

Mesoscopic photovoltaic effect

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We have studied the response of mesoscopic conductors to microwave electric fields. This response is found to be nonlinear, with the generation of a dc voltage, V_{dc} , whose magnitude is proportional to the microwave power. This “photovoltaic” effect is sensitive to temperature, magnetic field, and the precise location of the scattering centers within the sample. Our results are generally in good agreement with recent theoretical predictions. However, the temperature dependence of V_{dc} is somewhat weaker than expected, and in some samples the behavior of V_{dc} in response to changes in the polarity of the magnetic field is not understood.

I. INTRODUCTION

Studies of small, so-called mesoscopic, conductors have revealed a variety of interesting properties.¹ For example, mesoscopic systems exhibit large fluctuations in their conductance; these are known as universal conductance fluctuations (UCF), and have been the subject of numerous theoretical and experimental studies in the past few years.^{2-4,1} An interesting feature of UCF is that the motion of only a single scattering center can lead to relatively large changes in the conductance.^{5,1} These fluctuations are sample specific, and reflect the detailed, microscopic arrangement of the disorder within the system.

It has recently been pointed out that the precise arrangement of the disorder will be of crucial importance for another property of mesoscopic systems. If a system has a center of inversion, symmetry does not allow certain types of nonlinear behavior. However, in a mesoscopic system the disorder will break inversion symmetry, leading to nonlinear effects that might otherwise be forbidden.⁶ While this is not a surprising point of principle, it turns out that these nonlinearities can be quite significant. In fact, recent experiments⁷⁻⁹ have observed some of these nonlinearities at low frequencies; i.e., with $\omega\tau_\phi < 1$, where τ_ϕ is the electron phase coherence time. In this paper we report the observation^{10,11} of nonlinear behavior in the high-frequency regime, $\omega\tau_\phi > 1$. The high-frequency behavior exhibits a number of interesting features, including a close connection to UCF. The nonlinearity we have studied has been termed a photovoltaic effect;^{12,13} an ac electric field produces a dc voltage, V_{dc} , in response. In our experiments we have investigated this effect in films of Ag, Sb, Au, and AuFe. We find that V_{dc} varies aperiodically with temperature and magnetic field. Our results also confirm that V_{dc} is sensitive to the precise locations of the scattering centers within the system; this is reflected in time-dependent fluctuations of V_{dc} that appear to be caused by the motion of individual scattering centers.

II. THEORY

The nature of the photovoltaic response is closely related to UCF, and can be understood from the following qualitative argument due to Fal’ko and Khmel’nitskii.^{12,13} A sample with conductivity σ and volume V in an ac electric field E_{ac} will absorb an energy $\sigma E_{ac}^2 V$ per unit time, corresponding to $\sigma E_{ac}^2 V/\hbar\omega$ photons. In the weak-field limit, an electron will absorb at most one photon, so this will also be the number of electrons that are excited by the ac field. These excited electrons will diffuse in random directions; because of the asymmetry induced by the disorder, different numbers will diffuse towards the two sides of the sample (we assume here a two lead arrangement), producing a dc voltage. The magnitude of this asymmetry will be of order the ratio of the universal conductance fluctuations, $\sim e^2/h$, to the total conductance. The electrons in question will be within an energy $\hbar\omega$ of the Fermi energy. From UCF theory, one knows that the motion of electrons within an energy range $E_c \sim \pi^2\hbar\tau_\phi^{-1}$ will be correlated, and hence will contribute dc voltages that are “in phase,” while the contributions from electrons whose energies differ by more than E_c will add incoherently. Putting all of these effects together, the total dc voltage in the high-frequency limit is predicted to be^{12,13}

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

where we have also included the numerical factors obtained from a quantitative calculation for two dimensions. Here $\zeta(x)$ 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 the electron phase coherence length, L_ϕ . Note that $\overline{V_{dc}}$ is the ensemble average, rms voltage. If one considers an ensemble of statistically similar, but microscopically distinct samples, any particular one may exhibit a value somewhat smaller or larger than this, and the sign of V_{dc} will also be random. One can effectively generate such an en-

semble by applying a magnetic field. This field will, if it is sufficiently large, shift the electron phases, thereby changing the diffusion asymmetry, and hence the value of V_{dc} . Varying the strength of an applied magnetic field should thus cause fluctuations of V_{dc} that are of order $\overline{V_{dc}}$ in (1).

Equation (1) is appropriate when L is less than or comparable to the phase coherence length, L_ϕ . When L_ϕ is smaller than the size of the system, which will tend to be the case at high temperatures, the sample will no longer behave as a single coherent region, as assumed in (1). Rather, it will behave approximately as a number of independent subsystems, each of size $L_\phi \times L_\phi$, with each subsystem exhibiting its own, independent dc voltage. In analogy with the behavior of UCF, subsystem averaging¹ will then yield a reduction of $\overline{V_{dc}}$ with decreasing L_ϕ ; i.e., an overall reduction of $\overline{V_{dc}}$ with increasing temperature. The precise temperature dependence depends on a number of factors; for our case, the theory predicts¹²

$$\overline{V_{dc}} \sim \tau_\phi^{3/2} T^{-1/2}, \quad (2)$$

hence the temperature dependence of $\overline{V_{dc}}$ depends on that of the phase coherence time, τ_ϕ . Since τ_ϕ generally varies as $\sim T^{-p}$ with $p \geq 1$ (in two dimensions), we would expect a fairly strong temperature dependence for $\overline{V_{dc}}$ when $L > L_\phi$.

III. EXPERIMENTAL SETUP

The samples were made from sputtered Ag, and evaporated Sb, Au, and AuFe films, which were patterned with optical lithography. Typical dimensions were¹⁴ $1 \mu\text{m} \times 1 \mu\text{m} \times 100 \text{ \AA}$, with sheet resistances of ~ 10 – 100Ω , and resistivities of 10 – $50 \mu\Omega \text{ cm}$. The sample was connected to much larger regions of the same film on two ends, and thus the measurements were performed in a two lead geometry. The experiments were conducted at temperatures as low as 1.4 K . Previous studies in our laboratory¹⁵ have found electron phase coherence lengths¹⁶ $L_\phi \sim 1 \mu\text{m}$, or larger (depending on the sheet resistance) for macroscopic films of these materials at 1.5 K . The apparatus was similar to that described previously.¹⁷ The sample was located in a microwave cavity, and frequencies near 8.4 and 30 GHz were employed. The microwave amplitude was modulated at 30 Hz , and the voltage produced at this frequency was detected with a lock-in amplifier. The absolute magnitude of the microwave field in the sample was accurately known from our previous work;¹⁷ this was, of course, essential for making a quantitative comparison with the theory.

Because of the modulation of the microwave source, the photovoltaic signal had a frequency of 30 Hz , but there was also some “pickup” in the electrical setup which gave a small signal at 30 Hz even in the absence of a photovoltaic effect. Despite efforts to reduce this pickup, it was generally not possible to completely eliminate it. However, this signal was usually small compared to V_{dc} (typically no more than 25%), was independent of temperature from 300 to 77 K , and was similar in magnitude

for both large and small samples. It was also proportional to E_{ac} , and hence could be readily distinguished from $\overline{V_{dc}}$, which varied as E_{ac}^2 [see Ref. 10 and Eq. (1)].¹⁸ In all of the measurements shown below, the magnitude of E_{ac} was kept fixed, and the contribution of pickup was measured at room temperature and subtracted off to obtain V_{dc} . 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 in the present experiments.

IV. RESULTS

Figure 1 shows typical results for V_{dc} as a function magnetic field for a Ag sample at several different temperatures. It is seen that V_{dc} is an aperiodic function of both T and H . So long as the sample was not “disturbed” (i.e., not exposed to switching transients, etc.), these results were completely reproducible. Hence, they closely resemble the behavior of UCF.¹ We should also note that, although it is not shown in Fig. 1, V_{dc} was here an even function of H ; i.e., $V_{dc}(+H) = V_{dc}(-H)$. We believe that this is to be expected for the following reason. As discussed above, V_{dc} arises from the difference in the number of excited electrons that diffuse towards the two leads, and this asymmetry is closely related to UCF. According to the theory¹⁹ we know that for a two lead arrangement, such as ours, the conductance is an even function of H . Hence, we expect that V_{dc} will also be an even function, as observed. While the results in Fig. 1 were obtained with a Ag sample, very similar results were obtained with Sb and Au samples.

The magnitude of $\overline{V_{dc}}$ at the lowest temperature in Fig. 1 was $\sim 4 \text{ nV}$. At this temperature $L_\phi \simeq 8000 \text{ \AA}$,

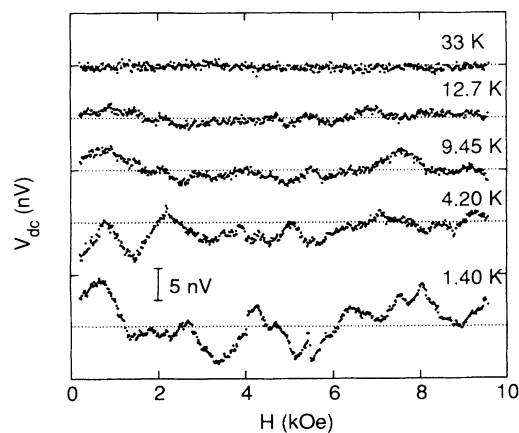


FIG. 1. V_{dc} as a function of H for a Ag sample at several temperatures, as indicated in the figure. The amplitude of the microwave field, E_{ac} , was 20 V/m . The sample was $1 \times 1 \mu\text{m}^2$, and had a sheet resistance of 8Ω . Note that the dotted lines are the horizontal axes ($V_{dc} = 0$) for the different temperatures.

hence under these conditions the sample was nearly a single coherent region, and (1) should apply. Evaluating (1) we find $(\overline{V_{dc}})_{\text{theor}} = 3 \text{ nV}$, in excellent agreement with the experimental value.

The overall magnitude of V_{dc} in Fig. 1 is seen to become smaller as T is increased, in agreement with the arguments given above. This is shown also in Fig. 2 where we plot the rms value $\overline{V_{dc}}$, as obtained from the data in Fig. 1 (plus additional data at other temperatures), as a function of T . The solid curve in Fig. 2 is a fit²⁰ to a power-law of the form $\overline{V_{dc}} \sim T^{-p_1}$, which yielded $p_1 = 0.6 \pm 0.1$. It is seen that while this power-law form provides a qualitative description of the data, there are deviations, especially at the highest temperatures. Nevertheless, from (2) we find $p_1 = 3p/2 + 1/2$, where p is the exponent that characterizes the temperature dependence of τ_ϕ . Since we expect^{21,22} $p \geq 1$, this yields $p_1 \geq 2$, which is much larger than found experimentally. That is, the observed temperature dependence is much weaker than predicted. As noted above, the data are not well described by a power law at high temperatures, so comparisons with the fitted value of p_1 must be viewed with caution. However, the predicted value of p_1 is so large that we would have expected the magnitude of $\overline{V_{dc}}$ to be completely negligible above 4 K, but this is certainly not found experimentally. The reason for this discrepancy is not understood, although it is possible that we are in some sort of crossover region that separates the low-temperature regime, in which $\overline{V_{dc}}$ should be temperature independent, from the high-temperature regime, where $\overline{V_{dc}}$ is predicted to be strongly temperature dependent.

The aperiodic fluctuations of V_{dc} observed in Fig. 1 can be characterized by an "average" period, or correlation field, H_c . We have derived H_c by Fourier analysis of the results for V_{dc} as a function of H . The Fourier transforms exhibit a peak that identifies H_c . The inset to Fig. 2 shows results for H_c as a function of T .

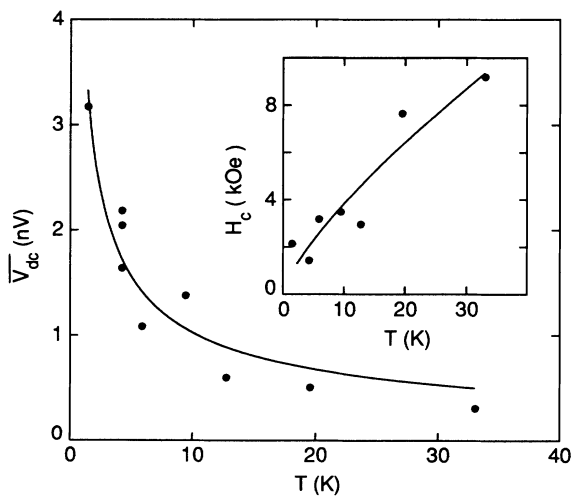


FIG. 2. $\overline{V_{dc}}$ (the rms value of V_{dc}) as a function of T for the sample considered in Fig. 1. Inset: H_c as a function of T . The solid lines show fits to power laws, as discussed in the text. The amplitude of the microwave field, E_{ac} , was 20 V/m.

According to the theory, and from analogous results for UCF, changing the flux through a phase coherent region by an amount of order a flux quantum, $\Phi_0 = h/e$, is equivalent to "changing" the system, hence we expect²³ $H_c \simeq \Phi_0/L_\phi^2$. The solid line in the inset to Fig. 2 is a fit to the form $H_c \sim T^{-p_2}$, which gave $p_2 = 0.75 \pm 0.25$. If $H_c \sim L_\phi^2$, we expect $p_2 = p \geq 1$. Our results are thus just consistent with the theory. We should also note that the absolute magnitude of H_c at the lowest temperature is in agreement with the (independently) measured magnitude of L_ϕ .

It is also interesting to consider how small changes in temperature affect V_{dc} . According to the theory, one would expect the "fingerprint" pattern $V_{dc}(H)$ to vary substantially for temperature changes, such that $k_B \Delta T = E_c \sim \pi^2 \hbar \tau_\phi^{-1}$. As discussed in connection with the theory (1), this is the energy scale over which phase coherence is maintained. For smaller temperature changes the patterns should be very similar, while for larger temperature variations $V_{dc}(H)$ should change completely. It is seen from Fig. 1 that the patterns $V_{dc}(H)$ at different temperatures are uncorrelated; this is expected since these temperatures are spaced more widely than ΔT . Figure 3 shows results for $V_{dc}(H)$ for a Ag sample, at two relatively nearby temperatures. Here we see that the two patterns are similar, although not quite identical. For example, they exhibit similar maxima at $\sim 5.5 \text{ kOe}$, but the minima near 7 kOe and the behavior below about 4 kOe differ somewhat. Given the known phase-breaking time for this sample we estimate $E_c/k_B \sim 0.8 \text{ K}$. The data sets in Fig. 3 were taken at temperatures spaced by almost precisely this amount, so according to the theory we would expect the two patterns $V_{dc}(H)$ to be somewhat (but not completely) similar, as is observed.

We noted above that V_{dc} was generally reproducible so long as the sample was not exposed to switching transients, etc. We also found that thermal cycling to room

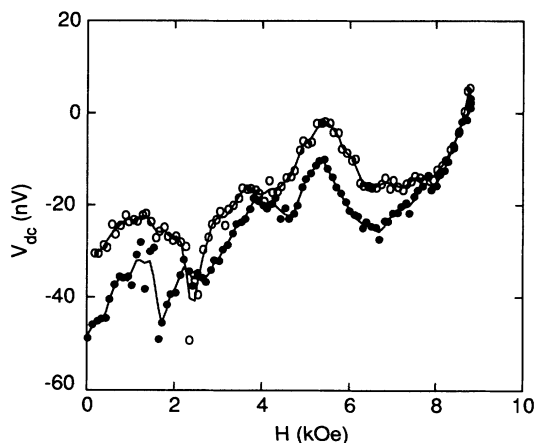


FIG. 3. Variation of V_{dc} with H for a Ag sample with dimensions $1.5 \times 1.5 \mu\text{m}^2$, and a sheet resistance of 50Ω . The open circles were obtained at 4.2 K, while the filled circles were obtained at 3.3 K. The microwave frequency was 8.4 GHz and the field strength was 20 V/m. The smooth curves are simply guides to the eye.

temperature and back resulted in completely different results for V_{dc} as a function of H , although \bar{V}_{dc} was unchanged. As in studies of UCF, this appears to be due to a rearrangement of scattering centers within the system. In a few samples, it was possible to observe this process take place directly at low temperatures. Figure 4 shows such results for V_{dc} as a function of time for a Ag sample. Abrupt, discontinuous changes in V_{dc} , i.e., switching events, are readily observed, and the behavior resembles so-called telegraph noise. We believe that these events are due to the back and forth motion of one, or a small number of scattering centers. Similar effects have been observed previously through measurements of time-dependent conductance fluctuations.^{24,5,1} An important difference is that here the fluctuations are as large as the "signal," while the fluctuations of the conductance are generally only a few tenths of a percent. In addition, the fluctuations in V_{dc} are easily resolved at 4.2 K, which is about an order of magnitude larger than the highest temperature at which the analogous conductance fluctuations are usually observed.^{24,5} Hence, measurements of V_{dc} might, in some cases, prove more useful in studies of impurity motion in metals.

In Fig. 1 we showed the behavior of V_{dc} as a function of H . It was noted that for those measurements $V_{dc}(+H) = V_{dc}(-H)$, and we argued that this is consistent with the behavior of UCF in a two lead measurement. This symmetry is illustrated explicitly in Fig. 5, which shows results for V_{dc} for a Sb sample at low fields, for both polarities of H . To within the experimental error, the behavior is independent of the sign of H .

While most samples exhibited behavior like that shown in Fig. 5, a few did not. Such a case is shown in Fig. 6, which gives results for a Ag sample. Here four field sweeps are shown; the directions in which the field was swept are shown by the arrows. Let us first concentrate on sweeps (a) and (b), in which the magnitude of the

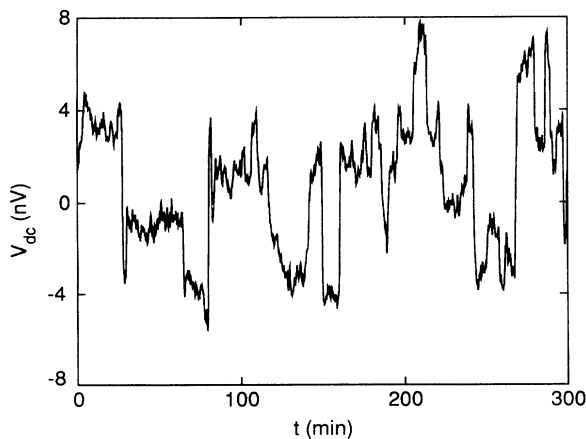


FIG. 4. V_{dc} as a function of time for a Ag sample with dimensions $2 \times 2 \mu\text{m}^2$, and a sheet resistance of 8Ω . The abrupt, essentially discontinuous changes of V_{dc} are believed to be due to the motion of single scattering centers. The amplitude of the microwave field, E_{ac} , was 20 V/m .

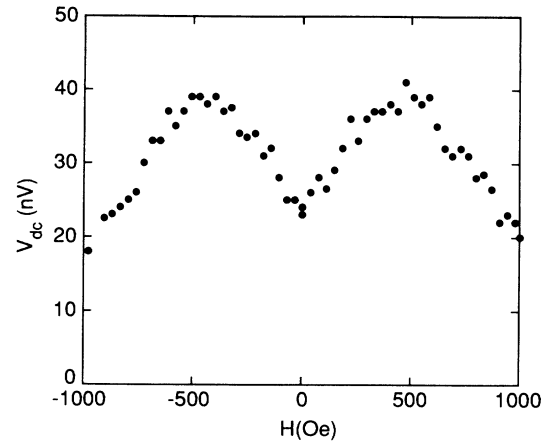


FIG. 5. V_{dc} as a function of H for a $2.0 \times 2.0 \mu\text{m}^2$ Sb sample with a sheet resistance of 100Ω . The microwave frequency was 8.4 GHz , the field strength was 20 V/m , and the temperature was 4.2 K .

field was swept from zero to larger values. Here we see that the behavior is approximately *antisymmetric*; i.e., $V_{dc}(+H) \sim -V_{dc}(-H)$. Sweeps (c) and (d) in Fig. 6 were obtained by sweeping the field towards zero from large values. Here again V_{dc} is approximately antisymmetric upon reversal of H . However, comparing sweeps (a) and (c) [or equivalently (b) and (d)] we also see that the value of V_{dc} depends on the direction in which the field is swept, or equivalently, the field history. Such hysteresis is clearly suggestive of magnetic effects. While this sample was composed of nominally pure Ag, the purity was only 99.999%, so we would not be surprised to find a few ppm of magnetic impurities. It was noted earlier that for two lead measurements of the conduc-

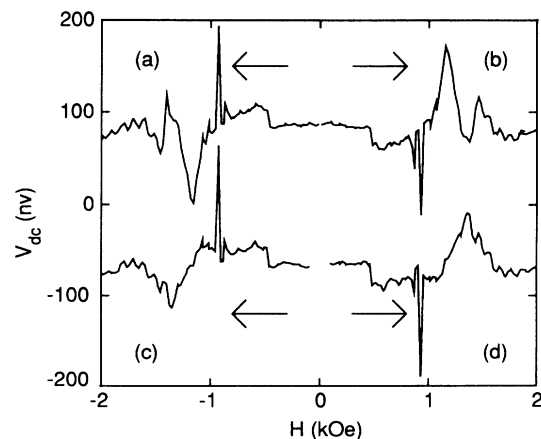


FIG. 6. V_{dc} as a function of H for a $1.5 \times 1.5 \mu\text{m}^2$ Ag sample with a sheet resistance of 40Ω . The microwave frequency was 8.4 GHz , field strength was 20 V/m , and the temperature was 4.2 K . The arrows indicate the direction in which the field was swept. The smooth curves are guides to the eye.

tance one expects $G(+H) = G(-H)$, but this is only true for nonmagnetic systems. More generally one has¹⁹ $G(+H, +M) = G(-H, -M)$. Thus, if there are magnetic spins in the sample, and they do not change direction

when H is reversed, then the conductance should change when the field is reversed. Given the close connection between UCF and the photovoltaic effect, we would expect similar behavior for V_{dc} .

While this argument seems to provide a qualitative explanation for the lack of symmetry of V_{dc} upon field reversal, it does *not* appear to explain why the behavior should be approximately *antisymmetric*. Nevertheless, we proceeded to investigate this problem further by studying samples of Au doped with Fe; typical concentrations were 0.1 at. % Fe. When compared with the nominally pure samples (i.e., as in Fig. 6), the AuFe samples had a greater tendency to display antisymmetric behavior, although they did not always do so. Some results for a AuFe sample are shown in Fig. 7. All of the data are shown in Fig. 7(a), and for clarity we have replotted portions of it in Figs. 7(b) and 7(c). In Fig. 7(b) we have plotted only the data for fields below 3 kOe, with the horizontal axis being $|H|$. Here it is seen that, aside from an approximately constant offset, the behavior of V_{dc} is very nearly antisymmetric. Figure 7(c) shows the data for fields larger than 3 kOe, again plotted as a function of $|H|$. In this region the behavior is symmetric, to within the experimental error. The results in Fig. 7 can be understood, at least qualitatively, if one assumes that the Fe spins are frozen in direction at low fields, but that in large fields they are "unlocked" and able to follow the direction of H . Such behavior would not be expected for isolated spins, but the Fe concentration in this sample was fairly high, so interactions between the spins, which would presumably be required for them to be frozen in direction in low fields, could well have been important. Moreover, the field at which the behavior changed from antisymmetric to symmetric [compare Figs. 7(b) and 7(c)] became smaller as the temperature was increased, which is consistent with this picture. It thus appears that the antisymmetric behavior found for V_{dc} , at least in AuFe, may be due to the presence of magnetic impurities. The cause in the other samples, i.e., Fig. 6, is not as clear, unless one assumes that their purity was somewhat less than originally believed.

V. CONCLUSIONS

In summary, we have observed nonlinear behavior in mesoscopic metal films exposed to a high-frequency electric field. The results are in good overall agreement with the theory of Fal'ko and Khmel'nitskii, although the temperature dependence is somewhat weaker than predicted by the theory and requires further study. In addition, in both nominally pure samples and in samples doped with magnetic impurities, V_{dc} exhibits some interesting features upon field reversal. We hope to examine these problems more in the near future.

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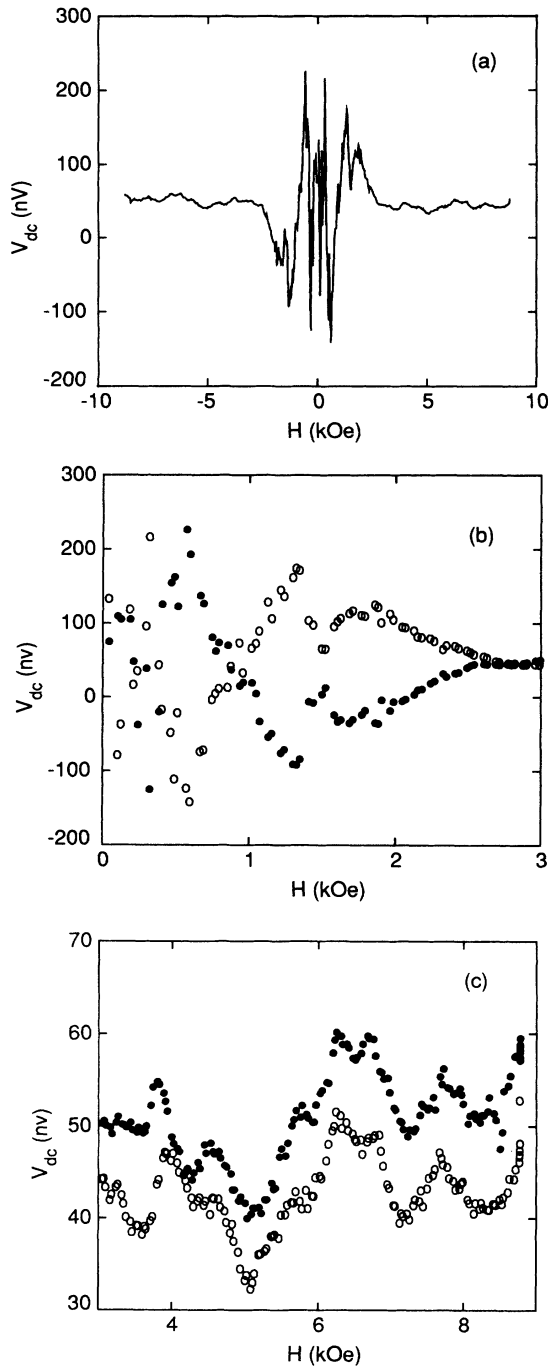


FIG. 7. V_{dc} as a function of H for a $1.0 \times 1.0 \mu\text{m}^2$ Au sample doped with 0.1 at. % Fe. The sheet resistance was 17Ω , the microwave frequency was 8.4 GHz, the field strength was 20 V/m, and the temperature was 1.4 K. (a) All of the data. (b) Expanded view of the data at low fields plotted as a function of $|H|$. The filled symbols are for $H > 0$ while the open symbols are for $H < 0$. (c) Data at high fields plotted as a function of $|H|$. The symbols have the same meaning as in (b).

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- ¹¹Experiments similar to ours have recently been reported by 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)]. These workers observed the photovoltaic effect in a semiconductor structure. However, the temperature dependence was not reported, and as we will see below from our results, this dependence is much weaker than predicted by the theory. In addition there were no observations of the switching events (as in our Fig. 3 below) which strongly imply that the impurity arrangement plays a crucial role in this effect.
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- ¹³V. I. Fal'ko, Europhys. Lett. **9**, 785 (1989).
- ¹⁴Similar results were obtained for sample sizes in the range $1 \times 1 \mu\text{m}^2$ to $10 \times 10 \mu\text{m}^2$. $\overline{V_{dc}}$ was found to become smaller as the sample size was increased, as predicted by the theory. For the samples considered in Figs. 1-3, $D \sim 25 \text{ cm}^2/\text{s}$ and at 1.5 K $\hbar\omega/k_B T \sim 3$ while $\omega\tau_\phi \sim 50$.
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- ²¹At low temperatures, typically below 4 K for films like ours, one expects $p = 1$, corresponding to electron-electron scattering in two dimensions (Ref. 22), and this is in good agreement with the behavior found previously for similar films (Ref. 15). At higher temperatures electron-phonon scattering becomes important, and leads to a larger value of p .
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- ²³At low temperatures, the size of a phase coherent region cannot become larger than the size of the system, so one would expect H_c to be given by an expression of the form $H_c = \Phi_0/L_{\text{eff}}^2$, with $L_{\text{eff}}^{-2} = L_\phi^{-2} + L^{-2}$. However, in our case $L^{-2} < L_\phi^{-2}$, even at our lowest temperatures, so it is safe to just use L_ϕ in place of L_{eff} .
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