

Spin-polarization enhancement of dilute ^3He - ^4He solutions through porous media

A. Ohi and T. Nakayama

Department of Applied Physics, Hokkaido University, Sapporo 060, Japan

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The flow of ^3He quasiparticles in a dilute ^3He - ^4He solution through a porous medium is investigated theoretically on the basis of the percolation model. It is pointed out that the chemical potential μ of ^3He quasiparticles is related to the percolation density p of the porous medium. This allows us to control the flow rate for polarized ^3He quasiparticles through the porous medium by magnetic fields. It is suggested that highly spin-polarized ^3He - ^4He solutions are achieved by this mechanism at millikelvin temperatures.

One of the important problems in the field of low-temperature physics concerns the experimental achievement of spin-polarized quantum systems such as the $\text{H}\uparrow$, $\text{D}\uparrow$, or $^3\text{He}\uparrow$ phase, where completely new phenomena could be expected due to the polarization of nuclear spins. Many ideas to produce these systems have been proposed so far.¹⁻³ It is, however, difficult to reach the degenerate Fermi-fluid regime in gases, since the condensation to the liquid states generally sets in before the effects of quantum degeneracy become important. The most promising system studying the degenerate case is a dilute $^3\text{He}\uparrow$ - ^4He solution which provides unique insights about many effects common to different types of degenerate Fermi systems,⁴ including transport properties.⁵ It is sure that dilute solutions can be polarized almost perfectly by the application of a large magnetic field. This brute force method is, however, applicable to the case of very low concentration of ^3He in HeII of 0.01% because magnetic fields at hand are limited below several ten Tesla. Mullin⁶ has suggested that degenerate $^3\text{He}\uparrow$ systems are produced by using a Nuclepore filter. The idea is based on the fact that, in polarized gas, \uparrow and \downarrow spins have different Fermi velocities so that the dominant spin species diffuse down a channel at a faster rate.

In this Rapid Communication, we propose a novel mechanism utilizing a porous medium by which a highly spin-polarized dilute ^3He - ^4He solution with a *high concentration* of ^3He can be achieved. The schematic diagram of our system is given in Fig. 1, in which a dilute ^3He - ^4He solution is partially polarized in one chamber and polarized ^3He quasiparticles flow through the porous

medium into a second chamber filled by HeII . The porous medium is modeled as a percolating network consisting of spherical pores (diameter a) and cylindrical channels (length l and radius d with distribution) connecting pores.⁷ The zero-point energy of ^3He quasiparticles confined in a porous medium can be expressed as a function of sizes of pores or channels. For instance, the zero-point energy of ^3He quasiparticles in a cylindrical channel with the cross section πd^2 becomes $V_{\text{ch}} \approx \pi^2 \hbar^2 / 4m_3^* d^2$, where m_3^* is the effective mass of ^3He quasiparticles in a dilute solution. It is crucial that the zero-point energies of ^3He quasiparticles in pores are lower than those in channels due to the condition $a \geq 2d$. As a result, when ^3He quasiparticles diffuse in the porous medium, they feel the random potential barrier V_{ch} corresponding to the random distribution of channel radii d 's. Only ^3He quasiparticles with kinetic energy $E_{k\sigma}$ (σ is the spin index) higher than the barrier height V_{ch} can diffuse into the adjacent pores (the bond is opened up). This means that the percolation density p for diffusive ^3He quasiparticles is related to their energy. We can neglect the tunneling effect of ^3He quasiparticles through narrow channels because of its very low tunneling probability. Thus the bond-percolation density $p(E_{k\sigma})$ for ^3He quasiparticles with the energy $E_{k\sigma}$ is expressed as

$$p(E_{k\sigma}) = p_{st} \int_0^{E_{k\sigma}} F(V_{\text{ch}}) dV_{\text{ch}}, \quad (1)$$

where p_{st} is the bond-percolation density for the original porous network. The function $F(V_{\text{ch}})$ is the density of the distribution of potential barrier V_{ch} , which we determine from the inverted random void percolation model.^{8,9} In this model, the distribution function is expressed by $F(V_{\text{ch}}) = 3V_{\text{min}}(1 - V_{\text{min}}/V_{\text{ch}})^{1/2}/2V_{\text{ch}}^2$, where V_{min} is the bottom energy of potential wells. We can derive from Eq. (1) that the percolation density $p(E_{k\sigma})$ is expressed by $p(E_{k\sigma}) = p_{st}(1 - V_{\text{min}}/E_{k\sigma})^{3/2}$. It is important to note that ^3He quasiparticles with kinetic energy lower than some critical energy E_c , defined by $p(E_c) = p_c$, cannot diffuse in the porous medium. Here p_c is the critical percolation density.¹⁰

The diffusion constant of ^3He quasiparticles with the energy $E_{k\sigma}$ is given in the following general form:^{8,11}

$$D(E_{k\sigma}) \sim \frac{l^2}{\bar{\tau}(E_{k\sigma})} [p(E_{k\sigma}) - p_c]^{1-\beta}, \quad (2)$$

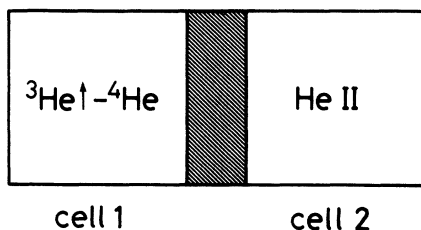


FIG. 1. Schematic diagram of our system. Cells 1 and 2 are separated by a thin porous medium with thickness L , where the cells 1 and 2 are filled initially with a partially polarized ^3He - ^4He solution and HeII , respectively.

where t and β are the exponents for the conductivity and for the order parameter (the probability belong to the infinite cluster) of the percolating network, respectively.¹² The channel length is given by l . The averaged hopping time of diffusive particle with $E_{k\sigma}$ is given by $1/\bar{\tau}(E_{k\sigma}) \sim E_{k\sigma}^{1/2}$.¹³

The diffusion constant for polarized ^3He quasiparticles in a dilute ^3He - ^4He solution with the chemical potential μ under a magnetic field B is defined by $\mathbf{J}_\sigma = -D_\sigma(\mu, B, T)\nabla n_\sigma$, where \mathbf{J}_σ and n_σ are the flow density and the number density of ^3He quasiparticles with the spin σ , respectively. The diffusion constant $D_\sigma(\mu, B, T)$ is obtained by averaging the diffusion constant $D(E_{k\sigma})$ for a diffusive ^3He quasiparticle with $E_{k\sigma}$ as, by using the number density $n(E_{k\sigma})$ of ^3He quasiparticles,

$$D_\sigma(\mu, B, T) = \frac{\int_{E_c}^{\infty} D(E_{k\sigma}) n(E_{k\sigma}) dE_{k\sigma}}{\int_{E_c}^{\infty} n(E_{k\sigma}) dE_{k\sigma}}, \quad (3)$$

where the number density $n(E_{k\sigma})$ is given by the product of the density of states (DOS) for ^3He quasiparticles and the Fermi distribution function expressed by $f(E_{k\sigma}) = 1/\{\exp[-(E_{k\sigma} - \gamma B\sigma - \mu)/k_B T] + 1\}$. Here γ is ^3He nuclear magnetic moment (0.778 mK/T) and $\sigma = \pm 1$. The density of states $\rho(E_{k\sigma})$ for polarized ^3He quasiparticles has a continuous spectrum for $E_{k\sigma} > E_c$ and a discrete one for $E_{k\sigma} < E_c$. This is due to the fact that ^3He quasiparticles for $E_{k\sigma} < E_c$ are localized in the porous medium. The DOS per unit volume for the continuous spectrum for $E_{k\sigma} > E_c$ is given by

$$\rho(E_{k\sigma}) = \frac{m_3^{*3/2}}{\sqrt{2}\pi^2\hbar^3} \sqrt{E_{k\sigma}}. \quad (4)$$

Using Eqs. (2)-(4), we find that the diffusion constant for zero magnetic field shows the universal critical behavior in the vicinity of $\mu = E_c$ as a function of the chemical potential μ as $D \sim (\mu - E_c)^{t-\beta}$. Under a magnetic field B , the diffusion constant for ^3He quasiparticles behaves as $D_\sigma \sim (\mu + \gamma B\sigma - E_c)^{t-\beta}$. This indicates that the mobility edge E_σ^m for ^3He quasiparticles is different for \uparrow and \downarrow spins under a magnetic field, where the mobility edge is defined as the minimum energy at which ^3He quasiparticles can flow, i.e., $E_\sigma^m = E_c - \gamma B\sigma$. Hereafter, ^3He quasiparticles with parallel spin to the magnetic field and with anti-parallel spin are denoted by $^3\text{He}\uparrow$ and $^3\text{He}\downarrow$, respectively. In the case of $\mu = E_c$, we see that $^3\text{He}\uparrow$ quasiparticles diffuse dominantly according to the power law on the magnetic field B as

$$D_\uparrow \sim B^{t-\beta}, \quad (5)$$

but $^3\text{He}\downarrow$ quasiparticles do not diffuse through the porous medium (see Fig. 2).

By taking the suitable values for our percolating system, the pore diameter $a = 50 \text{ \AA}$, the channel length $l = 50 \text{ \AA}$, and the percolation density of the porous medium $p_{st} = 0.5$, the mobility edge at zero magnetic field is calculated as $E_c = 70 \text{ mK}$. Using these values and the known values of the exponents t and β for the $d=3$ percolating network, the diffusion constants for \uparrow spins at $\mu = E_c$ are computed in Fig. 3 from Eq. (3) as a function of magnetic field B at various temperatures. We find that the diffusion

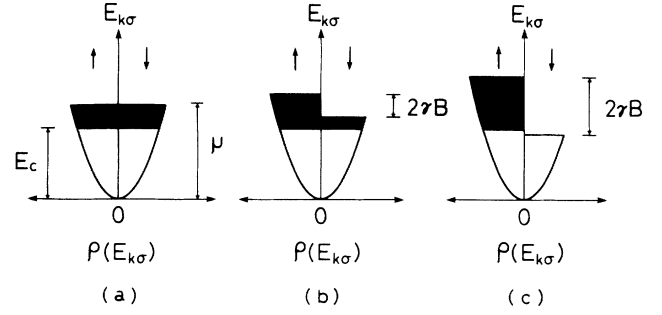


FIG. 2. Schematic illustrations of the density of states (DOS) under various magnetic fields. The DOS's are shown as a function of the kinetic energy $E_{k\sigma}$ of ^3He quasiparticles with spin σ in a dilute ^3He - ^4He solution with chemical potential μ (a) under zero magnetic field and (b), (c) magnetic field B . The magnetic field for (c) is higher than that of the case (b). The shaded regions indicate that ^3He quasiparticles can diffuse into porous media, i.e., $E_{k\sigma} > E_c$. Figures (b) and (c) correspond to the case I [$n_{\uparrow 1}(0) > n_{\downarrow 1}(0) > n_c$] and the case II [$n_{\uparrow 1}(0) > n_c > n_{\downarrow 1}(0)$], respectively.

constant shows the power-law dependence of Eq. (5) in the region $B \lesssim 10 \text{ T}$.

Let us consider the situation in which ^3He quasiparticles in a dilute ^3He - ^4He solution in one cell can flow through a porous medium into the second cell filled by superfluid ^4He . As argued above, the magnitude of the diffusion constant becomes different among \uparrow and \downarrow spins because these particles have different kinetic energies under high magnetic fields, i.e., the dominant spin species

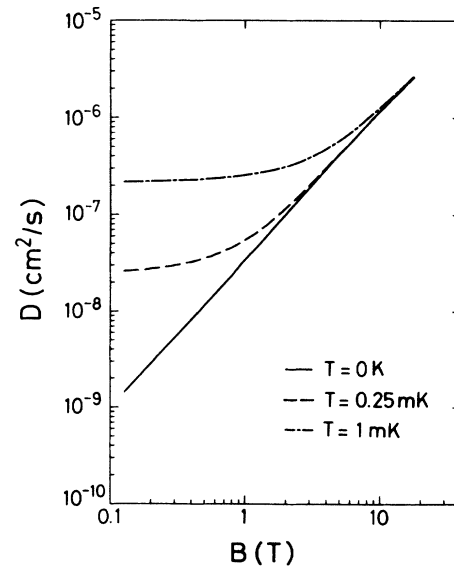


FIG. 3. The diffusion constants D_\uparrow of ^3He quasiparticles in dilute ^3He - ^4He solution with $\mu = E_c$ are calculated as a function of magnetic fields from Eq. (3) for various temperatures. We have used the following values of the parameter: the pore diameter $a = 50 \text{ \AA}$, the channel length $l = 50 \text{ \AA}$, and the percolation density of the porous medium $p_{st} = 0.5$, respectively. The critical energy is obtained as $E_c = 70 \text{ mK}$ and the known exponents for percolating nets, $\beta = 0.4$ and $t = 2.0$, are used.

diffuses faster. This implies that the spin polarization in the second cell will be enhanced. In the next paragraph we will discuss the time development of the spin polarization in each cell.

Consider the case that two cells of each volume V_1 and V_2 are separated by a thin porous medium with the thickness L ($z=0$ and $z=L$). The current density of ${}^3\text{He}$ quasiparticles with spin σ is written by

$$J_\sigma = -D_\sigma \frac{\partial n_\sigma}{\partial z}, \quad (6)$$

where the diffusion constant D_σ is defined by Eq. (3), and n_σ is the number density of ${}^3\text{He}$ quasiparticles with spin σ in the porous medium. The time derivative of the density $n_{1\sigma}$ in cell 1 is proportional to the current density J_σ as

$$\frac{dn_{1\sigma}}{dt} = -\frac{A}{V_1} J_\sigma(0), \quad (7a)$$

where A is the effective cross section of the porous medium at $z=0$. The identical equation holds for the density $n_{2\sigma}$ in the other cell,

$$\frac{dn_{2\sigma}}{dt} = \frac{A}{V_2} J_\sigma(L). \quad (7b)$$

The current density of ${}^3\text{He}$ quasiparticles in the porous medium can be determined by Eq. (6) and the continuity condition for the density of the form $\partial n_\sigma / \partial t = \partial J_\sigma / \partial z$. It is obvious that, as long as the total void space in the porous medium is much smaller than that of each cell, the density reaches the steady state faster ($\partial n_\sigma / \partial t = 0$) in the porous medium. Thus the current density is obtained by integrating Eq. (6) with respect to z from 0 to L ,

$$J_\sigma = -\frac{1}{L} \int_{n_{1\sigma}}^{n_{2\sigma}} D_\sigma dn_\sigma, \quad (8)$$

where $n_{1\sigma}$ and $n_{2\sigma}$ indicate the boundary conditions for n_σ at $z=0$ and $z=L$, respectively. The time derivative of the density for ${}^3\text{He}$ quasiparticles with spin σ in the cell i is obtained from Eqs. (7) and (8),

$$\frac{dn_{i\sigma}}{dt} = (-1)^{i+1} \frac{A}{LV_i} \int_{n_{1\sigma}}^{n_{2\sigma}} D_\sigma dn_\sigma \quad (i=1,2). \quad (9)$$

We have iteratively computed these equations at $T=2$ mK for the following initial conditions (see also the caption of Fig. 4): the case I in which $n_{1\uparrow}(t=0) > n_{1\downarrow}(t=0) > n_c$, and the case II in which $n_{1\uparrow}(t=0) > n_c > n_{1\downarrow}(t=0)$. Here the critical density n_c , where ${}^3\text{He}$ quasiparticles in the cell cannot diffuse into the porous medium, is given by¹⁰

$$n_c = \frac{(2m_3^* E_c)^{3/2}}{6\pi^2 \hbar^3}. \quad (10)$$

The density in cell 2 is zero for both types. The time dependence of the polarization in the cell i is defined by

$$M_i(t) = \frac{n_{i\uparrow}(t) - n_{i\downarrow}(t)}{n_{i\uparrow}(t) + n_{i\downarrow}(t)}. \quad (11)$$

Let us consider, at first, the case I. Since the relevant velocities of ${}^3\text{He}$ quasiparticles are different for \uparrow and \downarrow spin, ${}^3\text{He}\uparrow$ quasiparticles diffuse faster into cell 2. As a

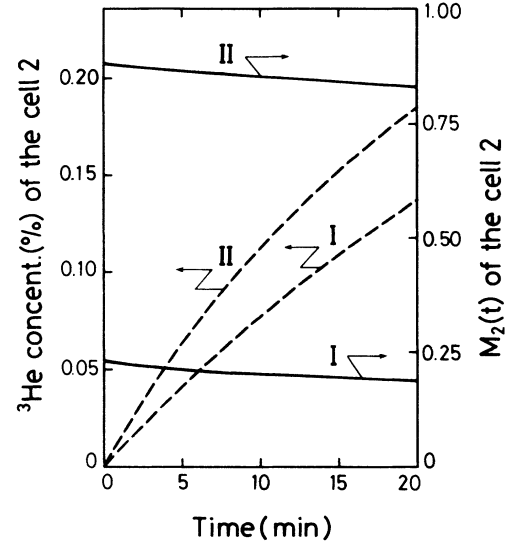


FIG. 4. The time development of the polarization ratio $M_2(t)$ (solid lines) and ${}^3\text{He}$ concentration (dashed lines) in cell 2 for two cases. These two I and II are realized for the cases that 0.59% ${}^3\text{He}$ - ${}^4\text{He}$ solution ($E_F=80$ mK) in cell 1 is polarized under magnetic fields $B=2$ and 12 T at $T=2$ mK; i.e., the initial conditions are taken as $n_c=5.15 \times 10^{19}/\text{cm}^3$, $n_{1\uparrow}(t=0)=6.40 \times 10^{19}/\text{cm}^3$, $n_{1\downarrow}(t=0)=6.00 \times 10^{19}/\text{cm}^3$ for I, and $n_{1\uparrow}(t=0)=7.29 \times 10^{19}/\text{cm}^3$, $n_{1\downarrow}(t=0)=5.11 \times 10^{19}/\text{cm}^3$ for II, respectively. These conditions produce the initial polarization ratio $M_1(0)=0.03$ and 0.18, respectively. We have used the following values of parameters: the volume of cells 1 and 2 are $V_1=4V_2=4$ cm^3 , the thickness of the porous medium $L=1$ μm , the effective area $A=1$ cm^2 , and other parameters the same as those in Fig. 3.

next step, ${}^3\text{He}\downarrow$ quasiparticles flow into cell 2. The time development of the polarization ratio in cell 2 is calculated in Fig. 4, as well as the time dependence of ${}^3\text{He}$ concentration. The mechanism of polarization enhancement of this type is analogous to that suggested by Mullin.⁶ The case II is unique where ${}^3\text{He}\uparrow$ quasiparticles flow dominantly into cell 2. This is because ${}^3\text{He}\downarrow$ quasiparticles do not diffuse through the porous medium. As a result, the polarization in cell 2 becomes almost unity and in cell 1 decreases from the initial one. One sees from the calculated curves for the case II in Fig. 4 that the concentration of ${}^3\text{He}$ quasiparticles in cell 2 increases monotonically as a function of time. This implies that an almost complete spin-polarized ${}^3\text{He}\uparrow$ system with a high concentration of ${}^3\text{He}$ quasiparticles can be produced. It should be emphasized that the mechanism of spin-polarization enhancement of the type II is unique compared with that of the type I because we utilize the fact that the mobility edge exists for the system. It should be noted that the spin-relaxation time due to the interaction with a wall coated by ${}^4\text{He}$ is much larger than the diffusion time $\tau_D \sim L^2/D$ (L is the size of sample) of polarized ${}^3\text{He}$ quasiparticles through the porous medium (for the case of thickness 1 μm , τ_D is of the order of 1 s).¹⁴

In summary, we have studied the mechanism of spin-polarization enhancement for ${}^3\text{He}$ quasiparticles in dilute

^3He - ^4He solutions through a porous medium. It is crucial that the mobility edge for ^3He quasiparticles is different for \uparrow and \downarrow spins under high magnetic fields. Using this magnetic technique, it is possible to produce the degenerate Fermi system with a very high polarization ratio. In particular, an almost completely polarized system with a high concentration of ^3He quasiparticles can be achieved

for the case that the number density of one spin species (\uparrow or \downarrow) under magnetic fields is smaller than the critical density n_c .

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¹For a review, see, for example, G. Frossati, *Jpn. J. Appl. Phys.* **26**, suppl. 26-3, 1833 (1987), and references therein.

²For a review, see, for example, I. F. Silvera and J. T. M. Warraren, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (Elsevier, Amsterdam, 1986), Vol. X, p. 139, and references therein.

³For a review, see, for example, D. S. Betts, F. Laloe, and M. Leduc, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (Elsevier, Amsterdam, 1989), Vol. XII, p. 45, and references therein.

⁴A. E. Meyrovich, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (Elsevier, Amsterdam, 1987), Vol. XI, p. 1.

⁵W. J. Mullin and K. Miyake, *J. Low. Temp. Phys.* **53**, 313 (1983).

⁶W. J. Mullin, *Phys. Rev. Lett.* **57**, 2710 (1986).

⁷We should note that fluids confined in porous media provide interesting phenomena from the percolating aspects and this subject is a topic of much current interest since the work of P. G. de Gennes and E. Guyon, *J. Mec.* **17**, 403 (1978).

⁸A. Ohi and T. Nakayama, this issue, *Phys. Rev. B* **41**, 7322 (1990).

⁹S. Feng, B. I. Halperin, and P. N. Sen, *Phys. Rev. B* **35**, 197 (1987).

¹⁰The diffusion of ^3He quasiparticles is treated as a problem of percolation in a classical point of view from the following reason. A wave packet of ^3He quasiparticles has a spatial extent over $\delta x \sim \hbar/\delta p$, where the thickness of the energy shell at T K is $\delta p \approx 2k_B T/v_F$. This leads us to take the spatial extent as $\xi_0 \approx 0.5/T(\text{\AA})$ by using the Fermi velocity $v_F = 1.3 \times 10^3$

cm/sec for 0.5% solution, i.e., a wave packet of ^3He quasiparticle extends over a few hundred angstrom in the millikelvin temperature region (e.g., $\xi_0 \approx 100 \text{\AA}$ at 5 mK). Since we consider the situation where the average distance between pores is of the order of 100 \AA , the localization effect due to the quantum interference of ^3He quasiparticles is irrelevant.

¹¹Y. Gefen, A. Aharony, and S. Alexander, *Phys. Rev. Lett.* **50**, 77 (1983).

¹²D. Stauffer, *Introduction to Percolation Theory* (Taylor and Francis, London, 1985).

¹³The average time $\bar{\tau}(E_{k\sigma})$ is obtained from the hopping time $\tau(E_{k\sigma}, V_{\text{ch}})$. This is the time that ^3He quasiparticles with the energy $E_{k\sigma}$ diffuse into the adjacent pore through the channel with the barrier height V_{ch} , that is, $\bar{\tau}(E_{k\sigma}) = \int \tau(E_{k\sigma}, V_{\text{ch}}) F(V_{\text{ch}}) dV_{\text{ch}}$. The hopping time $\tau(E_{k\sigma}, V_{\text{ch}})$ is given by $1/\tau(E_{k\sigma}, V_{\text{ch}}) = S(V_{\text{ch}})v_\sigma/S_{p\sigma}a$, where v_σ is the velocity of ^3He quasiparticles with $E_{k\sigma}$. The effective area of a porous medium is $S_{p\sigma} = \pi a^2$, and the cross section of channel $S(V_{\text{ch}}) = \hbar^2 \pi^3 / 4m_\sigma^* V_{\text{ch}}$, respectively. As a result, we have

$$\bar{\tau}(E_{k\sigma}) = \frac{6a}{[2(E_{k\sigma} - V_{\text{min}})/m_\sigma^*]^{1/2}} \left[\ln \frac{1 + (1 - V_{\text{min}}/E_{k\sigma})^{1/2}}{1 - (1 - V_{\text{min}}/E_{k\sigma})^{1/2}} - 2(1 - V_{\text{min}}/E_{k\sigma})^{1/2} \right].$$

From this we see that the dominant $E_{k\sigma}$ dependence for $\bar{\tau}(E_{k\sigma})$ becomes $\bar{\tau}(E_{k\sigma}) \sim E_{k\sigma}^{-1/2}$ because of $E_{k\sigma} \gg V_{\text{min}}$.

¹⁴L. Himbert, V. Lefevre-Seguin, P. J. Nacher, J. Dupontroc, M. Leduc, and F. Laloe, *J. Phys. (Paris) Lett.* **44**, L523 (1983).

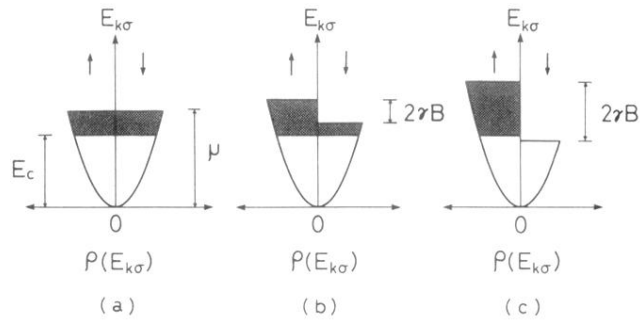


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