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Identification of local spin fluctuations by weak localization

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While some $3d$ impurities in a metal host possess a magnetic moment, the majority of d impurities are nonmagnetic at low temperatures. This can be due to the Kondo effect or local spin fluctuations (LSF's). We present two experimental examples for LSF's and show that weak localization is an excellent tool to investigate LSF's. The LSF's are enhanced by the Stoner factor due to the exchange interaction. This enhancement of the LSF's causes a dephasing of the conduction electrons. The dephasing is proportional to the temperature and can be measured by weak localization.

The magnetic properties of d impurities in a metal host have been intensively studied over the last three decades. While some $3d$ impurities possess a magnetic moment, the majority of d impurities are nonmagnetic at low temperatures. This can be due to the Kondo effect or local spin fluctuations (LSF's). However, experimentally it is difficult to distinguish between these two phenomena. In this paper we suggest that an investigation with weak localization can identify LSF's. We present two experimental examples and calculate the dephasing influence of LSF's on weak localization.

The theoretical properties of $3d$ impurities are generally discussed within the simplified Friedel-Anderson model.[1,2] In the mean-field approximation the Friedel-Anderson model is equivalent to two d resonances with a width $\Lambda=\pi V^2 g_0$, where V is the s-d hopping matrix element and g_0 is the free-electron density of states at the Fermi energy. We denote the density of states of the d resonance at the Fermi energy with g_d. Then the mean-field solution yields a magnetic d impurity if $g_d U>1$ and a nonmagnetic impurity for $g_d U<1$ (U is the exchange interaction). In the nonmagnetic state the impurity shows an enhanced susceptibility $\chi_0/(1-Ug_d)$, which results in local spin fluctuations at finite temperatures. Here χ_0 is the bare Pauli spin susceptibility $\chi_0=2g_d\mu_B^2$.

The experimental investigation of LSF's is rather difficult with conventional methods. In this paper we want to show that weak localization is a very sensitive tool for studying the LSF's of transition-metal impurities. Weak localization is a quantum correction to the conduction which is due to interference effects between the impurity scattered electron waves. In the past we and others have demonstrated that the method of weak localization is an excellent tool for studying magnetic impurities as predicted by Hikami *et al.*[3] Recently we have investigated the dephasing of weak localization in Au films which were covered with a fraction of a monolayer of vanadium impurities.[4] The V-surface impurities caused a large magnetic dephasing of the coherent electronic wave functions (which is observed in the broadening of the magnetoresistance curve by the $3d$ impurities). Similar results have been obtained for Cr, Mn, Fe, and Co surface impurities on Au.[5] However, for Ti surface impurities, we observed a dephasing rate which was by a factor of 200 smaller than for the magnetic $3d$ impurities and which showed a temperature dependence of T^α, where α was between 1.2 and 1.4. The experimental results for the temperature dependence of the dephasing cross section of the Ti surface impurities are plotted in Fig. 1. (The cross section is given in dimensionless units of $\sigma_F k_F^2/4\pi$.) The Ti coverage on top of the Au is 0.05 atomic layers in this experiment. We obtained similar results with $\alpha=1.0$ for a Ni coverage of 0.05 atomic layers. In this case the dephasing is caused by the Ni pairs because the dephasing cross section increases linearly with the Ni coverage. The experimental procedures and evaluation of these experiments is quite analogous to the Au/V experiment. In this paper we want to show that LSF's are the likely origin of the described dephasing.

Let us first consider a d impurity in the absence of an exchange interaction. Then the impurity's contribution to the susceptibility is $\chi_0=2g_d$ (in units of μ_B^2). If we introduce artificially a magnetic moment μ at the impurity then its contribution to the free energy is $\mu^2/2\chi_0$. At finite temperature we obtain a finite expectation value for μ^2 which is $\langle\mu^2\rangle=2g_d k_B T=\chi_0 k_B T$. This fluctuation of the local mag-

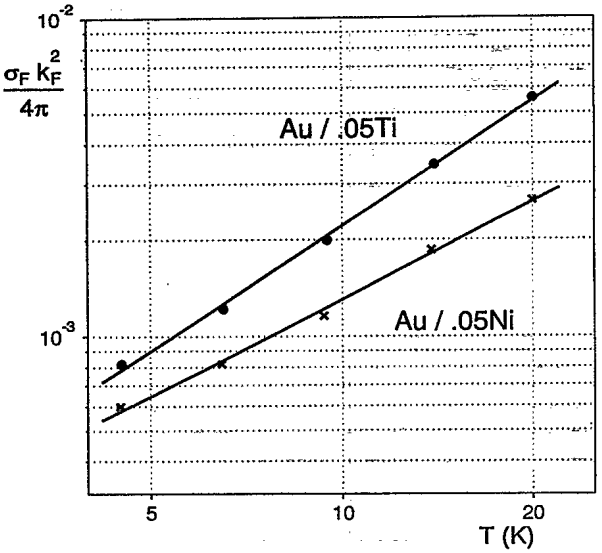

FIG. 1. The temperature dependence of the dephasing cross section (in units of $4\pi/k_F^2$) of Ti and Ni surface impurities on the surface of Au (coverages are 1/20 atomic layers).

netic moment is just due to statistical fluctuations in the occupation of the spin up and the spin down states. It does not alter the scattering properties of the impurity. A conduction electron of energy ϵ is scattered by the impurity according to the phase shift $\delta_2(\epsilon)$ at the energy ϵ, independent of the fluctuation moment.

However, this changes when we include the effect of the exchange interaction. For a resonance (or band) with exchange interaction the Pauli susceptibility is enhanced by the enhancement factor $(1-Ug_d)^{-1}$. Therefore the susceptibility becomes $\chi_0/(1-Ug_d)$. As a consequence one obtains also enhanced LSF's with $\langle\mu^2\rangle=\chi k_BT=2g_dk_BT/(1-Ug_d)$. The physics of this enhancement is easily understood. First consider a bare fluctuation of the magnetic moment at the impurity, which is created statistically and ignores the Coulomb repulsion. This moment alters the balance of spin-up and spin-down d states, lowering, for example, the spin-down resonance and lifting the spin-up resonance. The important consequence is that the scattering amplitude of a conduction electron with an energy ϵ (close to ϵ_F) is altered because the Friedel phase shifts for spin-up and spin-down electrons, δ_{2+} and δ_{2-}, are changed. The difference $\delta_{2+}-\delta_{2-}$ also fluctuates in time, and the thermal expectation value of $(\delta_{2+}-\delta_{2-})^2$ is given by the enhanced portion of the fluctuation of the magnetic moment $\langle\mu^2\rangle$. The relation between phase shift fluctuations and magnetic fluctuations is given by the following relation:

$$\left(\frac{5}{\pi}\right)^2\langle(\delta_{2+}-\delta_{2-})^2\rangle=\langle\mu^2\rangle Ug_d=2g_0k_BT\,\frac{Ug_d}{1-Ug_d}\,. \tag{1}$$

In the following we will show that this fluctuating phase shift has a slight dephasing effect on the quantum interference of the conduction electrons which is proportional to $(\delta_{2+}-\delta_{2-})^2$ and can be experimentally determined. It yields interesting information about the enhancement of LSF's.

Below we calculate the dephasing cross section of a LSF impurity in weak localization. We will report the result for three different cases: (i) the one-dimensional case with slowly varying LSF, (ii) the three-dimensional case with slowly varying LSF, (iii) the three-dimensional case with fast LSF's.

In the one-dimensional case we assume that the LSF's occur only in one direction, for example the z direction. While this may be realistic in special situations we expect that in the majority of cases the magnetic moment of the LSF's can point into any direction. Then the expectation value of $\langle\mu^2\rangle$ is by a factor of 3 larger. Furthermore the LSF's are considered as slow when the fluctuating moment μ remains essentially stationary during the time scale of weak localization. The longest time in weak localization is the inelastic-scattering time. On the other hand, the LSF's are considered as fast when μ changes its value and direction on a much shorter time scale. The essential criterion is whether a scattered-electron amplitude and its time reversed counterpart see the same (fluctuating) moment at the impurity.

Before discussing the calculations we wish to summarize the results: (a) local spin fluctuations of the Friedel-Anderson impurity cause an additional dephasing of weak localization; (b) the dephasing is proportional to the temperature; (c) the characteristic dephasing cross section σ_F in the dimensionless units of $4\pi/k_F^2$ (which will be used throughout this paper) is given by

$$\frac{\sigma_Fk_F^2}{4\pi}\simeq\frac{5}{2}\langle(\delta_{2+}-\delta_{2-})^2\rangle=z\,\frac{\pi^2}{5}\,\frac{g_dk_BUg_d}{1-Ug_d}\,, \tag{2}$$

which yields the corresponding dephasing rate $1/\tau_F=n_d\sigma_Fv_F$ of the LSF's. Here z is equal to 1 for one-dimensional LSF's and $z=3$ for three-dimensional LSF's. v_F is the Fermi velocity and n_d the concentration of d impurities; (d) for the magnetoconductance of weak localization in two dimensions, i.e., for a thin film, this well-known equation is obtained,

$$\frac{\Delta G}{G_{00}}=-\left[\Psi\left(\frac{1}{2}+\frac{H_1}{H}\right)-\Psi\left(\frac{1}{2}+\frac{H_2}{H}\right)+\frac{1}{2}\,\Psi\left(\frac{1}{2}+\frac{H_3}{H}\right)-\frac{1}{2}\,\Psi\left(\frac{1}{2}+\frac{H_4}{H}\right)\right], \tag{3}$$

where $G_{00}=e^2/2\pi^2\hbar$. The field H_ν represent scattering rates according to the relation

$$H_n=\frac{1}{\tau_n}\,\frac{1}{4eD}=\frac{1}{\tau_n}\,\frac{1}{4}\,\hbar eg_0\rho, \tag{4}$$

where D is the diffusion constant, and ρ the resistivity of the film.

In the case of fast three-dimensional LSF's one obtains for the characteristic fields;

$$H_1=H_0+H_{so}+H_d,\quad H_2=H_i+\frac{4}{3}\,H_{so}+\frac{1}{2}\,H_F,$$
$$H_3=H_i+\frac{1}{2}\,H_F,\quad H_4=H_i+\frac{4}{3}\,H_{so}+\frac{1}{2}\,H_F. \tag{5}$$

From the evaluation of the experimental magnetoresistance curves one may determine the dephasing cross section $\sigma_F/2$ due to LSF's.

In the following we sketch the derivation of the above results for the case of slow one-dimensional enhanced LSF's. The derivation of the one-dimensional case is slightly shorter but contains all the essential physics. The other two cases are quite analogous.

The scattering amplitude $f(\Omega)$ of an impurity is determined by the Friedel phase shifts δ_l for different angular momenta. For a d impurity $f(\Omega)$ is dominated by its d amplitude with the scattering phase δ_2. The other contributions are neglected. For a Stoner enhanced fluctuation there are different phase shifts $\delta_{2\pm}$ for spin up and spin down. We abbreviate $\delta_{2\pm}$ by $\delta_\pm$. Then the scattering amplitude is in this approximation:

$$f_\pm(\Omega)=\frac{1}{k}\,5\,\exp(i\delta_\pm)\sin\delta_\pm P_2(\cos\Theta).$$

The scattering cross section is

$$\frac{\sigma_\pm k_F^2}{4\pi}=5\sin^2\delta_\pm$$

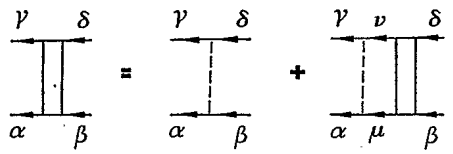

FIG. 2. The (graphical) Dyson equation for weak localization showing the spin indices.

and the corresponding scattering rates are

$$\frac{1}{\tau_{\pm}}=n_d v_F \sigma_{\pm},$$

where n_d is the concentration of impurities. The total averaged scattering rate for a conduction electron is $1/\tau=1/\tau_0+1/\tau_{so}+1/\tau_d$, where $1/\tau_d=(1/\tau_+ + 1/\tau_-)/2$.

To calculate the quantum correction of the conductance in weak localization one has to consider the particle-particle propagator. It is given by a Dyson equation which is graphically shown in Fig. 2 showing also the spin indices. The diagram in Fig. 2 is an inverted fan diagram. Therefore the scattering of the top and the bottom propagators occur at different times. It makes an important difference whether the fluctuation of the d impurity, i.e., the phase shifts δ_+ and δ_-, are the same at these different times. In the case of slow fluctuations we assume that they are the same and in the case of fast fluctuations we take the other extreme and assume that they are uncorrelated. The Dyson equation has the form (see for example the review articles Refs. 6–10).

$$\Gamma_{\alpha\beta\gamma\delta}=\Gamma^0_{\alpha\beta\gamma\delta}+\sum_{\mu,\nu}\Gamma^0_{\alpha\mu\gamma\nu}\Pi_{\mu\nu}\Gamma_{\mu\beta\nu\delta}.$$

Here $\Pi_{\mu\nu}$ is the product of two Green functions, a restarted one $G_\mu(k,\epsilon)$ with the spin index μ and an advanced one $G_\nu(k+q,\epsilon+\omega)$ with the spin index ν. The Green functions depend only through their Zeeman shifts $\mu_B H$ on the spin index, and for practical purposes this shift can be neglected. This yields

$$\Pi(q,\omega)\simeq\frac{2\pi g_d}{\hbar}[1+i\omega\tau-Dq^2\tau].$$

The dashed line is denoted as $\Gamma^0_{\alpha\beta\gamma\delta}$ and represents the scattering of both propagators by the same scattering centers. It is equal to the product of the scattering matrix element of the upper propagator times the conjugate complex matrix element of the lower propagator. The propagators may change their spin at the scattering event. The spins are denoted as α, β, γ, and δ. In the presence of potential and spin-orbit scattering one finds for $\Gamma^0_{\alpha\beta\gamma\delta}$,

$$\Gamma^0_{\alpha\beta\gamma\delta}=\frac{\hbar}{2\pi g_0}\left[\frac{1}{\tau_0}\delta_{\alpha\beta}\delta_{\gamma\delta}-\frac{1}{3\tau_{so}}\sum_i\sigma^i_{\alpha\beta}\sigma^i_{\gamma\delta}\right],$$

where $\sigma_{\alpha\beta}$ are the Pauli spin matrices.

The phase shift treatment of the LSF's does not yield spin flip scattering, so the d impurity contributes only to $\Gamma^0_{\alpha\alpha\beta\beta}$. This adds to Γ^0_{++++} and Γ^0_{----} the term $1/\tau_d$ (inside the bracket). For Γ^0_{++--} and Γ^0_{--++} we find an additional term $1/\tau_{+-}=1/\tau_d-1/\tau_F$ which we define and derive below.

In the following we average immediately over the positive and negative values μ of the LSF's. Then the product of the two scattering amplitudes in Γ^0_{++--} is

$$\frac{1}{2}\left\{\frac{1}{k^2}5^2\exp(i\delta_+)\sin\delta_+\exp(i\delta_-)\sin\delta_-+\text{c.c.}\right\}-P_2^2(\cos\Theta).$$

The integration over all angles gives a cross section σ_{+-},

$$\frac{\sigma_{+-}k_F^2}{4\pi}=5\cos(\delta_+-\delta_-)\sin\delta_+\sin\delta_-$$

which yields a rate $1/\tau_{+-}=n_d\sigma_{+-}v_F$. Therefore one finds $\Gamma^0_{++--}=\Gamma^0_{--++}$,

$$\Gamma^0_{++--}=\frac{\hbar}{2\pi g_0}\left(\frac{1}{\tau_0}+\frac{1}{3\tau_{so}}+\frac{1}{\tau_{+-}}\right).$$

The (weak localization) quantum correction to the conductivity is proportional to the sum of Γ_{++++}, Γ_{----}, Γ_{+--+}, and Γ_{-++-}. With the Dyson equation for weak localization one obtains for $\Gamma_{++++}=\Gamma_{----}$

$$\Gamma_{++++}=\frac{\hbar}{2\pi g_0\tau}\frac{1}{Dq^2\tau-i\omega\tau+4\tau/3\tau_{so}}$$

and for $\Gamma_{+--+}=\Gamma_{-++-}$

$$\Gamma_{+--+}=\frac{1}{2}\frac{\hbar}{2\pi g_0\tau}\left\{-\frac{1}{Dq^2\tau-i\omega\tau+\tau/\tau_F}+\frac{1}{Dq^2\tau-i\omega\tau+4\tau/3\tau_{so}+\tau/\tau_F}\right\},$$

where $1/\tau_F=1/\tau_d-1/\tau_{+-}$. The dephasing rate $1/\tau_F$ can be expressed by the corresponding cross section σ_F with $1/\tau_F=n_d v_F\sigma_F$, where

$$\frac{\sigma_F k_F^2}{4\pi}=\frac{5}{2}\sin^2(\delta_+-\delta_-).$$

Finally one replaces $-i\omega$ by $1/\tau_i$, the inelastic rate, and obtains for the one-dimensional slowly varying LSF's the characteristic fields in Eq. (3). While H_1 is the same as in Eq. (5) the other characteristic fields are $H_2=H_i+(4/3)H_{so}$, $H_3=H_i+H_F$, $H_4=H_i+(4/3)H_{so}+H_F$.

In the case of three-dimensional slow LSF's the orientation of the LSF's can point in any direction. Then one can express the spin-up and spin-down electrons in the basis of this quantization direction. This introduces two amplitudes α and β which depend on this direction. Formally one obtains spin-flip processes and the calculation contains more algebra but is otherwise quite analogous. Finally one has to average the coefficients $|\alpha|^2$ and $|\beta|^2$ over all directions. The result differs only in the explicit form of the characteristic fields for which one finds $H_2=H_i+(4/3)H_{so}+(1/3)H_F$, $H_3=H_i+(2/3)H_F$, $H_4=H_i+(4/3)H_{so}+(2/3)H_F$.

In the case of fast LSF's the upper and lower propagator see uncorrelated phase shifts $\delta_{\pm}$ at the impurity because they see the d impurity at different times. This yields for the three-dimensional LSF's the fields which are listed in Eq. (5).

One can experimentally determine the fluctuation cross section σ_F. For the dilute titanium impurities on the surface of gold we observed at 9.5 K a dephasing of weak localization which corresponds to a value of $k_F^2\sigma_F/4\pi=2\times10^{-3}$. For a d impurity the density of states can be expressed in terms of Γ (half the resonance width) and the number of d electrons of the impurity;

$$g_d=\frac{5}{\pi\Gamma}\sin^2\delta_2.$$

Assuming for the Ti impurity two d electrons yield $\delta_2=\pi/5$ and $g_d\simeq0.55/\Gamma$. If we take for $\Gamma\simeq0.5$ eV then we find $[Ug_d]/(1-Ug_d)=0.8$. This yields a value of Ug_d for Ti of 0.45 and a Stoner enhancement factor of about 1.8.

The authors tried to show that weak localization is a very effective method to study LSF's. This method is particularly effective for surface impurities where the spin fluctuating impurities are evaporated after the pure host film is investigated. With this procedure one can determine the small additional dephasing due to the surface impurity. This method is much more accurate than only investigating the sample with the impurities already present.

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