

## Photovoltaic effect in Au and Au-Fe microjunctions

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Recent work has shown that when a small conductor is exposed to a microwave electric field at low temperatures, a dc voltage is established. We have studied this photovoltaic (PV) effect as a function of magnetic field,  $H$ , in Au microjunctions, some of which were doped with small concentrations (typically  $\sim 50$  ppm) of Fe. In the pure (undoped) samples the PV effect was independent of the sign of  $H$ ; i.e., it was an even function of  $H$ . In contrast, the doped junctions displayed a PV signal which was an odd function of the magnetic field. This change in the symmetry of the PV effect appears to be associated with the localized magnetic moments of the Fe, but a theoretical explanation of this behavior is not currently available.

### I. INTRODUCTION

A mesoscopic system is one whose dimensions are comparable to or smaller than the distance over which the conduction electrons are able to travel while maintaining phase coherence. This coherence makes possible a number of quantum interference phenomena, including universal conductance fluctuations and Aharonov-Bohm-like oscillations of the conductance of multiply connected systems, which have attracted a great deal of attention in the past several years.<sup>1-3</sup> A related effect, which has only recently been observed, concerns the response of a mesoscopic sample to an ac electric field. Fal'ko and Khmel'nitskii<sup>4,5</sup> have predicted that such a field would lead to a dc voltage, and studies of this photovoltaic (PV) effect in the microwave regime have been reported in both semiconductor<sup>6,7</sup> and metal film structures.<sup>8,9</sup> While the general behavior is in fairly good agreement with the theory, some interesting questions remain unresolved. In particular, in previous work on small metal systems which contained magnetic impurities<sup>9</sup> the PV signal responded in an unexpected manner to changes in the sign of an applied magnetic field. However, the concentration of magnetic ions in those samples was rather large, and it is conceivable that effects such as the formation of a spin-glass phase, or clustering of the magnetic ions might be responsible for those results. In this paper we report further more detailed studies of the symmetry properties of the PV effect in Au and Au-Fe microjunctions with much lower Fe concentrations than in the previous work. Our results demonstrate that the presence of Fe at nominal concentrations of only  $\sim 50$  ppm can indeed have a pronounced effect on the symmetry of the PV signal.

### II. THEORY

The qualitative nature of the photovoltaic response can be understood from the following argument due to Fal'ko and Khmel'nitskii.<sup>4,5</sup> A sample of conductivity  $\sigma$

which is exposed to a high-frequency ac electric field  $E_{ac}$  will absorb an energy  $\sigma E_{ac}^2 V$  per unit time, where  $V$  is the sample volume. This energy will go towards exciting electrons above the Fermi level, and these electrons will diffuse in different directions. In a perfectly symmetric sample which is attached to two symmetric leads, an equal number of electrons will diffuse towards either end of the sample. However, if the sample is not perfectly symmetric the numbers of electrons diffusing in the two directions will not be equal, resulting in a dc voltage. All real systems will contain some disorder, and hence are asymmetric to some extent; thus all such systems should exhibit a nonzero photovoltaic effect. The surprising aspect (at least to us) is that in a mesoscopic system the magnitude of this PV effect can be quite significant.

A quantitative calculation of the PV voltage gives<sup>5</sup>

$$\overline{V_{dc}} = 4\pi\sqrt{\zeta(3)}\frac{e}{G}\left(\frac{\omega}{\tau_\phi}\right)^{1/2}\left(\frac{eE_{ac}L}{\pi\hbar\omega}\right)^2, \quad (1)$$

where  $\zeta(x)$  is the zeta function,  $e$  is the electronic charge,  $G$  is the conductance, and  $L$  is the length of the system, which is assumed to be less than (or comparable to) the electron phase coherence length  $L_\phi$ . For parameters appropriate to our experiments, (1) yields a PV voltage of order 1-10 nV.

It is important to note that  $\overline{V_{dc}}$  in (1) is the ensemble average, rms voltage. If one considers an ensemble of statistically similar but microscopically distinct samples, any particular member of the ensemble may exhibit a value somewhat smaller or larger than this, and the sign of  $V_{dc}$  will also be random. These fluctuations in the value of  $V_{dc}$  are closely related to universal conductance fluctuations (UCF),<sup>2,10,11</sup> which are fluctuations in the conductance of an ensemble of small disordered systems. Indeed, calculations show that in the PV effect, the asymmetry in the diffusion, i.e., the fraction of excited electrons that diffuse toward one end of the system as opposed to those that diffuse in the opposite direction, is proportional to  $\delta G/G$ , where  $G$  is the conductance of

the system, and  $\delta G \sim e^2/h$  is the magnitude of universal conductance fluctuations.<sup>5</sup>

The close relation between the PV effect and UCF suggests how  $V_{dc}$  will be affected by a magnetic field. Insofar as UCF are concerned, it is well established that the application of a sufficiently large magnetic field is equivalent to changing the microscopic arrangement of the disorder, for the following reason.<sup>1-3,10,11</sup> The semiclassical electron trajectories in a disordered system are diffusive, since the elastic mean free path is very short, with each step in a diffusion path corresponding to scattering from an impurity or other scattering center. The total probability for an electron to travel from  $\mathbf{r}_1$  to  $\mathbf{r}_2$  is determined by the sum of the amplitudes for the different paths which go from  $\mathbf{r}_1$  to  $\mathbf{r}_2$ . The interference between the amplitudes for different paths is extremely important, and leads to a number of effects including weak localization. Any process which affects this interference can thus have a profound effect on the electron transport. A magnetic field will shift the relative phases for different trajectories, thereby affecting the overall interference and yielding a change (i.e., fluctuation) in the conductance. In this way, the application of a magnetic field can be equivalent to changing the arrangement of the disorder in the system. Since very similar physics is responsible for the PV effect, we expect that applying a magnetic field should produce a change in the PV signal which is equivalent to changing samples. Measurements of  $V_{dc}$  as a function of  $H$  should thus yield a fluctuating result, whose rms value is given by (1). The field change required to effect a “full” change in  $V_{dc}$  should be of the order of the field required to produce one quantum of flux through an area  $L_\phi \times L_\phi$ .

This connection between the PV effect and UCF also suggests how  $V_{dc}$  will be affected by changes in the sign of  $H$ . It has been shown<sup>12,13</sup> that for a nonmagnetic system, a two-lead measurement of the conductance should satisfy  $G(H) = G(-H)$ . This result is valid even in the presence of universal conductance fluctuations. The connection between UCF and the PV effect then suggests that one should expect  $V_{dc}(H) = V_{dc}(-H)$  in this case. This is in agreement with previous experiments<sup>9</sup> (and with our results; see below) on nonmagnetic samples, which have found that the PV signal is independent of the sign of  $H$ .

Considerations of the symmetry of  $G$  with respect to changes in the sign of  $H$  become more complicated when the sample is magnetic. According to the theory (again for a two-lead measurement), if the magnetization  $M$  of the sample changes sign when the sign of  $H$  is reversed, then the symmetry of  $G$  is maintained;<sup>12</sup> that is,  $G(H, M) = G(-H, -M)$ . This is the situation that we would expect to apply to our experiments. Our samples have relatively low concentrations of magnetic impurities; hence the impurity spins are far apart and should behave as “free” spins, in which case the magnetization is always parallel to  $H$ . Since we expect the PV effect to possess symmetry properties similar to those of  $G$ , we anticipate  $V_{dc}(H) = V_{dc}(-H)$  in a system doped with a small concentration of magnetic spins, as in the Au-Fe samples we have studied.

However, as will be seen below, our results indicate that the symmetry of  $V_{dc}$  with respect to changes in the sign of  $H$  is not always consistent with the simple arguments given above. One can readily imagine situations for which these arguments would not apply. For example, even though the average concentration of magnetic spins may be small there could nevertheless be “clustering” of the magnetic ions, leading to small regions where their concentration is relatively high. In such regions the system might be a spin-glass or a ferromagnet. If so, then one would no longer necessarily expect the sign of  $M$  to change when the direction of  $H$  is reversed, and one might also find hysteresis. If the interactions between the impurity spins are strong then  $M$  might be independent of  $H$ , at least for small fields, in which case measurements of the conductance would essentially be comparing  $G(H, M)$  to  $G(-H, M)$ . To the best of our knowledge there is no simple general relation between these two quantities. In such a case one would presumably not expect the behavior of  $V_{dc}(H)$  to be correlated with  $V_{dc}(-H)$ . Of course, the magnetic behavior might be more complicated, and  $M(H)$  might not be simply related to  $M(-H)$ , so that again one would expect no simple relation between  $V_{dc}(H)$  and  $V_{dc}(-H)$ . We will return to these points below, after we have presented our results for the PV effect in Au-Fe.

Returning to (1), we note that this prediction is applicable only when the sample dimensions are smaller than the phase coherence length  $L_\phi$ . If this is not the case, then different regions of the sample, each of which is of order  $L_\phi$  in size, will contribute independent PV voltages, resulting in a decrease in the overall magnitude of  $\overline{V_{dc}}$ . In our experiments, the sample size was typically comparable to (and never more than a factor of 2 or 3 larger than)  $L_\phi$ , so this self-averaging was never very large. According to the theory<sup>4</sup> it should not have depressed  $\overline{V_{dc}}$  by more than a factor of about 3–10. Since our work has focussed on the *qualitative* behavior of  $V_{dc}$  as a function of  $H$ , the precise magnitude of  $V_{dc}$  will not be important in our analysis. We will therefore not require a quantitative estimate of the effect in our analysis.

### III. EXPERIMENTAL SETUP

The samples were made from films of Au and Au-Fe which were patterned with optical lithography and ion milling.<sup>9,14</sup> The samples had a two-lead constriction (i.e., microbridge or junctionlike) geometry, similar to that described previously,<sup>9,15</sup> which is shown schematically in the inset of Fig. 1. Typical junctions were 1  $\mu\text{m}$  long, 1  $\mu\text{m}$  wide, and 150  $\text{\AA}$  thick, with a sheet resistance of  $\sim 3 \Omega$ . The film leading away from the microjunction was continuous with much wider regions of the same film, and was in turn connected with much thicker Au (99.999%) or Ag (99.9999%) films to which contacts were attached. Thus, the contact films immediately adjacent to the microcontact were composed of the same material as the junction itself.

The films were prepared by thermal evaporation onto glass substrates at room temperature. The nominally

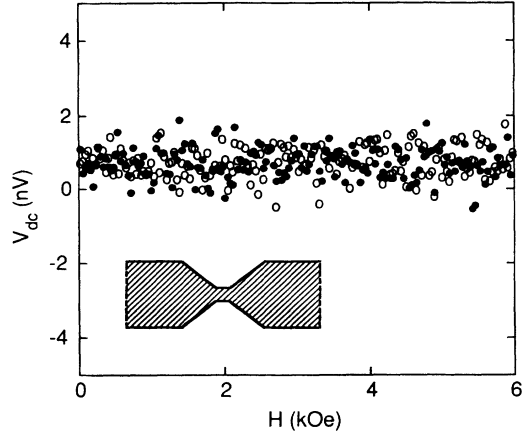


FIG. 1.  $V_{dc}$  as a function of  $H$  for a large ( $6 \times 6 \text{ mm}^2$ ) Au sample at 4.2 K. The open symbols were taken with the field swept upwards, while the closed symbols were obtained with  $H$  swept down. The inset shows the sample geometry.

pure Au had a purity of 99.999%, with an Fe concentration of 1 ppm, and a total concentration of other magnetic impurities of less than 2 ppm. The Au-Fe films were prepared by coevaporation of measured amounts of the pure Au together with Au-Fe alloy wire. The latter contained 0.07 at. % Fe, and had a diameter of  $75 \mu\text{m}$ . Hence, we were essentially diluting the Fe concentration of the original Au-Fe alloy material, and in this way we prepared Au-Fe films with Fe concentrations ranging from 1 to 150 ppm. For the Au-Fe samples we have studied in this work, the Fe concentration was in the range 20–125 ppm. The experimental results reported below were all performed at 4.2 K. Similar, but less extensive results were obtained at 1.4 K. Measurements of the magnetoresistance of coevaporated films were used to infer the phase coherence lengths of each sample.<sup>16,17</sup> These lengths (at 4.2 K) ranged from  $\sim 6000 \text{ \AA}$  for the pure Au films, to  $\sim 3000 \text{ \AA}$  for the samples with 125 ppm Fe. The phase coherence length decreased with increasing Fe content due to the increased importance of magnetic scattering.<sup>16</sup>

The apparatus was similar to that described previously.<sup>9,18</sup> The magnetic field was applied perpendicular to the plane of the film, with the microwave electric field directed in the plane of the sample, and a microwave frequency of 8.4 GHz. The microwave amplitude was modulated at either 77 or 103 Hz, and the voltage produced at this frequency was detected with a lock-in amplifier. The magnitude of the microwave field was held fixed for all of the measurements reported below, and was estimated from previous work<sup>18</sup> to be  $\sim 15 \text{ V/m}$ .

To check that the PV signal we observed was indeed a mesoscopic effect, we studied the behavior of a large,  $6 \times 6 \text{ mm}^2$  sample, which should of course not exhibit an appreciable PV effect, and Fig. 1 shows some typical results. It is seen that  $V_{dc}$  as a function of  $H$  is featureless; it is dominated by the noise of the measuring electronics. To within this noise level there was no PV signal in our large samples. In comparison, we will see below that

the PV signals for the mesoscopic samples are typically a few nV or larger, and vary significantly with  $H$ . One can also see from Fig. 1 that the average value of the signal is  $\sim 0.5 \text{ nV}$ , which one might be tempted to identify as a PV effect in this sample. However, the fact that this voltage was independent of field, in contrast to both the theoretically expected behavior of the PV effect and the behavior observed in the mesoscopic samples, leads us to conclude that it is not a PV effect, but is due rather to interference between the modulation circuit which drives the microwave source, and the detection electronics. Similar pickup, which was independent of temperature, can be seen in all of the data shown below, and its magnitude varied from run to run (although it was constant for a given run). Fortunately, the pickup level was not large compared to the variations of the PV signal with  $H$ . In this paper we will be interested mainly in the overall symmetry of the PV signal, and as will be seen below, the presence of this offset of  $V_{dc}$  due to the pickup will not affect our analysis. We should also note that Joule heating by the ac field could conceivably be a problem. However, we expect that effects due to heating would be proportional to the temperature derivative of the resistance,  $dR/dT$ . We found no correlation between  $V_{dc}$  and  $dR/dT$ , so we believe that Joule heating was not important.

#### IV. RESULTS

Figure 2 shows results for a nominally pure Au sample. Here and in all of the other results shown below the measuring temperature was 4.2 K (similar but less extensive results were obtained at 1.4 K). This sample, and the others considered below, had junction dimensions of  $\sim 1 \times 1 \mu\text{m}^2$ . The filled and open circles show results obtained by sweeping the field up and down (respectively), with a positive field polarity. The results were independent of the speed with which the field was swept, or even if the sweep was stopped and continued

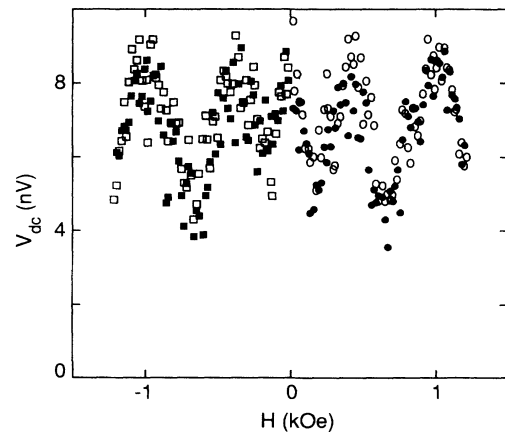


FIG. 2.  $V_{dc}$  as a function of  $H$  for a  $1 \times 1 \mu\text{m}^2$  Au sample, at 4.2 K. The open symbols were taken with the field swept towards increasing magnitude, while the closed symbols were taken in sweeps towards decreasing fields.

later. The boxes show results obtained for corresponding sweeps with a negative field polarity (the filled boxes were obtained with increasing  $|H|$  while the open boxes correspond to decreasing  $|H|$ ). It can be seen from Fig. 2 that the PV signal is in this case an even function of  $H$ , and that it is independent of the direction with which the field is swept. These results for  $V_{dc}(H)$  were quite reproducible; repeated sweeps yielded the same results (to within the uncertainties) so long as the sample temperature was maintained at 4.2 K. When the sample was allowed to warm to 77 K, and then recooled to 4.2 K, a completely different PV curve was found. However, the *symmetry* of  $V_{dc}(H)$  was *always* the same; it was *always* an even function of  $H$ . This symmetry is consistent with the arguments given in Sec. II above, and thus seems to be consistent with the theory. Behavior similar to that shown in Fig. 2 was found in other pure Au samples. The value of  $\overline{V}_{dc}$  as obtained from the rms fluctuation magnitude in Fig. 2 is  $1.3 \times 10^{-9}$  V. If we allow for subsystem averaging which should depress the value of  $\overline{V}_{dc}$  by a factor of 2-4 (since the sample size is about a factor of 1.5 larger than  $L_\phi$ ), then this compares fairly well with the theory (1) which predicts<sup>19</sup>  $7 \times 10^{-9}$  V.

Figure 3 shows results for a Au-Fe sample with an Fe concentration of 125 ppm. As in Fig. 2 we show results for positive and negative fields (circles and squares, respectively), and for increasing and decreasing field sweeps (filled and open symbols, respectively). Here, in contrast to Fig. 2, we see that  $V_{dc}$  is definitely *not* an even function of  $H$ . The PV signal here is an *antisymmetric* function of  $H$ , i.e., aside from the pick-up, the sign of  $V_{dc}$  changes when the sign of  $H$  is changed. The value of  $\overline{V}_{dc}$  obtained from the data in Fig. 3 is  $1.8 \times 10^{-9}$  V, which, after allowing for subsystem averaging,<sup>20</sup> compares reasonably well with the theoretical prediction of  $8 \times 10^{-9}$  V.

It is useful to obtain a quantitative measure of the relation between  $V_{dc}(H)$  and  $V_{dc}(-H)$  for both the pure Au sample of Fig. 2, and the Au-Fe sample of Fig. 3.

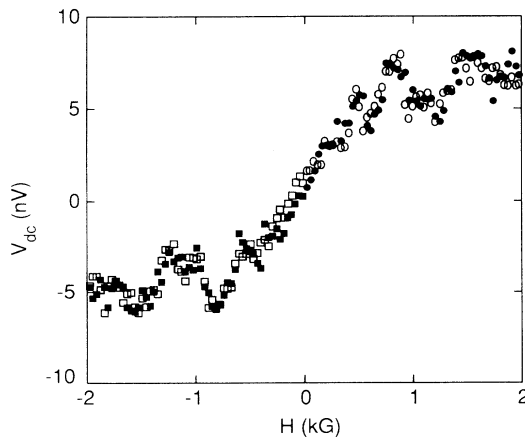


FIG. 3.  $V_{dc}$  as a function of  $H$  for a  $1 \times 1 \mu\text{m}^2$  Au-Fe sample, at 4.2 K. The Fe concentration was 125 ppm. The open symbols were taken with the field swept towards increasing magnitude, while the closed symbols were taken in sweeps towards decreasing fields.

Considering first the pure Au sample, Fig. 4(a) shows the quantity  $V_{dc}(H) - V_{dc}(-H)$  as a function of  $|H|$ . This quantity is seen to be zero to within the experimental scatter. The linear correlation between  $V_{dc}(H)$  and  $V_{dc}(-H)$  was calculated,<sup>21</sup> and found to be  $r = 71\%$ . It is not clear at present if the deviation of  $r$  from unity is significant, or is simply due to the experimental uncertainties. In Fig. 4(b) we consider the Au-Fe sample of Fig. 3, and show  $V_{dc}(H) + V_{dc}(-H)$  as a function of  $|H|$ . To within the experimental scatter, this quantity is seen to be a constant (the pickup discussed above), which simply confirms that  $V_{dc}$  is an odd function of  $H$ , as we concluded from Fig. 3. The correlation coefficient in this case was found to be  $-92\%$ , confirming that  $V_{dc}(H)$  and  $V_{dc}(-H)$  are almost completely anticorrelated.

For the Au-Fe sample in Fig. 3,  $V_{dc}$  was independent of the direction in which the field was swept, but this

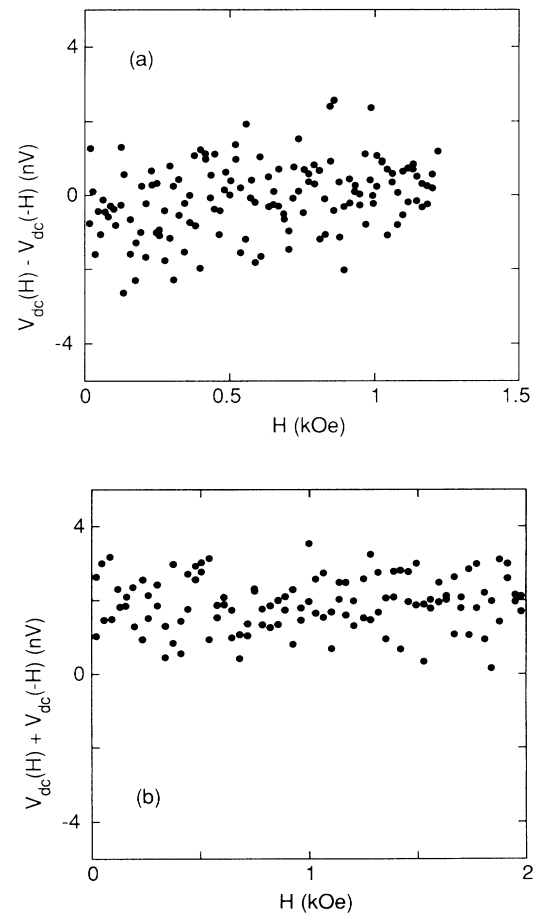


FIG. 4. (a)  $V_{dc}(H) - V_{dc}(-H)$  as a function of  $H$ , as calculated for a pure Au sample from the data in Fig. 2. This illustrates that the PV effect in this case is predominantly an *even* function of  $H$ , and that any antisymmetric component of  $V_{dc}$  is smaller than the experimental uncertainties. (b)  $V_{dc}(H) + V_{dc}(-H)$  as a function of  $H$ , as calculated for a Au-Fe sample from the data in Fig. 3. This illustrates that the PV effect in this case is predominantly an *odd* function of  $H$ , and that any symmetric component of  $V_{dc}$  is smaller than the experimental uncertainties.

was not always the case, as illustrated in Fig. 5. Here we show results for a Au-Fe sample with an Fe concentration of 20 ppm. Figures 5(a)–5(c) were all obtained with the *same* sample, the only difference being that it was ther-

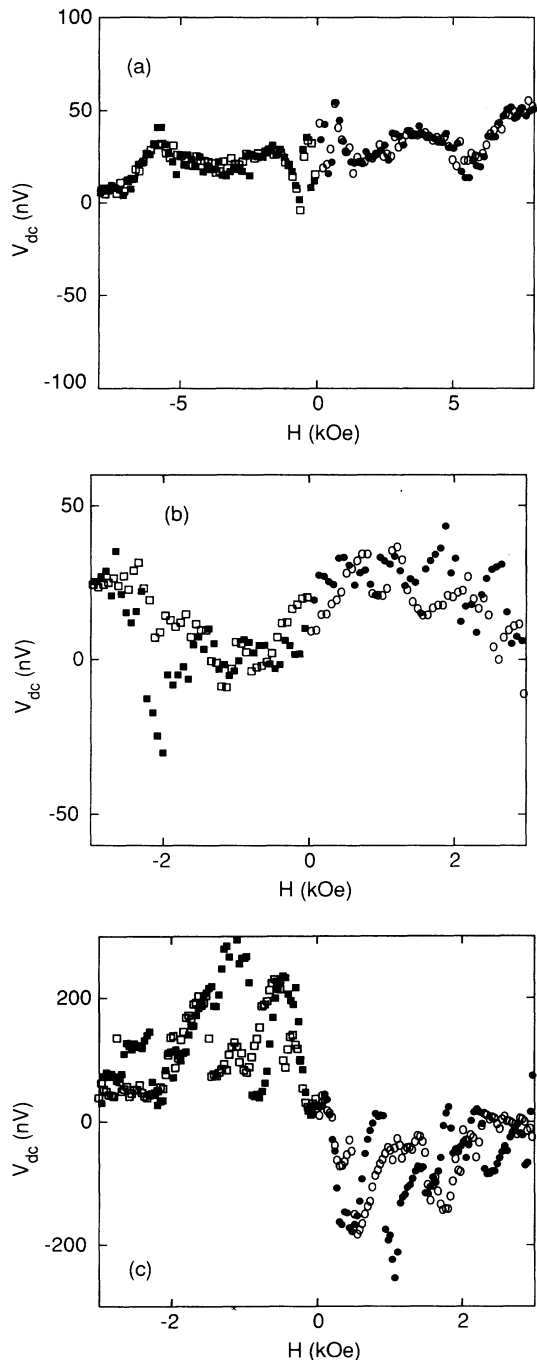


FIG. 5.  $V_{dc}$  as a function of  $H$  for a  $1 \times 1 \mu\text{m}^2$  Au-Fe sample, at 4.2 K. The Fe concentration was 20 ppm. The open symbols were taken with the field swept towards increasing magnitude, while the closed symbols were taken in sweeps towards  $H = 0$ . The data in (a)–(c) were all obtained with the same sample; the temperature was cycled to 77 K between scans. Note that the data in (b) were taken first, followed by (c), and then (a).

mally cycled to 77 K between measurements. In Fig. 5(a) the behavior is quite similar to that seen in Fig. 3;  $V_{dc}$  is an odd function of  $H$  and is essentially independent of the direction in which the field is swept. Note also that the magnitude of  $V_{dc}$  in Fig. 5(a) is somewhat larger than that seen in Figs. 2 and 3, and predicted from the theory (1). We will return to this point below.

In Fig. 5(b) we see a pronounced hysteresis; i.e., a dependence on the direction in which the field is swept. If one compares results for  $H >$  and  $H <$  0,  $V_{dc}$  is still an odd function of  $H$  so long as one compares measurements obtained only with the same sweep direction. That is,  $V_{dc}(H) = -V_{dc}(-H)$  for sweeps in which  $|H|$  was increasing; this is also true for decreasing sweeps, but the detailed form of  $V_{dc}(H)$  is different than found in the upward sweeps.  $V_{dc}$  in Fig. 5(b) is also comparable in size to that found in Fig. 5(a). In Fig. 5(c) the magnitude of  $V_{dc}$  is even larger, almost a factor of 4 larger than in Fig. 5(a) (and a factor of  $\sim 20$  larger than seen in Figs. 2 and 3), and the hysteresis is again large. The results in Fig. 5 are representative of those obtained with several other Au-Fe samples; it was found that the behavior of  $V_{dc}$  sometimes changed markedly after thermal cycling. It was not uncommon to observe hysteresis, and the magnitude of  $V_{dc}$  could change by nearly a factor of 10 after cycling. We should also note that this behavior did not appear to be due to an overall degradation of the sample, as it was found that the magnitude of  $V_{dc}$  could either increase or decrease after thermal cycling.

## V. DISCUSSION

As discussed in Sec. II, according to the theory we expect that for nonmagnetic systems  $V_{dc}$  should be an even function of  $H$ . This follows from the symmetry of the conductance in a two-lead geometry,  $G(H) = G(-H)$ , along with the close theoretical connection between universal conductance fluctuations and the PV effect. Such symmetric behavior was always found with our pure Au samples (i.e., Fig. 2), and in the nonmagnetic samples studied previously by our group.<sup>8,9</sup> The magnitude of  $\overline{V_{dc}}$  found with our Au samples was also in reasonable accord with the theory (1), so we conclude that for these samples theory and experiment are in good agreement.

The behavior of the Au-Fe samples is harder to understand. The hysteresis which was sometimes observed is clearly suggestive of magnetic effects, which is perhaps not surprising given that these samples contain Fe. However, on further examination our results are puzzling in several respects. First, the arguments relating the PV effect to UCF (Ref. 22) indicate that, at least for a nonmagnetic system,  $V_{dc}(H) = V_{dc}(-H)$ . However, it is not at all clear that the behavior should ever be an *odd* function of  $H$ . For a magnetic system one should not be surprised to find  $V_{dc}(H) \neq V_{dc}(-H)$ , since the magnetization may not change sign when the field is reversed. However, to find *antisymmetric* behavior is clearly a much stronger result.<sup>23</sup> We do not know of any argument as to how such behavior might arise. Second, the presence of hysteresis in  $V_{dc}$  suggests that  $M$  depends on how the field is cycled before a measurement. This would not be surprising in a

concentrated magnetic system, but our samples have relatively little Fe. While we do not know of any direct measurements of the spin-glass ordering temperature for Au-Fe alloys with concentrations similar to ours, one can use previous studies of the resistivity<sup>24</sup> and susceptibility<sup>25</sup> of these and more concentrated alloys to estimate an ordering temperature of  $\sim 0.1$  K or less for a concentration of 50 ppm, which is well below the range we have studied. Moreover, previous work<sup>24</sup> has shown that the Fe concentrations in the samples in Figs. 3 and 4 were low enough that the Kondo effect in bulk alloys is a linear function of the Fe concentration in our temperature range, implying that interactions between the Fe spins should be negligible. For a system of free spins,  $M$  should depend on  $H$  alone and not on past history; the observation of hysteresis suggests that in our case the spins are not free to follow  $H$ . Such "pinning" of the spin direction would be found, for example, in spin glasses, but our Fe concentrations would seem to be too low for such behavior. An obvious possibility here is that there is clustering of the Fe, although it is also conceivable that the Ruderman-Kittel-Kasuya-Yosida interactions between the Fe spins may be important.<sup>26</sup> Studies of the spin scattering through the weak-localization magnetoresistance<sup>27</sup> in similarly prepared Au-Fe films suggest that most (and perhaps all) of the Fe is present in the form of isolated spins, although a small amount of clustering cannot be ruled out. Indeed, the hysteresis which was sometimes observed implies fairly strongly that a magnetically ordered phase, probably as a result of clustering, was present in our Au-Fe samples. However, even if there was clustering of the Fe, it is *still* not clear how this could account for the antisymmetric behavior of  $V_{dc}$ , since as noted above we do not expect there to be any general relation between  $G(H, M)$  and  $G(-H, M')$ , or between  $V_{dc}(H, M)$  and  $V_{dc}(-H, M')$ , when  $|M| \neq |M'|$ .

It is also interesting to consider the field scale of the fluctuations of  $V_{dc}$ . In analogy with the magnetofingerprints common to UCF, we expect that in order for changes in the magnetic field to produce an effectively "different" sample,  $\Delta H L_\phi^2 \sim \phi_0$ , where  $\phi_0 = h/e$  is the flux quantum. For the pure Au samples,  $L_\phi \sim 6000$  Å, which gives  $\Delta H \sim 110$  Oe. Unfortunately, we do not have enough data to allow a calculation of the appropri-

ate correlation function, so we cannot give a quantitative result for the field correlation scale. The theoretical estimate of 110 Oe given above is somewhat smaller than one might infer qualitatively from Fig. 2, but until we have more extensive data we can only conclude that the observed field correlation scale is not inconsistent with the theory. For the Au-Fe sample in Fig. 5,  $L_\phi \sim 3000$  Å, which gives  $\Delta H \sim 450$  Oe, which is again at least qualitatively consistent with our results.

An important feature of the results for Au-Fe is that relatively large values of  $V_{dc}$  are sometimes observed, as in Fig. 5. Such large values were never found in the pure Au samples. These large values of  $V_{dc}$  are much greater than the typically 1–10 nV predicted by the theory (1), which may indicate that in this case  $V_{dc}$  arises from a mechanism different from that considered by Fal'ko and Khmel'nitskii. However, so far as we are aware, no other mechanisms have been suggested to date. Nevertheless, the similar symmetry properties of  $V_{dc}$  seen in the different cases, Figs. 3 and 5, i.e., when  $V_{dc}$  is of the expected size and when it is much larger than expected, may suggest a common mechanism.

In conclusion, our results indicate that the presence of magnetic impurities can have a profound effect on the symmetry of the PV effect, but how this happens is not understood. Our results raise the possibility that there may be other mechanisms, in addition to the one discussed by Fal'ko and Khmel'nitskii, which can contribute to the PV effect in mesoscopic systems. In addition, the variation of the PV effect with field may indicate some rather surprising behavior of the magnetization in these dilute spin systems. Both of these phenomena deserve further study.

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<sup>1</sup>See, for example, *Mesoscopic Phenomena in Solids*, edited by B. L. Al'tshuler, R. A. Webb, and P. A. Lee (Elsevier, Amsterdam, 1991).

<sup>2</sup>S. Washburn and R. A. Webb, *Adv. Phys.* **35**, 375 (1986).

<sup>3</sup>*Nanostructure Physics and Fabrication*, edited by M. A. Reed and W. P. Kirk (Academic, San Diego, 1989).

<sup>4</sup>V. I. Fal'ko and D. E. Khmel'nitskii, *Zh. Eksp. Teor. Fiz.* **95**, 328 (1989) [*Sov. Phys. JETP* **68**, 186 (1989)].

<sup>5</sup>V. I. Fal'ko, *Europhys. Lett.* **9**, 785 (1989).

<sup>6</sup>A. A. Bykov, G. M. Gusev, Z. D. Kvon, D. I. Lubyshev, and V. P. Migal, *Pis'ma Zh. Eksp. Teor. Fiz.* **49**, 13 (1989)

[*JETP Lett.* **49**, 13 (1989)].

<sup>7</sup>A. A. Bykov, G. M. Gusev, Z. D. Kvon, and B. I. Fomin, *Zh. Eksp. Teor. Fiz.* **97**, 251 (1990) [*Sov. Phys. JETP* **70**, 140 (1990)].

<sup>8</sup>J. Liu and N. Giordano, *Physica B* **165&166**, 279 (1990).

<sup>9</sup>J. Liu, M. A. Pennington, and N. Giordano, *Phys. Rev. B* **45**, 1267 (1992).

<sup>10</sup>B. L. Al'tshuler, *Pis'ma Zh. Eksp. Teor. Fiz.* **41**, 530 (1985) [*JETP Lett.* **41**, 648 (1985)].

<sup>11</sup>P. A. Lee and A. D. Stone, *Phys. Rev. Lett.* **55**, 1622 (1985); P. A. Lee, A. D. Stone, and H. Fukuyama, *Phys. Rev. B* **5**, 1039 (1987).

<sup>12</sup>M. Büttiker, *IBM J. Res. Dev.* **32**, 317 (1988).

- <sup>13</sup>A. D. Benoit, S. Washburn, C. P. Umbach, R. B. Laibowitz, and R. A. Webb, *Phys. Rev. Lett.* **57**, 1765 (1986).
- <sup>14</sup>D. W. Palmer and S. K. Decker, *Rev. Sci. Instrum.* **44**, 1621 (1973).
- <sup>15</sup>T. L. Meisenheimer and N. Giordano, *Phys. Rev. B* **39**, 9929 (1989).
- <sup>16</sup>G. Bergmann, *Phys. Rep.* **107**, 1 (1984).
- <sup>17</sup>The analysis of the magnetoresistance measurements followed the approach described in detail by D. E. Beutler and N. Giordano, *Phys. Rev. B* **38**, 8 (1988). We also note that these measurements did not show any signs which would indicate clustering of the Fe [for a discussion of these signs see, for example, C. Van Haesendonck, J. Vranken, and Y. Bruynseraede, *Phys. Rev. Lett.* **58**, 1968 (1987)].
- <sup>18</sup>J. Liu and N. Giordano, *Phys. Rev. B* **39**, 9894 (1989).
- <sup>19</sup>In evaluating (1) we have assumed an electric field strength of 15 V/m and a phase coherence time  $\tau_\phi = 1.2 \times 10^{-10}$  s. The phase coherence time was obtained from the phase coherence length of 6000 Å, as measured from the weak-localization magnetoresistance, together with a diffusion constant of 30 cm<sup>2</sup>/s, as estimated from the measured resistivity at low temperatures and nearly-free-electron theory.
- <sup>20</sup>The phase breaking length for this sample was 3000 Å. Thus its size as compared to  $L_\phi$  was only slightly larger than that of the pure Au sample considered in Fig. 2, while its phase breaking time [see (1)] was slightly shorter.
- <sup>21</sup>See, for example, *Numerical Recipes*, edited by W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling (Cambridge University Press, Cambridge, 1986).
- <sup>22</sup>We should also note that studies of UCF in Au-Mn alloys with magnetic ion concentrations similar to ours [see A. Benoit, S. Washburn, C. P. Umbach, R. A. Webb, and D. Maily, in *Anderson Localization*, edited by T. Ando and H. Fukuyama (Springer-Verlag, Berlin, 1988), p. 346, and also Ref. 2] did not observe any unexpected variations of the conductance with field reversal.
- <sup>23</sup>A. D. Stone (private communication).
- <sup>24</sup>J. W. Loram, T. E. Whall, and P. J. Ford, *Phys. Rev. B* **2**, 857 (1970).
- <sup>25</sup>V. Cannella and J. A. Mydosh, *Phys. Rev. B* **6**, 4220 (1972).
- <sup>26</sup>A. Yu. Zyuzin and B. Z. Spivak, *Pis'ma Zh. Eksp. Teor. Fiz.* **43** 185 (1986) [*JETP Lett.* **43**, 234 (1986)].
- <sup>27</sup>N. Giordano (unpublished).