Crossover from Mott to Efros-Shklovskii variable-range-hopping conductivity in In_xO_y films

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Crossover from Mott variable-range-hopping conductivity to the Coulomb-gap Efros-Shklovskii (ES) variable-range-hopping conductivity has been observed in amorphous indium oxide films. The hopping exponent $x \simeq 0.56$ in the activated Coulomb-gap regime is greater than the $x = 0.50$ value predicted by Efros and Shklovskii. The experimental value of $x \simeq 0.56$ is in excellent agreement with the computational calculations of Mobius and Richter, who suggest that $x = 0.55$. The experimental ratios for $T_{\text{Mott}}/T_{\text{ES}} \simeq 54$ are in close agreement with the prediction of Castner that $T_{\text{Mott}}/T_{\text{ES}} = 81$. Experimental values for the crossover temperatures, which separate the two hopping regimes, are consistent with predicted values. The Coulomb-gap energy Δ_{CG} is estimated to range from a few tenths of a meV for films close to the metal-insulator transition to several meV's for films quite deep in the insulating regime.

I. INTRODUCTION

Although the Mott prediction for variable-range-hopping (VRH) conductivity was published over 20 years ago,[1] and the Efros-Shklovskii prediction for VRH in the "Coulomb gap" has appeared in the literature more than 15 years ago,[2] only very recently have there appeared experimental studies of the crossover from Mott to Efros-Shklovskii VRH conductivity.[3,4] However, some of the experimental results are in rather poor agreement with the predictions as some of the samples were not sufficiently insulating to satisfy several criteria of the two theories. This work spans a wider range of samples that include films quite close to the metal-insulator transition (MIT) and also include films quite deep in the insulating region that exhibit resistances in the GΩ range at liquid-helium temperatures.

II. THEORETICAL BACKGROUND

Recently, Castner has written a review article on hopping transport.[5] This paper draws heavily upon the theoretical results that Castner has summarized, as well as some of his interesting predictions that describe the crossover from Mott to Efros-Shklovskii (ES) VRH conductivity.[5,6]

Provided that the density of states (DOS) near the Fermi energy $N(E_F)$ is a slowly varying function of energy, Mott first predicted the hopping form of the electrical resistivity (conductivity) on the insulating side of the metal-insulator transition for a three-dimensional (3D) material:[1]

$$\rho(T) = \rho_0 \exp(T_{\text{Mott}}/T)^{1/4} , \tag{1}$$

where the characteristic temperature T_{Mott} is found from the slope of the R versus T data. The Mott theory is based upon the Miller-Abrahams pair hopping expression for the phonon-assisted hopping rate;[7] the Miller-Abrahams derivation assumes the existence of an electron localization length ξ. Knowledge of the Mott characteristic temperature T_{Mott} allows the calculation of the localization length ξ:[1,5]

$$\xi = \{18/[k_B T_{\text{Mott}} N(E_F)]\}^{1/3} , \tag{2}$$

where we have used the value of $N(E_F) = 10^{45}/\text{J m}^3$ as suggested by Ovadyahu for indium oxide films.[8] Note that the localization length ξ is expected to diverge to infinity as the metal-insulator transition is approached from the insulating side.[9]

For the Mott theory to be valid, the electron must "hop" a mean distance $\bar{R}_{\text{hop,Mott}}$ that is considerably greater than the nearest-neighbor impurity separation and considerably greater than the localization length ξ. Often, a multihop path will involve three or more sites. Schirmacher recently suggested that the most common hops occur either directly between the initial and final sites or indirectly between a third additional site.[10] The mean hopping distance $\bar{R}_{\text{hop,Mott}}$ and the mean hopping energy difference between sites $\bar{\Delta}_{\text{hop,Mott}}$ can be derived directly from Mott's arguments and will play important roles shortly in determining criteria:[1,5]

$$\bar{R}_{\text{hop,Mott}}/\xi = \tfrac{3}{8}(T_{\text{Mott}}/T)^{1/4} \tag{3}$$

and

$$\bar{\Delta}_{\text{hop,Mott}} = \tfrac{1}{4} k_B T (T_{\text{Mott}}/T)^{1/4} . \tag{4}$$

Mott did not consider the Coulomb interaction between hopping sites. The important "Coulomb-gap" problem was developed by Pollak, Srinivasan, Efros and Shklovskii, and Shklovskii and Efros.[11–14] The theory predicts a power-law dependence in the density of states $N(E)$ near the Fermi energy going as

$$N(E) = N_0 |E - E_F|^{\gamma} , \tag{5}$$

with the exponent γ taking on the value $\gamma = 2$ for 3D films.[13,14] The power-law dependence of the DOS occurs within the "Coulomb gap" Δ_{CG}:[14]

$$\Delta_{\text{CG}} = k_B T_{\text{CG}} \simeq e^3 N(E_F)^{1/2}/\epsilon'^{3/2} , \tag{6}$$

where T_{CG} is the characteristic "Coulomb-gap" temperature, $N(E_F)$ is the unperturbed DOS at the Fermi energy, and ϵ' is the static dielectric response. The static dielectric response is composed of the normal host lattice dielectric response ϵ_{host} and an anomalous contribution:[5]

$$\epsilon' = \epsilon_0 \epsilon'_r = \epsilon_{host} + 4\pi e^2 N(E_F) \xi^2 . \quad (7)$$

Recall that ϵ_0 is the permittivity constant ($\epsilon_0 = 8.85 \times 10^{-12}$ c^2/nm^2); and that for many metallic oxide films, the dielectric coefficient or relative permittivity $\epsilon_{r,host}$ ranges between $10 < \epsilon_{r,host} < 30$. Near the metal-insulator transition, the anomalous contribution can be much greater than the host value owing to the divergence of the localization length ξ. Critical behavior in the dielectric response was first observed by Castner *et al.*[15] Note that if the Mott characteristic temperature is known, then one can estimate the static dielectric response using Eqs. (2) and (7) and thus estimate the Coulomb-gap energy Δ_{CG} via Eq. (6).

The Efros-Shklovskii theory predicts the following resistivity versus temperature dependence for all dimensions:[13,14]

$$\rho = \rho_0 \exp(T_{ES}/T)^{1/2} . \quad (8)$$

Knowledge of the characteristic Efros-Shklovskii temperature T_{ES} also allows one to calculate the magnitude of the static dielectric response:

$$\epsilon' = \epsilon_0 \epsilon'_r = \beta_1 e^2 / (k_B T_{ES} \xi) , \quad (9)$$

provided that the localization length ξ is known. β_1 is a constant on the order of 3. The mean hopping distance $\bar{R}_{hop,ES}$ and the mean hopping energy difference between sites $\bar{\Delta}_{hop,ES}$ are given, respectively, by [5,13,14]

$$\bar{R}_{hop,ES}/\xi = \tfrac{1}{4}(T_{ES}/T)^{1/2} \quad (10)$$

and

$$\bar{\Delta}_{hop,ES} = \tfrac{1}{2} k_B T (T_{ES}/T)^{1/2} . \quad (11)$$

Again, for the ES theory to have meaning, the average hop distance $\bar{R}_{hop,ES}$ must be greater than the nearest-neighbor distance and greater than the localization length.

If the crossover from Mott to Efros-Shklovskii VRH conduction occurs in a material, Castner has made some intriguing predictions.[5,6] He notes that if the sample is not too far away from the metal-insulator transition, then the host dielectric response ϵ_{host} may be neglected and the dielectric response of the material can be approximated by $\epsilon' \simeq 4\pi e^2 N(E_F) \xi^2$. Using this approximation, Castner predicts the following ratios:[5,6]

$$T_{Mott}/T_{ES} = 18(4\pi)/\beta_1 = 81 \quad \text{if } \beta_1 = 2.8 , \quad (12)$$

$$T_{Mott}/T_{CG} = 18(4\pi)^{3/2} = 801 , \quad (13)$$

and

$$T_{ES}/T_{CG} = \beta_1 (4\pi)^{1/2} = 9.9 \quad \text{if } \beta_1 = 2.8 . \quad (14)$$

If T_{Mott} can be determined with sufficient accuracy, then Eq. (13) is an extremely useful expression for estimating the Coulomb-gap characteristic temperature T_{CG} since no adjustable parameters appear in this ratio. However, in our case, the T_{ES}'s are known with better accuracy than the T_{Mott}'s, and Eq. (14) was used instead to estimate the T_{CG}'s. Unfortunately, Eq. (14) contains the parameter β_1 that depends upon the compensation factor K. For $K = 0.5$, then $\beta_1 = 2.8$;[14] for our In_xO_y material, the films are uncompensated and the value for β_1 is not known.

Although films that exhibit the crossover from Mott to Efros-Shklovskii VRH conductivity do suggest the existence of the Coulomb-gap energy, these transport measurements in no way provide direct evidence for the existence of the gap. Only experimental results from phonon conductivity spectroscopy developed by the groups of Eisenmenger and Lassmann can directly establish the existence of the gap.[16,17]

As long as one makes measurements at sufficiently high temperatures such that the Mott hopping energy $\bar{\Delta}_{hop,Mott}$ is considerably greater than the Coulomb-gap energy Δ_{CG}, then the hopping electrons will be affected by a relatively smooth DOS despite the existence of the gap; in this case, the exponent γ in the power-law expression for the DOS is approximately zero. Thus one expects to see Mott VRH conductivity as long as $\bar{\Delta}_{hop,Mott} > 2\Delta_{CG}$.[5,6] This criterion leads to the constraint that the Mott regime must occur in the temperature interval T that satisfies the condition

$$T > (\tfrac{8}{18})^{4/3} T_{Mott}/(4\pi)^2 = T_{Mott}/465 . \quad (15)$$

Since typical values for T_{Mott} range from 1000 to 100 000 K, this criterion is easily satisfied for many materials.

In order to observe Efros-Shklovskii VRH conductivity, one requires that $\bar{\Delta}_{hop,ES} < \bar{\Delta}_{CG}$.[5,6] Thus one expects to see Efros-Shklovskii conductivity as long as the measurement temperatures T satisfy the criterion

$$T < T_{ES}/(\beta_1^2 \pi) = T_{ES}/24.6 \quad \text{if } \beta_1 = 2.8 . \quad (16)$$

Note that this criterion is quite sensitive to the parameter β_1. Since many materials exhibit small magnitudes for the Efros-Shklovskii characteristic temperature, $1 < T_{ES} < 500$ K, very low temperatures in the mK and liquid-^{3}He temperature regimes are often required to observe the Coulomb-gap conductivity. There are not many materials which exhibit the crossover temperature in the convenient temperature regime of 10–50 K; and we will see that amorphous indium oxide films are exceptional in this case. Both criteria given by Eqs. (15) and (16) can be satisfied in the more strongly insulating In_xO_y films.

We note that the crossover occurs when $\bar{\Delta}_{hop,Mott} \simeq \bar{\Delta}_{hop,ES}$. Using Eqs. (4) and (11), the temperature where the crossover occurs, T_{cross}, is given by

$$T_{cross} = 16 T_{ES}^2 / T_{Mott} . \quad (17)$$

Since all three temperatures can be determined experimentally, this prediction can be easily tested; Eq. (17) has the nice feature that no theoretical adjustable parameters appear in it. Note that the crossover temperature T_{cross} is typically twice as large as the characteristic Coulomb-

gap temperature T_{CG} since

$$T_{cross}/T_{CG}=\frac{4}{9}\frac{\beta_1^2}{\pi^{1/2}}\simeq 2 \quad \text{if } \beta_1=2.8 \ . \tag{18}$$

We mention that Mobius and Richter have studied a system of localized electrons interacting via the long-range Coulomb interaction using computer experiments. They found that the DOS decreased considerably faster near the Fermi energy than predicted by the analytical studies; their results can be fairly well approximated by the power law of Eq. (5) with exponents $\gamma=2.7\pm0.1$ for the 3D case and $\gamma=1.5\pm0.05$ for the 2D case.[18,19] Hamilton and Pollak have shown that the exponent γ of the power-law DOS dependence can be related to the hopping conductivity exponent x in the expression $\rho(T)=\rho_0\exp(T_0/T)^x$:[20,21]

$$x=(\gamma+1)/(\gamma+d+1) \ , \tag{19}$$

where d is the dimensionality of the film. For the Mott case where $\gamma=0$, then $x=\frac{1}{4}$ for 3D films; for the Efros-Shklovskii analytical case where $\gamma=2$, then $x=\frac{1}{2}$ for 3D films; and for the computer studies of Mobius and Richter where $\gamma=2.7$, then $x=0.55\pm0.01$ both for 2D and 3D films. Thus it should be experimentally possible to differentiate between the analytical and computational predictions.

Mobius also points out that many 3D disordered materials in the hopping regime exhibit a resistivity ρ that is well described by a scaling law[22,23]

$$\rho(T,n)=\rho_0\phi(T/T_0(n)) \ , \tag{20}$$

where n denotes the impurity concentration and ρ_0 is a constant that is independent of T, n, and the preparation conditions. In particular in the exponential regime, the following form:

$$\rho(T,n)=\rho_0\exp[T_0(n)/T]^{1/2} \tag{21}$$

fits much of the experimental data very well.[22,23] Thus it should also be possible to check the universal behavior of ρ_0 in the indium oxide films.

In summary, the ratios T_{Mott}/T_{ES}, T_{Mott}/T_{CG}, and T_{ES}/T_{CG} given by Eqs. (12), (13), and (14) assume that the two conditions $\bar{R}_{hop}>\xi$ and $\epsilon_{host}\ll 4\pi e^2N(E_F)\xi^2$ are both simultaneously satisfied. Both criteria can be met in samples located "sufficiently deep" into the insulating side of the MIT; such samples have large values for T_{Mott} and T_{ES}. Yet, the samples should not be "so strongly" insulating that the contribution $4\pi e^2N(E_F)\xi^2$ approaches the host dielectric response ϵ_{host} in magnitude; in this case the localization length ξ approaches the impurity Bohr radius or equivalently the value of $0.25(1/n_c)^{1/3}$ from the Mott criterion;[24,25] this limit is typically 10 Å for indium oxide.

III. EXPERIMENTAL PROCEDURES

Only the amorphous form of indium oxide exhibits crossovers from Mott to Efros-Shklovskii conductivity in the convenient temperature interval around 25 K. The transparent polycrystalline form of indium oxide, In_2O_{3-x}, has characteristic temperatures that are one to two orders of magnitude smaller than those of the amorphous In_xO_y form; crossovers in the polycrystalline In_2O_{3-x} material have not yet been observed. Since the polycrystalline form is obtained by heating the substrate to temperatures greater than 150 °C, we intentionally maintained the microscope glass slide substrates at room temperature.[26] High-purity In_2O_3 powder was thermally evaporated at 0.25 Å/s from alumina-coated molybdenum boats in an oxygen partial pressure ranging from 2×10^{-5} to 8×10^{-5} mm Hg.[27,28] Film thicknesses were 460 Å.

The oxygen partial pressure during the evaporation controls the number of donor impurities—namely, oxygen vacancies; the greater the oxygen partial pressure, the smaller the vacancy concentration. Since each oxygen atom within the lattice attempts to close its d shell with two electrons, each vacancy contributes two electrons to the carrier concentration n. Recently, Bregman, Shapira, and Aharoni have demonstrated in indium-tin oxide films that the carrier concentration n and the electrical conductivity σ decrease directly with increasing oxygen partial pressure.[29] Note that we did not intentionally dope our films with any other impurity; thus the compensation in our films should be very small with $K\simeq0$. Our low compensation should be contrasted to compensations of $K\simeq0.45$ used in the CdSe studies of Ref. 4.

IV. ANALYSIS, RESULTS, AND DISCUSSION

The R versus T data were analyzed using a procedure suggested by Zabrodskii and Zinov'eva.[30] Assuming that

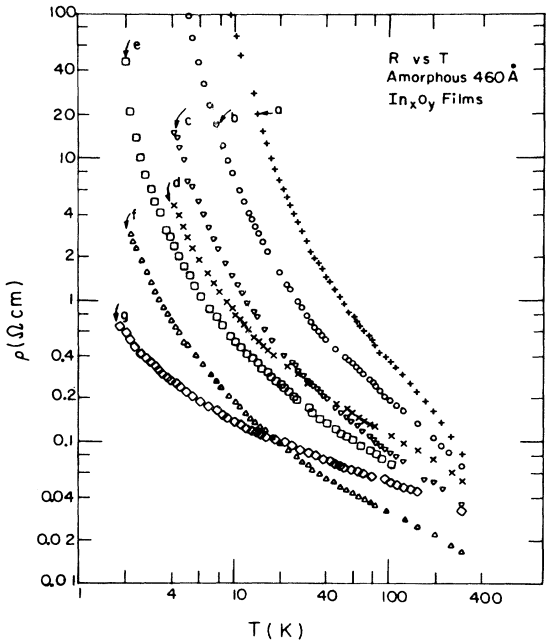

FIG. 1. Resistivity vs temperature data for 460-Å amorphous In_xO_y films. The films differ from one another owing to the number of oxygen vacancies in the material. The number of vacancies is determined by the evaporation rate and amount of oxygen gas introduced during the evaporation.

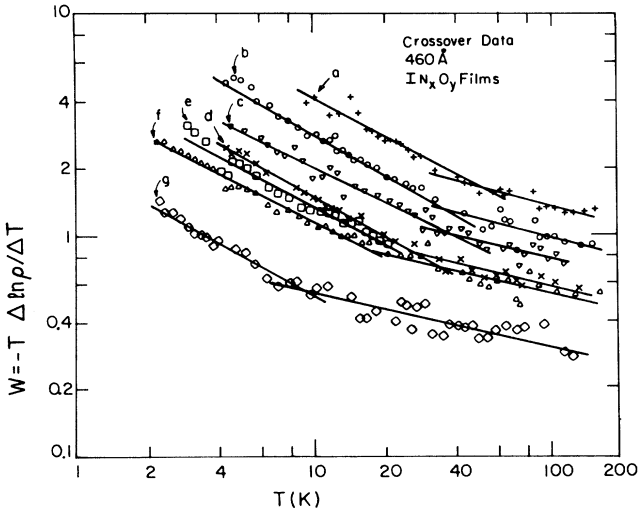

FIG. 2. Plot of w vs T on a log-log plot; w is the slope of $\log_{10}\rho$ vs $\log_{10}T$. The solid lines are the least-squares fits to the $\log_{10}w$ vs $\log_{10}T$ plots. The slopes of the solid lines yield the hopping exponents x and the y intercepts of the lines are related to the characteristic hopping temperatures T_0. For clarity, the "high"-temperature w points of curve e have been omitted. The fitting parameters of all the curves are listed in Table I.

the resistivity data can be fitted to the general hopping law expression $\rho(T)=BT^{-m}\exp(T_0/T)^x$, then the parameter $w(T)=-\partial\log_{10}\rho/\partial\log_{10}T$ is also equal to $w(T)=m+x(T_0/T)^x$. For computational purposes, it was convenient to calculate values of w from the ρ versus T data using the expression

$$w(T)=-\frac{\partial\log_{10}\rho}{\partial\log_{10}T}=\frac{-\partial\ln\rho}{\partial\ln T}\simeq-\bar{T}\frac{\Delta\ln\rho}{\Delta T}\ , \qquad (22)$$

where $\Delta\ln\rho=\ln\rho_1-\ln\rho_2$, $\Delta T=T_1-T_2$, and $\bar{T}=(T_1+T_2)/2$; ρ_1 and ρ_2 are two resistivity values at the closely spaced temperatures of T_1 and T_2. For an exponential hopping dependence of ρ where the second term of $w(T)$ is assumed to be much larger than the first term, that is $m\ll x(T_0/T)^x$, then $\log_{10}w(T)\simeq\log_{10}(xT_0^x)-x\log_{10}T$. This last expression has the form of a linear equation $y=A-Sz$ where $y=\log_{10}w$ and $z=\log_{10}T$. Thus a plot of $\log_{10}w$ versus $\log_{10}T$ yields not only the value of the exponent x from the slope S, but the y intercept A yields the value for the characteristic temperature $T_0=(10^A/x)^{1/x}$.

The resistivity data for the amorphous In_xO_y films are shown in Fig. 1. From these data values for the w's of the logarithmic slopes were calculated using Eq. (22) and are shown in Fig. 2. The w's for each film exhibit, as seen from Fig. 2, a high-temperature interval where the slope of $\log_{10}w$ is considerably smaller than the slope of $\log_{10}w$ in the low-temperature interval. The method of linear regression (least-squares method) was used to determine the best slope S and the best intercept A for both the high- and low-temperature intervals. The fitting results are summarized in Table I where values for the exponent x, the characteristic temperature T_0, and the prefactor ρ_0 are presented. The x's are known to an accuracy of ±0.04 and the T_0's to an accuracy of $\pm100\%$. Note that the values of T_0's are very sensitive to both the y intercept A and to the exponent x since $T_0=(10^A/x)^{1/x}$. The fitting parameters x and T_0 describe the "best fitting" lines through the $\log_{10}w$ data and these lines are shown as the solid lines in Fig. 2; the intersection of the two lines for each data set was used to define the value for the crossover temperature T_{cross}.

Two rather surprising results are seen from Table I. At low temperatures the exponent x is consistently greater than the value of $\frac{1}{2}$ predicted analytically by Efros and Shklovskii.[13,14] In fact, the mean value of x for all the films is 0.56 in very close agreement with the Mobius and Richter computational results of 0.55.[18,19] Perhaps the theoretical model should be refined. The other interesting result is the value of the prefactor ρ_0 in the low-temperature ES regime. The value of ρ_0 does not vary by more than a factor of 2 from film to film; this behavior does support Mobius's claim of universality for ρ_0.[22,23] In contrast in the Mott high-temperature regime, values of ρ_0 vary by a factor of 10 from film to film. Note that all the films are 3D since $\bar{R}_{\text{hop}}<d_{\text{thickness}}$.

In Table II, we compare the experimental results with the theoretical predictions. According to Castner, $T_{\text{Mott}}/T_{\text{ES}}\simeq81$ provided that $\beta_1=2.8$;[5] our mean value for this ratio is 54. Considering our inability to determine the T_{Mott}'s with much accuracy, the experimental result seems quite consistent with the theory and in conflict with the experimental findings of Zhang *et al.* that $T_{\text{ES}}\propto(T_{\text{Mott}})^{2/3}$.[4] However, our films are much

TABLE I. Fitting parameters to the conductivity hopping law $\rho(T)=\rho_0\exp(T_0/T)^x$.

Film		High-T's resistivity data			Low-T's resistivity data		
Symbol	Designation	x	$T_0=T_{\text{Mott}}$ (K)	ρ_0 (Ω cm)	x	$T_0=T_{\text{ES}}$ (K)	ρ_0 (Ω cm)
+	a	0.281	27 590	0.031 0	0.564	352	0.0514
○	b	0.277	9 780	0.006 02	0.598	140	0.0692
▽	c	0.262	6 765	0.004 30	0.521	140	0.0305
×	d	0.274	1 700	0.012 8	0.590	49.9	0.0652
□	e	0.250	4 320	0.005 56	0.552	52.3	0.0450
△	f	0.276	1 240	0.004 33	0.545	40.9	0.0216
◇	g	0.244	292	0.015 2	0.580	8.9	0.0571

TABLE II. Comparison between experimental and theoretical results and some theoretical predictions.

		Experimental results				Theoretical results						
Symbol	Film Designation	T_{Mott} (K)	T_{ES} (K)	$T_{\text{Mott}}/T_{\text{ES}}$	T_{cross} (K)	T_{cross} (K)	T_{CG} (K)	Δ_{CG} (meV)	ξ (Å)	ϵ_r'	$\bar{R}_{\text{hop,Mott}}/\xi$	$\bar{R}_{\text{hop,ES}}/\xi$
+	*a*	27 590	352	78.4	52	72	35.5	3.06	36	471	$4.8/T^{1/4}$	$4.7/T^{1/2}$
○	*b*	9 780	140	70.0	38	32	14.1	1.21	51	941	$3.7/T^{1/4}$	$3.0/T^{1/2}$
∇	*c*	6 765	140	48.3	39	46.5	14.1	1.21	58	1205	$3.4/T^{1/4}$	$3.0/T^{1/2}$
×	*d*	1 700	49.9	34.0	26.5	23.5	5.0	0.43	91	3022	$2.4/T^{1/4}$	$1.8/T^{1/2}$
□	*e*	4 320	52.3	82.6	19	10.1	5.3	0.46	67	1621	$3.0/T^{1/4}$	$1.8/T^{1/2}$
△	*f*	1 240	40.9	30.3	17.5	21.6	4.1	0.36	101	3736	$2.2/T^{1/4}$	$1.6/T^{1/2}$
◇	*g*	292	8.9	33	9.6	4.3	0.9	0.08	164	9784	$1.6/T^{1/4}$	$0.75/T^{1/2}$

more "insulating" than the CdSe films studied by Sarachik's group.[4] Shown in Table II is a comparison between the experimental and theoretical values [from Eq. (17)] of the crossover temperature T_{cross}; again there is agreement to within a factor of 2. Also included are estimated values for the Coulomb-gap temperature T_{CG} and energy Δ_{CG} as well as values for the localization length ξ [from Eq. (2)] and the relative dielectric constant ϵ_r' [from Eq. (7)]. The values of ξ and ϵ_r' seem reasonable; and the criterion that $\epsilon_{\text{host}} \ll 4\pi e^2 N(E_F)\xi^2$ is easily met. The high-temperature criterion of Eq. (15) for Mott hopping seems to be well satisfied by all the films, and even the criterion that $\bar{R}_{\text{hop,Mott}} \geq \xi$ seems to be satisfied. However, in the Efros-Shklovskii regime, the criterion of Eq. (16) fails badly for all the films, and the criterion that $\bar{R}_{\text{hop,ES}} \geq \xi$ is not satisfied except in the most insulating films. Perhaps the reason that the criterion of Eq. (16) fails is owing to the value used for $\beta_1 = 2.8$; our data suggest that $\beta_1 \simeq 1.4$; but then the ratio $T_{\text{Mott}}/T_{\text{ES}}$ should be 162 rather than 81. It is not clear how to resolve this discrepancy.

In conclusion, the indium oxide crossover data seem quite consistent with the Mott and Efros-Shklovskii theories. Our results do suggest the existence of the Coulomb gap; it now remains an experimental challenge to observe the gap directly.

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