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Photovoltaic effect in small superconducting–normal-metal systems

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We have observed an unusual photovoltaic effect in small metallic systems in which some portion of the sample is superconducting. In these systems, an applied microwave field can induce a dc voltage (the ‘‘photovoltage’’), V_{dc}. We have found that this voltage can be an antisymmetric function of magnetic field, i.e., $V_{\mathrm{dc}}(+H)=-V_{\mathrm{dc}}(-H)$. It also exhibits aperiodic fluctuations as a function of both H, and the strength of the microwave field. Results for several different sample geometries suggest that it is due to the inverse Josephson effect, although the samples are not obviously reminiscent of weak link structures. [S0163-1829(96)01329-X]

I. INTRODUCTION AND BACKGROUND

Several recent experiments in our laboratory have been devoted to the photovoltaic (PV) effect in mesoscopic systems.[1–3] The application of a microwave field to such a system results in an induced dc voltage, V_{dc}, even when there is no current (dc or ac) applied via external leads. The mesoscopic PV effect arises from the broken inversion symmetry inherent in a disordered system, and has much in common with universal conductance fluctuations (UCF's).[4–6] These are fluctuations in the conductance of a mesoscopic metallic structure due to small changes in the sample. These changes can be small variations in an applied magnetic field, or the motion of individual electron scattering centers. In either case, a fluctuation in the conductance of order e^2/h results. In accord with this analogy, we have observed fluctuations in V_{dc} (including Aharonov-Bohm oscillations) with mesoscopic ring samples in response to variations in magnetic field.[7]

During the course of this work, several samples were found to exhibit an unexpected dependence of V_{dc} on magnetic field, H, which we attributed to the presence of magnetic impurities.[2,3] In this paper we describe the results of additional studies of such anomalous samples, and show that contrary to our initial suspicions their behavior was *not* due to magnetic impurities. Results for different sample geometries suggest instead that the behavior was due to the inverse Josephson effect in Josephson junctions formed inadvertently. It is certainly not surprising that the combination of microwave fields and Josephson junctions can give rise to dc voltages; this is well known from work on tunnel junctions and other types of superconducting weak links. However, in our samples the geometry is somewhat different from that of conventional weak links, and the precise nature of our junctions is not obvious. Nevertheless, the apparent simplicity of our sample geometry may prove useful for further studies of such effects.

II. EXPERIMENTAL METHOD

In this paper we present detailed results for two samples. The first was similar in all respects to those reported on previously.[1–3] It was made from a 150 Å thick evaporated Au film, which had a sheet resistance of ~2.5 Ω. It was patterned using photolithography into a microbridge geometry with a small, roughly square region, $\sim 1\times 1$ $\mu\mathrm{m}^2$ in size, which was continuous with much wider regions of the film, which acted as contact films. This sample was similar to those we have used in previous studies of the mesoscopic PV effect.[1–3] We will see below that the key feature of this sample was that the external leads were attached to the contact films using In-Sn solder [see Fig. 1(a) for a schematic], which is superconducting below about 6 K. We will refer to this sample as the ‘‘slug’’ sample, since, as will become clear shortly, it seems to have some properties in common with the Josephson devices of the same name described by Clarke.[8] This is the *only* sample considered below which had leads attached with superconducting solder; the others all had leads attached with Ag paint (which was nonsuperconducting).

The second sample was also patterned from a Au film into a $\sim 2\times 2$ $\mu\mathrm{m}^2$ microbridge geometry similar to that described above. In the center of the bridge region a dot of In which was ~2 μm diameter and 500 Å thick was deposited[9] on top of the Au; i.e., it completely covered the Au in the vicinity of the microbridge, as illustrated in Fig. 1(b). The resistivity of the In at 4.2 K was ~10 μΩ cm, implying that it was a continuous film (i.e., not granular). This sample will be referred to as the ‘‘In-dot’’ sample. We have also studied a number of other samples with geometries related to the In-dot sample, including cases in which the In dot was off center; i.e., on top of the Au film but positioned several (up to 10 or more) μm away from the bridge region. Results for these cases will be mentioned as appropriate.

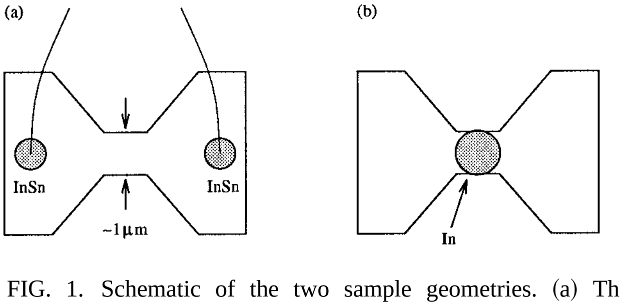

FIG. 1. Schematic of the two sample geometries. (a) The ‘‘slug’’ geometry in which contact is made to the Au film by two InSn solder pads. (b) The In-dot geometry in which a small In dot is deposited on top of the Au film. In these samples the contact was made via (nonsuperconducting) silver paint.

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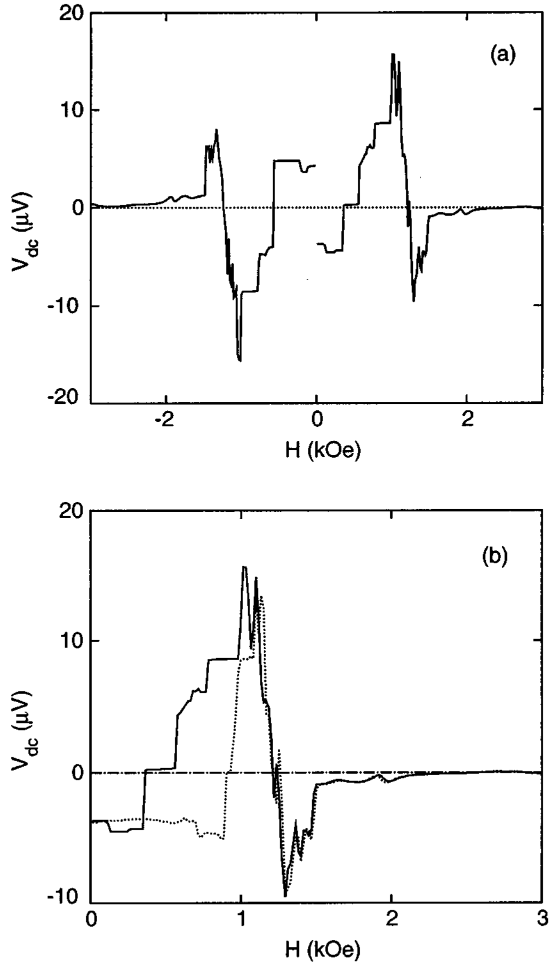

FIG. 2. V_{dc} vs H for the slug sample at 1.4 K. The microwave electric field was ≈ 4 V/m. (a) Results obtained by sweeping H toward zero from large positive and large negative values. (b) Results for just positive polarities, but with different sweep directions. The solid curve was obtained by sweeping H down from large positive values, while the dotted curve was measured while sweeping H up toward large positive values.

The samples were mounted in a microwave cavity which had a resonant frequency of 8.4 GHz for its 210 mode. They were located at a maximum of the electric field, with this field directed in the plane of the film. The cavity was inside a vacuum can which was usually filled with liquid He to minimize Joule heating of the sample by the microwave field (such heating was believed to be unimportant for all of the data shown in this paper). This was all positioned inside a superconducting solenoid which provided a magnetic field perpendicular to the plane of the film. The microwave field was modulated at 150 Hz, and the sample voltage measured with a lock-in amplifier using a transformer coupled preamplifier. This scheme allowed us to conveniently measure the very small signals which were encountered in the mesoscopic PV effect. We will refer to the voltage measured in this way as V_{dc} even though it was not a strictly dc measurement.

III. RESULTS

Figure 2 shows some typical results for the photovoltaic signal, V_{dc}, as a function of magnetic field at 1.4 K for the slug sample. In Fig. 2(a) we show the results for both polarities of H; in each case the field was swept toward zero, and it is seen that the signal was an approximately *antisymmetric* function of H; i.e., $V_{dc}(+H) \approx -V_{dc}(-H)$. The two curves are not continuous at $H=0$ as the signal was hysteretic, as shown in Fig. 2(b), which shows results for increasing and decreasing field sweeps, both with a positive field polarity. This behavior is similar as that reported for the anomalous samples in Refs. 2 and 3 (see Figs. 3, 4, and 5 in Ref. 3). We also note that this PV signal is approximately 3 orders of magnitude larger than that expected,[4,5] and observed,[1,2] for the mesoscopic PV effect (see also below), so it seems clear that some other physics is responsible. Several features of the results in Fig. 2 suggest that superconductivity was involved. First, the PV signal was reduced to the level expected for the mesoscopic PV effect (a few nV) at large fields; in Fig. 2 we see that this reduction occurred above a few kOe, and it is natural to associate this with the critical field of a superconductor. Second, the signal sometimes exhibited an approximately ‘‘stepwise’’ structure; i.e., there were plateaus or ranges of H over which it was approximately constant. Several such plateaus are seen below about 1kOe in Fig. 2. Such quantized voltages remind one immediately of the ac Josephson effect. Third, we also studied samples which were essentially identical to the slug sample, except that the contact leads were attached with Ag paint, so that they contained no superconducting regions of any kind. These nonsuperconducting samples *never* exhibited the giant PV signals, hysteresis, or antisymmetric behavior noted above.

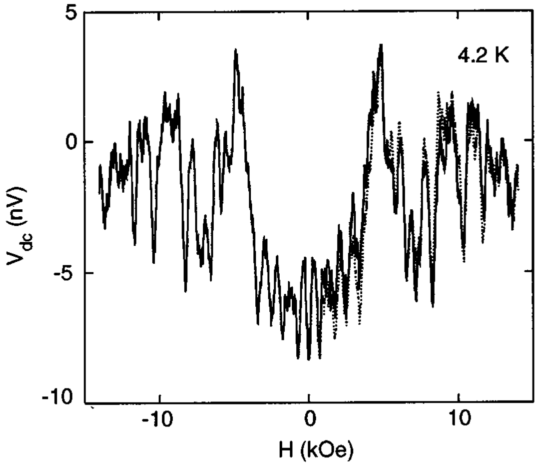

FIG. 3. V_{dc} vs H at 4.2 K for the In-dot sample. The microwave field was ≈ 4 V/m. These results were independent of the direction of the field sweep. The dotted curve shows the data for $H<0$ plotted as a function of $|H|$, to illustrate the reproducibility of the results.

Since the results for the slug sample strongly implied that superconductivity was connected with the behavior in Fig. 2, we studied the In-dot sample, along with a number of similar samples. They all had nonsuperconducting leads and contacts, with the only superconductor present being a small ‘‘dot’’ of In, typically a few μm in size, near the location of the Au microjunction. The intent was to make the geometry of the superconducting region as simple as possible. We used In for the superconductor, since it had a much sharper transition than the In-Sn solder in the slug sample. Some PV

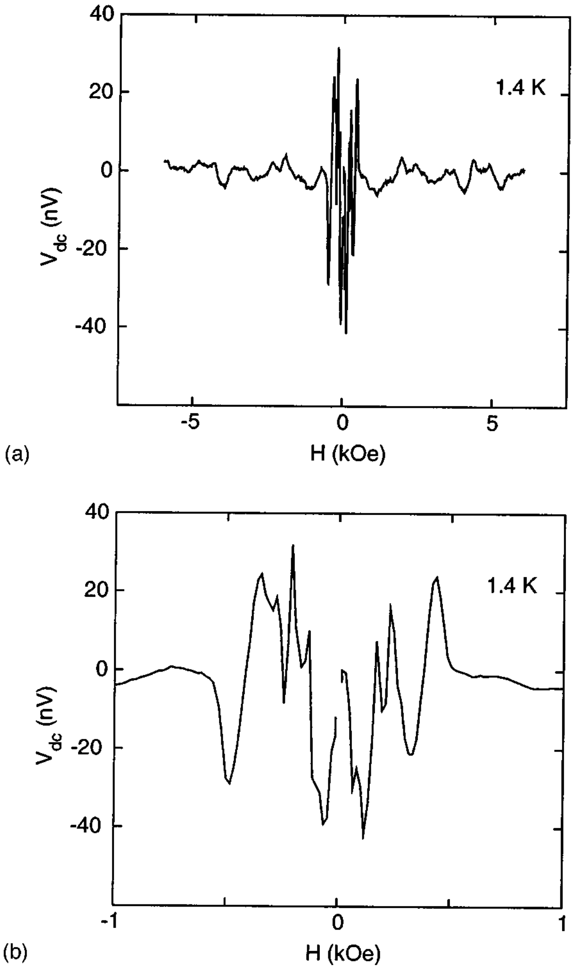

FIG. 4. V_{dc} vs H at 1.4 K for the In-dot sample, at the same microwave intensity as in Fig. 3. (a) shows all of the data, while (b) gives an expanded view of the behavior at low fields.

results for the In-dot sample described in Sec. II are shown in Figs. 3 and 4. At 4.2 K the PV signal, Fig. 3, was of order a few nV. These aperiodic fluctuations as a function of H are in good agreement with the theory of the mesoscopic PV effect,[4,5] and have an origin similar to that of universal conductance fluctuations.[6] Within the experimental noise level, this mesoscopic PV effect was a symmetric function of field, $V_{dc}(+H) \approx V_{dc}(-H)$ and did not exhibit any hysteresis. Similar behavior was found at other temperatures above the T_c of the In. The results in Fig. 3 also illustrate the reproducibility of the behavior. If the sample was maintained at low temperatures, field scans performed several hours apart were generally reproducible to nearly the thickness of the curves in Fig. 3. Thermal cycling to 77 K often produced quantitative changes in the aperiodic pattern. This was expected, since, as in the case of universal conductance fluctuations, this PV ''magnetofingerprint'' reflects the detailed spatial arrangement of the scattering centers, which is known to change with thermal cycling.[6]

In Fig. 4 we show PV results for the same In-dot sample at 1.4 K, i.e., below the T_c of the In. At low magnetic fields the PV signal was nearly an order of magnitude larger than found at 4.2 K, and V_{dc} was approximately (but not exactly) antisymmetric, in sharp contrast to the symmetric behavior seen at 4.2 K. Above about 500 Oe the magnitude of the PV signal dropped abruptly to a level similar to that seen at 4.2 K, and in this high field regime the signal was again a symmetric function of H. Although we do not show the data here, we note that V_{dc} exhibited hysteresis in low fields, but not at high fields.

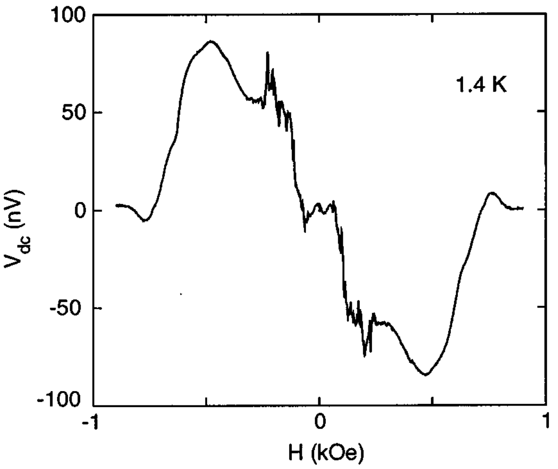

FIG. 5. V_{dc} vs H at 1.4 K for the In-dot sample, at a microwave field of ≈ 2 V/m, illustrating the antisymmetric behavior at low fields.

From the results for the In-dot sample it seems clear that there were two separate contributions to the PV signal. One was a symmetric function of field, had a magnitude of a few nV, persists to high fields, and was present at both 4.2 and 1.4 K. We have already identified this component as the mesoscopic PV effect. The other contribution was an approximately antisymmetric function of H, was much larger than the mesoscopic PV signal, and was quenched in high fields. Data at other temperatures showed that with this sample, the antisymmetric PV signal was only found below the T_c of the In. This antisymmetric component thus had all of the properties of the PV signal found at low fields in the slug sample.

V_{dc} was also a strong function of microwave power. Results for the In-dot sample at a second value of the microwave field are shown in Fig. 5. Here the antisymmetric component was larger than in Fig. 4, and the ''degree'' of antisymmetry is even clearer. The PV signal as a function of the microwave field is shown in Fig. 6. Again we find aperiodic fluctuations of V_{dc}; we are certainly not in any sort of ''linear response'' regime.

The unusual dependence on microwave field is highlighted further in Fig. 7, which shows the magnetic field dependence of V_{dc} for the In-dot sample at a smaller value of the microwave field than in Fig. 5. Here V_{dc} increased by 2 orders of magnitude, and also exhibited quantized voltages. This is similar to the behavior exhibited by the slug sample (Fig. 2), and is again reminiscent of the inverse Josephson effect.

Other samples, with geometries similar to that of the In-dot sample, exhibited similar behavior, even when the In was located well away (~ 10 μm) from the microbridge region. It was thus not necessary that the dot be located precisely at the center of the junction.

All of the evidence suggests that the antisymmetric PV signals displayed by the In-dot samples and the slug sample

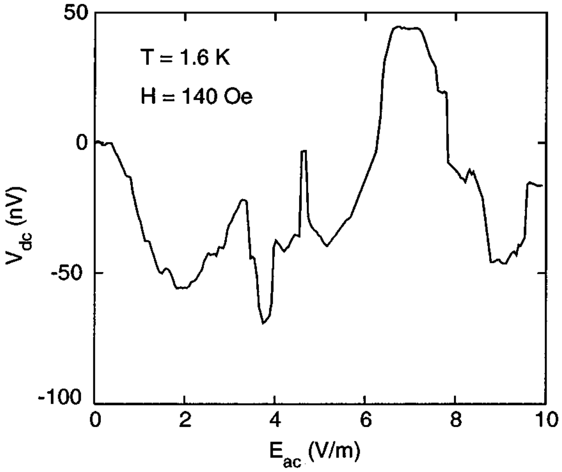

FIG. 6. V_{dc} as a function of the magnitude of the microwave electric field, E_{ac}, at 1.6 K and 160 Oe for the In-dot sample.

have a common origin associated with the superconducting regions. We believe that it was due to the inverse ac Josephson effect, and will give our arguments for this conclusion in the next section.

IV. DISCUSSION

In view of the systematics of the antisymmetric PV signal, and the clear connection with superconductivity, we believe that it arises from the inverse Josephson effect. In measurements of the usual ac Josephson effect, the current-voltage (I-V) characteristic of a Josephson junction is monitored in the presence of microwave radiation of frequency ω. This characteristic is generally found to exhibit regions of constant I, Shapiro steps, at values of V corresponding to the Josephson relation

$$V = n\hbar\omega/2e, \tag{1}$$

where n is an integer (or, in some cases, the ratio of two integers). The inverse Josephson effect is observed in junctions which are unbiased; i.e., for which there is no applied dc current. In this case the voltage across the junction is simply measured in the presence of microwave radiation. The inverse effect appears to have been first observed by Langenberg and co-workers in tunnel junctions,[10] and has since been discussed by a number of other workers.[11,12] This effect has generally been studied in systems containing conventional ''well-defined'' Josephson junctions. However, it has also been reported in systems such as packed powders of Al or Nb particles,[13,14] and more recently in high-T_c powder samples,[15,16] in which the junctions are presumed formed at the contacts between superconducting grains.

The theory of the inverse Josephson effect seems to be much less well developed than the theory for the usual biased junction case. Nevertheless, experiments and theory give the following picture. First, the inverse effect does not always exhibit voltages given by (1). The voltage can be quantized according to (1) or unquantized, depending on the ''quality'' of the junction, and the magnitude of the microwave power and magnetic field. Generally, high quality junctions show quantized voltages while low quality ones, such as found in packed powders or the high-T_c samples, do not (note, however, that unquantized behavior is often found in tunnel junctions, which are usually considered to be more ''ideal'' Josephson junctions than other types of weak links). Second, in the case of unquantized voltages, the voltage changes sign when the polarity of the magnetic field is reversed.[10] Third, when the voltage is unquantized, it is an aperiodically fluctuating function of the microwave power.[10–12]

These are precisely the properties of the PV signal we have observed. Our results correspond well to those found for ''low quality'' junctions, as V_{dc} often did not obey the quantization condition (1) (the Josephson voltage corresponding to 8.4 GHz with $n=1$ is 17.4 μV). The behavior as a function of both magnetic field and microwave power are similar to that reported in Refs. 10 and 11 and 12. Given the strong similarity of our results to those reported in other Josephson systems (such as for the ''low quality'' junction in Fig. 6 of Ref. 11) we believe that the evidence is very strong that our PV signal is due to the inverse Josephson effect.

However, this interpretation raises an interesting question. Namely, precisely where are the Josephson junctions in our samples? In our slug type samples it seems plausible that the junctions were formed at an oxide layer where the In-Sn solder made contact to the Au films, in analogy with the Clarke slug devices.[8] It would not be surprising for these structures to contain several such weak link regions. These junctions would certainly not be the same from sample to sample; indeed, one could easily imagine that some samples with superconducting solder contacts may not have any such junctions. This would explain why many of the samples studied previously[1,2] did not exhibit the anomalous PV behavior we have described. However, in our In-dot samples the voltage was measured across opposite ends of a (normal) Au film; the In, which was continuous, was apparently in contact with the Au film only in one singly connected region. This is very different from the usual superconducting-normal-superconducting weak links in which the Josephson

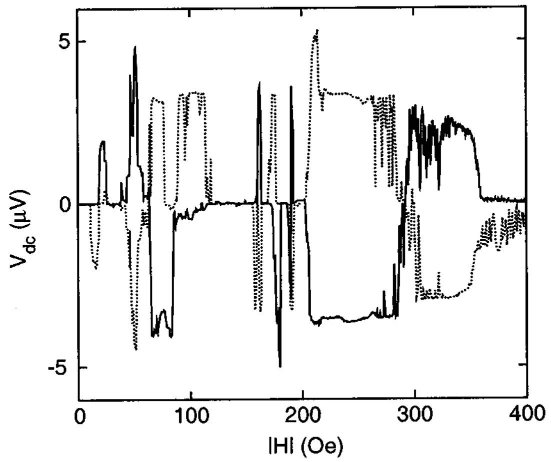

FIG. 7. V_{dc} as a function of the magnitude of the magnetic field at 1.4 K for the In-dot sample. The microwave field was ≈2 V/m. To illustrate the approximately antisymmetric behavior, we have plotted the results as a function of $|H|$; the solid curve shows results for $H>0$ while the dotted curve is for $H<0$.

effect is well known to occur (and from the packed powder samples). In particular, our In-dot samples do not appear to possess two ''weakly coupled'' superconducting regions, which is thought to be a requirement for the realization of a Josephson junction. It is thus not obvious to us exactly where the junctions are located in the In-dot type samples. While we cannot rule out the possibility of inadvertently formed Josephson junctions within the In dot, we do not, at present, understand how a superconducting-normal-superconducting weak link can be formed from a single continuous In dot on top of a Au film. One possibility that has been mentioned concerns the interdiffusion of Au and In. The presence of such an intermediate region would certainly complicate matters, and may lead to junction formation, although we do not at present understand how this could occur. One might also imagine that some sort of proximity induced Josephson effect[17] could play a role. That is, the In could induce superconductivity in the Au underlayer, and a junction could then be formed between these two superconductors. However, precisely how (or if) this could occur is not clear to us.

On the other hand, the geometry of our In-dot samples is extremely simple. In fact, it is hard to imagine a simpler system, since it would appear to contain only a single superconductor–normal-metal interface, whose dimensions and other properties could, with some additional effort, be precisely controlled and characterized. Indeed, one might even hope that the In-dot geometry would provide a ''model'' system in which to study the inverse Josephson effect. Such a model system would be very useful, since the difference in the behaviors of high and low quality junctions, while evident from the experimental properties, does not appear to be well understood in terms of the basic junction properties. However, our understanding, or rather lack of it, concerning the In-dot geometry suggests that it is premature to employ it as such a model system.

In summary, we have studied photovoltaic effects in a variety of samples containing superconducting regions, and have observed behavior indicative of the inverse Josephson effect. Our In-dot sample geometry may prove useful for further experiments in this area.

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