples that are relatively large. Thus, to be able to compare our measurements with the theoreticel predictions we have to take into account random fluctuations of electron states. These fluctuations (Thouless 1977) are caused by inelastic scattering which destroys the phase coherence of the electron. If we denote by τ_{in} the time that an electron spends in an eigenstate of energy then we can introduce a "phase coherence length"

$$L_{Th} = (D\tau_{in})^{1/2} \tag{7}$$

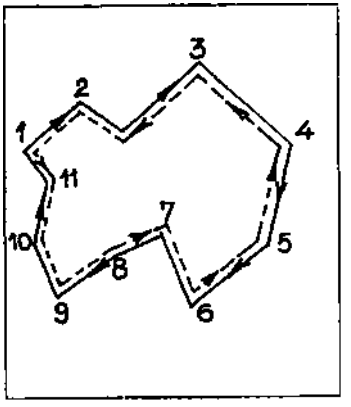

FIG. 4

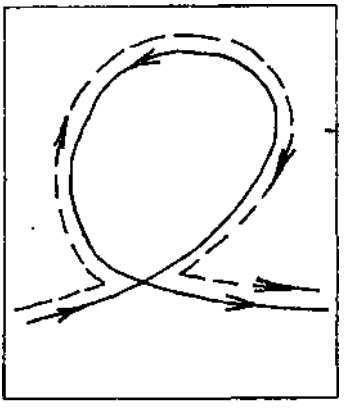

FIG. 5

where the diffusion constant $D = v_F^2\tau/d$. If we substitute L_{Th} in place of the minimal L into the theory pertaining to T = 0 then the results are valid for cases where $T > 0$. Since we are talking about quantum interference it is obvious that a large number of quasi-elastic scatterings that an electron is likely to undergo during the time τ_{in} will not change its phase by an amount large compared to π. It is therefore better to talk about the "Phase-coherence time" τ_ϕ rather than τ_{in} which implies a "discontinuous" event (Altshuler, et al 1981). Thus τ_ϕ would be the time an electron takes to undergo a phase change of the order of 2π. At low temperatures $\tau_\phi >> \tau$ where τ is the time the electron spends in an eigenstate of momentum.

Since τ_ϕ depends on the temperature as $\tau_\phi \propto T^{-p}$, where p is a parameter that is dependent upon the scattering mechanisms, we may say that $L_{Th} = aT^{-p/2}$. Taking this as our scale limits we may writte (4), (5) and (6) as

$$\sigma_{3D} = \sigma_0 + \frac{e^2}{h\pi^3}\ \frac{1}{a}\ T^{p/2} \qquad (8)$$

$$\sigma_{2D} = \sigma_0 + \frac{p}{2}\ \frac{e^2}{h\pi^2}\ \ln\left(\frac{T}{T_0}\right) \qquad (9)$$

$$\sigma_{1D} = \sigma_0 - \frac{ae^2}{h\pi}\ T^{-p/2} \qquad (10)$$

As the temperature falls the localisation effects are more pronounced and the conductivity falls. In the context of "weak localisation" therefore this is the origin of the negative α coefficient. Any "strong localisation" effects such as "hopping" will also contribute to a negative α coefficient.

3. MAGNETORESISTIVITY AND SPIN-ORBIT INTERACTION

It can be shown (Lee, 1980) that electron scattering on magnetic impurities ruins the phase coherence. Therefore at scales larger than $L_s = (D\,\tau_s)^{1/2}$ (where τ_s is the spin-flip relaxation time) the conductivity will be essentially scale independent. This comes from the fact that the spin-flip Hamiltonian does not conserve the time-reversal symmetry. This can be readily visualised by observing that an electron may undergo a series of spin-flip interactions (see for ex. Fig. 4) in certain order on the time scale. Each interaction may tip the spin by a given angle. The complementary wave going the other way will undergo the same processes but in reverse order. Three dimensional rotations belong to a non-Abelian group so that final spin states are different for the partial waves and the interference is destroyed. The action of an imposed magnetic field also acts in the way shifting the phase (Altshuler et al, 1980) which is rather evident if we recall the effect of the vector potential on closed electron orbits.

It is therefore to be expected that simple systems containing localised electrons will exhibit a negative magnetoresistance which will express itself at relatively low fields, especially at low temperatures, due to a relatively large L_{Th}.

Systems in which there exists a strong spin-orbit interaction will show an *increase* in conductivity for all scales larger than ℓ and $L_{s0} \approx (D\tau_{s0})^{1/2}$, where τ_{s0} is the relaxation time for the spin-orbit scattering. Again for self-crossing trajectories (Fig. 5) and for backscattered electrons the action of the superimposed magnetic field will be additive. The spin-orbit interaction Hamiltonian conserves the time-reversal symmetry so that partial-wave spins tilt in opposite directions. One of the rules of Quantum mechanics shows that the spin has to turn by 4 in order for the spin function to reassume its original form. A rotation by 2 will throw the partial waves exactly out of phase. It turns out that in the presence of strong spin-orbit interaction this is exactly what happens, that is, most of the partial waves in the Langer-Neal diagram are shifted out of phase. Thus conductivity is enhanced, the spin-orbit interaction having almost cancelled the constructive interference (Bergmann, 1982). It is also to be expected that a superimposed magnetic field will tend to re-establish the original localisation by tilting the partialwave spins further until they are once again in phase. This is indeed the case both in 2D and 3D systems.

We may point out at this juncture that the dimensionality of a system will depend on how L_{Th} compares with the smallest dimension of a given sample. Since L_{Th} depends on the temperature it is possible to go from a 3D "regime" to a 2D "regime" by changing the temperature. Also if the wave length of thermal phonons exceeds the electron mean-free-path ($\lambda_{Th} > \ell$) (or the diameter of the sample wire) there will be electron heating in the electric field showing an ln V dependence of the conductivity (Altshuler et al, 1981).

4. ELECTRON-ELECTRON INTERACTION

What has been said so far pertains to a gas of noninteracting electrons in systems of relatively generous mean-free-paths. Altshuler and Aronov (1979) used a model of disordered Fermi liquid to explore systematically electron-electron interaction. They concluded that under some conditions these contributions may be significant.

There are two reasons why a detailed discussion of this problem exceeds the scope of this short review. In the first place the calculations and the arguments involved are relatively long and complicated to be briefly described in an acceptably transparent

way. A more important reason is the fact that the e-e interaction does not qualitatively change the picture outlined above. Indeed at all, except very low, temperatures the electron-pohonon interaction relaxation times are much shorter than analogous e-e relaxation times. There is a possible exception to be expected in systems with relatively high superconducting transition temperatures namely the effect of electron scattering on Cooper Pairs above T_c. This, so called Maki-Thompson diagram, may contribute significantly to the magnetoresistance in the same sense as the spin-orbit effect i.e. to disrupt the coherence of electron partial waves. This will occur in fields that satisfy the condition

$$\frac{2eH}{hc} > \frac{1}{D\tau_{in}} \tag{11}$$

Generally this contribution will be small in systems in which $|\tilde{\lambda}| \ll 1$; where $|\tilde{\lambda}|$ is the renormalized electron-pohonon coupling constant. As to the e-e relaxation time τ_{ic}, Schmid gives it as

$$\frac{1}{\tau_{ic}} = \frac{\pi}{8}\frac{k_B^2 T^2}{\epsilon_F} + \frac{3}{2}\left(\frac{1}{k\ell}\right)^{3/2}\frac{(k_B T)^{3/2}}{\epsilon_F^{1/2}} \tag{12}$$

we can observe that these are relatively long relaxation times at most experimentally relevant temperatures.

5. COULOMB SCREENING

The Coulomb anomaly in disordered systems comes from the diffusive character of the electron motion. Thus the electron-electron interaction is retarded since a sudden change of charge distribution cannot at once be screened by slowly diffusing electrons. For small momentum transfers this may imply significant corrections in the e-e interaction. In the presence of spin-orbit interaction these contributions appear to be small (Lee and Ramakrishnan, 1982). The correction to the quantum interference is based on a relaxation time given as

$$\tau_T = \frac{h}{2\pi k_B T} \tag{13}$$

6. ELECTRON-PHONON INTERACTION

Bergmann (1971) has pointed out that the Anderson theorem, which states that the superconducting transition temperature is independent of impurity content of a system, must be modified when the system in question is such that the impurities significantly modify the phonon spectrum. This is mostly the case in highly disordered systems where the electron-phonon interaction tends to rise. In the context of the problem

discussed here, the electron-phonon interaction introduces an additional T^2 – dependent term in the expression for τ_i (Keck and Schmid, 1976)

$$\frac{1}{\tau_i} = \frac{n}{LMm} \left(\frac{1}{c^3}\right) \frac{1}{\ell} (k_B T)^2 \int_0^\infty \frac{x\,dx}{\sin h(x)} \qquad (14)$$

where n is the electron density, L is the number of atoms in a unit volume and c is the mean sound velocity in the system. Again the interaction tends to disrupt the electron coherence and so it contributes to the de-location of electrons.

7. COMPARISON WITH EXPERIMENT

As a typical case of a system with a negative α coefficient we may examine Zr_2Ni metallic glass doped with varying amounts of hydrogen that we have been investigating (Babić et al., 1982). In Fig. 6 we see a series of curves showing the temperature profile of resistance of samples doped with hydrogen in several concentrations. We observe that the values of α become more negative with increasing hydrogen concentration. The reason for this increase is to be found in greater disorder introduced by the dopant, that is the random potential fluctuations increase, but we may also note that the influence of spin-orbit delocalisation will decrease since additional scattering centers will be provided by the dopant atoms.

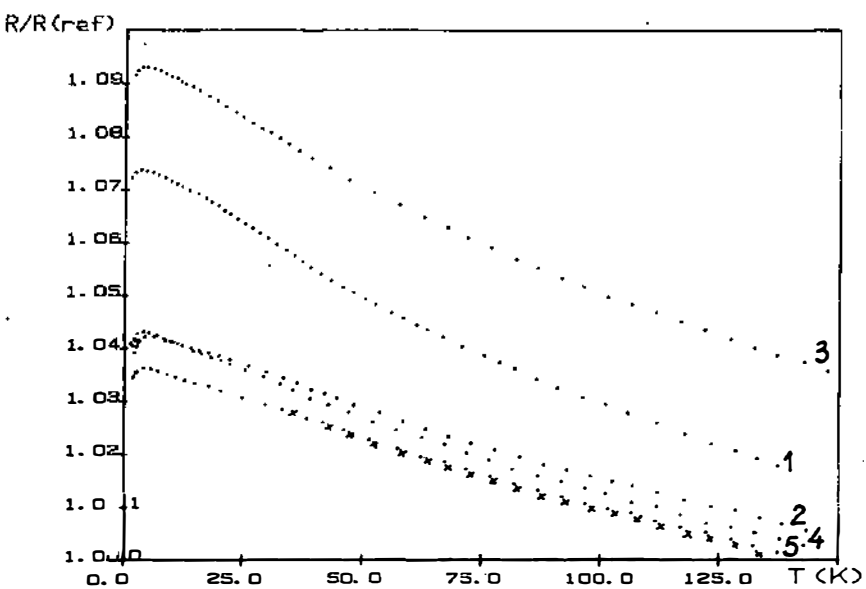

FIG. 6

A very good example of direct measurement of spin-orbit interaction in quasi-two-dimensional systems is due to Bergmann (Fig. 7). A pure Mg-film is shown to exhibit negative magnetoresistance, but when the same film is covered with a thin layer of gold the magnetoresistance progressively turns positive with increased thickness of the gold film.

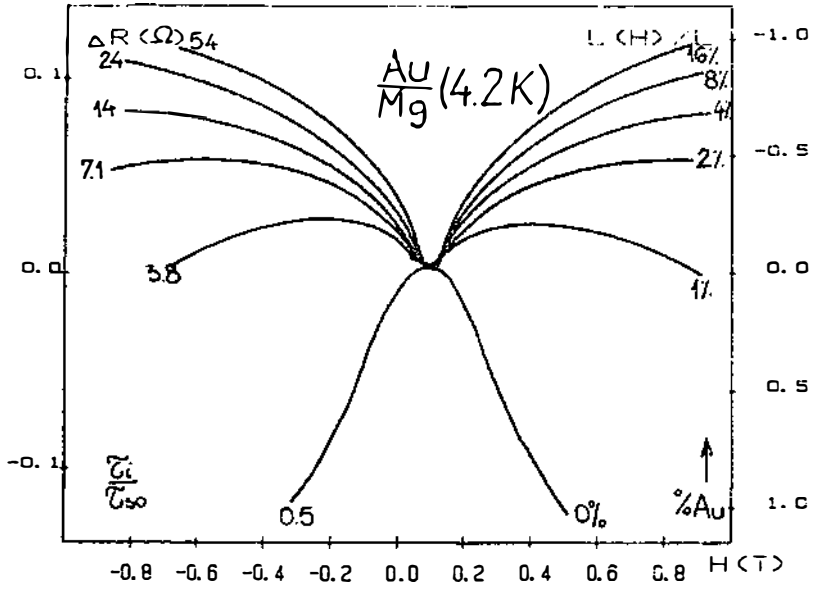

FIG. 7

In three-dimensional systems the field-induced "compensation" of the spin-orbit interaction is manifested at high fields. We can see (Fig. 8) the results of such measurements on a $Zr_{43}Cu_{57}$ system due to Bieri et al. (1984). The gradient of the magnetoresistance curves is strongly temperature dependent. The full curves are calculated using an expression due to Altshuler et al.:

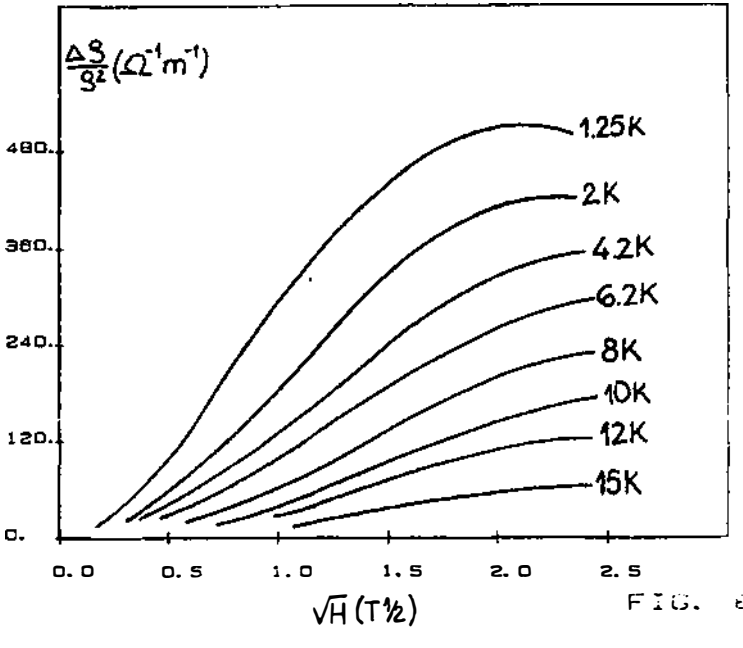

FIG. 8

$$\frac{\Delta\rho}{\rho^2} = \frac{e^2}{2\pi^2\hbar}\left(\frac{eH}{\hbar}\right)^{1/2}\left[\left(\frac{1}{2}+\beta\right)f_3\left(\frac{H}{H_i}\right) - \frac{3}{2}f_3\left(\frac{H}{H_{s0}}\right)\right] \quad (15)$$

where $H_i = \frac{h}{4eD\tau_i}$; $H_{s0} = \frac{h}{4eD}(\tau_i^{-1} + 2\tau_{s0}^{-1})$

and β is the coefficient of the Maki-Thompson contribution (i.e. the quenching of superconducting fluctuations above T_c). We may also note that at higher fields and/or lower temperatures expression (15) is inadequate probably due to a dependence on the magnetic field.

We have carried out magnetoresistance measurements on the $(Zr_2Ni)_{1-x}H_x$ system at various temperatures using samples with different dopant concentrations. The results are shown in Fig. 9. We can at once notice how strongly the dopant supresses the spin-orbit contribution to the magnetoresistance as well as the Maki-Thompson interaction. This can readily be explained if we note than the hydrogen atoms are located inside

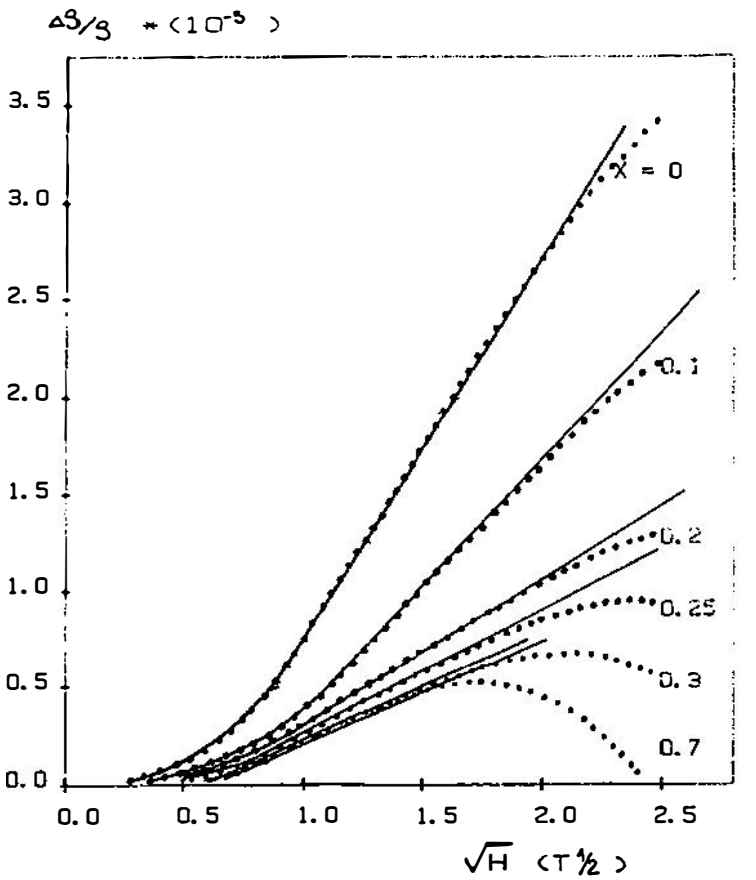

FIG. 9

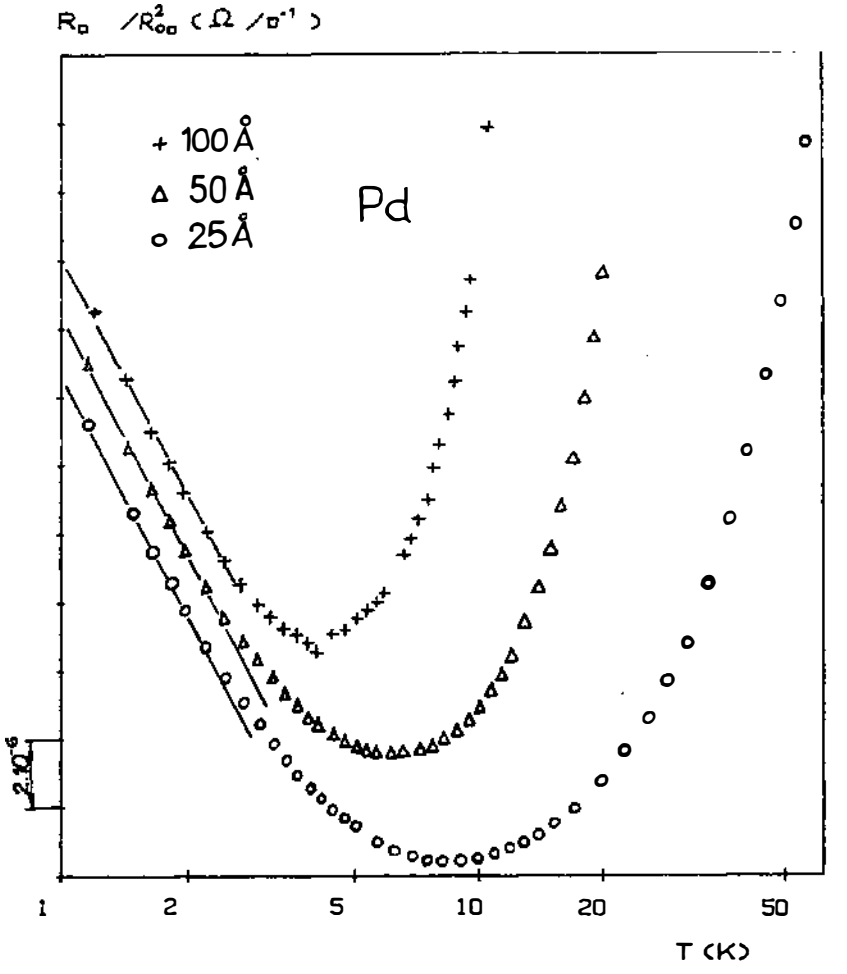

FIG. 10.

the elementary tetrahedra of the matrix which are rich with Zr-atoms. The dopant provides additional scattering centers for electrons and thus the relative contribution of the spin-orbit interaction is diminished. On the other hand increased concentrations of the dopant supress the T_c and with it the Maki-Thompson contribution to the magnetoresistance. Apart from depleting the density of electron states at the Fermi level the hydrogen increases the electron-phonon coupling and thus the pair-breaking process in the system. In addition to the aforementioned effects there is probably a contribution of electron scattering on paramagnons at the lowest temperature used (1.7 K). The origin of these paramagnons is at present obscure.

The tendency of the resistivity curves (Fig. 6) to dip at the lowest temperatures may be due to the same phenomenon but this remains yet to be confirmed.

It is also interesting to observe the results of Dumoulin et al. (1984) who measured the resistivity of Pd and Pd-H films. They find a ln T dependence in agreement with the theory but also a significant contribution of the hydrogen to the inelastic relaxation time τ_i which is in agreement with our observations (Figs. 10 and 11). In films with high T_c the localisation is almost cancelled by superconducting fluctuations (Fig. 12).

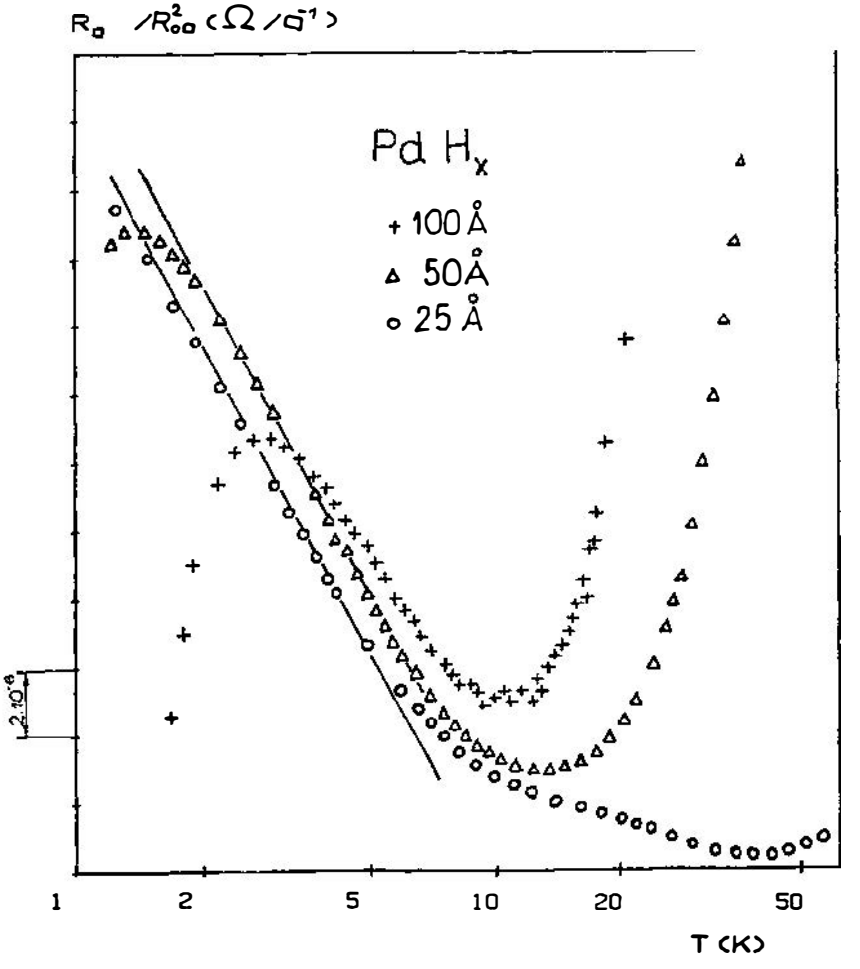

FIG. 11.

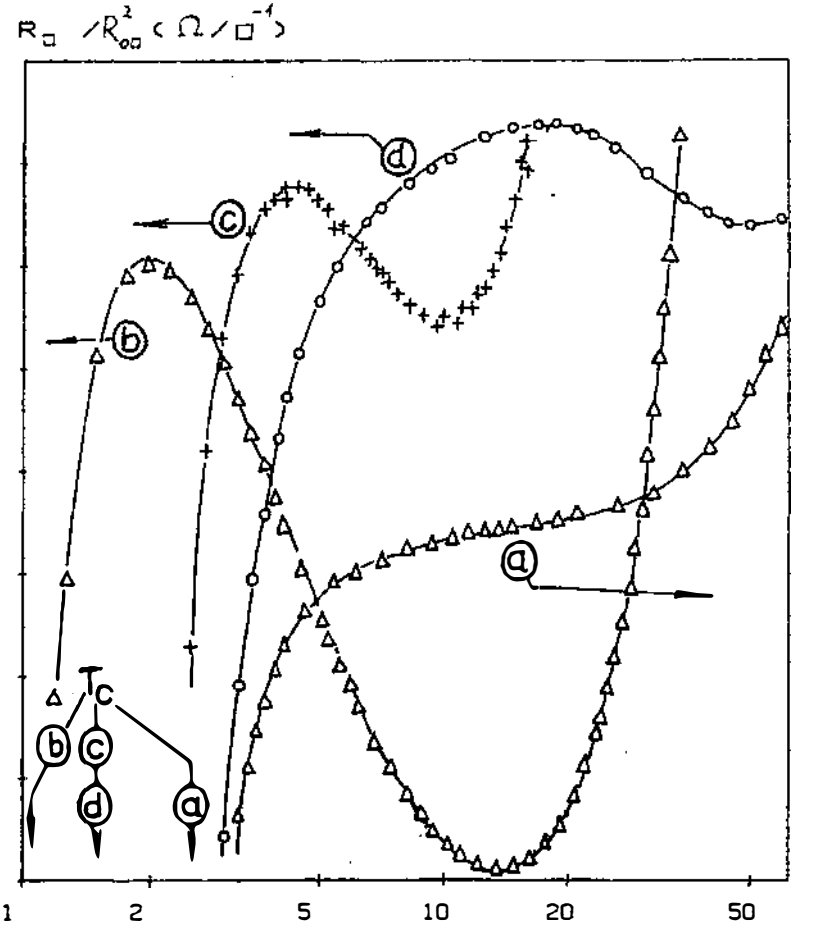

FIG. 12.

8. CONCLUSIONS

We have seen that quantum interference effects in disordered systems are a ubiquitous phenomenon which is well described by the standard perturbation theoretical approach. In systems that exhibit a strong spin-orbital interaction a light atomic dopant has a profound influence. This influence can be used to explore the topology of the host matrix and it can be used to elucidate the mechanisms of short range chemical ordering in metallic glases. Since hydrogen also modifies the host matrix its influence on the d-band of the host atoms can be used to study the two-band conduction. Phenomena such as paramagnon formation at low temperatures and a possible two-level hydrogen state ar also intriguing and should be further studied.

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