Magnetoresistance in GeAu thin films exhibiting variable-range-hopping conductivity

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(Received 22 December 1994)

Measurements are reported for the magnetoresistance of $Ge_{1-x}Au_x$ thin film samples, which are known from previous measurements to exhibit variable-range-hopping conductivity. Films with $0.075 < x < 0.138$ were studied in magnetic fields from 0 to 4.0 Tesla and in the temperature range 10 to 1.2 K. Magnetoresistance of both positive and negative signs was observed. At a fixed magnetic field, the form of the dependence of resistance upon temperature, for both Mott($T^{-1/4}$) and Shklovskii($T^{-1/2}$) types of variable-range-hopping conduction samples, was found to be unaffected by the presence of magnetic fields in the range studied. A simple ansatz for the hopping exponent in the case of a magnetic field is presented, combined with optimized hopping conduction theory as developed by Mott, which is shown to explain the present data in a natural way.

I. INTRODUCTION

The present understanding of the magnetoresistance in materials exhibiting variable-range-hopping conductivity (VRHC) is rather uncertain, despite a considerable amount of literature of the subject.[1–13] There is no generally accepted theory to explain the large magnetoresistance of both algebraic signs which has been reported.

As discussed in our previous work,[14] the electronic behavior of thin film $Ge_{1-x}Au_x$ is consistent with the theory of weak localization and variable-range hopping in the presence of a Coulomb gap (S&E VRHC), as developed by Shklovskii and Efros [7], extending the work of Miller and Abrahams[15] and Mott[16] on the case with no Coulomb gap (Mott-VRHC). The temperature dependence of the resistivity of our samples, in the low-bias limit and in zero field, accurately follows the VRHC form predicted by *optimized* hopping theory:[7,16,17]

$$\rho(T)=\rho_0 \exp\left[\frac{T^*}{T}\right]^p, \tag{1}$$

where the temperature scaling parameter T^* strongly depends upon the Au concentration on the insulator side of the metal-insulator transition. Most of our thin film samples have $p=1/2$ (S&E VRHC). A few samples with higher Au concentration ($x>0.135$) were also aged in air at room temperature for over six months, resulting in their changing to a Mott VRHC ($p=1/4$).

The present work explores magnetoresistance in $Ge_{1-x}Au_x$ thin films. The measurements show that Eq. (1) remains valid in the temperature range 1.2–10 K for fields between 1.0 to 4.0 T. That is, the temperature dependence of the resistivity in the presence of an external magnetic field can be described as

$$\rho(T,H)=\rho_0(H) \exp\left[\frac{T^*(H)}{T}\right]^p, \tag{2}$$

where $\rho_0(H)$ and $T^*(H)$ are only dependent on the field applied. This behavior was also observed by Tokumoto *et al.*[3] in their investigation of *n*-type InSb at temperatures below 1 K. More recently, Inov and Shlimak also observed such temperature-independent T^* behavior in NTD GaAs and NTDGe systems.[4] As we will discuss in the later section, it is not possible to explain these results using the existing theory. A simple ansatz for the tunneling exponent used to calculate resistivity in VRHC theory is given in Sec. III B, which when "optimized" by the method of Mott[16,17] succeeds in explaining the observed behavior.

II. EXPERIMENT

The fabrication and mounting of the samples has been described in detail in a previous paper.[14] Briefly, the samples were prepared by rapid evaporation of a mechanical mixture of Ge and Au, using a diffusion-pumped filament evaporator fitted with an LN_2 trapped glass bell jar, with base pressure 10^{-6} torr. $Ge_{1-x}Au_x$ thin films 2000 Å thick were deposited on the [100] surface of Si substrates ($\rho > 1500\ \Omega$ cm at room temperature). The evaporation rate was manually controlled at 50–50 Å/sec. Interdigitated electrodes (needed to form connections to these high-resistivity samples) were formed by evaporating thin Au films (about 700 Å) on the top of the $Ge_{1-x}Au_x$ film and patterning the Au thin film photolithographically. Connection to the samples was made through Al (1% Si) wires ultrasonically bonded to the thin film electrodes and to copper-clad PC board sample holder. The area of an individual device was 6.0×6.0 mm^2.

The samples were mounted in a pumped liquid-^{4}He cryostat fitted with a sample heater and a superconducting magnet. The magnetic field was applied (perpendicular)(parallel) to the plane of the $Ge_{1-x}Au_x$ thin film, which was perpendicular or parallel to the predominant direction of current flow in the films. The electric resistance of each sample was measured with a computerized four-terminal measurement system at zero field and at several field settings.

Figure 1 shows the typical results of temperature-

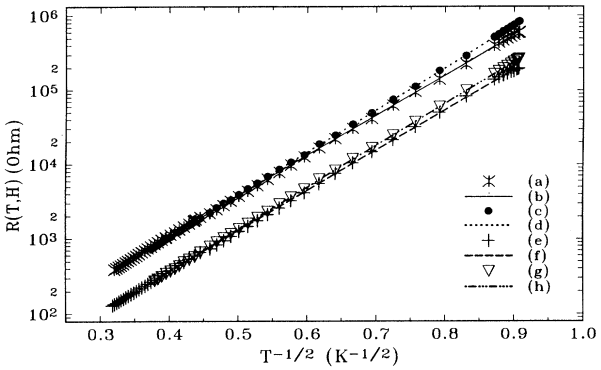

FIG. 1. Ln-linear plot of low-bias resistivity vs $T^{-1/2}$ for thin film Ge-Au samples in zero [(a) for sample $G1$ and (e) for sample $G2$] and in 4.0 T magnetic fields [(c) for sample $G1$ and (g) for sample $G2$]. The straight line fits [(b), (d), (f), and (h)] indicate that low-bias resistivities are dominated by variable-range-hopping conduction (S&E VRHC samples, $x=0.076$) according to Eq. (2) of the text, both with and without a magnetic field.

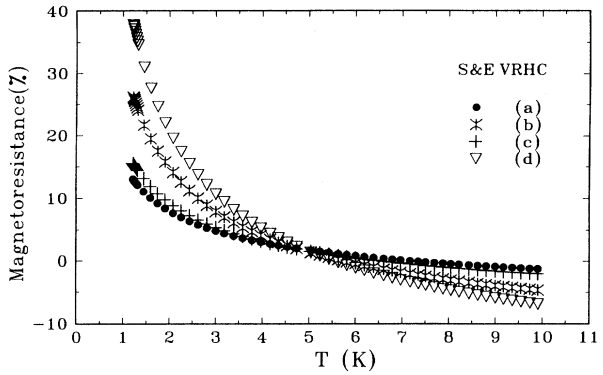

FIG. 3. Magnetoresistance of thin film $Ge_{1-x}Au_x$ (S&E VRHC samples, $x=0.076$). Curves (a) and (b) are the measurements of two samples at 4.0 T. Curves (c) and (d) are the measurements of the two samples at 2.5 T. The plot indicates there is a crossover temperature at which magnetoresistance changes its sign.

resistance dependence of two samples having Au concentration $x=0.076$ and following S&K VRHC, both with [sample $G1(c)$ and sample $G2(g)$ in Fig. 1] and without applied field [sample $G1(a)$ and sample $G2(e)$ in Fig. 1]. The resistance is seen to increase (positive magnetoresistance) under an applied field if the temperature is low enough. At higher temperature the resistance is reduced by the presence of the field (negative magnetoresistance), although this is not easily seen on Fig. 1. We call the crossover temperature between these two behaviors T_{np}. The fact that both curves (with and without applied field) in Fig. 1 are logarithmically linear shows that the form of the temperature dependence of the resistance still follows Eq. (1) in the presence of the applied field. Figure 2 shows the same information for two samples [(a) and (c) for sample $G3$, and (e) and (g) for sample $G4$] having Coulomb gapless (Mott VRHC) behavior, and Au concentration $x=0.138$. Apparently, the effect of the field is to change the parameter T^* and the prefactor ρ_0 in Eq. (1) in a temperature-independent way.

As shown by the straight line fits (b), (d), (f), and (h) in Fig. 1 and Fig. 2, the joint dependence of resistivity upon temperature and field found from the present measurements can be fit by the empirical expression

$$\rho(T,H)=\frac{\rho_0(H=0)}{1+\alpha H^2}\exp\left[\frac{T^*(H=0)}{T}(1+\beta H^2)\right]^p . \quad (3)$$

Here β and α weakly depend on the strength and the direction of the applied field, and $p=1/2$ for S&E VRHC and $p=1/4$ for Mott VRHC. When $\beta H^2 \ll 1$ and $\alpha H^2 \ll 1$, Eq. (3) can be written as

$$\frac{1}{H^2}\ln\left[\frac{\rho(H)}{\rho(H=0)}\right]\approx p\beta\left[\frac{T^*(H=0)}{T}\right]^p-\alpha . \quad (4)$$

It follows from Eq. (3) that total effective magnetoresistance depends on field quadratically and is given by

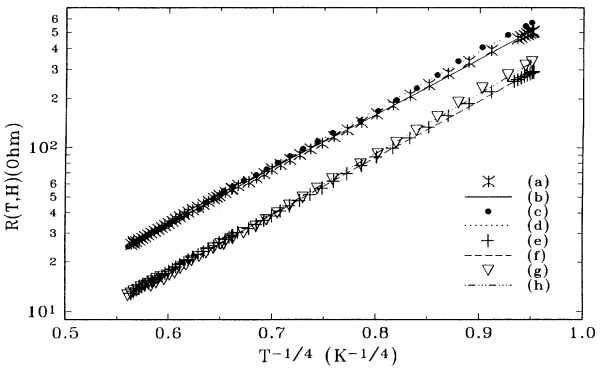

FIG. 2. As in Fig. 1, but for Mott VRHC samples of $Ge_{1-x}Au_x$ with $x=0.138$ (sample $G3$ and sample $G4$).

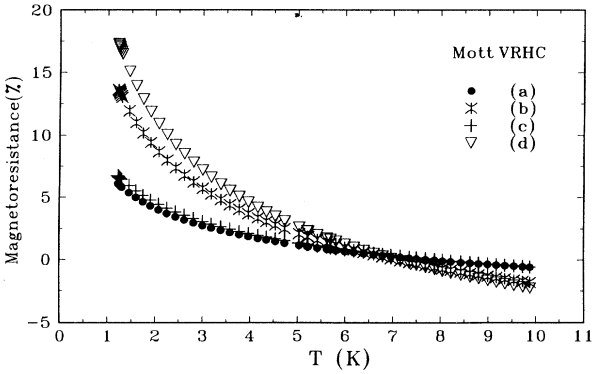

FIG. 4. As in Fig. 3, but for Mott VRHC samples of $Ge_{1-x}Au_x$ with $x=0.138$.

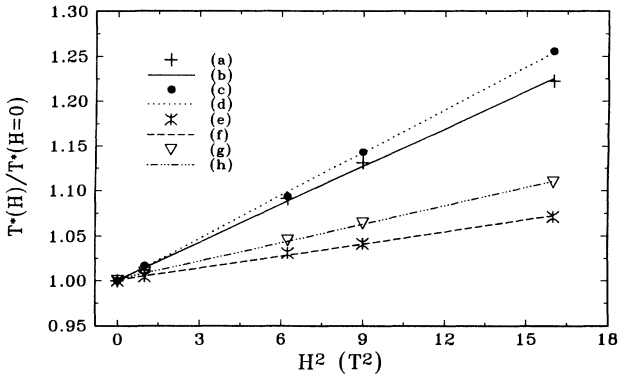

FIG. 5. Experimental results for $T^*(H)/T^*(H=0)$ for both S&E VRHC [(g) for sample $G1$ and (e) for sample $G2$], and Mott VRHC samples [(a) for sample $G3$ and (c) for sample $G4$]. The straight line fits [(b), (d), (f), and (h)] imply that $T^*(H)/T^*(H=0)=1+\beta H^2$. The fitted parameters are given in Table I. The difference in the slope of each group of samples is due to the field orientation.

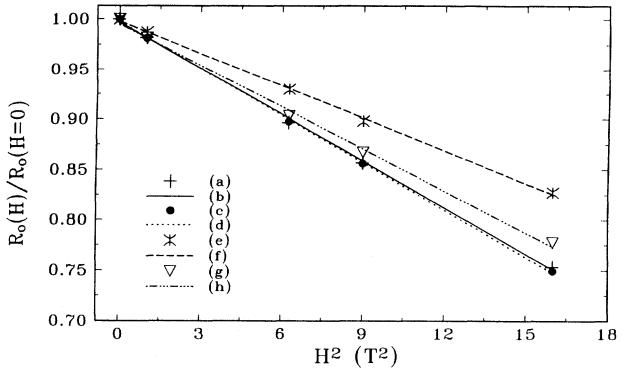

FIG. 6. Experimental results for $R_0(H)/H_0(H=0)$ for both S&E VRHC [(g) for sample $G1$ and (e) for sample $G2$], and Mott VRHC samples [(a) for sample $G3$ and (c) for sample $G4$]. The straight line fits [(b), (d), (f), and (h)] imply that $R_0(H)/R_0(H=0)=1/(1+\alpha H^2)$. The fitted parameters are given in Table I. The difference in the slope of each group of samples is due to the field orientation.

$$\frac{\rho(H)-\rho(H=0)}{\rho(H=0)} \approx \left\{ p\beta \left[\frac{T^*(H=0)}{T} \right]^p - \alpha \right\} H^2 . \quad (5)$$

Equation (5) gives a crossover temperature

$$T_{np} = \left[p\frac{\beta}{\alpha} \right]^{1/p} T^*(H=0) \quad (6)$$

at which the magnetoresistance changes its sign, and therefore magnetoresistance vanishes. Experimentally, this temperature is weakly dependent on magnetic fields. Figures 3 and 4 show the experimental results for the magnetoresistance of thin film $Ge_{1-x}Au_x$ of both VRHC types.

From the above discussion, the effect of an applied magnetic field is to increase the T^* parameter and to decrease the prefactor of the resistivity for a given sample. Both effects are quadratic in the field magnitude, as shown in Figs. 5 and 6. Similar data were obtained for a number of samples with different x values as summarized in Table I. (In Table I, $\|$ is for the field parallel to the plane of the sample, and $\perp$ is for the field perpendicular to the plane of the sample. Measurements showed that parameters of α and β depended weakly on field orientation, both of α and β being slightly larger when the field was applied perpendicular to the samples' plane.)

III. DISCUSSION

A. Previous theory of magnetoresistance in VRHC

Magnetoresistance is always expected to be positive in a metal.[18] However, in semiconductors, magnetoresistance may be positive or negative. Theories have proposed to explain positive and negative magnetoresistance in the variable-range-hopping regime.[1,2,7,11–13,19] In general, positive magnetoresistance results from the shrinkage of the wave functions of localized electronic states due to the external magnetic field. This shrinkage along the direction perpendicular to that of the field applied decreases the probability of hopping, which increases the resistivity. On the other hand, negative magnetoresistance results from the scattering of the tunneling electrons from impurities. Among the great variety of such random scattering sequences, the scattering amplitude might be positive and negative. The hopping probability

TABLE I. Results of fitting parameters describing magnetoresistance of $Ge_{1-x}Au_x$ thin film samples. The direction of the field relative to the film plane is indicated by the symbol in parentheses.

Au Atom (%)	p	$T^*(H=0)$ (K)	$\alpha(T^{-2})$ (2.5 T)	$\alpha(T^{-2})$ (4.0 T)	$\beta(T^{-2})$ (2.5 T)	$\beta(T^{-2})$ (4.0 T)
0.076	1/2(S&E)	158.3	0.0129($\|$)	0.0119($\|$)	0.0056($\|$)	0.0049($\|$)
0.076	1/2(S&E)	157.9	0.0134($\|$)	0.0124($\|$)	0.0058($\|$)	0.0047($\|$)
0.076	1/2(S&E)	157.0	0.0177($\perp$)	0.0173($\perp$)	0.0066($\perp$)	0.0071($\perp$)
0.138	1/4(Mott)	3506.3	0.0166($\|$)	0.0171($\|$)	0.0143($\|$)	0.0135($\|$)
0.138	1/4(Mott)	3494.3	0.0175($\|$)	0.0143($\|$)	0.0139($\|$)	0.0122($\|$)
0.138	1/4(Mott)	4166.7	0.0187($\perp$)	0.0200($\perp$)	0.0146($\perp$)	0.0135($\perp$)

is determined by the interference between many paths associated with hopping sites. The magnetic field destroys this quantum interference, resulting in a tendency for the resistance to decrease with increasing magnetic field for perpendicular orientations of field and current. An additional mechanism involving the narrowing of impurity bands by the applied field[13] is believed to also lead to a negative magnetoresistance at low and moderate fields.

Shklovskii and Efros extensively reviewed the analytical predictions for the effect of magnetic fields upon resistivity in both types of VRHC materials.[7] They defined a magnetic scale length $\lambda=(\hbar c/eH)^2$, and a hopping radius projection in the plane perpendicular to the field $\rho=(x^2+y^2)^{1/2}$. In the weak field limit, which corresponds to $\lambda>a$ and $\rho\ll\lambda^2/a$ (a is the localization length), they predicted

$$\ln\left[\frac{\rho(T,H)}{\rho(T,H=0)}\right]=0.0015\frac{a^4}{\lambda^4}\left\{\frac{T^*(H)}{T}\right\}^{3/2} \tag{7}$$

for VRHC with a density of states having a parabolic Coulomb gap (S&E VRHC), and

$$\ln\left[\frac{\rho(T,H)}{\rho(T,H=0)}\right]=0.0025\frac{a^4}{\lambda^4}\left\{\frac{T^*(H)}{T}\right\}^{3/4} \tag{8}$$

for VRHC with a constant density of states (Mott VRHC). In the strong field limit, which corresponds to reversing both inequalities given above, the prediction was

$$\ln\left[\frac{\rho(T,H)}{\rho(T,H=0)}\right]\approx\left\{\frac{T^*(H)}{T}\right\}^{\frac{\mu+1}{\mu+3}}, \tag{9}$$

with $\mu=0$ for Mott VRHC and $\mu=2$ for S&E VRHC. For the present samples the localization radius a is in the range 50–1000 Å, and as a consequence all fields below about 10 T correspond to the low-field limit. Figures 7

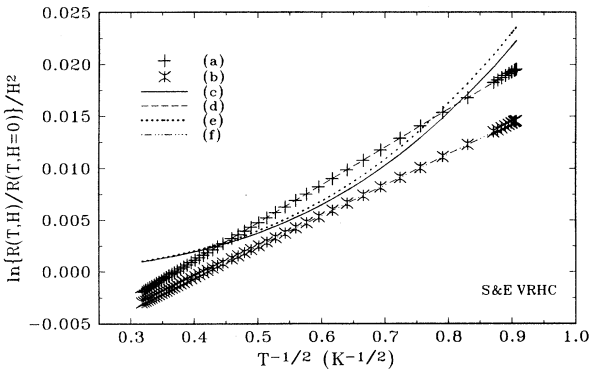

FIG. 7. Comparison of S&E model and the present ansatz with data for one S&E VRHC ($p=1/2$) sample. Single and double crosses correspond to experimental results for $\ln\{R(H)/R(H=0)\}/H^2$ when $H=2.5$ T [labeled (a)] and 4.0 T [labeled (b)], respectively. Curves (e) and (c) are the predictions of the S&E model, Eq. (7). The straight line fits (d) and (f) are the predictions of Eq. (21). Similar results were obtained from testing a number of Ge_xAu_{1-x} thin film samples exhibiting S&E type variable-range-hopping conductivity.

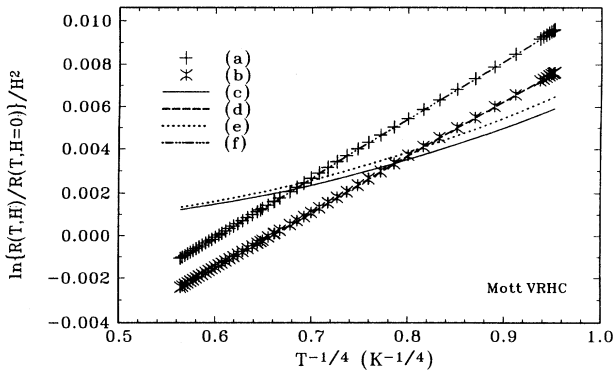

FIG. 8. As in Fig. 7, but for two Mott VRHC samples ($p=1/4$). Similar results were obtained from testing a number of Ge_xAu_{1-x} thin film samples exhibiting Mott type variable-range-hopping conductivity.

and 8 show the present data compared with these predictions. Clearly the data do not follow this theory. As mentioned above, and shown in Figs. 1 and 2, the form of $\rho(T)$ is found experimentally to be unchanged by applied magnetic fields, for both Mott VRHC and S&E VRHC samples. Only theories with this behavior can give a reasonable fit to the whole range of temperature considered. In the next section we present a quantitative explanation of this behavior.

B. Anisotropic optimized hopping

To develop an expression for the resistance in an applied magnetic field which agrees with the present data, we return to the basic theory of VRHC developed by Mott.[7,16] Central to this theory is the hopping exponent ξ_{ij}, which determines the probability of an electron making a transition from one site to a neighboring one. The resistivity associated with the two hopping sites is given by $\rho_{ij}=\rho_{ij}^{(0)}\exp(\xi_{ij})$. In the absence of any applied fields, hopping is spherically symmetric and ξ takes the form

$$\xi=\frac{2r}{a}+\frac{\epsilon}{k_bT}. \tag{10}$$

As discussed in previous work[7,14,20–23] the electric field of the applied bias induces a preferred direction into the energy term of the above expression. Net charge transport results, with a resistivity of the form of Eq. (1). In the present case, additional anisotropy is expected due to the applied field. The influence of a magnetic field can be interpreted as a parabolic potential barrier ("magnetic barrier" as defined by Shklovskii and Spivak[19])

$$\delta\epsilon\sim\frac{\hbar^2(x^2+y^2)}{8m^*\lambda^4} \tag{11}$$

in the Schrödinger equation for an electron in the ground state. As a consequence, the electron has to tunnel not only through a energy barrier ϵ, but also through the field-dependent potential barrier. Therefore a simple ansatz is to take for the tunneling exponent

$$\xi=\frac{2r}{a}+\frac{\epsilon}{k_bT}+\frac{(x^2+y^2)}{b\lambda^4}\frac{a^3}{r}\ , \tag{12}$$

with the parameters λ and a (zero-field localization length) defined as in the previous paragraph, and b a dimensionless constant. This expression has the effect of suppressing tunneling in the direction transverse to the field, by amount which is quadratic in the field and which also is more pronounced for longer hops. The general notion of introducing an ellipsoidal hopping range is due to previous work,[7] but this particular form of ξ is new.

Insight into the meaning of Eq. (12) can be obtained by moving the ϵ/k_bT term to the left-hand side and squaring the result, keeping only the leading magnetic field term. This gives

$$\xi'^2=\left[\xi-\frac{\epsilon}{k_bT}\right]^2=\frac{4r^2}{a^2}\left[1+\frac{(x^2+y^2)}{r^2}\frac{a^4}{b\lambda^4}\right]\ . \tag{13}$$

To identify the right-hand side as the square of a term representing a hopping distance scaled by half the localization length a, we find that the hopping distance is the distance from the origin to an ellipsoid of revolution, satisfying

$$\frac{x^2+y^2}{A^2}+\frac{z^2}{C^2}=1\ . \tag{14}$$

The volume of such an ellipsoid is just $4\pi A^2C/3$, with

$$A^2=\frac{a^2\xi'^2}{4(1+a^4/b\lambda^4)}\quad\text{and}\quad C^2=\frac{a^2\xi'^2}{4}\ . \tag{15}$$

This is the hopping volume introduced by Mott,[16] generalized to the present anisotropic case. To obtain the optimized hopping exponent, we introduce the appropriate density of states $g(\epsilon)$ per unit volume and unit energy measured from ϵ_F, and require that there be exactly one state within the hopping volume. This constraint permits the spatial coordinates to be eliminated from the equation for ξ' in favor of the density of states per unit volume. We can take the density of states per unit volume to be given by

$$N(\epsilon)=N_0\epsilon^n\ , \tag{16}$$

where $N_0=g(\epsilon_F)$ and $n=1$ for Mott VRHC, and $N_0=2\kappa^3/(\pi e^6)$ and $n=3$ for S&E VRHC (κ is the dielectric constant). Solving Eqs. (14) and (15) for ξ gives

$$\xi=\frac{2}{a}\frac{(1+a^4/b\lambda^4)^{1/3}}{(4\pi/3)N(\epsilon)]^{1/3}}+\frac{\epsilon}{k_bT}\ . \tag{17}$$

Optimizing this expression by minimizing $\xi(T,\epsilon;a,b,\lambda)$ with respect to ϵ gives

$$\xi_{\rm opt}=\frac{2(\nu+1)}{(2\nu k_b)^{\nu/(\nu+1)}a^{1/(\nu+1)}}\times\frac{(1+a^4/b\lambda^4)^{1/3(\nu+1)}}{\left[\frac{4\pi}{3}N_0\right]^{1/3(\nu+1)}}\frac{1}{T^{\nu/(\nu+1)}}\ , \tag{18}$$

where $\nu=n/3$. Referring to the definitions of $\xi(T,H)$ and $T^*(H=0)$ in Refs. 7, 14, and 16, we see that Eq. (18) can be rewritten for the cases of S&E VRHC and Mott VRHC respectively as

$$\xi_{\rm opt}^{\rm S\&E}=\left\{\frac{T^*_{\rm S\&E}(H=0)}{T}\left[1+\frac{a^4}{3b\lambda^4}\right]\right\}^{1/2}=\left\{\frac{T^*_{\rm S\&E}(H)}{T}\right\}^{1/2} \tag{19}$$

and

$$\xi_{\rm opt}^{\rm Mott}=\left\{\frac{T^*_{\rm Mott}(H=0)}{T}\left[1+\frac{a^4}{b\lambda^4}\right]\right\}^{1/4}=\left\{\frac{T^*_{\rm Mott}(H)}{T}\right\}^{1/4}\ . \tag{20}$$

Equations (19) and (20) are of the same form found in the absence of applied magnetic field. The parameter $T^*(H)$ is subject to a small positive correction quadratic in the field, leading to an increase in the resistivity whenever a magnetic field is applied.

Thus a simple treatment of the anisotropic tunneling introduced by applied magnetic fields leads to a result in qualitative agreement with the present (as shown in Figs. 1 and 2) and previous experiments (Tukumoto *et al.*[3] and Ionov and Shlimak[4]).

C. Negative and positive magnetoresistance

The present experimental results actually imply that not only is the parameter T^* appearing in the exponent affected by an applied magnetic field, but the prefactor in Eq. (1) is also slightly changed. The change in the prefactor needed to fit the data has the opposite tendency as the change in T^*, tending to decrease the resistivity. The theoretical ansatz described in the preceding paragraphs does not include such an effect. However, it has been pointed out that the effect of applied magnetic field upon resistivity is also *weakened* by impurity scattering,[7,2,8,24] which tends to smear out the anisotropy caused by the field. It was also suggested that negative magnetoresistance would be quadratically dependent on the field.[2,8,13,24,25] Taking an additional factor of the form $\exp\{-f(H)\}=\exp\{-\alpha H^2\}$ in the resistivity-temperature relation and expanding to lowest nonvanishing order in H^2, one obtains

$$\rho(T,H)=\frac{\rho(H=0)}{1+\alpha H^2}\exp\left\{\frac{T^*(H=0)}{T}(1+\beta H^2)\right\}^p\ , \tag{21}$$

which is identical to the empirical expression obtained from fits to the present experimental data, with $\beta^{\rm S\&E}=a^4e^2/(3b\hbar^2c^2)$ and $\beta^{\rm Mott}=3\beta^{\rm S\&E}$. Here b and α in Eq. (21) are free parameters which can be determined by fitting experimental data.

IV. SUMMARY

Experimental results have been presented for the dependence of resistivity upon magnetic field and temperature, in thin film $Ge_{1-x}Au_x$ samples exhibiting VRHC.

The results were consistent with previous experimental results, but disagreed qualitatively with the theory of Shklovskii and Efros. A simple theoretical ansatz was exhibited in which the anisotropy introduced into the tunneling process by the presence of the magnetic field was accounted for. Following this idea through the optimization procedure of Mott to find the $\rho(T,H)$ dependence it implies, a formula was obtained which agrees with the experimental data for magnetoresistance of VRHC samples, both those with and without a Coulomb gap in the density of states.

Further experiments with very heavily doped samples (larger α and β) in very strong fields (greater or of the order of 10 T) could be undertaken to verify the higher-order field dependence predicted by the formulas presented here, and to further study field orientation dependence in VRHC regime.

ACKNOWLEDGMENTS

This work was supported by the Center for Particle Astrophysics, University of California at Berkeley, National Science Foundation, Subcontract No. 22310NM-6, and by Temple University.

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