Thermodynamic and thermoelectric properties of (Ga,Mn)As and related compounds

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Various experimental results providing information on thermodynamic density of states in (Ga,Mn)As are analyzed theoretically assuming that holes occupy GaAs-like valence bands. Allowing for Gaussian fluctuations of magnetization, the employed model correctly describes a critical behavior of magnetic specific heat found experimentally in (Ga,Mn)As near the Curie temperature T_C [S. Yuldashev *et al.*, Appl. Phys. Express **3**, 073005 (2010)]. The magnitudes of room-temperature thermoelectric power, as measured for GaAs:Be and (Ga,Mn)As [M. A. Mayer *et al.*, Phys. Rev. B **81**, 045205 (2010)], are consistent with the model for the expected energy dependencies of the hole mobility. The same approach also describes temperature variations of conductance specific to the Anderson-Mott localization, found for various dimensionality (Ga,Mn)As nanostructures at subkelvin temperatures [D. Neumaier *et al.*, Phys. Rev. Lett. **103**, 087203 (2009)]. We conclude that the examined phenomena do not provide evidence for an enhancement of density of states by the presence of an impurity band at the Fermi energy in ferromagnetic (Ga,Mn)As. Furthermore, for (Ga,Mn)As we provide expected values of both electronic specific heat at low temperatures $T \ll T_C$ and magnetization as a function of the magnetic field at T_C.

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I. INTRODUCTION

Extensive studies of hole-controlled ferromagnetic semiconductors, such as (Ga,Mn)As and p-(Cd,Mn)Te, have allowed one to demonstrate a variety of functionalities specific to these systems.[1,2] At the same time, however, results of various experiments have faced us with a number of challenges suggesting that the understanding of these materials is far from satisfactory. For instance, recent studies of (Ga,Mn)As using two kinds of tunneling spectroscopy have led to two entirely contradictory pictures of electronic states in the vicinity of the Fermi energy E_F. According to scanning tunneling microscopy,[3] and in agreement with the Altshuler and Aronov[4] and Finkelstein[5] descriptions of the Anderson-Mott localization for the spin-polarized band carriers,[6] the one-electron density of states (DOS) attains a minimum at E_F and its depression extends over an energy range of the order of the momentum relaxation rate, $\hbar/\tau \sim 100$ meV in (Ga,Mn)As.[7] In contrast to the one-electron DOS probed in tunneling experiments, the DOS for charge excitations as well as thermodynamic DOS, $\rho_F = \partial p/\partial E_F$, are only weakly renormalized by carrier correlation and disorder in doped semiconductors on the metal side of the Anderson-Mott transition, where the ratio of the interparticle distance to an effective Bohr radius is relatively small, $r_s = (4\pi p/3)^{-1/3}/a_B^* \lesssim 2.4$.

A rather different picture emerges from resonant tunneling spectroscopy of (Ga,Mn)As quantum wells.[8,9] According to the interpretation of the accumulated findings, the Fermi level resides in a narrow impurity band located $\sim$50 meV above the edge of hole subbands. These subbands are well resolved by resonant tunneling spectroscopy and, thus, virtually unaffected by disorder.[8,9]

The impurity-band model implies an enormous thermodynamic DOS of carriers at the Fermi level. An unusually large effective mass emerges also within some interpretations of the optical[10] and transport[11] properties of (Ga,Mn)As. Historically, large magnitudes of effective masses in, for example, heavy-fermion systems[12] and Kondo alloys[13,14] were discovered by studies of thermodynamic and thermoelectric effects. Therefore, it is of particular importance to describe theoretically those properties of (Ga,Mn)As, which provide information on thermodynamic DOS. Since this DOS, in contrast to the one-electron DOS probed in tunneling experiments, does not exhibit a Coulomb anomaly at E_F,[4] it should show a significant enhancement if relevant electronic states are indeed characterized by a large effective mass.

In this paper, various experimental results providing information on thermodynamic DOS in (Ga,Mn)As are analyzed theoretically assuming that holes occupy GaAs-like valence bands, described here by the six-band $k \cdot p$ model with effects of the p-d exchange interaction included within the molecular field approximation.[15,16] We show that the employed model, taking into account Gaussian fluctuations of magnetization, correctly describes a critical behavior of magnetic specific heat found experimentally in (Ga,Mn)As around the Curie temperature T_C.[17] We also provide the expected field dependence of magnetization at T_C within the mean-field approximation. Furthermore, we show that the magnitudes of room-temperature thermoelectric power, as measured for GaAs:Be and (Ga,Mn)As,[18] are consistent with our model for the expected energy dependencies of the hole mobility. We then turn to low temperatures $T \ll T_C$ and present computed values of the Sommerfeld coefficient γ, describing electronic specific heat, and magnetization as a function of the magnetic field at T_C. We also employ the same approach to discuss temperature variations of conductance specific to the Anderson-Mott localization, determined experimentally for various dimensionality (Ga,Mn)As nanostructures at subkelvin temperatures.[19] We conclude that the examined phenomena do not provide any evidence for an enhancement of thermodynamic DOS by

the presence of an impurity band at E_F in ferromagnetic (Ga,Mn)As.

II. CRITICAL BEHAVIOR OF SPECIFIC HEAT

Recently, a critical behavior of specific heat in $Ga_{1-x}Mn_xAs$ was resolved experimentally for two samples with Mn concentration $x = 1.6\%$ and 2.6%, which showed insulating and metallic behavior, respectively.[17]

In order to describe these findings we employ the Ginzburg-Landau approach, taking into account critical fluctuations in the Gaussian approximation.[20] We specify by $\sigma_i(x)$, $i = 1,2,\ldots,n$, a local magnitude of an n-dimensional order parameter (e.g., a magnitude of local spin density) around x in a d-dimensional block. The block Hamiltonian (the free energy functional) $H[\sigma(x)]$ at temperature T and in magnetic field h can then be written as an expansion in powers of $\sigma(x)$ and $\nabla\sigma(x)$,[20]

$$H[\sigma(x)]/k_BT = \int d^dx\,[a_0 + a_2\sigma^2 + a_4(\sigma^2)^2 + c(\nabla\sigma)^2 - h\sigma], \quad (1)$$

where

$$\sigma^2 = \sum_{i=1}^{n}[\sigma_i(x)]^2, \quad (2)$$

$$(\nabla\sigma)^2 = \sum_{\alpha=1}^{d}\sum_{i=1}^{n}\left(\frac{\partial\sigma_i}{\partial x_\alpha}\right)^2. \quad (3)$$

The temperature-dependent coefficients a_k ($k = 0,2,4$) and c describe the free-energy cost associated with a change in the magnitude and in the local direction of the order parameter, respectively.

Within the Gaussian approximation and taking into account only terms that are singular on approaching the ordering temperature T_C, the specific heat assumes a critical behavior given by[20] $C = C^{\pm}t^{d/2-2}$, where $C^+ = nC_0$ and $C^- = 2^{d/2}C_0$ apply to the $T > T_C$ and $T < T_C$ cases, respectively; $t = |T - T_C|/T_C$ and

$$\frac{C_0}{k_B} = \frac{1}{2}(2\pi)^{-d}(a_2'T_C/c)^{d/2}\int d^dk'\,(1+k'^2)^{-2}, \quad (4)$$

where a_2' defined by $a_2 = a_2'(T - T_C)$ shows no singular dependence on temperature around T_C.

The integral in Eq. (4) is convergent for $0 < \Re(d) < 4$ (a Debye cutoff is necessary for higher dimensions) and is given by

$$\int d^dk'\,(1+k'^2)^{-2} = \frac{d\pi^{d/2}}{\Gamma(1+d/2)}\frac{(2-d)\pi}{4\sin(d\pi/2)}, \quad (5)$$

which yields $\pi/2$, π, and π^2 for d equal to 1, 2, and 3, respectively.

In order to determine the magnitudes of a_2' and c for (Ga,Mn)As we note that, within the p-d Zener model,[15,21,22] the free-energy functional consists of two contributions. The first describes the free energy of localized spins of magnitude S in the absence of carriers, whereas the second is the free energy of the Fermi liquid of holes in the valence band in the molecular field of a prescribed configuration of the localized spins (for such a description to be possible it is required that the dynamics of the localized spins are much slower than that of the carriers). The form of particular contributions to the Ginzburg-Landau free-energy functional was determined[15,22,23] employing magnetization of localized spins $M(x) \equiv \sigma(x)M_{sat}/S$ as an order parameter, where the saturation magnetization is related to the magnetic moment $g\mu_BS$ and the effective concentration of localized spins N_0x_{eff} in a standard way, $M_{sat} = g\mu_BSN_0x_{eff}$, where the g factor $g \approx 2.0$. Neglecting interactions between localized spins in the absence of carriers, and assuming that the carrier liquid is strongly degenerate,[15,22] we obtain

$$a_2'T_C = \frac{3x_{eff}N_0}{2S(S+1)} \quad (6)$$

and, in terms of the magnetic stiffness A,[23,24]

$$c = \lim_{M\to 0}\frac{A(M)M_{sat}^2}{S^2M^2k_BT_C} \equiv \frac{(N_0\beta x_{eff})^2}{k_BT_C}\frac{B}{2}, \quad (7)$$

where B is a property of the electronic subsystem, N_0 is the cation concentration, and β is the p-d exchange constant. Using the definition of D_{nor} introduced in Ref. 23 for the case of $d = 3$,

$$D_{nor} = \frac{4(S+1)k_F^2D}{k_BT_C}, \quad (8)$$

where $D = x_{eff}N_0S\beta^2B = 2A(M_{sat})/x_{eff}N_0S$ (we have neglected the difference between B and the analogous quantity at $T = 0$) and $k_F = \sqrt[3]{3\pi^2p}$, we can write

$$\frac{C_0}{k_B} = \frac{3\sqrt{3}}{2\pi}\frac{k_F^3}{D_{nor}^{3/2}} = \frac{9\sqrt{3}\pi}{2}\frac{p}{D_{nor}^{3/2}}. \quad (9)$$

Since, according to Eq. (9), C_0 depends solely on the carrier density p and the normalized spin wave stiffness D_{nor}, where $D_{nor} = 1$ for s-type spin-1/2 carriers in a parabolic band and $D_{nor} \approx 10$ for the GaAs valence band,[23,24] it can be expected that, similarly to thermodynamic DOS ρ_F, C_0 does not show any critical behavior across the metal-insulator transition (MIT).

In Ref. 17, experimental data were presented for the specific heat of two (Ga,Mn)As films. According to x-ray diffraction measurements and x-ray microanalysis, the Mn concentration x is 1.6% in the sample A and 2.6% in the sample B, with T_C of 40 and 52 K, respectively. Due to relatively low x values, these samples are expected to be close to the MIT. This expectation is confirmed by temperature dependencies of resistance, which indicate that samples A and B are on the insulating and metallic side of the MIT, respectively.[17] According to magnitudes of the Hall resistance ρ_H at room temperature, the hole concentrations are 2.7×10^{19} and 4.5×10^{19} cm^{-3} for samples A and B, respectively. Since, however, ρ_H tends to diverge near the MIT (Ref. 25) (if measured at temperatures below the impurity binding energy, $\sim$1000 K in GaAs:Mn), it leads to underestimated values of the hole concentrations. Furthermore, owing to critical fluctuations in the local DOS near the MIT,[3,15,26] only a part of the sample volume is ferromagnetic.[15,26] This means that the apparent values of low-temperature spontaneous magnetization are smaller than the expected magnitudes of saturation magnetization $M_{sat} =$

$g\mu_B S N_0 x_{\rm eff}$, as observed.[18,27,28] It is worth noting that while scanning tunneling microscopy and its theoretical description[3] provide information on the fluctuations of single-particle DOS, to our knowledge there are no corresponding data on the local behavior of thermodynamic DOS (such data could help to quantitatively assess the expected reduction in the magnitude of spontaneous magnetization near the MIT).

Assuming that $x_{\rm eff} < x$ only due to the presence of interstitial Mn, we have $x_{\rm eff} = x - 2x_{\rm i}$, where x and $x_{\rm i}$ are the total and interstitial Mn contents. Similarly, the hole concentration is $p = N_0(x - 3x_{\rm i})$.[29] Using these equations and the mean-field theory,[15] we determine the values of $x_{\rm eff}$ and p, which reproduce the experimental values of $T_{\rm C}$. We find $x_{\rm eff} = 1.6\%$ and $p = 3.5 \times 10^{20}$ cm^{-3} for sample A, whereas for sample B the values are 2.0% and 3.8×10^{20} cm^{-3}, respectively. As expected, the magnitudes of the hole concentrations obtained in this way are larger than the ones obtained from the Hall resistance, as quoted above. Similarly, the measured values of spontaneous magnetization[17] are smaller by factors 4.7 and 4.0 r than $M_{\rm sat}$ calculated for the values of $x_{\rm eff}$ for samples A and B, respectively. We note that the contribution of holes' magnetic moment to experimentally available magnetization may account for its reduction by less than 20%.[30]

We use the above sample parameters as well as GaAs Luttinger parameters and $N_0\beta = -1.2$ eV.[15,16] The values of B have been determined within the six-band $k \cdot p$ model,[23,24] in which we have neglected $E_{\mathbf{k}}^{++}$. The numerical value is $B = 0.17$ eV^{-1} nm^{-1} for both samples. These numbers yield $C_0 = 0.13$ J/(mol K) for sample A and $C_0 = 0.14$ J/(mol K) for sample B. The corresponding dependencies of the specific heat on temperature are shown in Fig. 1, where experimental data from Figs. 3 and 4 of Ref. 17 are depicted with dots. We see that the theory describes quite reasonably the experimental values, particularly if one can assume that, owing to magnetic anisotropy, the system is effectively Ising-like.

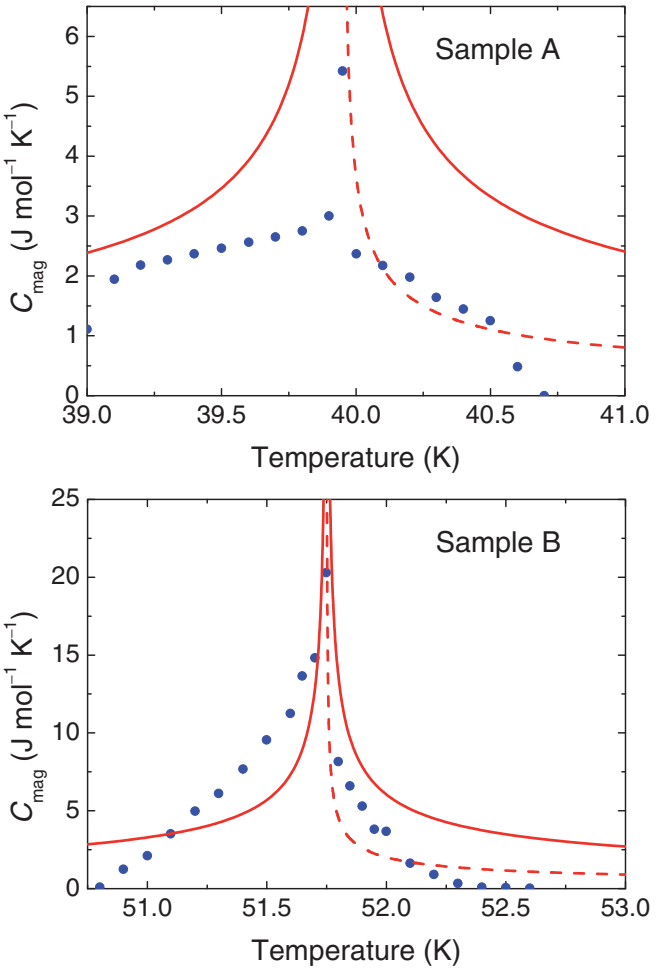

FIG. 1. (Color online) Theoretical temperature dependence of the magnetic specific heat calculated with no adjustable parameters for sample A (upper panel) and sample B (lower panel). The solid and dashed lines correspond to the Heisenberg ($n = 3$) and Ising ($n = 1$) models, respectively. Dots represent experimental data [after Yuldashev *et al.* (Ref. 17)].

III. CRITICAL BEHAVIOR OF MAGNETIZATION

Within the Gaussian approximation, the temperature and field dependencies of magnetization $M(T,H)$ are given by the mean-field formula. Since theoretically expected values of spontaneous magnetization $M(T)$ were already presented,[16,31] here we discuss only $M(H)$ at $T = T_{\rm C}$. This dependence is determined by the coefficient a_4 in Eq. (1) according to

$$\bar{\sigma} = (h/4a_4)^{1/3}. \tag{10}$$

Within the p-d Zener model $a_4 = a_{4(S)} + a_{4(c)}$, where

$$a_{4(S)} = \frac{9(2S^2+2S+1)}{40S^3(S+1)^3} N_0 x_{\rm eff} \approx 6.2 \times 10^{-3} N_0 x_{\rm eff}, \tag{11}$$

is the localized-spin contribution and $a_{4(c)}$, the carrier contribution, is calculated numerically from the expansion of the carrier free energy in the spin splitting parameter $B_G = A_{\rm F}\beta M/(6g\mu_B)$,

$$F_c(B_G) = F_c(0) - p_2 B_G^2 + p_4 B_G^4, \tag{12}$$

where $A_{\rm F}$ is the Landau parameter describing the correlation-induced enhancement of the carrier spin susceptibility, $A_{\rm F} = 1.2$,[15,32,33] whereas the expansion coefficients p_i are determined by the band-structure parameters and the hole concentration. We obtain

$$M^3 = \frac{(g\mu_B N_0 x_{\rm eff})^4}{4k_B T_{\rm C}(a_{4(S)} + a_{4(c)})} \mu_0 H, \tag{13}$$

where

$$k_B T_{\rm C} = A_{\rm F}\beta^2 N_0 x_{\rm eff} S(S+1) p_2/54 \tag{14}$$

and

$$a_{4(c)} = A_{\rm F}(\beta N_0 x_{\rm eff}/6)^4 p_4/k_B T. \tag{15}$$

The computed magnitudes of the slope $M(H)^3/H$ at various hole concentrations and effective Mn contents are presented in Fig. 2. The decrease of the slope with increasing hole concentration, seen in the plot for low x, is due to an increase of $T_{\rm C}$. The value for sample A is $0.039\,({\rm emu/cm^3})^3/{\rm Oe}$, which corresponds to the solid line shown in Fig. 3 against the experimental data.[17] As seen, the theoretical values of magnetization $M(H)$ satisfactorily reproduce the character of the field dependence observed experimentally. However, the

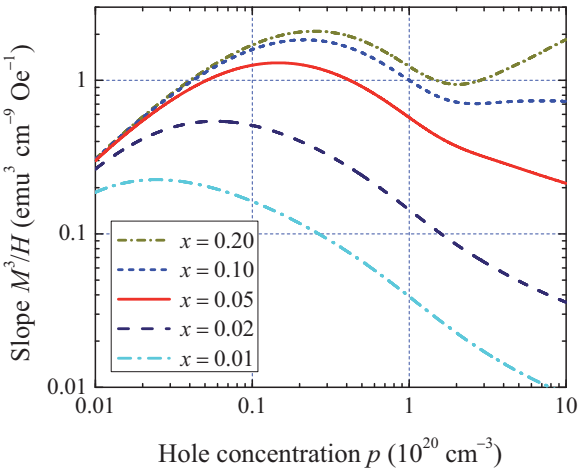

FIG. 2. (Color online) Computed values of the proportionality coefficient between $M(H)^3$ and H for Mn magnetization M in the magnetic field H at T_C. The calculation has been performed within the p-d Zener model for various effective Mn contents x in (Ga,Mn)As.

absolute magnitudes of computed magnetization are greater than the experimental ones by a factor of about 2.45. We attribute this discrepancy to a reduction of the ferromagnetic phase volume by critical fluctuations in the local DOS in the vicinity of the MIT, as discussed in the previous section. In fact, this factor is even smaller than the ratio of saturated and low-temperature spontaneous magnetization, determined to be 4.7 for the sample in question, as discussed in the previous section.

IV. HIGH-TEMPERATURE THERMOELECTRIC POWER

In general, thermoelectric power contains diffusion and phonon drag contributions,[34] as well as, in the ferromagnetic case, a magnon drag term. The magnitudes of the drag terms scale with phonon and magnon relaxation times, so that they dominate at low temperatures, particularly in annealed

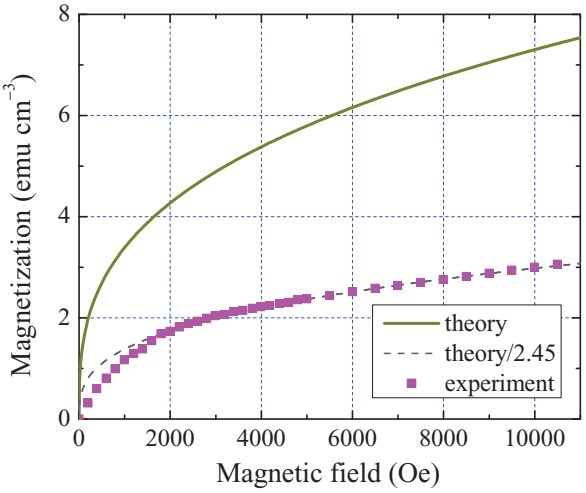

FIG. 3. (Color online) Comparison of theoretical (solid line) and experimental (symbols) field dependence of magnetization at T_C for sample A of Yuldashev *et al.*[17] The dashed line shows theoretical values reduced by a factor of 2.45 to match experimental data.

(Ga,Mn)As samples,[35–39] where a reduced concentration of Mn interstitials may suppress relevant scattering.

Awaiting a quantitative theory of phonon and magnon scattering in (Ga,Mn)As, we limit our considerations to high temperatures, where the diffusion term is expected to dominate. Furthermore, we assume that the measured magnitude of the thermoelectric power (Seebeck coefficient) is not perturbed by buffer or substrate contributions. For spherical bands with arbitrary dispersion, the diffusion thermopower for carriers with charge $+e$ is given by[40]

$$S = \frac{k_B}{e} \frac{\left\langle \frac{E-E_F}{k_B T} \mu(E) \right\rangle}{\langle \mu(E) \rangle}, \tag{16}$$

where f_0 in

$$\langle A \rangle = \int dE \left(-\frac{\partial f_0}{\partial E} \right) A(E) k^3(E) \tag{17}$$

is the Fermi-Dirac distribution function and $k^3(E)$ describes the spherical band. The mobility μ depends on the hole energy E with respect to the top of the corresponding valence-band subband as

$$\mu(E) = \mu_0 E^r, \tag{18}$$

where the exponent r depends on the mechanism which limits the carrier mobility. We have $r \approx -1/2$ for scattering by acoustic phonons and $r \approx 3/2$ for scattering by ionized impurities.

In the case of a bulk GaAs-like semiconductor (zinc-blende structure, valence-band maximum at the Γ point), there are two kinds of carriers: heavy and light holes. Therefore, both the numerator and the denominator in Eq. (16) are sums of the contributions from each of the subbands. Therefore, the resulting thermopower coefficient is a weighted average of the coefficients calculated separately for each subband at the Fermi level, which is determined by the total concentration of holes distributed over both subbands. Since the weights include the unknown parameter μ_0, which in general has a different value for each subband, we cannot determine this average. However, we can consider S calculated for the heavy and light hole subbands separately as the limiting cases, as the final result has to fall within the range spanned by them.

The results of computations carried out for 300 K and employing the six-band $k \cdot p$ model[16] are presented in Fig. 4. The thermoelectric power S is shown separately for the heavy and light holes. It is expected that the heavy holes dominate for the case of scattering on ionized impurities, while the opposite is true for scattering on acoustic phonons (we assume that the Fermi energy is small compared to the spin-orbit splitting of the valence band).

The theoretical data are compared to experimental values obtained at room temperature by Mayer *et al.*[18] for a series of GaAs:Be and (Ga,Mn)As samples, in which hole density was changed by irradiation with high-energy Ne^+ ions that introduce compensating donor defects. We note that for these (Ga,Mn)As samples the content of substitutional Mn is $x = 0.045$, and the MIT occurs at the hole concentration $p_c \approx 3 \times 10^{20}$ cm^{-3},[18] a value about two orders of magnitude higher than p_c for GaAs:Be. Nevertheless, the difference between the magnitude of S in (Ga,Mn)As and GaAs:Be is only slight,

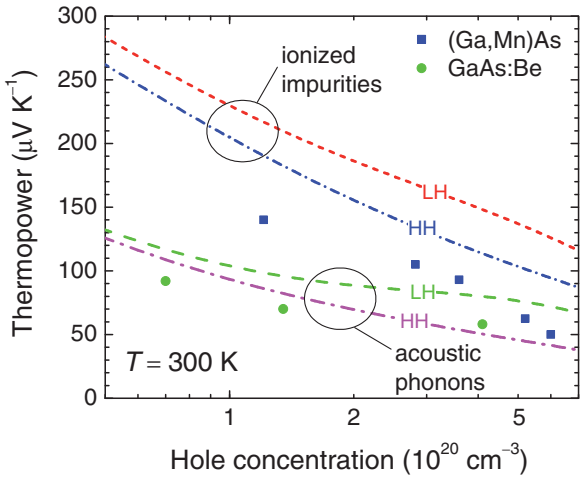

FIG. 4. (Color online) Room-temperature thermoelectric power in (Ga,Mn)As and GaAs:Be as a function of hole density changed by irradiation with high-energy Ne^+ ions [after Mayer *et al.* (Ref. 18)]. Lines are calculated using the standard six-band model and GaAs parameters (Ref. 16) assuming that either ionized impurity or acoustic phonon scattering dominates. The actual value of thermopower S should lie between lines obtained for heavy and light hole bands in each case.

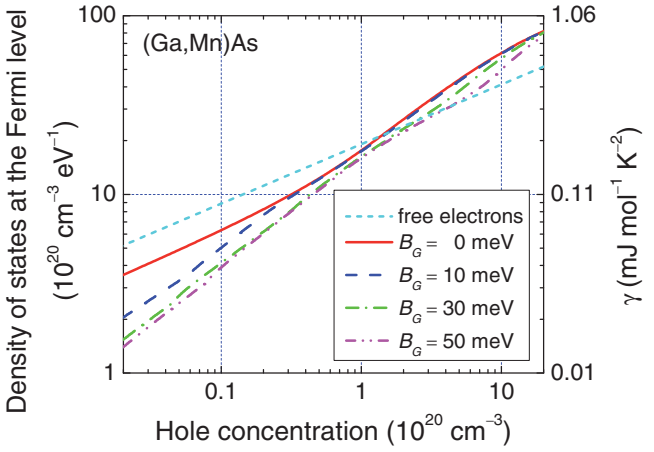

FIG. 5. (Color online) Thermodynamic density of states as a function of the hole concentration in (Ga,Mn)As for various values of the parameter B_G characterizing spin splitting of the hole subbands. The corresponding values of the Sommerfeld electronic specific-heat coefficient γ are shown on the right axis.

which shows that the MIT actually has little effect on the thermodynamic DOS, as could be expected for the Anderson-Mott localization.

We expect that acoustic phonon scattering, for which $r = -1/2$, is relevant in GaAs:Be at 300 K. In contrast, in the case of (Ga,Mn)As, owing to the proximity to the MIT, the mobility is expected to increase with the carrier energy. At the same time, an additional compensation by interstitial Mn makes ionized impurity scattering, for which $r = 3/2$, more significant. Thus, we can conclude that the data for (Ga,Mn)As fall in the range expected for hole transport in the GaAs valence band.

V. LOW-TEMPERATURE ELECTRONIC SPECIFIC HEAT

In doped semiconductors on the metal side of the Anderson-Mott localization, the magnitude of the Fermi-liquid parameter is relatively small, $r_s = (4\pi p/3)^{-1/3}/a_B^* \lesssim 2.4$, so that Landau's renormalization of the specific heat and thermodynamic DOS by carrier-carrier interactions is of minor quantitative importance. Hence, we compute the electronic specific heat in the low-temperature limit according to $C_V = \gamma T$, with the Sommerfeld constant

$$\gamma = \frac{\pi^2}{3} k_B^2 \rho_F, \tag{19}$$

where $\rho_F = \partial p/\partial E_F$ is determined for holes in the valence band with no carrier-carrier interactions taken into account. In Fig. 5 we plot the dependence of ρ_F and γ on the hole concentration p for (Ga,Mn)As with various values of the spin splitting parameter $B_G = A_F \beta M/(6g\mu_B)$. The numerical values for samples A and B of Yuldashev *et al.* are 0.37 and 0.39 mJ mol^{-1} K^{-2}, respectively, hence the values of γT_C are small compared to the critical anomaly of the specific heat, validating the results shown in Fig. 1.

VI. LOW-TEMPERATURE CONDUCTIVITY

Near the MIT, temperature and magnetic field dependencies of conductivity $\sigma(T,H)$ are determined by quantum phenomena specific to Anderson-Mott localization.[4,41] These striking effects result from single-particle interferences of scattered waves and/or from scattering-driven interferences of carrier-carrier interaction amplitudes. It was suggested within this framework that the magnitude of conductivity changes in (Ga,Mn)As at low temperatures points to the value of DOS actually expected for the GaAs valence band.[26] More recently, Neumaier *et al.*[19] carried out comprehensive studies of conductance in various dimensionality structures of ferromagnetic (Ga,Mn)As at subkelvin temperatures. Since the external magnetic field has no effect on $\sigma(T)$ in this regime,[42] the single-particle Anderson localization term, destroyed presumably by the demagnetizing field, does *not* contribute to $\sigma(T)$ in this ferromagnetic semiconductor below 1 K. At the same time, the study of $\sigma(T)$ at the dimensional crossover 2D $\rightarrow$ 3D allowed one to determine the magnitude of the diffusion coefficient $D = \sigma/(e^2\rho_F)$.[19] In this way the value of DOS at the Fermi level for charge excitations ρ_F was determined and found to be slightly smaller than the one expected for the GaAs valence band.[16,19]

To supplement the above analysis, we consider $\sigma(T)$ within the universality class for which the transport proceeds in two subbands, whose splitting is much larger than k_BT but much smaller than the Fermi energy. Furthermore, we make use of the value of the Landau parameter describing the correlation-induced enhancement of the carrier spin susceptibility and the Curie temperature $A_F = 1.2$.[15,32,33] Our goal is to describe the magnitudes of parameters a characterizing the rate of change of $\sigma(T)$ displayed in Fig. 1 of Ref. 19 for various dimensionality systems. In the two-dimensional (2D) case $a = F_{2D}e^2/(\pi ht)$, where according to theory developed for a simple isotropic band, in the

presence of a sizable spin splitting, $F^{2D} = (1 - F/4)\ln 10$, where $F = 2(A_F - 1)$.[4,41] The theoretical value obtained for the sample thickness $t = 42$ nm, $a = 1.6 \times 10^7 e^2/h$ m is seen to be in good agreement with the experimental finding $1.8 \times 10^7 e^2/h$ m.[19]

We now apply the same procedure to the one-dimensional (1D) and three-dimensional (3D) cases, $d = 1$ and 3,[19] where $a \sim D^{1-d/2}$. Here, $F^{1D} = 1.37$ and $F^{3D} = 1.04$ are anticipated theoretically for $F = 0.4$.[4] For the experimental values $\sigma = 6.95 \times 10^8 e^2/h$ m and $3.95 \times 10^8 e^2/h$ m as well as for $\rho_F = 1.98 \times 10^{46}$ 1/J m^3 and 1.45×10^{46} 1/J m^3, as expected for the disorder-free (Ga,Mn)As valence band at the hole concentrations in question,[16,19] we obtain $a = -3.1 \times 10^7 (e^2/h)(\mathrm{K}^{1/2}/\mathrm{m})$ for $d = 1$ and $0.93 \times 10^7 e^2/(h\,\mathrm{m\,K}^{1/2})$ for $d = 3$, which are close to the experimental values $a = -2.5 \times 10^7\,(e^2/h)(\mathrm{K}^{1/2}/\mathrm{m})$ and $1.5 \times 10^7 e^2/(h\,\mathrm{m\,K}^{1/2})$, respectively, shown in Figs. 1(b) and 1(d) of Neumaier *et al.*[19] As seen, this approach suggests a slightly higher DOS compared to that expected for the GaAs-like valence band.

It is worth noting that other authors,[43,44] analyzing $\sigma(T)$ up to 4 K, found that $\sigma(T) = \sigma_0 + AT^\alpha$, where $\alpha = 1/3$. This dependence was interpreted in terms of a renormalization-group equation[4] applicable close to the MIT, where $\sigma_0 < AT^\alpha$ and then $1/3 \lesssim \alpha \lesssim 1/2$ in the 3D case.[41,45] Furthermore, the apparent value of α can be reduced above 1 K by a crossover to the regime, where the effect of scattering by magnetic excitations onto quantum corrections to conductivity becomes significant.[26]

VII. CONCLUSIONS

We have considered the thermodynamic and thermoelectric properties of (Ga,Mn)As including the critical behavior of specific heat and magnetization, high-temperature thermoelectric power, low-temperature electronic specific heat, and low-temperature conductivity. The available experimental data[17–19] are consistent with the *p-d* Zener model[15] in which the carriers reside in a GaAs-like valence band. In particular, the critical behavior of specific heat[17] can be reasonably well described assuming Gaussian fluctuations of magnetization. The magnitudes of the thermoelectric power at room temperature[18] are consistent with the theoretical results for scattering mechanisms expected to limit the magnitude of hole mobility. The data for low-temperature electronic specific heat are provided in order to stimulate corresponding experimental investigations. Finally, the magnitudes of density of states obtained assuming the valence-band model are supported by the temperature dependence of conductivity at subkelvin temperatures, determined by disorder-modified carrier-carrier interaction effects, significant near the metal-insulator transition.[19] The proximity to this transition also results in a reduction of the portion of Mn spins which are aligned by itinerant holes and, thus, contribute to the long-range ferromagnetic order.

It would be interesting to find out whether the experimental results discussed here could also be interpreted within the impurity-band models. We note that no quantitative theories of pertinent ferromagnetic characteristics, such as Curie temperature, magnetic anisotropy, or exchange stiffness, have been developed within particular variants of those models.[46] Moreover, according to recent studies,[47] the impurity-band models appear to be in conflict with the microscopic theory of the (Ga,Mn)As band structure for the Mn concentrations relevant for ferromagnetism. Nevertheless, the premise that the holes relevant for ferromagnetism reside in a narrow impuritylike band, detached from the valence band, has been systematically proposed in order to explain various nonstandard findings of optical[10,48–50] or transport[9,11,51] studies on (Ga,Mn)As. A typical enhancement of the density of states and effective mass over the free electron value (and thus over the hole mass in *p*-type GaAs; see Fig. 5) proposed in these works varies between 10 (Ref. 48) and 30 (Ref. 11). Since the magnitude of thermoelectric power at high temperatures is directly proportional to the density of states ρ_F, whereas the temperature dependence of low-temperature conductivity to the square root or the inverse square root of ρ_F in 3D and 1D, respectively, we conclude that the experimental results discussed in Secs. IV and VI are inconsistent with impurity-band models of electronic states in ferromagnetic (Ga,Mn)As. Furthermore, we note that the agreement between experimental and theoretical magnitudes of critical specific heat (Sec. II) is obtained if the structure of the GaAs valence band is carefully taken into account. No such comparison can be performed for the case of impurity-band models for which no theoretical predictions concerning the magnitude of the spin-wave stiffness D_{nor} are available.

At the same time, it has been demonstrated that a number of puzzling features of (Ga,Mn)As optical properties can be explained within the valence-band picture, particularly the infrared conductivity[52] and magnetic circular dichroism.[16,52,53] Nevertheless, we would like to emphasize that in many cases, the proximity of the Anderson-Mott localization may actually account for a nonstandard behavior of transport and optical phenomena in carrier-controlled ferromagnetic semiconductors.[26] These phenomena may not be easily tackled theoretically as the current theories of the Anderson-Mott transition rather than predicting the absolute values of static or dynamic conductivities, provide information only on critical exponents as well as on the dependencies of quantum corrections to conductivity on the magnetic field, frequency, and temperature.[4,5,45] Since, according to the results presented in Fig. 5, the effective mass of holes increases with the hole density, the region perturbed significantly by quantum localization effects extends deeply into the metallic phase in (Ga,Mn)As.

We conclude that there is no experimental evidence for the enhanced density of states at the Fermi level, expected within the impurity-band models of ferromagnetism in (Ga,Mn)As. Instead, however, the properties of this ferromagnet are strongly affected by hole localization effects.

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