Quantum Interference Effects in Lithium Ring Arrays

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We report detailed measurements on the weak-localization corrections in well-defined and -characterized arrays of quasi one-dimensional normal-metal rings. The Bohm-Aharonov oscillations are very well resolved. Measurements on sets of samples of varying ring size and geometry determine the distinct effects of these parameters and allow an unambiguous, quantitative comparison to the weak localization theory for ring geometries.

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Al'tshuler, Aronov, and Spivak[1,2] (AAS) predicted that the weak-localization corrections to the resistivity of an isolated metal ring produce a resistance component oscillatory in the magnetic flux through the ring. The effect has been observed in experiments on thin-film cylinders by Sharvin *et al.*[2] and then others[3] in planar ring arrays,[4,5] and has had further theoretical substantiation,[6,7] including comprehensive calculations of the special properties of arrays by Doucot and Rammal.[8] Many of the general properties have been observed in the first experiments on individual arrays by Pannetier *et al.*[4] Here we describe the properties of sets of array samples which individually show these general properties more clearly and completely than the earlier work and together show the systematics of the geometry dependence of the arrays. Further, they are composed of quasi one-dimensional wires whose properties are independently determined; the oscillatory behavior is particularly well resolved and is measured over a wide range of temperature and magnetic field. Therefore, an absolute and quantitative comparison to the existing theories can be made.

The weak-localization corrections arise from the interference between a backscattered electronic state and its time-reversed state, so that the interference amplitude is periodic in the two-electron quantum of magnetic flux $\Phi_0 = hc/2e$. The processes are distinct from the recently discovered hc/e effects observable only in isolated rings,[9] although many of the experimental considerations and some of our general conclusions apply to both problems. The size of the interference effects depends on the range of coherence of the diffusing electrons and is limited by temperature-dependent inelastic scattering processes, the dephasing of electrons in different trajectories by a magnetic field, and spin-flip and spin-orbit scattering by impurities. It is convenient to consider paired electron states to define an effective range of coherence for the electrons. Then the ranges for the triplet of spin-1 states and the single spin-0 state are respectively[10-13]

$$L_1(T,H) = [L_i(T)^{-2} + \tfrac{4}{3}L_{s.o.}^{-2} + \tfrac{2}{3}L_s^{-2} + L_H^{-2}]^{-1/2}, \quad L_0(T,H) = [L_i(T)^{-2} + 2L_s^{-2} + L_H^{-2}]^{-1/2}. \tag{1}$$

$L_i(T) = [D\tau_i(T)]^{1/2}$ is the diffusion distance between inelastic scattering events; diffusion distances for spin-flip and spin-orbit scattering are defined analogously. The magnetic length for a quasi one-dimensional sample is $L_H \equiv \sqrt{3}\Phi_0/\pi HW$ for a "wire" of dimension W transverse to the field. Quasi one dimensionality occurs when all of the diffusion lengths exceed the wire width and thickness. Then the backscatter interference corrections to resistance may be expressed as the small fraction change of the ordinary resistance:

$$\frac{\Delta R}{R_0} = \frac{e^2}{\pi\hbar}\frac{R_\square}{W}\{\tfrac{3}{2}L_1F(L_1) - \tfrac{1}{2}L_0F(L_0)\} \tag{2}$$

if the "wires" are thin films of sheet resistance $R_\square$. The result of AAS is for a wire turned into itself to form an isolated ring. For a square "ring" of side S,

$$F(L) = \frac{\sinh(4S/L)}{\cosh(4S/L) - \cos(2\pi\Phi/\Phi_0)} = 1 + 2\sum_{n=1}^{\infty} e^{-n(4S/L)}\cos\left(\frac{2\pi n\Phi}{\Phi_0}\right). \tag{3}$$

Here $\Phi = HS^2$ and the functional dependences on T and H are suppressed. The series form is always rapidly convergent and invites the appealing interpretation of the sum terms as corresponding to n-fold circulations of the unit cell by the electrons. It also is apparent that the wire magnetoresistance can be viewed as an envelope (defined, for example, by the points for $\Phi = \Phi_0/2$) on which the oscillations are superimposed. One of the simpler arrays con-

sidered by Doucot and Rammal is a string or "necklace" of rings:

$$F(L)=\frac{(2S/L)\cosh(2S/L)-\sinh(2S/L)}{2(2S/L)\sinh(2S/L)}+\frac{(1/2)\sinh(2S/L)}{[\cosh^2(2S/L)-\cos^2(\pi\Phi/\Phi_0)]^{1/2}}. \quad (4)$$

Doucot and Rammal have also calculated $F(L)$ for other kinds of array including a square grid or mesh although no closed form is obtained in the other cases. Despite the rather different forms, Eqs. (3) and (4) have many of the same qualitative features. But the size of the oscillations and their envelope should be reduced in the arrays. The difference in the envelopes vanishes at small L/S, and all results reduce satisfyingly to that of a simple wire: $F(L)=1$. But even in this limit the array predicts a reduction in the periodic component [the $\cos(2\pi\Phi/\Phi_0)$ term] of $\frac{1}{4}$ for a necklace and $\frac{1}{8}$ for a square mesh. Because of their complexity, either of the forms may be used to produce reasonable fits to experimental data on a single necklace sample, for example, if the several diffusion lengths are used as free parameters. One of the special, and demonstrated,[11,12,14] properties of one-dimensional systems is that the magnetoresistance of a simple wire determines the absolute values of the diffusion lengths. By applying Eq. (2) to analyze $R(T,H)$ for a wire sample [$S\equiv\infty$, $F(L)=1$] in this experiment we measure the values of the diffusion lengths so that all the parameters relevant to the weak-localization phenomena are known.

Our samples were nominally pure lithium films evaporated at low temperature. Details of their preparation and measurement appear elsewhere.[5,12] Lithium, the lightest metal, is chosen for its small intrinsic spin-orbit scattering; the spin-dependent scattering actually observed is small and varies $\approx 100\%$ from run to run. The films had thickness $d=21\pm1$ nm, and $W=55\pm7$ nm with the spread representing the differences between samples. For each individual sample the width was defined to ≈ 2 nm. W was less than all the characteristic lengths in the problem, so that the wires were quasi one dimensional with $R_\square/W=31.5\pm1$ Ω/μm. These values correspond to film sheet resistance $R_\square=1.6$ Ω, or resistivity $\rho=3.5$ $\mu\Omega$-cm.

Figure 1 includes sketches of the sample geometries studied. The control consists of long thin film strips. "Necklace" samples consist of square rings strung end to end as shown and a "mesh" is a two-dimensional square array. The average ring edge dimensions were $S=0.50$, 0.71, and 1.00 μm. Each sample had a total length of 1 mm. The necklace and control samples were composed of 25-μm ($>> L_1, L_0$) segments arranged in series and in parallel to produce individual samples with a few kilohms in resistance. $R(T,H)$ for the control sample gave the following values for the diffusion distances controlling Eq. (2): $L_i(T)=(1.85\pm0.1$ μm-K$)T^{-1}$; $L_{s.o.}=2.3\pm0.2$ μm; $L_S=3.1\pm0.2$ μm. To show the dramatic effects of relatively strong spin-orbit scattering, we will also show data from another set of otherwise similar samples with smaller $L_{s.o.}$.

Figure 1 shows the magnetoresistance for six samples at $T=0.13$ K. At this low temperature the coherence ranges are limited primarily by spin-dependent scattering but are still large: $L_1=1.8$ μm and $L_0=2.23$ μm, lengths comparable to the circumference of the smallest rings. The curve for the control sample (top) is characteristic of the one-dimensional localization phenomenon; below it are the curves for three necklaces and two meshes. The ring-array specimens show oscillations superimposed on an envelope decreasing with H. The envelopes represent the coherent effects in the wires comprising the rings and should be compared to the control curve. The oscillations represent the extra effects of electrons traveling the circumference of a ring or rings coherently. The oscillations vanish at high temperatures or fields [$F(L)\rightarrow 1$] and all the envelopes coincide as they should. Because the curves do not overlap at low temperatures, it is immediately clear that Eq. (3) cannot describe all of the phenomena. However, the important general features it shares with the array results are manifested. The oscillation amplitude falls off with

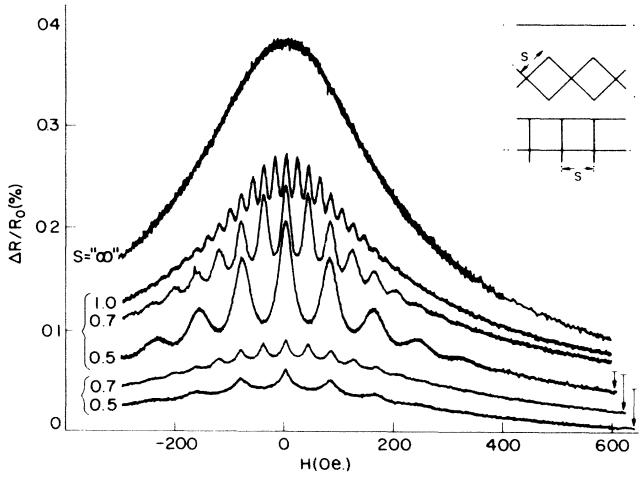

FIG. 1. The magnetoresistance $R(T,H)$ for $T=0.13$ K for the wire control sample (top), three necklace arrays (next three curves), and two meshes (bottom two curves). The upper-right-hand sketches define the control, necklace, and mesh geometry. The size, S, of the unit-cell side is indicated next to each curve. Some of the curves have been displaced vertically for clarity.

temperature (Fig. 2), field, and cell size in ways which may be roughly mapped onto each other in accordance with the symmetries of Eqs. (1)–(4). The oscillations have period Φ_0/S^2 within $\approx 1\%$; this shows that it is the *average* cell area which determines the period.[6] At high temperatures or fields the oscillations are smooth and sinusoidal, but at low temperatures and fields the "harmonic" structure represented by the higher-order terms in the summation in Eq. (3) cause a sharpness in the peaks which becomes more marked for smaller S and for the meshes relative to the necklaces. The nth-order terms may be interpreted as representing n-fold circulations of electrons around a loop or combination of loops with consequent n-fold coupling to the loop flux. The increased sharpness of the oscillations for the mesh, which has more such combinations, shows that these extra closed paths contribute significantly. This difference in behavior and the apparent suppression of the oscillations and their envelope occur because the electron coherence length is so long at this low temperature as to allow a significant interaction between the array elements.

One quantitative test of the theory is presented in Fig. 2. The low-field amplitude of the oscillating resistance component is plotted vs T for the three necklaces and for the mesh with $S = 0.71$ μm (dashed curve). The resistance change for the simple wire and the theoretical curve for this case also appear near the top of the figure. For simplicity, theoretical results for the oscillation amplitude are shown for $S = 0.71$ μm only. At high temperatures, the necklace and mesh data approach each other and the single-ring theory (solid curve A). At low temperature, the single-ring theory seriously overestimates the oscillation amplitude and, of course, does not predict the difference between the two arrays. The array theory for the necklace (curve B) and for the mesh (curve C) agrees with the respective data at low temperatures but falls factors ~ 4 and ~ 8 below the data at high temperatures. Because of the strongly exponential nature of the oscillation amplitude at small L, the disagreement is in a sense minor. An increase of the diffusion lengths by only 15% brings the necklace data into agreement for all T for all of the necklace samples. However, this is not justified by our measurements and to achieve the same agreement for the mesh data would require a larger (25%) "adjustment."

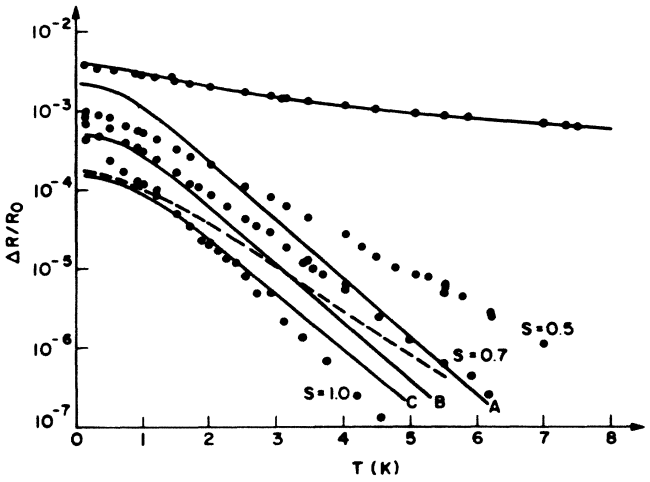

FIG. 2. The temperature dependence of the oscillating component of resistance for the arrays. For perspective, we show, near the top of the figure, data and the weak-localization theory for the total magnetoresistance for the simple wire sample used as a control. The oscillation amplitude for the three necklaces of cell size S appear as dots; the data for the mesh with $S = 0.7$ μm is represented as a dashed curve. The nearby solid curves correspond to $S = 0.7$ μm and are the predictions of the single-ring calculation (curve A) and of the array calculations for a necklace (curve B) and a mesh (curve C). The nominal errors in the data are comparable to the dot size; the error bar at bottom right is the uncertainty in the theoretical curves A–C at the highest temperatures.

Fits to magnetoresistance curves like those in Fig. 1 provide another quantitative test. The necklace theory produces reasonable facsimiles, with all qualitative properties and the systematic geometry dependence observed at this low temperature regarding both the oscillations shape and the envelope. However, the shape of the low-field (therefore, most structured) oscillations is not precisely produced by the theory. This is in modest disagreement with earlier work[2,8] where the diffusion lengths were used as free or almost free parameters. The situation is further illustrated by the data in Fig. 3 which show data analogous to those in Fig. 1 but for a set of samples with somewhat stronger spin-orbit scattering. The data are of independent interest because they show the reversal in oscillation phase evidenced previously only by the behavior in the opposing limits of weak and strong spin-orbit scattering. The reversal is related to the well-known "spin-orbit dip" observed in the magnetoresistance of homogeneous systems[7] and in the envelope in the figure. It occurs at a field which is dependent on the unit-cell size as well as the diffusion lengths. Both the single-ring and the array theories predict such behavior with approximately the right T, H, and S dependence. However, exact fits are never obtained and the diffusion lengths corresponding to the best fits generally are much too large. For the rather demanding case in Fig. 3, even the necklace theory fails rather substantially by this kind of criterion, but it always produces an approximation to the data which is clearly superior to the single-ring theory at low temperatures.

In summary, the weak-localization theory as represented by the array work of Doucot and Rammal produces all the general features of the effects ob-

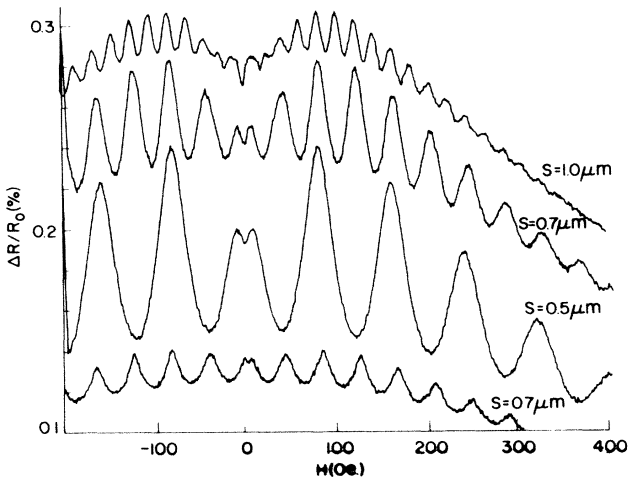

FIG. 3. $R(T,H)$ at $T = 0.13$ K plotted analogously to Fig. 1 for a second set of samples with a higher spin-orbit scattering rate and a consequent S-dependent reversal of the oscillation sign. The top three curves are for the necklace geometry; the bottom curve is for a mesh.

served in our ring arrays, including those specific to arrays as distinguished from a single ring and such a remarkable detail as the phase reversal shown in Fig. 3. The theory does not appear to produce a precise replication of our data within the experimental error and systematically underestimates the oscillation amplitude at small coherence range. Although the latter may arise from a small, undetermined systematic error in our diffusion-length determinations, the evidence indicates a failure of the theory in this regime where, in fact, the single-ring theory becomes more accurate. But the array theory is quantitatively accurate to an extent of practical importance. Our results indicate that determinations of the diffusion lengths from either the array envelope (preferably) or the oscillation amplitude (if one is not too near the phase-reversal region) would be in error by at most 25%. The application of the single-ring theory to interpret, for example, the envelopes in Fig. 1 would produce much larger errors since the suppression of the envelopes is such a large effect. One should note that this suppression is a rather general effect arising from the nonlocal nature of the conductivity at small scales. Analogous effects will influence related measurements like those on a small wire or ring with leads attached.[8] Our results may be used to estimate the size of such distortions.

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[13]The complete form Eqs. (2) and (3) appears nowhere in the literature but the calculation is contained piecemeal for the different elements. The equation includes a modification of the prefactor of the results of AAS. We are grateful to E. Abrahams for pointing out the error in the original result and to Professor Abrahams, R. Bhatt, Y. Imry, D. E. Prober and P. Santhanam for assistance in producing the complete form.