Weak Antiferromagnetism of Monolayers and Multilayers of V on Au Films

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Weak localization and the anomalous Hall effect are used to study the magnetic properties of submonolayers, monolayers, and multilayers of vanadium on the surface of Au films. Dilute V atoms possess a strong magnetic moment. For a monolayer the magnetic scattering is reduced by a factor of about 40. This suggests a strongly reduced moment of the V compared with the dilute V coverage. From the anomalous Hall effect we conclude that the magnetic structure is antiferromagnetic. For multilayers the moment per V atom progressively diminishes but is still finite for 16 atomic layers of V.

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The formation of magnetism in monolayers and multilayers has attracted considerable interest over several decades. It started with the search for (magnetically) dead layers of normally magnetic materials. In recent years the interest shifted towards the formation of magnetic monolayers and multilayers in metals which are normally nonmagnetic. One interesting prediction is the formation of magnetic moments in monolayers of V on the surface of Ag (and Au). Fu, Freeman, and Oguchi [1] and Bluegel *et al.* [2] predicted that a monolayer of V on the (100) surface of Ag is magnetic. The prediction for the V moment is about $2\mu_B$. According to Bluegel *et al.* the structure of the monolayer is antiferromagnetic (see, for example, the review article by Freeman and Wu [3]).

The magnetic behavior of V on Ag surfaces has been experimentally investigated by several groups [4–8]. Drube and Himpsel [4] observed unoccupied $3d$ states of submonolayers of V (and Mn) on Ag(111) which according to their interpretation suggested a ferromagnetic exchange splitting of the V $3d$ states. Stampanoni *et al.* [5] and Fink *et al.* [8] were searching for the originally predicted ferromagnetism of V monolayers on Ag and did not observe ferromagnetic behavior. Rau *et al.* [7] studied V films between 1 and 7 monolayers on Ag(100) by means of electron capture spectroscopy. They observed ferromagnetic behavior with a thickness dependent Curie temperature. Moodera and Meservey [6] investigated quenched films of V, Ag/V, and Au/V on Pb. They measured the reduction of the superconducting transition temperature of Pb by the coverage with V, Ag/V, and Au/V, respectively. They concluded that the V is antiferromagnetic up to a thickness of 1.5 monolayers of V. For larger V thicknesses their measurements suggested ferromagnetic behavior of the V.

In this paper we will use two methods to investigate the magnetic properties of V on the surface of Au. The first one is weak localization which is a powerful method to study characteristic scattering times in thin disordered metal films [9]. Magnetoresistance measurements correspond to a time-of-flight experiment which yields the inelastic, spin-orbit, and magnetic scattering times. The fact that the coherence of the conduction electrons is destroyed by magnetic scattering roughly after the magnetic scattering time results in an increase of the width of the magnetoresistance curves which can be well evaluated with the theory by Hikami, Larkin, and Nagaoka [10]. This method does not depend on the formation of a ferromagnetic state, it is very sensitive to the existence of magnetic moments.

Our second method is the anomalous Hall effect (AHE) which has been very useful in proving the existence of dead Ni layers on the surface of polyvalent metals [11]. The AHE is very efficient in observing ferromagnetic behavior. Both methods combined permit the identification of a ferromagnetic or antiferromagnetic state (we define here as antiferromagnetic a state with compensated total magnetic moment).

Our film samples are prepared by *in situ* condensation onto a quartz substrate at helium temperature. In a typical experiment a Au film with a thickness of about 25 atomic layers and a resistance per square of about 120 Ω is condensed. The vacuum is better than 10^{-11} torr. The quench condensed film is homogenous. With the exception of some cases of epitaxial evaporation where one obtains monolayer by monolayer this yields probably the best flat films. After the evaporation the film is annealed to 40 K because quench condensed films have many lattice defect with atoms being in local energy minima. Since some of the measurements are performed at 20 to 25 K the defects would anneal and the film changes its local structure irreversibly. This is undesired and can be avoided by annealing to 40 K so that the structure does not change during the experiment.

The Au film is then investigated in the temperature range between 4.5 and 20 K. The Hall resistance and the magnetoresistance are measured. In the following evaporation steps V is condensed on top of the Au film. The V is evaporated from a thin wire and its evaporation rate is carefully calibrated. Details of the thickness and evaporation rate calibration have been published in earlier work [12]. In several experiments the V thickness is varied between 0.005 and 16 atomic layers by essentially doubling the thickness each time. After each evaporation the film is annealed to 35 K and the measurements are repeated.

A set of magnetoresistance curves for different V coverages, measured at 4.5 K is shown in Fig. 1. The upper curve represents the pure Au film, the second curve Au with a coverage of 0.42 atomic layer of V. This coverage shows the widest magnetoresistance curve and therefore the strongest magnetic scattering. For thicker V coverage the curves narrow again, demonstrating a reduction of the magnetic scattering. The width of the magnetoresistance curve is described by the dephasing field H_i^*. This field is proportional to $1/\tau_i + 2/\tau_s$, where τ_i is inelastic and τ_s is the magnetic dephasing time of the conduction electrons. The latter is due to magnetic scattering by the V atoms. (The proportionality constant is 0.34 ps T.) We evaluated the magnetoresistance curves with the theory of weak localization by Hikami, Larkin, and Nagaoka [10]. This theory yields the dephasing field H_i^* of the conduction electrons. In Fig. 2 H_i^* is plotted as a function of the V thickness. One observes a steep increase for small V thicknesses, a maximum at about a thickness of 0.4 atomic layer of V and a decrease for thicker V coverages. Since the initial increase is so strong, it is plotted in an expanded scale to show the linear increase for thicknesses up to $\frac{2}{100}$ of a monolayer.

These results prove clearly that the V on top of the Au film possesses a magnetic moment. They also suggest that the magnetic moment is relatively strong for very low coverage up to 0.02 atomic layer of V. The dephasing rate increases proportional to the number of V atoms. This suggests that in this range of coverage each V atom has the same magnetic moment and magnetic interaction with the conduction electrons and their effect is simply additive. Between a coverage of 0.02 and 0.4 atomic layer of V the magnetic dephasing still increases. However, the effect per V atom is strongly reduced. Above 0.4 atomic layer of V even the accumulated effect of all V atoms decreases with increasing V thickness and the (averaged) dephasing of an individual V atom diminishes much more rapidly. For 0.1 atomic layer the dephasing per V atom is already by a factor of 5 smaller than for the thinnest coverage and for one monolayer of V the reduction of the dephasing is about 40. For several monolayers of V the efficiency of the magnetic scattering is further reduced.

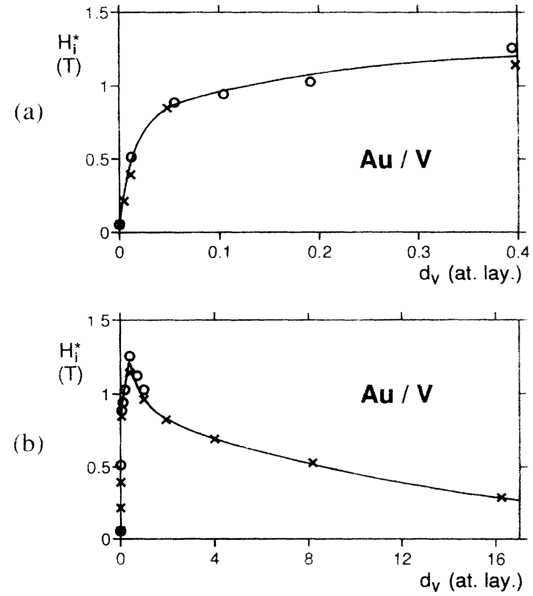

FIG. 2. The dephasing field (which is proportional to $1/\tau_i + 2/\tau_s$) as a function of the V thickness: (a) for small V thickness, (b) for the whole thickness range.

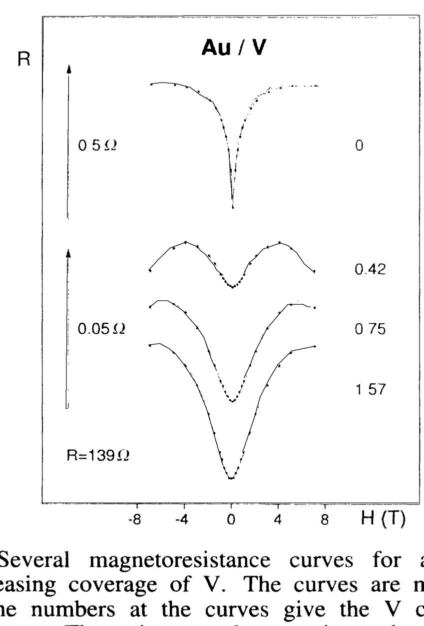

FIG. 1. Several magnetoresistance curves for a Au film with increasing coverage of V. The curves are measured at 4.5 K. The numbers at the curves give the V coverage in atomic layers. The points are the experimental values. The curves are fitted with the theory of weak localization.

The AHE is very well suited to identify a ferromagnetic order (see, for example, [13]). The anomalous Hall resistance is essentially proportional to the magnetization perpendicular to the film plane. For a ferromagnetic metal the magnetization increases linearly with the applied magnetic field B until B equals the saturation magnetization. Then the AHE saturates and one observes only the normal Hall effect (and possibly a high field susceptibility). For our measured Hall curves for V on Au the AHE contribution is very small but it clearly does not show a ferromagnetic behavior (for any V thickness). Details will be published elsewhere. Therefore we conclude that magnetic moments of the V atoms compensate each other as is the case in an antiferromagnetic layer.

The interesting question is whether the reduction of the magnetic scattering for V coverages above 0.02 atomic layer is due to the antiferromagnetic state or a reduction in the magnetic moment. There are very few investigations of weak localization in the antiferromagnetic state. Komaori, Kobayashi, and Sasaki [14] investigated the dephasing in Mn and CuMn films. But this experiment does not show the strength of the dephasing as a function of the coverage of the antiferromagnetic metal. Therefore we investigated the system Au/Mn and evaporated Mn in similar steps on the surface of the Au. We find that a thin coverage of 0.01 atomic layer of Mn on Au causes a similar dephasing as V. However, when we increase the Mn coverage the dephasing field increases strongly with the Mn coverage until at about 0.1 atomic layer of Mn the dephasing is so strong that the magnetoresistance curves can no longer be evaluated. (An additional magnetoresistance appears which is not due to weak localization but to spin-flip scattering.)

We conclude from the above Au/Mn experiment that the antiferromagnetic structure does not strongly reduce the dephasing which in first approximation is proportional to the number of magnetic atoms. This leads us to the conclusion that in our Au/V system the magnetic scattering of the individual V atoms is strongly reduced with increasing V coverage. It is extremely unlikely that the magnetic moments remain constant and only their interaction with the conduction electrons is reduced. As a matter of fact the opposite assumption makes much more sense; that the average magnetic moment drops with increasing V thickness. This suggests a strong reduction of the average V magnetic moment for a monolayer of V on Au although we cannot derive the exact value of the V moment. Such a reduction of the V moment is in contrast to the theoretical calculations.

In the theoretical papers the magnetic moment for a single monolayer of V on top of a metallic substrate is calculated for a ferromagnetic [1,15] and an antiferromagnetic [2] structure. According to the numerical calculations the antiferromagnetic structure has the lower energy; the magnetic moments in both structures agree almost perfectly. For V on top of the Ag(100) surface the calculated moment is $2\mu_B$ [2].

Almost all theoretical investigations calculate only the properties of single monolayers of V on a metallic substrate. An exception is an early paper by Fu, Freeman, and Oguchi [1] which compares the magnetic properties of a single and a double monolayer of V on Ag. In this investigation the authors considered only a ferromagnetic state of the V. They found for two layers of V a reduction of the magnetic moment in the surface layer to $1.15\mu_B$ and the moment in the subsurface layer was less than $0.05\mu_B$. If one assumes that the main contribution to the energy comes from the formation of the magnetic moments and the structure yields only a small influence then one might extrapolate this result also to the antiferromagnetic state.

This theoretical result of a large magnetic moment in the surface layer is definitely not confirmed by our experiments. The theory suggests that for a V thickness of more than a monolayer the magnetic moment drops. This result is confirmed by our experiment. Between one monolayer and 16 atomic layers the magnetic scattering drops. However, even for 16 atomic layers we still find some magnetic scattering. This can be best seen from the temperature dependence of the effective dephasing field. The latter is plotted in Fig. 3 for different V coverages in a log-log plot. The dephasing field of the pure Au film is much smaller and has a slope of 1.9 which is close to the T^2 law. For a pure V film we found also roughly a T^2 dependence for the inelastic field. In both cases the dephasing is caused only by inelastic processes and $1/\tau_i$ is essentially proportional to T^2. However, for V on top of Au the temperature dependence of H_i^* in Fig. 3 does not show a simple power law. The reason is that H_i^* is proportional to the sum of the inelastic rate $1/\tau_i$ and twice the magnetic scattering rate $1/\tau_s$. The latter is in first approximation temperature independent and distorts the power law. One recognizes that this distortion reduces with increasing V coverage but does not disappear, not even for 16 atomic layers of V. The conclusion is that the magnetic dephasing reduces but is still present in a 16 atomic layers thick V film on top of Au. There is no clear theoretical prediction for the decay of the magnetic moment at the interface with increasing V thickness.

We compare our experimental results with the theoretical predictions. (a) In agreement with the theory we find an antiferromagnetic ordering for V on Au. (b) We observe a very strong magnetic scattering for V coverages below 0.02 atomic layer. This is in qualitative agreement with the theoretical value of $2\mu_B$ for a monolayer of V. (c) The strong reduction of the magnetic scattering

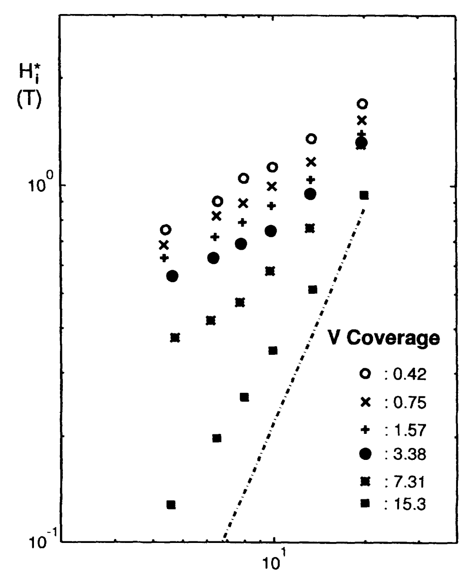

FIG. 3. The temperature dependence of the dephasing field for different V coverages. Films without magnetic scattering have a T^p dependence with $p \simeq 2$ (see dashed line).

for a monolayer of V on Au suggests a magnetic moment of the V which is much smaller than the theoretical value. (d) We find a reduced magnetic scattering for increasing V thickness in qualitative agreement with the theoretical calculation for two monolayers of V on Ag.

In the theoretical calculations an overlayer of V with perfect periodic structure was considered whereas our overlayers are disordered and rather statistical in their structure. One might argue that this difference in structure is the reason for the reduced magnetic signature of our V atoms. However, this argument is incorrect. The moment formation of the V is favored when the overlap of its wave function with its neighbors is reduced. In a disordered overlayer the perfect overlap is reduced compared with that of the periodic structure. Therefore one would rather expect that in a disordered overlayer the tendency towards magnetism is slightly enhanced over that in a periodic film. Obviously our experimental results represent an interesting challenge for the theory.

One might suspect that the interface of Au/V develops a layer of magnetic V which becomes independent of the V thickness when d_V exceeds a critical thickness. This would be a serious statement because it suggests that even bulk V might have a magnetic surface. (In the theoretical calculations the role of the normal metal is to block the hybridization of neighboring V d orbits. The normal metal acts to a certain extent as an inert material.) Rau *et al.* [16] reported a ferromagnetic V surface layer on the (100) surface of bulk V. We examined this question by preparing first a pure V film with a thickness of 36 atomic layers and measured its dephasing field. It showed a T^p dependence on the temperature with p slightly larger than 2. This is the typical result for nonmagnetic metal films and does not support a magnetic surface on bulk V. Then we covered the V film with increasing thicknesses of Au up to a thickness of 13 atomic layers. The resulting dephasing field H_i^* changed only according to the proximity effect of the two films (which means that it averages between the inelastic field of V and Au). It still showed a power law as a function of the temperature with p close to 2. In the third step we covered the Au with V (up to a thickness of 6 atomic layers). This introduced a strong magnetic scattering. Therefore in our experiment the surface of V as well as the interface Au/V do not develop magnetic moments. Only when the V thickness is sufficiently small does the moment occur and its size decays with increasing V coverage. The surprising effect is that for a V film of 16 atomic layers on top of Au the magnetism of the V is still affected by the film thickness. There must be a large correlation length involved in this phenomenon.

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