Energy spectrum and parameters of deep impurity level in $Pb_{1-x}Ge_xTe$ alloys doped with Yb

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In this paper, the galvanomagnetic properties $(4.2 \le T \le 300 \text{ K})$ of $Pb_{1-x}Ge_xTe(x \le 0.04)$ alloys doped with ytterbium were investigated under both atmospheric pressure and hydrostatic compression up to 12 kbar. The variation of the energy diagram of $Pb_{1-x}Ge_xTe\langle Yb \rangle$ under variation of the Ge concentration and under pressure were proposed. It was shown that change of energy spectrum under pressure leads to the insulator-metal transition due to the redistribution of electrons between the valence band and the Yb-induced deep level. By comparing theoretical and experimental dependences of Fermi energy and free hole concentration on pressure the main parameters of Yb-induced deep level were determined. [S0163-1829(99)09919-1]

I. INTRODUCTION

It has been known that doping narrow gap $A^{IV}B^{VI}$ semiconductors with mixed valence impurities (In, Ga, Tl) leads to the emergence of deep impurity levels in the energy spectrum of semiconductors and the principal modification of its electrical and optical properties.¹⁻³ Impurity states possess donor-acceptor properties and if the impurity concentration is rather high (≈ 1 at. %) the Fermi energy is pinned within the impurity level partially filled with electrons. Due to a high density of the impurity states the Fermi-energy position is independent of the concentration of other impurities, intrinsic and radiation-induced defects and depends on the energy position of the deep level only.⁴⁻⁶ The impurity levels can be situated in the forbidden gap as well as in the allowed bands depending on the alloy composition, the temperature, the pressure, and the magnetic field. If the deep impurity level lies in the forbidden gap, it is possible to obtain solid solutions with an extremely low-charge carrier concentration close to the intrinsic one.^{7,8}

After exciting doped alloys by some external factors, for example, infrared illumination, or applying a quantizing magnetic field, all relaxation processes connected with capturing nonequilibrium charge carriers by impurity-induced centers exhibit the long-term character usually explained by the Jahn-Teller nature of the defect impurity centers.^{9–11} Accordingly, capturing electrons in the defect center leads to a change of its lattice surrounding and to a decrease in the electronic energy of the defect state. Thus, the states with various number of captured electrons become separated from each other by autolocalizing barriers, which drastically reduces the rate of carrier transitions between the localized and band states. It results, in particular, in the anomalously high-impurity photosensitivity below a critical temperature T_c and the persistent photoconductivity effect at temperatures $T \ll T_c$ after the infrared illumination of the doped alloys. For $Pb_{1-x}Sn_xTe$ alloys doped with In, T_c \approx 20 K whereas for PbTe doped with Ga the critical temperature reaches the values about 80 K.^{12,13}

It has recently been found that ytterbium in $Pb_{1-x}Ge_xTe$ $(x \le 0.06)$ alloys also exhibits a mixed valence. It has been shown that *p*-type alloys are paramagnetic due to selfionization of Yb ions $(Yb^{2+} \rightarrow Yb^{3+} + e_{band})$ and transition of these ions to the magnetically active Yb^{3+} state $(4f, {}^{13}s=1/2)$.¹⁴ By analogy with the behavior of wellknown mixed valence impurities one can assume that the ytterbium impurity can form deep Jahn-Teller electronic states in the energy spectrum of $Pb_{1-x}Ge_xTe$ alloys. The investigations of the galvanomagnetic and photoelectric properties of the alloys confirm this assumption. It has been revealed that in Pb_{1-x}Ge_xTe ($x \le 0.06$) the Yb-induced deep level is situated near the top of the valence band either in the forbidden gap or in the valence band.^{15,16} High photosensitivity of the alloys at temperatures $T < T_c \approx 30$ K and the persistent photoconductivity effect at T=4.2 K have also been established.¹⁷ However, neither the exact position of the Yb-induced deep level in alloys with various Ge concentration nor the main parameters of this level have been determined up to now.

The investigation of the galvanomagnetic properties of the doped alloys based on PbTe under pressure and in quantizing magnetic fields is one of the most promising ways of obtaining information about the energy spectrum and parameters of impurity levels in these alloys.^{1,2} The variation of the mutual arrangement of the deep level and the band edges under the condition of the Fermi-level pinning makes it possible to observe redistribution of electrons between localized and band states and insulator-metal transitions induced by a variation of pressure or a magnetic field. In this case, the investigation of the pressure or magnetic-field dependence of the free-carrier concentration during the insulator-metal or metal-insulator transition allows one to obtain the unique information about the energy spectra of band and localized electrons, to estimate the impurity level width and to restore its density-of-state function.

In the present paper, the galvanomagnetic properties of $Pb_{1-x}Ge_xTe$ doped with Yb were investigated under both

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TABLE I. Parameters of the investigated samples $Pb_{1-x}Ge_xTe\langle Yb \rangle$ at T=4.2 K.

Sample	x	$C_{\rm Yb}$, at. %	Туре	R_H , cm ³ /C	$ ho,~\Omega~{ m cm}$	$p, {\rm cm}^{-3}$	μ_H , cm ² /Vs
Yb15-1	0.0085	0.75	р	1.8×10^{3}	7.2×10^{-1}	2.8×10^{15}	2.5×10^{3}
Yb15-2	0.0085	0.75	р	9.2×10^{2}	3.4×10^{0}	5.5×10^{15}	2.7×10^{2}
Yb60-1	0.038	0.86	р	$> 5 \times 10^{7}$	$> 6 \times 10^{4}$	$< 10^{11}$	$> 8 \times 10^{2}$
Yb60-2	0.038	0.86	р	$> 2 \times 10^{6}$	$> 8 \times 10^{2}$	$< 10^{12}$	$> 3 \times 10^{3}$

atmospheric pressure and hydrostatic compression. It was assumed that the variation of Ge content and the application of hydrostatic compression would make it possible to change the position of Yb-induced level with respect to the top of the valence band, to change the energy spectrum of the alloys by varying the Ge concentration and applying pressure and to determine the main parameters of Yb-induced deep states.

II. EXPERIMENTAL DETAILS

Pb_{1-x}Ge_xTe ($x \le 0.04$) doped with Yb single crystals were grown by a modified Bridgeman method described in Ref. 15. Concentrations of germanium and ytterbium in the alloys was checked by the energy dispersive x-ray flouorescence analysis.¹⁴ The parameters of the investigated samples at atmospheric pressure and T=4.2 K are presented in Table I.

In each sample the temperature dependence of resistivity ρ and the Hall constant $R_{\rm H}$ (4.2 \leq T \leq 300 K, B \leq 0.1 T) were measured in the chamber shielded from the external background illumination. Then the sample Yb60-1 with the Ge content x = 0.038 was investigated under high hydrostatic pressure. Pressures up to 12 kbar were obtained in the beryllium bronze chamber with kerosene-oil-pentane pressure transmitting medium. The pressure was applied at room temperature and the chamber was allowed to cool slowly in a thermal contact with either liquid nitrogen or liquid helium. The pressure calibration was carried out at helium temperature by measuring the superconducting transition temperature in high-purity tin as a function of pressure in the chamber. The temperature dependences of the resistivity and the Hall constant in a weak-magnetic field were measured at every value of pressure. Besides, at pressures P > 4 kbar the Shubnikov-de Haas effect and the magnetic-field dependences of the Hall constant in strong-magnetic fields (T =4.2 K, $B \le 7$ T) were studied.

III. RESULTS AND DISCUSSION

A. Galvanomagnetic properties and the energy spectrum of $Pb_{1-x}Ge_xTe\langle Yb \rangle$ alloys

The investigation of the galvanomagnetic effects in $Pb_{1-x}Ge_xTe\langle Yb \rangle$ alloys shows the strong dependence of its properties upon the alloy composition. For the alloys with the Ge content x=0.0085, rather low values of the resistivity and the Hall constant and therefore, high-carrier concentration at T=4.2 K are typical, while the alloys with the Ge content x=0.038 are high resistive and characterized by low (close to the intrinsic) values of the charge-carrier concentra-

tion (see Table I). The temperature dependences of the resistivity and the Hall constant measured when the samples were shielded from the external background illumination are shown in Fig. 1. The important feature of these curves is the presence of the low-temperature activation region of impurity conductivity, which is indicative of the existence of the impurity level in the forbidden gap of the investigated alloys. From the slope of the low-temperature activation region in the $\rho(1/T)$ dependence the activation energy of the Yb-induced level were determined. It was found that its value $\Delta E_{\rm Yb} = E_{\rm Yb} - E_v \approx 2$ meV in the alloys with the Ge content x = 0.0085 and $\Delta E_{\rm Yb} \approx 22$ meV for the alloys with x = 0.038.

In the alloys with a lower forbidden gap (x=0.0085) the temperature dependences of the resistivity and the Hall constant also exhibit a high-temperature activation region (see curves 2 and 2' in Fig. 1) due to the intrinsic ionization of electrons. Unfortunately, this activation region is unsuitable for determination of the gap because at the experimental temperatures it was only the beginning of the intrinsic ionization region that was observed.

The obtained values of the activation energy ΔE_{Yb} together with the well-known data on the energy spectrum of $Pb_{1-x}Ge_xTe$ and $Pb_{1-x}Yb_xTe$ alloys^{18,19} allow us to propose the variation of the energy diagram of $Pb_{1-x}Ge_xTe$ doped with Yb under variation of the Ge content (Fig. 2). It is known that an increase in the Ge content leads to an increase



FIG. 1. Temperature dependence of the Hall constant (1, 2) and resistivity (1', 2') in $Pb_{1-x}Ge_xTe\langle Yb \rangle$. 1,1'-x=0.038, 2,2'-x = 0.0085.



FIG. 2. Change of the energy spectrum of $Pb_{1-x}Ge_xTe\langle Yb \rangle$ under the variation of the Ge content x at T=4.2 K.

in the Pb_{1-x}Ge_xTe forbidden band with the rate $dE_g/dx \approx 20 \text{ meV/at.}\%$ (Ref. 18) and doping with Yb leads to an increase of the Pb_{1-x}Yb_xTe forbidden gap with the rate $dE_g/dx = 25 \pm 5 \text{ meV/at.}\%$.¹⁹ So one can assume that in PbTe the Yb-induced deep level should be situated in the valence band near its top. Upon an increase in the Ge content the Yb level moves to the top of the valence band, intersects it at $x \approx 0.01$ and enters the forbidden gap. Assuming that the position of the Yb-induced level with respect to the valence-band top depends linearly upon the alloy composition, the rate of the activation energy increase with increasing Ge content can be estimated as $d\Delta E_{Yb}/dx \approx 7 \text{ meV/at.}\%$.

B. The insulator-metal transition induced by pressure in $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (x = 0.038)

Application of hydrostatic pressure leads to significant changes in the galvanomagnetic properties of the $Pb_{1-x}Ge_{x}Te\langle Yb \rangle$ (x=0.038) alloy. In the pressure range P <5 kbar the resistivity and the Hall constant at low temperatures decrease sharply by about six orders of magnitude (Figs. 3 and 4). At higher pressures, these parameters decrease slower and attain the values of $10^{-4} \Omega$ cm and $1 \text{ cm}^3/\text{C}$, respectively. The character of the temperature dependences of the resistivity and Hall constant also changes under pressure. As the pressure increases, the slope of the activation region in the $\rho(1/T)$ dependence as well as in the $R_H(1/T)$ dependence decrease approaching zero at some critical pressure P^* . As the pressure increases further, the temperature dependence of the resistivity assumes a metallic character typical of the undoped PbTe-based alloys. The temperature dependence of the Hall constant at $P > P^*$, however, exibits an unusual feature: the Hall constant monotonically increases with increasing temperature.

The investigation of transverse magnetoresistance of the sample shows the emergence of the Shubnikov-de Haas oscillations at pressures about P^* (Fig. 5). Since the oscillation frequency is determined by the free-carrier concentration in the sample, the comparison of the oscillation curves measured at various pressures gives information about changes in the carrier concentration under pressure. It was found that as the pressure increases, the oscillation frequency mono-



FIG. 3. Temperature dependence of the resistivity in $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (*x*=0.038) under various pressures. *P* (kbar): 1-0, 2-1.6, 3-2.3, 4-3.3, 5-3.9, 6-4.4, 7-6.8, 8-8.7, 9-12.0.

tonically increases, at rather high pressures (about 12 kbar) the oscillation curves reveal the so-called beats, connected with the presence of more than one characteristic frequency. The existence of several characteristic frequencies can be explained under the assumption that there are several types of charge carriers in the metallic phase of the alloy.

In this case, the Hall constant vs the magnetic-field dependence should exhibit some features of, for example, the nonlinear Hall voltage vs the magnetic-field dependence or the reversal of the Hall voltage sign with increasing pressure. However, careful measurements of the Hall constant vs magnetic-field dependence shows that this dependence does not exhibit any nonlinearity. Besides, the beats can be ex-



FIG. 4. Temperature dependence of the Hall constant in $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (x=0.038) under various pressures. *P* (kbar): 1-0, 2-1.6, 3-2.3, 4-3.3, 5-3.9, 6-4.4, 7-6.8, 8-8.7, 9-12.0.



FIG. 5. Transverse magnetoresistance oscillations in $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (*x*=0.038) under various pressures. *P* (kbar): 1-4.7, 2-6.8, 3-8.7, 4-12.0.

plained by the splitting of the four equivalent valleys in L points of the Brillouin zone to a couple of singlet and triplet valleys due to the low-temperature structural transition from the cubic to the rombohedral phase.²⁰ In order to prove this assumption further experiments with exact control of the orientation of the samples relative to the direction of the magnetic field are required.

The experimental data, in particular, the decrease of the slopes of the activation regions in the $\rho(1/T)$ and $R_H(1/T)$ dependences, and the change in the character of the temperature dependences of the resistivity and the Hall constant with increasing pressure are indicative of the transition of the investigated sample from the insulator to the metallic phase under pressure. The emergence of the Shubnikov–de Haas oscillations and the increase of its frequency in the metallic phase confirm this conclusion and imply the increase of the free-carrier concentration with a further increase in pressure.

C. Variation of the energy spectrum of $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (x=0.038) under pressure

The experimental results obtained allow us to propose the variation of the energy diagram of $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (x =0.038) alloy under pressure (Fig. 6). Under atmospheric pressure the Yb-induced deep level is situated in the forbidden gap near the top of the valence band. Under pressure the width of the forbidden gap at the L point of the Brillouin zone in $Pb_{1-x}Ge_xTe$ alloys decreases. We assume that the pressure coefficient of the forbidden gap dE_g/dP in the investigated alloys is the same as that of PbTe and equals 7.4 meV/kbar.21 With decreasing forbidden gap the deep level approaches the valence-band edge. In order to determine the rate of the Yb level moving with respect to the top of the valence band the values of the activation energy $\Delta E_{
m Yb}$ were calculated from the slopes of the activation regions in the $\rho(1/T)$ dependences (Fig. 3) and the activation energy vs. pressure dependence was constructed (Fig. 7). It was found that with increasing pressure the $\Delta E_{\rm Yb}$ decreases almost lin-



FIG. 6. Change of the energy spectrum of $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (x=0.038) under pressure at T=4.2 K.

early with the rate $d\Delta E_{\rm Yb}/dP \approx 5$ meV/kbar and becomes zero at a critical pressure $P^* \approx 4.5$ kbar.

At about the same pressure the Yb-induced level intersects the top of the valence band and the insulator-metal transition occurs, which is connected with the emergence of free holes due to the flow of electrons from the valence band to the free states in the deep level (Fig. 6). A further increase in pressure results in the Yb-induced level moving deeper into the valence band and a further increase in the free-hole concentration due to the pinning of the Fermi level by the Yb-induced states.

The effect of Fermi-level pinning in the metallic phase also allows us to explain the unusual character of the temperature dependences of the Hall constant at $P > P^*$ mentioned above (see curves 6–9 in Fig. 4). The observed increase of the Hall constant can be accounted for by the movement of the Yb-induced level to the top of the valence



FIG. 7. Dependence of the activation energy of Yb-induced deep level in $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (x=0.038) on pressure.



FIG. 8. Dependence of the free-hole concentration in $Pb_{1-x}Ge_xTe\langle Yb\rangle$ (x=0.038) on pressure. Solid curves were calculated in terms of the model given by Eqs. (1)–(4) with the bandwidth $\sigma=7$ meV and k: 1-0, 2-1/8, 3-3/8.

band with increasing temperature. This movement makes electrons flow from the impurity level to the valence band resulting in decreasing the free-hole concentration in the valence band of the alloys.

In order to estimate the main parameters of the Ybinduced level in the frame of proposed model the pressure dependences of the free-hole concentration and the Fermilevel position in the metallic phase were constructed. The experimental values of the free-hole concentration (solid squares in Fig. 8) were calculated from the values of the Hall constant at T=4.2 K. The free-hole concentration estimated from the period of the Shubnikov–de Haas oscillations coincides within 10% with that obtained from the Hall-effect data. The position of the Fermi level E_F (solid circles in Fig. 9) was calculated from the values of the free-hole concentration, using Kane's energy-momentum relation with the values of the parameters typical of $Pb_{1-x}Sn_xTe$ ($x \approx 0.2$) presented in Ref. 22.

It is interesting to note (Fig. 9) that the Fermi level enters the valence band at a significantly lower pressure than the critical pressure P^* , at which the middle of the impurity band should enter the valence band. Thus, in the finite pressure range the activation of electrons from the valence band to the Yb-induced level and the existence of free holes in the valence band at helium temperatures are observed simultaneously. This fact allows us to conclude that the Yb-induced level (impurity band) should have a considerable width (5–10 meV) and under pressures up to the insulator-metal transition point should be less than half filled with electrons (Fig. 6). In any other case the emergence of free holes must occur exactly at the pressure P^* , when the activation energy of the Yb-induced level becomes zero.

Moreover, in the whole investigated pressure range the pressure dependence of the Fermi level position lies under the dashed line, which corresponds to the position of the middle of the Yb-induced band under pressure (Fig. 9).



FIG. 9. Dependence of the activation energy of the Yb-induced level in the insulator phase and Fermi energy in the metallic phase in $Pb_{1-x}Ge_xTe\langle Yb \rangle$ (x=0.038) on pressure. Solid curves were calculated in terms of the model given by Eqs. (1)–(4) with the bandwidth $\sigma=7$ meV and k: 1-0, 2-1/8, 3-3/8.

Thus, one can assume that up to the maximum pressure the impurity band remains less than half filled with electrons.

The sharp increase of the free-hole concentration immediately after the insulator-metal transition and the lack of the saturation of the free-hole concentration obviously imply the extremely high concentration of the empty states in the impurity band. Taking into account that at the maximum pressure P = 12 kbar the concentration of free holes was $p \approx 3$ $\times 10^{18}$ cm⁻³ and in the whole pressure range the impurity level was less than half-filled with electrons, the density of states in this level can be estimated as $N_{\rm Yb} > 10^{19}$ cm⁻³.

D. Parameters of deep Yb-induced states in $Pb_{1-x}Ge_xTe$ alloys

In order to determine quantitatively the main parameters of the Yb-impurity band the theoretical and experimental pressure dependences of the free-hole concentration and the Fermi-level position were compared. The theoretical dependences were calculated using the model of the energy spectrum of the alloy under pressure mentioned above (Fig. 6). According to this model, the flow of electrons from the valence band to the impurity band occurs in such a way that for all the values of pressure *P* total free-hole concentration in the valence band p(P) and the density of empty states in the Yb-induced band $p_{Yb}(P)$ are equal to the initial concentration of empty states in the impurity band under atmospheric pressure $p_{Yb}(0)$

$$p_{\rm Yb}(0) = p(P) + p_{\rm Yb}(P).$$
 (1)

The concentrations of the empty states in the Yb-induced band were calculated assuming that the density-of-states function in the Yb-induced band $g_{Yb}(E)$ can be approximated by the Gaussian-type curve and under atmospheric pressure the impurity band is partially filled with electrons

$$p_{\rm Yb}(P) = \int_{E_F}^{\infty} g_{\rm Yb}(E) dE, \qquad (2)$$

$$g_{\rm Yb}(E) = \frac{N_{\rm Yb}}{\sigma \sqrt{2\pi}} \exp\left[-\frac{(E - E_{\rm Yb})^2}{2\sigma^2}\right],\tag{3}$$

$$p_{\rm Yb}(0) = N_{\rm Yb}(1-k),$$
 (4)

where σ is the width of the Yb-induced band, $N_{\rm Yb}$ is the total capacity of the Yb-induced band and k is the initial occupancy of the band with electrons under the atmospheric pressure.

Thus, in Eqs. (1)–(4) there are three variable parameters $N_{\rm Yb}$, σ , and k. The preliminary calculations showed that in this case there exist several sets of parameters, which provide coincidence between the experimental and theoretical data. In order to avoid such a situation one parameter — $N_{\rm Yb}$ was independently estimated and fixed. Taking into account our estimation (see above), $N_{\rm Yb} > 10^{19}$ cm⁻³, we supposed that every impurity atom takes part in the formation of impurity states and gives one electronic state to the impurity band. The total capacity obtained in such a way was $N_{\rm Yb} \approx 10^{20}$ cm⁻³.

So in our calculations of the theoretical pressure dependences of the free-hole concentration and Fermi-level position only two parameters (the width of the Yb-induced band σ and the occupancy of this band with electrons at atmospheric pressure k) were varied. It was found that the variation of k influences mostly the first region of the p(P) dependence characterized by a sharp increase in the free-hole concentration, while the variation of σ influences mostly the second region of this dependence characterized by a slow increase in the free-hole concentration (Fig. 8). With increase in k the insulator-metal transition point shifts toward higher values of pressure and a sharper increase in the free-hole concentration in the metallic phase takes place. The increase of σ slightly moves the insulator-metal transition point to a lower pressure, but at the same time leads to a significantly slower increase of the free-hole concentration in the second region of the p(P) dependence. One can see that in this case it is only one set of parameters that would provide the best fit of the theoretical and experimental curves. These parameters were found to be $k \approx 1/8$ and $\sigma \approx 7$ meV (curve 2 in Figs. 8 and 9).

In order to estimate the reliability of the parameters obtained the above calculations were also carried out under changes of σ and k near their average values (curves 1 and 3 in Figs. 8 and 9). Calculations show that both parameters possess a satisfactory stability to their variations. Taking into account the errors obtained in the electron occupancy of the band and in the impurity band width, we can finally assume these parameters to be $k = 1/8 \pm 1/16$ and $\sigma = 7 \pm 3$ meV, respectively.

It is also interesting to note that in the investigated pressure range in Fig. 9 all the calculated curves lie under the dashed line, indicating the position of the middle of the impurity band. It means that regardless of the exact value of the parameter *k* the impurity band is less than half filled with electrons due to the extremely high density of the impurity states. According to our estimations, the impurity band becomes half filled with electrons only at the pressure of $P \approx 50$ kbar.

IV. CONCLUSIONS

The experimental results obtained allow us to conclude the following.

(1) Doping of the $Pb_{1-x}Ge_xTe$ ($x \le 0.04$) alloys with Yb leads to the appearance of Yb-induced deep level (impurity band); the energy position of this level with respect to the valence-band top is strongly dependent on the alloy composition. In PbTe, this level is situated in the valence band, with an increase of the Ge content in the alloys the impurity level moves to the top of the valence band, intersects it at $x \approx 0.01$ and goes deep into the forbidden gap with the rate $d\Delta E_{Yb}/dx \approx 7$ meV/at. %.

(2) Hydrostatic compression of $Pb_{1-x}Ge_xTe$ (x=0.038) (Yb) leads to the decrease of the activation energy of the Yb-induced level with the rate $d\Delta E_{Yb}/dP \approx 5$ meV/kbar and insulator-metal transition due to the pinning of Fermi level by the impurity band and redistribution of electrons between the valence band and the Yb level under pressure.

(3) In the investigated alloys the Yb-induced impurity band has an extremely high capacity $N_{Yb} \approx 10^{20}$ cm⁻³ and a considerable width $\sigma = 7 \pm 3$ meV. At pressures P<12 kbar this band is less than half filled with electrons and the initial occupancy of the band under atmospheric pressure $k \approx 1/8$. The density-of-state function in the impurity band can be satisfactorily described by the Gaussian-type curve.

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